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Coherence in Spontaneous Emission in the Presence of a Dielectric

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A quantum-electrodynamic theory of spontaneous emission in the presence of a dielectric has been worked out, and formulas are presented for the change in the lifetime and the Lamb shift of a two-level atom, as a result of the presence of a dielectric. Curves are given for the behavior of the damping as a function of the distance of the atom from the interface. The frequency shift for the case of a conductor is also evaluated.

Over the last few years, a large number of papers have appeared which deal with the problem of coherence in spontaneous emission from a system of many atoms¹ (Dicke-type superradiance). Here I discuss another type of coherence which arises while analyzing the radiation from an excited atom in the close vicinity of a dielectric.² I have worked out a complete quantumelectrodynamic theory of spontaneous emission in the presence of a dielectric and in this communication summarize some of the results. In particular, I discuss how the lifetime and the Lamb shift of the atom change as a result of the presence of the dielectric. Such changes depend on the dielectric constant and the distance of the atom from the interface and should be experimentally observable.

The interaction Hamiltonian between an atom and the radiation field is given by

$$H_1 = -\int \vec{p}(\vec{r}) \cdot \vec{E}(\vec{r}) d^3 \gamma, \qquad (1)$$

where the dipole approximation has been made and we have ignored the contact term $2\pi \int |\vec{p}_{\perp}|^2 d^3 r$. This term is not considered as we are interested in distance-dependent effects only. The electric field is a second-quantized field operator. The field should now be quantized, not in free space, but in the space containing the dielectric and the free space. I assume, for simplicity, that the dielectric occupies the domain $-\infty \le z \le 0$ and that the atom is located at $\vec{r} = \vec{a} (a_x = a_y = 0, a_z = a)$. The quantization of the field has been carried out in a beautiful paper by Carniglia and Mandel.³ One would, however, *not* need to quantize the field explicitly. In this approach I have avoided the problem of explicit quantization by resorting to an appropriate response function and the fluctuation-dissipation theorem.⁴

It has been shown that for a two-level atom, the damping coefficient γ and the shift in the energy separation of the two levels Ω (which would be referred to as Lamb shift) are given, to the lowest order in the coupling coefficient, by

$$\gamma(\vec{\mathbf{a}},\,\omega) + i\Omega(\vec{\mathbf{a}},\,\omega) = \int_0^\infty d\tau \,Q(\vec{\mathbf{a}},\,\tau)e^{\,i\omega\tau},\tag{2}$$

where ω is the energy separation between two atomic levels and Q is the mean value of the *anticommutator* of the "free field" operators at two space-time points, i.e.,

$$Q(\mathbf{\vec{a}}, \tau) \equiv \langle \{ \mathbf{\vec{d}} \cdot \mathbf{\vec{E}}_{0}(\mathbf{\vec{a}}, \tau), \mathbf{\vec{d}} \cdot \mathbf{\vec{E}}_{0}(\mathbf{\vec{a}}, 0) \} \rangle.$$
(3)

In (3), \tilde{d} is the dipole-moment matrix element, and \vec{E}_0 is the free-field operator, i.e., it evolves according to the unperturbed Hamiltonian. I emphasize that in deriving (2), I did *not* make use of any particular *mode expansion* of the field operators. For the case of an atom emitting in entirely *free* space, it was also noticed by Bullough and Caudrey⁵ that the Lamb shift can be computed from the anticommutator of the field operators at two time points. Note that we are dealing with an inhomogeneous problem and that is why Q is a function of \tilde{a} . From (2) it follows that the Lamb shift Ω is related to the damping γ by the dispersion relation

$$\Omega(\mathbf{\vec{a}},\,\omega) = \pi^{-1} \mathbf{P} \int_0^{\omega} d\omega_0 \,\gamma(\mathbf{\vec{a}},\,\omega_0) \\ \times [(\omega_0 + \omega)^{-1} - (\omega_0 - \omega)^{-1}], \quad (4)$$

with P standing for the principal part. Therefore,

a + 90

once the damping is known, the Lamb shift can be computed from Eq. (4). Let us now introduce $D(\vec{a}, \tau)$, defined by

$$D(\vec{a}, \tau) \equiv [\vec{d} \cdot \vec{E}_0(\vec{a}, \tau), \vec{d} \cdot \vec{E}_0(\vec{a}, 0)].$$
(5)

