Structured Light for Second-Harmonic Spectroscopy in Mie-Resonant AlGaAs Nanoparticles

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Abstract: We employ doughnut-shaped cylindrical vector beams to observe the enhanced second-harmonic generation from individual subwavelength AlGaAs nanoparticles which support both electric and magnetic multipolar Mie-type resonances at the fundamental and double frequencies.

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1. Introduction

Dielectric resonant nanophotonics and meta-optics are rapidly developing research areas with potential applications for novel on-chip light sources, wavefront-controlling metasurfaces, and ultrafast subwavelength metadevices [1]. A strong confinement of the electromagnetic fields inside resonant subwavelength dielectric structures leads to the increase of the efficiency of *nonlinear optical processes at the nanoscale*. High-index semiconductor nanoparticles supporting multipolar Mie-type resonances have been studied extensively due to their low losses, in comparison with metallic plasmonic structures, and due to the opportunity to achieve the interplay of the electric and magnetic multipolar modes. Nonlinear optical response of such resonant nanostructures is considered for both closely packed and individual nanoparticles. The nature of multipolar modes is controlled by the geometry and constituent material of resonators, while the light coupling efficiency mostly depends on the structure of the excitation beam.

Cylindrical doughnut-shaped vector beams have already been used for the harmonic generation to develop nonlinear microscopy techniques [2]. In addition, radially and azimuthally polarized vector beams have recently been employed for the selective third-harmonic generation in silicon nanodisks [3]. Here we report on both experimental and numerical studies of nonlinear optical response of individual subwavelength semiconductor resonators pumped with structured light and conduct, for the first time to our knowledge, the nonlinear spectroscopy of the enhanced second-harmonic generation (SHG) in isolated AlGaAs nanodisks excited by an azimuthal pump beam near the magnetic dipolar resonance, and by a radial pump beam with a coupling to the eigenmodes at double frequencies.

2. Materials and Methods

Nanodisks are fabricated from a wafer consisted of a GaAs substrate, a sacrificial layer of $Al_{0.51}In_{0.49}P$ and a 650-nm [100]-grown $Al_{0.2}Ga_{0.8}As$ layer by the subsequent application of electron beam lithography, dry etching, selective wet-etching methods and pillar transferring to a BK7 glass substrate. We employ two types of fabricated nanodisks: with the diameters of 870 nm and 935 nm, respectively. We generate a second-harmonic signal in individual nanodisks by a tunable femtosecond pulsed laser pump; pulse duration is around 300 fs, a repetition rate is 21 MHz. We tune the spectral range of a central wavelength within 1520-1670 nm. We generate two orthogonal states of cylindrical vector beams (azimuthal and radial polarizations) using an achromatic half-wave plate and a silicon-based highly-efficient q-plate dielectric metasurface reported previously [4]. The SHG signal from a single resonator is collected by an objective lens with a numerical aperture 0.9 and detected by a cooled CCD camera, and then it is normalized over a spectral function of the experimental setup including filter transmission, pump power, and detector quantum efficiency spectra. The origin of a nonlinear signal is checked by the direct spectral measurements and by analyzing the corresponding quadratic power dependence.

3. Results and Discussions

Magnetic multipoles are excited resonantly in a single AlGaAs disk by an azimuthal pump beam, whilst the considered nanoparticles reveal a non-resonant behavior under the radially polarized excitation, see Figs. 1(a, b, e, f). The experimental results on the SHG spectroscopy of AlGaAs resonators of both radius values with cylindrical vector beams are presented in Figs. 1(c, g). The enhancement of a SHG signal is observed for the case of a nanoparticle which is resonant at the fundamental wavelength, but also there is the enhancement of the nonlinear response provided by a non-resonant particle pumped by a radially polarized laser beam.

