Collective Resonances in Nanoparticle Oligomers

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This thesis is an account of research undertaken at the Nonlinear Physics Centre between January 2013 and November 2016, while I was enrolled in the degree of Doctor of Philosophy at the Australian National University. The research was conducted under the supervision of Andrey Miroshnichenko within the Research School of Physics and Engineering. I declare that the material presented within this thesis is my own work except where stated otherwise, and has never been submitted for any degree at this or any other institution of learning.

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Abstract

The study of nanostructured artificial media for optics has expanded rapidly over the last few decades, coupled with improvements of fabrication technology that have enabled investigation of previously unrealisable optical scattering systems. Such development is complemented by renewed impetus to understand the physics of optical scattering from complex subwavelength geometry and nanoparticle systems. Here I investigate specifically the optical properties of closely packed arrangements of nanoparticles, known as nanoparticle oligomers, which provide an intuitive platform for analytical and numerical study on the formation and interplay of collective resonances. I consider both plasmonic nanoparticles, and also high-refractive-index dielectric nanoparticles that support Mie-type electric and magnetic dipole resonances. Specific outcomes of this study are listed as follows. (i) A new model is presented for optical Fano resonances, which is based on interference between nonorthogonal eigenmodes of the associated scattering object. This is demonstrated to correctly describe Fano resonances in both plasmonic and high-refractive-index dielectric nanoparticle oligomers; it also revealed capacity for two-channel Fano interference in the magnetic dipolar response from the dielectric oligomers. (ii) Polarisation-independent scattering and absorption losses are shown to be enforced by $n$-fold discrete rotational symmetry, $C_n$ ($n \geq 3$), and reciprocal degeneracy of eigenmodes. (iii) A new form of circular dichroism is presented, which occurs due to the interaction of nonorthogonal resonances, and impacts the ratio of radiative scattering loss to dissipative absorption loss experienced by reciprocal plane waves. Geometric asymmetry and optical chirality are also reviewed to quantify the minimum symmetries that must be broken to allow other circular dichroism effects in chiral and achiral scattering objects. The sequence of general theoretical conclusions (i)-(iii) serve to build the understanding of optical scattering from nanoparticle systems while removing existing ambiguities.
Notation

Use of fonts

\( u \) scalar
\( \mathbf{u} \) vector (function of frequency)
\( \mathbf{u} \) vector (function of time)
\( \hat{\mathbf{u}} \) unit vector
\( \mathbf{\bar{u}} \) matrix
\( \hat{u} \) operator

Generic terms

\( t \) time
\( \omega \) angular frequency
\( \mathbf{r} \) position vector
\( V, \Omega_V \) a volume and its surface

Electromagnetism

\( \epsilon_0, \mu_0 \) permittivity and permeability of background medium
\( \mathbf{E}(\mathbf{r}, \omega), \mathbf{E}(\mathbf{r}, t) \) electric field
\( \mathbf{J}(\mathbf{r}, \omega), \mathbf{J}(\mathbf{r}, t) \) electric current
\( \mathbf{P}(\mathbf{r}, \omega), \mathbf{P}(\mathbf{r}, t) \) electric polarization
\( \mathbf{p}(\omega), \mathbf{p}(t) \) electric dipole moment
\( \mathbf{B}(\mathbf{r}, \omega), \mathbf{B}(\mathbf{r}, t) \) magnetic B-field
\( \mathbf{H}(\mathbf{r}, \omega), \mathbf{H}(\mathbf{r}, t) \) magnetic H-field
\( \mathbf{m}(\omega), \mathbf{m}(t) \) magnetic dipole moment
\( P(\omega), \sigma(\omega) \) power and cross-section
\( \mathbf{j}_v(\mathbf{r}, \omega), \mathbf{p}_v(\omega), \mathbf{m}_v(\omega) \) currents and dipole moments of an eigenmode \( |v⟩ \)
\( \lambda_v(\omega) \) eigenvalue of an eigenmode \( |v⟩ \)

Symmetry operations

\( \hat{C}_n \) rotation about a principle axis by \( \frac{2\pi}{n} \) radians
\( \hat{\sigma}_v, \hat{\sigma}_d \) reflection plane parallel to the principle axis
\( \hat{\sigma}_h \) reflection plane perpendicular to the principle axis
\( \hat{S}_n \) improper rotation: both \( \hat{C}_n \) and \( \hat{\sigma}_h \)
\( \hat{i} \) point inversion through the origin
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The study of artificial optical media grew from a desire to capitalise on the broad advancements in nanoscale fabrication capacity, and to enable new optical functionality that cannot be realised with conventional materials. Here I will introduce, first: the reason and motivation for research into artificial media and nanoparticle optical systems generally, and second: the physics involved in the optical scattering from nanoparticles. Discussion on these topics is not intended to be comprehensive, but to provide sufficient background and context for the technical Chapters 2-5. This introductory Chapter will then end with a brief overview of the broader goals of the thesis and each of the coming chapters.

1.1 Artificial nanostructured materials for optics

Optical technology is a well-established tool throughout research, manufacturing, and technology; its mark found on some of the foremost achievements of society, from optical microscopy in the discovery of bacteria and microbiology, to the fabrication of semiconductor microprocessors that now saturate our digital age. Yet contemporary demands increasingly call for optical functionality that would lie beyond the limits of conventional, diffractive optics. Indeed, many elusive facets of nature would become readily accessible once we can observe the structure and dynamics of subcellular networks, or protein structure and its folding [1, 2]; or the greater part of energy consumed by computation could be suppressed once we can miniaturise optical communication networks to replace their electrical counterparts within computing infrastructure, and microchips themselves [3, 4].

As part of this push away from conventional optics, the development of artificial media for optics has been receiving a surge of renewed interest over the last two decades. The growing capacity to accurately deposit [5], etch [6] or assemble [7, 8] nanostructured materials has enabled investigation of previously unimagined optical systems, particularly those grouped largely as metamaterials: artificial media made by massed concatenation of subwavelength-sized resonant nanoparticles or general nanostructured inclusions. Conceptually, metamaterials aim to mimic the construction of natural materials from atoms, or molecules, by substituting nanoparticles or analogous inclusions as artificial meta-atoms that remain small relative to the operating wavelength of light. With control over the geometry of the constituent meta-atoms, and their arrangement, it becomes possible to design the optical properties of the collective media. This freedom has allowed artificial media, or nanostructures generally, to be employed for rapidly diversifying research pursuits. Some particular examples of note are: the enhancement of spontaneous emission from nearby molecules [9–12], combined with sharp frequency selective sensing [13, 14] of even single molecules [15]; the imaging of light [16, 17] or objects [18]; control over propagation and state of light [19–22]; and the enhancement of nonlinear harmonic generation,
wavemixing and switching effects [23–26]. However, until very recently, the original concept of bulk metamaterials was impeded at optical frequencies by the limited capacity of three-dimensional material fabrication at application-relevant volumes. Seemingly in a bid to bypass this challenge, optical metamaterials research made a shift toward single- or few-layer realisations of metamaterials, so-called *metasurfaces*, as two-dimensional analogues of the existing metamaterials. The promise underlying optical metasurfaces was perhaps conveyed most concisely by Pfeiffer and Grbic [27], recognising that the surface equivalence principle (Stratton-Chu formulation [28]) implies that a surface of both electric and magnetic currents can perform a reflectionless transformation between any two sets of electric and magnetic fields on opposing sides. In essence, any optical operation becomes possible if we can impose an arbitrary polarisation and magnetisation distribution; this being a functionality that artificial nanostructured media can aim to provide. During the initial surge of metamaterials research, metallic structures were designed to support loop currents in response to light, and create the effective magnetic response to imprint a magnetisation distribution. Perhaps the most iconic of these geometries was the split ring resonator, which permitted a magnetic response using a split in a ring resonator (loop antenna) to reduce the symmetry and permit coupling into oscillating circulating current from a normally incident plane wave [29]. See illustration in Figure 1.1.

![Figure 1.1: Qualitative illustration of magnetic response from nanoparticles. A pair of nested ring resonators with different resonant frequencies, which produce induced currents $J$ that are $\pi$ out of phase, will not allow a net circulation of current (left). However, introducing two splits to suppress part of the current in each ring leads to net circulating current (centre). Alternatively, a high-refractive-index dielectric sphere can instead use the retardation of an applied plane wave’s electric field to produce a circulating polarization current (right).](image)

However, a persisting challenge for this approach was the inherent Ohmic losses of metals leading to unavoidable dissipation of light. While reflection-based operation could be viable and support efficiencies in excess of 50% with metals [30], the maximum operational transmission efficiency, even with metasurfaces, was at [31] or below [32] 50%. Such limitations from dissipative losses were largely resolved by the predictions [33,34] and realisations [35–38] that simple nanoparticles made of low-loss and high-refractive-index dielectric materials, such as silicon or germanium, would inherently support both electric and *magnetic* dipolar optical resonances, with comparable magnitude to the electric dipole resonances of gold or silver nanoparticles. Furthermore, as shown nicely in the Supplementary Information of [37], the magnetic dipole resonance becomes the lowest-frequency resonance as the refractive index of a dielectric sphere increases, with the second resonance being an electric dipole. As such, it wasn’t necessary to fabricate loops or other complicated geometries: high-index dielectric nanoparticles would support resonant circulation...
of polarisation current with very simple geometries such as arrays of spheres, disks, or bars. Subsequent control over both electric and magnetic resonant responses then enabled the metasurface concept presented by Pfeiffer and Grbic [27] to create arbitrary polarisation and magnetisation distributions on a surface, but now with low losses and twin resonances to obtain full $2\pi$ phase control [39]. Current dielectric metasurfaces can now function at above 90% transmission efficiency for focusing [21, 40], holography [21, 41], polarisation control and exotic beam forming [21, 22, 42].

While the original premise of metamaterials was to replicate continuous media [43] with constituent elements much smaller than the operational wavelength, this is rarely the case: the majority of metamaterials and metasurfaces have lattice periods on the order of a wavelength, and therefore have some semblance to photonic crystals [44]. Here I will instead make the claim that the key conceptual distinction of optical metamaterials is that their constituent elements are resonant at the operational wavelength in isolation, being now referred to as nanoantennas. The isolated resonances are what allows almost arbitrary spatial contrast in the phase, amplitude and orientation of imprinted polarisation and magnetisation distributions, and what allows optically thin elements to alter macroscopic forms of light. Indeed, I would argue that the most distinctive physical freedom of metamaterials, the one that supposes to provide a functionality beyond the existing photonic crystals, is the resonant properties of the nanoantennas themselves. My studies have focused on presenting and developing analyses for operational principles of nanoantennas, and particularly those that support multiple interacting resonances. I have particularly been interested in the formation of resonances and collective optical responses arising from coupling between nanoparticles, as the analogy and precursor of artificial materials being formed from constituent nanoantennas. Specifically, I focus on groups of several nanoparticles arranged in closely packed clusters, so-called nanoparticle oligomers.

The concept of nanoparticle oligomers was introduced for metal nanoparticles [45], where they could allow more complicated optical responses, while simultaneously offering simplification in fabrication, which still typically favours basic nanoparticle geometries such as spheres and other primitive shapes. The subsequent transition to incorporate high-index dielectric nanoparticles that support electric and magnetic dipole moments into nanoparticle oligomers, provides an analogue to the local resonant sources of polarisation and magnetisation that nanoantennas represent for artificial materials. This is particularly interesting given nanoparticle oligomers operate between isolated nanoparticle response and collective media response; collective resonances exist in nanoparticle oligomers, but the resonant properties of any individual nanoparticle remain. Indeed, oligomers provide a window to explore the evolution between isolated and collective resonant responses that we utilise in the pursuit of artificial media. This thesis will not emphasize any specific implementation, instead the investigation aims to provide relevant insight on the principles of collective resonances in nanoantennas and artificial nanostructured materials generally.
1.2 Optical scattering from nanoparticles

Here I briefly discuss how distributions of currents and charges presented by the classical electromagnetism model can be parametrised to resemble material phenomena like polarisation and magnetisation. This aims to give a contextual precursor to models for the optical scattering from nanoparticles presented in Chapter 2, and the subsequent parametrisation of currents into the resonant eigenmodes considered in Chapter 3.

We must appropriately begin with Maxwell’s Equations, which describe the evolution of real electric $E$ and magnetic $B$ fields at the position $r$ and time $t$, existing in a homogeneous background medium with permittivity $\epsilon_0$ and permeability $\mu_0$, prescribing a speed of light $c_0 = (\epsilon_0 \mu_0)^{-\frac{1}{2}}$, with some charge $\rho$ and current $J$.

\[ \nabla \cdot B = 0 \]  \hspace{1cm} (1.1)
\[ \nabla \times E + \frac{\partial}{\partial t} B = 0 \]  \hspace{1cm} (1.2)
\[ \nabla \cdot E = \frac{\rho}{\epsilon_0} \]  \hspace{1cm} (1.3)
\[ \nabla \times B - c_0^{-2} \frac{\partial}{\partial t} E = \mu_0 J \]  \hspace{1cm} (1.4)

Note that these equations implicitly assume the continuity of charge $\nabla \cdot J = -\frac{\partial \rho}{\partial t}$, seen by taking the divergence of (1.4), and then substituting (1.3). To define interaction with matter, we consider the Lorentz force imparted on any given charge $q$, moving with a velocity $\mathbf{v}_q$ at a location $r$ and at time $t$.

\[ \mathbf{F}_q = q \mathbf{E}(r,t) + q \left( \mathbf{v}_q \times \mathbf{B}(r,t) \right) \]  \hspace{1cm} (1.5)

Notably, the magnetic field isn’t needed to describe the electromagnetic force on $q$ in the inertial reference frame where it is stationary. The electric field $\mathbf{E}_{q'}$ at position $r$ and time $t$, produced by some arbitrarily moving charge $q'$, located at $r'$ at the retardation-adjusted time $t' = t - \frac{R}{c_0}$, with $R = |r - r'|$, was presented by Feynman, §28 [46], but is equivalent [47] to fields described by Liénard-Wiechert potentials [48].

\[ \mathbf{E}_{q'}(r,t) = \mathbf{E}(r,t) - \mathbf{E}(r,t) = \sum_{q'} \mathbf{E}_{q'}(r,t) \]  \hspace{1cm} (1.6)

Here $\hat{n}$ is the unit vector pointing from $r'$ to $r$, meaning: $r - r' = R \hat{n}$. By then writing the total electric field at $(r, t)$ as a sum over the fields generated by an arbitrary number of $q'$, $\mathbf{E}(r,t) = \sum_{q'} \mathbf{E}_{q'}(r,t)$, the physical force on $q$ is given by $\mathbf{F}_q = q \mathbf{E}$. Albeit not remotely practical, we could correctly model all electromagnetic interaction using only electric fields and different reference frames for each $q$. However, the need for both electric and magnetic fields emerges by defining materials with polarisation $P$, but also magnetisation $M$ that interacts directly with $B$, both of which can be presented as the density of electric dipole moments and magnetic dipole moments per unit volume [48]. In this regard, while the assumption of point dipoles distributed within some background volume is physically reasonable, given existence of atoms and the like, we are concerned with artificial media constructed from nanoparticles that have nonzero volume. This suggests we should make the distinction of introducing polarisation and magnetisation in terms of currents and charge. We therefore consider density distributions of charge and current, $\rho(r) \to d\rho \equiv \rho(r) \, \Delta r^3$, $\mathbf{J}(r) \to d\mathbf{J} \equiv \mathbf{J}(r) \, \Delta r^3$, to define the electric dipole $\mathbf{p}$ and magnetic...
dipole \( \mathbf{m} \) of some volume \( V \) centred about a point \( \mathbf{r} \).

\[
p = \int_{V'} (r' - r) \rho(r') \, d^3r', \quad \mathbf{m} = \frac{1}{2} \int_{V'} (r' - r) \times \mathbf{J}(r') \, d^3r' \tag{1.7}
\]

The equivalent density of point dipoles per unit volume at a point \( \mathbf{r} \), will then be the density of \( p \) or \( m \) in a volume \( V \to 0 \) about \( \mathbf{r} \).

\[
P(\mathbf{r}) = \lim_{V \to 0} \frac{1}{V} \int_{V'} (r' - r) \rho(r') \, d^3r' / V, \quad M(\mathbf{r}) = \lim_{V \to 0} \frac{1}{2} \frac{1}{V} \int_{V'} (r' - r) \times \mathbf{J}(r') \, d^3r' / V \tag{1.8}
\]

Now, a local misalignment of \( \mathbf{E} \) with some \( p \) in the limit of \( V \to 0 \) constitutes a local restoring torque \( \tau_p \) acting on \( p \), and similarly for \( \mathbf{B} \) with \( m \).

\[
\tau_p = \left( \lim_{V \to 0} p \right) \times \mathbf{E}(\mathbf{r}), \quad \tau_m = \left( \lim_{V \to 0} m \right) \times \mathbf{B}(\mathbf{r}) \tag{1.9}
\]

If the timescale for movement of charge and current is small compared to that for variation in the applied field, we can then assume local alignment of \( P \) with \( \mathbf{E} \), and \( M \) with \( \mathbf{B} \), because of the torques \( \tau_p \), \( \tau_m \). More generally, we can consider fields that oscillate harmonically at an angular frequency \( \omega \), for which we can expect some steady state average effect of the torque acting to align dipole moments to the corresponding fields. This thereby lets us enstate a general tensor relationship between respective phasors, specifically the one of permittivity and permeability: \( P = (\bar{\varepsilon} - \epsilon_0) \mathbf{E} \), and \( M = (\bar{\mu} - \mu_0) \frac{1}{\mu_0} \mathbf{B} \). Such relationships are useful because any arbitrary distribution of time-varying fields, \( \mathbf{u} \) for generality, can be expressed as a spectral distribution of harmonic phasors \( \mathbf{u} \) in a causal Fourier representation.

\[
\mathbf{u}(\mathbf{r}, t) = \int_{-\infty}^{\infty} \mathbf{u}(\mathbf{r}, \omega) e^{-i\omega t} \, d\omega, \quad \mathbf{u}(\mathbf{r}, \omega) = \int_{-\infty}^{t_0} \mathbf{u}(\mathbf{r}, t) e^{i\omega t} \, dt \tag{1.10}
\]

I have chosen to end the time integral at a time \( t_0 \), rather than covering the full \((-\infty, \infty)\) interval, to represent the absence of \( \mathbf{u} \) beyond the current time \( t_0 \) in any causal measurement. This allows us to consider \( \mathbf{u} \) as being derived from an observed \( \mathbf{u} \), though it does mean that \( \mathbf{u} \) treats \( \mathbf{u}(t) = 0 \) for \( t > t_0 \). The absence of italics will be used herein to denote complex phasors in the frequency domain.

We now simply recognise that the tensor relationship of \( P, M \) with \( \mathbf{E}, \mathbf{B} \), which defines materials, can arise from the charge and current distributions in (1.8), provided the restoring torques \( \tau_p \) and \( \tau_m \) in (1.9). The expression for \( \tau_p \) in (1.9) follows by substituting \( \mathbf{F} = \rho \mathbf{E} \) from (1.5) and \( p \) from (1.7) into \( \mathbf{p} \times \mathbf{E} \), and assuming \( V \to 0 \) for the electric field to be considered as uniform \( \mathbf{E}(\mathbf{r}') = \mathbf{E}(\mathbf{r}) \). The torque \( \tau_m \) in (1.9) can be derived for point magnetic dipoles [49], or from \( \tau_p \) using an electromagnetic duality transformation \( (\mathbf{E}, c_0 \mathbf{B}, p, c_0 \mathbf{m}) \to (c_0 \mathbf{B}, -\mathbf{E}, c_0 \mathbf{m}, -p) \), though this effectively treats \( \mathbf{m} \) as being due to (non-physical) magnetic charge [50]. Otherwise the derivation of \( \tau_m \) can be considered using just currents, see box. In all cases, the physical notion from the tensor relationship of \( P \) with \( \mathbf{E} \), and \( M \) with \( \mathbf{B} \), which defines material, is now that the electric and magnetic fields are heralds of linear and rotational forces between distributed charges. This will have been at least historically convenient, because it corresponds to the way natural materials respond: atoms and other neutral compositions of charged matter dominantly behave as electric and magnetic dipoles. But let us now consider the artificial case of
**Introduction**

Magnetic dipoles from a current distribution. Let us define the current $J_m$, associated with an $m$, such that it only contains components that contribute to $m$ from (1.7). This means that: $J_m(r') \cdot (r' - r) = 0$ and $J_m(r') \cdot m = 0$. Consider the quantity $m \times B(r)$ with (1.7), where $B$ is the total magnetic field, not just that due to $J_m$.

$$m \times B(r) = \int_{r' \in V} \left[ (r' - r) \times (J_m(r') \times B(r)) + J_m(r') \times (B(r) \times (r' - r)) \right] \, dr'^3 \quad (1.11)$$

Here we have applied the vector identity $(a \times b) \times c = a \times (b \times c) + b \times (c \times a)$. If we take the limit $V \to 0$ to assume $B(r') = B(r)$ and perform a substitution for the local force $F = J_m \times B$ from (1.5), the first term of (1.11) is the torque $\tau_m$. The second term of (1.11) can be expanded with a vector triple product $a \times (b \times c) = b(a \cdot c) - c(a \cdot b)$ to make use the orthogonality of $J_m$ and $(r' - r)$.

$$J_m(r') \times (B(r) \times (r' - r)) = -(r' - r) \cdot (J_m(r') \cdot B(r)) \quad (1.12)$$

This is zero if $J_m$ is perpendicular to $B$, however $B$ is able to be arbitrarily oriented, such as when it is dominated by external sources. The second term of (1.11) is therefore not forbidden by our existing constraints on $J_m$. The expression for torque $\tau_m$ in (1.9) appears to require we make a further assumption about $J_m$ and/or $V$ to make the second term of (1.11) zero, while keeping the first term nonzero. At the very least, if $J_m$ is invariant under arbitrary coordinate rotations about $m$, we can visualise $J_m$ in uniform rings of circulation about $m$, i.e. where $J_m(r')$ at fixed $|r' - r|$ is oriented with uniform magnitude in the direction of $(r' - r) \times m$. The contribution to the torque from (1.12) integrated over any such ring is then orthogonal to $B$ and $m$, hence at least parallel to $\tau_m$ in (1.9).

A given nanoparticle, occupying a volume $V$, which too only supports an electric and a magnetic dipole moment, and we can write these dipole moments in the frequency domain, i.e. $p$ and $m$, using (1.10).

$$p = \int_V (r - r') \rho(r') \, dr'^3, \quad m = \frac{1}{2} \int_V (r - r') \times J(r') \, dr'^3 \quad (1.13)$$

We can place a hypothetical bounding sphere around the nanoparticle and use the result of Devaney and Wolf [51], which states that all fields external to any sphere that bounds an arbitrary object will be precisely described by the radiation of the complete set of spherical harmonic multipoles. Moreover, the radiation of the given dipole moments $p$ and $m$ in (1.13) can be described by some corresponding $a_1$ and $b_1$ scattering coefficients [52]:

$$p = \frac{6\pi i \epsilon_0}{k^3} \left[ \frac{1}{\sqrt{2}} (a_{11} - a_{1-1}) \hat{x} + \frac{i}{\sqrt{2}} (a_{11} + a_{1-1}) \hat{y} + a_{10} \hat{z} \right] \quad (1.14a)$$

$$m = \frac{6\pi i}{k^3} \sqrt{\frac{\epsilon_0}{\mu_0}} \left[ \frac{i}{\sqrt{2}} (b_{11} - b_{1-1}) \hat{x} + \frac{i}{\sqrt{2}} (b_{11} + b_{1-1}) \hat{y} + b_{10} \hat{z} \right] \quad (1.14b)$$

Further details on the spherical multipole decomposition, and spherical nanoparticles, are provided in the second box. It is also worth mentioning that, unlike (1.14), the general relationship between the dipole moments $p$ and $m$ in (1.13) and the total $a_1$ and $b_1$ coefficients, will require a series of additional correction terms to the dipole moments, where each correction term radiates identically to the $a_1$ or $b_1$ coefficient [69]. These
correction terms generally become more relevant as the volume \( V \), of the physical system in (1.13), approaches the order of the wavelength of light. However, if we instead have prescribed knowledge of the \( a_1 \) and \( b_1 \) coefficients, then we can choose to define effective dipole moments \( \mathbf{p} \) and \( \mathbf{m} \) of point dipoles that will radiate identically to the known \( a_1 \) and \( b_1 \) coefficients. This is what is done for the case of spheres in (1.21). Such effective dipole moments can be different to those defined in (1.13), given they will account also for the additional correction factors. In any case, we return to our earlier discussion: the mentioned result of Devaney and Wolf [51], combined now with the equivalence of radiation from \( \mathbf{p} \) and \( \mathbf{m} \) to that described by \( a_1 \) and \( b_1 \), allows us to conclude that all fields external to the smallest bounding sphere around the given dipolar nanoparticle are the same as the fields radiated by point dipoles with moments \( \mathbf{p} \) and \( \mathbf{m} \). This means that an oscillating or circulating current in a nanoparticle is indistinguishable, at points external to a bounding sphere, to a point of “true” polarisation or magnetisation. This is a conclusion at the heart of artificial optical materials: it explains why sufficiently small nanoparticles are able to serve, at least optically, as building blocks of artificial materials in a manner analogous to atoms in conventional materials. There are some residual distinctions, such as effective magnetisation from a nanoparticle will be induced by the anti-symmetric component of the electric field over a volume \( V \neq 0 \) of \( \mathbf{m} \) in (1.7), and not the magnetic field, which is relevant to \( V \to 0 \). However, this distinction is also a reason for the access to strong magnetic responses when using nanoparticles: the larger displacements \( (\mathbf{r} - \mathbf{r}') \) of circulating currents in (1.13), relevant to \( \mathbf{m} \). Yet, we must ultimately recognise that the models of dipoles, polarisation and magnetisation still inherently correspond to material as found in nature, and we have no particular guarantee that assemblies of arbitrarily shaped subwavelength nanoantennas will respond to fields as per linear or rotational movement of current. Indeed, the macroscopic description of polarisation and magnetisation has already been recognised to not align with even layered media [56], and particularly as we reduce symmetry, the resonant optical responses of even single nanoparticles cease to align with purely electric or magnetic dipoles [57]. As such, it is not necessarily appropriate to use a homogenised polarisation and magnetisation description for nanostructured media, indeed different models and analyses are necessary to model the scattering of light from assemblies of nanoparticles. This is the reason why alternate modelling approaches are presented in Chapter 2, and it also illustrates a motivation to parametrise optical responses instead according to eigenmodes in Chapter 3 onwards.
The spherical multipole decomposition. Details here on the spherical multipole decomposition are from §9 of [48], and also from [53], details on the scattering from a sphere are from §4 of [54]. The vector spherical harmonics $X_{lm}$ are origin-dependent vector fields defined from the scalar spherical harmonics $Y_{lm}$ as per:

$$X_{lm} = \frac{1}{\sqrt{l(l+1)}} (r \times \nabla) Y_{lm}, \quad Y_{lm} = \sqrt{\frac{2l+1}{4\pi} \frac{(l-m)!}{(l+m)!}} P^m_l(\cos \theta) e^{im\phi} \quad (1.15)$$

Here $\theta$ and $\phi$ are the azimuthal and polar angles of $\mathbf{r}$, and $P^m_l$ is the associated Legendre function. The scattering coefficients $a_{lm}$ and $b_{lm}$ denote complex amplitudes for the decomposition of a given $\mathbf{E}$-field distribution in terms of the vector spherical harmonics:

$$\mathbf{E}(\mathbf{r}) = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} \frac{a_{lm}}{k} \mathbf{\nabla} \times \left( f_l(k|\mathbf{r}|) \mathbf{X}_{lm} \right) + b_{lm} g_l(k|\mathbf{r}|) \mathbf{X}_{lm} \quad (1.16)$$

The functions $f_l(k|\mathbf{r}|)$ and $g_l(k|\mathbf{r}|)$ are normalisation factors for the $a_{lm}$ and $b_{lm}$ coefficients of the form: $f_l(k|\mathbf{r}|) = A_l h_l^{(1)}(k|\mathbf{r}|) + B_l h_l^{(2)}(k|\mathbf{r}|)$, where $A_l$ and $B_l$ are free $l$-dependent coefficients, and $h_l^{(1)}$ and $h_l^{(2)}$ are the $l$-th order spherical Hankel functions of the first and second kind. The specific normalisation I use corresponds to that for homogeneous spheres in §4 of [54], which provides analytic solutions for $a_{lm}$ and $b_{lm}$ under plane wave illumination, known as Mie Theory [55]. Being spheres, the orientation of the plane wave polarisation can be neglected, subsuming the $m$ dependence to define scalar $a_l$ and $b_l$ coefficients. Specifically, for a sphere of radius $n$, and refractive index $n$, the $a_l$ and $b_l$ coefficients due to a plane wave with unit amplitude at the origin are expressed in terms of Ricatti-Bessel functions: $\psi_l(x) = x j_l(x)$ and $\xi_l(x) = x h_l^{(1)}(x)$, and their derivatives with respect to $x$ (denoted here as $\psi'$ and $\xi'$), where $j_l$ is the $l$-th order spherical Bessel function of the first kind.

$$a_l = \frac{n \psi_l(nr) \psi'_l(r) - \psi_l(r) \psi'_l(nr)}{n \psi_l(nr) \xi'_l(r) - \xi_l(r) \psi'_l(nr)} \quad (1.17)$$

$$b_l = \frac{\psi_l(nr) \psi'_l(r) - n \psi_l(r) \psi'_l(nr)}{\psi_l(nr) \xi'_l(r) - n \xi_l(r) \psi'_l(nr)} \quad (1.18)$$

For the specific case of $l = 1$, we can use the following substitutions:

$$\psi_1(x) = \frac{\sin x}{x} - \cos x, \quad \psi'_1(x) = \frac{\cos x}{x} - \frac{\sin x}{x^2} + \sin x \quad (1.19)$$

$$\xi_1(x) = e^{ix} \left( - 1 - \frac{i}{x} \right), \quad \xi'_1(x) = e^{ix} \left( - i + \frac{1}{x} + \frac{i}{x^2} \right) \quad (1.20)$$

We then obtain a simple relationship between the dipole moments of spheres and the spherical scattering coefficients for a plane wave with field $\mathbf{E}_0, \mathbf{H}_0$ at the origin.

$$\mathbf{p} = \frac{6\pi i}{k^3} a_1 \mathbf{E}_0, \quad \mathbf{m} = \frac{6\pi i}{k^3} b_1 \mathbf{H}_0 \quad (1.21)$$
1.3 Outline of context statement

This thesis presents a set of analysis tools that were developed to rigorously quantify the collective optical resonances of coupled nanoparticles in oligomer arrangements, before then presenting arguments that use these tools to explore a priori the properties of geometry and resonances that impose specific optical effects. The three specific optical effects I emphasize are: Fano resonances in dielectric nanoparticle oligomers (Chapter 3), polarisation-independent scattering and absorption (Chapter 4), and circular dichroism in absorption (Chapter 5), which are the key results presented over the six works in Appendix A. This thesis ultimately aims to collate and contextualise these six works, in addition to introducing retrospective insights. The relations between each paper and the Chapters are illustrated in Figure 1.2.

Figure 1.2: The six papers from Appendix A with their relationships denoted by arrows, and the corresponding Chapters denoted by colour.

Regarding the structure of the thesis itself, Chapter 2 firstly presents to models that were used in my studies to analytically quantify, and numerically simulate, optical scattering in nanoparticle systems. This is essentially providing a means for retrospective analysis of a fixed nanoparticle system, from which Chapter 3 then shows that the eigenmodes of these models provide a useful basis to quantify any given geometry. This particular chapter then also presents the argument as to why nonorthogonal eigenmodes are necessary for interference phenomena to exist, particularly Fano resonances. In the following two Chapters 4 and 5, I am able to consider the optical properties of an undefined geometry in terms of a generic set of eigenmodes. This serves as an attempt to relate realistic geometric design considerations to a designated optical effect by investigating corresponding necessary properties of the generic geometry’s eigenmodes. Chapter 4 uses this approach to relate geometric symmetry to the degeneracies of eigenmodes, and a subsequent derivation that discrete rotational symmetry leads to polarization-independent scattering and absorption properties. Chapter 5 then illustrates a different form of prospective geometric relation, starting instead from an assumption of nonorthogonal eigenmodes, from which a new form of circular dichroism in absorption is shown to be possible. Here the knowledge that Fano
resonances imply nonorthogonal eigenmodes, from Chapter 3, allows us to repurpose empirical knowledge of oligomer geometries that support Fano resonances to realise circular dichroism in absorption with planar chiral oligomers. This then completes the thesis; the chapters having served to trace successive steps that illustrate a manner in which one can impose specified scattering quantities from geometric design freedoms by analysing the necessary properties of eigenmodes. The specific summary of each chapter is now listed.

- Chapter 2 defines the induced current model and the coupled dipole model, which I use for derivations, analysis and numerical simulation of optical scattering in the subsequent chapters. These two models are amended versions of the models presented in the works of Appendix A, each having the benefit of cumulative and retrospective developments encountered during my studies. This Chapter is therefore intended to serve as a reference for similar modelling, and also for justifying my choices in utilising these specific models.

- Chapter 3 presents the analysis technique for describing Fano resonances as interference between the nonorthogonal eigenmodes, and its implementation for nanoparticle oligomer systems. This contrasts plasmonic and high-index dielectric nanoparticle oligomers, culminating in a demonstration of Fano interference occurring between multiple magnetic dipolar resonances, specific to dielectric nanoparticle oligomers.

- Chapter 4 discusses the effects of geometric symmetry and reciprocity on the optical properties of resonant nanostructures. This revisits the derivation of polarisation-independent scattering and absorption losses due to discrete rotational symmetry in [A.1], using retrospective knowledge of reciprocal eigenmode degeneracy in [A.6].

- Chapter 5 considers instead the absence of symmetry and discusses both geometric and optical chirality, relating to their influence on scattering properties of nanostructures and their resonances. It reviews circular dichroism effects from the perspective of symmetries, before presenting a new form of circular dichroism in the material absorption that is attributed to interaction between nonorthogonal resonances.
Models for optical scattering

Chapter 2

For the context of investigating scattering from nanoparticle systems, it is necessary to have models to describe scattering systems that resemble neither homogenised media nor simple Rayleigh scatterers. Here I compile the two modelling approaches used in the papers of [A], where I remove variations between papers while also introducing some retrospective insights. The first section presents the description of scattering in terms of combined free currents and polarisation currents, which allows us to directly consider the physical source of fields within any nanostructured optical system, thereby providing a largely unapproximated model for optical scattering. We then present the coupled dipole model as a practical simplification that allows direct investigation and straightforward simulation for the dominant resonances of compact nanoparticle systems. This model is tailored specifically for describing nanoparticle oligomer geometries, and particularly those consisting of high-index dielectric nanoparticles.

2.1 Induced current model

We begin our analysis of linear optical scattering systems by acknowledging there is no need to recognise the distinction between oscillating free current and polarisation current. Any tensor conductivity $\bar{\sigma}$ and susceptibility $\bar{\chi}$ can be incorporated into an effective permittivity $\bar{\epsilon}$ that relates electric field $\mathbf{E}$ to a total electric current $\mathbf{J}$ containing both free and polarisation currents.

$$\bar{\epsilon} \equiv (\bar{\chi} + 1)\epsilon_0 - \bar{\sigma}/i\omega \Rightarrow \mathbf{J}(\mathbf{r}, \omega)e^{-i\omega t} = -i\omega [\bar{\epsilon}(\mathbf{r}) - \epsilon_0] \cdot \mathbf{E}(\mathbf{r}, \omega)e^{-i\omega t} \quad (2.1)$$

There is also the simplification that most optical materials have a negligible permeability difference to the background medium, allowing us to neglect the radiation from any magnetisation current. It is also notationally convenient to use the $\mathbf{H}$-field, which is now defined relative to the uniform background permeability $\mathbf{B} = \mu_0 \mathbf{H}$. To relate the currents $\mathbf{J}$ to fields $\mathbf{E}_s, \mathbf{H}_s$ they radiate, we can make use of the dyadic Green’s function $\mathbf{G}_0$.

$$\mathbf{G}_0(\mathbf{r}, \mathbf{r}') = \left[ \mathbf{I} + \frac{1}{k^2} \nabla \nabla \right] \frac{e^{ikR}}{4\pi R} \left( \begin{array}{c}
\begin{array}{c}
(1 + \frac{i}{kR} - \frac{1}{k^2 R^2}) \mathbf{I} - \left(1 + \frac{3i}{kR} - \frac{3}{k^2 R^2}\right) \hat{n}\hat{n}^T
\end{array}
\end{array} \right) \quad (2.3)$$
where $R = |\mathbf{r} - \mathbf{r}'|$ and $\mathbf{n}$ is the unit vector pointing from $\mathbf{r}'$ to $\mathbf{r}$, in other words: $R\mathbf{n} = \mathbf{r} - \mathbf{r}'$. This is a solution for the dyadic wave equation:

$$\nabla \times \nabla \times \tilde{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}') - k^2 \tilde{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}') = \delta(\mathbf{r} - \mathbf{r}')\mathbf{I}$$

(2.4)

where $\delta$ is a Dirac delta function and $\mathbf{I}$ is the identity matrix. This is notably relevant for the wave equation for the electric field shown in (2.5), which is obtained from substituting (1.2) into (1.4) while assuming harmonic $e^{-i\omega t}$ time dependence.

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}) - k^2 \mathbf{E}(\mathbf{r}) = -i\omega\mu_0\mathbf{J}(\mathbf{r})$$

(2.5)

As such, the dyadic Green’s function specifies the radiation from a Dirac delta point source of electric current at $\mathbf{r}'$. The electric field $\mathbf{E}_s$ radiated by an arbitrary distribution of electric current can therefore be expressed using the dyadic Green’s function by integrating the electric fields generated from each point of electric current [58].

$$\mathbf{E}_s(\mathbf{r}) = i\omega\mu_0 \left( \text{P.V.} \int \left[ \tilde{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}') - \mathbf{L} \frac{\delta(\mathbf{r} - \mathbf{r}')}{k^2} \right] \cdot \mathbf{J}(\mathbf{r}') \, d\mathbf{r}'^3 \right)$$

(2.6)

$$\mu_0 \mathbf{H}_s(\mathbf{r}) = \frac{1}{i\omega} \nabla \times \mathbf{E}_s(\mathbf{r})$$

from (1.2) (2.7)

Here $k$ is the wavenumber, $\omega$ is the angular frequency, $\epsilon_0$ and $\mu_0$ are the permittivity and permeability of the background medium, and the volume of the scattering object is assumed to be finite. The P.V. implies a principal value exclusion of $\mathbf{r}' = \mathbf{r}$ when performing the integration, and $\mathbf{L}$ is the source dyadic necessary to account for the shape of the infinitesimal volume that forms this exclusion [58]. Source dyadics have been derived for different shaped exclusions, the simplest being spheres or cubes: $\mathbf{L} = \frac{1}{3}$, but more complicated expressions have also been derived for ellipsoids, cylinders, rectangular parallelepiped and others [58]. The different source dyadics are necessary to ensure the same electric field is obtained, irrespective to the shape of the exclusion. This point becomes necessary in numerical simulations, which consider discretised meshes of continuous objects and thereby exclude self-interaction in a single mesh element, which requires a source dyadic that can account for the shape of the exclusion volume. From the expression (2.6), and referring to (2.3), can now take the opportunity to define explicitly the near-field as the component of $\mathbf{E}_s$ that scales with $R^{-2}$ or $R^{-3}$, and the far-field as the component scaling with $R^{-1}$. The time-average power flux density $\frac{1}{2}(\mathbf{E}_s^\ast \times \mathbf{H}_s)$ of the far-field scales as $R^{-2}$, while its total flux area expands with $R^2$, meaning the total energy of these scattered fields do not decay: they propagate and can be observed at distances very far from the source. By the same reasoning, the near-field will decay the further from the source they get: they do not propagate, but will dominate the total field at very small $R$. We can now solve for the current induced by an external electric field $\mathbf{E}_0$. The total internal field $\mathbf{E} = \mathbf{E}_0 + \mathbf{E}_s$ is related to the current through the effective permittivity in (2.1), therefore we can combine this with $\mathbf{E}_s$ in (2.6), to relate the induced currents and the external field.

$$-i\omega(\bar{\epsilon}(\mathbf{r}) - \epsilon_0) \cdot \mathbf{E}_0(\mathbf{r}) = \mathbf{J}(\mathbf{r}) - \frac{k^2}{\epsilon_0} [\bar{\epsilon}(\mathbf{r}) - \epsilon_0] \left( \text{P.V.} \int \left[ \tilde{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}') - \mathbf{L} \frac{\delta(\mathbf{r} - \mathbf{r}')}{k^2} \right] \cdot \mathbf{J}(\mathbf{r}') \, d\mathbf{r}'^3 \right)$$

(2.8)

In the absence of magnetisation, (2.8) will determine the currents induced in any finite object under any arbitrary excitation, then (2.6) will describe the fields radiated by this
current. It is, however, highly nontrivial to obtain general solutions for (2.8), even with
very simple geometries. Therefore, I use (2.8) primarily as an analytical model for investigat-
ging general principles of scattering systems. In Chapter 3 the current model is used to
relate far-field interference features to the nonorthogonality of different resonant distribu-
tions of current, and in Chapters 4 and 5 it is used to explore consequences of symmetry
and asymmetry. In these pursuits, I focus on how the energy within any given current
distribution is lost. Specifically, we consider two broad loss channels: power transported
elsewhere as electromagnetic radiation, or power transported elsewhere as anything that
isn’t electromagnetic radiation. The former is radiative losses, which will represent the
power of scattering when the currents are induced by externally applied electromagnetic
fields, and the latter is dissipative losses, which we will call absorption, and encompasses
heat generation, photocurrents, and other linear loss mechanisms\(^1\). Assuming a lossless
background medium, the total radiated power can be calculated by considering any sur-
face encompassing the current distribution, which is finite by our initial definitions, and
performing a surface integral of the normal component of time-averaged Poynting vector
for the scattered fields \(S_s = \frac{1}{2} \text{Re}\{E_s \times H_s\}\), as per (2.6) and (2.7). Such a calculation is
relatively straightforward to implement numerically, but analytically we can rely on the
derivations of Markel [59], who calculated the total radiation by an arbitrary arrangement
of electric dipoles bound by an infinite spherical surface.

\[
P_{\text{scat}} = \frac{1}{2} \sqrt{\frac{\varepsilon_0}{\mu_0}} (4\pi\varepsilon_0)^{-2} \left( \sum_i \frac{8\pi k^4}{3} |\mathbf{p}_i|^2 + 4\pi k \sum_{j \neq i} \text{Im} \left\{ \mathbf{p}_i \cdot (4\pi k^2 \mathbf{G}_0(\mathbf{r}_i, \mathbf{r}_j) \cdot \mathbf{p}_j \right\} \right) \quad (2.9)
\]

Here I have added coefficients as necessary adjustment factors to express Markel’s result
in SI units for \(\mathbf{p}\). We also need to note that the right hand term \(\mathbf{G}_0(\mathbf{r}_i, \mathbf{r}_j) \cdot \mathbf{p}_j\) is related
to the radiated electric field at \(\mathbf{r}_i\) by the dipole at \(\mathbf{r}_j\), the expression for which I will later
cover in (2.20). The next step simply requires translating (2.9) to apply to continuous
systems. We first write a current distribution \(\mathbf{J}(\mathbf{r})\) as an equivalent polarization distribution
\(\mathbf{P}(\mathbf{r}) = -i\omega \mathbf{J}(\mathbf{r})\). Each infinitesimal volume \(d\mathbf{r}\), over which \(\mathbf{P}\) can be considered constant,
will thereby have a dipole moment defined as \(\mathbf{p}(\mathbf{r}) = \mathbf{P}(\mathbf{r}) d\mathbf{r}\). Our continuous current
representation can then be represented with the set of dipole moments \(\{\mathbf{p}(\mathbf{r})\}\), which we
can substitute into (2.9), and noting that the summations in (2.9) will become integrals.

\[
P_{\text{scat}} = \frac{1}{2} \sqrt{\frac{\varepsilon_0}{\mu_0}} (4\pi\varepsilon_0)^{-2} \int \left[ \frac{8\pi k^4}{3} |\mathbf{P}(\mathbf{r}_i)|^2 d\mathbf{r}_i \right]
+ (4\pi)^2 k^3 \left( \text{p.v.} \int \text{Im} \left\{ \mathbf{P}^*(\mathbf{r}_i) \cdot \left( \mathbf{G}_0(\mathbf{r}_i, \mathbf{r}_j) - \mathbf{G}_0(\mathbf{r}_j, \mathbf{r}_i) \right) \cdot \mathbf{p}(\mathbf{r}_j) \right\} d\mathbf{r}_j^3 \right) \quad (2.10)
\]

We can now neglect the first term in (2.10) because it is proportional to \(d\mathbf{r}^3\) after volume
integration. Meaning this term will get arbitrarily small as the voxel gets arbitrarily small,
and it is physically equivalent to the radiation of the single voxel in isolation: it is the only
term that remains if we define \(\mathbf{P}(\mathbf{r}_j) = 0\) at all \(\mathbf{r}_j \neq \mathbf{r}_i\). The expression for scattered power

\[\text{implies}\]

\[\text{is}\]

\[\text{that}\]

\[\text{only}\]

\[\text{become}\]

\[\text{relevant}\]

\[\text{at}\]

\[\text{high}\]

\[\text{electric}\]

\[\text{field}\]

\[\text{intensities}.\]
after neglecting such terms can now be written in terms of currents given: $J = -i\omega P$.

$$P_{\text{scat}} = \frac{\omega \mu_0}{2} \left( \text{P.V.} \int \int \text{Im} \left\{ J^*(r) \cdot \left( \mathbf{G}_0(r, r') - \mathbf{L} \frac{\delta(r - r')}{k^2} \right) \cdot J(r') \right\} \text{d}r^3\text{d}r'^3 \right)$$  \hspace{1cm} (2.11)

$$= -\frac{1}{2} \int \text{Re} \{ J^*(r) \cdot \mathbf{E}_a(r) \} \text{d}r^3 , \quad \text{by substituting (2.6)} \hspace{1cm} (2.12)$$

To now calculate the total absorbed power there are two immediate options: perform a power balance calculation globally, or perform a power balance locally (at each point) and integrate over the whole space. The global power balance refers to calculating absorption as the difference between the net input power $P_{\text{in}}$ entering the system and the net electromagnetic power leaving the system, which is a surface integral of the total time-averaged Poynting vector leaving any bounding surface $\Omega$ around the scattering object.

$$P_{\text{abs}} = P_{\text{in}} - \oint_{\Omega} \frac{1}{2} \text{Re} \{ \mathbf{E}^*(r) \times \mathbf{H}(r) \} \cdot d\mathbf{A}$$  \hspace{1cm} (2.13)

Here $d\mathbf{A}$ is the area differential element represented by a surface-normal vector. This calculation is straightforward to implement numerically for scattering of propagating electromagnetic waves, because $P_{\text{in}} = 0$, but otherwise it will require the value of $P_{\text{in}}$ to calculate absorption. The general advantage of this calculation approach in (2.13) is that it doesn’t require knowledge of either the scattering object’s material or specific geometry, other than requiring a bounding surface. The disadvantage is that we need to know $P_{\text{in}}$, which becomes challenging whenever the excitation source is complicated, as might be the case for modelling nonlinear mixing or harmonic generation processes. It is then often instead better to use knowledge of the material and geometry to calculate the local power balance within the material. Draine [60] derived an expression on the total absorption in arbitrary systems of electric dipole moments, by calculating the local difference between the radiative and total losses of each individual dipole.

$$P_{\text{abs}} = -\frac{1}{2} \sqrt{\frac{\epsilon_0}{\mu_0}} \frac{k}{\epsilon_0} \sum_i \frac{k^3}{6\pi} |p_i|^2 + p_i^* \cdot \text{Im} \{ \bar{\alpha}_i^{-1} \cdot p_i \}$$  \hspace{1cm} (2.14)

We can again translate this expression to consider a continuous current system using the discretisation previously considered for scattered power, which amounted to $J(r)\text{d}r^3 \equiv -i\omega p(r)$, and implies that $\bar{\alpha}_r \epsilon_0 = (\bar{\epsilon}(r) - \epsilon_0)\text{d}r^3$. When performing this substitution from $p$ to $J$, the term $|p_i|^2 \rightarrow |p_{\text{r}}|^2 = \frac{2}{\omega} \text{d}r^6$ will notably remain proportional to $\text{d}r^3$ after the volume integration. As such, this term becomes arbitrarily small and can be neglected, meaning we neglect the contribution to absorption from each voxel in isolation, which is reasonable given each voxel is infinitesimally small.

$$P_{\text{abs}} = -\frac{1}{2\omega} \int J'(r) \cdot \text{Im} \{ (\bar{\epsilon}(r) - \epsilon_0)^{-1} \} \cdot J(r) \text{d}r^3$$  \hspace{1cm} (2.15)

Note that, by writing in terms of the total electric field $\mathbf{E}$ using $J = -i\omega(\bar{\epsilon}(r) - \epsilon_0)\mathbf{E}$, the expression in (2.15) becomes:

$$P_{\text{abs}} = \frac{1}{2} \int \text{Re} \{ \mathbf{E}'(r) \cdot J(r) \} \text{d}r^3$$  \hspace{1cm} (2.16)
The final way to quantify optical response is the total amount of power that interacts with the given object; this quantity is the sum of $P_{\text{scat}}$ and $P_{\text{abs}}$, and it is known as the extinction.

$$P_{\text{ext}} = P_{\text{scat}} + P_{\text{abs}}$$

$$= -\int \left( \frac{1}{2} \text{Re}\{\mathbf{J}(\mathbf{r}) \cdot \mathbf{E}_s(\mathbf{r})\} + \frac{1}{2\omega} \mathbf{J}^*(\mathbf{r}) \cdot \text{Im}\{(\bar{\epsilon}(\mathbf{r}) - \epsilon_0)^{-1}\} \cdot \mathbf{J}(\mathbf{r}) \right) \, d^3 \mathbf{r} \quad (2.17)$$

If we now consider the input power of our system to be written in terms of an externally applied field $\mathbf{E}_0$, it is then possible to write the scattered field $\mathbf{E}_s$ as the difference of total and incident fields $\mathbf{E}_s = \mathbf{E} - \mathbf{E}_0$, and use (2.1) to equate $\mathbf{E} = \frac{1}{\omega}(\bar{\epsilon}(\mathbf{r}) - \epsilon_0)^{-1} \cdot \mathbf{J}$. If we now substitute $\mathbf{E}_s = \frac{1}{\omega}(\bar{\epsilon}(\mathbf{r}) - \epsilon_0)^{-1} \cdot \mathbf{J} - \mathbf{E}_0$ into (2.17) we are then able to obtain a simplified expression for extinction.

$$P_{\text{ext}} = \frac{1}{2} \int \text{Re}\{\mathbf{E}_0^*(\mathbf{r}) \cdot \mathbf{J}(\mathbf{r})\} \, d^3 \mathbf{r} \quad (2.18)$$

This is now precisely a volume integral of the time-averaged power imparted locally by the fields $\mathbf{E}_0$ on the currents $\mathbf{J}$ [61], and therefore shows that extinction is the total amount of power removed from the excitation fields $\mathbf{E}_0$. Extinction is often therefore considered to be equal to the difference of input and output power ($1 - \text{transmission}$) through surfaces or media, but this should be treated carefully as it neglects the power of the scattered field that is co-propagating with the incident field. An ideal $\lambda/2$ waveplate has 100% transmission when rotating the linear polarisation of an incident plane wave by $\pi/2$, but thereby produces 100% extinction, because none of the original plane wave with original polarisation remains. Similarly, reflectionless metasurfaces have also been shown to allow near 100% co-polarized transmission, while simultaneously altering the phase of this transmission anywhere over the complete $2\pi$ range of phase shifts, defined relative to the original illumination [39]. That the resonators in the metasurface are able to change the phase of a wavefront implies they must be the source of significant scattering, and hence extinction, yet transmission remains near 100%. As such, one generally cannot use the extinction calculated through equations such as (2.17) or (2.18) to infer transmission.