We are dealing with spontaneous emission and hence the fields at t = 0 are in the *vacuum* state. The vacuum field can be viewed as a thermal field at zero temperature; then, by the fluctuation-dissipation theorem, Q and D are related by⁴

$$\hat{Q}(\vec{\mathbf{a}},\,\omega) = \hat{D}(\vec{\mathbf{a}},\,\omega), \quad \omega > 0,$$

= $-\hat{D}(\vec{\mathbf{a}},\,\omega), \quad \omega < 0.$ (6)

where the Fourier transforms are defined by

$$\psi(t) = \int_{-\infty}^{\infty} (d\omega/2\pi) \hat{\psi}(\omega) e^{-i\omega t}.$$
(7)

From Eqs. (2)–(7) it fillows that $\gamma(\mathbf{a}, \omega)$ is given by

$$\gamma(\vec{\mathbf{a}},\,\omega) = \mathrm{Im}\sum_{ij}d_{i}d_{j}\chi_{ij}(\vec{\mathbf{a}},\,\vec{\mathbf{a}},\,\omega),\tag{8}$$

where $\chi_{ij}(\vec{a}, \vec{a}, \omega)$ is the response of the electric field at a point \vec{a} to an applied (external) polarization at the point \vec{a} , i.e., to $\vec{P}_{ex} = \vec{P}_{ex} \delta(\vec{r} - \vec{a})$ $\times e^{-i\omega t}$. I have thus formulated the problem of calculating $\gamma(\vec{a}, \omega)$ and therefore $\Omega(\vec{a}, \omega)$ in terms of the response. The response function appearing in (8) is easily obtained from the solution of Maxwell's equations with proper account of boundary conditions. All the equations (2)-(8) are applicable to any geometrical arrangement.⁶

I now specialize to the case mentioned earlier, i.e., when the dielectric occupies the volume $-\infty \le z \le 0$ and the atom is located at \vec{a} . I further assume that the dipole moment of the atom is randomly distributed in the *x*-*y* plane, and hence in the subsequent derivation carry out an averaging over the orientation of the dipole moment. We solve the equations (\vec{B} standing for the magnetic field)

$$\nabla \times \nabla \times \mathbf{E} - k_0^{2} (\mathbf{E} + 4\pi \mathbf{P}_{ext}) = 0,$$

$$\nabla \times \mathbf{E} = -c^{-1} \partial \mathbf{B} / \partial t,$$

$$\nabla \cdot (\mathbf{E} + 4\pi \mathbf{P}_{ext}) = 0,$$

$$\mathbf{P}_{ext} = \mathbf{P}_{ex} \delta(\mathbf{r} - \mathbf{a}), \quad z \ge 0;$$

$$\nabla \times \nabla \times \mathbf{E} - k_0^{2} \epsilon_0 \mathbf{E} = 0, \quad \nabla \cdot \mathbf{E} = 0,$$

$$\nabla \times \mathbf{E} = -c^{-1} \partial \mathbf{B} / \partial t, \quad k_0 = \omega / c, \quad z < 0,$$
(10)

subject to the boundary conditions that tangential components of $\vec{\mathbf{E}}$ and $\vec{\mathbf{B}}$ and the normal components of $\epsilon \vec{\mathbf{E}}$ and $\vec{\mathbf{B}}$ are continuous across the interface z = 0, to obtain the response function $\chi_{ij}(\vec{\mathbf{a}}, \vec{\mathbf{a}}, \omega) [\equiv \vec{\mathbf{E}}_i(\vec{\mathbf{a}}, \omega)/\vec{\mathbf{P}}_j]_{\text{ex}}$. A straightforward but long calculation shows that

