Microcavity-induced modification of the dipole-dipole interaction

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We consider the feasibility of controlling the energy transfer between an unexcited and an excited atom using microcavities. The excitation-transfer probability is related to the strength of the dipole-dipole interaction and thus the excitation-transfer rates could be increased by enhancing the dipole-dipole interaction. We demonstrate this enhancement with an explicit three-dimensional calculation for a metallic cavity, which we characterize with a realistic dielectric function. We also demonstrate the shifts in cavity resonances from their ideal positions $m\lambda/2$ (m is an integer and λ is the wavelength). [S1050-2947(98)01101-9]

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I. INTRODUCTION

It is now understood that spontaneous emission from an atom is very sensitive to the environment in which the atom is radiating. The subject of enhancement and inhibition of spontaneous emission has been extensively discussed since the very early work of Purcell [1]. More specifically, the changes in lifetimes and radiative shifts in various environments have been calculated and measured [2–9]. There is in fact a modification of all the radiative characteristics including, for example, the van der Waal forces [6] between the atom and the metallic wall. Clearly, by choosing the environment appropriately one has a control over the spontaneous emission. We can now enquire if the interactions between atoms will be modified by the environment [10,11]. In particular, we could examine if the energy-transfer rate between an excited and an unexcited atom could be enhanced by using a suitable environment. This is what we study in this paper. The energy-transfer rate is related in general to the retarded dipole-dipole interaction and thus we study the possibilities of controlling the excitation-transfer rates. More specifically, we study this transfer rate when the atoms are contained in a microcavity. The control of excitation transfer could be very important in many different contexts [12] including the operations of microscopic laser and maser systems involving one or a few atoms. The dipole-dipole interaction can be computed in a number of different ways [3,4,10]. A general theory of the quantum electrodynamical effects in the presence of dielectric and conducting surfaces was developed by one of the authors [3,4]. This theory was *fully quantum in nature and showed that the retarded dipole-dipole interaction was related to the real part of the response function* $\chi_{\alpha\beta}(\vec{r}_1,\vec{r}_2,\omega)$ [see Eq. (2) below], where $\vec{r}_1$ and $\vec{r}_2$ are the locations of the two atoms. The response functions depend on boundaries, cavity geometry, etc. Starting from the general expressions of Ref. [3], we produce exact numerical results. The different features of the paper can be summarized as follows. We study the dependence of the dipole-dipole interaction on the size of the cavity and bring forth the role of cavity modes and its cutoff frequency. We retain the full three-dimensional nature of the cavity. Contributions from both longitudinal and transverse modes of the cavity are thus important and are fully incorporated. We also investigate how the dipole-dipole interaction changes as a function of the interdipolar distance in, say, $\lambda/2$ cavities. Our studies reveal the range for which dipole-dipole interaction effects can be enhanced significantly. Moreover, we show how the results of a realistic description of the boundary in terms of a complex dielectric function could be different from the ones obtained by treating metal as a perfect conductor. Inclusion of mirror losses is shown to lead to cavity-induced resonances with finite width that are shifted from $m\lambda/2$ (m is an integer).

The response function $\chi_{\alpha\beta}(\vec{r}_1,\vec{r}_2,\omega)$ is defined as the αth component of the electric field at $\vec{r}_1$ produced by an oscillating dipole of unit magnitude at $\vec{r}_2$ and oriented along the β direction. In terms of the response function, the electric field at $\vec{r}$ due to a dipole of strength $\vec{p}$ at $\vec{r}_2$ is given by

$$E_\alpha(\vec{r})=\sum_\beta \chi_{\alpha\beta}(\vec{r},\vec{r}_2,\omega)p_\beta . \tag{1}$$

The electric field can be calculated from the solution of Maxwell's equations with a source polarization term $\vec{p}\,\delta(\vec{r}-\vec{r}_2)e^{-i\omega t}+\text{c.c.}$ The Maxwell's equations are to be solved subject to the boundary conditions. A quantum electrodynamic treatment [4] shows that the cooperative decay γ_{12} and the dipole-dipole interaction Ω_{12} are related to χ via

$$\Omega_{12}-i\gamma_{12}\equiv-\frac{1}{\hbar}\sum_{\alpha,\beta} p_\alpha p_\beta \chi_{\alpha\beta}(\vec{r}_1,\vec{r}_2,\omega), \tag{2}$$

where the p_α's are the components of the dipole matrix element connecting the two states of the atom. In free space