Before concluding, it is worth recognising that the quantities of scattering, absorption and extinction, are regularly considered in terms of their corresponding cross-section $\sigma$, which refers to the cross-sectional area of the illuminating plane wave that contains the power $P$. In other words, a plane wave with intensity $I_0 = |\mathbf{E}_0|^2$ will have a power flux density of $\frac{1}{2} \sqrt{\frac{\epsilon_0}{\mu_0}} I_0$, and the area of a cross-section $\sigma$ is therefore related to the corresponding power $P$ as per:

$$P = \frac{1}{2} \sqrt{\frac{\epsilon_0}{\mu_0}} I_0 \sigma$$

(2.19)

The use of cross-sections is practical because a realistic light source that has a finite beamwidth. A cross-section therefore prescribe a limit on the maximum power of scattering, absorption or extinction for any fixed input light intensity. From now onwards, we will consider cross-sections rather than total power loss. However, the current model remains very nontrivial to find general continuous solutions for simple geometries. In the next section, I will therefore outline the use of the dipole model, as a way of imposing discretisation to replace the integrals here with finite sums, and thereby allow explicit
solutions for scattering response to be found as matrix equations. More specifically, we will turn our attention to arrangements of simple nanoparticles in oligomer geometries.

2.2 Coupled dipole model

Here we consider the optical responses of nanoparticles placed in oligomer arrangements as to produce more complex optical features. The constituent nanoparticles of oligomers are typically both sufficiently simple and subwavelength in size, for their lowest-energy resonances to resemble dipoles at optical or near-infrared wavelengths. Given electric $p$ and magnetic $m$ dipole moments radiate identically to the $a_1$ and $b_1$ spherical scattering coefficients, see (1.14) and (1.21), we can again make use of the conclusion from Devaney and Wolf [51]. Specifically, the scattered field of a dipolar nanoparticle with only the moments $p$ and $m$ will be described by only $a_1$ and $b_1$ coefficients, hence the near fields external to the smallest bounding sphere around the nanoparticle itself can be exactly modelled by that of $p$ and $m$. We can therefore use a dipole model as a simplified alternative to the current model for nanoparticle oligomers with only minor limitations in accuracy, for even closely spaced nanoparticles\(^2\). In essence, the dipole model enables us to analyse the dominant optical properties of a collective nanoparticle oligomer by considering only the dominant optical resonances of the individual nanoparticles.

We now first consider the case of small plasmonic nanoparticles, where the individual nanoparticle response is dominantly an electric dipole, and we can therefore use the dipole approximation [60,62] to describe the optical properties of the oligomer. Moreover, analogous to (2.6) and (2.7) for currents, we can write the scattered electric and magnetic fields from a system of electric dipole moments $\{p_i\}$ in terms of dyadic Green’s functions.

\[
\mathbf{E}_a(\mathbf{r}) = \frac{k^2}{\epsilon_0} \sum_i \mathbf{\tilde{G}}_0(\mathbf{r}, \mathbf{r}_i) \cdot \mathbf{p}_i \tag{2.20}
\]

\[
\mathbf{H}_a(\mathbf{r}) = \frac{1}{i\omega\mu_0} \nabla \times \mathbf{E}_a(\mathbf{r}) = \frac{k\epsilon_0}{i} \sum_i \nabla \times \mathbf{\tilde{G}}_0(\mathbf{r}, \mathbf{r}_i) \cdot \mathbf{p}_i \tag{2.21}
\]

Here the Green’s function is the same as that defined for currents in (2.3), but we can also write the effect of $\mathbf{\tilde{G}}_0$ acting on arbitrary vector $\mathbf{u}$:

\[
\mathbf{\tilde{G}}_0(\mathbf{r}, \mathbf{r}') \cdot \mathbf{u} = \frac{e^{ikR}}{4\pi R} \left[ \left( 1 + i \frac{kR}{k^2R^2} \right) \mathbf{u} - \left( 1 + 3i \frac{kR}{k^2R^2} \right) (\hat{n} \cdot \mathbf{u}) \hat{n} \right] \tag{2.22}
\]

\[
\nabla \times \mathbf{\tilde{G}}_0(\mathbf{r}, \mathbf{r}') \cdot \mathbf{u} = ik \frac{e^{ikR}}{4\pi R} \left( 1 + i \frac{kR}{k^2R^2} \right) \hat{n} \times \mathbf{u} \tag{2.23}
\]

Here $\hat{n}$ is the unit vector pointing from $\mathbf{r}$ to $\mathbf{r}'$, so that: $\mathbf{r} - \mathbf{r}' = R\hat{n}$. The dipole moment of the $i^{th}$ point dipole (located at $\mathbf{r}_i$) can generally be related to the total electric field at $\mathbf{r}_i$, being the sum of $\mathbf{E}_0$ and $\mathbf{E}_a$, using a tensor electric dipole polarisability, i.e. $\mathbf{p}_i = \mathbf{\alpha}_e^{(0)} [\mathbf{E}_0(\mathbf{r}_i) + \mathbf{E}_a(\mathbf{r}_i)]$. By substituting $\mathbf{E}_a$ from (2.20), we then obtain an expression for each electric dipole moment $\mathbf{p}_i$ in an arbitrary dipole system as a function of the

\(^2\) An example of the validity of near-field predicted by the dipole model can be found in the Supplemental Material of [A.3].
externally applied electric field distribution $E_0$:

$$p_i = \bar{\alpha}_E^{(i)} c_0 E_0(r_i) + \bar{\alpha}_E^{(i)} k^2 \sum_{j \neq i} \bar{G}_0(r_i, r_j) \cdot p_j$$  \hspace{1cm} (2.24)$$

For a system of $N$ dipoles, the expression in (2.24) forms a matrix equation of rank $3N$, which we can then solved for any arbitrary excitation as per an ordinary matrix equation. Note that, unlike the current model in (2.8), the source dyadic can be neglected here given we are able to exclude a single point, rather than shaped volumes, to avoid the $R^{-3}$ singularity of $\bar{G}_0$ when $r_i = r_j$. In Figure 2.1a, we present the validity of this model for a symmetric trimer arrangement of gold nanospheres. For the spherical nanoparticles, the electric and magnetic dipole polarisabilities are scalars and defined in terms of the $a_1$ and $b_1$ scattering coefficients from Mie theory [54,55]:

$$\alpha_E = \frac{6i\pi a_1}{k^3}, \quad \alpha_H = \frac{6i\pi b_1}{k^3}$$  \hspace{1cm} (2.25)$$

Note that I use the convention for electric and magnetic dipole polarizabilities: $p = \alpha_E c_0 E$, $m = \alpha_H H$. The extinction spectra calculated from the dipole model are compared to those calculated using commercial electromagnetic simulation software, CST Microwave Studio, which uses a frequency domain solver that is based on the finite element method [63]. The details of finite element methods are beyond the scope of this thesis, but I will at least outline that they aim to solve a boundary value problem for the general class of wave equations where a differential operator $\hat{L}$ acts on an unknown vector field $\psi$, and is equal to some imposed driving vector field $f$, that is: $\hat{L} \psi = f$. One relevant manner by which such equations can be solved, is to minimise a corresponding functional defined in terms of an unknown $\psi$. Moreover, by defining a so-called test or trial function for $\psi$ as a linear combination of some known basis functions, the minimisation of the functional can be equated to a matrix equation for the coefficients of each basis function, and is solvable by matrix inversion. A minimised trial function will then approach the correct solution for $\psi$. Details of the above steps, which correspond to the Ritz Method, can be found in §2 of [63]. However, the distinguishing feature of the finite element method, is that it subdivides the full simulation volume into a volumetric mesh of smaller domains, and defines different trial functions in each voxel. This allows a simpler set with only a handful of basis functions to be used for each voxel’s trial function. Furthermore, it makes the calculation method more easily translatable to complex geometries, as one can simply reduce the size of each voxel until the fixed basis functions can correctly represent the minimised trial function. In this sense, CST recreates a given scattering geometry as a finite volumetric mesh, then superimposes an external electric and magnetic field distribution as the driving term $f$, much like $E_0$ in (2.8). It then performs its own version of a finite element method calculation, which has been able to provide quantitative agreement with experiment, such as in Fig. 6 of [A.3].

Even with very small gaps between the spheres, the dipole model offers an accurate prediction of the trimer’s response in Figure 2.1a, with the exception of that coming from the single particle electric quadrupole response. To next use the dipole model to consider high-index dielectric nanoparticles, we must also account for the potential magnetic dipole resonance in addition to the electric dipole resonance. To this end, the dipole model in (2.24) can then be extended to include both electric and magnetic dipoles. It is
straightforward to define magnetic dipoles induced by the total magnetic field with magnetic polarizabilities, such as denoted in (2.25), but we must also recognize that the total magnetic field will now include magnetic field radiated by electric dipoles (2.21), and also vice versa [65]. As such, we can write electric and magnetic fields radiated by a system of electric and magnetic dipoles:

\[ \mathbf{E}_s(\mathbf{r}) = \frac{k^2}{\varepsilon_0} \left( \sum_i \tilde{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}_i) \cdot \mathbf{p}^{(i)} - \frac{1}{ikc_0} \nabla \times \tilde{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}_i) \cdot \mathbf{m}^{(i)} \right) \]  
\[ \mathbf{H}_s(\mathbf{r}) = k^2 \left( \sum_i \tilde{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}_i) \cdot \mathbf{m}^{(i)} + \frac{c_0}{ik} \nabla \times \tilde{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}_i) \cdot \mathbf{p}^{(i)} \right) \]

This leads to the two equations in (2.28) for the electric and magnetic dipole moments induced by external electric and magnetic field distributions: (2.28a) from the total electric field and (2.28b) from the total magnetic field.

\[ \mathbf{p}^{(i)} = \tilde{\alpha}_E^{(i)} \mathbf{E}_0(\mathbf{r}_i) + \tilde{\alpha}_E^{(i)} k^2 \left( \sum_{j \neq i} \tilde{\mathbf{G}}_0(\mathbf{r}_i, \mathbf{r}_j) \cdot \mathbf{p}^{(j)} - \frac{1}{ikc_0} \nabla \times \tilde{\mathbf{G}}_0(\mathbf{r}_i, \mathbf{r}_j) \cdot \mathbf{m}^{(j)} \right) \]  
\[ \mathbf{m}^{(i)} = \tilde{\alpha}_H^{(i)} \mathbf{H}_0(\mathbf{r}_i) + \tilde{\alpha}_H^{(i)} k^2 \left( \sum_{j \neq i} \tilde{\mathbf{G}}_0(\mathbf{r}_i, \mathbf{r}_j) \cdot \mathbf{m}^{(j)} + \frac{c_0}{ik} \nabla \times \tilde{\mathbf{G}}_0(\mathbf{r}_i, \mathbf{r}_j) \cdot \mathbf{p}^{(j)} \right) \]  

Figure 2.1: A comparison of the extinction cross-section calculated using the dipole model in (2.28) with that calculated using CST Microwave Studio. Calculations are for 150 nm nanospheres made of (a) gold and (b) silicon, when arranged as symmetric trimers with varying separation \( g \) between nanoparticles. Figure taken from Chapter 10 of [64].
To summarize notation: $\mathbf{p}^{(i)} (\mathbf{m}^{(i)})$ is the electric (magnetic) dipole moment of the $i^{th}$ nanoparticle, $\mathbf{G}_0(\mathbf{r}_i, \mathbf{r}_j)$ is the free space dyadic Green’s function between the locations of the $i^{th}$ and $j^{th}$ dipoles, $\tilde{\alpha}_E^{(i)} (\tilde{\alpha}_H^{(i)})$ is the tensor electric (magnetic) dipole polarisability of the $i^{th}$ particle, $c_0 = 1/\sqrt{\epsilon_0 \mu_0}$ is the speed of light in the background medium and $k$ is the background wavenumber. As can be seen in Figure 2.1b, this coupled electric and magnetic dipole model is able to accurately predict the extinction of a trimer, with the exception of the single nanoparticle’s magnetic quadrupole response. Once again, the quantities we consider from the dipole model will be the cross-sections. We can again define the scattering cross-section from the integral of the far-field scattered power, now following the derivations by Merchiers et al. [66].

$$\sigma_s = \frac{k}{\epsilon_0^2 I_0} \text{Im} \left\{ \sum_i \epsilon_0 \mathbf{E}_0^*(\mathbf{r}_i) \cdot \mathbf{p}_i + \epsilon_0 \mu_0 \mathbf{H}_0^*(\mathbf{r}_i) \cdot \mathbf{m}_i \right\}$$

$$+ \mathbf{p}_i^* \cdot \left( \frac{i k^3}{6 \pi} + (\tilde{\alpha}_E^{(i)})^{-1} \right) \cdot \mathbf{p}_i + \epsilon_0 \mu_0 \mathbf{m}_i^* \cdot \left( \frac{2}{3} i k^3 + (\tilde{\alpha}_H^{(i)})^{-1} \right) \cdot \mathbf{m}_i \right\} \quad (2.29)$$

Here $I_0$ is the average intensity of a plane wave excitation to relate the area of a cross-section $\sigma$ to the total power $P$ as per (2.19). The absorption cross-section can next be calculated from the local losses of the internal electric and magnetic field [60].

$$\sigma_a = -\frac{k}{\epsilon_0^2 c_0} \text{Im} \left\{ \mathbf{p}_i^* \cdot \left( \frac{i k^3}{6 \pi} + (\tilde{\alpha}_E^{(i)})^{-1} \right) \cdot \mathbf{p}_i + \epsilon_0 \mu_0 \mathbf{m}_i^* \cdot \left( \frac{2}{3} i k^3 + (\tilde{\alpha}_H^{(i)})^{-1} \right) \cdot \mathbf{m}_i \right\} \quad (2.30)$$

The extinction cross-section is again written as the sum of absorption and scattering.

$$\sigma_e = \frac{k}{\epsilon_0 I_0} \text{Im} \left\{ \sum_i \mathbf{E}_0^*(\mathbf{r}_i) \cdot \mathbf{p}_i + \mu_0 \mathbf{H}_0^*(\mathbf{r}_i) \cdot \mathbf{m}_i \right\} \quad (2.31)$$

We now conclude with a pair of comparisons between the current and dipole models. Firstly, (2.24) is the discrete equivalent to current model in (2.8), and the extension to include magnetic dipoles in (2.28) is therefore analogous to accounting for magnetisation currents. This also means that (2.28) becomes invariant under duality transformations: $(\mathbf{E}, \mathbf{c}_0 \mathbf{B}) \rightarrow (\mathbf{c}_0 \mathbf{B}, -\mathbf{E})$ with $(\mathbf{p}, \mathbf{c}_0 \mathbf{m}) \rightarrow (\mathbf{c}_0 \mathbf{m}, -\mathbf{p})$, and any electric and magnetic dipoles satisfying (2.28a) necessarily also satisfy (2.28b), or vice versa, as discussed in [A.3]. Secondly, we can note that the scattering and absorption from each dipole in isolation is accounted for in the dipole model, (2.9) and (2.14), but can be neglected for a point in the current model, (2.11) and (2.14), given it is an infinitesimally small voxel of current. The influence of an individual source in the current model is only felt on the surrounding media, and so the current model explicitly describes a purely collective response. On the other hand, the dipole model still accounts for losses from individual dipoles, in recognition that they can represent a significant resonant object. In a nanoparticle oligomer, we therefore account for the isolated response of each nanoparticle, yet their coupling ensures that no nanoparticle can be considered independently, and so the overall optical response remains a collective property of the oligomer. Nanoparticle oligomers exist on the boundary between collective and isolated optical responses, and subsequently present a unique challenge to understand their collective optical properties. In the next Chapter, I present the use of eigenmodes to model the collective resonances of oligomers directly.
Quantifying resonances and their interference

This Chapter presents the approach developed during my studies for modelling resonances using the eigenmodes of nanoparticle systems, and the subsequent conclusion that interference between resonances, and particularly Fano resonances, can be described by the overlap of nonorthogonal eigenmodes. I also take some time to contrast the derivation of eigenmodes and Fano resonances between high-refractive-index dielectric nanoparticle oligomers and plasmonic nanoparticle oligomers. The aim is to combine the key results of the works [A.2,A.3,A.5] in Appendix A, which have explored and developed these particular areas for nanoparticle oligomers. This culminates in the demonstration of Fano interference between multiple magnetic dipoles when using dielectric nanoparticle oligomers in [A.3] .

3.1 Resonances in terms of eigenmodes

One of the key properties of nanoantennas for artificial materials is their capacity to support a resonant optical response. Understanding the characteristics of a resonance is therefore important for tailoring it toward the given application, be it electric or magnetic field localisation, directional scattering, or any number of other potential properties. In this regard, the most common way to characterise a resonance quantitatively is with a multipolar decomposition [54, 67]. This is obtained by projecting the scattered field onto vector spherical harmonics to obtain a spherical multipole decompositions, or by projecting the internal current distributions onto Cartesian multipoles, and potentially also their various correction factors, to obtain a Cartesian multipole decomposition [52, 68, 69]. Yet these multipolar decompositions ultimately remain a choice of basis for the scattering responses, and one where multipoles depend on the choice of origin; a multipole expansion is performed about an origin to describe the fields in the surrounding space. There may be an intuitive choice of origin for simple nanostructures, but it can be more ambiguous for complex nanostructures, or arrangements of nanoparticles. Additionally, there is the practical problem that any given multipole is not necessarily going to align with a given resonance of a considered nanoantenna, which can lead to a large number of multipoles being necessary to describe a single resonance. In this context, it was desirable to quantify resonances for a fixed object in a unique and origin-independent manner, particularly for later quantifying the interaction between resonances. We therefore began to consider the eigenmodes of the current model in the presence of a driving field. Moreover, (2.8) has an associated eigenmode equation, where an eigenmode $|v\rangle$ has a current distribution $j_v$ and
eigenvalue \( \lambda_v \) that satisfies:

\[
i \omega \lambda_v \mathbf{j}_v(\mathbf{r}) = -[\bar{\epsilon}(\mathbf{r}) - \epsilon_0]^{-1} \cdot \mathbf{j}_v(\mathbf{r}) + \frac{1}{\epsilon_0} \int \bar{\mathbf{G}}_0(\mathbf{r}, \mathbf{r}') \cdot \mathbf{j}_v(\mathbf{r}') \, d\mathbf{r}'^3
\]  

(3.1)

These eigenvalues represent scalar impedances (or susceptibilities \( \lambda^{-1} \)) and the eigenmodes represent the associated origin-independent basis of stable current distributions. These have a number of desirable properties:

- By nature of being eigenmodes, the set of eigenmodes also represents the only basis for the optical response where each basis vector represents a current distribution that is subject to energy conservation in isolation. In a formal sense, the real component of the eigenvalue must be greater than zero to be passive, or less than zero to be active, where active means inputting net energy into the system and passive refers to being not active. This follows from the sign of the extinction losses (2.18), when using eigenmodes as solutions to (2.8): \( \mathbf{E}_0 \rightarrow \lambda_v \mathbf{j}_v, \mathbf{J} \rightarrow \mathbf{j}_v \).

- The complex frequency where an eigenvalue becomes zero corresponds to a self-sustaining field distribution, which is a formal way to define a resonance [70]. In fact, given the eigenmodes form almost always\(^1\) a complete and linearly independent basis, every such self-sustaining resonance must be associated with at least one zero eigenvalue. Note that ’self-sustaining’ refers to a distribution of currents and fields whose magnitude does not decay in time, and requires no external input of energy.

That is: a solution to (2.8) with \( \mathbf{E}_0 = 0 \) and a nontrivial current distribution \( \mathbf{J} \neq 0 \).

An eigenmode decomposition therefore uniquely maps to the complete set of resonances at complex frequencies, while also providing a complete set of necessarily passive basis vectors (assuming the absence of gain media) at real frequencies, and further connecting these two physical attributes together in a consistent and origin-independent modal framework. However, it does introduce an issue in that the current model (2.8) contains loss and is thereby generally non-Hermitian, meaning its eigenmodes are not necessarily orthogonal. The specific excitation of each eigenmode in the current model can still be found through the impact of reciprocity, or time-reversal symmetry of (1.1)-(1.4), on the eigenmodes of any arbitrary system. This is discussed further in Chapters 4 and 5, but for now it suffices that Onsager’s arguments [71, 72] or the Fluctuation Dissipation Theorem [73], require that the dyadic Green’s function and permittivity tensor must be symmetric, although complex and not necessarily Hermitian.

\[
\bar{\mathbf{G}}_0(x, x') = \bar{\mathbf{G}}_0(x', x), \quad \bar{\mathbf{G}} = \bar{\mathbf{G}}_0^T, \quad \bar{\epsilon} = \bar{\epsilon}^T
\]  

(3.2)

The overall operator of the eigenvalue equation (3.1) then represents a complex symmetric matrix, and there are a number of ways to show that this makes any two nondegenerate eigenmodes \( \mathbf{j}_v, \mathbf{j}_w \) orthogonal under unconjugated complex projections, see Chapter 7 of [74]. For an example, one can write the matrix in Gantmacher’s normal form [75], which enforces such orthogonality between nondegenerate eigenmodes [76].

\[
\int \mathbf{j}_v(\mathbf{r}) \cdot \mathbf{j}_w(\mathbf{r}) \, d\mathbf{r}'^3 = 0, \quad \text{when } \lambda_v \neq \lambda_w .
\]  

(3.3)

\(^1\)See discussion on exceptional points following Figure 3.2.
The excitation $a_v$ of any eigenmode $\mathbf{j}_v$ can then be determined through unconjugated dot products between the eigenmode and the driving field distribution, analogous to the more familiar use of true complex projections for the excitation of orthogonal eigenmodes.

$$
\lambda_v a_v = \frac{\int \mathbf{j}_v(r) \cdot \mathbf{E}_0(r) \, dr^3}{\int \mathbf{j}_v(r) \cdot \mathbf{j}_v(r) \, dr^3} \quad (3.4)
$$

Therefore, despite the eigenmodes being nonorthogonal, any given eigenmode’s excitation is determined entirely by the given eigenmode’s current distribution and the driving field. The only exception to (3.4) is $\int \mathbf{j}_v \cdot \mathbf{J}_v = 0$, which can generally be disregarded as accidental, although in Chapter 5, and specifically (5.6), I discuss eigenmodes whose symmetry enforces $\int \mathbf{j}_v \cdot \mathbf{J}_v = 0$, and require a different calculation of $a_v$. However, turning now to our consideration of nanoparticle oligomers, we can consider analogous eigenmodes of systems made from purely electric dipoles. An eigenmode $|v\rangle$, now having electric dipoles $\mathbf{p}_v$, will satisfy (2.24) as:

$$
\mathbf{p}_v^{(i)} = \xi_E^{(i)} \epsilon_0 a_v \mathbf{p}_v^{(i)} + \xi_E^{(i)} \sum_{j \neq i} k^2 \mathbf{G}_0(r_i, r_j) \cdot \mathbf{p}_v^{(j)} \quad (3.5)
$$

However, when considering the model including magnetic dipoles (2.28), the eigenmodes need to be simultaneously constructed of both electric dipoles and magnetic dipoles, which have different units. To address this difference of units, my initial approach of [A.2] was to separate (2.28) and consider different eigenmode equations for electric and magnetic dipoles, with cross terms to describe driving of electric dipoles by an applied magnetic field, and the driving of magnetic dipoles by an applied electric field. In effect, this approach finds the eigenmodes of either the electric or the magnetic dipole systems, and their polarisabilities (eigenvalues), irrespective of the effect they have on the other dipole system. However, while this remains a full description of the dipole system, and it provides information on the resonances of electric and magnetic systems in the presence of each other, it does not describe the simultaneous stable oscillations of both the electric and magnetic dipoles. To consider the resonances of the collective system, we must consider both electric and magnetic dipoles together for single eigenmodes. In this regard, it is desirable to introduce relative scaling between the electric and magnetic dipoles, and between the electric and magnetic fields, to maintain fixed units of polarisability for the resulting eigenvalues. Moreover, the units can be standardised if the magnetic dipoles are scaled by a factor of $\epsilon_0^{-1}$, and the magnetic field by a factor of $\sqrt{\mu_0/\epsilon_0}$. This also makes the eigenmodes (not eigenvalues) independent of $\epsilon_0$ and $\mu_0$, as might be expected given polarisabilities are defined relative to an arbitrary background.

An eigenmode $|v\rangle$, having electric dipoles $\mathbf{p}_v$ and magnetic dipoles $\mathbf{m}_v$, will then satisfy (2.28) as:

$$
\lambda_v \mathbf{p}_v^{(i)} = (\xi_E^{(i)} \epsilon_0)^{-1} \mathbf{p}_v^{(i)} - \frac{k^2}{\epsilon_0} \left( \mathbf{G}_0(r_i, r_j) \cdot \mathbf{p}_v^{(j)} + \frac{1}{ik} \nabla \times \mathbf{G}_0(r_i, r_j) \cdot [\epsilon_0^{-1} \mathbf{m}_v^{(j)}] \right) \quad (3.6a)
$$

$$
\lambda_v [\epsilon_0^{-1} \mathbf{m}_v^{(i)}] = (\xi_H^{(i)} \epsilon_0)^{-1} [\epsilon_0^{-1} \mathbf{m}_v^{(i)}] - \frac{k^2}{\epsilon_0} \left( \mathbf{G}_0(r_i, r_j) \cdot [\epsilon_0^{-1} \mathbf{m}_v^{(j)}] - \frac{1}{ik} \nabla \times \mathbf{G}_0(r_i, r_j) \cdot \mathbf{p}_v^{(j)} \right) \quad (3.6b)
$$

This expression describes a matrix equation for eigenmodes of the electric and magnetic...
dipole system describing $N$ nanoparticles, however the associated $6N \times 6N$ matrix will, notably, not be symmetric when there is non-negligible coupling between the electric and magnetic dipoles. Therefore the corresponding eigenmodes will not maintain orthogonality analogous to that in (3.3) for currents.

I now want to conclude with some discussion related to [A.5], and consider the derivation of eigenmodes for nanoparticle oligomers using the eigenmodes of its different resonant subsystems. The aim of this particular work was to try and reconcile our eigenmode model with plasmonic hybridisation theory [77], which is an existing method to determine the collective optical response of metallic nanoparticle systems that support localised plasmon resonances. While the underlying model in plasmonic hybridisation theory treats individual metallic nanoparticles as electron gas density distributions and is generally quite different to the models presented in Chapter 2, it has an intuitive and conceptual goal. By dividing a given metal nanoparticle system into two or more resonant subsystems with sufficiently simple properties, the collective properties can be deduced from how these subsystems combine. The theory itself originally neglected retardation of interaction between charges, meaning it focused on smaller nanoparticle systems than I consider, but an amended version was later proposed to account for retardation [78], therefore justifying its use on systems including nanoparticle oligomers. In [A.5], I aimed to parallel the construction procedure of plasmonic hybridisation theory, but instead being relevant to our models for optical scattering in Chapter 2: deriving the collective eigenmodes of nanoparticle oligomers from the eigenmodes of their interacting subsystems. In doing this, we wanted to provide alternate and simplified commentary for the derivation of resonances without requiring the same complexity of plasmonic hybridisation theory. Moreover, the plasmonic hybridisation calculation is nontrivial for even simple systems, such as concentric spheres [79] and two-particle dimers [80], which has led it to becoming more regularly used as a conceptual tool to designate experimental and numerical observations of scattering that does not resemble that of the constituent nanoparticles in isolation. Our intention was to provide an alternate avenue to allow simplified derivations of collective resonance formation, but also to extend this approach to apply to high-index dielectric nanoparticles. In this regard, symmetric nanoparticle trimers were used as an example geometry, given these did not have a general solution in plasmonic hybridisation theory at the time. We also later used asymmetric nanoparticle dimers as a second example in §10.3.3 of [64], because this particular geometry was becoming of particular interest for high-index dielectric nanoparticles [81–84]. Our proposed method used the eigenmodes of each isolated subsystem as basis vectors for the collective optical response, and used the radiation from each of these basis vectors to quantify coupling channels between the basis vectors. This allowed us to re-express the eigenvalue problem in (3.6) with sets of a few coupled equations. General expressions for the collective eigenmodes and eigenvalues could then be found directly from these coupled equations, and were able to provide quantitative agreement to full-wave numerical simulations, and an experimental measurement of transmission through a dielectric nanoparticle trimer. This demonstrated that the eigenmode model presented here could replicate the dominant collective resonance formation of plasmonic hybridisation theory, while simultaneously offering dramatic simplifications with quantitatively reliable modelling. Given we are able encapsulate much of the effect of plasmonic hybridisation through a simple dipole model, I will turn our attention to the formation of specific resonant interference features known as Fano resonances in the coming section. Moreover, Fano resonances in plasmonic nanoparticle oligomers were regularly attributed to the presence of plasmonic hybridisation, largely because it enabled
non-radiative coupling between resonances [85–87]. The use of eigenmodes now offers an unambiguous way to quantify resonances, and is therefore a platform on which to quantify and understand interactions between resonances. In the coming section, I present my work toward understanding both Fano resonances and modal interference generally, and particularly the implementation of nanoparticle oligomers to realize such features.

### 3.2 Eigenmode interference and Fano resonances

One particular area that has garnered significant attention in recent years is the study of Fano resonances in nanoparticle oligomers and other cluster structures [45, 88]. For the context of nanoparticle scattering systems, Fano resonances have come to refer to a resonant interference in the total scattered power that is typically observed as a spectrally sharp, asymmetric lineshape in the extinction. The name itself owes to an asymmetric lineshape in the energy spectrum of atomic photoionisation explained by Fano [89] to be due to constructive and destructive two-channel interference between photoionisation from a broad ground state and from a discrete autoionised state of an atom. Yet analogous asymmetric spectra appear in a wide range of optical, atomic and mechanical systems that support at least two-channel interference [90]. To this background, Fano resonances in plasmonic nanoparticle scattering systems became predominantly described by the interference between a strongly scattering “bright resonance” and a weakly scattering “dark resonance” [86, 88]. By nature of being a poor radiator, a dark resonance is expected to be less damped, and thereby spectrally sharp, while a bright resonance is heavily damped by radiation losses and spectrally broad. Notably, this depicts a coupled oscillator model [91–93], where a harmonically driven (bright) oscillator \( m_1 \) is damped by coupling \( \gamma \) to an undriven (dark) oscillator \( m_2 \), such as the illustration in Figure 3.1a.

Here the feedback acting on \( m_1 \) due to a resonance in \( m_2 \) creates interference in the energy dissipation of the driving force provided by \( m_1 \) (the extinction). This interference can be either constructive or destructive, given the expected \( \pi \) change in relative phase acquired over the resonance of \( m_2 \), enabling a characteristic asymmetric Fano lineshape in extinction. However, it is less clear how to apply the coupled oscillator model if we can’t identify distinct resonant subsystems for \( m_1 \) and \( m_2 \), or if the choice becomes arbitrary or ambiguous. Furthermore, there is an implicit requirement that the coupling \( \gamma \) is considered as small for individual oscillators \( m_1 \) and \( m_2 \) to resemble the resonances that one can physically observe in the total channel system, Figure 3.1b. If the identified resonant subsystems for \( m_1 \) and \( m_2 \) are instead strongly coupled, then the physical resonances of the two-channel system will not resemble the isolated resonances of \( m_1 \) and \( m_2 \), Figure 3.1c. However, there is still a two-channel system that allows interference between resonances, hence a continued propensity for Fano resonances. The bright and dark mode depiction is simply no longer representative of the strongly coupled system, indeed both eigenmodes are bright, meaning they both couple directly to the applied force. Notably, a case where we can’t clearly identify distinct resonant subsystems for \( m_1 \) and \( m_2 \) is likely also a case where they are strongly coupled, which is generally the situation arising in my consideration of nanoparticle oligomers. At least when utilising coupled plasmonic nanoparticles, there are indeed a host of oligomers, and nanoparticle arrangements generally, from which Fano resonances can arise [94]. However, it was subsequently predicted that Fano resonances should also occur in symmetric oligomers made of silicon nanoparticles [95], where there is no plasmon hybridisation between nanoparticles, previously considered responsible for providing the coupling \( \gamma \) in Figure 3.1a. To this background, the formal treatment
Figure 3.1: (a) The coupled oscillator model for two-channel Fano resonances; only the mass $m_1$ receives a direct external driving force, but it is coupled by $\gamma$ to a second mass $m_2$. Qualitative illustration of two regimes where the interfering resonances will correspond to: (b) isolated resonances of $m_1$ and $m_2$, and (c) distributed resonances of the collective system.

proposed in [A.2] was developed to model Fano resonances directly from the resonances of the collective system without requiring separation into distinct resonant subsystems. This model identified a common theme underlying the physics of Fano resonances in both plasmonic and all-dielectric oligomers: the existence of Fano resonances can be attributed to the fact that the eigenmodes of these scattering systems are not orthogonal and, therefore: they can directly interfere with each other in extinction. As I will present herein, this is a general and rigorous conclusion applicable to all interference between resonances that impacts the extinction.

We begin by noting that both the current model (2.8) and the dipole model (2.28) describe open, radiative systems. As such, even in the absence of material loss, the system does still exhibit radiation losses and is generally, therefore, non-Hermitian. The immediate consequence of this non-Hermicity is that the eigenmodes we have defined in (3.1), (3.5) and (3.6) are not necessarily orthogonal. To recognise the effect of this nonorthogonality, and its relation to the Fano resonances, we can refer to the extinction cross-section by combining (2.18) and (2.19).

$$\sigma_{\text{ext}} = \frac{1}{I_0} \sqrt{\frac{\mu_0}{\epsilon_0}} \text{Re} \left[ \int E_0^* \cdot J \ dr^3 \right].$$

We now recognise that an arbitrary applied field and its induced currents can be defined in terms of a linear superposition of the eigenmodes.

$$E_0 = \sum_v a_v \lambda_v j_v \quad \Rightarrow \quad J = \sum_v a_v j_v.$$
We are then able to rewrite the total extinction (5.8) in terms of eigenmodes and eigenvalues. Moreover, we can divide the extinction into two contributions: direct terms that provide contributions to extinction from individual eigenmodes, and also interference terms coming from the overlap between different eigenmodes.

\[
\sigma_{\text{ext}} = \frac{1}{I_0} \sqrt{\frac{\mu_0}{\varepsilon_0}} \sum_v \left( \text{Re}[\lambda_v] \int |a_v|^2 |j_v|^2 \, dr^3 \right) + \sum_{w \neq v} \text{Re}\left[ a_v^* a_w \lambda_v \int j_v^* \cdot j_w \, dr^3 \right].
\]

(3.9)

We firstly recognise that the direct terms must always be greater than zero if we assume the system is passive; an eigenmode is an isolated solution to (2.8) and so must always produce positive extinction in the absence of gain media: it can’t generate power. Secondly, each given eigenmode’s excitation, being the \(a_v\) coefficients of (3.8), is independent from the excitations of other eigenmodes as shown in (3.4). Therefore, the only way any interaction between two or more eigenmodes can affect the extinction cross-section is through interference terms. The existence of nonzero interference terms, is therefore required for Fano resonances to exist in our model, meaning Fano resonances can only be described here by the nonorthogonality of eigenmodes. Given eigenmodes map uniquely to resonances, this also coincidentally shows that the largely accepted condition for one resonance being dark (orthogonal to the incident field) is not a requisite for Fano resonances. These are the conclusions of [A.2] for nanoparticle oligomers, and analogous conclusions were also reached in parallel by a separate work [96] using models for plasmons in metallic nanoparticles systems. The absence of dark resonances in Fano resonances was further in agreement with other recent works where Fano resonances were proposed and observed to occur between resonances that were driven directly by the incident field [87,97]. However, even beyond the conclusion that dark resonances are not necessary requisites for Fano resonances, our model required that Fano resonances be due to nonorthogonal eigenmodes, and we could explicitly define requisites for nonorthogonal eigenmodes, as presented in [A.6]. For eigenmodes to be nonorthogonal, we must require either: (i) non-negligible retardation in coupling, \(k \neq 0\) or \(kR \neq 0\), to prevent \(G_0\) becoming real and symmetric, hence Hermitian with orthogonal eigenmodes; or (ii) multiple materials following the argument of (31)-(35) in [A.6].

The eigenmode decomposition (3.9) is also not specific to plasmonic nanoparticle systems, it simply requires that we can calculate eigenmodes, which meant we could equally explore interference that appeared in high-index dielectric nanoparticle oligomers, as was done in [A.2, A.3, A.5]. To this extent, a comparison between plasmonic and dielectric Fano resonant oligomers, heptamers, is given in Figure 3.2. Here I show an eigenmode decomposition of extinction of each heptamer, in which I plot the direct terms of extinction (3.9) from the dominant eigenmodes, overlaid with the total extinction. The difference between total extinction and the sum of direct terms is then the sum of interference terms due to nonorthogonal eigenmode overlap. For the case of the gold nanoparticle heptamer in Figure 3.2a, we have a typical scenario for a Fano resonance: the overlap of a broad resonance and a sharp resonance, which leads to destructive interference. This gold heptamer is modelled after the investigation in [45], and shows a classical example of a two-channel Fano resonance, albeit with interference between the resonances being due to eigenmode nonorthogonality. However, as seen in Figure 3.2b, the situation becomes dramatically more complicated for a silicon nanoparticle heptamer. The number of eigenmodes of this system is much higher. More formally, the number of eigenmodes of the
gold heptamer that can be excited by a plane wave is limited by symmetry to three pairs of polarisation-degenerate eigenmodes following the argument of [A.2], equations (2)-(10). This number increases to six, due to extra magnetic dipoles in the silicon quadrumer when repeating the same argument and neglecting the electric-magnetic dipole coupling (2.23). It then increases beyond six with electric-magnetic dipole coupling, because such coupling allows dipoles to be oriented parallel to the propagation direction; see (2.23) regarding cross-coupling in (2.28). The first consequence of more eigenmodes seen in Figure 3.2b is many more signatures of interference occurring across the extinction spectra. The second consequence is that we have to deal with the formation of exceptional points [98–100]. An exceptional point refers to a point degeneracy of two or more eigenvalues when the

Figure 3.2: (Dashed lines) Extinction spectra of (a) gold and (b) silicon heptamers, simulated using the dipole model (2.28) and showing the role of eigenmode interference in producing Fano resonances. (Solid lines) Overlaid direct terms to extinction, as per (3.9), for all three excited eigenmodes of the gold heptamers, and the six most dominant eigenmodes for the silicon heptamers. The grey regions hide wavelength bands near exceptional points. Both gold and silicon nanospheres are 150 nm in diameter. Figure taken from Chapter 10 of [64].
corresponding eigenmodes also become linearly dependent. The linear dependence of two or more eigenmodes subsequently implies that the span of the complete set of eigenmodes has reduced, indicating that the eigenmodes cease to be a complete basis for the response space. One can instead recover the complete basis by defining generalised eigenvectors, see Chapter 6 of [101]. Ultimately, however, the eigenmodes are a bad basis in the vicinity of an exceptional point: there is a component of the response space that is becoming orthogonal to the eigenspace. In Figure 3.2b we observe that the excitation magnitude of coalescing eigenmodes can diverge in the vicinity of an exceptional point, given there is a component of the excitation field becoming orthogonal to the eigenmodes while the still in the span of the eigenspace (i.e. when not precisely at the exceptional point). Exceptional points can exist even in simple plasmonic and dielectric oligomer systems, as found when deriving eigenmodes in [A.5] and in §10.3.3 of [64] with complex frequencies, but appear more regularly when there are more interacting eigenmodes. I have therefore attempted to divert attention away from the direct extinction terms of individual coalescing eigenmodes in Figure 3.2b, given they diverge as the wavelength nears an exceptional point. This nonetheless illustrates an unexpected level of complexity of interactions that arises there are many intercoupled resonances, the example being Figure 3.2a vs. 3.2b.

Yet [A.3] instead presents a controlled use of four silicon nanoparticles in a symmetric quadrumer geometry for the purposes of interfering two collective magnetic resonances. Referring to the illustration in Figure 1b of [A.3], the global circulation of electric dipoles resembles a magnetic dipole analogous to a circulation of current (1.7), while the parallel aligned magnetic dipoles of each nanoparticle will also radiate like a magnetic dipole analogous to a volume integral of magnetisation \( \mathbf{m} = \int_{V} \mathbf{M}(\mathbf{r}) dV \). If we were to treat this quadrumer as a point source, the net magnetisation of this point would have contributions from the circulating electric dipoles resembling circulating current (1.8), but also a second channel from aligned internal magnetic dipoles, which would more resemble a fictitious oscillating magnetic charge. While this is ultimately a consequence of incorrectly perceiving the quadrumer as a point source, there are tangible effects of the presence of these two channels for the quadrumer’s collective magnetic response: the two channels are coupled through internal electric-magnetic dipole coupling, and thereby enable two-channel interference and Fano resonances. This is precisely what is realized in [A.3]. A symmetric silicon quadrumer, and its microwave analogue, were both demonstrated to support magnetic-magnetic Fano interference when perceiving the response of the quadrumer as a single collective object. This magnetic Fano resonance was then later used to enhance the local internal magnetic dipole moments of silicon quadrumer arrays for the purpose of enhancing their third harmonic generation [102], using interference to build on previous demonstrations of third harmonic generation due to magnetic resonances of individual silicon disks [23]. As such, this demonstrates how considered use of high-index dielectric nanoparticles in oligomers can enable both unconventional optical properties and functional outcomes.
Quantifying resonances and their interference
Chapter 4

Symmetry effects and eigenmodes

Symmetry is one of very few tools where simple analytical arguments can provide quantifiable constraints on the otherwise nontrivial dependency shared between the geometry of a nanoparticle system and its collective optical response. Section 4.1 will provide a short introduction to the decomposition of optical scattering responses based on their properties under geometric symmetry transformations, and then 4.2 discusses the consequences of time-reversal symmetry. These ideas will then be combined in Section 4.3 to present analysis on the eigenmodes of geometries that have at least 3-fold discrete rotational symmetry, and the subsequent manifestation of polarisation-independent scattering and absorption.