$$\gamma(\vec{\mathbf{a}},\,\omega) = \gamma^{(0)} + \gamma^{(1)}(\vec{\mathbf{a}},\,\omega),\tag{11}$$

where $\gamma^{(0)}$ is equal to the usual expression $\frac{2}{3}|d|^2\omega^3/c^3$ and $\gamma^{(1)}$ is given by

$$\gamma^{(1)}(\vec{\mathbf{a}},\omega) = \frac{3}{4}\gamma^{(0)} \operatorname{Re} \int_0^\infty (\kappa d\kappa/\mu) e^{i\mu x} [\kappa^2 - 2\mu \kappa^2/(\epsilon_0\mu + \mu_0) - 2(\mu - \mu_0)^2/(\epsilon_0 - 1)], \qquad (12)$$

with

$$\mu^{2} = 1 - \kappa^{2}, \quad \mu_{0}^{2} = \epsilon_{0} - \kappa^{2}, \quad x = 2\omega a/c.$$
(13)

The ϵ_0 which appears in (12) is the dielectric function of the dielectric at the frequency ω . Equation (12) is our final expression for the damping coefficient. It does not appear possible to do the integrals analytically. I have evaluated (12) numerically as a function of a and the results are given in Fig. 1, where I have plotted the behavior of $\gamma^{(1)}$ for a number of cases. We see that $\gamma^{(1)}$ is an oscillating function of a. For small distances $\gamma^{(1)}$ increases and hence the lifetime goes down. It is clear from these curves that there are appreciable changes in the lifetime of the atom at distances much smaller than a wavelength. It is certainly feasible to deposit atoms at such distances by using the techniques developed by Drexhage.⁷

For the case of a perfect conductor, if we take the limit of infinite conductivity,⁸ then (12) leads to

$$\gamma_{\rm cond}{}^{(1)}(\bar{a},\,\omega) = -\frac{3}{2}\,\gamma^{(0)} \bigg[\frac{\sin x}{x} - \frac{\sin x}{x^3} + \frac{\cos x}{x^2} \bigg],\tag{14}$$

and then the Lamb shift is found to be

$$\Omega_{\text{cond}}(\vec{\mathbf{a}},\,\omega) = \frac{3\gamma^{(0)}}{\pi} \left\{ \frac{1}{x^2} \left[1 - \cos x \,\operatorname{Ci} x - \sin x \,\operatorname{Si} x \right] + \left(\frac{1}{x^3} - \frac{1}{x} \right) \left[\sin x \,\operatorname{Ci} x - \cos x \,\operatorname{Si} x \right] \right\},\tag{15}$$

where Si and Ci are the usual sine and cosine integrals.⁹ Note that for large distances $x \gg 1$, $\Omega_{\text{cond}} \sim x^{-4}$. I have plotted the behavior of $\gamma_{\text{cond}}^{(1)}$ in Fig. 1 as function of x. It should be noted that, for



FIG. 1. Curves I, II, and III show, respectively, the behavior of $\frac{4}{3}\gamma^{(1)}/\gamma^{(0)}$ as a function of $2\omega a/c$ for the cases of dielectrics with refractive indices 1.5, 2, and 5 (0.1 on the scale corresponds to 0.5 for the curve III). Curves IV and V represent, respectively, $\frac{2}{3}\gamma_{\text{cond}}^{(1)}/\gamma^{(0)}$ and $\pi\Omega_{\text{cond}}^{(1)}/3\gamma^{(0)}$.