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$$\chi^{(0)}_{\alpha\beta}(\vec{r}_1,\vec{r}_2,\omega)=\left(k^2\delta_{\alpha\beta}+\frac{\partial^2}{\partial x_\alpha \partial x_\beta}\right)\frac{e^{ik|\vec{r}_1-\vec{r}_2|}}{|\vec{r}_1-\vec{r}_2|}, \tag{3}$$

which on use in Eq. (2) leads to the well-known results

$$\gamma_{12}=\gamma\left\{\frac{\sin kR}{kR}-\frac{3\cos^2\theta-1}{2}\left[\frac{\sin kR}{kR}+3\left(\frac{\cos kR}{k^2R^2}-\frac{\sin kR}{k^3R^3}\right)\right]\right\}, \tag{4}$$

$$\Omega_{12}=\frac{3}{2}\gamma\left\{(1-3\cos^2\theta)\left(\frac{\sin kR}{k^2R^2}+\frac{\cos kR}{k^3R^3}\right)-(1-\cos^2\theta)\frac{\cos kR}{kR}\right\}, \tag{5}$$

where γ is the free-space decay rate of a single dipole, $\vec{R}=\vec{r}_1-\vec{r}_2$, and θ gives the angle between $\vec{p}$ and $\vec{R}$. Note further that in the limit $c\to\infty$, Eq. (5) reduces to the static form of the dipole-dipole interaction. The cooperative decay rate [13] $\gamma_{12}\to\gamma$ in the limit $\vec{R}\to 0$, whereas Ω_{12} diverges. The radiating atoms get coupled via terms such as Eq. (2). In fact, the density-matrix equation describing the reduced dynamics of the atomic system is [4,10]

$$\frac{\partial\rho}{\partial t}=-\sum_{i,j}\gamma_{ij}(S_i^+S_j^-\rho-2S_j^-\rho S_i^++\rho S_i^+S_j^-)-i\sum_{i,j}\Omega_{ij}[S_i^+S_j^-,\rho], \tag{6}$$

where the spin-$\frac{1}{2}$ operators $\vec{S}_i$ characterize the two-level atoms in the usual manner.

II. MODIFICATION OF THE DIPOLE-DIPOLE INTERACTION IN A REALISTIC CAVITY WITH LOSSES

There is an important modification of the above picture in the presence of a different environment, say, when the dipoles are placed near a dielectric interface or in a cavity. In the presence of a cavity the nature of the radiation field inside the cavity can undergo a significant change. The cavity leaves its imprint by means of its mode structure. For certain geometries there can be a cutoff beyond which no modes exist. Since there are no radiation modes beyond the cutoff the dipole-dipole interaction and spontaneous emission can significantly be suppressed. From a mathematical angle the major contribution of the cavity (environment) lies in the fact that response functions are now altered. In fact, now one has to solve the Maxwell equations with proper implementation of the boundary conditions, which can lead to an altogether different response function. However, since the general solution of the boundary-value problem can be decomposed in two parts, viz., the general solution of the homogeneous problem (assuming the absence of sources or dipoles) and a particular solution of the inhomogeneous problem (with sources), the response functions also can be accordingly decomposed. The particular solution determines the translationally invariant part of the response, i.e., the free-space response, whereas boundary conditions (the ''environment'') determines the relationship between the arbitrary constants of the homogeneous problem and thus leads to the contributions of the boundaries to the response function. Thus the total response consists of two parts and can be written as