4.1 Geometric symmetry

In this section, I provide a short summary on the use of group theory to quantify geometric symmetry in the models from Chapter 2, and also the eigenmodes in Chapter 3. The content will largely follow from more comprehensive works on the application of group theory to condensed matter systems [103, 104], and particularly [105], though presented here as relevant to specifically nanoparticle scattering systems. In this regard, I first restrict our consideration to specifically symmetry point groups, which consider geometric transformations acting on coordinates (points) in a given space. A symmetry point group $G$ refers to a particular set of transformation operations that form a group because each operation $\hat{R}$ in $G$ also has an inverse operation $\hat{R}^{-1}$ in $G$. The relevant symmetry for the models in Chapter 2 is going to be the invariance under a given transformation $\hat{R}$ of either: a permittivity distribution $\bar{\epsilon}(r)$, or an arrangement of discrete dipole polarisabilities $\bar{\alpha}_E, \bar{\alpha}_H$ located at points in space $r$. The application of a symmetry operation $\hat{R}$ on the corresponding geometric space can then be described by transforming each $r \rightarrow \hat{R}r$. However, to quantify symmetry operations on vector fields (e.g. $\mathbf{E}$, $\mathbf{B}$, $\mathbf{J}$, etc.), it is often intuitive to consider the equivalent transformation of the coordinate system from the inverse symmetry operation $\hat{R}^{-1}$, which is also necessarily in $G$. When $\hat{R}^{-1}$ acts on the coordinate system of a vector field $\mathbf{u}(r)$, which is distributed over our geometric space, the vector field will be transformed by $\hat{R}$ as $\mathbf{u}(r) \rightarrow \hat{R}\mathbf{u}(\hat{R}r)$. I should now emphasize that $\hat{R}$ here denotes an operation, while the explicit transformation will be enacted by some matrix $\bar{\mathbf{D}}^{(\hat{R})}$. The form of $\bar{\mathbf{D}}^{(\hat{R})}$ will depend on the properties of the space on which $\hat{R}$ acts, that is: the specific quantity a given $\mathbf{u}$ represents. As an example, parallel components of electric and magnetic fields have opposite parity under a reflection of the coordinate system $\hat{R} = \hat{\sigma}$. This follows because magnetic field is defined from the curl of electric field in (1.2); the magnetic field is related to local rotation of electric field at the given location, discussed in Section 1.2. As such, if $\hat{\sigma}(\mathbf{E} \cdot \hat{\mathbf{x}})\hat{x} = (\mathbf{E} \cdot \hat{x})\hat{x}$, for some direction $\hat{x}$,
then \( \hat{\sigma}(\mathbf{B} \cdot \hat{x}) \hat{x} = - (\mathbf{B} \cdot \hat{x}) \hat{x} \).

Returning now to our given symmetry group \( G \), but now also presuming to have a quantity \( u \), the set of symmetry operations \( \hat{R} \) in \( G \) will be described for \( u \) by some set of transformation matrices \( \overline{D}^{(\hat{R})} \). This set of matrices is called a *representation* of \( G \). Yet, much like our ability to consider \( E \) and \( H \) separately owing to their different transformation properties under reflection (*e.g.* rather than considering a concatenated 6-vector), we can choose to decompose our desired vector fields according to the transformation properties under symmetry operations. This is where the limiting concept of *irreducible representations* becomes useful. A given representation is *reducible* if there is a single unitary basis transformation that can express all matrices \( \overline{D}^{(\hat{R})} \) from the representation in the same diagonal matrix block form, as it indicates each block is a smaller dimensional representation of a subspace of the given \( u \). For example, if there is a single unitary matrix \( \hat{U} \) that allows a representation of matrices \( \overline{D}^{(\hat{R})} \) to be expressed in a block diagonal form seen below for all \( \hat{R} \) in \( G \), then the original representation can be reduced into the two representations having matrices \( \overline{D}^{(\hat{R})} \) and \( \overline{D}^{(\hat{R}')} \).

\[
\hat{U}^\dagger \cdot \overline{D}^{(\hat{R})} \cdot \hat{U} = \begin{pmatrix} \overline{D}^{(\hat{R})} & \overline{D}^{(\hat{R}')} \end{pmatrix}
\]

An irreducible representation \( \Gamma \) is a representation for \( G \), consisting a set of matrices \( \overline{D}^{(\hat{R})}_\Gamma \) for all \( \hat{R} \) in \( G \), which cannot be rewritten in a diagonal block form of lower dimensional representations. Conversely, this means that any symmetry operation can be decomposed into a matrix that is equivalent to a block diagonal form, where each block is a matrix in the irreducible representations of the corresponding operation. The specific \( \overline{D}^{(\hat{R})}_\Gamma \) matrices of irreducible representations are described by character tables, which list the character (trace) of each matrix \( \overline{D}^{(\hat{R})}_\Gamma \) in the given symmetry group. In Table 4.1, I write down a set of character tables for a few symmetries that will be used in this and the next section, and have corresponding example geometries illustrated in Figure 5.1a.

For the remainder of this Chapter, we will require only that the given \( \overline{D}^{(\hat{R})}_\Gamma \) exists, or consider exclusively one-dimensional irreducible representations, where each \( \overline{D}^{(\hat{R})}_\Gamma \) is just a scalar \( D^{(\hat{R})}_\Gamma \), and equal to its character. The main use of irreducible representations in this Chapter will be to separate a vector field or distribution \( u \) into a finite set of orthogonal distributions \( u_\Gamma \) that each transform according to a single irreducible representation \( \Gamma \). Here each \( \Gamma \) is an irreducible representation for a symmetry group \( G \), which consists only symmetry operations the given geometry is invariant under. In essence, we will show that one can always write a given \( u \) as a linear combination of some set of \( u_\Gamma \):

\[
u(r) = \sum_\Gamma a_\Gamma u_\Gamma(r)
\]

Here \( a_\Gamma \) are complex coefficients. The ability to write this decomposition isn’t immediately obvious, so we can start from the definition that the irreducible representations describe all possible transformations under the symmetry operations of \( G \), following from their definition as a basis for representations for the group \( G \) [105]. This is equivalent to saying that there is no portion of \( u \) that doesn’t lie in the span of some given \( u_\Gamma \), because it would indicate a component of \( u \) doesn’t transform under symmetry operations according to any
### §4.1 Geometric symmetry

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Table 4.1: Character tables for the $C_4$, $S_4$, $C_{4v}$ and $C_{4h}$ symmetry groups (see also the example illustrations in Figure 5.1a). Rows denote irreducible representations, columns denote symmetry operations: $\hat{E}$ is the identity, $\hat{i}$ is a point inversion, $\hat{C}_n$ and $\hat{S}_n$ are rotation and improper rotation by $\frac{2\pi}{n}$, $\hat{\sigma}$ is a reflection plane parallel ($\hat{\sigma}_v$, $\hat{\sigma}_d$) or perpendicular ($\hat{\sigma}_h$) to the principle rotation axis. Table indices are the trace of the matrix representation for the corresponding symmetry operation.

irreducible representation in $G$. If it can now be shown that $u_{\Gamma_i}$ and $u_{\Gamma_j}$ are orthogonal when $\Gamma_i \neq \Gamma_j$, we infer linear independence of each $u_{\Gamma}$ and therefore the ability to uniquely decompose $u$ into $u_{\Gamma}$ as per (4.2). This is precisely what is shown in the following box.
Orthogonality of basis vectors from different irreducible representations.

When applying symmetry operations $\hat{R}^{-1}$ to the coordinate system, the inner product between $u_{j_1}$ and $u_{j_2}$ will be conserved because symmetry operations $\hat{R}$ are necessarily unitary (e.g. to conserve geometry).

$$\int [\bar{D}_{j_1}(\hat{R}) \cdot u_{j_1}(r)]^* \cdot [\bar{D}_{j_2}(\hat{R}) \cdot u_{j_2}(r)]dr^3 = \int [u_{j_1}(r)]^* \cdot u_{j_2}(r)dr^3$$  \hspace{1cm} (4.3)

Here we have also used the fact that $\bar{x}$ and $\bar{R} \bar{x}$ are equivalent integration variables because $\hat{R}$ preserves the geometric integration volume, allowing us to describe the effect of any $\hat{R}$ entirely by the matrix representations $\bar{D}_{j_1}(\hat{R})$ and $\bar{D}_{j_2}(\hat{R})$ acting on $u_{j_1}$ and $u_{j_2}$, respectively.

We now utilise the Wonderful Orthogonality Theorem \[105\] and unitary $\hat{R}$.

$$\sum_R [\bar{D}_{j_1}(\hat{R})]_{ab} [\bar{D}_{j_2}(\hat{R})]_{a'b'} = \frac{N_G}{L} \delta_{\Gamma_1,\Gamma_2} \delta_{a,a'} \delta_{b,b'} \Rightarrow \sum_R [\bar{D}_{j_1}(\hat{R})]^* [\bar{D}_{j_2}(\hat{R})] = 0$$  \hspace{1cm} (4.4)

Here $a, a', b, b'$ denote matrix indices, $N_G$ is the number of $\hat{R}$ in the group $G$, $\delta$ denotes a Kronecker delta function, and $L$ is the dimension of both $\Gamma_1$ and $\Gamma_2$, where we can neglect $L_{\Gamma_1} \neq L_{\Gamma_2}$ due to the $\delta_{\Gamma_1,\Gamma_2}$ term. The right hand side of (4.4) also assumes $\Gamma_1 \neq \Gamma_2$. In any case, we can then consider a sum of $N_G$ identical inner products of $u_{j_1}$ and $u_{j_2}$.

$$N_G \int u_{j_1}^* \cdot u_{j_2} dr^3 = \sum_R \int [\bar{D}_{j_1}(\hat{R})]^* [\bar{D}_{j_2}(\hat{R})] u_{j_1} u_{j_2} dr^3, \text{ using (4.3)}$$

$$= 0, \text{ using (4.4)}$$  \hspace{1cm} (4.5)

Given there is always at least an identity operation in any $G$, we know $N_G \geq 1$, and therefore (4.5) shows $u_{j_1}$ and $u_{j_2}$ must be orthogonal if $\Gamma_1 \neq \Gamma_2$.

Let us now consider the more explicit case of some scattering object whose geometry is invariant under operations of some symmetry group $G$, which has a set of irreducible representations $\{\Gamma\}$. Using (4.2), we can separate any external field distribution $E_0$ into the individual components $\varepsilon_\Gamma$ that transform according to the corresponding $\Gamma$.

$$E_0(r) = \sum_\Gamma a_\Gamma \varepsilon_\Gamma(r)$$  \hspace{1cm} (4.6)

The current distribution $J_\Gamma$, induced by $\varepsilon_\Gamma$, can now be written as a linear combination of eigenmode current distributions $j_v$, following (3.1).

$$\varepsilon_\Gamma = \sum_v b_{\Gamma,v} \lambda_v j_v \quad \Rightarrow \quad J_\Gamma = \sum_v b_{\Gamma,v} j_v.$$

Importantly, we can show that each eigenmode $j_v$ of the geometry will, once excluding accidental degeneracies, transform according to only a single irreducible representation $\Gamma$. If this were not true, a single $j_v$ could be written as a linear combination of $u$ that each transform according to different irreducible representations $\Gamma$ using (4.2). Because each $\Gamma$ has a unique set of transformation properties, the number of linearly independent $u_\Gamma$ in the given $j_v$ specifies the number of $\hat{R}$ that can act on $j_v$ to create a new, linearly independent eigenmode $j'_v$, which must be degenerate with $j_v$ (having the same eigenvalue $\lambda_v$) given $\hat{R}$ is
equivalent to a coordinate transformation. In other words, the number of \( u_\Gamma \) is equal to the degeneracy order of \( j_v \). We can subsequently construct a set of \( j_v \) and all \( j'_v \) whose size is equal to, and thereby spans, the set of \( u_\Gamma \). This means, conversely, that we can write each \( u_\Gamma \) as a linear combination of the set of \( j_v \) and \( j'_v \), which means each \( u_\Gamma \) is an eigenmode with eigenvalue \( \lambda_v \). Hence we have reached our original statement that each eigenmode transforms according to a single \( \Gamma \), once excluding accidental degeneracies. We can also conclude that \( j_\Gamma \) in (4.7) transforms according to \( \Gamma \), given it is a linear combination of \( j_v \). These results will be used in the derivations of the next section to constrain the scattering and absorption properties of rotationally symmetric nanostructures.

### 4.2 Reciprocity and time-reversal symmetry

In this section, I want to explore a specific conclusion necessary for arguments I consider in the remainder of this thesis: any applied electric field distribution \( E_0 \) experiences exactly the same rate of energy loss (extinction) as the distribution \( E_0^* \). When \( E_0 \) is a plane wave with wavevector \( k \), \( E_0^* \) is a co-polarised plane wave with wavevector \( -k \), and this invariance of extinction from plane waves propagating in opposite directions is one basic conclusion attributed to reciprocity in optics. So I begin with reciprocity, the most common definition of which for optics is an equivalence of some measurement when interchanging source and detector. While clearly dependent on the chosen definition of the detector’s measurement, there are a number relevant theorems to this extent in optics, of which Potton [106] provides an extensive review. The most commonly utilised version of reciprocity, and relevant to optical far-field scattering, can be paraphrased in a manner analogous to that presented in §4 of [106] when summarising such reciprocal theorems.

- The amplitude of a scattered plane wave propagating along direction \( \hat{s}_\beta \) with polarization state \( \hat{E}_\beta \), due to an incident plane wave propagating along \( \hat{s}_\alpha \) with polarization \( \hat{E}_\alpha \).

**Equal to:**

- The amplitude a scattered plane wave propagating along \( -\hat{s}_\alpha \) with polarization \( \hat{E}_\alpha \), due to incident plane wave propagating along \( -\hat{s}_\beta \) with polarization \( \hat{E}_\beta \).

However, the models for scattering used in this thesis align instead with volumetric field distributions, so I instead focus entirely on the Lorentz Reciprocity Theorem seen in (4.8), a derivation of which can be found in §89 of [107].

\[
\int_V E_\alpha \cdot J_\beta - E_\beta \cdot J_\alpha \, dV = \int_{\Omega_V} E_\alpha \times H_\beta - E_\beta \times H_\alpha \, d\Omega \tag{4.8}
\]

This defines a relationship between any two solutions of Maxwell’s Equations denoted by subscript \( \alpha, \beta \): the fields \( E_\alpha, H_\alpha \) and \( E_\beta, H_\beta \), with corresponding current distributions \( J_\alpha \) and \( J_\beta \). For compact source currents with finite energy, the surface integral on the right hand side of this equation will go to zero whenever we can assume one of two conditions. In the case of a background with any dissipative loss \( \text{Im}\{\epsilon_0,\mu_0\} < 0 \), one can simply take the volume \( V \) as infinite, such that any fields at the boundary \( \Omega_V \) have propagated infinitely far from their finite energy sources, and hence the fields are zero. Alternatively, we can assume a homogeneous background exists at an infinite distance from our compact sources with finite energy, which means the only fields on the boundary of an infinite spherical volume \( V \) will be both plane waves, and propagating radially outwards (along \( \hat{r} \)). As such, on the boundary \( \Omega_V \), we can substitute \( H \) as per a plane wave \( E = \sqrt{\mu_0 \epsilon_0} \hat{r} \times H \), when using
Symmetry effects and eigenmodes

the scalar triple product relation \( \mathbf{a} \cdot (\mathbf{b} \times \mathbf{c}) = \mathbf{b} \cdot (\mathbf{a} \times \mathbf{c}) \). This then equates the surface integral to zero:

\[
\int_{\Omega} \mathbf{E}_\alpha \times \mathbf{H}_\beta - \mathbf{E}_\beta \times \mathbf{H}_\alpha \, d\Omega = \int_{\Omega} \hat{\mathbf{r}} \cdot (\mathbf{E}_\alpha \times \mathbf{H}_\beta - \mathbf{E}_\beta \times \mathbf{H}_\alpha) \, d\Omega \\
= \int_{\Omega} \sqrt{\varepsilon_0 \mu_0} (\mathbf{E}_\alpha \cdot \mathbf{E}_\beta - \mathbf{E}_\beta \cdot \mathbf{E}_\alpha) \, d\Omega = 0 \quad (4.9)
\]

The two assumptions we just considered will generally account for all radiative systems of interest, hence we can reliably use a simpler reciprocity equation without the surface integral:

\[
\int_V \mathbf{E}_\alpha \cdot \mathbf{J}_\beta - \mathbf{E}_\beta \cdot \mathbf{J}_\alpha \, dV = 0 \quad (4.10)
\]

To reach the desired conclusion, being the equal extinction experienced by applied fields \( \mathbf{E}_0 \) and \( \mathbf{E}_0^* \), let us define \( \mathbf{J}_\alpha \) as the currents induced by some \( \mathbf{E}_0 \), and \( \mathbf{J}_\beta \) as the currents induced by \( \mathbf{E}_0^* \). We can then consider four solutions to Maxwell’s Equations grouped into two sets:

(i) \( \mathbf{E}_0 \) and \( \mathbf{E}_0^* \) are imposed separately, and respectively induce currents \( \mathbf{J}_\alpha \), \( \mathbf{J}_\beta \), with total fields \( \mathbf{E}_\alpha \), \( \mathbf{E}_\beta \).

(ii) The two currents \( \mathbf{J}_\alpha \) and \( \mathbf{J}_\beta \) are imposed separately in the background medium of (i), respectively producing scattered fields \( \mathbf{E}_\alpha - \mathbf{E}_0 \) and \( \mathbf{E}_\beta - \mathbf{E}_0^* \).

Both (i) and (ii) share the same current distributions \( \mathbf{J}_\alpha \) and \( \mathbf{J}_\beta \), and we can therefore take the difference of (4.10) due to (i) and (ii), which gives the result we are interested in:

\[
\int_V \mathbf{E}_0 \cdot \mathbf{J}_\beta \, dV = \int_V \mathbf{E}_0^* \cdot \mathbf{J}_\alpha \, dV \quad (4.11)
\]

By referring to the definition of extinction in (5.8), and remembering that \( \mathbf{J}_\alpha \) is induced by \( \mathbf{E}_0 \) and \( \mathbf{J}_\beta \) is induced by \( \mathbf{E}_0^* \), the conclusion in (4.11) states that \( \mathbf{E}_0 \) and \( \mathbf{E}_0^* \) experience precisely the same power loss, as desired. However, before concluding that reciprocity is therefore responsible for this conclusion, we must recognise that the derivation of the Lorentz Reciprocity Theorem (4.8) itself assumes permittivity \( \hat{\varepsilon} \) and permeability \( \hat{\mu} \) are symmetric matrices. The symmetry of these matrices is generally considered to be a consequence of Onsager’s Reciprocity \([71, 72]\), which in turn was derived from the existence of microscopic reversibility: the equal probability of any two time-separated random fluctuation events occurring in the opposite order for a system in thermal equilibrium. More specifically, Onsager showed that, when given a system in thermal equilibrium, a matrix \( \mathcal{M} \) that relates the time evolution of any set of time-averaged displacement quantities \( q \) to the rate of change with \( q \) of the maximum system entropy \( S \) at thermal equilibrium, will be symmetric. That is: \( \frac{du}{dt} = \sum_{ij} M_{ij} \frac{\partial S}{\partial q_j} \) and \( M_{ij} = M_{ji} \). The relevant application to electric displacement \( \mathbf{D} \) seems to arise by recognising \( \partial S/\partial \mathbf{D}_j \) acts as a force on \( \mathbf{D} \) and is thereby proportional to the local electric field, hence the reciprocal relation enforces symmetry on permittivity defined through \( \mathbf{D} = \hat{\varepsilon} \mathbf{E} \). However, the later application of the Fluctuation Dissipation Theorem \([73]\) to generalised susceptibilities, being frequency domain tensors that relate physical quantities to the generalized forces they experience, gets the same matrix symmetry requirements more directly, as discussed in §125 of \([61]\), or indeed in §96 of \([107]\) for specifically permittivity and permeability.
§4.2 Reciprocity and time-reversal symmetry

I will now define time-reversal symmetry in electromagnetism, and provide a generic argument as to why time-reversal symmetry implies microscopic reversibility. The reversal of time transformation \( t \to -t \) will leave Maxwell’s Equations in (1.1)-(1.2) unchanged, provided the current and magnetic field are time-antisymmetric, i.e. \( E(t) \to E(-t), \ B(t) \to -B(-t), \ \rho(t) \to \rho(-t), \ J(t) \to -J(-t) \). From a physical perspective, \( J \to -J \) is needed to preserve the continuity of charge equation \( \nabla \cdot J = -\frac{\partial \rho}{\partial t} \), and \( B \to -B \) is needed to preserve the Lorentz force (1.5). From the ability to reverse time without changing Maxwell’s Equations, we have an implicit freedom in any electromagnetic system to define the causal progression of time as either \( t \) increasing or decreasing. When now presented with a steady state solution to Maxwell’s Equations that is distributed over all time, this solution must simultaneously be valid for both directions of causal time. As such, the evolution of this system from its state at time 0 to \( t_0 \) is identical to and that describing evolution from the same state at time 0 to \( -t_0 \). The conditions that specify a probability for any random fluctuation of fields or currents to occur in this steady state system at time 0 must therefore be precisely the same as at time \( -t_0 \). Given that the definition of time 0 is actually arbitrary in a steady state solution, our previous argument starting from a time origin of \( t_0 \) applies to the probability of random fluctuations at time 0 and time \( 2t_0 \). This is the microscopic reversibility required by Onsager’s arguments, and it therefore suggests that time-reversal symmetry is necessary for Lorentz reciprocity, and our subsequent conclusion that the applied field distributions \( E_0 \) and \( E_0^* \) experience equal extinction. To go even further, in the box I show that the equal extinction of \( E_0 \) and \( E_0^* \) can even follow directly from time-reversal, without introducing permittivity or permeability, let alone their symmetry. This is significant given the earlier review by Potton [106], because it was able to conclude that reciprocity is distinct from time-reversal symmetry. I will continue to refer to the equal extinction of \( E_0 \) and \( E_0^* \) as being due to reciprocity, not time-reversal symmetry, in recognition that I needed to assume linearity and include the Lorentz force experienced by charges, which is not fundamentally different to the assumptions used in the derivation of the Lorentz Reciprocity Theorem.

---

1 Given the magnetic field can be attributed to relative velocity of different inertial reference frames, see §13 [108], and that this velocity changes sign if \( t \to -t \), so the time-antisymmetry of \( B \) should be expected.
**Symmetry effects and eigenmodes**

The same argument shows that system A, by its definition as the fixed field distribution $q$, experiences the same extinction in the background medium (as the origin of currents). In other words, the external field $E_0$ will induce $-J_0^{*}$.

The reflection (2.18) of $E_0^{*}$ from $J_0$ is the negative to that of $E_0$ from $J$: the energy lost per unit time changes sign, as largely expected given we reversed causal time. Conversely, the causal extinction defined as energy lost per unit causal time remains unchanged. The statement (i) then follows.

### Derivation of (ii)

The current model (2.8) describes the coupling between two solutions to Maxwell's Equations: system A as $E_0$ in the background medium, and system B as a static distribution of charge $q$ in the background medium (as the origin of currents). System A, by its definition as the fixed field distribution $E_0 e^{-i\omega t}$, does not depend on the choice of causal time. The optical properties of system B are governed by the forces $F_q$ experienced by the charges $q$, which also don’t depend on the direction of causal time: using the definition of time-reversal, $E(t) \rightarrow E(-t)$, $B(t) \rightarrow -B(-t)$, $\rho(t) \rightarrow \rho(-t)$, $J(t) \rightarrow -J(-t)$, it follows that $F_q(t) \rightarrow F_q(-t)$ in (1.5). We now recognise that the extinction of $E_0$, as in (2.18), is the time-averaged rate of energy transfer from system A into system B. As neither system A nor system B depend on the direction of causal time in isolation, a difference in energy transferred from system A to system B, which depends only on the choice of causal time, implies a preferred direction of overall time evolution. We know such a preference does not exist from our capacity to perform an inconsequential time-reversal transformation on Maxwell's Equations. The statement (ii) then follows.
4.3 Discrete rotational symmetry

Here I use the eigenmode model of Chapter 3 to derive the result presented in [A.1]: $C_n$ ($n \geq 3$) discrete rotational symmetry of an object forces the total power it scatters (2.11) and absorbs (2.15) to be independent from the angle of linear polarisation for plane waves propagating parallel to the $n$-fold rotation axis. This result is interesting because both the far-field scattering pattern and the near-field distribution remain highly dependent on the incident plane wave’s polarisation angle, even while the total scattering and total absorption remain independent. The reason to return to this result is to retrospectively understand what symmetry has actually done to the individual resonances causing this effect. Of the specific relevance is equations (6) to (12) of [A.6], where it was derived that, because the permittivity $\tilde{\epsilon}$ is a symmetric matrix required by Onsager’s reciprocity, degeneracy is enforced between pairs of eigenmodes belonging to irreducible representations that are complex conjugates of each other. This result then affects geometries whose symmetry is invariant under the operations of the $C_n$ ($n \geq 3$) group, such as the $E$ irreducible representation for $C_3$ in Table 4.2, for which rotations are equated to phase shifts.\(^2\)

We will show here that this same reciprocity induced degeneracy derived in [A.6] implies the polarisation-independent scattering and absorption cross-sections derived in [A.1].

<table>
<thead>
<tr>
<th></th>
<th>$\hat{E}$</th>
<th>$\hat{C}_3$</th>
<th>$(\hat{C}_3)^2$</th>
<th>examples for $[x, y, z]$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A$</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>$z$</td>
</tr>
<tr>
<td>$E$</td>
<td>1</td>
<td>$\phi$</td>
<td>$\phi^*$</td>
<td>$x + iy$</td>
</tr>
<tr>
<td></td>
<td>1</td>
<td>$\phi^*$</td>
<td>$\phi$</td>
<td>$x - iy$</td>
</tr>
</tbody>
</table>

Table 4.2: Character table for the $C_3$ symmetry group. Rows denote irreducible representations, columns denote symmetry operations ($\hat{E}$ is the identity, $\hat{C}_3$ is a rotation by $\frac{2\pi}{3}$), indices are the character of the corresponding matrix representation for the symmetry operation ($\phi = e^{\frac{2\pi i}{3}}$).

To begin, we reiterate that the $E_1$ irreducible representation of $C_n$ can be separated into two, one-dimensional irreducible representations that are complex conjugates of each other, which we will denote as $E_{+1}$ and $E_{-1}$. Using the aforementioned result from [A.6] that the eigenmodes of $E_1$ will come in degenerate pairs between $E_{+1}$ and $E_{-1}$ due to reciprocity, we can treat $E_1$ as a single two-dimensional irreducible representation. Moreover, we consider any excitation field $\mathbf{E}_\Gamma$ that transforms according to a single $\Gamma = E_1$ (4.7), and write out its corresponding eigenmode decomposition.

$$\mathbf{E}_\Gamma = \sum_v b_v \lambda_v \mathbf{j}_{\Gamma,v} \quad \Rightarrow \quad \hat{C}_n \mathbf{E}_\Gamma = \sum_v b_v \lambda_v [\hat{C}_n \mathbf{j}_{\Gamma,v}]$$

Here $\hat{C}_n$ denotes the symmetry operation in $C_n$ for rotation by $\frac{2\pi}{n}$. The symmetry rotated eigenmode $\hat{C}_n \mathbf{j}_{\Gamma,v}$ is also generally able to be linearly independent to $\mathbf{j}_{\Gamma,v}$, given $\mathbf{j}_{\Gamma,v}$ can contain both $E_{+1}$ and $E_{-1}$ transformation properties. A plane wave propagating parallel

\(^2\)Note that the degeneracy of left- and right-circulating eigenmodes, belonging to the $E_1$ irreducible representation of $C_n$ ($n \geq 3$), has some similarities with a preliminary example considered by Onsager in (4.1)-(4.11) of [71]. There, for hexagonal and tetragonal crystals with $C_3$ and $C_4$ symmetries, Onsager derived the enforced equivalence of clockwise and counter-clockwise heat currents due to microscopic reversibility (defined in Section 4.2).
to the principal axis of $C_n$ transforms according to $E_1$, given LCP and RCP respectively transforming according to $E_{-1}$ and $E_{+1}$. These plane waves are therefore encompassed by our consideration of a generic $\mathcal{E}_\Gamma$, and we now simply need to derive the consequences of $E_1$ eigenmode degeneracy on the total scattering (2.11) and total absorption (2.15) of $\mathcal{E}_\Gamma$. Moreover, for each $j_{r,v}$, we can define a new degenerate eigenmode $j_{r,v}^{(\theta)}$, which is rotated by an angle $\theta$.

$$j_{r,v}^{(\theta)} = (\cos \theta - \sin \theta \cot \frac{\pi}{n})j_{r,v} + \sin \theta \csc \frac{\pi}{n}[\hat{C}_n j_{r,v}]$$  \hspace{1cm} (4.14)

Here rotation is defined relative to $j_{r,v}$ by treating $\hat{C}_n j_{r,v}$ as a second basis vector oriented with a rotated angle of $2\pi/n$. We can also define a new excitation field $\mathcal{E}_\Gamma^{(\theta)}$ that is rotated by an angle $\theta$ relative to $\mathcal{E}_\Gamma$ in (4.13), with corresponding induced currents $\mathcal{J}_\Gamma^{(\theta)}$.

$$\mathcal{E}_\Gamma^{(\theta)} = \sum_v b_v \lambda_v j_{r,v}^{(\theta)}, \hspace{1cm} \mathcal{J}_\Gamma^{(\theta)} = \sum_v b_v \hat{j}_{r,v}^{(\theta)}$$  \hspace{1cm} (4.15)

We next split each $j_{r,v}$ into terms of its constituent degenerate eigenmode pairs $\dot{j}_{\pm v}$ and $j_{\pm v}$ that transform according to $E_{+1}$ and $E_{-1}$, respectively. The $\hat{C}_n$ operation used in (4.13) and (4.14) is then specifically described by scaling $\dot{j}_{\pm v}$ and $j_{\pm v}$ by $e^{i2\pi n}$ and $e^{-i2\pi n}$.

$$j_{r,v} = A_v \dot{j}_{+v} + B_v j_{-v} \Rightarrow \hat{C}_n j_{r,v} = A_v \dot{j}_{+v} e^{i2\pi n} + B_v j_{-v} e^{-i2\pi n}$$  \hspace{1cm} (4.16)

Here $A$ and $B$ are complex scalars, and (4.16) also shows that the arbitrary rotation by $\theta$, defined in (4.14), is a unitary operation.

$$\left| (\cos \theta - \sin \theta \cot \frac{\pi}{n}) + \sin \theta \csc \frac{\pi}{n} e^{\pm i2\pi/n} \right|^2 = 1$$  \hspace{1cm} (4.17)

To consider the expressions for scattering (2.11) and absorption (2.15), let us first consider a modified inner product $\beta_{vw}$ between $\dot{j}_{\pm v}$ and $\bar{M} \cdot \dot{j}_{\mp w}$, where $\bar{M}$ is some matrix.

$$\beta_{vw} = \int |\dot{j}_{\pm v}(r')| \cdot \bar{M}(r', r) \cdot \dot{j}_{\mp w}(r) dr^3$$  \hspace{1cm} (4.18)

We can now utilise $\hat{R}$ being unitary, and substitute its operation with the corresponding scalar matrix representations $\hat{R} j_{\pm v}(r) = D_{\Gamma \pm v}^{(\hat{R})} j_{\pm v}(r)$, to write $\beta_{vw}$ in a new form.

$$\beta_{vw} = \int |\dot{j}_{\pm v}(r')|^* \cdot |\hat{R}^{-1} \hat{R} \cdot |\bar{M}(r', r) \cdot |\hat{R}^{-1} \hat{R} \cdot \dot{j}_{\mp w}(r) dr^3$$  \hspace{1cm} (4.19)

Notably, if $\bar{M}$ satisfies $\bar{M}(\hat{R} r, \hat{R} r') = \hat{R} \cdot \bar{M}(r, r') \cdot \hat{R}^{-1}$, and we commute scalars $D_{\Gamma}$ through $\bar{M}$, we can go one step further.

$$\beta_{vw} = \int |D_{\Gamma \pm v}^{(\hat{R})} \dot{j}_{\pm v}(r')|^* \cdot |\hat{D}_{\Gamma \mp w}^{(\hat{R})} \bar{M}(\hat{R} r, \hat{R} r') \cdot \dot{j}_{\mp w}(r)| dr^3$$  \hspace{1cm} (4.20)

We can now set up an equation analogous to (4.5) by considering the sum of (4.20) over
all symmetry operations $\hat{R}$ in $C_n$.

$$N_{C_n} \beta_{vw} = \sum_{\hat{R}} \beta_{vw} = 0, \quad \text{using (4.4) and (4.20)} \quad (4.21)$$

Here $N_{C_n}$ denotes the number of symmetry operations in $C_n$. We now note that (4.21) can only be true if $\beta_{vw} = 0$, because there is always the identity operation in $C_n$, and hence $N_{C_n} \geq 1$. Comparing (4.18) to (4.21) provides the following statement:

$$\int [j_{\pm v}(r')]^* \cdot [\tilde{M}(r', r)] \cdot j_{\mp w}(r) d^3r' d^3r = 0 \quad (4.22)$$

We are now able to put everything together. It follows from (4.22), with (4.14), (4.16) and (4.17), that our modified inner products between eigenmodes are conserved under rotations (4.14) by $\theta$.

$$\int [j_{\Gamma v}(r')]^* \cdot [\tilde{M}(r', r)] \cdot j_{\Gamma w}(r) d^3r' d^3r = \int [j_{\Gamma v}(r')]^* \cdot [\tilde{M}(r', r)] \cdot j_{\Gamma w}(r) d^3r' d^3r \quad (4.23)$$

By combining (4.15) and (4.23), we reach a similar conclusion for $J_{\Gamma}$ under rotations by $\theta$.

$$\int [J_{\Gamma v}(r')]^* \cdot [\tilde{M}(r', r)] \cdot J_{\Gamma w}(r) d^3r' d^3r = \int [J_{\Gamma v}(r')]^* \cdot [\tilde{M}(r', r)] \cdot J_{\Gamma w}(r) d^3r' d^3r \quad (4.24)$$

Two important choices of $\tilde{M}$ are $(\tilde{G}_0(r, r') - \tilde{L} \delta(r-r'))$ in (2.11) and $\text{Im}\{((\tilde{\epsilon} - \epsilon_0)^{-1})$ in (2.15), for which (4.24) shows that $P_{\text{scat}}$ and $P_{\text{abs}}$, are invariant to the rotation angle $\theta$. This thereby completes a derivation of the result in [A.1] starting from the eigenmode degeneracy due to reciprocity presented in [A.6]. The conclusion is also slightly more general, given that any external field distribution $E_0 = \epsilon_{\Gamma}$ that transforms entirely according to the irreducible representation $\Gamma = E_1$, will be scattered and absorbed by exactly the same amount irrespective to any rotation in orientation of $E_0$ about the principal axis of $C_n$. This result encompasses, but is not necessarily restricted to, normally incident plane waves. Additionally, the argument we just presented also applies to $S_4$ symmetry groups owing to its analogous complex conjugate irreducible representations, noting that $S_4$ does not contain a $C_n$ ($n \geq 3$) subgroup.
Asymmetry and chirality

This will be the converse to the previous Chapter on symmetry, being a discussion on the absence of symmetry. I first define chirality as a geometric concept, then provide some basic arguments as to why this concept can have counterparts in optical scattering, and their relation to both chiral and achiral scattering objects. In the second section, I focus specifically on the differences in scattering from oppositely handed, circular-polarised plane waves, particularly the known effects of circular dichroism and circular conversion dichroism, and the specific geometric symmetries that suppress them. I then discuss circular dichroism in absorption, first discussed in [A.6], as a further distinct effect that originates from differing optical responses between reciprocal plane waves.

5.1 Defining chirality in optics

A physical object is said to be chiral if it cannot be arranged through any number of rotations and translations into coincidence with its mirror image, implying the given object is a different object to its mirror image. This is a definition of chirality, and we can use it to also define left and right enantiomers as the distinct pair of mirror images for a single chiral object. We now relate chirality to symmetry. Any mirror image operation applied to a given object can be described by two consecutive operations acting on the coordinate system: a reflection plane $\hat{\sigma}$ as the mirror, and an arbitrary rotation $\hat{C}_\theta$ to account for the orientation of the mirror. These two operations acting on coordinates $(x, y, z)$ can next be written as transformation matrices, $\bar{D}^{(\hat{\sigma})}$ and $\bar{D}^{(\hat{C}_\theta)}$, in (5.1) and (5.2), which notably commute with each other: $\bar{D}^{(\hat{C}_\theta)} \bar{D}^{(\hat{\sigma})} = \bar{D}^{(\hat{\sigma})} \bar{D}^{(\hat{C}_\theta)}$. This is seen by writing $\bar{D}^{(\hat{C}_\theta)}$ as the product of three rotations about each axis, as in (5.2), then directly confirming the commutivity for each of these rotation matrix with $\bar{D}^{(\hat{\sigma})}$.

\[
\bar{D}^{(\hat{\sigma})} = \begin{pmatrix}
-1 & 0 & 0 \\
0 & 1 & 0 \\
0 & 0 & 1
\end{pmatrix}
\] (5.1)

\[
\bar{D}^{(\hat{C}_\theta)} = \begin{pmatrix}
1 & 0 & 0 \\
0 & \cos \theta_x & \sin \theta_x \\
0 & -\sin \theta_x & \cos \theta_x
\end{pmatrix} \begin{pmatrix}
\cos \theta_y & 0 & -\sin \theta_y \\
0 & 1 & 0 \\
\sin \theta_y & 0 & \cos \theta_y
\end{pmatrix} \begin{pmatrix}
\cos \theta_z & \sin \theta_z & 0 \\
-\sin \theta_z & \cos \theta_z & 0 \\
0 & 0 & 1
\end{pmatrix}
\] (5.2)

Given our initial definition of chirality permits any arbitrary rotation after performing the mirror image, where we have just seen that arbitrary rotation commutes with reflection, the orientation of the reflection plane isn’t important when defining the two enantiomers of a chiral object. Conversely, it means an object that is symmetric about any reflec-
Asymmetry and chirality

The plane will always be the same object as its mirror image, meaning it is not chiral, or rather: it is achiral. Similarly, point inversion, which can be written as the product of three perpendicular mirror planes and correspondingly understood as switching enantiomer handedness three times (an odd number), will also imply achirality. By confining ourselves to known geometric symmetry operations, we can now define chirality as being equivalent to the absence of any planes of reflection symmetry $\hat{\sigma}$, points of inversion symmetry $\hat{i}$, or axes of improper rotation symmetry $\hat{S}_n$ (rotation $\hat{C}_n$ and perpendicular reflection $\hat{S}_h$). See the example geometries exhibiting each symmetry in Figure 5.1a.

We now consider the chirality of electromagnetic fields occupying some given volume $V$, for which I will draw a distinction between local chirality ($V \to 0$) and global chirality ($V \not\to 0$). In a spherical volume $V \to 0$, the local fields $\mathbf{E}(t)$ and $\mathbf{B}(t)$ at the sphere origin will be chiral if there is a component of $\mathbf{E}$ parallel to $\mathbf{B}$. That is to say: fields with $\mathbf{E} \cdot \mathbf{B} \neq 0$ have no planes of reflection, axes of improper rotation, or points of inversion symmetry. This is a consequence of $\mathbf{E}$ having opposite parity to $\mathbf{B}$ under reflection and inversion operations, which is imposed by the Maxwell curl equation (1.2). The absence of symmetry is therefore illustrated in Figure 5.1b by depicting $\mathbf{B}$ as the corresponding circulation of $\mathbf{E}$. Chirality for local fields now obtains physical significance when considering the interaction between fields and objects that are sufficiently small to be considered as points. Moreover, if the local fields are achiral, the converse statement is that there is some local reflection, inversion or improper rotation transformation on the coordinates of $V$ that leave these achiral local fields unchanged. If there is a small chiral point particle or molecule in $V$,
the achiral coordinate transformation that leaves the fields unchanged would necessarily transform the small chiral point particle into its opposite enantiomer. This means that any interaction between achiral local fields and different enantiomers of a chiral molecule is equivalent to a coordinate transformation, which we can define as being a trivial difference. Chiral local fields are therefore necessary to allow nontrivial differences in interaction, one key example of this being the total electromagnetic power absorbed by the molecule. Local field chirality is therefore a relevant concept for molecular sensing or manipulation that can distinguish between enantiomers [109,110]. However, to be more explicit, such investigations have sought to introduce a bias in the signed magnitude of local helicity density $\text{Im}\{E^* \cdot H\}$, which has been show to create a net difference in both the absorption and radiation of electromagnetic fields by oppositely handed chiral molecules [11,111].

For nanoparticle oligomers, the relevant definition of optical chirality also relates to the geometric concept of chirality. However, owing to the non-negligible size of oligomers and nanostructures relative to the considered wavelength, we must attribute optical chirality in nanoparticle oligomers to field distributions in volumes $V \neq 0$. This implicitly loosens the conditions for realising a chiral field distributions. As an example of such, the spatial field distributions $E(r,t)$ for oppositely handed, circular-polarised plane waves in a homogeneous background are mirror images, and cannot be superimposed onto each other, as seen in Figure 5.2a. These fields therefore have a form of global chirality even though the fields are locally achiral at any single point: $E(t)$ and $B(t)$ are orthogonal in any plane wave. When considering such global chirality of fields in the current model for optical scattering (2.8), the relevant volume $V \neq 0$ is that internal to the scattering object: the induced currents $J$, and hence their scattering response in (2.6), will inherit the symmetry transformation properties of the applied electric field distribution $E_0$ internal to $V$, as discussed following (4.7). The significance of chirality of the internal fields is now different to that of local optical chirality. Local optical chirality implicitly relied on the assumption that the volume $V \to 0$ is a homogeneous sphere, to disregard any variation of $V$ under mirror, inversion or improper rotation operations on the coordinate system. Here the volume $V \neq 0$, which is relevant for scattering, is only invariant under the symmetry operations of corresponding the symmetry group for the given scattering object. In the first instance, we can consider what happens if the the scattering object is chiral. The enantiomers of internal fields will exist in different enantiomers of the scattering object: any reflection, inversion, or improper rotation of the coordinate system flips the handedness of both the internal field distribution and the scattering object. Consequently, a left-circular polarised (LCP) plane wave in a chiral $V$ never has symmetric equivalence to that of a right-circular polarised (RCP) plane wave in the same $V$, and this permits differences between the resulting scalar quantities like total scattering (2.11) and total absorption (2.15). This thereby resembles the situation previously considered for small chiral particles and local field chirality. However, we can now consider the case where the scattering object is achiral, in which both enantiomers of chiral internal fields will exist in the same object: there is a coordinate transformation that preserves $V$ while flipping the handedness of the internal fields. Such enantiomers of the chiral internal fields are then equivalent to each other under a coordinate transformation, which makes scalar quantities like total scattering (2.11) and total absorption (2.15) conserved for each enantiomer. However, while we can define equivalent enantiomers for any $E_0$ distribution, it is not guaranteed that these enantiomers align with co-propagating LCP and RCP plane waves confined to $V$, which is the relevant comparison for effects such as circular dichroism, to be discussed in the next Section. Moreover, if there is no sequence of symmetry opera-
tions from the symmetry of group for \( V \) to transform one spiral enantiomer of a circular polarised plane wave in Figure 5.2a into its co-propagating enantiomer, the internal fields of oppositely handed plane waves will have no symmetric equivalence, in precisely the same manner as true chiral geometries. For example, see Figure 5.2b, which illustrates the enantiomers of a circularly polarised plane wave superimposed onto the \( S_4 \) achiral geometry of Figure 5.1a, and these are not related via symmetry operations of \( S_4 \) group. This is the first tangible distinction between globally and locally chiral fields: many “chiral” optical effects from chiral molecules can arise from achiral nanoparticles, simply because the internal fields of the relevant circular-polarised plane waves are inequivalent under the symmetry transformations of the nanoparticles. This distinction between global and local chirality has arisen in many contexts for artificial nanostructured systems, which is what I discuss in the next section.

Figure 5.2: Depiction of global optical chirality: (a) enantiomers formed from the electric field distributions of two oppositely handed circular-polarised plane waves (image taken from [A.6]), (b) enantiomers of the same plane waves when superimposed onto an achiral volume, the \( S_4 \) geometry in Figure 5.1a.

### 5.2 Circular dichroism effects

Here I discuss the differences experienced between LCP and RCP plane waves in: the power dissipated from the incident field (circular dichroism), and the power of the transmitted cross-polarization (circular conversion dichroism). This is a precursor to Section 5.3, where I will introduce and discuss the analogous differences in power dissipated from the total fields (circular dichroism in absorption).