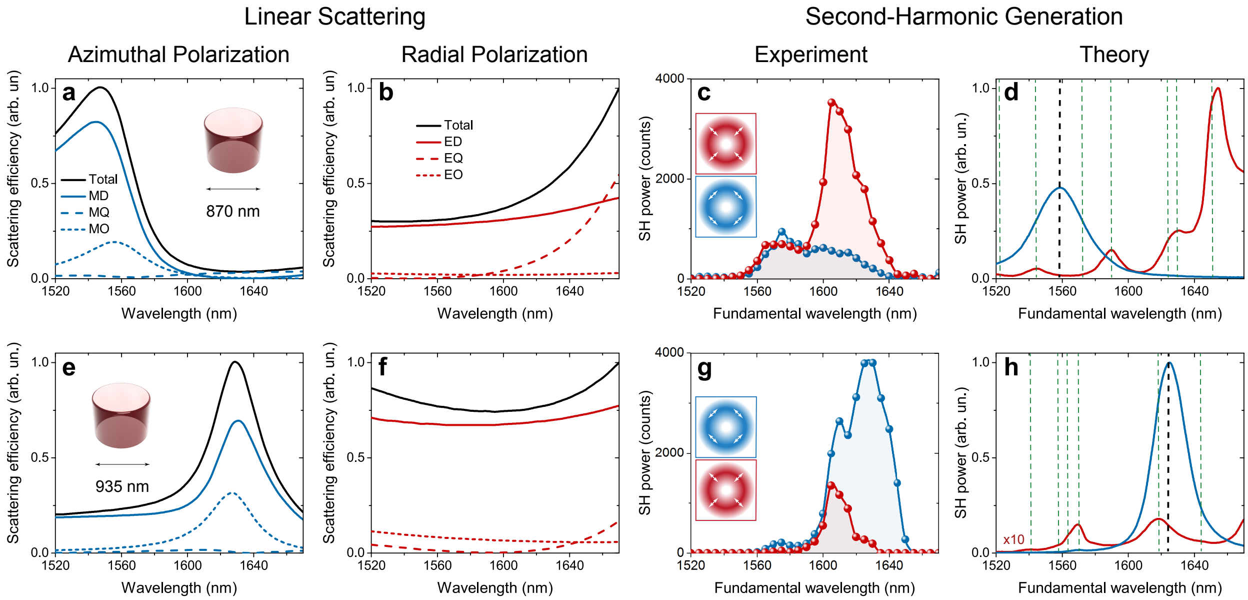

Fig. 1 Nonlinear spectroscopy with structured light. (a, b), (e, f) Multipolar decomposition of the scattering efficiency of a single AlGaAs nanodisk with a height of 650 nm and diameter of (a, b) 870 nm and (e, f) 935 nm excited by a vector beam with (a, e) azimuthal and (b, f) radial polarizations; solid black line represents the total sum of multipolar contributions; the total scattering efficiency is independently normalized on the maximum value for each case; electric dipole (ED) and magnetic dipole (MD); electric and magnetic quadrupoles, EQ and MQ; electric and magnetic octupoles, EO and MO. (c, g) Experimentally measured SHG spectrum of a single AlGaAs nanoresonator with a diameter of (c) 870 nm and (g) 935 nm, respectively, pumped by structured light; blue dots correspond to the azimuthally polarized pump beam, red dots – to the radial polarization, as shown in the insets; white arrows illustrate the polarization distributions in the central cross-section of the cylindrical vector beams. (d, h) Calculated SHG spectra for (d) 870-nm and (h) 935-nm disks pumped by an azimuthal (blue line) and radial (red line) vector beams; black and green dashed vertical lines mark the positions of the eigenmodes at the fundamental and double frequencies, respectively.

In order to provide the theoretical interpretation of the observed effects, we perform numerical simulations of the generated second-harmonic electromagnetic fields and the analysis of the nanodisk eigenmodes using the finite-element method in COMSOL Multiphysics. The spectral positions of particle eigenmodes do not depend on the type of the external excitation, and they are defined only by the parameters of the sample. The dominant contribution to the SHG enhancement of the nanoparticle pumped by an azimuthally polarized light is made by the mode at the fundamental frequency, while the spectral features of the nonlinear response generated by the radially polarized excitation are caused by the coupling of the generated second-harmonic fields to the particle eigenmodes at the double frequency and, moreover, the radially polarized pump can be even more efficient for SHG effects.

4. Conclusions

We have employed azimuthally and radially polarized cylindrical vector beams to generate the second harmonic from isolated AlGaAs nanoparticles, and we have conducted nonlinear spectroscopic measurements. Theoretical interpretation of the observed spectral features of a nonlinear signal is based on the calculations of the eigenmodes of the nanoparticle excited at the fundamental and double frequencies. Our results open new ways for the efficient light coupling to resonant modes of high-index nanoparticles for advanced photonic nanoscale metadevices.

5. References

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