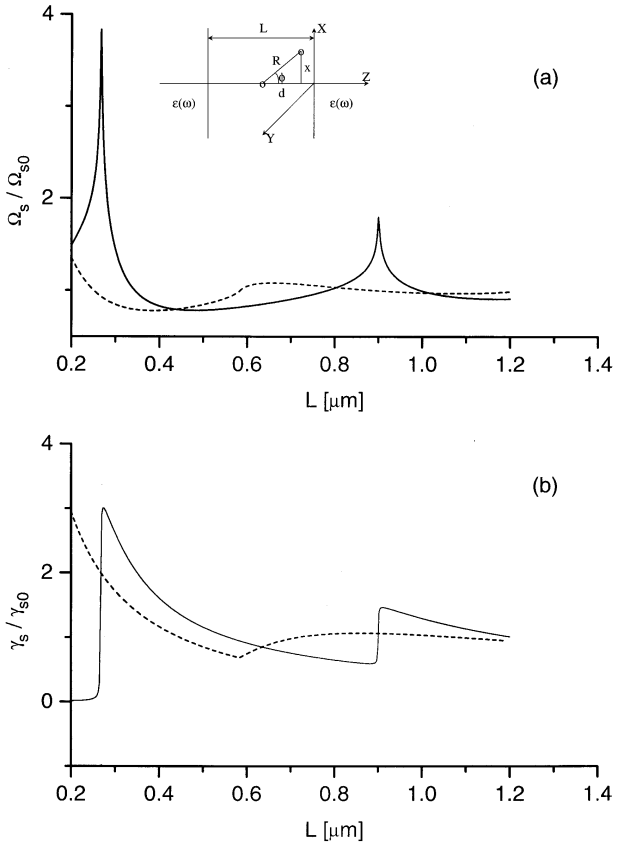

FIG. 1. (a) Dipole-dipole interaction normalized to the free-space value Ω_s/Ω_{s0} and (b) cooperative decay normalized to the free-space value γ_s/γ_{s0} as functions of the cavity length L for $x=d=0.07\ \mu$m. The solid (dashed) curves are for x- (z-) oriented dipoles. Other parameters are $\lambda=0.633\ \mu$m and $\varepsilon(\omega)=-16.3+0.53i$. The inset shows the schematic view of the cavity with two dipoles (marked by small circles) in it.

$$\chi_{\alpha\beta}(\vec{r},\vec{r}_0,\omega)=\chi^{(0)}_{\alpha\beta}(\vec{r},\vec{r}_0,\omega)+\chi^{(1)}_{\alpha\beta}(\vec{r},\vec{r}_0,\omega), \tag{7}$$

where $\chi^{(1)}(\vec{r},\vec{r}_0,\omega)$ is the contribution of the environment to the linear response. The literature on the calculation of response functions for various environments is vast and a rigorous and comprehensive treatment can be found in Ref. [3]. Depending on the nature and size of the environment, the effects on the evolution on a single dipole and dipole-dipole interaction can be multifaceted. For example, one can manipulate the decay rates and control the frequency shifts of a single dipole. We show that there is also control of the dipole-dipole interaction as well, simply by manipulating the environment.

In what follows, we restrict ourselves to a planar cavity of length L (see the inset of Fig. 1) bounded on both sides by a medium characterized by a dielectric function $\varepsilon(\omega)$, which in general is complex. One of the dipoles is located at the center of the cavity at $\vec{r}_0=(0,0,-L/2)$. The other dipole is located at $\vec{r}=(x,0,-L/2+d)$, $d<L/2$. The response functions were

calculated in Ref. [3] for the above-mentioned geometry [see Eqs. (5.1)–(5.24) in Ref. [3]]. For example, both dipoles have the same orientation (x or z oriented) for which the surface-dependent contributions to linear-response functions are given by

$$\chi_{xx}^{(1)} = -i\int_0^\infty ds\ \cos(w_0 d)\left[\frac{sw_0}{D_+}\{J_0(sx)-J_2(sx)\} + \frac{sk^2}{w_0 D_3}\{J_0(sx)+J_2(sx)\}\right], \tag{8}$$

$$\chi_{zz}^{(1)} = -2i\int_0^\infty ds\ \cos(w_0 d)\left[\frac{s^3}{w_0 D_-}J_0(sx)\right], \tag{9}$$

where J's are Bessel functions and the following notations are introduced:

$$D_\pm = 1 \pm \frac{w_0\varepsilon + w}{w_0\varepsilon - w}\, e^{-iw_0 L}, \tag{10}$$

$$D_3 = 1 + \frac{w_0 + w}{w_0 - w}\, e^{-iw_0 L}, \tag{11}$$

$$w_0^2 = k^2 - s^2, \quad w^2 = k^2\varepsilon - s^2. \tag{12}$$

Equation (9), for example, is obtained from Eq. (5.24) of Ref. [3] by moving from Cartesian variables u and v to polar coordinates s and θ [i.e., we set $s=\sqrt{u^2+v^2}$ and $\theta = \arctan(v/u)$] and performing integration with respect to the polar angle. Thus contributions from all the spatial harmonics are taken into account and the integrals (8) and (9) represent explicit results for the three-dimensional cavity. It may also be noted from Eq. (12) that, for example, for real ε, the z component of the wave vector w_0 changes its character from real to purely imaginary as s becomes larger than k. Since integration is performed over all possible s's Eqs. (8) and (9) retain the contributions from both propagating and evanescent waves. It may be noted that the evanescent part does not contribute to the cooperative decay γ_{12}. Another pertinent point that should be noted from Eqs. (8) and (9) is that enhancement of the dipole-dipole interaction is expected near the poles of the integrand (zeros of $D_\pm$ and D_3). These denominators clearly arise from multiple passes of the field in the cavity. *These poles correspond to the excitation of the cavity modes* and we show later that there is indeed enhancement of the dipole-dipole interaction when the cavity modes come into play. The integrals given by Eqs. (8) and (9) are not amenable to analytical methods and we resort to numerical integration.