Circular dichroism is one of the typical hallmarks of chiral optical response: a difference in the extinction of co-propagating LCP and RCP plane waves. Circular dichroism is often seen as a measure of the magnitude of chiral optical response, given extinction is the total amount of light removed from the incident field (2.18) and a measure for the magnitude of total optical interaction. One of the highest densities of optical circular dichroism is achieved with chiral nanostructures, producing 35% absolute difference between the co-polarised transmission power of LCP and RCP, relative to the incident light power, at a thickness of \( \lambda/6 \) [112]. However, achiral nanoparticles have been observed to exhibit upwards of 80% extinction contrast between LCP and RCP for specific propagation directions [113]. Circular dichroism is therefore not specific to
chiral geometries. I now formally quantify symmetry conditions for circular dichroism in extinction. For notation of circular-polarised plane waves, I will use $+$ and $-$ to denote LCP and RCP relative to a specified $k$, meaning $E_0(\pm) = \frac{|E_0|}{\sqrt{2}} \left( \pm i \right) e^{i k \cdot r}$, where the two indices denote a right-handed orthonormal basis in the plane whose normal is directed along $+k$. We begin by incorporating the result of Section 4.2, where it was derived that incident fields $E_0^*$ and $E_0$ both experience the same extinction, which then constrains circular dichroism as follows:

Circular dichroism in extinction is forbidden if any sequence of geometric symmetry operations can arrange the distribution of $E_0(\pm)$ in $V$ to be proportional to the distribution in $V$ of either (i): $E_0(\mp)$, or (ii): $[E_0(\mp)]^*$. Neither condition (i) or (ii) will constrain a chiral scattering object, as they both require an achiral symmetry operation to relate $E_0(\pm)$ to $E_0(\mp)$ in any $V$. In other words, these conclusions are only relevant for an achiral scattering object. Specifically, (i) is the standard constraint that a sequence of geometric symmetry operations will allow us to relate co-propagating LCP and RCP plane waves as a coordinate transformation, for instance: a reflection symmetry plane parallel to the propagation direction, analogous to Figure 5.2a. However, (ii) is an additional and important constraint from reciprocity, because it forbids circular dichroism due to inversion symmetry and symmetry under reflection perpendicular to the propagation direction. A geometric inversion operation $\hat{i}$ performs $\hat{i}E_0(\pm) = -[E_0(\mp)]^*$, and we can apply (ii). A reflection operation $\hat{\sigma}_h$ about a symmetry plane perpendicular to the propagation direction will perform $\hat{\sigma}_hE_0(\pm) = [E_0(\pm)]^*$, and we can apply (ii). We can now define minimum symmetries that will suppress circular dichroism:

- The $C_{i}=S_2$ symmetry group forbids circular dichroism for any plane wave using (ii), i.e.

\[
E_0(\pm) \equiv \left( \pm \frac{1}{i} \right) e^{i k \cdot r} \longleftrightarrow \text{reflection} (\mp i) \quad e^{-i k \cdot r} \longleftrightarrow \text{reciprocal plane waves} \left( \mp \frac{1}{i} \right) e^{i k \cdot r} \quad (5.3)
\]

- The $C_{1h}=S_1$ symmetry group forbids circular dichroism for plane waves propagating either parallel, using (i), or perpendicular, using (ii), to the symmetry plane

I have called $C_i$ and $C_{1h}$ groups the minimum symmetries because they are the only groups that contain only an identity operation and a single achiral symmetry operation. The only achiral symmetry operation we appear to miss is improper rotation, but $\hat{S}_1$ is equal to reflection $\hat{\sigma}_h$ in $C_{1h}$, $\hat{S}_2$ is equal to inversion $\hat{i}$ in $C_i$, and $\hat{S}_3$ onwards exists only in larger symmetry groups given $\hat{S}_n \neq \hat{S}_n^{-1}$ for $n \geq 3$, which implies there must exist at least one more symmetry operation $\hat{R} = \hat{S}_n^{-1}$ in order to form a group.

<table>
<thead>
<tr>
<th>$A_\hat{E}$</th>
<th>$i$</th>
<th>$\hat{i}$</th>
<th>$\hat{\sigma}_h$</th>
<th>$\hat{\sigma}_h$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A_1$</td>
<td>1</td>
<td>1</td>
<td>$x^2, y^2, z^2$</td>
<td>$A_1$</td>
</tr>
<tr>
<td>$A_2$</td>
<td>1</td>
<td>-1</td>
<td>$x, y, z$</td>
<td>$B_1$</td>
</tr>
<tr>
<td>$A_3$</td>
<td>1</td>
<td>-1</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 5.1: Character tables for the (left) $C_i$ symmetry group, and (right) $C_{1h}$ symmetry group.
Circular conversion dichroism is the difference in the conversion efficiency of an LCP or RCP plane wave into the cross-polarised RCP or LCP plane wave. As previously discussed, circular dichroism in extinction is forbidden in a two-dimensional surface due to its implicit $C_{1h}$ symmetry, yet there were observations of a difference in the total transmitted power under illumination by LCP and RCP plane waves from such a surface, which was explained to be due to circular conversion dichroism [114,115]. In other words, the observed difference in the total transmitted power was a difference in the power of the cross-polarised transmission, while the co-polarised transmission was conserved. A symmetry that will forbid circular conversion dichroism is $C_n$ ($n \geq 3$) discrete rotational symmetry, provided $k$ is parallel to the principal axis of $C_n$. This was first derived by Fernandez-Corbaton [116], in the context of the conservation of helicity $|E \pm \sqrt{\mu_0/\epsilon_0}H|^2$ in transmission, but we can also quickly re-illustrate the result here from the perspective of symmetry operations acting on plane waves. We begin by noting that a rotation by any angle $\phi$ about the propagation axis of a circularly polarized plane wave is equivalent to a phase shift.

\[
\begin{pmatrix}
\cos(\phi) & -\sin(\phi) \\
\sin(\phi) & \cos(\phi)
\end{pmatrix}
\begin{pmatrix}
1 \\
\pm i
\end{pmatrix}
= \begin{pmatrix}
1 \\
\pm i
\end{pmatrix} e^{\mp i\phi}
\] (5.4)

As such, an LCP or RCP plane wave $E_0^{(\pm)}$ with $+\hat{k}$ parallel to the principal axis of $C_n$ will transform according to the $E_{\pm 1}$ irreducible representation of $C_n$. This follows because a symmetric rotation by $\frac{2\pi}{n}$ of the coordinate system is equivalent to applying a uniform phase shift of $e^{\mp i\frac{2\pi}{n}}$ to $E_0^{(\pm)}$, and extending this to multiples of $\frac{2\pi}{n}$ writes out the $E_{\pm 1}$ transformation properties, e.g. see Table 4.2 for $n = 3$. The physical locations lying on the principal axis $r = z$ remain static with any symmetric rotation operation in $C_n$, i.e. $\hat{C}_n z = z$, hence the total fields $E(z)$ then transform as $E(z) \rightarrow \hat{C}_n E(\hat{C}_n z) = \hat{D}(\hat{C}_n) E(z)$. Given rotations of the coordinate system are equivalent to phase changes of $E_0^{(\pm)}$, as according to $E_{\pm 1}$, it follows that $E(z)$ must also transform under rotations as the same uniform phase shifts, and it therefore transforms according to $E_{\pm 1}$. A cross-polarised plane wave $E_0^{(\mp)}$ can then only exist in $E(z)$ if it transforms according to $E_{\pm 1}$, following the orthogonality in (4.5), if it propagates in the $-\hat{k}$ direction. As such, there is no conversion of circular polarisation in transmission, being $E_0^{(\mp)}$ in the $+\hat{k}$ direction. A similar argument also follows to conclude there is no conserved polarisation in reflection, $E_0^{(\pm)}$ in the $-\hat{k}$ direction. Notably, $C_n$ ($n \geq 3$) is a chiral symmetry group, meaning it contains no achiral symmetry operations. As such, rotationally symmetry scattering objects that are also chiral, can exhibit circular dichroism in extinction while exhibiting no circular conversion dichroism.
Circular dichroism in absorption is the difference in the loss due to material absorption (2.15) experienced by LCP and RCP plane waves, and it can exist separate of the two forms circular dichroism introduced in the previous section. Moreover, the work in [A.6] showed circular dichroism in absorption in a system with $C_{2mn}$ ($2m \geq 3$) symmetry that forbids both: circular dichroism in extinction ($C_i \subset C_{2mn}$), and circular conversion dichroism ($C_{2m} \subset C_{2mn}$) for normal incidence plane waves. I have referred to this effect as circular dichroism in absorption, in recognition that dissipative material loss is necessary to ensure that the scattering is not equal to the extinction. Notably, however, the absence of circular dichroism in extinction, and simultaneous presence of circular dichroism in absorption, means circular dichroism must also be occurring in scattering as the difference of extinction and absorption. The secondary conclusion of [A.6] was that the observed circular dichroism in the absorption required nonorthogonal eigenmodes, and the magnitude of the dichroism could be enhanced through Fano interference, because it implies the existence of highly nonorthogonal eigenmodes as discussed in Chapter 3. In this section, I will explore further why nonorthogonal eigenmodes were necessary for circular dichroism in the absorption of LCP and RCP plane waves.

We firstly recognise that the $E_1$ irreducible representation of $C_n$ ($n \geq 3$), which we used in the previous section to describe the symmetry transformation properties for normally incident plane waves under the corresponding rotations, is separated in $C_{nh}$ into either: $E_{1u}$ and $E_{1g}$ irreducible representations if $n$ is even, or $E'_{1}$ and $E''_{1}$ if $n$ is odd. These irreducible representations respectively corresponding to whether the given quantity is even or odd parity under inversion $\hat{i}$ (n even), or reflection $\tilde{\sigma}_h$ (n odd). For ease of notation, I will now just focus on $E_{1u}$, but analogous arguments will apply to $E_{1g}$, $E'_{1}$ and $E''_{1}$. Now, as was the case with the $E_1$ irreducible representation of $C_n$ in Chapter 4, the eigenmodes $j_v$ of $E_{1u}$ occur in degenerate pairs $j_{v+}$ and $j_{v-}$ between the complex conjugate irreducible representations $E_{1u+}$ and $E_{1u-}$. We can consider the $k$ of $E_0^{(\pm)}$ to be aligned with the $\pm$ notation such that $E_0^{(\pm)}$ is a linear combination of eigenmodes $j_{v\pm}$.

\[
E_0^{(\pm)} = \sum_v \lambda_v a_{v\pm} j_{v\pm} + \sum_v \lambda'_v a'_{v\pm} j'_{v\pm} \quad (E_{1g} \text{ component})
\]

Here the $a_v$ cannot be calculated using (3.4), because $j_{v\pm}$ and $j_{v\mp}^*$ belong to different irreducible representations $E_{1u\mp}$ and $E_{1u\mp}$, and are therefore orthogonal using (4.5). This means that $\int j_{v\pm} \cdot j_{w\mp} = 0$, which breaks the approach to calculate the excitation amplitude $a_{v\pm}$ through unconjugated projections in (3.4). Instead, the $a_{v\pm}$ can be calculated using: $\int j_{v\pm} \cdot j_{w\mp} = 0$ if $\lambda_v \neq \lambda_w$, as derived in (12) of [A.6].

\[
\lambda_v a_{v\pm} = \frac{\int j_{v\pm} \cdot E_0^{(\pm)}}{\int j_{v\mp} \cdot j_{v\pm}} \quad (5.6)
\]

By relating co-propagating LCP and RCP plane waves through the inversion symmetry operation $E_0^{(\pm)} = -[iE_0^{(\pm)}]^*$, combined the unitarity of the inversion operation $i \cdot i = \hat{E}$, (5.6) is actually implying the excitation amplitude $a_{v\pm}$ is proportional to the true
projection of $E_0^{(\mp)}$ onto $j_{a\mp}$.

$$\lambda_v a_{v\mp} = \frac{\int [E_0^{(\mp)}]^* \cdot j_{a\mp}}{\int j_{c\mp} \cdot j_{a\mp}}$$  \hspace{1cm} (5.7)

This is sufficient to now show that the extinction due to each eigenmode will be conserved for LCP and RCP plane waves, by substituting $J^{(\pm)} = \sum_v \lambda_v^{-1} [\lambda_v a_{v\mp} j_{a\mp}]$ with (5.7) into the expression for extinction (2.18).

$$P_{\text{ext}} = \frac{1}{2} \int \Re \{ [E_0^{(\mp)}]^* \cdot J^{(\pm)} \} = \sum_v \frac{1}{2} \Re \left\{ \lambda_v^{-1} \left( \frac{\int [E_0^{(\mp)}]^* \cdot j_{a\mp}}{\int j_{c\mp} \cdot j_{a\mp}} \right) \right\}$$  \hspace{1cm} (5.8)

Note that the dependence on $+$ or $-$ has disappeared from the extinction for each independent $v$, rather than the sum of all $v$. As such, the degenerate eigenmodes $j_{v+}$ and $j_{v-}$ are associated with the same extinction under their respective excitation from $E_0^{(+)}$ and $E_0^{(-)}$.

Yet these two eigenmodes are importantly not chiral enantiomers of each other, $j_{v\mp}$ is proportional to the same $j_{v\mp}$ under inversion, and inversion is an achiral symmetry operation. Additionally, if the eigenmodes are nonorthogonal, there is no reciprocal symmetry relationship $j_{v\mp} \propto j_{v\pm}^{*}$ as per the discussion on circular dichroism in extinction, because this would correspond to $j_{v\pm}$ being orthogonal to all eigenmodes by following the result $\int j_{v\pm} \cdot j_{w\pm} = 0$ for $\lambda_v \neq \lambda_w$, which we previously used to define (5.6). Therefore, by nature of $j_v$ being nonorthogonal to some $j_w$, we know that $j_{v+}$ and $j_{v-}$ must be nontrivially distinct current distributions, having no symmetric relationship through spatial symmetry operations, nor through time-reversal. Consequently, by making an oligomer of coupled nanoparticles with both strongly dissipative nanoparticles and strongly radiative nanoparticles, we can translate any differences in the distribution of $j_{v+}$ and $j_{v-}$ into differences of absorption and scattering. This is precisely what is done in [A.6], using nanoparticle oligomer with small nanoparticles (dissipative component) surrounding a large nanoparticle (radiative component).

This scenario was explored further in [A.4], where we instead considered the difference in absorption and scattering of co-polarised plane waves propagating in opposite directions, irrespective of polarization. The previous circular dichroism in absorption and scattering between LCP and RCP plane waves of an object with $C_{nh}$ symmetry, is likened to a difference in the scattering and absorption of reciprocal plane waves for a scattering object with $C_n$ symmetry. For such a situation, there is a simple one-dimensional analogy of an ensemble of lossy and radiative nanoparticles: an ideal 100% absorbing surface next to an ideal 100% reflective surface, see Figure 5.3a. Reciprocal plane waves propagating in opposite directions are absorbed if incident on the absorbing surface first or scattered if incident on the reflective surface first; both cases have 100% extinction, satisfying reciprocity, but the portion of absorption and scattering varies completely between reciprocal plane waves. In [A.4], this to the situation was liken to the two spheres illustrated in Figure 5.3b, one absorbing and one radiative, which could be modelled as point electric dipoles. Using equally sized gold spheres, but artificially increasing the dissipative absorption of gold in one sphere, differences in scattering and absorption appeared between reciprocal plane waves. To make the effect stronger with realistic material parameters, the geometry was then simply adjusted to be two parallel crosses ($C_{4v}$ symmetry) that
could support much larger dipole moments than spheres at a comparable separations. The arm thickness of one cross was then varied to make it a better or worse radiator, and thereby control its dissipative absorption to resemble the lossy sphere in Figure 5.3b. This geometry then predicted comparable differences in absorption between reciprocal plane waves, see Figure 3 of [A.4], as the circular dichroism in absorption seen in Figure 4 of [A.6]. By then adding reflection planes to increase symmetry of the cross geometry to $C_nv$ ($n \geq 3$), the absorption and scattering behaviour did not depend\footnote{$C_n$ symmetry to remove linear polarization dependence (Chapter 4), and $\sigma_v$ reflection symmetry parallel to the propagation direction to remove dependence on LCP and RCP.} on the polarization of this incident field, and thereby only depended on the propagation direction of the plane wave.
Asymmetry and chirality
Conclusions

It is apparent that the field of metamaterials has been maturing; operational principles are gradually becoming clearer, and functional devices more frequent. Many challenges now tend toward implementation in manufacturing and technology. However, a frontier the research community can significantly impact is developing understanding of the physics in metamaterial operation, and removing existing ambiguities. This particular pursuit will be relevant for future opportunities that now lie in how the concepts of metamaterials, and the advancements in fabrication, can be utilised to explore new physics. In this thesis I have focused on developing both principles and modelling approaches for the collective optical behaviour of nanoparticle oligomers. These systems have provided an avenue to granularly quantify the formation of collective resonances from constituent elements, which has enabled delivery of the following list of key results over the course of my studies:

- A new model for Fano resonances in optical scattering systems was presented, and is based on the interference between the nonorthogonal eigenmodes. This was then implemented to correctly describe Fano resonances occurring in both plasmonic and high-refractive-index dielectric nanoparticle oligomer systems. Chapter 3; [A.2].

- The derivation of collective resonances in plasmonic and dielectric nanoparticle systems from their resonant subsystems, was shown to be translatable to the coupled dipole model while maintaining quantitatively accurate predictions. This was demonstrated to enable simplified calculations, interpretation and analysis of Fano resonances in symmetric arrangements of three plasmonic or dielectric nanoparticles. Chapter 3; [A.5].

- High-refractive-index dielectric nanoparticle oligomers were shown to enable interference effects and Fano resonances between multiple collective magnetic resonances, and thereby behave analogous to two-channel point sources of magnetisation that do not occur naturally. Chapter 3; [A.3].

- Polarisation-independent scattering and absorption losses, while still allowing non-trivial variation of the near-field, was shown to be enforced by symmetry through the combination of $n$-fold cyclic $C_n$ ($n \geq 3$) discrete rotational symmetry and reciprocal eigenmode degeneracy. Chapter 4; [A.1, A.6].

- A new form of circular dichroism due to the interaction of nonorthogonal resonances was presented, and impacts the ratio of radiative scattering loss to dissipative absorption loss. This was explained to be analogous to differences in scattering and absorption between reciprocal plane waves. Chapter 5; [A.4, A.6].
Bibliography


Appendix A

Thesis publications

A.1 Optically isotropic responses induced by discrete rotational symmetry of nanoparticle clusters

Summary

The most robust and general approach to eliminate dependence of scattering on the incident polarization of light will come from the overall symmetry of the scattering geometry, given this is not affected by the operating wavelength or the material properties. There had previously been no comprehensive investigations on this topic except for some example case studies on specific symmetries, particularly given the intrinsic absorption spectra was neglected almost unanimously. In this work, we performed a rigorous investigation on symmetry induced polarisation-independent scattering and absorption. Discrete $n$-fold ($n \geq 3$) rotational symmetry is shown to provide polarisation-independent optical responses, not only in far-field properties, such as extinction, but also for intrinsic absorption, which is usually considered to result from a near-field that is itself highly polarisation-dependent. This paper clarifies both physically and mathematically the uncertainty around the topic of symmetry induced polarisation-independence.

Notes and errata

- While presented derivations use the dipole model, this model can be generalised to continuous current distributions by simply performing a substitution of $J(r) \, dr^3 = -i\omega p(r)$, as was done in Chapter 2. Furthermore, given we consider both electric and magnetic dipoles simultaneously, the derivations of this work actually account for magnetisation current density defined as $J_m(r) \, dr^3 = -i\omega m_r$. The conclusions and derivations of this work will therefore apply generally to any object consisting both permittivity and permeability, which is not made evident in the work itself.

- The curl of the dyadic Green’s function is a factor of $ik$ larger than it should be, however all equations are correct once making the substitution: $\nabla \times \tilde{G}^0(r_i, r_j) \rightarrow \frac{1}{ix} \nabla \times G^0(r_i, r_j)$. 

65
Optically isotropic responses induced by discrete rotational symmetry of nanoparticle clusters

Ben Hopkins,* Wei Liu, Andrey E. Miroshnichenko and Yuri S. Kivshar

Fostered by the recent progress of the fields of plasmonics and metamaterials, the seminal topic of light scattering by clusters of nanoparticles is attracting enormous renewed interest gaining more attention than ever before. Related studies have not only found various new applications in different branches of physics and chemistry, but also spread rapidly into other fields such as biology and medicine. Despite the significant achievements, there still exists unsolved but vitally important challenges of how to obtain robust polarisation-invariant responses of different types of scattering systems. In this paper, we demonstrate polarisation-independent responses of any scattering system with a rotational symmetry with respect to an axis parallel to the propagation direction of the incident wave. We demonstrate that the optical responses such as extinction, scattering, and absorption, can be made independent of the polarisation of the incident wave for all wavelengths. Such polarisation-independent responses are proven to be a robust and generic feature that is purely due to the rotational symmetry of the whole structure. We anticipate our finding will play a significant role in various applications involving light scattering such as sensing, nanoantennas, optical switches, and photovoltaic devices.

1 Introduction

The current surging interest in various applications of nanoscale light-matter interactions, including biosensing,4–6 nanoantennas,6,9 photovoltaic devices9 and many others, has triggered enormous effort into the old and fundamental problem of the manipulation of a particle’s scattering and absorption characteristics.7,8 In the recently emerging fields of nanophotonics, various novel phenomena have been demonstrated involving interaction of nanoparticles with light, such as super-scattering,9,10 control of the direction of the scattered light by metasurfaces,11,12 coherent perfect absorption of light by surface plasmons,13,14 Fano resonances in nanoscale structures15,16 and plasmonic oligomers.17–21 At the same time, the interest in artificial magnetic responses that was fostered by the field of metamaterials has lead to the observation of artificial magnetic modes in nanoparticles and, since then, many related novel scattering features based on the interplay of both electric and magnetic responses have been demonstrated.22–28

To make further breakthroughs in different applications based on particle scattering, there is a fundamental challenge to overcome: polarisation dependence. The dependence of an optical response on polarisation comes from the fact that most structures have dominantly electric responses, which are highly dependent on the polarisation of the incident field. The simplest structure that does not exhibit polarisation-dependent scattering properties is a single spherical particle. According to the Mie theory the total extinction, scattering and absorption cross-sections do not depend on the incident polarisation angle, although the scattering diagram will exhibit some angle-dependent properties.7 It is possible to achieve a polarisation-independent scattering diagram by overlapping the electric and magnetic dipole responses of a single spherical nanoparticle,29,30 but such effects can only be achieved by rigorous engineering of the structure and can only happen in specific spectral regimes. However, it has also been experimentally observed that some plasmonic oligomer structures with discrete symmetries exhibit completely polarisation-independent extinction cross-section spectrums.31–33 This all leads to the question of what the necessary conditions are for an arbitrary system to have polarisation-independent scattering properties.

Inspired by the concepts of symmetry-induced degenerate states in quantum mechanics34 and mode degeneracy in uniform waveguides,35–37 there have been some studies about symmetry-induced polarisation-independent scattering by clusters of particles.38–40 However, as far as we know, there are no rigorous and systematic investigations of this topic. Additionally, in previous studies, usually only the dependence of extinction or scattering spectra on polarisation is investigated and the intrinsic loss spectra is neglected, which can be quite important in its own right, particularly for photovoltaic devices and biological applications.
In this paper we show that established group theory concepts\textsuperscript{40,41} can be used to deduce the effect of symmetry on discrete dipole scattering systems. We then perform a systematic investigation on the optical responses of structures with an $n$-fold ($n \geq 3$) rotational symmetry, where the $n$-fold symmetry axis is parallel to the direction of propagation of the incident plane wave [see Fig. 1]. Such $n$-fold symmetry implies that the optical properties of the system will be identical when rotating the whole structure by $2\pi/n$ radians. But, as we analytically prove, the extinction, scattering and even absorption cross-sections are all identical for rotations of 

\[ p = a_\parallel \mathbf{E} \] (1a)

\[ m = a_\parallel \mathbf{H} \] (1b)

where $\mathbf{E}$, $\mathbf{H}$ are the electric and magnetic fields acting on the particle and $a_\parallel$, $a_\perp$ are the scalar effective polarisabilities as defined below in terms of Mie Theory dipole scattering coefficients $a_1$, $b_1$.

In this approximation the scattered fields from a single particle in free space are then described using dyadic Green’s functions

\[ \mathbf{E}'(r) = k^2 \left[ \frac{1}{r_0} G^0(r, r_0) \mathbf{p} - \frac{\mu_0}{r_0} \left( \mathbf{\nabla} \times G^0(r, r_0) \right) \mathbf{m} \right] \] (3a)

\[ \mathbf{H}'(r) = k^2 \left[ G^0(r, r_0) \mathbf{m} + \frac{1}{\sqrt{\mu_0 r_0}} \left( \mathbf{\nabla} \times G^0(r, r_0) \right) \mathbf{p} \right] \] (3b)

where free space dyadic Green’s functions are defined as

\[ G^0(r, r_0) = G^{(3)}(r, r_0) = a_0 G^{(3)} + b_0 \mathbf{n}_y \otimes \mathbf{n}_y \] (4a)

\[ \mathbf{\nabla} \times G^0(r, r_0) = g^{(3)} = d_y \left( \mathbf{n}_y \otimes \mathbf{n}_y - I^{(3)} \right)^{\frac{1}{2}} \] (4b)

In these the $\mathbf{n}_y \times I^{(3)}$ operator is expressed explicitly as $(\mathbf{n}_y \otimes \mathbf{n}_y - I^{(3)})^{\frac{1}{2}}$ and we have defined scalars

\[ a_0 = \frac{e^{\mathbf{h}_z}}{4 \pi r_0} \left( 1 + \frac{i}{k r_0} - \frac{1}{k^2 r_0^2} \right) \] (5a)

\[ b_0 = \frac{e^{\mathbf{h}_z}}{4 \pi r_0} \left( 1 - \frac{3i}{k r_0} + \frac{3}{k^2 r_0^2} \right) \] (5b)

\[ d_y = \frac{e^{\mathbf{h}_z}}{4 \pi r_0} \left( 1 + \frac{i}{k r_0} \right) \] (5c)

where $r_0 = |r - r_0|$ and $\mathbf{n}_y = \frac{1}{r_0} (r_y - r_y)$.

In an arbitrary system constructed from $N$ particles the fields acting on the $i^{th}$ particle will be the sum of both the externally applied incident fields and the scattered fields from all the
other particles. Therefore the expressions for the dipole moments of the \( i \)th particle are

\[
P_i = a_{\text{e},i} E_i^G + a_{\text{m},i} H_i \tag{6a}
\]

\[
m_i = a_{\text{h},i} H_i + a_{\text{m},i} E_i \tag{6b}
\]

where \( E_i^G \) and \( H_i \) are the externally applied electric and magnetic fields at \( r_i \).

The dipole equations in eqn (6) show that each incident field vector can be equated to some linear combination of the dipole moments and, as such, there will exist some \( 6N \times 6N \) interaction matrix, \( M^{(6N)} \), to relate all the dipole moments to the incident field vectors. To write this relation in the form of a matrix equation we define a state consisting of all dipole moments, \( |p\rangle \), and a state consisting the incident fields acting on each dipole, \( |e\rangle \).

\[
|p\rangle = \left( p_1, \ldots, p_N, \sqrt{\mu_0} m_1, \ldots, \sqrt{\mu_0} m_N \right) \tag{7a}
\]

\[
|e\rangle = \left( E_1^G, \ldots, E_N^G, \sqrt{\mu_0} H_1, \ldots, \sqrt{\mu_0} H_N \right) \tag{7b}
\]

The matrix equation that relates the dipole moments to the incident field is then written as

\[
e_i^G|e^{(6N)}\rangle = \hat{M}^{(6N)}|p\rangle \tag{8}
\]

where \( \hat{M}^{(6N)} \) is a diagonal matrix containing the electric and magnetic scalar polarisabilities, \( a_e \) and \( a_m \), of each particle.

To generate a general expression for the interaction matrix, we define four smaller matrices; two that couple electric or magnetic dipoles together, \( M^{(6)}_{\text{ee}} \) and \( M^{(6)}_{\text{hh}} \), and two that couple electric to magnetic dipoles and vice versa, \( M^{(6)}_{\text{eh}} \) and \( M^{(6)}_{\text{he}} \). These matrices are constructed from the dyadic Green’s functions as to match the concatenation of vectors in \( |p\rangle \) and \( |e\rangle \) [see eqn (7)]

\[
\hat{M}^{(6)}_{\text{ee}} = k^2 \hat{a}_{\text{ee}} \hat{M}^{(6)}_{\text{G}} \tag{9a}
\]

\[
\hat{M}^{(6)}_{\text{hh}} = -k^2 \hat{a}_{\text{hh}} \hat{M}^{(6)}_{\text{G}} \tag{9b}
\]

\[
\hat{M}^{(6)}_{\text{eh}} = k^2 \hat{a}_{\text{eh}} \hat{M}^{(6)}_{\text{G}} \tag{9c}
\]

\[
\hat{M}^{(6)}_{\text{he}} = k^2 \hat{a}_{\text{he}} \hat{M}^{(6)}_{\text{G}} \tag{9d}
\]

where \( \hat{a}_{\text{ee}} \) is a diagonal matrix containing the electric or magnetic scalar polarisability of each particle and \( \hat{M}^{(6)}_{\text{G}} \), \( \hat{M}^{(6)}_{\text{G}} \) are defined as

\[
\hat{M}^{(6)}_{\text{G}} = \begin{pmatrix}
\hat{G}^{(3)} & -\hat{G}^{(3)}_2 & \cdots & -\hat{G}^{(3)}_N \\
-\hat{G}^{(3)}_2 & \hat{G}^{(3)} & \cdots & -\hat{G}^{(3)}_N \\
\vdots & \vdots & \ddots & \vdots \\
-\hat{G}^{(3)}_N & -\hat{G}^{(3)}_N & \cdots & \hat{G}^{(3)}
\end{pmatrix} \tag{10a}
\]

The complete interaction matrix as defined in eqn (8) to relate incident field to dipole moments is then constructed as

\[
\hat{M}^{(6N)} = \hat{I}^{(6N)} + \begin{pmatrix}
\hat{M}^{(6N)}_{\text{ee}} & \hat{M}^{(6N)}_{\text{eh}} \\
\hat{M}^{(6N)}_{\text{he}} & \hat{M}^{(6N)}_{\text{hh}}
\end{pmatrix} \tag{11}
\]

As such all further analysis will use the matrix equation, eqn (8), to study the optical responses of particle systems.

3 Polarisation invariance and symmetry

In this section we derive an expression for the commutation relation between the interaction matrix (see eqn (8)) and a generic symmetry operation in order to implement group theory principles and restrict the shape of the interaction matrix based on the symmetry of a given system. We will consider the case of general particle systems that have a rotational symmetry described by an \textit{n-fold axis}; an axis about which any number of \( 2\pi/n \) rotations will leave the system unchanged. This symmetry is also referred to as cyclic symmetry and the corresponding group of operations that represent this symmetry are denoted \( C_n \) (the cyclic group). In doing this, we will show that the extinction, scattering and absorption cross-sections are all independent of the incident field polarisation in any system with cyclic symmetry.

In the coupled dipole equations (eqn (6)), the only terms that contain information on the geometrical structure of a system are the dyadic Green’s functions, \( \hat{G}^{(3)}_{\text{ee}} \) and \( \hat{G}^{(3)}_{\text{eh}} \). Moreover, it follows from the definitions in eqn (4) that both \( \hat{G}^{(3)}_{\text{ee}} \) and \( \hat{G}^{(3)}_{\text{eh}} \) will transform as a change of basis when any unitary operation, \( \hat{U}^{(3)} \), is applied uniformly to a system’s position vectors

\[
\hat{G}^{(3)}_{\text{ee}} \rightarrow \hat{U}^{(3)} \hat{G}^{(3)}_{\text{ee}} \hat{U}^{(3)\dagger} \tag{12a}
\]

\[
\hat{G}^{(3)}_{\text{eh}} \rightarrow \hat{U}^{(3)} \hat{G}^{(3)}_{\text{eh}} \hat{U}^{(3)\dagger} \tag{12b}
\]

For \( \hat{G}^{(3)}_{\text{eh}} \) to transform in this way, we must also acknowledge that it can always be expanded as a Taylor series. This is true because the magnitude of every component in \( (n_\phi \otimes n_\phi - I) \) will be less than or equal to one and therefore a Taylor series for \( (n_\phi \otimes n_\phi - I) \), and hence \( \hat{G}^{(3)}_{\text{eh}} \), will always converge. So the overall interaction matrix must also transform in an analogous manner given its construction in terms of dyadic Green’s functions (shown in eqn (9)-(11)) and because every \( \hat{G}^{(3)}_{\text{ee}} \) and \( \hat{G}^{(3)}_{\text{eh}} \) will transform uniformly according to eqn (12)

\[
\hat{M}^{(6N)} \rightarrow \hat{U}^{(6N)} \hat{M}^{(6N)} \hat{U}^{(6N)\dagger} \tag{13}
\]

where \( \hat{U}^{(6N)} \) is defined such that it applies \( \hat{U}^{(3)} \) to the position vector of every particle in \( \hat{M}^{(6N)} \).
de ned for eqn (8). It is then straightforward to rearrange eqn (7)
\(R^{(i)}r_j = r_i\) for each \(i, j \in \{1, \ldots, N\}\). (15)

Therefore we can express eqn (12) in the following manner
\(R^{(i)}(\tilde{R}^{(j)})^T = \tilde{G}_D^{(i)}\) (16a)
\(R^{(i)}R^{(j)}(\tilde{R}^{(i)})^T = \tilde{G}_D^{(j)}\) (16b)

Noticeably this means that \(R^{(i)}\) will not necessarily act as a
symmetry operation on these dyadic Green’s functions despite
being a symmetry operation on the structure. It does, however,
show that there must exist a single permutation matrix, \(R^{(i)}\),
for each \(R^{(i)}\) such that
\[R^{(i)}M^{(i)}(\tilde{R}^{(j)}N^{(j)})^T = (\tilde{R}^{(i)}N^{(j)})^T M^{(i)}(\tilde{R}^{(j)})^T\]\ (17a)
\[R^{(i)}R^{(j)}M^{(j)}(\tilde{R}^{(i)}N^{(i)})^T = (\tilde{R}^{(i)}N^{(i)})^T M^{(j)}(\tilde{R}^{(j)})^T\] (17b)

where \(M^{(i)}\) and \(M^{(j)}\) are the quadrants of \(M^{(i)}\) as defined
in eqn (10) and \(R^{(i)}\) is defined in a manner analogous to eqn (14).
It then follows that there will also exist a permutation matrix,
\(R^{(i)}\), which is constructed from four identical \(R^{(i)}\) quadrants and satisfies
\[R^{(i)}M^{(i)}R^{(j)}(\tilde{R}^{(j)}N^{(j)})^T = (\tilde{R}^{(i)}N^{(j)})^T M^{(i)}(\tilde{R}^{(j)})^T\] (18)

We can consider all non-zero components in these permutation
matrices as \(R^{(i)}\) matrices and therefore they will always commute
with the \(R\) matrices and also the \(\tilde{a}^{(i)}\) matrices defined for eqn (8). It is then straightforward to rearrange eqn (17) and (18) to show that the general, symmetric commutation relation of the interaction matrix is
\[R^{(i)}M^{(i)}R^{(j)}(\tilde{R}^{(j)}N^{(j)})^T = \tilde{R}^{(i)}N^{(j)}(\tilde{R}^{(j)}N^{(j)})^T\] (19)

In order to now consider the aggregate optical response of a
symmetric system we refer to the general expressions for the
extinction, absorption and scattering cross-sections of any
arbitrary system with dipole moments \(p\) and incident field \(e\)
(defined in eqn (7))
\[\sigma = \frac{k}{\epsilon_0}\text{Im}\{\langle e|p\rangle\}\] (20a)
\[\sigma = -\frac{k}{\epsilon_0}\text{Im}\left\{\frac{k^2}{6\pi}\langle p|p\rangle + \text{Im}\left\{\langle p|\tilde{a}|p\rangle\right\}\right\}\] (20b)
\[\sigma = \sigma_e - \sigma_a\] (20c)

It is then desirable to re-express all three cross-sections in
terms of the incident field state and three distinct matrices
using eqn (8)
\[
\begin{pmatrix}
\sigma_e = \text{Im}\{\langle e|M^{(i)}f^{(i)}|e\rangle\} \\
\sigma_a = \langle e|M^{(i)}f^{(i)}|e\rangle + \text{Im}\{\langle e|M^{(i)}f^{(i)}|e\rangle\} \\
\sigma = \sigma_e - \sigma_a
\end{pmatrix}
\]
(21a)
(21b)
(21c)

where we have defined the \(M^{(i)}\) matrices as
\[M^{(i)}f^{(i)} = \frac{k}{|E|^2} \left(\tilde{M}^{(i)}f^{(i)}\right)^{-1} \tilde{a}^{(i)}\] (22a)
\[M^{(i)}f^{(i)} = -\frac{k^2}{6\pi |E|^2} \left(\tilde{M}^{(i)}f^{(i)}\right)^{-1} \tilde{a}^{(i)}\] (22b)
\[M^{(i)}f^{(i)} = -\frac{k^2}{6\pi |E|^2} \left(\tilde{M}^{(i)}f^{(i)}\right)^{-1} \tilde{a}^{(i)}\] (22c)

Given that \(R^{(i)}f^{(i)}\) will commute with both the \(\tilde{M}^{(i)}\) and
\(\tilde{a}^{(i)}\) matrices, it therefore follows that \(R^{(i)}f^{(i)}\) must also
commute with any of the \(M^{(i)}\) matrices we have just defined.
That is to say all three inner products seen in eqn (21) can
ultimately be written in the one form
\[
\langle e|M^{(i)}f^{(i)}|e\rangle
\] (23)

where \(R^{(i)}f^{(i)}\) commutes with \(M^{(i)}\).

For this reason we can analyse the effect of the symmetric
commutation relation (eqn (19)) on all cross-sections at once
by considering an arbitrary \(M^{(i)}\) and evaluating eqn (23).
Moreover, to begin this analysis we separate the \(M^{(i)}\)
matrix into the quadrants that act between the electric and/or magnetic
fields
\[M^{(i)} = \begin{pmatrix} M^{(i)}_{11} & M^{(i)}_{12} \\
M^{(i)}_{21} & M^{(i)}_{22} \end{pmatrix} \] (24)

By doing this we can write the general inner product of eqn
(23) in terms of the individual electric and magnetic field states
\[
\langle e|M^{(i)}f^{(i)}|e\rangle = \langle e|M^{(i)}_{11}f^{(i)}|e\rangle + \langle e|M^{(i)}_{12}f^{(i)}|e\rangle + \langle e|M^{(i)}_{21}f^{(i)}|e\rangle + \langle e|M^{(i)}_{22}f^{(i)}|e\rangle
\] (25)

where \(|e\rangle = \begin{pmatrix} E_0^e \\
E_0^h \end{pmatrix}\) and \(|f^{(i)}\rangle = \begin{pmatrix} H_0^e \\
H_0^h \end{pmatrix}\)

This can be simplified further if we only consider incident
fields that are plane waves. Moreover, we can define a new
matrix, \(A^{(i)}\), that will introduce the appropriate phase differences
to relate the field at each particle to a single field vector of
the incident plane wave for the electric and magnetic
components. That is to say we define \(A^{(i)}\) as
\[A^{(i)} = \begin{pmatrix} a_{11}f^{(i)} \\
a_{12}f^{(i)} \\
a_{21}f^{(i)} \\
a_{22}f^{(i)} \end{pmatrix} \] (26)
such that $|e'\rangle = \hat{A}^{(3N)}|e_\alpha\rangle$ and $|h'\rangle = \hat{A}^{(3N)}|h_\alpha\rangle$ where $|e_\alpha\rangle = \begin{pmatrix} E_\alpha \\ \vdots \\ E_\alpha \end{pmatrix}$ and $|h_\alpha\rangle = \sqrt{\frac{c}{\hbar}} \begin{pmatrix} H_\alpha \\ \vdots \\ H_\alpha \end{pmatrix}$.

We are then able to rewrite eqn (25) in terms of a single incident field vector for both electric and magnetic fields

$$
\langle e|\hat{M}^{(3N)}|e\rangle = \langle e_\alpha| \begin{pmatrix} \hat{A}^{(3N)} \end{pmatrix}_{3\times1}^\dagger \begin{pmatrix} A^{(3N)} \end{pmatrix}_{3\times1} |e_\alpha\rangle 
+ \langle e_\alpha| \begin{pmatrix} A^{(3N)} \end{pmatrix}_{3\times1} \begin{pmatrix} \hat{A}^{(3N)} \end{pmatrix}_{3\times1}^\dagger |h_\alpha\rangle 
+ \langle h_\alpha| \begin{pmatrix} A^{(3N)} \end{pmatrix}_{3\times1}^\dagger \begin{pmatrix} \hat{A}^{(3N)} \end{pmatrix}_{3\times1} |e_\alpha\rangle 
+ \langle h_\alpha| \begin{pmatrix} \hat{A}^{(3N)} \end{pmatrix}_{3\times1} \begin{pmatrix} A^{(3N)} \end{pmatrix}_{3\times1}^\dagger |h_\alpha\rangle. 
$$

(27)

However, if we now express the four inner products in eqn (27) as sums, it becomes apparent that we can express the whole equation in terms of four 3 × 3 matrices

$$
\langle e|\hat{M}^{(3N)}|e\rangle = E_\alpha^e \sum_\delta \bar{E}^{(3N)}_\delta E_\delta^e + H_\delta^e \sum_\pi \bar{H}^{(3N)}_\pi E_\delta^e + E_\delta^e 
\times \sqrt{\frac{c}{\hbar}} \sum_\pi \bar{E}H^{(3N)}_{\pi \delta} H_\delta^e + H_\delta^e \sqrt{\frac{c}{\hbar}} \sum_\pi \bar{H}H^{(3N)}_{\pi \delta} H_\delta^e 
\Rightarrow \hat{M}^{(3N)}_\text{mat} \times \bar{E}H^{(3N)}_{\pi \delta} H_\delta^e = \hat{M}^{(3N)}_\text{mat} \times \bar{E}H^{(3N)}_{\pi \delta} H_\delta^e. 
$$

(28)

where we have denoted the 3 × 3 matrices that make up $\bar{E}E^{(3N)}$, $\bar{E}H^{(3N)}$, $\bar{H}E^{(3N)}$, and $\bar{H}H^{(3N)}$ according to row and column indices $E_{\alpha \beta}^{(3N)}$.

In summary, we have shown that any of the inner products from eqn (21) can be written in the form

$$
\langle e|\hat{M}^{(3N)}_f|e\rangle = E_\alpha^e \sum_\delta \bar{E}^{(3N)}_\delta \bar{E}H_{\pi \delta}^{(3N)} E_{\alpha \beta}^e + H_\delta^e \sum_\pi \bar{E}H_{\pi \delta}^{(3N)} H_{\alpha \beta}^e + E_{\alpha \beta}^e \sum_\delta \bar{E}E^{(3N)}_\delta H_{\alpha \beta}^e + H_{\alpha \beta}^e \sum_\delta \bar{H}E^{(3N)}_\delta H_{\alpha \beta}^e. 
$$

(29)

It is relatively straightforward to show that each $\hat{M}^{(3N)}_f$, $\hat{M}^{(3N)}_g$, $\hat{M}^{(3N)}_h$, and $\hat{M}^{(3N)}_i$ will commute with the symmetry operators $\hat{R}^{(3)}$. Given that each $\hat{M}^{(3N)}_f$ commuted with $\hat{R}^{(3)}$, specifically, because $\hat{R}^{(3N)}$ is constructed from four identical quadrants (see eqn (17)), it follows that each of the quadrants of $\hat{M}^{(3N)}$ must necessarily commute with $\hat{R}^{(3)}$. In other words

$$
\hat{R}^{(3N)} \hat{M}^{(3N)}_{f(i)} = \hat{M}^{(3N)}_{f(i)} \hat{R}^{(3N)}. 
$$

(30)

The transformation of each $\hat{M}^{(3N)}_{f(i)}$ with the $\hat{A}^{(3N)}$ matrix in eqn (27) does not affect the existing commutation relation given $\hat{A}^{(3N)}$ is diagonal and constructed of multiples of the 3 × 3 identity matrix and therefore commutes directly with both $\hat{R}^{(3N)}$ and $\hat{H}^{(3N)}$. The following sum over all 3 × 3 matrix components of $\bar{E}E^{(3N)}$, $\bar{E}H^{(3N)}$, $\bar{H}E^{(3N)}$, and $\bar{H}H^{(3N)}$ in eqn (28) will then absorb and ignore the permutation matrix leaving the commutation relation solely in terms of $\hat{R}^{(3)}$

$$
\hat{R}^{(3N)} \hat{R}^{(3)} = \hat{R}^{(3)} \hat{R}^{(3N)} \Rightarrow \hat{R}^{(3)} \sum_\delta \bar{E}E_{\delta} = \hat{R}^{(3)}. 
$$

(31)

As such the $\hat{M}^{(3N)}_f$, $\hat{M}^{(3N)}_g$, $\hat{M}^{(3N)}_h$, and $\hat{M}^{(3N)}_i$ matrices will all commute with the symmetry operators $\hat{R}^{(3)}$. From this point onward we will only be dealing with these 3 × 3 matrices, so can neglect the indices and other notation to write this commutation relation as simply

$$
\hat{R} \hat{M} = \hat{M} \hat{R}. 
$$

The commutation relation in eqn (31) can be used to deduce constraints on the given matrix, $\hat{M}$, depending on which symmetry group $\hat{R}$ corresponds to. In the following argument we will consider only the constraints arising from the $C_n$ group, however there will be analogous procedures for other symmetry groups. In any case, the elements of the $C_n$ group can be expressed as

$$
\{ \hat{I}, \hat{C}_n^1, \hat{C}_n^2, \ldots, \hat{C}_n^{n-1} \} 
$$

(32)

where $\hat{C}_n$ is a rotation about the symmetry axis of $2\pi/n$.

