Atomic resonance interaction in dielectric media

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A theory is presented for the retarded resonance interaction in dielectric media between a ground state atom and an excited atom. We demonstrate in the retarded limit large deviations compared to earlier incorrect evaluations of this important quantity.

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We investigate the influence of retardation on the resonance interaction of a ground state, and an excited, Drude model atom when the atoms are immersed in a dielectric medium. The dipole-dipole mechanism has been proposed as a way to create entangled states for quantum logic using both molecules [1] and quantum dots [2]. Resonance interaction and the related Förster energy transfer [3] go to the very heart of biophysics. Resonance interaction has very broad applications and it has also been exploited to create cold molecules [4]. We showed recently [5] how earlier theoretical results for the retarded resonance interaction in free space were incorrect in the retarded limit. In this Brief Report we present first the basic theory for the interaction between a ground state atom and an excited atom immersed in a dielectric medium. The extension to molecules is straightforward. We then derive simple analytical asymptotes for the long-range interaction and show that there are large deviations when the results are compared to earlier, incorrect results.

We consider two identical atoms where one is initially in its ground state and the other is in an excited state. This whole system can also be represented by a superposition of states: one symmetric and one antisymmetric with respect to interchange of the atoms. While the symmetric state is likely to decay into two ground state atoms, the antisymmetric state can be long lived. The system can thus be trapped in the antisymmetric state [6]. The energy migrates back and forth between the two atoms until either the two atoms move apart or a photon is emitted away from the system. First order dispersion interactions are caused by the coupling of the system [i.e., by the separation (r) dependence of the energy difference between the two states].

By writing down the equations of motion for the excited system, it is straightforward to derive the zero temperature Green function for two identical (and isotropic) atoms [5,7]. The transition moments of the two atoms are assumed to be parallel and to make an angle θ with the axis that joins the two atoms. The extension to considering two different atoms in a dielectric medium is straightforward and the resonance frequencies (ωs) of the system are given by the solution of the following equations:

\[ 1 - \alpha(1|\omega)\alpha(2|\omega)T(\rho|\omega)^2 = 0, \]

\[ \alpha(j|\omega) = \frac{\alpha(j|0)}{1 - \omega^2/\omega_j^2 - i\omega\gamma/\omega_j^2}, \]

where ωs is the polarizability of atom s, r is the distance between the atoms, and \( \rho \) is the atom-atom distance. The dielectric function, this expression can be simplified further to obtain

\[ T(\rho|\omega) = \left\{ \begin{array}{l}
\left[ 1 - 3\cos^2(\theta) \right] \frac{\omega^2}{\rho^2c^2} \frac{1}{\rho} \\
+ \sin^2(\theta) \frac{\omega^2}{c^2} e^{i\omega c \rho /c^2} \frac{\rho}{\epsilon}. \end{array} \right. \]

(3)

Here \( T(\rho|\omega) \) is the field susceptibility [7,8] in a material with dielectric function \( \varepsilon(\omega) \), and \( \alpha(j|\omega) \) is the polarizability of atom j. In the above expression we have used a two-level approximation for the polarizability with oscillator frequency \( \omega_j \) and dissipation frequency \( \gamma_j \) (via, e.g., radiation damping). The influence of the finite velocity of light (c) and the background material is contained in the field susceptibility. In the case of two identical atoms the above resonance condition can be separated in one antisymmetric and one symmetric part. Since the excited symmetric state has a much shorter lifetime than the antisymmetric state, the system can be trapped in an excited antisymmetric state. The resonance interaction energy of this antisymmetric state is

\[ U(\rho) = \hbar \left[ \omega_s(\rho) - \omega_s(\infty) \right]. \]

(4)

Since the relevant solution of Eq. (1) is the pole of the antisymmetric part of the underlying Green function [5] we can in a standard way [9] deform a contour of integration around this pole to obtain a simple and exact expression for the resonance interaction energy,

\[ U(\rho) = \frac{\hbar}{\pi c} \int_0^\infty d\xi \ln[1 + \alpha(1|i\xi)T(\rho|i\xi)]. \]

(5)

In the nonretarded limit we can take the limit of infinite velocity of light to obtain

\[ U = \frac{\hbar}{\pi \rho^3} [3\cos^2(\theta) - 1] \int_0^\infty d\xi \frac{\alpha(i\xi)}{\varepsilon(i\xi)}. \]

(6)

If the resonance frequency of the atom occurs at a much lower frequency than the first adsorption frequency of the dielectric function, this expression can be simplified further to obtain
In the large $\rho$ limit one can replace both $\omega(i\xi)$ and $\varepsilon(i\xi)$ with their respective static limits and evaluate the integral to obtain

$$U \approx \frac{\hbar \alpha(0) \omega}{2\varepsilon(0)\rho^3} [3\cos^2(\theta) - 1].$$  

This is the dominating term in the resonance interaction energy in the retarded limit (at zero temperature). It can be contrasted with the oscillating forms obtained in earlier treatments. These earlier incorrect treatments [5,7] ignored the coupling between the two interacting atoms via the electromagnetic field. We can easily rederive these results by replacing $T(\rho|\omega) \rightarrow T(\rho|\omega_r)$. With this substitution Eq. (5) leads to an oscillating form in the retarded limit.

We conclude that the generally accepted [10–12] form for the retarded resonance interaction is not correct. The resonance interaction does not oscillate but follows simple power laws. The fact that the interaction takes place in a dielectric medium has two effects in the two limiting cases considered in Eqs. (7) and (8). The first effect is a reduction of the speed of light ($c \rightarrow c/\sqrt{\varepsilon}$) in the retarded limit. The second effect is a screening of the interaction potential ($U \rightarrow U/\varepsilon$).

[8] L. Knöll et al., in Coherence and Statistics of Photons and Atoms (Ref. [2]).