III. NUMERICAL RESULTS

In our numerical study we assumed the bounding medium to be silver with $\varepsilon(\omega) = -16.3 + i0.53$ at wavelength $\lambda = 0.633\ \mu$m [14]. We first look at the real and imaginary parts of the responses as a function of the size (L) of the cavity for specific choices of interdipolar distance given by x and d. Recall that the real [imaginary] part of χ determines the dipole-dipole interaction Ω_s (cooperative decay γ_s), where s ($=x,z$) denotes the orientation of the dipoles with

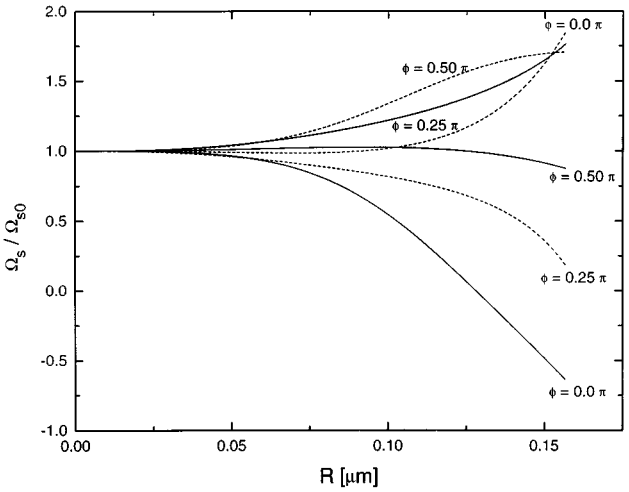

FIG. 2. Dipole-dipole interaction normalized to the free-space value Ω_s/Ω_{s0} as a function of R for a $\lambda/2$ cavity for z-oriented dipoles (dashed curves) and x-oriented dipoles (solid curves). Different curves are labeled by the corresponding value of φ. Other parameters are as in Fig. 1.

an identical direction of oscillation. We present the results for Ω_s [γ_s] normalized to its corresponding free-space value $\Omega_{s0} = -\mathrm{Re}(\chi_{ss}^{(0)})$ [$\gamma_{s0} = \mathrm{Im}(\chi_{ss}^{(0)})$] in Fig. 1(a) [Fig. 1(b)]. The choice of this normalization is obvious since our aim is to bring out the cavity-induced modifications. The solid [dashed] curve gives the results for x-oriented [z-oriented] dipoles. It is clear from Fig. 1 that for large L cavity effects are negligible and the dipole-dipole interaction and decay rate reduce to their free-space values. The behavior of Ω_x is quite distinct from that of Ω_z. Since the z-oriented dipoles do not sense the longitudinal modes (with field oriented along the normal to the cavity axis), there are no drastic changes near the frequencies corresponding to these cavity modes. On the other hand, Ω_x shows a drastic enhancement close to the cavity resonances at $\lambda/2$, $3\lambda/2$, etc. Note also the shift of the resonances from $\lambda/2$, $3\lambda/2$, etc., because of the finite (negative) real and nonzero imaginary part of ε. This is in contrast to the ideal cavity results that were obtained by means of modal expansion of the cavity field (see, for example, the paper by Kobayashi, Zheng, and Sekiguchi in Ref. [10]). Below $\lambda/2$ cavity propagating longitudinal modes do not exist and hence there is a suppression of the decay rate γ_s [see the solid curve in Fig. 1(b)]. However, even for $L<\lambda/2$ the dipole-dipole interaction cannot be totally suppressed because of the presence of the evanescent waves emitted by the dipoles. Similar results to those in Fig. 1 were obtained (not shown) for a smaller distance between the dipoles. For smaller distances between the dipoles the cavity effects are less pronounced. In terms of the image theory [9], the proximity of one of the dipoles to the mirror implies that with larger interdipolar distance the second dipole is closer to its image and this leads to the more pronounced effect. These features are shown in Fig. 2 for the zz (dashed curves) and xx (solid curves) orientation, where we have plotted Ω_s/Ω_{s0} as a function of R for a $\lambda/2$ cavity for various angles φ, namely, $\varphi = 0, 0.25\pi, 0.5\pi$. It may also be noted that for shorter R, cavity effects are negligible, which is quite expected since the images of the dipoles are further away from

their originals. Figure 2 thus demonstrates the possibility of significant change in the dipole-dipole interaction. It should be borne in mind that we have assumed atoms to be well localized. When required, the effects of small dispersion in the atomic position can obviously be included in the theory.