For practicality this group can also be represented in a matrix form with a Cartesian basis in which the z-basis vector is parallel to the symmetry axis. The general form of a matrix element in such a representation is expressed in terms of the irreducible representations of the $C_n$ group as

$$
\hat{R} \equiv \left( \begin{array}{c|c} \hat{U} & \begin{pmatrix} R_{11} \\ 0 \\ R_{22} \end{pmatrix} \\ \hline \begin{pmatrix} 0 \\ R_{22} \\ 0 \end{pmatrix} \end{array} \right). 
$$

(33)

where $R_{11}$ and $R_{22}$ are the first conjugate pair of one-dimensional (degenerate) irreducible representations ($E_1$, $R_{33}$ is the symmetric 1-dimensional irreducible representation ($A_1$) and $\hat{U}$ is a unitary matrix used to describe the appropriate similarity transform for a Cartesian basis.$^{40,41}$

$$
R_{11} = \{ 1, \omega, \omega^2, \ldots, \omega^{n-1} \}, \quad \omega = e^{i \frac{2\pi}{n}} 
$$

(34a)

$$
R_{22} = \bar{R}_{11} 
$$

(34b)

$$
R_{33} = \{ 1, 1, 1, \ldots, 1 \} 
$$

(34c)
It is important to acknowledge that this decomposition of \( \hat{R} \) in terms of distinct irreducible representations requires that \( n \geq 3 \) because there are precisely \( n \), 1-dimensional irreducible representations in the \( C_n \) group. That is to say the \( C_n \) symmetry group only has two such irreducible representations and subsequently a matrix representation of its operators can’t be expressed in the manner of eqn (33). To continue with the \( C_n \) \( n \geq 3 \) case; we now divide the \( M \) matrix in a manner corresponding to that done for \( \hat{R} \) in eqn (33)

\[
M = \begin{pmatrix}
\hat{M}_{11} & \hat{M}_{12} \\
\hat{M}_{21} & \hat{M}_{22}
\end{pmatrix}.
\] (35)

As such we can then write the commutation relation in eqn (31) as the following four equations

\[
R_{12} M_{22} = M_{22} R_{33}
\] (36a)

\[
\begin{pmatrix} R_{11} & 0 \\ 0 & R_{22} \end{pmatrix} \left( \hat{U} M_{12} \right) = \left( \hat{U} M_{12} \right) \begin{pmatrix} R_{11} & 0 \\ 0 & R_{22} \end{pmatrix}
\] (36b)

\[
\begin{pmatrix} R_{11} & 0 \\ 0 & R_{22} \end{pmatrix} \left( \hat{M}_{11} U^* \right) = \left( \hat{M}_{11} U^* \right) \begin{pmatrix} R_{11} & 0 \\ 0 & R_{22} \end{pmatrix}
\] (36c)

\[
\hat{U} \begin{pmatrix} R_{11} & 0 \\ 0 & R_{22} \end{pmatrix} \hat{U} R_{12} = R_{12} \hat{U} \begin{pmatrix} R_{11} & 0 \\ 0 & R_{22} \end{pmatrix} \hat{U}.
\] (36d)

Eqn (36a) is trivial as \( M_{22} \) and \( R_{12} \) are both scalars, hence there are no restrictions on \( M_{22} \) and we can just consider it as some scalar \( B \in \mathbb{C} \). Eqn (36b) and (36c) are not so trivial and describe four scalar relationships between irreducible representations of the form

\[
R_{i,a} = a R_{j,a}
\] (37)

where \( i \neq j, a \in \mathbb{C} \).

Noticeably, the only way non-trivial solutions to this sort of equation could exist is if the distinct irreducible representations were equal to each other. However it is obvious that distinct irreducible representations cannot be equal to each other by their definition (e.g. see eqn (34)) and hence the only valid solution is that both sides of eqn (36b) and (36c) are equal to zero. Then, given \( \hat{U} \) is invertible, we can conclude that both \( M_{11} \) and \( M_{12} \) must therefore consist of only zeros. The final equation from the commutation relation is then eqn (36d), which we can similarly rearrange to relate the \( R_{11} \) and \( R_{22} \) irreducible representations to each other

\[
\begin{pmatrix} R_{11} & 0 \\ 0 & R_{22} \end{pmatrix} U M_{11} U^* = U M_{11} U^* \begin{pmatrix} R_{11} & 0 \\ 0 & R_{22} \end{pmatrix}
\] (38)

We can see that the off-diagonal terms of \( M' \) must be zero as they produce a scalar relationship in the form of eqn (37) between \( R_{11} \) and \( R_{22} \). As it happens, this is the only constraint we can apply and subsequently \( M' \) will be of the form

\[
M' = \begin{pmatrix} a & 0 \\ 0 & b \end{pmatrix}
\] (39)

where \( a, b \in \mathbb{C} \).

We can then get the corresponding \( M' \) directly from the definition of \( M' \) in eqn (38)

\[
M_{11} = M' U^* \hat{U} = \frac{1}{2} \left( \begin{array}{cc} a + b & (a - b)i \\ (b - a)i & a + b \end{array} \right) = \begin{pmatrix} A & C \\ -C & A \end{pmatrix}
\] (40)

where \( A, C \in \mathbb{C} \).

In conclusion each matrix from eqn (29) in a system with \( C_n \) symmetry will be of the form

\[
M = \begin{pmatrix} A & C \\ -C & A \end{pmatrix}
\] (41)

where \( A, B, C \in \mathbb{C} \).

However, if the incident field is propagating in the direction of the symmetry axis (\( E_{x1}^0, E_{y1}^0 = 0 \) then we can relate \( H_0 \) to \( E_0 \) as

\[
H_0^p = \frac{\varepsilon_0}{\mu_0} \begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} E_0^0
\] (42)

The combination of electric and magnetic inner products in eqn (29) can then be reduced to just a single, purely electric, inner product

\[
\langle e | M^{(0N)} | e \rangle = E_{x1}^0 M_{x1}^0 E_0 + H_{x1}^0 M_{x1}^0 H_0 + E_{y1}^0 M_{y1}^0 H_0 + H_{y1}^0 M_{y1}^0 H_0
\] (43)

\[
= E_1^0 \left( M_{x1}^0 + \frac{\varepsilon_0}{\mu_0} M_{y1}^0 \right) E_0^0
\]

Here the double-prime indicates that the matrix has been multiplied by the matrix seen in eqn (42), which noticeably commutes with- and does not change the general form of any matrix defined as per eqn (41). Therefore, with no z-component of the incident field and a single matrix in the form of eqn (41), any of the inner products used to define the cross-sections in eqn (21) can be written as

\[
\langle e | M^{(0N)} | e \rangle = A_E |E_1|^2 + C_E |E_1|^2
\] (44)

where \( A_E, C_E \in \mathbb{C} \).

In this paper we consider only linearly polarised light and so the \( E_z \) and \( E_y \) components are in phase. As such, the term proportional to \( C_E \) in eqn (44) can be removed and we are left with

\[
\langle e | M^{(0N)} | e \rangle = A_E |E_1|^2 + |E_1|^2
\] (45)

The cases of other polarisations will be addressed in another paper. However, for linearly polarised light, the expressions for...
the cross-sections in eqn (21) can be simplified using eqn (45) to become

\[ \sigma_v = \text{Im}\{A_1\sqrt{|E_x|^2 + |E_y|^2}\} \quad (46a) \]

\[ \sigma_s = (A_2 + \text{Im}\{A_3\sqrt{|E_x|^2 + |E_y|^2}\}) \quad (46b) \]

\[ \sigma_a = \sigma_v - \sigma_s = (\text{Im}\{A_1 - A_3\sqrt{|E_x|^2 + |E_y|^2}\}) \quad (46c) \]

Noticeably this shows that all the cross-sections are independent of polarisation. Specifically, we have shown that any system with at least 3-fold cyclic symmetry (C3) will have polarisation-independent extinction, scattering and absorption cross-sections for linearly polarised plane waves traveling parallel to the symmetry axis.

While this is the main result of the paper, it is also worth noting that the derivations of eqn (29) and (31) are for arbitrary symmetry operations and therefore provide a foundation on which to evaluate cross-sections for operations (R) corresponding to different symmetries. In this way it is possible to, for instance, consider the tetrahedral (T) or pyramidal (Cnv) symmetry groups, both of which can be handled by directly applying Schur’s Lemma applied to Schur’s symmetry groups, both of which can be handled by directly applying Schur’s Lemma to show isotropic and axial polarisation-independent cross-sections.

4 Examples of light scattering

To demonstrate the validity of our approach we employed two methods to study the light scattering by some oligomer structures with n-fold symmetries [see Fig. 2-4]. Firstly we used CST Microwave Studio to calculate the exact total extinction, scattering and absorption cross-sections as well as the near-field profiles of the corresponding structures at resonance. And, secondly, we employed the dipole approximation and dyadic Green’s function method to obtain the cross-sections and the distribution of optically-induced electric and magnetic dipoles in the individual nanoparticles.\(^\text{42}\) Fig. 2 shows the extinction, scattering and absorption cross-sections of a trimer consisting of three silicon nanospheres with 3-fold symmetry. Fig. 3 shows the extinction, scattering and absorption cross-sections for a heptamer consisting of seven gold nanospheres with 6-fold symmetry. All particles of these oligomer-like structures are in the same transverse plane, so the excitation field is identical for all particles. We can also lower the symmetry of a structure by shifting some particles along the propagation axis. For example, in Fig. 4 we present a structure with 3-fold symmetry which was derived from a gold heptamer structure shown in Fig. 3. To construct the new structure we shifted two equilateral trimers of the outer ring in opposite directions from the central particle evenly spaced along the propagation axis, and then twisted each with respect to the other. The final structure is then chiral with 3-fold symmetry. Thus, according to our theoretical prediction, we expect that it should be polarisation-invariant. We used Palik’s data for permittivity of the various materials.\(^\text{44}\) All the presented results support our derivation that these structures will exhibit polarisation-independent optical properties for any incident polarisation angle, which does not necessarily coincide with the rotational symmetry of the structures. These figures also show that the total absorption is polarisation-independent even though the near-field distribution varies with the incident polarisation. It allows us to conclude that, although all structures exhibit some degree of geometrical anisotropy, their optical response is isotropic. And the only requirement that we impose is that the structure supports n-fold symmetry with \(n \geq 3\). For completeness we also acknowledge that structures with only \(C_2\) symmetry are known to be polarisation-dependent\(^\text{43}\) and therefore can conclude that \(n \geq 3\) is a requirement for the n-fold symmetry.

Finally, based on the coupled dipole approximation method,\(^\text{44}\) the optical response of structures with arbitrary geometries and complex refractive index can be approximated by an ensemble of discrete dipoles. Thus, our results can be
easily generalised to any structure with n-fold symmetry. Fig. 5 shows the results of direct numerical simulations of a continuous structure with 3-fold symmetry, which, for simplicity, is modeled as $r(\theta) = R[1 + \cos(1.5\theta)]$ with $R = 200$ nm (on the transverse plane) and $h = 100$ nm (along longitudinal direction) and is made of gold. It still exhibits polarisation-independent optical response, in full agreement with our approach above. This proves that our statement is quite universal and can be applied to any system.

5 Conclusions

We have studied the optical response of nanoparticle structures with an n-fold ($n \geq 3$) rotational symmetry excited by an incident plane wave propagating parallel to the symmetry axis. We have demonstrated that polarisation-independent responses (in terms of the cross-section of scattering, absorption and
extinction) can come solely from the overall rotational symmetry of a structure without any condition placed on other elements of a given system. We have presented specific examples which support our general theory. Such robust polarisation-independent features are expected to play an important role in various applications including nanoantennas, sensing, imaging, solar cells, and other applications in chemistry, biology, and medicine.

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A.2 Revisiting the physics of Fano resonances for nanoparticle oligomers

Summary

The interference phenomena known as Fano resonances generated significant interest in metal nanoparticle oligomers, where they offered characteristically thin spectral lineshapes and significant field enhancement. Yet Fano resonances were predicted to occur also in all-dielectric oligomers [95], which suggested a mechanism other than plasmonic hybridisation of modes was able to lead to Fano resonances. In this work, we revisit the optical responses from general nanoparticle oligomers and acknowledge the non-Hermitian interactions, which allows us to rigorously show that Fano resonances can be described purely from the overlap interference between nonorthogonal collective eigenmodes. This unifies the understanding of Fano resonances in both plasmonic and dielectric nanoparticle oligomers, and demonstrates that low-loss dielectric oligomers can provide comparable outcomes to what had been an exclusively plasmonic pursuit.

Notes and errata

- The expression for the extinction cross section in equation (23) is missing a factor of $\mu_0$, the correct expression is (2.31).

- The discussion of FIG. 1 suggests a maximum of four, doubly degenerate, eigenmodes can be excited by a plane wave normally incident on an rotationally symmetric ring of three or more electric and magnetic dipole pairs. This is true provided one does not define eigenmodes as simultaneously containing both electric and magnetic dipoles, whereupon coupling between electric and magnetic dipoles allows these dipoles to be oriented parallel to the principal rotation axis. It subsequently follows that more than four eigenmodes, when defined according to the discussion in Chapter 3, can be excited by a normal-incidence plane wave. However, the related derivation of the maximum two, doubly degenerate, eigenmodes being excitable for each rotationally symmetric ring of three or more electric xor magnetic dipoles, following equations (2)-(10), remains unchanged.

- The curl of the dyadic Green’s function is a factor of $ik$ larger than it should be, however all equations are correct once making the substitution: $\nabla \times \hat{G}^0(r_i, r_j) \rightarrow \frac{1}{4\pi} \nabla \times \tilde{G}^0(r_i, r_j)$. 
Revisiting the physics of Fano resonances for nanoparticle oligomers

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We present a robust approach for interpreting the physics of Fano resonances in planar oligomer structures of both metallic and dielectric nanoparticles. We reveal a key mechanism for Fano resonances by demonstrating that such resonances can be generated purely from the interference of nonorthogonal collective eigenmodes, which are clearly identified based on the coupled-dipole approximation. We prove analytically a general theorem to identify the number of collective eigenmodes that can be excited in ring-type nanoparticle oligomers and further demonstrate that no dark-mode excitation is necessary for the existence of Fano resonances in symmetric oligomers. As a consequence, we unify the understanding of Fano resonances for both plasmonic and all-dielectric oligomers.

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I. INTRODUCTION

Recently, a lot of attention has been paid to the physics of Fano resonances [1] in nanoscale oligomer structures composed of plasmonic nanoparticles. The current understanding of Fano resonances in these symmetric oligomers relies on an interference of super- and subradiant collective modes, such as those created from the interaction between a symmetric ring of nanoparticles and a single central nanoparticle [2–4]. With very few exceptions [5,6], this interference is described for specific plasmonic oligomers, where a directly excited superradiant mode interferes destructively with an indirectly excited “dark mode” (or trapped mode). By dark mode we refer to any mode that cannot couple directly with an incident plane wave. More formally, when given an oligomer’s symmetry group, if the irreducible representation of a particular mode is mutually exclusive to the irreducible (or reducible) representation of the incident field, then that mode cannot be excited by the incident field, and it is defined as dark [7]. However, we use a common extension to this definition and also consider dark modes as being modes whose projection onto the incident field is zero. Thus, such dark modes cannot couple directly to the incident field, although they may transform according to the same irreducible representation as the incident field. The current understanding is that the latter form of dark modes can still be excited in symmetric oligomers through the coupling to a bright plasmonic mode via near-field hybridization [3,8–19]. While the concept of dark modes inducing Fano resonances in oligomer structures has been discussed in many studies [20–23], the associated definition of the system’s modes is deduced from molecular analogues that do not fully resemble the electromagnetic coupling in oligomer systems. For this reason, such modes do not represent the real eigenmodes of the oligomer system. This observation has led to the necessary distinction between bare (approximate) and dressed (real) modes in the existing literature when regarding plasmon hybridization from the perspective of quantum optics [5,14,24,25].

Recently, it was predicted that Fano resonances should also occur in all-dielectric symmetric oligomers [26] despite the absence of the necessary hybridization of modes required to excite dark modes. As such, this development calls for the study of different mechanisms that also lead to Fano-type interference and resonances.

By revisiting the mechanisms that underpin the general optical properties of oligomer structures, in this paper we present a general symmetry theory for analyzing their electric and magnetic responses. We show that generic oligomer structures can be characterized entirely in terms of distinct electric and magnetic collective eigenmodes. In this approach, we are able to unify both plasmonic and all-dielectric oligomers. We are then able to build upon work by Frimmer et al. [5] to show conclusively that Fano resonances created in symmetric oligomers can be explained entirely in terms of interference of these eigenmodes. Importantly, this theory does not rely on the hybridization of modes, and any mode can only be reliably excited if it couples with an incident field. Therefore, only bright eigenmodes will be excited except in accidental cases. To demonstrate this finding, we revisit experimental results that demonstrated Fano resonances in gold heptamers and show that they occur purely due to the interference of the system’s bright eigenmodes and do not involve dark-mode interference, with which they were previously attributed.

We are further able to address the types of oligomer systems that can support Fano resonances on the basis that two or more nondegenerate eigenmodes must be excited to permit an interference. We prove that the response of any symmetric ring-type oligomer consisting of arbitrarily many nanoparticles will be described by precisely two nondegenerate eigenmodes. Subsequently, we are able to conclude and numerically demonstrate that ring-type oligomers can support Fano resonances as well. This idea seems to contradict current understanding of the origin of Fano resonances in oligomers, where it is typically shown that the removal of a central particle should destroy the resonance [20,27]. The common practice of adding a central particle to an oligomer system is then reevaluated as simply increasing the number of (excitable) nondegenerate eigenmodes in the nanoparticle system.
To amplify the full implications of this eigenmode theory, we also consider all-dielectric oligomers where the individual constituent particles have nontrivial electric and magnetic responses [28–32]. We provide formal and rigorous definitions of the distinct electric and magnetic eigenmodes present in these systems. As such, we are able to precisely define and justify the existence of purely magnetic Fano resonances in terms of the physics discussed in this paper. Subsequently, there is immediate relevance of this theory to the emerging field of all-dielectric nanophotonics as it provides an explicit theoretical framework for analyzing both electric and magnetic responses in oligomers. However, it is also worth acknowledging that the generality of our approach makes it important to broader fields, including molecular optics and antenna design.

II. EIGENMODE ANALYSIS OF OLIGOMERS

The modes of plasmonic oligomer structures are typically understood based on their electric dipole moments or surface charge distributions [14,16,19], and therefore, the analysis of these modes can be carried out from the perspective of the coupled-dipole approximation [33–35]. In this approximation, the incident field \(|E_0\rangle\) can be linked to the induced dipole moments \(|p\rangle\) using an interaction matrix \(M\):

\[
|E_0\rangle = M |p\rangle.
\]

Here the use of the bra-ket notation for the incident field and dipole moments is simply to emphasize that each state is the concatenation of all the dipole-moment vectors in the given system. In the following, we derive constraints for a general oligomer’s basis vectors and then eigenmodes using the group theory. In this regard, it has previously been shown that the interaction matrix must transform according to the symmetry group of the corresponding system’s geometry [36]. The particular symmetry group of the oligomers we are interested in is \(D_{nh}\), where \(N\) is arbitrary. Therefore, the components of a normally incident electric field \(E_{0,x}\) and \(E_{0,y}\) transform according to the irreducible representation \(E \equiv E_i^\dagger (E_{i,x})\) for odd (even) \(N\) [37]. Then we consider the in-plane components of the electric dipole moments as transforming according to some reducible representation \(P\). Formally, the number of nondegenerate eigenmodes interacting with the electric field is given by the number of times the irreducible representation \(E\) is contained in \(P\) [37]. From this, we can deduce the dimension of the space that spans all responses of the system to such an incident field, given that each associated eigenmode is doubly degenerate because \(E\) has a rank of 2.

Let’s consider a plane wave that is normally incident on an oligomer that contains a symmetric ring of \(N\) particles, the particles of which are located at the points

\[
\begin{align*}
\mathbf{r}_i &= -r \sin \varphi_i \mathbf{e}_x + r \cos \varphi_i \mathbf{e}_y, \\
\varphi_i &= \frac{2\pi}{N} (i - 1), \quad i = 1, \ldots, N.
\end{align*}
\]

The basis vectors for the response of this ring can then be determined by using the projection-operator technique [37,38]. Moreover, we apply all projection operators to some initial state \(|e\rangle\) in order to produce different linear combinations of all the basis vectors associated with the irreducible representation \(E\) and subsequently deduce the dimension of space of the corresponding responses. Now we note that a dipole moment at the location of a particular particle is moved through every location on the ring by symmetric rotations. As such, an initial state that has a dipole moment at only one location on the ring must be linearly independent from all its symmetrically rotated versions. By extrapolation, two such initial states that occupy the one location and are linearly independent will, in conjunction with their respective symmetry-rotated versions, form a basis for a \(2N\)-dimensional space. However, we are only considering responses that transform according to an irreducible representation and are thereby neglecting the \(z\) direction; hence, the space of responses cannot have more than \(2N\) dimensions. In other words, any response of the system that transforms according to \(E\) will be spanned by the projections of these two initial states. To this end we can define the initial state \(|e\rangle\) to have only one nonzero dipole moment at the first particle and then apply the projection operators [39] to obtain the subsequent polarizations of the other dipoles. For the symmetry operations \(g\) and their matrix representations \(\hat{D}(g)\), the projection operator is defined as

\[
P_{\mu \nu}^{(E)} = \sum_{g} D_{\mu \nu}^{(E)}(g) g.
\]

where we neglect normalization. The idea is that the operator \(P\) acting upon any state \(|p\rangle\) will yield either a zero or a linear combination of basis vectors that transform according to the vector representation \(E\). Hence, we can start with the formal definition of our test state

\[
|e\rangle \equiv \left( e, 0, \ldots, 0 \right), \quad e = (e_x, e_y, 0).
\]

Here \(e_x\) and \(e_y\) are arbitrary in order to produce two linearly independent initial states by varying the polarization of the vector \(e\). All possible basis vectors can then be deduced by varying the indices \(\mu\) and \(\nu\) in \(P_{\mu \nu}^{(E)}\). For the following we also introduce the notation to extract the \(i\)th dipole moment from a state

\[
|\{p\}\rangle_i \equiv p_{i, x} e_x + p_{i, y} e_y, \quad i = 1, \ldots, N.
\]

We have chosen to neglect the \(z\) direction given that there is no operation in \(E\) that transforms an \(x\)- or \(y\)-polarized dipole moment onto the \(z\) direction. Now, because there is only a limited set of symmetry operators that will transform the first particle’s dipole moment onto that of the \(i\)th particle, it can be checked using the full nonlocal definitions [36] of each \(g\) that

\[
[P_{\mu \nu}^{(E)} |e\rangle] = \hat{P}_{\mu \nu} [\hat{D}(R_1) \hat{D}(\sigma_z)]_{\mu \nu} R_{1, x} R_{1, y} + \hat{D}(R_1)_{\mu \nu} R_{1, z},
\]

where \(R_1\) is a local rotation by the angle \(\psi_i\), \(\sigma_z\) is a local reflection about the \(yz\) plane, and their matrix representations are

\[
\hat{D}(R_1) = \left( \begin{array}{cc} \cos \varphi_i & -\sin \varphi_i \\ \sin \varphi_i & \cos \varphi_i \end{array} \right), \quad \hat{D}(\sigma_z) = \left( \begin{array}{cc} -1 & 0 \\ 0 & 1 \end{array} \right).
\]

Noticing, in neglecting the \(z\) direction we have not considered the \(R_1 \sigma_x\) (also known as \(S_z\)) or \(R_1 C_x\) operations; however, these will be described by exactly the same symmetry operations and matrix representations as \(R_1\) and \(R_1 \sigma_z\), respectively, for \(E\). For this reason, these extra terms can be neglected by normalization. In summary, Eq. (6) is just a way to write the
components of the operated states obtained by acting with $P_{\mu,i}$ upon $|\epsilon\rangle$. However, it allows us to obtain explicit forms for the matrix $P_{\nu,j}$.

$$P_{11,j} = \begin{pmatrix} 2 \cos^2 \psi_j & 0 \\ 2 \cos \psi_j \sin \psi_j & 0 \end{pmatrix} \quad (8a)$$

$$P_{12,j} = \begin{pmatrix} 0 & 2 \sin^2 \psi_j \\ 2 \cos \psi_j \sin \psi_j & 0 \end{pmatrix} \quad (8b)$$

$$P_{21,j} = \begin{pmatrix} 2 \cos \psi_j \sin \psi_j & 0 \\ 2 \sin^2 \psi_j & 0 \end{pmatrix} \quad (8c)$$

$$P_{22,j} = \begin{pmatrix} 0 & 2 \cos \psi_j \sin \psi_j \\ 2 \cos \psi_j \sin \psi_j & 0 \end{pmatrix} \quad (8d)$$

Evaluating Eq. (6) then yields only four nonzero combinations:

$$P_{11,j} \epsilon_i = \begin{pmatrix} 2 \cos^2 \psi_i \\ 2 \cos \psi_i \sin \psi_i \end{pmatrix} \quad (9a)$$

$$P_{21,j} \epsilon_i = \begin{pmatrix} 2 \cos \psi_i \sin \psi_i \\ 2 \sin^2 \psi_i \end{pmatrix} \quad (9b)$$

$$P_{12,j} \epsilon_i = \begin{pmatrix} 2 \sin^2 \psi_i \\ -2 \cos \psi_i \sin \psi_i \end{pmatrix} \quad (9c)$$

$$P_{22,j} \epsilon_i = \begin{pmatrix} -2 \cos \psi_i \sin \psi_i \\ 2 \cos^2 \psi_i \end{pmatrix} \quad (9d)$$

In this sense, we have shown that these four basis vectors span all responses of the system that transform according to the irreducible representation $E$. For later reference, it is also worth noting that this approach will also hold for a magnetic incident field and the associated responses, where $E \equiv E_0^z (E_1)$ for even (odd) $N$. This would be done by simply substituting in a negative version of $\sigma_z$ (and $\sigma_y$) and following the exact same argument as just presented here. In any case, we have the freedom to create a more intuitive set of basis vectors from linear combinations of the basis vectors presented in Eq. (8) [40]:

$$P_1^{(1x)} = \frac{1}{2N} (P_{11,j} \epsilon_i + P_{12,j} \epsilon_i) = \frac{\epsilon_i}{\sqrt{N}} \quad (10a)$$

$$P_1^{(1y)} = \frac{1}{2N} (P_{21,j} \epsilon_i + P_{22,j} \epsilon_i) = \frac{\epsilon_i}{\sqrt{N}} \quad (10b)$$

$$P_1^{(2x)} = \frac{1}{2N} (P_{11,j} \epsilon_i - P_{12,j} \epsilon_i)$$

$$= \frac{1}{\sqrt{N}} \left[ \cos 2\psi_i \epsilon_i + \sin 2\psi_i \epsilon_i \right] \quad (10c)$$

$$P_1^{(2y)} = \frac{1}{2N} (P_{21,j} \epsilon_i - P_{22,j} \epsilon_i)$$

$$= \frac{1}{\sqrt{N}} \left[ \sin 2\psi_i \epsilon_i - \cos 2\psi_i \epsilon_i \right] \quad (10d)$$

It is then relatively straightforward to see that mode $1x$ only couples with the $x$-polarized wave and, similarly, mode $1y$ only couples with the $y$-polarized wave, and modes $2x$, $2y$ do not couple with the incident field at all; in that sense we can identify them as dark modes.

At this point we would like to acknowledge that we have proven an important result: any response from a symmetric ring of particles that transforms according to $E$ (e.g., those excited by a normal-incidence plane wave) will be spanned by explicitly four linearly independent basis vectors. In regard to eigenmodes, this means that any such response will be described by no more than two doubly degenerate eigenmodes, irrespective of how many particles the ring contains. This is a generalization of previous results obtained for particular values of $N$ [12]. Moreover, we have shown here that oligomers made of multiple rings contain two doubly degenerate eigenmodes per ring. This agrees with the existing studies of mechanical vibrations of the molecules. For instance, the benzene molecule has two $D_6h$ rings made of carbon and hydrogen atoms and four (doubly degenerate) in-plane vibration modes in total [41]. We can also consider the effect of a single central particle, the response of which will be described by a single dipole moment vector at the origin. Such an electric dipole moment must always transform according to $E$ (when neglecting the $z$ direction once again), and therefore, the space of the associated responses will increase by two, which adds one more doubly degenerate eigenmode. So for a given polarization of the incident wave, the number of modes that can be excited in oligomers consisting of one ring without (with) a central particle is restricted to only two (three) modes. This is, counterintuitively, not dependent on the number of particles in the ring that make up the nanoparticle system. We stress here that the approach up to now has not identified the eigenmodes of the system. What is presented in Eq. (10) was obtained based solely on the symmetry considerations and served to provide basis vectors for the response. Equation (10) is, in fact, a generalization of the existing understanding of modes in oligomers, and it shows that mode hybridization is required to excite the dark modes ($2x$ and $2y$). The opposing argument as to why these deduced dark modes actually cannot exist in the real system of eigenmodes is then somewhat subtle. The key is that the symmetry approach produces orthogonal modes by definition, whereas the interaction matrix [see Eq. (21) in the next section], which describes the coupling between the dipole moments of each particle, is non-Hermitian [42]. As such, the eigenmodes of the system are not orthogonal except in accidental cases. This means that the space spanned by the dark basis vectors ($2x$ and $2y$) can be spanned by nonorthogonal bright vectors in the real eigenmode system. In this sense the projection operator approach has forced orthogonality onto the space of the system’s responses and found dark basis vectors by necessity. Moreover, an orthogonal basis which includes the incident-field vectors ($1x$ and $1y$) requires that the remaining basis vectors be orthogonal to the incident field. Admittedly, this does not explicitly rule out the existence of dark eigenmodes, which transform according to $E$, as we discuss in the following section.

### III. EXAMPLES OF NANOPARTICLE OLIGOMERS

In what follows, we present numerical simulations of the excitation of electric and magnetic eigenmodes in different plasmonic and all-dielectric oligomer structures to demonstrate the application of our theory. Calculations are performed.
numerically using the coupled electric and magnetic dipole approximation [33], where each constituent particle in an oligomer is modeled as a single electric and magnetic dipole. A full description of this model, including the definition of the electric and magnetic eigenmodes, is provided in the next section. Here we focus on the implications of the arguments provided in the previous section. To first illustrate the arguments on the number of eigenmodes that transform according to \( E \) in a ring-type oligomer, we consider the simplest system that meets the associated symmetry requirements: a \textit{trimer} with \( D_3 \) symmetry. In Fig. 1, we show the extinction spectra of plasmonic (silver) and dielectric (silicon) trimers with their associated decomposition into both electric and magnetic eigenmodes (the definitions of electric and magnetic eigenmodes are provided in the following section). In doing this decomposition, we notice that the magnetic modes have a negligible role in the plasmonic trimer, whereas both electric and magnetic eigenmodes are excited in the dielectric trimer. In both cases, there are only two eigenmodes ever excited by the incident field for the electric and magnetic cases, which is in full agreement with our theory. The reason why two, rather than four, eigenmodes are important for both electric and magnetic responses is that the polarization of the nondegenerate eigenmodes is defined to match the polarization of the incident field, and hence, we only excite one eigenmode in each doubly degenerate eigenspace.

We start with a silver trimer, supporting a not-so-pronounced Fano resonance at the intersection of the two electric eigenmodes. In other words, the Fano resonance occurring at the point where two eigenmodes have equal magnitude suggests that the interference of eigenmodes plays an important role, especially given the electric dipole moment profiles in Fig. 1(c) show that these eigenmodes are out of phase with each other. Similarly, for the silicon trimer, there exists a well-pronounced Fano line shape, but this time at the equivalent intersection of two magnetic eigenmodes. Noticeably, the increase of the magnetic response also corresponds to a reduction in the overall electric response, which is due to the coupling between electric and magnetic dipoles. Further intricacies of the dielectric system are apparent in that there is also a (smaller) secondary interference line shape occurring at the peak of the subradiant electric eigenmode, which interferes with the superradiant electric eigenmode. That is to say, there appears to be both electric and magnetic Fano-like interference occurring in one trimer.

Additionally, it is also worth noting that there exists an apparent relationship between the location of the single-particle resonance and the location of the Fano resonance. This is at least intuitive given that the subradiant modes (modes ii and iv) in Figs. 1(c) and 1(f) have a comparatively low projection onto the incident field, and subsequently, their excitation is significantly dependent on interparticle coupling. As such, the single-particle resonance represents a wavelength where the field radiating from each individual particle as a direct response to the incident field is maximized, and it is therefore not surprising to see more significant excitation of the subradiant modes.

To demonstrate that the number of nondegenerate eigenmodes that can be excited by a normal-incidence plane wave is fixed independently of the number of particles in the given ring,
of two eigenmodes. However, to fully justify the claim
that the Fano resonance results from the interference of
these eigenmodes, the extinction cross section in Fig. 2(a)
is decomposed into the components that come from each
eigenmode and also the component coming from the interference
between them. This interference term is a consequence of the
eigenmodes not being orthogonal, a point which we can illustrate by considering an incident field written in terms of
our two nondegenerate eigenmodes,

\[ |E_0⟩ = a_1|v_1⟩ + a_2|v_2⟩ \]  
\[ |p⟩ = a_1λ_1|v_1⟩ + a_2λ_2|v_2⟩. \]

Here the coefficients of each eigenmode in the induced dipole
moment solution (i.e., the \(a_1λ_1\) terms) are the eigenmode
amplitudes that are plotted in Figs. 1 and 2; they represent the
magnitude of excitation for each eigenmode. However, the

extinction cross section \([35]\) is proportional to

\[ σ_e \propto \text{Im}⟨|E_0⟩|p⟩⟩, \]  
\[ ⟨E_0|p⟩ = ⟨a_1^*|v_1⟩ + a_2^*|v_2⟩(a_1λ_1|v_1⟩ + a_2λ_2|v_2⟩) \]  
\[ = |a_1|^2λ_1|v_1|^2 + |a_2|^2λ_2|v_2|^2 \]  
\[ + a_1^*a_2λ_2|v_1⟩|v_2⟩ + a_1^*a_1λ_1|v_1⟩|v_1⟩. \]

In this expression, the “direct terms” correspond to the
collection of each eigenmode to the extinction without
accounting for the interplay between eigenmodes (the “interference terms”). It is precisely the direct terms for each
eigenmode and the sum of interference terms seen in Eq. (12c)
that are presented in Fig. 2(a) for the silver nanosphere octamer.
It is important to note that each of the direct terms can produce
only a positive contribution to the extinction as the direct terms
represent the extinction in the case where the incident field has
been structured to consist of only the one given eigenmode.
Indeed, if this were not the case, the energy conservation
law would be violated. However, as seen in Fig. 2(a), the
interference terms can lead to a negative contribution to
extinction. Such a negative contribution has been observed previously \([5]\); however, it is important that it is the interference
alone that can produce a negative extinction component. In
other words, no eigenmode can feed energy into the incident
field. It is also worth noting that if we were to have an
orthogonal basis of eigenmodes, the interference terms would
all go to zero, as is the case with analogous inner products in
quantum mechanics and other Hermitian spaces. In this
sense, we can conclude that the Fano line shape is not a result
of energy coupling into a nonradiative dark mode, but rather
the interference between the two eigenmodes. Additionally, a
further effect of a nonorthogonal basis of eigenmodes is that
the decomposition of the incident field into eigenmodes has to
be solved through a set of coupled equations. Specifically, if we were to consider the decomposition of some incident field
in terms of eigenmodes

\[ |E_0⟩ = \sum_i a_i|v_i⟩, \]  
\[ \langle v_j|E_0⟩ = \sum_i a_i|v_j⟩|v_i⟩ \quad ∀ j = 1, \ldots, N. \]

In this way, it is worth noting that even if the eigenmodes are
themselves decoupled, their excitations are coupled. In regard
to the possibility of dark eigenmodes, Eq. (14) means that
an eigenmode can be excited even if it has a zero projection
onto the incident field (i.e., if it is dark). In this case we can
actually use the derived basis vectors in Eq. (10) to get an
explicit expression for the dark eigenmode of the arbitrary
system. Moreover, because an arbitrary eigenmode of a ring-
type oligomer can be written as a linear combination of the
set of orthogonal basis vectors provided in Eq. (10), a dark
eigenmode having a zero projection onto both 1x and 1y (i.e.,
being also the x- and y-polarized incident field) ensures that
it is expressible entirely in terms of the remaining two basis
vectors: $2x$ and $2y$. Then, given that such an eigenmode is doubly degenerate because it transforms according to $E$, we know it must have a two-dimensional eigenspace and that there will subsequently be two orthogonal eigenmodes that span this dark eigenspace. In other words, if there is a dark eigenmode, then the basis vectors $2x$ and $2y$ from Eq. (10) will span precisely the associated dark eigenspace, and therefore these two basis vectors will be the dark eigenmodes of a ring-type oligomer. However, it is nonetheless worth noting that the existence of dark modes that transform according to $E$ is not guaranteed in any oligomer, and it is unlikely that they would be frequency independent as $2x$ and $2y$ are. Thus, dark eigenmodes would be isolated much like the pure superradiant eigenmodes (i.e., making the incident field an eigenmode), which are the other basis vectors presented in Eq. (10). In this regard, all numerically calculated eigenmodes in this paper are explicitly bright at all calculated frequencies.

To demonstrate the applicability of this eigenmode theory to experiment, we reconsider the gold heptamer made by Hentschel et al. [20] in Fig. 3. This investigation observed a Fano resonance experimentally and associated it with coupling into a dark mode. However, based on our analysis, one can explicitly see that such Fano resonances are described by the inference of two bright eigenmodes. Moreover, Fig. 3(b) shows the eigenmode decomposition of the system’s response (solid line) and also the projections of each eigenmode onto the incident electric field (dashed line), which are all nonzero.

This is not the same as the previously supported explanation in a number of papers, which attributed the Fano resonance to a bright mode with a dark mode. It is worth noting that the bright modes from our simulation appear to have dipole moment profiles that qualitatively match those of the modes and dark modes proposed by Fan et al. [16] for a similar gold ring heptamer structure. The key point of our analysis applied to this structure is therefore not that the mode profiles are largely different, but rather that the true modes are bright instead of dark. However, we can conclude that the Fano resonance in this system is explicitly due to the interference between bright modes only, and it does not involve any dark mode.

IV. DIELECTRIC OLIGOMERS

The electric dipole response is known to be the dominant contribution to the optical properties of a single spherical particle as its size is made arbitrarily small, the so-called Rayleigh approximation. This is the principle on which the coupled electric dipole approximation is based [35]. However, for high-refractive-index dielectric materials such a situation can change. In particular, when the wavelength inside a particle matches its diameter, a magnetic dipole mode can be excited due to resonant excitation of the circulating displacement current. In this way it is possible for the magnetic dipole response of a dielectric particle to be on the same order of magnitude as the electric dipole response [33]. Importantly, provided the particle also remains small compared to the wavelength, it will also be substantially larger than its electric quadrupole response, which is otherwise the next dominant order of multipole after the electric dipole response in plasmonic materials [44]. Therefore, in such a regime, it is necessary to take into account the magnetic dipole responses for high-refractive-index dielectric materials, and subsequently, an all-dielectric oligomer requires full treatment of both electric and magnetic dipole moments to characterize its behavior. In the following, we show that the electric and magnetic responses of symmetric oligomers can be considered independently, with distinct electric and magnetic eigenmodes as opposed to combined electromagnetic eigenmodes.

To begin, the general expressions for the electric and magnetic dipole moments of the $i$th particle in an oligomer can be defined using the coupled electric and magnetic dipole equations [33].

$$\mathbf{p}_i = \alpha_{E,i} E_0 \mathbf{r}_0 + \alpha_{E,i} E_0 k^2 \left( \sum_{j \neq i} \frac{1}{\epsilon_0} G_0^\rm{E}(\mathbf{r}_i, \mathbf{r}_j) \mathbf{p}_j \right) - \frac{m_i}{\epsilon_0} \nabla \times \hat{G}_0^\rm{M}(\mathbf{r}_i, \mathbf{r}_j) \mathbf{m},$$  

(15a)
where \( \mathbf{p}_i \) (\( \mathbf{m}_i \)) is the electric (magnetic) dipole moment of the \( i \)th particle, \( \tilde{G}_i^0(\mathbf{r}, \mathbf{r}_{ji}) \) is the free-space dyadic Green’s function between the \( i \)th and \( j \)th particles, \( \alpha_{E,i} \) (\( \alpha_{H,i} \)) is the electric (magnetic) polarizability of the \( i \)th particle, and \( \kappa \) is the wave number of light. The free-space dyadic Green’s functions are defined as

\[
\tilde{G}_i^0(\mathbf{r}, \mathbf{r}_{ji}) = \hat{G}_{ij} + \hat{b}_j \mathbf{n}_{ji} \otimes \mathbf{n}_{ji},
\]

(16a)

\[
\mathbf{V} \times \tilde{G}_i^0(\mathbf{r}, \mathbf{r}_{ji}) = \hat{g}_{ij} \mathbf{n}_{ji} \otimes \mathbf{n}_{ji} - \hat{I} \mathbf{l},
\]

(16b)

where \( \hat{I} \) is the identity matrix, \( \mathbf{n}_{ji} \) is the unit vector pointing from the \( j \)th to \( i \)th particle, the \( \mathbf{n}_{ji} \otimes \hat{I} \mathbf{l} \) operator is expressed explicitly as \( (\mathbf{n}_{ji} \otimes \mathbf{n}_{ji}) - \hat{I} \mathbf{l} \mathbf{l} \), and the following scalars have been defined:

\[
\alpha_{E} = 6 \pi a_1 / k^3,
\]

(18a)

\[
\alpha_{H} = 6 \pi a_1 / k^3.
\]

(18b)

The coupled-dipole equations in Eq. (15) can then be written in an equivalent matrix notation:

\[
e_0 |E_0\rangle = (\hat{a}_E^{-1} - k^2 \hat{M}_E)|p\rangle + \sqrt{e_0 \mu_0 k^2} \hat{M}_g |m\rangle,
\]

(19a)

\[
|H_0\rangle = (\hat{a}_H^{-1} - k^2 \hat{M}_H)|m\rangle - \sqrt{e_0 \mu_0 k^2} \hat{M}_e |p\rangle,
\]

(19b)

where \( \hat{a}_E \) (\( \hat{a}_H \)) is a diagonal matrix containing the electric (magnetic) polarizabilities of each particle and \( \hat{M}_E \) or \( \hat{M}_H \) contains the appropriate collection of dyadic Green’s functions to act between the dipoles,

\[
\hat{M}_E = \begin{pmatrix}
0 & \hat{G}_{12} & \cdots & \hat{G}_{1N} \\
\hat{G}_{12} & 0 & \cdots & \cdots \\
\vdots & \vdots & \ddots & \vdots \\
\hat{G}_{1N} & \cdots & \cdots & 0
\end{pmatrix},
\]

(20a)

\[
\hat{M}_g = \begin{pmatrix}
0 & \hat{g}_{12} & \cdots & \hat{g}_{1N} \\
\hat{g}_{12} & 0 & \cdots & \cdots \\
\vdots & \vdots & \ddots & \vdots \\
\hat{g}_{1N} & \cdots & \cdots & 0
\end{pmatrix},
\]

(20b)

Given the matrix equations seen in Eq. (19), it is apparent that there is flexibility in whether the incident field is written as a function of electric and magnetic dipole moments or whether the electric or magnetic dipole moments are written as a function of the incident electric and magnetic fields. Moreover, by rearranging, we can write Eq. (19) in the following alternate form:

\[
|p\rangle = e_0 \hat{M}_e \left( |E_0\rangle - \frac{\mu_0}{e_0} \hat{M}_h |H_0\rangle \right),
\]

(21a)

\[
|m\rangle = \hat{M}_g \left( |H_0\rangle + \frac{e_0}{\mu_0} \hat{M}_h |E_0\rangle \right),
\]

(21b)

where the \( \hat{M} \) matrices have been defined as

\[
\hat{M}_e = (\hat{a}_e^{-1} - k^2 \hat{M}_E) + k^2 \hat{M}_e (\hat{a}_H^{-1} - k^2 \hat{M}_G)^{-1} \hat{M}_g^{-1},
\]

(22a)

\[
\hat{M}_h = (\hat{a}_H^{-1} - k^2 \hat{M}_H) + k^2 \hat{M}_h (\hat{a}_G^{-1} - k^2 \hat{M}_G)^{-1} \hat{M}_e^{-1},
\]

(22b)

\[
\hat{M}_g = k^2 \hat{M}_g (\hat{a}_G^{-1} - k^2 \hat{M}_G)^{-1},
\]

(22c)

\[
\hat{M}_h = k^2 \hat{M}_h (\hat{a}_E^{-1} - k^2 \hat{M}_E)^{-1}.
\]

(22d)

Here the matrices \( \hat{M}_e \) and \( \hat{M}_h \) describe the response of the system to applied forcings, whereas \( \hat{M}_g \) and \( \hat{M}_e \) describe the component of bianisotropic forcing on the electric and magnetic dipoles that comes from the magnetic and electric fields, respectively (both \( \hat{M}_g \) and \( \hat{M}_h \) are singular). The obvious change in using the form of the coupled-dipole equations in Eq. (21) is that the electric and magnetic dipole moments can be calculated separately from each other. That is to say, rather than using one rank \( 6N \) matrix to describe the electromagnetic system [42], we are able to instead use the two rank \( 3N \) matrices (\( \hat{M}_g \), and \( \hat{M}_h \)) to describe how the system responds electrically and magnetically. The reason this particular formulation is important is because the electric and magnetic forcings are distinct from each other, and hence, each forcing transforms according to a single irreducible representation (when considering a normal-incidence plane wave). This therefore makes the electromagnetic dipole system directly subject to the eigenmode arguments provided in the previous section. It is important to note that no additional approximations have been made in writing the dipole equations in the manner of Eq. (21); this formulation still represents the full electromagnetic dipole system.

Now, for the specific case of oligomers with \( D_{6h} \) symmetry, we are able to restrict the observed effect of the bianisotropic forcings. This is specifically because, under normal plane-wave excitation, the incident field is acting uniformly on all dipoles in planar systems, and therefore, it is only the sum of dipole moments in the complete oligomer, the “total dipole moment,” that affects the extinction cross section. Moreover, the extinction cross section can be written as

\[
\sigma_e = \frac{e_0 |E|^2}{4 \pi} \Im \left( \sum_i E_i \mathbf{p}_i + H_i \mathbf{m}_i \right),
\]

(23a)

\[
= \frac{e_0 |E|^2}{4 \pi} \Im \left( E_0 \left( \sum_i \mathbf{p}_i \right) + H_0 \left( \sum_i \mathbf{m}_i \right) \right).
\]

(23b)
In a physical sense this means that only the total dipole moment of such a structure can couple directly to an arbitrarily polarized incident field. So, by using Eq. (21), the total dipole moments can be written as a function of the electric and magnetic incident-field vectors

\[
\sum_l p_l = \varepsilon_0 \hat{M}_{el} E_0(r_0) - \sqrt{\varepsilon_0 \mu_0} \hat{M}_{el} H_0(r_0),
\]

\[
\sum_l m_l = \hat{M}_{bh} H_0(r_0) + \frac{\varepsilon_0}{\mu_0} \hat{M}_{el} E_0(r_0).
\]

where we have defined the \(3 \times 3\) matrices \(\hat{M}\) as

\[
\hat{M} = \sum_{3 \times 3 \text{ blocks}} \hat{M}_{el,bh,eh,he}.
\]

However, given \(D_{N_R}\) contains a \(C_N\) symmetry subgroup, it has previously been shown [36] that we will have forms for the summed \(\hat{M}\) matrices that always commute with \(\hat{M}\):

\[
\hat{N} \propto \begin{pmatrix} a & c & 0 \\ -c & a & 0 \\ 0 & 0 & b \end{pmatrix}, \quad a,b,c \in \mathbb{C}.
\]

For this reason, applying the reflection symmetry operation to position vectors in Eq. (25) in the manner described by Eq. (26) will require that both the aggregate electric to magnetic coupling matrices (\(\hat{M}_{el}\) and \(\hat{M}_{bh}\)) be equal to their own negative. In other words, these matrices must be zero, and subsequently, the bianisotropic forcing will have no effect on the total dipole moments or extinction cross section. This can be understood from the perspective that \(D_{N_R}\) symmetry is not chiral, which is typically the catalyst that leads to such bianisotropy. So, for a planar oligomer with \(D_{N_R}\) symmetry, the dipole moments of the full electromagnetic system [Eq. (24)] will reduce to

\[
\sum_l p_l = \varepsilon_0 \hat{M}_{el} E_0(r_0). \tag{29a}
\]

\[
\sum_l m_l = \hat{M}_{bh} H_0(r_0). \tag{29b}
\]

As such, when considering far-field measures such as extinction [Eq. (23)], we are able to use the following modified dipole equations with no loss of generality or accuracy:

\[
|p\rangle = \varepsilon_0 \hat{M}_{el} |E_0\rangle, \tag{30a}
\]

\[
|m\rangle = \hat{M}_{bh} |H_0\rangle. \tag{30b}
\]

In this way we can use the two matrices, \(\hat{M}_{el}\) and \(\hat{M}_{bh}\), to define and calculate separate electric and magnetic eigenmodes rather than combined electromagnetic eigenmodes. For all simulations presented in this paper, we calculate the eigenmodes numerically from specifically these two matrix definitions.

To demonstrate how Fano resonances can occur in all-dielectric oligomers we will now consider the silicon heptamer seen in Fig. 4. Here there are three magnetic eigenmodes and three electric eigenmodes that interfere with each other to produce multiple Fano resonances and other sharp features.
First, in regard to the analysis of all-dielectric oligomer Fano resonances [26], we can see that the resonance of the central particle corresponds to the largest Fano resonance. This relationship can be reconciled with the eigenmode decomposition presented here given the dominance of the central particle in the eigenmode denoted by vi in Fig. 4(f). However, as seen in Fig. 4(d), there are also two other Fano resonances resulting from the sharp resonances of eigenmodes iii (electric) and v (magnetic), which do not correspond to the central particle resonance. When removing the central particle, only one of these Fano resonances remains with only a slight frequency shift and broadening, as seen in Fig. 4(a). This can be expected given that the interfering (magnetic) eigenmode profiles look similar in both heptamer and hexamer cases when neglecting the central particle. That is to say, this Fano resonance comes primarily from the ring-type oligomer in both cases. Fano resonances in ring-type oligomers have been demonstrated earlier in this paper, so here we will instead highlight the more subtle effect that adding or removing the central particle has on this one resonance. Specifically, the shift and narrowing of this particular Fano resonance, from adding the central particle, can be seen to be a result of a narrower excitation of the corresponding eigenmode, but it is also worth noting that the mode itself becomes more subradiant in the heptamer case given the central particle oscillates out of phase with the surrounding hexamer. In this sense the resonance in the heptamer closer resembles a “definition Fano resonance” where a narrow subradiant resonance interferes with a broadband superradiant mode. This subradiance and increased similarity to textbook Fano resonance conditions correspondingly results in the Fano resonance occurring at the peak of the subradiant mode in the heptamer rather than the intersection between modes in the hexamer. As such, we are able to differentiate interference between two resonant eigenmodes and these definition Fano resonances, although, noticeably, both lead to similar line shapes and sharp features in extinction, so the necessity for such a distinction is not particularly obvious. However, the resulting resonance observed in extinction is not always simple given systems such as the heptamer can have more than two interfering eigenmodes. For instance, we observe that a definition Fano resonance is occurring from the interference of super- and subradiant electric eigenmodes (i and iii, respectively) in Fig. 4(b), but the Fano line shape becomes skewed by the third electric eigenmode. Understandably, a more thorough analysis can be done on this system. However, what we have aimed to demonstrate here is that the approach presented in this paper offers a richer framework to understand the variety of electromagnetic responses and their interactions in all-dielectric oligomers.

V. CONCLUSIONS

We have rigorously shown that there exists a common physical mechanism leading to Fano resonances in both plasmonic and all-dielectric symmetric oligomers. The observed Fano line shapes are attributed to the fact that the eigenmodes of oligomers are not orthogonal and therefore that they can interfere with each other. We have also proved a key theorem to demonstrate that there exists a fixed number of eigenmodes that can be excited by a plane wave under normal incidence in a symmetric oligomer and that this number is independent of the number of nanoparticles in the oligomer. As such, we have shown that Fano resonances can be realized in completely symmetric oligomers excited by normal-incidence plane waves without involving additional complexity in the system. The presented approach has been directed at addressing the need for an analytical background in the rapidly growing field of all-dielectric systems and oligomers. In addition, we have also demonstrated that Fano resonances do not require the excitation of dark modes. The mechanism presented here therefore also has an impact on the current surging interest in the study of the far-field effects of dark modes [45–47] because it facilitates similar effects through a simpler mechanism. Finally, it is also worth noting that the approach presented in this paper is sufficiently general to be applicable to broader fields, such as molecular optics, antenna design, and other applications that are able to utilize dipole-dipole interactions.

Note added. Recently, a related study has been published by Forestiere et al. [48]. The authors employed a very different approach, but they came to the same conclusion that Fano resonances in plasmonic oligomers are induced by the interference of collective eigenmodes without requiring the excitation of dark modes.

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A.3 Interplay of magnetic responses in all-dielectric oligomers to realize magnetic Fano resonances

Summary

Optically induced magnetic response was one of the key motivations in the development of metamaterials, while there was also a more focused interest in optically magnetic responses that exhibit plasmonic-like interference features, particularly Fano resonances. Here it is demonstrated that oligomers made of high-index dielectric nanoparticles can support multiple dimensions of magnetic dipolar response; the magnetic dipole moment of an individual dielectric nanoparticle is shown to interact with the collective magnetic response of a nanoparticle oligomer. Using these two channels of magnetic dipolar response, a sharp and magnetic-magnetic Fano resonance was demonstrated in a symmetric high-index dielectric nanoparticle quadrumer. Such interference features had previously only been produced by the interaction between the electric and magnetic responses of asymmetric structures or nanoparticle clusters, demonstrating that high-index dielectric nanoparticle oligomers are a fully functional magnetic counterpart for plasmonic nanoparticle systems.

Notes and errata

- In equations (4) and (5), and impacting Figure 4, there are no additional scaling factors added between \( p \) and \( m \), and between \( \mathbf{E} \) and \( \mathbf{H} \), which were necessary to define eigenmodes with single units, as used in (3.6). The substitution \( \epsilon_0 = \mu_0 = c_0 = 1 \) here will provide the same eigenmodes as defined in Chapter 3.

- The curl of the dyadic Green’s function is a factor of \( ik \) larger than it should be, however all equations are correct once making the substitution: \( \nabla \times \hat{G}_0 \rightarrow \frac{1}{ik} \nabla \times \hat{G}_0 \).
Interplay of Magnetic Responses in All-Dielectric Oligomers To Realize Magnetic Fano Resonances
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ABSTRACT: We study the interplay between collective and individual optically induced magnetic responses in quadrumers made of identical dielectric nanoparticles. Unlike their plasmonic counterparts, all-dielectric nanoparticle clusters are shown to exhibit multiple dimensions of resonant magnetic responses that can be employed for the realization of anomalous scattering signatures. We focus our analysis on symmetric quadrumers made from silicon nanoparticles and verify our theoretical results in proof-of-concept radio frequency experiments demonstrating the existence of a novel type of magnetic Fano resonance in nanophotonics.

KEYWORDS: Fano resonance, nanoparticle oligomer, silicon, optical magnetism, symmetry, nanophotonics, quadrumer

A key concept in the study of optical metamaterials has been the use of geometry to engineer and boost the magnetic response from metallic nanostructures.1−5 Indeed, while simple metallic nanoparticles have a negligible magnetic response, a split ring resonator, or a properly arranged cluster of three or more particles, is able to sustain localized magnetic resonances.6−7 An alternate route to optical magnetism is based on single nanoparticles made from high-index dielectrics, because each such nanoparticle can exhibit an inherent magnetic response.8−11 In this work, we combine the use of the magnetic responses arising from both (i) individual nanoparticles and (ii) design geometry, to provide two distinct dimensions for purely magnetic dipolar response. Specifically, we present a comprehensive study of the magnetic interplay between individual and collective magnetic responses in symmetric clusters of four nanoparticles, known as quadrumers, and discuss the appearance of a novel type of magnetic Fano resonance. The ability to achieve directional Fano resonances with magnetic-type responses was considered for particles with negative permeability,16 but here we demonstrate magnetic dipolar Fano resonances in the total cross section of nanostructures made of conventional materials. Nanoparticle quadrumers were chosen because they have previously been shown to exhibit significant magnetic responses when light couples into a resonant circulation of displacement current associated with a large magnetic dipole moment.5,13−16 It was further demonstrated that, by breaking a quadramer’s geometric symmetry, it is possible to get interaction between this collective magnetic response and the in-plane electric response, an interaction that can lead to sharp magneto-electric hybrid Fano resonances.7,15 However, as we show in the following, dielectric nanoparticles enable a different coupling mechanism between magnetic resonances, one that does not require breaking geometric symmetry; the z-polarized magnetic field (where z is the quadramer’s principle axis) produced by a resonant circulation of current is, under certain conditions, able to couple to the inherent magnetic response of the individual nanoparticles. Here, we demonstrate that, by using an s-polarized plane wave at oblique incidence to induce a resonant circulating current across the cluster, we can couple the collective magnetic response of a symmetric all-dielectric quadramer into the inherent magnetic response of its individual dielectric particles. The interference between magnetic responses can then be tailored to produce distinctive and sharp magnetic Fano resonances. This form of optically induced magnetic–magnetic coupling and interference is a unique characteristic of properly designed, dielectric nano-clusters. This work thereby presents a new way to tailor the magnetic responses of all-dielectric metamaterials and nano-antenna devices, a result that complements the recent interest in dielectric nanostructures that utilize simultaneous excitation and tailoring of electric and magnetic optical responses.17−21
RESULTS AND DISCUSSION

Consider the optical response of a symmetric quadrumer when it is excited by a plane wave, whose propagation direction and electric field polarization lie in the plane of the quadrumer (s-polarization), as shown in Figure 1a. The conventional magnetic response to such an excitation is supported by a collective circulation of electric displacement current around all four nanoparticles. However, if the nanoparticles are made of a high-index dielectric material such as silicon, the individual nanoparticles are also able to couple directly with the applied magnetic field, sustaining internal circulating polarization currents. In this sense, a silicon quadrumer can produce magnetic responses to both electric and magnetic components of the applied plane wave, as depicted in Figure 1b. In this figure, the red arrows denote an electric dipole, and the blue circular arrows denote the circulation of displacement current that supports an out-of-page magnetic dipole. Importantly, an interplay may be induced between the two magnetic responses through the locally enhanced, z-polarized magnetic field sustained by the collective circulation of displacement current, which establishes a coupling channel to the magnetic responses of the individual nanoparticles.

We begin by understanding this magnetic interplay, for which we will model the interactions between nanoparticles using the coupled electric and magnetic dipole approximation (CEMDA). The mathematical description of this dipole model in free space is based on the following equations:

\[
\mathbf{p} = \alpha_0 \varepsilon_0 \mathbf{E}_0(\mathbf{r}) + \alpha_0 k^2 \sum_{j \neq i} \mathbf{G}_0(\mathbf{r}_i, \mathbf{r}_j) \mathbf{p}_j - \frac{1}{\varepsilon_0} \mathbf{V} \times \mathbf{G}_0(\mathbf{r}_i, \mathbf{r}_j) \mathbf{m}_i
\]

\[
\mathbf{m}_i = \alpha_0 \mathbf{H}_0(\mathbf{r}_i) + \alpha_0 k^2 \sum_{j \neq i} \mathbf{G}_0(\mathbf{r}_i, \mathbf{r}_j) \mathbf{m}_j
\]

(1b)

where \( \mathbf{p} \) (\( \mathbf{m} \)) is the electric (magnetic) dipole moment of the \( i \)th particle, \( \mathbf{G}_0(\mathbf{r}_i, \mathbf{r}_j) \) is the free space dyadic Greens function between the \( i \)th and \( j \)th dipole, \( \alpha_e \) (\( \alpha_m \)) is the electric (magnetic) polarizability of a particle, \( c_0 \) is the speed of light, and \( k \) is the free-space wavenumber. In Figure 2a, we show that this dipole model is accurate in modeling full-wave simulations of the silicon nanosphere quadrumer performed using CST Microwave Studio, confirming that, in this spectral range, the optical response of such nanoparticle quadrumeres is dominated by dipole interactions among the individual particles. This is the case because higher order coupling in such systems will occur only with smaller interparticle separations (see also the Supporting Information). Figure 2b shows the electric field distribution at the collective magnetic resonance of the cluster, confirming that the response of the silicon quadrumer includes a collective circulation of electric field around the particles. However, we can also see the additional dynamic produced by dielectric nanoparticles: the magnetic response induced by circulating transverse electric dipoles is accompanied by a magnetic response from each of the individual nanoparticles. This is the unique effect we are interested in.

The magnetic response in an individual silicon nanosphere is an internal circulation of polarization current, which can be seen in Figure 2b. In the CEMDA we choose to homogenize this circulating current distribution into a discrete source of magnetization current: the magnetic dipole. This implies that the key to strong interaction between individual and collective magnetic responses resides in the strong coupling between induced electric and magnetic dipoles in each inclusion. To describe this coupling, we can consider the eigenmodes of the quadrumer system. As we will show, the eigenmodes of the full, electromagnetic, system can be constructed from the eigenmodes of the decoupled electric and magnetic dipole systems. Therefore, we are going to break apart our dipole...
system of eq 1 into two decoupled equations: one equation for the electric dipoles and one equation for the magnetic dipoles.

\[ \mathbf{p} = a_0 e_\alpha \mathbf{E}_0(\mathbf{r}) + a_k k^2 \sum_{j \neq i} \hat{g}_0(\mathbf{r}_j, \mathbf{r}) \mathbf{p}_j \]  
\[ \mathbf{m} = a_0 h_\alpha \mathbf{H}_0(\mathbf{r}) + a_k k^2 \sum_{j \neq i} \hat{g}_0(\mathbf{r}_j, \mathbf{r}) \mathbf{m}_j \]  

(2a)

(2b)

We can then define two sets of eigenmodes from these equations: one for the electric dipoles and one for the magnetic dipoles. Using state notation, we can refer to the electric dipole eigenmodes as \( |e\rangle \) and the magnetic dipole eigenmodes as \( |m\rangle \). The eigenmode approach in the dipole approximation offers a huge simplification when combined with symmetry, because it allows us to determine certain eigenmodes without calculation. The key is that, for both electric and magnetic dipole systems, the dipole approximation restricts the number of eigenmodes for each equation to 12 (three dimensions by four dipoles). Each of these eigenmodes can only transform according to a single irreducible representation of the quadrumer’s symmetry group, referred to as \( D_{4h} \). Further details regarding the \( D_{4h} \) symmetry group’s irreducible representations and the implications for eigenmodes are provided in the Supporting Information. For the analysis here, it suffices to say that there are only eight irreducible representations in \( D_{4h} \) for 12 eigenmodes, and therefore the eigenspace associated with certain irreducible representations must be one-dimensional. Moreover, given that any vector in a one-dimensional space is an eigenvector by default, we are able to derive a number of eigenmodes by simply finding dipole moment profiles that transform according to certain irreducible representations. Eight such dipole moment profiles are shown in Figure 3.

![Basis vectors for the optical response of the quadrumer’s electric and magnetic dipoles](image)

Figure 3. Basis vectors for the optical response of the quadrumer’s electric and magnetic dipoles, named according to their associated irreducible representation. Due to finite dimensions and symmetry constraints, each basis vector shown here is an eigenmode of the electric or magnetic dipole equation (eq 2a or eq 2b).

Each of these dipole moment profiles is the sole basis vector for a single irreducible representation and is therefore an eigenmode of the electric or magnetic dipole equations in eq 2, irrespective of wavelength or the choice of material, size, or any parameter that conserves the symmetry of the quadrumer. It is worth noting that this same procedure for finding eigenmodes is applicable to other symmetries when using the dipole approximation, particularly the \( D_{4h} \) symmetry groups for small values of \( n \).

We need to now consider the interaction between electric and magnetic dipole systems. An eigenmode of either electric or magnetic dipoles can be substituted into eq 1 to determine the resulting state of magnetic or electric dipoles (\( |m_{i\alpha}\rangle \) or \( |p_{i\alpha}\rangle \), respectively) that it will induce due to magnetoelectric coupling:

\[ \mathbf{m}_{i\alpha} = a_0 k^2 e_\alpha \sum_{j \neq i} \mathbf{V} \times \hat{g}_0(\mathbf{r}_j, \mathbf{r}_i) \mathbf{e}_j \]  
\[ \mathbf{p}_{i\alpha} = -a_0 k^2 h_\alpha \sum_{j \neq i} \mathbf{V} \times \hat{g}_0(\mathbf{r}_j, \mathbf{r}_i) \mathbf{h}_j \]  

(3a)

(3b)

However, it has been shown that the operation describing the coupling between states of electric and magnetic dipole moments must commute with the geometry’s symmetry operations. Subsequently, the \( |e\rangle \) and \( |m\rangle \) dipole moments must both transform according to the same irreducible representation, and similarly for the \( |h\rangle \) and \( |p\rangle \) dipole moments. Therefore, if we consider the eigenmodes of electric and magnetic dipoles in Figure 3, the requirement for symmetry conservation specifies which electric dipole eigenmode can couple into which (if any) magnetic dipole eigenmode, and vice versa. As a result, the \( A_{2g} \) or \( B_{1g} \) eigenmodes (cf. Figure 3) are able to only magnetoelectrically couple into the other \( A_{2g} \) or \( B_{1g} \) eigenmode, whereas the \( A_{1g} \) and \( B_{2g} \) eigenmodes are not able to couple into any eigenmodes due to a symmetry mismatch. This conclusion allows us to take our analysis of the decoupled electric and magnetic dipole equations and use it to determine the full, electromagnetic, eigenmodes of the real system. Indeed, we would not necessarily expect an eigenmode of this complex scattering system to be a current distribution described by purely electric or magnetic dipoles; we would expect it to be a combination of both. If we now restrict ourselves to considering only the \( A_{2g} \) (magnetic-like) eigenmodes of an all-dielectric quadrumer, the two eigenmodes from the decoupled electric and magnetic dipole equations (cf. Figure 3) form basis vectors for the \( A_{2g} \) eigenmodes, \( |v\rangle \), of the electromagnetic system.

\[ |v\rangle = a_e |e\rangle + b_h |h\rangle \]  

(4)

where \( a_e \) and \( b_h \) are complex scalars. Notably, we should expect to have two distinct electromagnetic eigenmodes here because there are two distinct basis vectors and, subsequently, a two-dimensional eigenspace for the quadrumer’s response. To derive expressions for \( a_e \) and \( b_h \), we can write the electromagnetic eigenvalue equation for eq 1:

\[ a_e e_\alpha = a_e \alpha e_\alpha e_\alpha e_\epsilon + a_k k^2 \sum_{j \neq i} a_j \hat{g}_0(\mathbf{r}_j, \mathbf{r}_i) e_{\epsilon} - \frac{b_h}{\epsilon_0} \]  
\[ \mathbf{V} \times \hat{g}_0(\mathbf{r}_j, \mathbf{r}_i) \mathbf{h}_j \]  

\[ b_h h_\alpha = b_h \alpha h_\alpha h_\alpha h_\epsilon + a_k k^2 \sum_{j \neq i} b_j \hat{g}_0(\mathbf{r}_j, \mathbf{r}_i) \mathbf{h}_j + a_c c_0 \]  
\[ \mathbf{V} \times \hat{g}_0(\mathbf{r}_j, \mathbf{r}_i) \mathbf{e}_j \]  

(5a)

(5b)
where $\lambda_1$ is the eigenvalue of the electromagnetic eigenmode. However, eqs 5a and 5b are not independent (further details of this relation are provided in the Supporting Information) because one can be obtained from the other using duality transformations. In other words, a solution for $a_i$ and $b_i$ that satisfies either eq 5a or eq 5b must also satisfy the complete eq 5 and describe an electromagnetic eigenmode of eq 4. So, if the basis vectors (e) and (h) are normalized to a magnitude of 1, we can project eq 5a and eq 5b onto $e_i$ and $h_i$ (respectively) and sum over all i to obtain the two solutions for eq 5:

$$a_i = \frac{\sum e_k^* P_{\lambda}(\lambda)}{\alpha_k e_0 (\lambda_1 - \lambda_2)}$$

(6)

$$b_i = \frac{\sum h_k^* m_{\lambda}(\lambda)}{\alpha_k (\lambda - \lambda_2)}$$

(7)

where $\lambda_1$ and $\lambda_2$ are the eigenvalues of (e) and (h) in the decoupled electric and magnetic dipole equations in eq 2:

$$a_k e_{\lambda\lambda} = 1 - \alpha_k^2 \sum_{i,j} e_k^* \tilde{G}_0(r_i, r_j) e_j$$

(8a)

$$a_k e_{\lambda\lambda} = 1 - \alpha_k^2 \sum_{i,j} h_k^* \tilde{G}_0(r_i, r_j) h_j$$

(8b)

Each of the two ratios in eq 6 and eq 7 describes a distinct eigenmode for the electromagnetic system according to eq 4. It is worth noting that, if either numerator or denominator go to zero in eq 6 or eq 7, the starting basis vectors of the decoupled electric or magnetic dipole equations were already eigenmodes of the full system. In regard to the $\lambda_2$ (magnetic-like) eigenmodes of an all-dielectric quadrumer, our derivations show that, when both eigenmodes are far from resonance and magnetoelectric coupling can be neglected, one eigenmode will be purely composed of magnetic dipoles. However, as they approach resonance, magnetoelectric coupling cannot be neglected and the two ratios of $a_i$ and $b_i$ will need to be calculated to determine their form as electromagnetic eigenmodes (see Figure 4c). In such a situation, the two eigenmodes, $w_1$ and $w_2$, are explicitly nonorthogonal to each other with the overlap defined by

$$\langle w_2 | w_1 \rangle = a_1^* a_2 + b_1^* b_2$$

(9)

We can therefore expect interference between these eigenmodes as they approach resonance. Indeed, as shown in Figure 4, the Fano resonance feature in the silicon quadrumer of Figure 2 is produced entirely by the interference between these two eigenmodes. To serve as a broader qualification of this magnetic Fano resonance feature, we also investigate its parameter dependence. In Figure 5a and b, we vary the size of the gap between nanoparticles and the size of each individual nanoparticle, respectively. It can be seen that the Fano resonance is dependent on a number of these coupling parameters, but in a way that is different from electric Fano resonances in plasmonic oligomers. Perhaps most notably, the magnetic Fano resonance is significantly shifted with small changes in size of the constituent nanoparticles, and closely spaced nanoparticles are favorable for electric Fano resonances in plasmonic oligomers, but can destroy the magnetic Fano resonance here. More specifically, the magnetic Fano resonance feature is heavily dependent on the spacing between neighboring nanoparticles; increasing or decreasing the spacing can quickly diminish the Fano feature. On the other hand, varying the size of the particles while holding the gap constant is able to conserve the Fano resonance and shift it spectrally (shown in the shaded region of Figure 5b). This behavior can be expected because the action of increasing the size of the particles while holding the gap constant is very similar to uniformly scaling Maxwell’s equations, and silicon permittivity has relatively minor dispersion in this spectral range. Otherwise, the final parameter we consider in Figure 5c is the angle of incidence to address the practical limitations for in-plane excitation of nanoscale optical structures. It can be seen that the Fano resonance will persist up to roughly 45° incidence, beyond which the magnetic response is dominated by the normal-incidence responses.

**Experimental Verification.** To verify and validate the theoretical arguments presented above, we look for the existence of magnetic Fano resonances in a high-index cluster experimentally. In this regard, one practical option is to mimic the scattering properties of silicon nanoparticles using MgO-TiO$_2$ ceramic spheres characterized by dielectric constant of 16 and dielectric loss factor of $(1.12-1.17) \times 10^{-4}$, measured at $9-12$ GHz. These ceramic spheres in the microwave range therefore have very similar properties to silicon nanospheres in the optical range, and they are subsequently a useful macroscopic platform on which to prototype silicon nanostructures. Here, they allow us to perform a "proof of concept" investigation into the properties of an isolated quadrumer with much more signal than would be expected from a single silicon.

### Figure 4. Electromagnetic eigenmode decomposition of the silicon quadrumer, calculated using the CEMDA, showing (a) the contribution of the $A_{2g}$ (magnetic-like) eigenmodes to the overall extinction. In (b) we decompose the $A_{2g}$ response in terms of its two eigenmodes, showing the creation of a Fano resonance from interference between them. To support this, we also show (c) the real components of the dipole moment profiles for each $A_{2g}$ eigenmode, calculated at the wavelength of the Fano feature.
nanosphere quadrumer. Indeed, such spheres have been used previously to predict the behavior of silicon nanoantennas.31 The MgO-TiO₂ quadrumer consists of four dielectric spheres with diameter $d = 15$ mm, and the size of the gap between the particles is $s = 5$ mm. The experimentally measured, and numerically calculated, total scattering of the quadrumer structure is shown in Figure 6. It can clearly be seen that a magnetic Fano resonance is produced at 5.4 GHz in both simulation and experiment. This is the first example of a magnetic–magnetic Fano resonance in a single symmetric metamolecule. Notably, this Fano resonance occurs in a spectral range where the single particle is not at resonance, which demonstrates its collective nature. Indeed, it appears near the intersection of the single particle’s electric and magnetic scattering contributions, reflecting that the overlap of eigenmodes (eq 9) is dependent on both electric and magnetic dipole polarizabilities.

**CONCLUSIONS**

We have presented a comprehensive study of the interplay between the collective optically induced magnetic responses of all-dielectric quadrumers and the individual magnetic responses of their constituent dielectric nanoparticles. We have been able to establish the theoretical basis behind the interaction between collective and individual magnetic responses in all-dielectric structures, providing a quantitative prediction of the interference between the quadrumer’s magnetic responses leading to magnetic–magnetic Fano resonance features. We have also been able to experimentally observe the existence of a sharp magnetic–magnetic Fano resonance in a dielectric quadrumer. Such Fano resonance features demonstrate the unique potential that suitably designed dielectric nanoclusters have for scattering engineering at the nanoscale, opening exciting and unexplored opportunities for dielectric nanophotonics.

**METHODS**

To fasten together the MgO-TiO₂ ceramic spheres for the experiment, we used a custom holder made of a styrofoam material with a dielectric permittivity of 1 (in the microwave frequency range). To approximate plane wave excitation, we employed a rectangular horn antenna (TRIM 0.75–18 GHz; DR) connected to the transmitting port of a vector network analyzer (Agilent E8362C). The quadrumer was then located in the far-field of the antenna, at a distance of approximately 2.5 m, and a second horn antenna (TRIM 0.75–18 GHz; DR) was used as a receiver to observe the transmission through the quadrumer. The extinction measurement shown in Figure 6b was then obtained as the difference between the measured transmission and unity transmission (i.e., with no quadrumer). For the theory, CST simulations were performed assuming plane wave excitation on a quadrumer located in free space. The CEMDA simulations, seen in Figure 2a, used electric and magnetic dipole polarizabilities that were derived from the scattering coefficients of Mie theory.34 In all simulations, silicon permittivity data were taken from Palik’s Handbook30 and the MgO-TiO₂ spheres were assumed to be dispersionless with a dielectric constant of 16 and dielectric loss factor of $(1.12−1.17) \times 10^{-4}$.

**ASSOCIATED CONTENT**

* Supporting Information

In the Supporting Information we provide additional information and derivations regarding the $D_{3h}$ symmetry group, the equivalence of the coupled electric and magnetic dipole equations (eq 1) under duality transformations, and the near-field accuracy of the dipole approximation. The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsphotonics.5b00082.

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**Notes**

The authors declare no competing financial interest.
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REFERENCES

Supporting Information:

Interplay of magnetic responses in all-dielectric oligomers to realize magnetic Fano resonances

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(4 Pages, 1 Table, 1 Figure)
D$_{4h}$ symmetry

The eight irreducible representations of the D$_{4h}$ symmetry group are depicted as the rows in Table S1 and the columns correspond to symmetry operations, being: rotations ($\hat{C}$), reflections ($\hat{\sigma}$), inversions ($\hat{i}$), improper rotations ($\hat{S}$) and the identity ($\hat{E}$). Each irreducible representation describes a distinct set of transformation behavior under the quadrumer’s symmetry operations. It follows that any given eigenmode can only transform according to a single irreducible representation and, consequently, that eigenmodes belonging to different irreducible representations must be orthogonal.

Table S1: Character table for the D$_{4h}$ symmetry group. The rows correspond to different irreducible representations and the columns are the symmetry operations. Each number in the table is the character (trace) of the matrix representation of each symmetry operation for the given irreducible representation.

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Duality transformations in dipole systems

We can consider the electric and magnetic field radiated by an arbitrary system of electric and magnetic dipoles:

\[ \mathbf{E}_r(r_i) = \frac{k^2}{\epsilon_0} \left( \sum_{j \neq i} \hat{G}_0(r_i, r_j) \mathbf{p}_j - \frac{1}{c_0} \nabla \times \hat{G}_0(r_i, r_j) \mathbf{m}_j \right), \]  
\[ (1) \]

\[ \mathbf{H}_r(r_i) = \frac{k^2}{\mu_0} \left( \sum_{j \neq i} \hat{G}_0(r_i, r_j) \mathbf{m}_j + c_0 \nabla \times \hat{G}_0(r_i, r_j) \mathbf{p}_j \right). \]  
\[ (2) \]

These two equations are, in fact, equivalent to each other under the duality transformation where \( \mathbf{E}_r \rightarrow \sqrt{\mu_0/\epsilon_0} \mathbf{H}_r, \mathbf{H}_r \rightarrow -\sqrt{\epsilon_0/\mu_0} \mathbf{E}_r, \mathbf{p} \rightarrow \mathbf{m}/c_0, \mathbf{m} \rightarrow -c_0 \mathbf{p} \). This follows directly from the duality transformation of the electrical displacement and B-field, when expressed in terms of electric and magnetic fields, polarization, and magnetization. Subsequently, if we return to the equations of the Coupled Electric and Magnetic Dipole Approximation (CEMDA) and rewrite them in terms of the radiated electric and magnetic field, we get:

\[ \mathbf{p}_i = \alpha_E \epsilon_0 \mathbf{E}_0(r_i) + \alpha_E \epsilon_0 \mathbf{E}_r(r_i), \]  
\[ (3) \]

\[ \mathbf{m}_i = \alpha_H \mathbf{H}_0(r_i) + \alpha_H \mathbf{H}_r(r_i). \]  
\[ (4) \]

Then, after dividing through by the respective dipole polarizabilities, it is clear to see that these equations simply state that the total field at any point is the sum of radiated and incident field. As such, the equivalence of radiated electric and magnetic fields under duality transformations is sufficient to make the two CEMDA equations equivalent under duality transformations.
Near field accuracy of the dipole model

The accuracy of the Coupled Electric and Magnetic Dipole Approximation (CEMDA) was demonstrated in Fig. 2a when modeling the extinction of the silicon nanoparticle quadrumer. However, it is also worth acknowledging that this method is also able to correctly deduce the near-field properties of this structure. To demonstrate this, in Fig. S1, we plot the distribution of the average (root mean square) electric field amplitude at the Fano resonance frequency, calculated from both CST and CEMDA. The CEMDA is inherently not able to produce the field distribution inside the nanoparticles, because the electric and magnetic fields diverge as they approach a point source. However, outside of the nanoparticles, the CEMDA is able to provide a near-quantitative match to the full CST simulation.

Figure S1: The distribution of the Root Mean Square (RMS) scattered electric field amplitude of the silicon quadrumer used in Fig. 2 in the main text, calculated using CST and the Coupled Electric and Magnetic Dipole Approximation (CEMDA). The simulations were performed at the wavelength of the quadrumer’s Fano resonance (also used in Fig. 2b), and show that the CEMDA is accurate in predicting the near field behavior of the quadrumer we consider in the main text.
A.4 Fano resonance enhanced nonreciprocal absorption and scattering of light

Summary

It is well recognized that the total losses from light incident on a optical scattering system does not change if you invert the propagation direction; a consequence of reciprocity as discussed in Section 4.2. An optical device that distinguishes between opposite propagation directions therefore typically requires one breaks reciprocity by typically using strong magnetic fields with magnetic materials, or an optical systes with properties that depend on a DC current. However, we show here that asymmetric plasmonic nanostructures can exhibit abrupt differences in absorption and scattering properties for light that propagates in opposite directions, without breaking reciprocity. Such behaviour is derived to occur in the presence of nonorthogonal eigenmodes, and we show it can thereby be driven and enhanced through Fano resonances. This thereby highlights a new avenue by which nanoscale optical devices are able to measure and distinguish between oppositely propagating light without requiring external magnetic fields or magnetic materials, to thereby avoid a number of fabrication and operation challenges.

Notes and errata

- In the eigenmode equation (4), $J$ should be replaced with $v_i$. The correct expression is (3.1).
- The use of the word ‘nonreciprocal’ is incorrect given the considered system is entirely reciprocal, the work instead refers to a difference of optical response due to reciprocal plane waves.
Abstract: We reveal that asymmetric plasmonic nanostructures can exhibit significantly different absorption and scattering properties for light that propagates in opposite directions, despite the conservation of total extinction. We analytically demonstrate that this is a consequence of nonorthogonality of eigenmodes of the system. This results in the necessity for modal interference with potential enhancement via Fano resonances. Based on our theory, we propose a stacked nanocross design whose optical response exhibits an abrupt change between absorption and scattering cross-sections for plane waves propagating in opposite directions. This work thereby proposes the use of Fano resonances to employ nanostructures for measuring and distinguishing optical signals coming from opposite directions.

Keywords: fano resonance; plasmonics; stereometamaterials; nanophotonics; reciprocity

1. Introduction

Reciprocity is one of the key concepts in optics because it is related to the equal transmission of oppositely-propagating plane waves through any structure or medium; a manifestation of the total optical loss being conserved [1–4]. Indeed, to induce a difference in transmission from oppositely-propagating
light requires optical elements that are driven by magnetic fields or materials to break the conditions of reciprocity [5–8]. Such nonreciprocal optical behavior is widely utilized in optical communications devices, and particularly optical isolators and circulators. There has subsequently been significant interest in miniaturizing these devices for incorporation into nanophotonic chips and the broader realization of integrated optics systems. However, there remain limitations on the minimum propagation length or interaction time that is needed to produce sufficient reciprocity breaking. Promising avenues exist to utilize gyromagnetic response [9] or engineered nonlinearity [10] to inherently break reciprocity, but here we instead present a way to produce nonreciprocal optical behavior in a linear system without breaking reciprocity. We begin by acknowledging that nanoantennas and metasurfaces, in general, have two separate loss channels: the radiative loss (scattering cross-section) and the dissipative material loss (absorption cross-section), both of which can be associated with distinct and measurable quantities. In particular, we show that the reciprocity constraint is not directly applicable to these independent loss channels. Indeed, this freedom has recently been demonstrated to allow circular dichroism in the absorption of planar structures [11] (arXiv). Here, we extend this approach to investigate nonreciprocal absorption and scattering cross-sections of three-dimensional nanostructures and stereometamaterials. First, we show that existence of nonorthogonal eigenmodes, in general, results in resonant interference phenomena associated with the Fano resonances. We then demonstrate that this nonorthogonality permits the absorption and scattering cross-section of an optical device to distinguish between opposite propagation directions, such as depicted in Figure 1.

**Figure 1.** A schematic representation of different optical loss channels: radiative (scattering) and dissipative (absorption). A planewave propagating in one direction will be channeled into far field scattering, while (simultaneously) an identical plane wave propagating in the opposite direction will be channeled into material absorption.

It suggests that absorption and scattering provide us with the independent measurements on oppositely-propagating signals in the vicinity of a Fano resonance. We further show that such a nonreciprocal optical device can be designed in a way to have optical response being independent of polarization. This behavior demonstrates that direction-isolating measurements are widely achievable
in passive optical nanodevices. Indeed, this proposes a new approach for ultrathin optical isolation of signal.

2. Eigenmodes, Interference, and Fano Resonances

It was recently demonstrated that Fano resonances can lead to circular dichroism in the absorption cross-section of planar chiral nanostructures [11]. The existence of such circular dichroism means that the absorption in the nanostructure was not constrained by reciprocity. In this article, we investigate further Fano resonances and their role in producing nonreciprocal absorption and scattering cross-sections of finite nanostructures. Below, we outline our approach and establish why Fano resonances can be associated with nonorthogonal eigenmodes. The existence of such nonorthogonal eigenmodes is then used to predict and demonstrate propagation-dependent absorption and scattering cross-sections.

The fields inside nanostructures can be associated with the induced currents and polarizations, which determine the complete near- and far-fields produced by the given configuration. In this approach all eigenmodes are defined within finite volumes. However, importantly, there is also a one-to-one correspondence between such eigenmodes and resonances [12], which provides an intuitive basis to capture the optical features of resonant nanostructures. The relationship between induced current, \( J \), and an arbitrary applied field, \( E_0 \), is expressible in terms of dyadic Green’s functions [13]:

\[
-i \omega [\epsilon - \epsilon_0] E_0(x) = J(x) - k^2[\epsilon(x) - \epsilon_0] \int_V \hat{G}_0(x, x') J(x') \, d^3x'
\]  

where \( k \) is the wavenumber, \( \omega \) is the angular frequency, \( V \) is the volume of the scatterer (assumed finite), and \( \hat{G}_0 \) is the free space dyadic Green’s function:

\[
\hat{G}_0(x, x') = \text{P.V.} \left[ \frac{\sigma}{k^2} \nabla \nabla + \frac{1}{k^2} \nabla^2 \right] \frac{e^{ik|x-x'|}}{4\pi \epsilon_0 |x-x'|} - \frac{\hat{L} \delta(x - x')}{k^2 \epsilon_0}
\]  

where \( \hat{L} \) is the source dyadic [13] and P.V. implies a principal value exclusion of \( x' = x \) when performing the integration in Equation (1). We have also defined permittivity (\( \epsilon \)) in terms of both the conductivity (\( \sigma \)) and the electric susceptibility (\( \chi \)) to define the current (\( J \)) as the sum of both polarization and conduction current, which is related to the total electric field as:

\[
\epsilon \equiv (\chi + 1)\epsilon_0 - \frac{\sigma}{i \omega} \quad \Rightarrow \quad J(x) = -i \omega [\epsilon(x) - \epsilon_0] E(x)
\]  

Equation (1) is therefore a general expression for modeling the complete optical response of an arbitrary scattering geometry. An eigenmode, \( v_i \), of any given structure can then be defined such that it will satisfy Equation (1) as:

\[
-i \omega [\epsilon - \epsilon_0] \lambda_i v_i (x) = J(x) - k^2[\epsilon - \epsilon_0] \int_V \hat{G}_0(x, x') v_i(x') \, d^3x'
\]
where \( \lambda_i \) is the eigenvalue of \( v_i \). Because the full set of these eigenmodes form a complete basis for the optical response, they allow us to model the entire range of bright and dark resonances in any given system. However, recently, it was demonstrated that such a basis of eigenmodes is not necessarily orthogonal [14]. The origin of such nonorthogonal eigenmodes can be understood as a consequence of the inherent radiation losses, where the Equation (1) corresponds to a non-Hermitian system. To consider the effect of this nonorthogonality, and its relation to the Fano resonances, we can refer to the extinction cross-section, which can be written in terms of the near-field overlap of induced current and incident field [11,15].

\[
\sigma_{\text{ext}} = \frac{1}{|E_0|^2} \sqrt{\frac{\mu_0}{\epsilon_0}} \text{Re} \left[ \int_{V_s} E_0^* \cdot J \, dV \right]
\]  

(5)

Additionally, an arbitrary applied field and the induced currents can be defined in terms of a linear superposition of the eigenmodes:

\[
E_0 = \sum_i a_i \lambda_i v_i \quad \Rightarrow \quad J = \sum_i a_i v_i
\]  

(6)

Using these expressions, we are able to rewrite the total extinction (Equation 5) in terms of eigenmodes and eigenvalues:

\[
\sigma_{\text{ext}} = \frac{1}{|E_0|^2} \sqrt{\frac{\mu_0}{\epsilon_0}} \sum_i \left( \text{Re}[\lambda_i] \left( \int_{V_s} |a_i|^2 |v_i|^2 \, dV \right) + \sum_{j \neq i} \text{Re}[a_i^* a_j \lambda_i^* \left( \int_{V_s} v_i^* \cdot v_j \, dV \right)] \right)
\]  

(7)

Notably, we have two sets of terms contributing to extinction: direct terms that provide contributions to extinction from individual eigenmodes, and also interference terms coming from the overlap between different eigenmodes. For orthogonal eigenmodes the interference term vanishes. Moreover, the presence of nonorthogonal eigenmodes leads to existence of the Fano resonances in this model. To demonstrate this, we first rule out any dependency of a given eigenmode’s excitation (i.e., the \( a_i \) coefficients of Equation (6)) on the excitations of other eigenmodes. This can be done very directly, because the dyadic Greens function is complex symmetric

\[
\hat{G}_0(x, x') = \hat{G}_0(x', x), \quad \hat{G}_0 = \hat{G}_0^T
\]  

(8)

Due to this symmetry, it is possible to write the overall operator of the eigenvalue equation (Equation (4)) as a complex symmetric matrix and in the normal form shown by Gantmacher [16]. Thus, such nondegenerate eigenmodes must be orthogonal under unconjugated projections [17].

\[
\int_V v_\alpha \cdot v_\beta \, dV = 0 \quad \text{when} \quad \lambda_\alpha \neq \lambda_\beta
\]  

(9)
Subsequently, the excitation of any given eigenmode is defined entirely by the eigenmode itself and the incident field:

\[ \lambda_i a_i = \frac{\int_V \mathbf{v}_i \cdot \mathbf{E}_0 \, dV}{\int_V \mathbf{v}_i \cdot \mathbf{v}_i \, dV} \]  

(10)

It shows that the excitation of any given eigenmode is independent from the excitations of any other eigenmode. This means that the only way an interaction between two or more eigenmodes affects the extinction is due to nonorthogonality of these eigenmodes resulting in the interference terms seen Equation (5). This is indeed the conclusion of earlier work for the Fano resonances arising in nanoparticle oligomers [14]. In particular, we have established that the excitation of eigenmodes is determined entirely from the eigenmode itself and the excitation field. As such, the unambiguous conclusion is that all optical Fano resonances can be considered as a consequence of interference between so-called “bright” eigenmodes. This is in good accordance with the bright mode Fano resonances that have been proposed and observed in recent years [14,18–20]. Furthermore, the existence of nonorthogonal eigenmodes allows us to describe nonreciprocal absorption and scattering cross-sections of lossy nanoscale structures.

3. Nonreciprocal Absorption and Scattering

We begin this section by considering three-dimensional geometries that have at least 3-fold discrete rotational symmetry and are excited by plane waves at normal incidence in order to utilize derivations provided in Ref. [11]. However, the use of such symmetry also prevents any circular cross polarization in transmission [21], which is often observed as circular conversion dichroism [22–24], and not of interest here. Using more formal notation, we focus on structures with \( C_n \) symmetry for \( n \geq 3 \). Notably, unlike the case of planar structures considered in Ref. [11], we do not have a plane of reflection symmetry perpendicular to the propagation direction and we, therefore, cannot relate the excitations of plane waves propagating in opposite directions. As such, we are automatically driven to account for the role of propagation direction. So, we begin by defining the current distribution, \( \mathbf{J} \), induced by a plane wave \( \mathbf{E}_0 \), and a current distribution, \( \mathbf{J}' \), induced by the reciprocal plane wave, \( \mathbf{E}_0^* \), where the * indicates a complex conjugate. These are source and response distributions satisfying Equation (1).

However, we can now acknowledge that a normally-incident plane wave can only excite eigenmodes that transform according to the two-dimensional, \( E \) irreducible representations of the rotational symmetry point group (e.g., see Table 1). A result from Ref. [11] then follows directly: if we assume the eigenmodes are orthogonal, then the total current intensity is conserved in the structure between reciprocal plane waves.

\[ \int_V \mathbf{J}'^* \cdot \mathbf{J} \, dV = \int_V \mathbf{J}''^* \cdot \mathbf{J}' \, dV \]  

(11)
Table 1. Character table for the 4-fold rotational symmetry group ($C_4$). Rows are irreducible representations, columns are symmetry operations, and each element denotes the trace of the associated symmetry operation’s matrix representation in the given irreducible representation.

<table>
<thead>
<tr>
<th>$C_4$</th>
<th>$E$</th>
<th>$C_4$</th>
<th>$C_2$</th>
<th>$C_4^3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A$</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>$B$</td>
<td>1</td>
<td>−1</td>
<td>1</td>
<td>−1</td>
</tr>
<tr>
<td>$E$</td>
<td>${1, i}$ &amp; $−1$ &amp; $−i$ &amp; ${1, −i}$ &amp; $−1$ &amp; $i$</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

If we have a uniform material loss throughout our geometry, this result means that the absorption cross-section is conserved for reciprocal plane waves. Thus, due to the fact that the extinction is already conserved by reciprocity, the scattering cross-section must too be conserved, as it is simply the difference between extinction and absorption. In other words, this result shows that nonorthogonal eigenmodes are necessary to permit nonreciprocal behavior in the absorption and scattering cross-sections. In practical terms, we need a structure that can exhibit a Fano resonance under plane wave excitation. Now, if we are considering a single, isotropic and homogeneous material, the eigenmodes of the Green’s function operator (depicted in Equation (1)) are the eigenmodes of the total system. The eigenmodes are therefore independent of material properties, which instead affect only the eigenvalues. Notably, $\hat{G}_0$ is a complex symmetric operator, which means that, when $\hat{G}_0$ is real, the whole operator is actually real symmetric and therefore Hermitian; making all its eigenmodes orthogonal. As the source dyadic ($\hat{L}$) is real, this means that retardation of coupling within the structure has to be nontrivial ($e^{i|\mathbf{x}−\mathbf{x}'|}$ $\not\rightarrow$ 1) in order to have nonorthogonal eigenmodes. This retardation condition provides a design guideline for generation of the Fano resonances. Nanoparticle oligomers were used as planar chiral geometries to satisfy this retardation condition in Ref. [11], but for nonplanar geometries we can instead look to the work done in the area of stereometamaterials [25]; three-dimensional structures commonly, albeit not always [26], constructed from stacked elements [27–29]. Here, the distance between stacked elements is able to guarantee nontrivial retardation in coupling.