IV. CONCLUSIONS

Using a very general response function approach, we studied the dipole-dipole interaction in a planar cavity with realistic metals as the bounding medium and reported the cavity-induced changes in the dipole-dipole interaction. We showed that for a particular orientation of the dipoles the dipole-dipole interaction can be significantly enhanced depending on the parameters of the cavity. We demonstrated the shift of the resonances as a function of the cavity length due to the realistic values of the dielectric constant of the bounding metal. We also showed the existence of a large enhancement in the region where the corresponding resonant interaction γ_s term vanishes.

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[1] E. M. Purcell, Phys. Rev. **69**, 681 (1946).

[2] K. H. Drexhage, in *Progress in Optics*, edited by E. Wolf (North-Holland, New York, 1974), Vol. 12, p. 163.

[3] G. S. Agarwal, Phys. Rev. A **11**, 230 (1975).

[4] G. S. Agarwal, Phys. Rev. A **12**, 1475 (1975); see also G. S. Agarwal, *Quantum Optics* (Springer, Berlin, 1974), p. 34, Eqs. (6.53), (6.57), and (6.58).

[5] D. Kleppner, Phys. Rev. Lett. **47**, 233 (1981); A. G. Vaidyanathan, W. P. Spencer, and D. Kleppner, *ibid.* **47**, 1592 (1981); G. S. Agarwal and S. V. ONeil, Phys. Rev. B **28**, 487 (1983); S. Haroche, in *Fundamental Systems in Quantum Optics*, Les Houches Summer School of Theoretical Physics, Session 53, edited by J. Dalibard, J. M. Raimond, and J. Zinn-Justin (North-Holland, Amsterdam, 1992), p. 769; D. Meschede, Phys. Rep. **211**, 203 (1992); J. E. Childs, K. An, R. R. Dasari, and M. S. Feld, in *Cavity Quantum Electrodynamics*, edited by P. R. Berman, Suppl. 2 to Advances in Atomic, Molecular, and Optical Physics (Academic, Boston, 1994), p. 325.

[6] E. A. Hinds, in *Cavity Quantum Electrodynamics* (Ref. [5]), p. 1.

[7] The surface-dependent contributions to atomic frequency shifts have been experimentally measured; see T. Wegener, J. Dircherl, and H. Walther, in *Atomic Physics 13*, edited by H. Walther, T. W. Hansch, and B. Neizert (American Institute of Physics, New York, 1993), p. 301.

[8] The effects of absorption in the dielectric are discussed by G. S. Agarwal and H. D. Vollmer, Phys. Status Solidi B **85**, 301 (1978); S. M. Barnett, B. Huttner, and R. Loudon, Phys. Rev. Lett. **68**, 3698 (1992).

[9] The image method was used to derive the decay of a dipole in a cavity with perfectly conducting mirrors; see, for example, P. W. Milonni and P. L. Knight, Opt. Commun. **9**, 119 (1973); J. P. Dowling, M. O. Scully, and F. De Martini, *ibid.* **82**, 415 (1991).

[10] The dipole-dipole interaction has recently attracted considerable attention; see, for example, G. V. Varada and G. S. Agarwal, Phys. Rev. A **45**, 6721 (1992); Takayoshi Kobayashi, Qianbing Zheng, and Takashi Sekiguchi, *ibid.* **52**, 2835 (1995); M. S. Tomas, *ibid.* **51**, 2545 (1995); E. V. Goldstein and P. Meystre, *ibid.* **53**, 3573 (1996); E. V. Goldstein, P. Pax, and P. Meystre, *ibid.* **53**, 2604 (1996); The dipole-dipole interaction in photonic band gaps has been discussed by S. John and T. Quang, *ibid.* **52**, 4083 (1995); S. Bay, P. Lambropoulos, and K. Molmer, Opt. Commun. **132**, 257 (1996); Gershon Kurizki, Phys. Rev. A **42**, 2915 (1990).

[11] J. M. Wylie and J. Sipe, Phys. Rev. A **30**, 1185 (1984); **32**, 2030 (1985).

[12] D. Y. Smith and D. L. Dexter, in *Progress in Optics*, edited by E. Wolf (North-Holland, Amsterdam, 1972), Vol. X, p. 165.

[13] For an experimental observation of interdipole decay see R. G. Devoe and R. G. Brewer, Phys. Rev. Lett. **76**, 2049 (1996).

[14] T. Inagaki, M. Motosuga, E. T. Arakawa, and J. P. Goudonnet, Phys. Rev. B **32**, 2548 (1985).