To demonstrate that this indeed provides nonorthogonal eigenmodes, we consider a simple dimer model of two electric dipoles, $p_1$ and $p_2$, with isotropic polarizabilities, $\alpha_1$ and $\alpha_2$, at locations, $r_1$ and $r_2$, along a principal axis, separated by a distance, $|r_1 − r_2| = d$. For such a system, we can use the relation between current and polarization in Equation (3), to simplify Equation (1) down to the known dipole model.

\[
\begin{align*}
\alpha_1 &\varepsilon_0 \mathbf{E}_0(r_1) = p_1 - \alpha_1 k^2 \hat{G}_0(r_1, r_2)p_2 \\
\alpha_2 &\varepsilon_0 \mathbf{E}_0(r_2) = p_2 - \alpha_2 k^2 \hat{G}_0(r_2, r_1)p_1
\end{align*}
\]

Given the reflection symmetry of the structure about the principal axis, there will be one set of eigenmodes with dipole moments polarized parallel to the principal axis and a separate (orthogonal) set
of eigenmodes that have dipole moments polarized perpendicular to the principal axis. This orthogonal separation is a result of the eigenmodes that are affected by such symmetric reflection operations being orthogonal to those that are not affected. More formally, it is a consequence of the eigenmodes being associated with different irreducible representations of the \( \mathcal{C}_\infty v \) point group. In any case, we are able to consider exclusively the dipole moments that are perpendicular to the principal axis, because they correspond to those that can be excited by a plane wave propagating along the principal axis. Moreover, the relationship between these dipole moments and the applied field can be written in a simplified form:

\[
E_0(r_1) = (\alpha_1 \epsilon_0)^{-1} p_1 - G_d p_2
\]

\[
E_0(r_2) = (\alpha_2 \epsilon_0)^{-1} p_2 - G_d p_1
\]

where \( G_d \equiv \frac{e^{ikd}}{4\pi \epsilon_0 d} \left( k^2 + \frac{i k}{d} - \frac{1}{d^2} \right) \)

From this, we can deduce analytical expressions for the two nondegenerate eigenmodes that are excitable by a planewave in this system:

\[
|v_{1,2} \rangle \equiv \begin{bmatrix} p_1 \\ p_2 \end{bmatrix} = \begin{bmatrix} \alpha_2 - \alpha_1 \pm \delta \\ 2\alpha_1 \alpha_2 \epsilon_0 G_d \end{bmatrix}
\]

where \( \delta \equiv \sqrt{(\alpha_2 - \alpha_1)^2 + (2\alpha_1 \alpha_2 \epsilon_0 G_d)^2} \)

Importantly, these two eigenmodes are not necessarily orthogonal provided that \( \alpha_1 \neq \alpha_2 \) and \( G_d \neq 0 \),

\[
\langle v_1 | v_2 \rangle = |2\alpha_1 \alpha_2 \epsilon_0 G_d|^2 + (\alpha_2 - \alpha_1 + \delta)^* \cdot (\alpha_2 - \alpha_1 - \delta)
\]

Therefore, these results demonstrate that nonorthogonal eigenmodes exist even in the dimer model, but only if there is coupling between the dipoles and the dipole polarizabilities are different. In other words, Equation (17) suggests that even an asymmetric dimer should be able to realize nonorthogonal eigenmodes and, therefore, nonreciprocal absorption and scattering. To demonstrate this, we consider an idealized dimer made from two nanoparticles of 200 nm diameter. We also assume that the material properties of nanoparticles are described by Drude model with a plasma frequency of 2.07 PHz. The particles are separated by a gap of 200 nm and excited with a plane wave along the dimer’s principal axis. This structure is illustrated in Figure 2a. The nonreciprocal absorption can be obtained by making one particle lossless and the other particle lossy with a damping factor of 600 THz (a very large damping). Importantly, because we define one nanosphere to be lossless, the absorption cross-section will only be related to the dipole moment intensity in the lossy particle, and not the integral over the complete geometry that we originally considered for continuous systems made from a single material, such as in Equation (11).
In Figure 2b, we show that the dipole model eigenmodes of this dimer (Equation (16)) are nonorthogonal across the majority of the visible frequency range. The extinction, absorption and scattering cross-sections of this dimer are then shown in Figure 2c. It demonstrates the difference in the scattering and absorption between oppositely-propagating planewaves at wavelengths where two resonances appear to overlap. Thus, even such a simple dual-layer geometry is able to provide strongly nonorthogonal eigenmodes and the nonreciprocal behavior. Notably, while we utilized different materials to produce nonorthogonal eigenmodes for this dimer, the coupling between dipoles is still a necessity for nonorthogonal eigenmodes (i.e., consider $G_d \to 0$ in Equation (17)). However, to consider a realistic dimer nanostructure, we have to acknowledge that material properties are largely fixed by fabrication and that we must instead consider geometry. On the simplest level, different sizes and shapes of nanoparticles will be associated with different dipole polarizabilities and thereby produce the same effect as material change did in Figure 2. Ultimately, the dipole responses in a nanoparticle
dimer do not provide sufficient freedom to induce significant modal interference with realistic material parameters. We can optimize the response by considering pairs of stacked, symmetric nanocrosses as seen in Figure 3 (similar in concept to the stereometamaterial in Ref. [27]). Arrays of such stacked nanocross geometries have previously exhibited a difference in effective impedance and hence reflection for opposite propagation directions [30]. For our analysis, a relative twist in the orientations of the nanocrosses also provides chirality and allows us to consider the lowest symmetry conditions that our earlier theoretical arguments applied to (i.e. the $C_n$ point groups).

**Figure 3.** Simulated nonreciprocal scattering and absorption cross-sections in stacked gold nanocrosses from linearly-polarized plane waves propagating in opposite directions ($\pm z$) along the rotational symmetry axis. Simulation results are shown to be the same for both chiral ($C_4$) and achiral ($C_{4v}$) structures, and both are independent of polarization. The calculation parameters are given in the text.

The dimensions of the stacked nanocrosses were chosen as to produce nonorthogonal eigenmodes and a Fano resonance by pushing the independent resonances of each nanocross together. This is done in Figure 3 using a pair of gold crosses separated by a 120 nm gap, and with dimensions: total length equal to 150 nm, arm width equal to 40 nm and thickness equal to 30 nm. We then reduced the arm width, $w$, of one cross to 25 nm in order to provide the necessary asymmetry. In Figure 3, we can clearly see that the absorption and scattering cross-sections differ dramatically between reciprocal plane waves, and independently of whether the structure is chiral or not. Interestingly, the lack of chirality tells us that the difference in scattering and absorption cannot be attributed to bianisotropic coupling, which was the mechanism for the impedance difference in Ref. [30]. In this regard, the symmetry of the individual crosses ensures that, in isolation, their optical responses are isotropic, yet notably: the optical response of the stacked crosses remains independent to the relative twist, which suggests that there is isotropic coupling between the two crosses. Such isotropic coupling would exist if the dominant
interaction between crosses was dipolar, as it would make the situation analogous to the coupled isotropic dipoles considered for Figure 2. Dipolar coupling would be expected to dominate with increasing separation between crosses, and, therefore, the twist-independent optical response we observe will be a consequence of the larger distance between the two crosses. Additionally, as is also suggested by the negligible effect of chirality, the choice of linear or circular polarization of the incident plane wave has no effect on any of the cross-sections. Indeed, in the chiral geometry, there appears to be negligible circular dichroism between oppositely-handed plane waves that propagate in the same direction for any of the cross-sections, simply because the associated curves are separated by less than 0.001 \( \mu \text{m}^2 \). Similarly, in regard to linear polarizations, the angle of linear polarization is of no significance because the discrete rotational symmetry is separately known to prevent any such angular dependence in all of the cross-sections [31]. Figure 3 therefore demonstrates that Fano resonances and modal interference in stacked nanocross geometries allow for significant variation in optical response depending on the propagation direction and irrespective of any polarization. All simulations were performed using CST Microwave Studio and gold data was taken from Johnson and Christy [32].

To finally provide an investigation into the role of the asymmetry about the \( z \)-axis for these stacked nanocrosses, we can consider varying the arm width, \( w \), of one nanocross while holding the other constant. The absorption, scattering and extinction cross-sections of the stacked nanocrosses through this parameter sweep are shown in Figure 4. Notably, when the two nanocrosses are identical (\( w = 40 \text{ nm} \)), the geometry has \( D_4 \) symmetry and is, thereby, invariant under a \( \pi \)-rotation perpendicular to the principal rotation axis, which makes reciprocal planewaves equivalent under geometric symmetry operations. In other words, we cannot have \( D_n \) symmetry if we want to produce nonreciprocal absorption and scattering. However, we can see that this invariance is related to direction of scattering and absorption splitting interchanging either side of \( D_n \) symmetry. Therefore, Figure 4 ultimately shows us that the difference in scattering and absorption is widely achievable in systems such as stacked nanocrosses that support closely overlapped and coupled resonances, and therefore modal interference. As such, this demonstrates that significant nonreciprocal measurements can be performed with stereometamaterial geometries.

4. Conclusions

We have shown that the distinct absorption and scattering cross-sections of nonplanar nanostructures can distinguish between reciprocal planewaves, provided the system has nonorthogonal eigenmodes. The relation of this effect to nonorthogonal eigenmodes was derived rigorously, and subsequently we established that the nonreciprocal absorption and scattering could be realized and enhanced through the Fano resonances. We have proposed that two stacked nanocrosses would provide the necessary retarded coupling to produce nonorthogonal eigenmodes and, thus, Fano resonances, while also providing sufficient design freedom for optimization. It was subsequently demonstrated that this geometry produces substantial differences in absorption and scattering cross-sections for plane waves that propagate in opposite directions. It was additionally shown that this difference would be independent of the plane wave’s polarization. This work subsequently establishes that modal interference and
Fano resonances enable a passive nanostructure to simultaneously measure and distinguish signal from reciprocal plane waves.

**Figure 4.** Simulation results for stacked gold nanocrosses, under excitation from circularly polarized plane waves given by propagation in either the $+z$ or $-z$ direction. The variation between right and left circular polarizations in the same propagation direction is negligible ($< 0.001 \mu m^2$) and making the associated curves overlapped here. The arm width, $w$, of one nanocross arm is varied from 20 to 60 nm, showing a significant difference in the absorption and scattering cross-section of reciprocal planewaves, provided the system does not have $D_1$ symmetry. The calculation parameters are given in text.

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Author Contributions

BH, AEM and ANP developed the theory. AEM and YSK supervised the research. BH, AEM and YSK wrote the manuscript.

Conflicts of Interest

The authors declare no conflict of interest.

References

A.5 Hybridisation and the origin of Fano resonances in symmetric nanoparticle trimers

Summary

The analysis of optical scattering properties for nanostructures is commonly based on the concept of mode hybridisation, particularly when considering plasmonic materials. Yet, in the vast majority of studies, hybridisation of plasmons is performed in only a qualitative manner, largely due to complexity of the quantitative calculation. Here we derive and present a simplified approach to the hybridisation procedure using the dipole model to investigate the optical response of a symmetric trimer. We are able to analytically follow the path by which hybridisation creates polarization-independent Fano resonances in symmetric nanoparticle trimers, a feature which has not yet been predicted in analyses that use plasmonic hybridisation theory. We are further able to show that the use of high-index dielectric particles provides an additional space of bianisotropic responses, which makes them more susceptible to Fano resonances compared to their plasmonic counterparts. The derived propensity of high-index dielectric trimers for Fano resonances is then demonstrated experimentally with full agreement to our theory. We are subsequently able to provide a validation of our hybridisation procedure and demonstrate that simple dielectric nanoparticle systems can lead to pronounced Fano resonances.

Notes and errata

- In section III, the scaling factors necessary to define eigenmodes with single units, are not included between \( p \) and \( m \), nor between \( \mathbf{E} \) and \( \mathbf{H} \). Consequently, the eigenmodes of section III depend on the choice of \( \epsilon_0 \) and \( \mu_0 \), unlike the eigenmodes as discussed in Chapter 3. One can perform the substitution \( \epsilon_0 = \mu_0 = c_0 = 1 \) in the expressions of section III to obtain the same eigenmodes as defined in Chapter 3.

- The expression for the doubly degenerate eigenmodes of a dielectric trimer is given as a function of the respective eigenvalue. It should be stated that the eigenvalue can be found relatively straightforwardly from the respective characteristic equation. An example of this is provided in §10.3.3 of [64].

- The curl of the dyadic Green’s function is defined to be a factor of \( ik \) larger than it should be in (B2), however all equations are correct once performing the substitution \( \nabla \times \tilde{G}_0 \rightarrow \frac{1}{ik} \nabla \times \tilde{G}_0 \).
Hybridization and the origin of Fano resonances in symmetric nanoparticle trimers

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We study the light scattering by plasmonic and dielectric symmetric trimers to investigate the existence of polarization-independent Fano resonances. Plasmonic hybridization theory is revealed to hide simple physics, and we instead provide a simplified model for hybridization to derive a plasmonic trimer’s eigenmodes analytically. This approach is demonstrated to accurately recreate full wave simulations of plasmonic trimers and their Fano resonances. We are subsequently able to deduce the grounds for modal interference in plasmonic trimers and the related formation of Fano resonances. However, by generalizing our simplified hybridization approach, we are also able to investigate the eigenmodes of all-dielectric trimers. We show that biaxialotropic coupling channels between high-index dielectric nanoparticles are able to increase the capacity for Fano resonances, even at normal incidence. We finally provide the first experimental measurements of sharp, polarization-independent Fano resonances from a symmetric all-dielectric trimer, with very good agreement with the predicted response from our simplified hybridization theory.

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I. INTRODUCTION

The Fano resonance [1] has become a well-recognized interference feature in the optical scattering response of many nanoscale structures. It attracted considerable attention due to its distinctly sharp extinction line shape and also due to anomalous near-field behavior for surface-enhanced Raman scattering [2], nonlinear response [3], and enhancement of circular dichroism [4]. While Fano resonances in the transmission of plasmonic surfaces and arrays can be readily described by the overlap of radiation in a single scattering channel [5,6], the mechanism for optical Fano resonances in compact plasmonic structures is often less apparent and has generally been explained through the use of a hybridization-theory argument [7]. This argument involves subdividing a structure into two or more subsystems with known properties and then deducing how their optical responses combine together in the near field. The way such optical responses combine, or hybridize, is regularly depicted as per molecular-orbital theory: the modes of each subsystem are added constructively or destructively to form a bonding mode and an antibonding mode, the latter of which exhibits suppressed scattering associated with the Fano resonance.

For the case of plasmonics, this involves treating individual nanoparticles as electron density distributions, a model which inherently accounts for quite comprehensive physics and subsequently requires nontrivial derivations for even the most simple structures, such as concentric spheres [8] and two-particle dimers [9]. However, these embodiments of hybridization theory also neglect retardation of the interaction between charges, making their results truly valid only for geometries that are bounded within negligible fractions of the operating wavelength. The absence of retardation also limits the model’s capacity to observe interference features, including Fano resonances, as is discussed in Ref. [10]. Such limitations have led to the amendment of plasmonic hybridization theory to account for retardation [11]; however, this understandably does not reduce the complexity of the theory. Indeed, using this model, even the comparatively simple case for plasmonic hybridization of a symmetric three-particle trimer has yet to be found. A consequence of such complexity is that hybridization is rarely performed explicitly. It has instead become a conceptual explanation for experimental and numerical observations in even quite complicated scattering systems for which hybridization solutions have never been found.

Here we instead begin from the premise that the dominant optical properties of nanoparticle geometries do not require the level of complexity found in plasmonic hybridization theory. We propose a simplified approach to hybridization when modeling a nanoparticle system in the dipole approximation, thereby considering only the dipole responses of individual nanoparticles and the coupling between them. To demonstrate the capacity of this simplification, we approach the outstanding problem of modal hybridization in symmetric trimers. In particular, the optical feature we focus on in symmetric trimers is the polarization-independent Fano resonance. This type of feature has previously been observed only in more complicated nanoparticle oligomers [12], where a conceptual hybridization can be performed in equivalent ways for arbitrary polarization angles. While the polarization independence of symmetric trimers is known [13–16], the existence of Fano resonances [17,18] and, indeed, their combination with polarization independence are not obvious in terms of hybridization, particularly because it is not possible to subdivide a trimer in a way that conserves its symmetry.

Here we are able to resolve this problem by performing the simplified hybridization procedure to find the eigenmodes of a plasmonic trimer analytically. We show that the doubly degenerate eigenmodes of trimers, those responsible for polarization-independent scattering, become inherently nonorthogonal in the presence of retarded coupling between particles. Such eigenmodes can therefore allow polarization-independent Fano resonances at normal incidence [18], and we provide full wave simulations to demonstrate these Fano resonances.
FIG. 1. (Color online) The eigenmodes of (a) a single particle and (b) a dimer with \( D_{2\text{h}} \) symmetry, when assuming only in-plane electric dipole responses from each particle. The dimer eigenmodes are labeled according to their associated irreducible representation. We also show the location of the extra particle(s) needed to form a trimer. In (c) and (d), we depict a generalized energy-level diagram for these eigenmodes and identify the coupling channels, labeled as \( A–D \).

resonances in plasmonic nanoparticle trimers, with good agreement with our analytical eigenmode results. However, by recognizing that our simplified approach to hybridization is portable to other models, we further investigate all-dielectric trimers in the coupled electric and magnetic dipole approximation [19] to account for the existence of magnetic dipolar responses of individual high-index dielectric nanoparticles. This analysis shows that previously reported bianisotropic coupling effects in dielectric oligomers [20] also play an important role in the optical response of all-dielectric geometries at normal incidence. The result is then a threefold increase in the number of doubly degenerate eigenmodes that can be excited by a normally incident plane wave in an all-dielectric trimer when compared to the plasmonic trimer. Moreover, all these eigenmodes are nonorthogonal and subsequently lead to a substantial increase in the presence and magnitude of polarization-independent Fano resonances at normal incidence.

We then verify these predictions in a radio-frequency experiment that mimics the optical transmission through a silicon nanosphere trimer. We observe a number of sharp, polarization-independent Fano resonances from the all-dielectric trimer at normal incidence. The eigenmodes calculated from our simplified hybridization approach are then able to provide a quantitative recreation of these experimental measurements and their explicit eigenmode decompositions. In doing so, we can unambiguously demonstrate the validity of our simplified approach to hybridization.

II. THE EIGENMODES OF PLASMONIC TRIMERS

For the purpose of performing hybridization, a trimer can be subdivided into only a dimer and a single particle; the other option would be the trivial case of dividing it into three single particles. Here we restrict ourselves to considering only the electric dipole response of each individual particle since it dominates for subwavelength plasmonic particles. We can therefore utilize the dipole approximation [21], where each particle’s dipole moment \( \mathbf{p} \) is related to the externally applied electric field \( \mathbf{E}_0 \) as

\[
\mathbf{p} = \alpha_\mathbf{E} \varepsilon_0 \mathbf{E}_0 \mathbf{p} + \alpha_\mathbf{E} k^2 \sum_{j \neq i} \hat{G}_0(\mathbf{r}_i, \mathbf{r}_j) \cdot \mathbf{p}_j,
\]

(1)

Here \( \alpha_\mathbf{E} \) is the electric dipole polarizability of a particle, and \( \hat{G}_0 \) is the free-space dyadic Green’s function, which acts on dipole moments as

\[
\hat{G}_0(\mathbf{r}', \mathbf{r}) \cdot \mathbf{p} = \frac{e^{ikR}}{4\pi R} \left[ \left( 1 + \frac{i}{kR} - \frac{1}{k^2 R^2} \right) \mathbf{p} - \left( 1 + \frac{3i}{kR} - \frac{3}{k^2 R^2} \right) (\mathbf{n} \cdot \mathbf{p}) \mathbf{n} \right].
\]

where \( k \) is the wave number, \( \mathbf{n} \) is the unit vector pointing from \( \mathbf{r} \) to \( \mathbf{r}' \), and \( R = |\mathbf{r} - \mathbf{r}'| \). In this model, the single-particle and dimer eigenmodes are both known and easily deduced and are shown in Figs. 1(a) and 1(b).

To derive how the dimer and single particle hybridize, we have to calculate the coupling channels between each dimer and single-particle eigenmode, labeled as \( A–D \) in Figs. 1(c) and 1(d). To find the coupling channels, we use Eq. (1) to calculate the dipole moments induced in the dimer by a single-particle eigenmode and project those onto the dimer eigenmodes (which form a complete basis for the dimer’s response) and vice versa. This gives us expressions for the coupling channels shown in Fig. 1(c).

\[
A = \frac{\alpha_\mathbf{E} e^{ikR}}{8\sqrt{2\pi} R} \left[ 3\kappa^2 + \frac{ik}{R} - \frac{1}{R^2} \right],
\]

(2a)

\[ B = \frac{-\sqrt{3} \alpha E e^{i k R}}{8 \sqrt{2} \pi R} \left[ k^2 + \frac{3 i k}{R} - \frac{3}{R^2} \right]. \]  

\[ C = \frac{\alpha e^{i k R}}{8 \sqrt{2} \pi R} \left( k^2 - \frac{5 i k}{R} + \frac{5}{R^2} \right). \]  

\[ D = B. \]

where we have normalized the single-particle and dimer modes to make all channels omnidirectional. All other coupling channels between the eigenmodes of the dimer and those of the single particle are zero, which is due to a geometry-induced symmetry mismatch between basis vectors. It is important to acknowledge that, unlike molecular-orbital hybridization, the hybridization of scattering structures does not have a perennial set of basis vectors (i.e., atomic orbitals) with known symmetry mismatch conditions to identify recurring coupling/hybridization channels in different geometries. In nanostructures, the coupling channels have to be derived for each given geometry in order to perform hybridization correctly. Importantly, our formulation of hybridization also relies on the basis vectors being eigenmodes of their associated subsystem. Indeed, if this were not the case, we would have to account for new coupling channels arising from interactions within each subsystem, which would substantially increase the number of coupling channels we have to consider and also their complexity.

In any case, to now find the trimer’s hybridized eigenmodes we recognize that an eigenmode’s dipole moment profile must satisfy the dipole equation [Eq. (1)] as

\[ v_i = \alpha_i \epsilon_0 \lambda_i v_i + \sum_{j \neq i} \alpha_j k^2 \hat{G}_0(\mathbf{r}, \mathbf{r}_j) \cdot v_j. \]  

We can then rewrite Eq. (3) as a system of linear equations by considering the dimer and single-particle eigenmodes (see Fig. 1) as orthogonal basis vectors. Moreover, we can consider an eigenmode \(|v_i\rangle\) as a linear combination of these basis vectors:

\[ |v_i\rangle = a_i |p_i\rangle + b_i |B_{3h}\rangle + c_i |B_{3v}\rangle + d_i |p_i\rangle \]  

\[ + e_i |B_{2v}\rangle + f_i |A_{x}\rangle. \]

As detailed in Appendix A, this basis allows us to solve Eq. (3) as a matrix equation to find analytical expressions for the hybridized eigenmodes of the trimer. Specifically, we obtain the following expressions for the hybridized eigenmodes:

\[ |v_1\rangle : \begin{cases} a_1 = \sqrt{3}, \\ b_1 = -1, \\ c_1 = -\sqrt{3}, \\ \lambda_1 = (\alpha \epsilon_0) - 1 + \frac{e^{i k R}}{8 \sqrt{2} \pi R} (3 k^2 + \frac{3 i k}{R} - \frac{3}{R^2}) \end{cases} \]  

\[ |v_{2v}, v_{3v}\rangle : \begin{cases} a_2, a_3 = \frac{\sqrt{3}}{2 \sqrt{2} \pi R} (5 k^2 + \frac{3 i k}{R} - \frac{3}{R^2}) \pm 2 \sqrt{6} \delta, \\ b_2, b_3 = \frac{-\sqrt{3}}{2 \sqrt{2} \pi R} (k^2 - \frac{3 i k}{R} + \frac{3}{R^2}) \pm 4 \sqrt{3} \delta, \\ c_2, c_3 = \frac{3 \sqrt{3}}{2 \sqrt{2} \pi R} (k^2 + \frac{3 i k}{R} - \frac{3}{R^2}), \\ \lambda_2, \lambda_3 = \lambda_0 \mp \delta, \end{cases} \]  

\[ |v_{2d}, v_{3d}\rangle : \begin{cases} d_2, d_3 = \frac{\sqrt{3}}{4 \sqrt{2} \pi R} (k^2 - \frac{3 i k}{R} + \frac{3}{R^2}) \pm 2 \sqrt{6} \delta, \\ e_2, e_3 = \frac{\sqrt{3}}{4 \sqrt{2} \pi R} (3 k^2 - \frac{3 i k}{R} - \frac{3}{R^2}), \\ f_2, f_3 = -\frac{3 i \sqrt{3}}{4 \sqrt{2} \pi R} (k^2 + \frac{3 i k}{R} - \frac{3}{R^2}), \\ \lambda_2, \lambda_3 = \lambda_0 \mp \delta, \end{cases} \]  

\[ \bar{v}_2, \bar{v}_3 : \begin{cases} d_4 = \sqrt{2}, \\ e_4 = -1, \\ f_4 = -\sqrt{3}, \\ \lambda_4 = (\alpha \epsilon_0) - 1 - \frac{e^{i k R}}{8 \sqrt{2} \pi R} (k^2 + \frac{3 i k}{R} - \frac{3}{R^2}). \end{cases} \]

Here all undefined coefficients [see Eq. (4)] are zero, and we have defined a central eigenvalue \(\lambda_0\) for the \(|v_2\rangle\) and \(|v_3\rangle\) eigenmodes, along with a splitting function \(\delta\) that produces their nondegeneracy.

\[ \lambda_0 := (\alpha \epsilon_0) - 1 - \frac{e^{i k R}}{16 \pi \epsilon_0 R} (k^2 - \frac{k}{R} + \frac{1}{R^2}). \]  

\[ \delta := \frac{1}{\alpha \epsilon_0 R} \sqrt{\frac{5 k^2}{2} + \frac{3(3 k^2 + 2 \pi i k - 3 \pi^2)}{2 R^2} + \frac{8 k^2}{5}}. \]

We can categorize the hybridized eigenmodes according to their irreducible representation in the trimer’s \(D_{3h}\) point group (see Table I). The \(|v_1\rangle\) eigenmode has azimuthally oriented dipole moments, and the \(|v_4\rangle\) eigenmode has radially oriented dipole moments, making them the trimer’s \(A_e\) and \(A_{1e}\) eigenmodes, respectively [13]. On the other hand, the \(|v_2\rangle\) and \(|v_3\rangle\) eigenmodes are the doubly degenerate \(E_e\) eigenmodes, which are responsible for polarization-independent behavior. Because we have obtained two distinct \(E_e\) eigenmodes, which is known to be the maximum number that a ring-type oligomer can exhibit in the dipole approximation [18], we know that the hybridization approach here has determined all of the \(E_e\) doubly degenerate eigenmodes for a plasmonic trimer.

It is worth pausing to acknowledge an interesting result of this analysis. If we were to consider the doubly degenerate eigenmodes in complex frequency space (\(s\) space) to find and/or analyze their resonances [23], there are points in \(s\) space where the expressions for \(|v_2\rangle\) and \(|v_3\rangle\) are the same, specifically when \(\delta = 0\). At such points, the two eigenmodes coalesce, and the eigenspace of these two eigenmodes subsequently reduces in dimension, indicating a nontrivial topology of the eigenspace. Such coalescence points are also known as

| TABLE I. Character table for the \(D_{3h}\) symmetry group. The rows correspond to different irreducible representations, and the columns are the symmetry operations. Each number is the trace of the associated operation’s matrix representation [22]. |

<table>
<thead>
<tr>
<th>(E)</th>
<th>(2C_3)</th>
<th>(3C_3^\prime)</th>
<th>(3\sigma_v)</th>
<th>(2S_3)</th>
<th>(3\sigma_e)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(A_1)</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>(A_2)</td>
<td>1</td>
<td>-1</td>
<td>1</td>
<td>1</td>
<td>-1</td>
</tr>
<tr>
<td>(E^2)</td>
<td>1</td>
<td>-1</td>
<td>0</td>
<td>2</td>
<td>-1</td>
</tr>
<tr>
<td>(E^*)</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>-1</td>
<td>-1</td>
</tr>
<tr>
<td>(E^0)</td>
<td>1</td>
<td>1</td>
<td>-1</td>
<td>-1</td>
<td>1</td>
</tr>
<tr>
<td>(E^\circ)</td>
<td>1</td>
<td>1</td>
<td>-1</td>
<td>0</td>
<td>1</td>
</tr>
</tbody>
</table>
exceptional points, and the coalescing eigenmodes are known to interchange expressions with each other, depending on the path taken through space in the vicinity of these points [24–26]. In a trimer, the locations of four such exceptional points can be found by solving \( \delta = 0 \) for a complex \( k \) in Eq. (7). An investigation into eigenspace topology is well beyond the scope of this work, and instead, it suffices that the eigenmodes and eigenvalues in Eq. (5) are exact expressions for that of a symmetric trimer when considering only the electric dipole response of each individual nanoparticle. Furthermore, using the expressions in Eq. (5), it follows that, in general, \(|v_2\rangle\) and \(|v_3\rangle\) will be nonorthogonal and written in terms of the eigenmode’s basis vector coefficients:

\[
\langle v_2 | v_3 \rangle = a_2^* a_3 + b_2^* b_3 + c_2^* c_3 = d_2^* d_3 + e_2^* e_3 + f_2^* f_3.
\]  

This means that polarization-independent Fano resonances could, indeed, be expected [18] to occur in plasmonic trimers, which is interesting given neither the dimer nor single particle is able to exhibit Fano resonances in isolation. In other words, the presence of coupling channels between the dimer and single particle has directly led to nonorthogonality in the trimer’s hybridized eigenmodes. This can be understood conceptually as the single particle providing a coupling channel into the \( |A_j\rangle \) and \( |B_{k_j}\rangle \) basis vectors, which are orthogonal to the incident field and thereby act as damped oscillators that are coupled to the remainder of the system. However, as discussed in Appendix A, it is also important that the coupling channels provide retardation in propagation for Eq. (8) to not sum to zero.

In any case, to now demonstrate Fano resonances of the nature we are describing, we can consider a symmetric trimer made of silver nanoparticles with a diameter of 100 nm, and we then vary the gap between particles from 40 nm down to touching. In Fig. 2, we show the evolution of the extinction cross section from this trimer for a normally incident plane wave. We provide two calculations for extinction: directly from CST MICROWAVE STUDIO (dashed line) and from the analytical eigenmodes of Eq. (5) (solid line). Notably, with the exception of the quadrupole resonance at high frequencies, there is a good match between the eigenmode analysis and the full wave simulations. Importantly, both approaches observe a small Fano resonance forming as the gap between particles is reduced. However, the advantage of using eigenmodes is we can attribute portions of the extinction to individual eigenmodes. Indeed, in Fig. 2(b), we can see that the Fano resonance is produced by interference [18] between the two eigenmodes.

Admittedly, while our model does not observe the single-particle quadrupole response, an extension our hybridization approach to models that account for quadrupole responses is not unfeasible. Such an extension would involve describing each nanoparticle using an electric dipole and an electric quadrupole, such as the model proposed in Ref. [27]. From this we would again be able to prescribe a set of coupling channels between the basis vectors composed of electric dipoles and also to and between new basis vectors composed of electric quadrupoles. We undertake a similar procedure in the next section when we treat individual particles as having both an electric dipole and a magnetic dipole. In any case, the results in Fig. 2 are able to validate the predictions and analysis of our approach to hybridization for a plasmonic trimer.

III. THE EIGENMODES OF ALL-DIELECTRIC TRIMERS

The analysis in the previous section corresponded to plasmonic nanoparticle trimers because we assumed only electric dipolar responses from individual nanoparticles. However, strong Fano resonances were predicted to exist in silicon all-dielectric trimers [17,18]. The hybridization of electric dipoles is not sufficient for considering all-dielectric oligomers because high-index dielectric nanoparticles have both electric and magnetic dipolar responses [28–30]. To investigate the eigenmodes of such systems, we can again employ our simplified approach to hybridization but instead use the coupled electric and magnetic dipole approximation [19], which will account for both electric and magnetic responses of individual nanoparticles.

From geometric symmetry principles, we know that this approach will result in two distinct sets of eigenmodes for the trimer: one which will transform under the trimer’s symmetry operations according to the \( E' \) irreducible representation and one which will transform according to the \( E'' \) irreducible representation (see Table I). The intuitive distinction between the two sets of eigenmodes is that they will be excited by either the electric or magnetic field in a normally incident plane wave for \( E' \) and \( E'' \), respectively. The different irreducible representations also mean that the trimer’s symmetry prevents \( E' \) eigenmodes coupling to the \( E'' \) eigenmodes and vice versa. If we could neglect the bianisotropic coupling between electric to magnetic dipoles, the \( E' \) eigenmodes would be those that...
we derived for plasmonic trimers [i.e., \(|v_2\rangle\) and \(|v_3\rangle\) in Eq. (5)], and the \(E''\) eigenmodes would be analogous to the same eigenmodes but constructed from magnetic dipoles rather than electric dipoles. However, by using the out-of-plane (\(z\)) direction, the \(z\)-oriented magnetic dipoles can be arranged into basis vectors that transform according to the \(E''\) irreducible representation, and the \(z\)-oriented electric dipoles can be arranged into basis vectors that transform according to the \(E'\) irreducible representation, as shown in Fig. 3(b). Hence, bianisotropic coupling between electric and magnetic dipoles will allow the electric dipole \(E'\) basis vectors to couple with the magnetic dipole \(E''\) basis vectors and vice versa for the \(E''\) basis vectors. From this point on, we shall consider only the \(E'\) eigenmodes because they are the eigenmodes excited by electric field and can therefore be more naturally related to the eigenmodes we derived for plasmonic trimers. The procedure for finding the \(E''\) eigenmodes is almost identical upon interchanging the electric and magnetic dipole polarizabilities.

We begin by extending the hybridization diagram of Fig. 1(d) to take into account both the new space of \(z\)-oriented \(E'\) responses and the bianisotropic coupling channels. In Figs. 3(a)–3(c), we show the complete set of basis vectors that hybridize to form the \(E'\) and \(E''\) eigenmodes of an all-dielectric trimer. These basis vectors can be separated according to their even or odd response under the dimer’s reflection symmetry operation because there are no coupling channels between the two resulting sets of basis vectors. One of these two sets is then sufficient to find instances of each doubly degenerate eigenmode (i.e., being the instances that are even/odd under the dimer’s reflection symmetry operation).

The even basis vectors offer a convenient opportunity to reduce the number of coupling channels because the trimer basis vector is an (orthogonal) eigenmode for the dimer. The new \(E\) and \(F\) coupling coefficients, shown in Fig. 3(d), can then be calculated by evaluating the magnetic and electric field radiated by electric and magnetic dipoles. See Appendix B for details.

\[
E_1 = \alpha_a \frac{1}{\sqrt{\varepsilon_0 \mu_0}} \frac{e^{ikR}}{4\sqrt{2}\pi R} \left( k^2 + \frac{ik}{R} \right),
\]

\[
E_2 = -\alpha_a \frac{1}{\sqrt{\varepsilon_0 \mu_0}} \frac{e^{ikR}}{4\sqrt{2}\pi R} \left( k^2 + \frac{ik}{R} \right),
\]

\[
F = \sqrt{2} E.
\]

We can then define an eigenmode of the dielectric trimer \(|w\rangle\) as a linear combination of single-particle and dimer eigenmodes.

\[
|w\rangle = a'|p\rangle + b'|B_0\rangle + c'|A\rangle + d'|E'\rangle.
\]
magnetic dipoles and is therefore unchanged from the previous analysis provided for plasmonic trimers. The remaining three-dimensional eigenspace can therefore be spanned by the two $E'$ eigenmodes in Eq. (5) ($|v_{y3}\rangle$ and $|v_{z3}\rangle$) and the $|E'_4\rangle$ basis vector. As such, all the remaining eigenmodes can be written as a linear combination of these three basis vectors, and therefore, all remaining eigenmodes must transform according to the $E'$ irreducible representation. Additionally, since the $E'$ eigenmodes transform according to a different irreducible representation than that of $|v_4\rangle$, we know they must be orthogonal to $|v_4\rangle$, and we can thereby write their general form as

$$|u_3\rangle : \begin{cases} a' = a', \\ b' = \sqrt{2}a' - \sqrt{3}c', \\ c' = c', \\ d' = d'. \end{cases} \quad (11)$$

If the bianisotropic coupling channels are negligible ($E, F \rightarrow 0$), the three basis vectors, $|v_{y2}\rangle$, $|v_{z2}\rangle$, and $|E'_4\rangle$, are the $E'$ eigenmodes of the trimer.

However, outside of this limit, we can use the dipole model to set up a rank-3 matrix equation to find the eigenmode solutions for $a'$, $c'$, and $d'$ (refer to Appendix B). These eigenmodes can be calculated analytically, but the result is cumbersome and does not provide any additional intuitive understanding. Therefore, it is sufficient to use a numerical approach, and this is what we do in the coming analysis. It is, however, worth acknowledging a conclusion here: because we have three linearly independent $E'$ basis vectors, a dielectric trimer will have three $E'$ eigenmodes, which is one more than the plasmonic trimer. Moreover, using the analogous argument, a dielectric trimer additionally has another three $E''$ eigenmodes that can be excited by the incident magnetic field. Therefore, the total number of doubly degenerate eigenmodes that can be excited in a dielectric trimer with a normally incident plane wave is six, and they are organized into two sets of three interfering eigenmodes. For comparison, a plasmonic heptamer (a central nanoparticle surrounded by a ring of six nanoparticles) has a single set of three interfering eigenmodes [18]. The dielectric trimer geometry therefore has double the propensity for Fano resonances of the plasmonic heptamer geometries. In this regard, the theoretical predictions for the existence of Fano resonances in all-dielectric trimers were previously made for silicon spherical nanoparticles [17,18], which exhibited Fano resonances in the optical frequency range. However, it is possible to create a macroscopic analog of silicon nanospheres in the microwave range, specifically using MgO-TiO$_2$ ceramic spheres characterized by a dielectric constant of 16 and a dielectric loss factor of $(1.12-1.17) \times 10^{-4}$ (measured at 9–12 GHz). These ceramic spheres in the microwave range therefore have properties very similar to those of silicon spheres in the optical range. As such, we are able to create a macroscopic analog of a silicon nanosphere trimer. This allows us to investigate the scattering properties of a single trimer with much less noise than would be possible for a single silicon nanosphere trimer.

In Fig. 4(a), we show an experimental setup. The trimer consists of three MgO-TiO$_2$ spheres with 15-mm diameter and 20-mm distance between the centers of the spheres. To fasten together the MgO-TiO$_2$ particles for the experiment, we used a custom holder made of a styrofoam material with a dielectric permittivity of 1 (in the microwave frequency range). To approximate plane-wave excitation and receive the signal scattered to the forward direction, we employed a pair of identical rectangular linearly polarized wideband horn antennas (TRIM, 1–18 GHz), connected to the coaxial ports of a vector network analyzer (Agilent E8362C). The trimer is located in the far-field of both antennas; the distance from the trimer to both the receiving and the transmitting antennas is approximately 1.5 m. The total extinction can then be extracted from the measured complex magnitude of the forward-scattered signal by means of the optical theorem [31]. Associated simulations of the experiment were also performed using the TIME DOMAIN solver of CST MICROWAVE STUDIO when assuming plane-wave excitation of the trimer in free space. The experimentally measured and numerically simulated extinction spectra are shown in Figs. 4(b) and 4(c). The extinction spectrum of an isolated MgO-TiO$_2$ sphere was also measured and simulated. In Fig. 4(b), we can see...
a pronounced Fano resonance at 4.8 GHz that is associated with the suppression of extinction. By varying the orientation of the trimer, it exhibits a polarization-independent response. The Fano resonance must subsequently be from interference between doubly degenerate eigenmodes, and we can therefore consider the hybridized eigenmodes derived according to our previous analysis. For this hybridization, we define electric and magnetic dipole polarizabilities from the $a_1$ and $b_1$ scattering coefficients of Mie theory [32].

The MgO-TiO$_2$ permittivity was assumed to be dispersionless with a dielectric constant of 16 and a dielectric loss factor of $(1.12-1.17) \times 10^{-4}$. By calculating the hybridized eigenmodes we get the extinction spectra shown in Fig. 5(a), which accurately reproduces the experiment’s extinction spectrum and Fano resonance. The discrepancy between the hybridization theory and experiment at high frequencies is due to the presence of a known [29, 30] magnetic quadrupole response in the individual spheres, which we simply do not take into account in our hybridization. Regarding, however, the Fano resonance at 4.8 GHz, we are able to decompose the simulated extinction spectrum into components coming from each isolated eigenmode, as seen in Fig. 5. The extinction depicted for each eigenmode is “isolated” in the sense that we are neglecting the extinction that can be attributed to the interference between eigenmodes [18]. The decomposition clearly shows that there are multiple eigenmodes that interfere destructively to form the main Fano resonance at 4.8 GHz. Moreover, modal interference is occurring at very similar frequencies in both the $E'$ and $E''$ responses; the Fano feature is coming from interference between $|E_{(a)}|^2$ and $|E_{(b)}|^2$ and between $|E_{(c)}|^2$ and $|E_{(d)}|^2$.

It is worth emphasizing that this is a simultaneous overlap of two optical Fano resonances, which are symmetrically exclusive by the nature of their distinct irreducible representations. The likely reason for this situation is that 4.8 GHz is also the frequency of an individual sphere’s magnetic dipole resonance, and at least one interfering eigenmode in both the $E'$ and $E''$ responses is dominated by magnetic dipoles. In other words, we have eigenmodes in both irreducible representations that are dependent on a single parameter, the magnetic dipole polarizability of a single MgO-TiO$_2$ sphere.

However, more generally, the correlation between a resonance of the single particle and modal interference is a recurring feature of the experiment. The single particle’s electric resonance at 6.5 GHz coincides with destructive interference in the $E'$ response, between the $|E_{(1)}|^2$ and $|E_{(2)}|^2$ eigenmodes. In Figs. 4(b) and 4(c), the single sphere’s magnetic quadrupole resonance at 7 GHz is also associated with a Fano resonance feature. It is interesting that the eigenmodes in our $E'$ response exhibit anomalous, resonant behavior in the vicinity of the (neglected) magnetic quadrupole resonance. Indeed, it is very likely that this frequency range should have significant multipolar coupling channels [27] that have been omitted in our hybridization. However, for the purposes of the work here, the key result is the largest Fano resonance at 4.8 GHz, which is fully described by our hybridization theory. This Fano resonance is a realization of the predicted propensity that all-dielectric trimers have towards Fano resonances.

IV. CONCLUSIONS

We have presented an explicit study on the hybridization of optical responses in both plasmonic and all-dielectric trimers. We presented a simplified hybridization model to allow us to derive the eigenmodes of these structures analytically and observed the formation of modal interference in eigenmodes excited by normally incident excitation. In this regard, plasmonic trimers were shown to exhibit nonorthogonality from retarded coupling channels, and dielectric trimers could further utilize bianisotropic coupling channels. A key prediction of our hybridization theory was then demonstrated experimentally: an all-dielectric trimer was shown to exhibit sharp, polarization-independent Fano resonances. The measurements were in good agreement with our simplified hybridization model and could therefore validate our approach. Our
conclusions subsequently demonstrate that the use of full plasmonic hybridization is not necessary to deduce the dominant eigenmodes of multiparticle geometries. Yet, importantly, our analysis has demonstrated that hybridized eigenmodes can be related to the set of intercoupled basis vectors in a simple form only if the basis vectors are, in isolation, eigenmodes of their associated subsystem. Indeed, this raises a more general point of contention against many empirical applications of hybridization concepts and establishes why a more considered utilization of hybridization concepts in terms of subsystem eigenmodes is necessary.

ACKNOWLEDGMENTS

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APPENDIX A: PLASMONIC TRIMER

The dipole model we use for a plasmonic trimer assumes only electric dipoles from the individual nanoparticles.

\[ \mathbf{p}_i = \alpha_i e_0 \mathbf{E}(\mathbf{r}_i) + \alpha_i k^2 \sum_{j \neq i} \mathbf{G}_0(\mathbf{r}_i, \mathbf{r}_j) \cdot \mathbf{p}_j. \]  

(A1)

where the coupling channels, A–D, are defined in Eq. (2) of the main text and \( \lambda_{B_{12}}, \lambda_{B_{13}}, \lambda_{B_{23}}, \) and \( \lambda_{A_b} \) are the eigenvalues of the corresponding eigenmodes in the isolated dimer.

\[ \lambda_{B_{12}} = (\alpha_{e} e_0)^{-1} + \frac{\epsilon_{d} k^2}{2 \pi e_0 R} \left( \frac{ik}{R} - \frac{1}{R^2} \right). \]  

(A5)

\[ \lambda_{B_{13}} = (\alpha_{e} e_0)^{-1} + \frac{\epsilon_{d} k^2}{4 \pi e_0 R} \left( \frac{ki}{R} - \frac{1}{R^2} \right). \]  

(A6)

\[ \lambda_{B_{23}} = (\alpha_{e} e_0)^{-1} - \frac{\epsilon_{d} k^2}{4 \pi e_0 R} \left( \frac{ki}{R} - \frac{1}{R^2} \right). \]  

(A7)

\[ \lambda_{A_b} = (\alpha_{e} e_0)^{-1} - \frac{\epsilon_{d} k^2}{2 \pi e_0 R} \left( \frac{ik}{R} - \frac{1}{R^2} \right). \]  

(A8)

These eigenvalues account for self-interaction of the dimer eigenmodes in the above matrix equation.

Here \( \alpha_e \) is the electric dipole polarizability of a particle, and \( \mathbf{G}_0 \) is the free-space dyadic Green’s function, which acts on dipole moments as

\[ \mathbf{G}_0(\mathbf{r}', \mathbf{r}) = \frac{\epsilon(ikR)}{4\pi R} \left( \left( 1 + \frac{i}{k R} - \frac{1}{k^2 R^2} \right) \mathbf{p} \right. \]

\[ \left. - \left( 1 + \frac{3i}{k R} - \frac{3}{k^2 R^2} \right) (\mathbf{n} \cdot \mathbf{p} \mathbf{n}) \right]. \]

where \( k \) is the wave number, \( \mathbf{n} \) is the unit vector pointing from \( \mathbf{r} \) to \( \mathbf{r}' \), and \( R = |\mathbf{r} - \mathbf{r}'| \). The associated eigenmode equation for a plasmonic trimer in this dipole model can then be written as

\[ \mathbf{v}_i = \alpha_e e_0 \mathbf{E}_i + \sum_{j \neq i} \alpha_j e_0 \mathbf{G}_0(\mathbf{r}_i, \mathbf{r}_j) \cdot \mathbf{v}_j. \]  

(A2)

If we define our eigenmode using the dimer and single-particle eigenmodes as basis vectors, we can write any associated eigenmode as

\[ |\mathbf{v}_i \rangle = a_i |\mathbf{p}_i \rangle + \sum_{j \neq i} c_j |\mathbf{B}_{i j} \rangle + d_i |\mathbf{B}_{i k} \rangle + e_i |\mathbf{B}_{i l} \rangle \]

(A3)

The six basis vectors are the dimer and single-particle eigenmodes we define in the main text. The eigenmode equation (A2) can then be written as a 6 × 6 matrix equation in terms of these basis vectors and the associated coupling channels:

\[ \begin{pmatrix} 1 & -A & -B & 0 & 0 & 0 \\ -A & \alpha_{e} e_0 \lambda_{B_{12}} & 0 & 0 & 0 & 0 \\ -B & 0 & \alpha_{e} e_0 \lambda_{B_{13}} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & -C \\ 0 & 0 & 0 & -C & \alpha_{e} e_0 \lambda_{B_{23}} & 0 \\ 0 & 0 & 0 & 0 & -D & \alpha_{e} e_0 \lambda_{A_b} \end{pmatrix} \begin{pmatrix} a_i \\ b_i \\ c_i \\ d_i \\ e_i \\ f_i \end{pmatrix} = \begin{pmatrix} a_i \\ b_i \\ c_i \\ d_i \\ e_i \\ f_i \end{pmatrix}. \]  

(A4)

The six solutions of Eq. (A4) provide the eigenmodes and eigenvalues for the plasmonic trimer in this dipole model. However, in regard to the nonorthogonality of \( |v_1 \rangle \) and \( |v_3 \rangle \), it is worth acknowledging that this nonorthogonality requires retarded coupling between the particles. Moreover, if we refer to the matrix in Eq. (A4) and the definitions of coupling channels in Eq. (2), we note that by neglecting retardation in the coupling between dipoles, we make the phase acquired through all coupling channels simply equal to that provided by the dipole polarizability. In other words, \( \lambda_{B_{12}}, \lambda_{B_{13}}, \lambda_{B_{23}}, \) and \( \lambda_{A_b} \) all become real numbers. This then makes the matrix in Eq. (A4) proportional to a real symmetric matrix, and hence Hermitian, which then guarantees it has orthogonal eigenvectors. As we already have orthonormal basis vectors, this then makes all eigenmodes orthogonal. As such, this shows that the retardation of coupling between particles is necessary for nonorthogonal eigenmodes.
APPENDIX B: DIELECTRIC TRIMER

To model the dielectric trimer, we use the coupled electric and magnetic dipole approximation, which is described by the following two (coupled) equations:

\[
p_i = \alpha_E e_0 \mathbf{E}_d(\mathbf{r}_i) + \alpha_E k^2 \left( \sum_{j \neq i} \hat{G}_d(\mathbf{r}_i, \mathbf{r}_j) \cdot \mathbf{p}_j - \frac{1}{\varepsilon_0} \nabla \times \hat{G}_d(\mathbf{r}_i, \mathbf{r}_j) \cdot \mathbf{m}_j \right),
\]

\[
\mathbf{m}_i = \alpha_H H_0(\mathbf{r}_i) + \alpha_H k^2 \left( \sum_{j \neq i} \hat{G}_d(\mathbf{r}_i, \mathbf{r}_j) \cdot \mathbf{m}_j + \varepsilon_0 \nabla \times \hat{G}_d(\mathbf{r}_i, \mathbf{r}_j) \cdot \mathbf{p}_j \right),
\]

where \( \mathbf{p}_i \) (\( \mathbf{m}_i \)) is the electric (magnetic) dipole moment of the \( i \)th particle, \( \hat{G}_d(\mathbf{r}_i, \mathbf{r}_j) \) is the free-space dyadic Green’s function between the \( i \)th and \( j \)th dipoles, \( \alpha_E (\alpha_H) \) is the electric (magnetic) polarizability of a particle, \( \varepsilon_0 \) is the speed of light, and \( k \) is the free-space wave number. The extra bianisotropic coupling terms are given according to

\[
\nabla \times \hat{G}_d(\mathbf{r}', \mathbf{r}) \cdot \mathbf{p} = \frac{e^{ik\mathbf{R}}}{4\pi R} \left( 1 + \frac{i}{kR} \right) \mathbf{n} \times \mathbf{p}.
\]

An eigenmode of the dielectric trimer, having electric dipoles \( \mathbf{v} \) and magnetic dipoles \( \mathbf{u} \), will therefore satisfy the coupled electric and magnetic dipole model [Eq. (B1)] as

\[
v_i = \alpha_E e_0 \lambda \mathbf{v}_i + \alpha_E k^2 \left( \sum_{j \neq i} \hat{G}_d(\mathbf{r}_i, \mathbf{r}_j) \cdot \mathbf{v}_j - \frac{1}{\varepsilon_0} \nabla \times \hat{G}_d(\mathbf{r}_i, \mathbf{r}_j) \cdot \mathbf{u}_j \right),
\]

\[
u_i = \alpha_H \mathbf{u}_i + \alpha_H k^2 \left( \sum_{j \neq i} \hat{G}_d(\mathbf{r}_i, \mathbf{r}_j) \cdot \mathbf{u}_j + \varepsilon_0 \nabla \times \hat{G}_d(\mathbf{r}_i, \mathbf{r}_j) \cdot \mathbf{v}_j \right).
\]

We can then use the dimer and single-particle basis vectors from Fig. 3 in the main text to write an expression that will describe the eigenmodes of a dielectric trimer that are invariant under the dimer’s reflection symmetry operation:

\[
|u_j\rangle = a'_i|p_j\rangle + b'_i|B_{2a}\rangle + c'_i|A_{2}\rangle + d'_i|E'_z\rangle.
\]

Analogous to the case for plasmonic trimers in Appendix A, we can rewrite Eq. (B3) as a \( 4 \times 4 \) matrix equation:

\[
\begin{pmatrix}
1 & -C & -D & -E_2 \\
-C & \alpha_E e_0 \lambda_{B_0} & 0 & -F_2 \\
-D & 0 & \alpha_E e_0 \lambda_{A_0} & 0 \\
-E_1 & -F_1 & 0 & \alpha_H \lambda_{E_0}
\end{pmatrix}
\begin{pmatrix}
a'_i \\
b'_i \\
c'_i \\
d'_i
\end{pmatrix}
= \lambda
\begin{pmatrix}
a'_i \\
b'_i \\
c'_i \\
d'_i
\end{pmatrix},
\]

where the coupling channels, \( C-F \), are defined in Eqs. (2) and (9) of the main text and the eigenvalue of the dimer eigenmode with \( z \)-oriented magnetic dipole moments is

\[
\lambda_{E_z} = a''_0^{-1} + \frac{e^{ikR}}{4\pi R} \left( k^2 + \frac{ik}{R} - \frac{1}{R^2} \right).
\]

We can then reduce Eq. (B5) to a \( 3 \times 3 \) matrix equation when searching for only the doubly degenerate eigenmodes given the eigenmode with radially oriented dipole moments \( |u_j\rangle \) remains an eigenmode of Eq. (B5). This is done by considering the eigenspace that is orthogonal to \( |v_j\rangle \). In effect, this means we substitute \( b'_i = \sqrt{2}a'_i - \sqrt{3}c'_i \) into Eq. (B5) to get the reduced matrix equation:

\[
\begin{pmatrix}
1 - \sqrt{2}C & \sqrt{3}C & -D & -E_2 \\
-D & \alpha_E e_0 \lambda_{A_0} & 0 & -F_2 \\
-E_1 - \sqrt{2}F_1 & \sqrt{3}F_1 & \alpha_H \lambda_{E_0}
\end{pmatrix}
\begin{pmatrix}
a'_i \\
c'_i \\
d'_i
\end{pmatrix}
= \lambda
\begin{pmatrix}
a'_i \\
c'_i \\
d'_i
\end{pmatrix},
\]

From this expression, it is straightforward to find numerical solutions to this matrix equation and obtain both the eigenmodes and eigenvalues. However, as an addendum, we can consider the relations between the \( a', b', c' \) coefficients of eigenmodes analytically. This will allow us to illustrate the interdependences of the collective eigenmodes on the resonances of the individual constituent subsystems. Moreover, we can use the second row of Eq. (B7) to first write a relationship between the \( a' \) and \( c' \) coefficients:

\[
-Da'_i = (\lambda_i - \lambda_{E_0}) a_E e_0 c'_i.
\]
From this relation, and using $F_1 = \sqrt{2}E_1$, the third row of Eq. (B7) can also provide a relationship between $c'$ and $d'$ coefficients:

$$\begin{align*}
(\lambda_i - \lambda_E)D_\alpha H d_i' & = E_1(3(\lambda_i - \lambda_A)\alpha E' + D\sqrt{6}c_i').
\end{align*}$$

These two relations are not sufficient to define eigenmodes analytically because we need the first row of Eq. (B7) to solve for $\lambda_i$, which becomes quite nontrivial. However, for our purposes, Eqs. (B8) and (B9) are sufficient to define the profile of an eigenmode for its given eigenvalue. Indeed, these two relations nicely illustrate how the hybridized eigenmodes become dependent on each basis vector’s resonant properties and all the coupling channels.

A.6 Circular dichroism induced by Fano resonances in planar chiral oligomers

Summary

The widespread desire for strong optical interactions with chiral molecules and proteins, present in many forms of biologicals and pharmaceuticals, has made nanoscale chiral materials and particles some of the most imminently applicable devices in the broader field of optical metamaterials. However, it is relatively well-accepted that surfaces which are chiral in only two-dimensions cannot exhibit circular dichroism at normal incidence, a common metric used to gauge the total chiral response of a scattering object or medium. In this work, a new form of circular dichroism that can exist in planar systems is presented, and is able to change the dominant loss mechanism of a plasmonic nanostructure between far-field radiation and near-field material absorption, both of which are key (and easily observable) forms of chiral response. To introduce this as a new circular dichroism effect, a ground-up description of its origin is provided, and it is shown that this form of circular dichroism is inherently linked to modal interference, and it can therefore be amplified through Fano resonances. This allows us to intuitively identify and demonstrate planar chiral oligomers as an obvious nanoparticle geometry that will exhibit this circular dichroism effect.

Notes and Errata

- The equation (14) for the dipole polarisability of a current voxel is missing a factor of $\epsilon_0$; the correct expression is in the text between (2.14) and (2.15). The cascaded effects of this mistake are that the expressions for absorption in equation (16) is also missing a factor of $\epsilon_0$; the correct expression is (2.15) after relating power $P$ to cross-section $\sigma$ as: $\frac{1}{2} \sqrt{\frac{\omega}{\mu_0}} |E_0|^2$. 
Abstract We present a general theory of circular dichroism in planar chiral nanostructures with rotational symmetry. It is demonstrated, analytically, that the handedness of the incident field's polarization can control whether a nanostructure induces either absorption or scattering losses, even when the total optical loss (extinction) is polarization-independent. We show that this effect is a consequence of modal interference so that strong circular dichroism in absorption and scattering can be engineered by combining Fano resonances with planar chiral nanoparticle clusters.

Circular dichroism induced by Fano resonances in planar chiral oligomers

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1. Introduction

The difference in absorption of left- and right-handed circularly-polarized (LCP and RCP) light by chiral structures has long been utilized for applications in molecular chemistry [1, 2], pharmaceuticals [3, 4], and optics [5, 6]. More recently, advances in nanotechnology have resulted in new types of chiral nanoantennas, colloidal nanoparticles, and metamaterials; nanostructured systems which provide unprecedented freedom to produce chiral responses that substantially exceed those of conventional materials. [7–13]

It is, however, planar nanostructures that have the least restrictions in design, particularly when considering fabrication, and, therefore, they are more suitable to capitalize on design freedom and produce the most effectual chiral responses. Yet, such two-dimensional nanostructures cannot be truly chiral because they have an inherent plane of mirror symmetry. In combination with reciprocity, this is known to prevent any difference in the total optical losses under LCP or RCP incident fields – a difference known as circular dichroism. Here, however, we consider separate contributions from the radiative and dissipative components of a system’s total loss. As we show, these components of the total loss, the scattering and absorption cross-sections, are not constrained by reciprocity and can subsequently exhibit circular dichroism in planar chiral geometries and metasurfaces (see Fig. 1). Here we focus on circular dichroism in the absorption cross-section. This definition is warranted from a practical perspective as, for instance, the absorption cross-section will encompass nonlinearity [14], photocurrent generation [15], heating [16], fluorescence [17] and photocatalysis [18]; quantities that are both measurable and foreseeably useful.

The effect of chirality on scattering and absorption is generally understood by how it impacts either the far-field scattering matrices or the effective medium parameters of a given structure [19, 20]. However, these two approaches have been tailored (historically) towards characterizing the chiral properties of conventional materials and do not provide information on what is physically occurring in the system. This means that it can be highly nontrivial to deduce how a general change in geometry will affect the far-field scattering matrices or effective parameters. In an attempt to address this, some recent works have considered the properties of the near-field of certain chiral structures to guide design empirically [21] or attempted to describe the response of chiral arrangements of nanorod dimers analytically [22,23]. Note here, the relation between the chiral response and the geometry is ultimately rooted in the current distribution that is induced by the given incident field. Specifically, these currents will encapsulate the complete optical response of any system in both the near- and far-field regions, and should thereby provide a deeper understanding of the observed optical responses. The approach we present here will be to simplify the analysis of structures using geometric symmetry rather than analytical approximations.

In this regard, there have been significant works on planar chiral metasurfaces and metamaterials whose constituent meta-atoms have a discrete rotational symmetry [24,25]. A number of key advantages of utilizing rotational symmetry in applications that require polarization-selective operation have further been proposed. In particular: invariance of
scattering and absorption loss to all linear polarizations\(^1\) [26], zero transmitted circular cross polarization [27] and others [28–31]. Therefore, while imposing symmetry conditions necessarily restricts the range of chiral nanostructures that can be considered for a given application, the remaining subset of nanostructures (which are both chiral and rotationally-symmetric at the same time) are desirable and have been just as effective at exhibiting chiral optical responses. We have subsequently focused here on symmetries that are rotationally symmetric and lack any mirror plane parallel to the rotational symmetry axis. We do not consider geometries that have a mirror plane parallel to their rotational symmetry axis, such as in the \(C_{nv}\) and \(D_{nh}\) point groups. This is because such mirror symmetry operations are able to transform an LCP plane wave into an RCP plane wave (and vice versa), thereby leaving the geometry unchanged when flipping the handedness of the excitation. This would subsequently make the optical responses to LCP and RCP plane waves symmetrically-equivalent. We therefore consider the following symmetry groups: \(C_n\), \(D_n\), \(S_n\) and \(C_{nh}\) (see Fig. 2). While the \(C_n\) and \(D_n\) point groups are chiral, it is important to acknowledge that \(S_n\) and \(C_{nh}\) point groups are, in fact, achiral; a result of their symmetry under improper rotation and planar mirror reflection operations, respectively. We focus on the \(C_{nh}\) point group in the main text, corresponding to geometries that are chiral in only two dimensions, a property which is often referred to as planar chiral. We also consider only the incident waves propagating along the symmetry axis. In this paper, we demonstrate how symmetry considerations can allow us to analytically extricate meaningful information from the currents induced in \(C_{nh}\) structures without calculating the explicit current distributions. From this foundation, we are able to link the origin of non-reciprocal circular dichroism in the absorption cross-section to modal interference and Fano resonances.

\(^1\) This is necessary to remove the cross-section’s dependence on the phase between LCP and RCP light.
and the free space Green’s function
\[ \tilde{G}(x, x') = \frac{\omega^2 \mu_0 P.V.}{k^2} \left( \frac{1}{k^2} \nabla \cdot \nabla \right) e^{i k |x-x'|} - \frac{\delta(x-x')}{\epsilon_0}. \]  
(2)

where \( \omega \) is the angular frequency of light and P.V. implies a principle value exclusion of \( x = x' \) when performing the integration in Eq. 1. For ease of notation, we can also define a tensor permittivity \( \bar{\varepsilon} \) in terms of the conductivity \( \bar{\sigma} \) and electric susceptibility \( \bar{\chi} \) to relate the induced current to the total electric field
\[ \bar{\varepsilon} \equiv (\bar{\chi} + 1)\epsilon_0 - \frac{\bar{\sigma}}{i\omega}. \]
\[ \Rightarrow J(x) = -i\omega (\bar{\varepsilon}(x) - \epsilon_0) E(x). \]  
(3)

As we are only considering \( x \) in the volume \( V_i \) with non-background permittivity, we can rewrite Eq. 1 as an integral equation for the induced current
\[ i\omega \epsilon_0 E_i(x) = -[\bar{\varepsilon}(x) - \epsilon_0]^{-1} J(x) + \int_{V_i} \tilde{G}(x, x') J(x') \, dx'. \]  
(4)

This integral equation is applicable to the fields of any arbitrary structure in the absence of magnetization. Now, for our purposes, Eq. 4 has an associated eigenmode equation, which has solutions \( v_i \) that satisfy
\[ i\omega \epsilon_0 v_i(x) = -[\bar{\varepsilon}(x) - \epsilon_0]^{-1} v_i(x) \]
\[ + \int_{V_i} \tilde{G}(x, x') v_i(x') \, dx'. \]  
(5)

These eigenmodes can be calculated numerically for simple systems based on discrete dipole approximation [33], but also for arbitrary continuous structures [34]. Now we can apply symmetry analysis to the structure under consideration. Each irreducible representation of the structure’s highest-order symmetry group will describe the transformation properties of a distinct set of eigenmodes. In the absence of accidental degeneracies, each of these eigenmodes will have a unique eigenvalue with a degeneracy level equal to the dimension of the associated irreducible representation [35]. For our purposes we are considering the \( C_n, S_n, D_n \) and \( C_{nh} \) symmetry groups, and, specifically, the two-dimensional \( E \) irreducible representations, which describe the transformation properties of any normally-incident planewave \( (E_0 \text{ in Eq. 4}) \) under the given group’s symmetry operations. Because of this, only the eigenmodes which transform according to these \( E \) irreducible representations can be excited. It is now important to notice here that we are working in a complex space. It implies that the two-dimensional \( E \) irreducible representations of the \( C_n, S_n \) and \( C_{nh} \) groups are written as two, one-dimensional irreducible representations that are complex conjugates of each other (for example, see Table 1). In regard to notation, we will refer to these two, one-dimensional irreducible representations as \( E^+ \) and \( E^- \) (i.e. where \( E^{+*} = E^- \)).

Taking into account that \( E^+ \) and \( E^- \) are different irreducible representations, each will describe the transformation properties of a distinct set of eigenmodes, which we refer here to as \( [\psi^+] \) and \( [\psi^-] \), respectively. From the geometrical symmetry perspective, the eigenmodes associated with different irreducible representations should not be degenerate [35]. However, such eigenmodes can become degenerate in some systems where additional symmetries are present. One of the key symmetries of electromagnetic theory that is neglected in a purely geometric argument is Lorentz reciprocity\(^2\). In the following we will prove that every eigenmode in \( E^+ \) will always be degenerate with one eigenmode of the associated \( E^- \) (and vice versa), purely due to the inherent Lorentz reciprocity of the Maxwell’s equations.

We start by taking an eigenmode \( (\psi^+) \), which transforms according to a one-dimensional \( E^+ \) irreducible representation. We also impose the standard normalization condition
\[ \int_V \psi_i^{++} \cdot \psi_i^+ \, dV = 1, \]  
(6)

where the dot denotes a vector dot product. Importantly, if \( \psi_i^+ \) transforms under symmetry operations according to \( E^+ \), we know that \( \psi^{++} \) must transform according to the complex conjugate irreducible representation, \( E^- \), because any operator describing a geometric symmetry operation in a Euclidean space will be real. We can therefore write \( \psi^{++} \) as some linear combination of the eigenmodes, \( [\psi^+] \), which transform according to the \( E^- \) irreducible representation
\[ \psi_i^{++} = \sum_j b_{ij} \psi_j^+, \quad b_{ij} \in \mathbb{C}. \]  
(7)

Notes \( \phi = e^{i\omega t} \)

| \hline
| \hline
| \hline
\end{tabular}

Table 1 The character table of the 3-fold rotational \( (C_3) \) symmetry group. Note here, the two-dimensional \( E \) representation is made up of two, one-dimensional irreducible representations that are complex conjugates of each other.

\( C_3 \) | \( E \) | \( C_3 \) | \( C_3^2 \) \\
--- | --- | --- | --- \\
\( A \) | 1 | 1 | 1 \\
\( E \) | 1 | \( \phi \) | \( \phi^2 \) \\
\( \phi \) | 1 | \( \phi^2 \) | \( \phi \) \\

\( ^2 \) Lorentz reciprocity itself can be seen as an application of the general Onsager reciprocity principle to electromagnetics, which in turn follows from the behavior of dissipative equilibrium systems under the time reversal operation [36]. However, for the discussion here, it is more convenient to use Lorentz reciprocity argument directly.
By substituting Eq. 7 into Eq. 6 we can rewrite the normalization of $v_i^+$ as

$$\sum_j b_{ij} \left( \int_{V_i} v_j^+ \cdot v_i^+ \, dV \right) = 1.$$  \hspace{1cm} (8)

For Eq. 8 to hold we know that there must be at least one $j$ such that

$$\int_{V_i} v_j^+ \cdot v_i^+ \, dV \neq 0.$$  \hspace{1cm} (9)

We now consider the role of the Lorentz reciprocity, a consequence of which is that both the dyadic Greens function and the permittivity tensor must be symmetric, albeit complex and not necessarily Hermitian

$$\tilde{G}(x, x') = \tilde{G}(x', x), \quad \tilde{\varepsilon} = \tilde{\varepsilon}^T, \quad \tilde{\varepsilon} = \tilde{\varepsilon}^T.$$  \hspace{1cm} (10)

Due to this symmetry, it is possible to write the overall operator of the eigenvalue equation (Eq. 5) as a matrix in the normal form shown by Gantmacher [37]. A result of this is that nondegenerate eigenmodes must be orthogonal under unconjugated projections [38]

$$\int_{V_s} v_i \cdot v_\beta \, dV = 0 \quad \text{when} \quad \lambda_i \neq \lambda_\beta.$$  \hspace{1cm} (11)

Hence Eq. 9 can be true only for some $v_i^+$ if there exists an eigenmode $v_i^-$ that is degenerate with $v_i^+$. Moreover, we must always be able to find at least one $v_i^-$ that satisfies Eq. 9 for each $v_i^+$. However, suppose now that multiple $v_i^-$ satisfied Eq. 9 for the one $v_i^+$; this would mean that each of these $v_i^-$ are degenerate with every other $v_i^-$. Such unenforced degeneracies between eigenmodes are accidental degeneracies and can be excluded from the general consideration. We can therefore expect each $v_i^-$ to be degenerate with a single $v_i^+$ (and vice versa). For ease of notation, we will use the subscript convention that $v_i^+$ is degenerate with $v_i^-$. We can now combine Eqs. 6 and 7 to write the following relation

$$b_{ij} \int_{V_i} v_j^+ \cdot v_i^- \, dV = \delta_{ij},$$  \hspace{1cm} (12)

where $\delta_{ij}$ is a Kronecker delta function. This completes the proof, we have shown that Lorentz reciprocity forces degeneracy between pairs of eigenmodes of complex conjugate irreducible representations in any symmetry group. For our purposes, we will specifically identify the $E^+$ irreducible representations of $C_n$, $S_n$, and $C_{nh}$ symmetry groups ($n \geq 3$) as being subject to this argument (the degeneracy already exists in $D_n$ from geometry alone). This result is applicable to all associated symmetric scattering systems irrespective of their specific dimensions or constituent materials.

To emphasize the significance of this degeneracy, we have to relate the eigenmodes to the applied fields that excite them. The behavior of the $E^+$ and $E^-$ irreducible representations in $C_n$, $S_n$, and $C_{nh}$ symmetry groups is that symmetric rotations are described by uniform phase shifts that are complex conjugates of each other. At normal incidence, it is the RCP and LCP plane waves that behave in this way and they can be, therefore, assigned to $E^+$ and $E^-$, respectively. As such, we can define the eigenmodes $\{v_i^+\}$ to correspond to those excited by an RCP plane wave and $\{v_i^-\}$ to those excitable by an LCP plane wave. Therefore the degeneracy we have just proven means that the modes that can be excited by LCP and RCP plane waves are degenerate. As such, changing the polarization of the incident plane wave between LCP and RCP cannot result in the excitation of modes that were previously forbidden by symmetry (dark modes). Moreover, the degeneracy of left- and right-handed eigenmodes highlights an important conceptual point on why the handedness of the incident field is able to affect optical response. While LCP and RCP plane waves are enantiomers, the degenerate eigenmodes they respectively excite cannot themselves be enantiomers because the geometry is not conserved under reflections. This permits nontrivial differences to exist in the distribution of eigenmodes that are excited by LCP and RCP plane waves. In other words, circular dichroism and other chiral scattering effects will be due to the varying magnitudes and phases of the excitations of degenerate eigenmodes. To now derive physically meaningful information from the current distributions, we need to calculate the extinction and absorption cross-sections to define circular dichroism. These two cross-sections can be calculated from current distributions similar to point-dipole systems in Ref. [39] by defining an infinite number of point dipoles $\{p\}$, where each dipole moment is defined for some infinitesimal volume $dV$ over which $J$ can be considered as constant

$$p_x = -\frac{1}{i \omega} \int_{dV} J(x')dV = -\frac{1}{i \omega} J(x) dV.$$  \hspace{1cm} (13)

Using the definition for induced current in Eq. 3 we can further define the polarizability of these dipoles as

$$\bar{\alpha}_x = |\tilde{\varepsilon}(x) - \varepsilon_0| \, dV.$$  \hspace{1cm} (14)

The extinction can then be determined from the projection of the complex conjugated incident field onto the induced current

$$\sigma_{\text{ext}} = \frac{k}{|E_0|^2} \varepsilon_0 \text{Im} \left\{ \sum_x E_0^x \cdot p_x \right\} = \frac{1}{|E_0|^2} \varepsilon_0 \left( \text{Im} \left\{ \int_{V_s} E_0^x \cdot J \, dV \right\} \right).$$  \hspace{1cm} (15)

Similarly, we can express the absorption in terms of the intensity of the induced currents by keeping only the lowest
order of $dV$

$$
\sigma_{\text{abs}} = -\frac{k}{\varepsilon_0 |E_0|^2} \sum_{\lambda} p_v^\lambda \cdot \left( \Im \left( \hat{a}_{s}^{-1} \right) + \frac{k^2}{6\varepsilon_0} \right) \cdot p_s.
$$

Hence, it is straightforward now to calculate the circular dichroism in extinction and absorption cross-sections from the currents that are induced by circularly-polarized plane waves with opposite handeness. Importantly, due to the optical theorem, the extinction cross-section depends only on the forward scattering amplitude whereas the scattering and absorption cross-sections depend on the far-field scattering at all angles (or, equivalently, on the full structure of the near-field). This already suggests that the scattering and absorption cross-sections will be more sensitive to the polarization in the structures with reduced symmetry, a hypothesis which we will confirm in the coming arguments.

3. Planar chiral oligomers

Circular dichroism is traditionally defined as a difference in optical loss between LCP and RCP plane waves propagating in the same direction. However, given the freedom of two polarizations and two propagation directions, there are four distinct excitations that can be applied by circularly-polarized plane waves. These four circularly-polarized plane waves be can represented in terms of their polarization depicted spatially along the propagation direction, where each of them resembles either one of two oppositely-handed helices (seen in Fig. 3). The remaining distinction is then the direction such a ‘polarization helix’ rotates in time. This depiction of an incident field corresponds to the spatial distribution of the applied electric field, which is what we use in our integral equation approach for the induced currents (Eq. 4).

In the expressions for the applied electric field, seen as insets in Fig. 3(a)-(d), we use the notation of two superscript plus and/or minus signs to indicate the sign of the imaginary unit in the polarization vector and exponential, respectively. This notation highlights which plane waves are complex conjugates (reciprocal), e.g. $E_0^{(\pm \pm)}$ is the complex conjugate of $E_0^{(\mp \mp)}$.

The reason circular dichroism is defined for one propagation direction only (i.e. two, not four, excitations) is reciprocity, which equates the extinction cross-section of oppositely propagating plane waves and, thus, the difference in extinction cross-section between LCP and RCP plane waves does not depend on the propagation direction [40]. This invariance between reciprocal plane waves can be observed in the electric field distributions ($E_0(x)$) that are complex conjugates of each other. So, if we then start from the earlier analysis of the induced currents in structures with $C_{nv}$, $D_{nh}$, $S_h$ or $C_{nh}$ symmetry, we are able to express the current ($\mathbf{J}$) induced by a circularly-polarized plane wave ($E_0$) in terms of eigenmodes

$$
E_0 = \sum_{i} a_i \lambda_i w^+_i \Rightarrow \mathbf{J} = \sum_{i} a_i w^+_i. \quad (17)
$$

Using Eq. 7, we can also define the current ($\mathbf{J}'$) induced by the complex conjugate field ($\overline{E_0}$)

$$
E_0 = \sum_{i} a_i^\prime \lambda_i^\prime w_i^\prime \Rightarrow \mathbf{J}' = \sum_{i} a_i^\prime w_i^\prime. \quad (18)
$$

This shows that the overall extinction of each degenerate eigenmode pair is not necessarily the same between $E_0$ and $\overline{E_0}$. However, despite the change in eigenmode excitation, we can use the result in Eq. 12 with Eqs. 17 and 18 to equate the extinction (Eq. 15) of complex conjugate applied fields (as expected)

$$
\int_{V_i} \mathbf{E}^0 \cdot J \, dV = \int_{V_i} \overline{E_0} \cdot \mathbf{J}' \, dV. \quad (19)
$$

This, therefore, demonstrates that the total extinction of each nondegenerate eigenmode can change between reciprocal plane-waves while the extinction is conserved. The conservation of extinction means a structure with a planar reflection symmetry (such as in $C_{nh}$ symmetry) cannot exhibit circular dichroism in extinction. Rigorously, applying a symmetric reflection operator ($\delta_h$) to the global reference frame of the generic system in Eq. 17 will not change the scatterer’s geometry by definition, but the applied field and the induced current become $\delta_h E_0$ and $\delta_h \mathbf{J}$ when expressed in terms of the new reference frame. Given that it is a unitary operation, the $\delta_h$ operator then cancels when evaluating the extinction for $\delta_h E_0$ and $\delta_h \mathbf{J}$, leaving the extinction unchanged

$$
\int_{V_i} (\delta_h E_0) \cdot (\delta_h \mathbf{J}) \, dV = \int_{V_i} \overline{E_0} \cdot \mathbf{J} \, dV. \quad (20)
$$

As the reflection operator changes the propagation direction of the incident field while leaving the polarization unchanged, $\delta_h E_0$ corresponds to an oppositely-handed polarization helix to that of $E_0$ (cf. Fig. 3(a) and (b), and Fig. 3(c) and (d)). Therefore Eq. 20, in conjunction with Eq. 19, means that all four excitations from a circularly-polarized plane-wave will produce the same extinction cross-section and, subsequently, this shows that no circular dichroism can occur in extinction for structures with $C_{nh}$ symmetry. Our argument also holds for structures with $S_h$ symmetry if we substitute the $\delta_h$ reflection operation for the $S_h$ improper rotation operation. As such, we have actually shown that neither $C_{nh}$ or $S_h$ symmetries permit circular dichroism in extinction. More specifically, we demonstrated that this conserved extinction does not require there to be a trivial difference between the currents induced by reciprocal plane-waves.

Up to this point, the direction that an applied field rotates in time has been discounted because of the reciprocity
condition in Eq. 19. However it is worth acknowledging that this result is only applicable to extinction and, subsequently, in the presence of material losses, reciprocity does not constrain the absorption and scattering cross-sections independently. Therefore, by introducing material losses to produce an absorption cross-section, neither the absorption nor scattering cross-section are necessarily invariant under reciprocal plane-waves. This is of particular interest for planar chiral scattering geometries, such as those with $C_{nh}$ symmetry, which we have shown cannot produce circular dichroism in the extinction cross-section due to the combination of reciprocity and planar reflection symmetry.

To consider the origin of such an effect, we will begin by proving that the total induced current intensity can only change between reciprocal excitations if the structure’s eigenmodes are nonorthogonal. Specifically, if we assume that the scattering structure’s eigenmodes are orthogonal to each other, we can define the excitations of each eigenmode by substituting Eq. 23 into Eq. 7

$$
\left(\int_{V_1} v_{ji}^r \cdot v_{ji}^r \, dV\right)^* = \left(\int_{V_1} v_{ji}^l \cdot v_{ji}^l \, dV\right) = \left(\int_{V_1} v_{ji}^r \cdot v_{ji}^l \, dV\right)^*.
$$

At the same time, Eq. 23, in conjunction with Eq. 12, means that the real projection of one eigenmode onto its degenerate partner has unit magnitude

$$
\left(\int_{V_1} v_{ji}^r \cdot v_{ji}^l \, dV\right)^* = \left(\int_{V_1} v_{ji}^l \cdot v_{ji}^r \, dV\right)^*.
$$

To combine this all, we start from the definitions of $\mathbf{J}$ and $\mathbf{J'}$ in Eqs. 21 and 22, then use the relations in Eqs. 24 and 25, in addition to our original assumption that all eigenmodes are orthogonal, to get the result that

$$
\int_{V_1} \mathbf{J}^* \cdot \mathbf{J} \, dV = \int_{V_1} \mathbf{J'}^* \cdot \mathbf{J'} \, dV.
$$

For geometries made of isotropic materials, this means that circular dichroism in absorption (Eq. 16) ceases to exist between complex conjugate excitations. In other words, we have proven that absorption circular dichroism can only exist between reciprocal plane-waves if such geometries have nonorthogonal eigenmodes. This peculiar requirement means circular dichroism in absorption can be considered...
as an interference effect in the same sense that Fano resonances are. Specifically, in Ref. [33], it was shown that the nonzero projections of one eigenmode onto another eigenmode are able to fully describe Fano resonances. We can therefore establish a link between circular dichroism in absorption and this eigenmode overlap by relating it to the $b_{ij}$ coefficient using Eqs. 7 and 12

$$
\int \mathbf{v}_{i}^{+} \cdot \mathbf{v}_{j}^{+} \, dV = b_{ij} \int \mathbf{v}_{i}^{-} \cdot \mathbf{v}_{j}^{+} \, dV. \tag{27}
$$

This shows us that $b_{ij}$ is proportional to the overlap between the nondegenerate eigenmodes $\mathbf{v}_{i}^{\pm}$ and $\mathbf{v}_{j}^{\pm}$. If we then refer to Eqs. 17 and 18, we can see that the excitation of nondegenerate eigenmodes should vary more between reciprocals excitations given the presence of a large $b_{ij}$. In other words, we should observe significant circular dichroism in absorption and scattering cross-sections when there is a large eigenmode overlap. Moreover, a large eigenmode overlap is known to exist, quite prominently, at a Fano resonance. So, to produce circular dichroism in absorption, we can take a structure known to produce Fano resonances and alter it to be planar chiral. In Fig. 4 we begin with a gold heptamer which is known to support Fano resonances [41] and then alter the nanoparticles in the outer ring to make it both planar chiral and having $C_{2v}$ symmetry. The choice of parameters was chosen so that the central particle resonance is overlapped with that of the outer ring [42]. As the outer ring in Fig 4 is a planar chiral structure, it will experience different current distributions in response to LCP and RCP plane waves (see Eqs. 17 and 18), but in isolation it does not experience significant circular dichroism. However, the collective structure exhibits a Fano resonance, which leads to significant circular dichroism in absorption and scattering. The extent of the circular dichroism is, in fact, sufficient to swap the dominance of scattering and absorption cross-sections using polarization. This therefore supports our derivation that nonreciprocal circular dichroism in absorption is an interference effect.

In Fig. 4, all nanoparticles are made from 20 nm thick gold, the central disk has a diameter of 140nm and each triangular nanoparticle has major and minor axes of 100 nm and 60 nm (respectively). The triangular nanoparticles have been placed at a radius of 170 nm away from the center of the disk and the major axis is oriented 65° off the radial vector. LCP and RCP are defined relative to a vector pointing towards the disk and the major axis is oriented 65° off the radial vector. LCP and RCP plane waves (see Eqs. 17 and 18), but in isolation it does not experience significant circular dichroism. This means, for instance, that dichroism between reciprocal, circularly-polarized plane-waves in absorption does not occur for a single isotropic material in the structure (i.e. if $\tilde{\mathbf{e}}(x) \rightarrow \epsilon$). Then, given $\mathbf{v}_{ij}^{+}$ is an eigenmode, we can utilize Eqs. 6 and 7 to get the result that

$$
b_{ij} = \delta_{ij} \quad \text{and} \quad \mathbf{v}_{ij}^{-} = \mathbf{v}_{ij}^{+}. \tag{30}
$$

From Eq. 11, this is sufficient to ensure that the eigenmodes are orthogonal. So, if a structure is made of a single isotropic material, nonorthogonal eigenmodes can only exist if there is only one uniform and isotropic material in the structure (i.e. if $\tilde{\mathbf{e}}(x) \rightarrow \epsilon$). This can be seen from the fact that any permittivity distribution $\tilde{\mathbf{e}}(x)$ represents the geometry and therefore has to be invariant under symmetry operations. So, if we were to define an incident field as

$$
\mathbf{E}_{0}(x) = - [\tilde{\mathbf{e}}(x) - \epsilon]^{-1} \mathbf{v}_{ij}^{+}(x). \tag{31}
$$

then this incident field would transform under symmetry operations according to $\mathbf{v}_{ij}^{+}(x)$ and we could subsequently express it as some linear combination of the eigenmodes $\{\mathbf{v}_{ij}^{\pm}\}$

$$
- [\tilde{\mathbf{e}}(x) - \epsilon]^{-1} \mathbf{v}_{ij}^{+}(x) = \sum_{j} u_{ij} \mathbf{v}_{j}^{+}(x). \tag{32}
$$

subsequently shows that circular dichroism in absorption is a robust and widely achievable feature of geometries that produce modal interference.

To investigate the associated physical dependencies of circular dichroism in the absorption cross-section, we need to identify a physical characteristic that causes nonorthogonal eigenmodes. This is in fact quite straightforward if we neglect the retardation of coupling within the structure, which can be done either resorting to the quasistatic approximation ($k \rightarrow 0$) or by working in the near-field limit ($k|x - x'| \rightarrow 0$) for small systems. In these cases, the free space Green’s function becomes entirely real. So we can take the complex conjugate of the eigenmode equation of our system (Eq. 5) to get the following equation

$$
i\omega_{\eta} \mathbf{v}_{ij}^{+}(x) = - (\tilde{\mathbf{e}}(x) - \epsilon_{0})^{-1} \mathbf{v}_{ij}^{+}(x)
+ \int d^{3}x' \tilde{\mathbf{G}}(x, x') \mathbf{v}_{ij}^{+}(x') \, d^{3}x', \tag{28}
$$

where

$$
\eta_{ij} = \frac{1}{i \omega_{\eta}} \left[ [\tilde{\mathbf{e}}^{\pm}(x) - \epsilon_{0}]^{-1} - (\tilde{\mathbf{e}}(x) - \epsilon_{0})^{-1} \right] - \lambda_{ij}. \tag{29}
$$

Notably, the complex conjugate of any eigenmode ($\mathbf{v}_{ij}^{+}$) will be an eigenmode in its own right if there is only one uniform and isotropic material in the structure (i.e. if $\tilde{\mathbf{e}}(x) \rightarrow \epsilon$). Then, given $\mathbf{v}_{ij}^{+}$ is an eigenmode, we can utilize Eqs. 6 and 7 to get the result that
Figure 4 Simulations demonstrating the role of interference for inducing circular dichroism in the absorption cross-section of a planar chiral heptamer with $C_{6v}$ symmetry. We observe the creation of significant circular dichroism in absorption in the vicinity of the Fano resonance, as is predicted given it corresponds to high modal overlap. Additionally, on the right hand side, we show that this circular dichroism can also be observed in the magnitude of the near-field chirality density [43] at the Fano resonance. Here the chirality density induced by an RCP plane-wave is less than half of that for the LCP case. The calculation parameters are given in text.

When neglecting retardation, $\vec{G}$ becomes real symmetric and, hence, the eigenmodes $\{\vec{g}\}$ are orthogonal and each coefficient $f_{ij}$ is the complex projection of $\vec{g}_i$ onto $\vec{v}_j$. The orthogonality of $\{\vec{g}\}$ in Eq. 33 then requires that there is at least one eigenmode $\vec{g}_l$ that exists in the linear combinations for both $\vec{v}_i^+$ and $\vec{v}_k^+$. Thus, we can explicitly write the complex inner product between $\vec{v}_i^+$ and $\vec{v}_k^+$ using their respective decompositions into the eigenmodes $\{\vec{g}\}$

\[
\int_{V_s} \vec{v}_i^+ \cdot \vec{v}_k^+ \, dV = \sum_l f_{il}^* f_{kl} \quad (where \ f_{il}, f_{kl} \neq 0).
\]

As such, the projection between eigenmodes will generally be nonzero. For instance, nonorthogonality would be guaranteed if there was only a single shared eigenmode ($\vec{g}_l$) in both $\vec{v}_i^+$ and $\vec{v}_k^+$. So, our original assumption, that all eigenmodes in $\{\vec{v}\}$ are orthogonal, is broken and we have therefore shown that at least one pair of nondegenerate eigenmodes will be nonorthogonal if the geometry consists of inhomogeneous and/or anisotropic materials. This also builds on our earlier result (Eq. 26) that circular dichroism in geometries made of isotropic materials can only occur when there are nonorthogonal eigenmodes, because the exception to that condition was the presence

Provided that $\vec{v}_i^+$ is not, by chance, an eigenmode of $(-\epsilon(x) - \epsilon)^{-1}$, there has to be at least one $k \neq i$ such that $u_{ik}$ is nonzero. Referring back to our original eigenmode equation (Eq. 5), this means that the Green’s function integral of $\vec{v}_i^+$ must produce equal and opposite components of each $\vec{v}_k^+$ to counterbalance the $\vec{v}_i^+$ components created by the permittivity distribution. Physically this means that inhomogeneous or anisotropic, lossy materials induce coupling between eigenmodes. To demonstrate that this makes at least one pair of eigenmodes nonorthogonal, we make the assumption that all eigenmodes are orthogonal. Then, substituting Eq. 32 into Eq. 5, requires that there is a nonzero projection between $\vec{v}_i^+$ and the scattered field of $\vec{v}_k^+$

\[
\int_{V_s} \vec{v}_i^+ \cdot (\int_{V_s} \vec{G}(x,x')\vec{v}_k^+ (x') \, d^3x') \, d^3x \neq 0.
\]

We can then consider each $\vec{v}_j^+$ as a linear combination of the eigenmodes $\{\vec{g}\}$ (with eigenvalues $\{\gamma\}$) of the Green’s function integral

\[
\vec{v}_i^+(x) = \sum_j f_{ij} \vec{g}_j(x)
\]

\[
\Rightarrow \int_{V_s} \vec{G}(x,x')\vec{v}_i^+ (x') \, d^3x' = \sum_j \gamma_j f_{ij} \vec{g}_j(x).
\]
of anisotropic materials, which we have just shown leads to nonorthogonal eigenmodes independently. Therefore we know that nonorthogonal eigenmodes are always necessary for nonreciprocal circular dichroism in absorption. Furthermore, we have derived that such nonorthogonal eigenmodes are a result of either retardation of coupling between currents in the structure or from anisotropic or inhomogeneous materials.

It is finally important to acknowledge that small differences in absorption and scattering have recently been observed numerically for a chiral oligomer with planar reflection symmetry [45]. Additionally, recent experimental observations have also reported a difference in heat generation from a planar chiral structure under excitation by LCP and RCP light [46]. These investigations support our derivations on the circular dichroism presented in this paper, but the observed difference in absorption was hypothesized to be a consequence of polarization-dependent near fields, which is not necessarily sufficient to produce a difference in the total absorption as shown in [26, 29]. It is nonetheless worth acknowledging that an alternate option is to measure circular dichroism in scattering and absorption from the far-field. As demonstrated in Ref. [47], the scattering and absorption cross sections can be experimentally measured from far-field light by using spatial modulation and interferometry to observe scattering from the interference of scattered field with the incident field. It has additionally been demonstrated in Ref. [48] that measuring the far-field extinction phase, in addition to amplitude, allows for measurement of the absorption cross section. Yet, we should also acknowledge that simpler measurements would be able to observe signs of circular dichroism from the scattering cross section. Measurements of transmission or reflection using a high numerical aperture lens have the capacity to capture a large portion of the scattering cross section and will therefore depend on LCP or RCP light. Indeed, there are a number of options to experimentally observe the circular dichroism presented here.

4. Conclusions

We have presented a rigorous analytical study of circular dichroism in nanostructures with rotational symmetry. It was shown that, because of reciprocity-enforced degeneracies, chiral scattering behavior cannot involve the excitation of modes that are inaccessible (dark) depending on polarization’s handedness. This observation led to the distinction of two forms of circular dichroism: the traditional form that originates from spatially-distinct excitations and is regularly observed in extinction, and the second form originating from spatially-identical excitations that rotate in opposite directions temporally (such as from reciprocal plane waves) and can be observed in absorption and scattering. To explain the peculiarities of the second form, we have shown that it can occur only if the scatterer has nonorthogonal eigenmodes. Necessary criteria for a scattering structure to have nonorthogonal eigenmodes was then shown to be retardation of coupling between currents and/or the use of multiple materials. Notably, these will also be necessary criteria for Fano resonances and other such modal interference effects that rely on nonorthogonal eigenmodes. A consequence of this analysis is that circular dichroism in absorption would be amplified at locations of significant modal interference, such as Fano resonances. To demonstrate a manifestation of this circular dichroism effect, we have proposed a planar chiral nanoparticle heptamer that exhibits a Fano resonance and observed that significant absorption circular dichroism occurs in the vicinity of the Fano resonance. Our conclusions subsequently suggest that there is a key relationship between the modal interference
and circularly-dichroic scattering in both linear and nonlinear responses of planar chiral systems.

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References

Appendix B

List of all publications


• G. Geraci, B. Hopkins, A.E. Miroshnichenko, B. Erkihun, D.N. Neshev, Y.S. Kivshar, S.A. Maier and M. Rahmani, Polarisation-independent enhanced scattering by tailoring asymmetric plasmonic systems, *Nanoscale* 8, 6021-6027 (2016)

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