short distances, $\gamma_{\text{cond}}^{(1)}$ is negative, and hence the lifetime of an atom in the close vicinity of a conductor increases. Such an effect has been observed by Drexhage.⁷ We again see from the curve that there could be appreciable changes in the Lamb shift, which should be experimentally observable since the effective damping in the case of a conductor is rather small. It should be further noted that $\gamma_{\text{cond}}^{(1)}$ can also be obtained from the arguments of an image dipole; however, the same argument will lead to an *incorrect* value,¹⁰

$$\Omega_{\rm cond}^{(1)}(\vec{a},\,\omega) = \frac{3\gamma^{(0)}}{2} \left[-\frac{\sin x}{x^2} + \left(\frac{1}{x} - \frac{1}{x^3}\right)\cos x \right],\tag{16}$$

of the Lamb shift. For small distances (16) leads to very large frequency shifts; e.g., $\pi\Omega_{\text{cond}}^{(1)}/3\gamma^0$ is equal to 0.57 and -11.28, respectively, according to (15) and (16) for x = 0.5.

For the case when the frequency of the atom coincides with one of the resonant (transverse) frequencies of the dielectric, the damping is again given by (14). The details of this method and the dynamical aspects (including radiation-reaction fields) of emission in the presence of a dielectric will be discussed elsewhere. Finally, I would like to mention that this method can also be generalized to the case of a radiation field which is *not* necessarily in the *vacuum* state.

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¹See the articles by G. S. Agarwal, R. Bonifacio, R. K. Bullough, I. R. Senitzky, and K. H. Drexhage, in *Coher-ence and Quantum Optics*, edited by L. Mandel and E. Wolf (Plenum, New York, 1973), pp. 157, 465, 121, 301, and 187, respectively; N. Rehler and J. H. Eberly, Phys. Rev. A <u>3</u>, 1735 (1971).

²Some aspects of radiation from an atom in the presence of a dielectric have been studied by C. M. Carniglia, L. Mandel, and K. H. Drexhage, J. Opt. Soc. Amer. <u>62</u>, 479 (1972), and by Drexhage in Ref. (1).

³C. K. Carniglia and L. Mandel, Phys. Rev. D <u>3</u>, 280 (1971).

(8a)

⁴For an account of the fluctuation-dissipation theorem, see, e.g., P. C. Martin, in *Many Body Physics*, edited by C. DeWitt and R. Belian (Gordon and Breach, New York, 1968), p. 37.

⁵R. Bullough and P. J. Caudrey, J. Phys. A: Proc. Phys. Soc., London 4, L41 (1971).

 6 It should be noted that if the two-level atom were treated as a harmonic oscillator, then (2), (4), and (8) should be replaced by

$$\gamma^{(-)}(\vec{a},\omega) + i\Omega^{(-)}(\vec{a},\omega) = \int_0^\infty d\tau \, D(\vec{a},\tau) e^{i\omega t},\tag{2a}$$

$$\Omega^{(-)}(\mathbf{\dot{a}},\omega) = -\pi^{-1} \int_0^\infty d\omega_0 \gamma^{(-)}(\mathbf{\dot{a}},\omega_0) [(\omega_0 + \omega)^{-1} + (\omega_0 - \omega)^{-1}],$$
(4a)

 $\gamma^{(-)}(\vec{a},\omega) = \operatorname{Im}\sum_{i,j} d_i d_j \chi_{ij}(\vec{a},\vec{a},\omega).$

Thus the damping is identical to that for a two-level atom, but the frequency shift is very different. Note further that (2a), (4a), and (8a) can also be obtained from classical considerations. I would also like to point out that $\gamma(\mathbf{a}, \omega) = \gamma(\mathbf{a}, \omega)$ only for the case when initially the field is in a vacuum state.

⁷K. H. Drexhage, Sci. Amer. <u>222</u>, No. 3, 108 (1970), and to be published.

⁸In quantum electrodynamics a perfect conductor seems to have been used in this sense [cf. H. B. Casimir and D. Polder, Phys. Rev. <u>73</u>, 360 (1948)]. Strictly speaking, one should include the dispersion of the dielectric function even for a conductor. At any rate, formulas (14) and (15) are quite instructive and that is why I have presented them.

⁹Handbook of Mathematical Functions, edited by M. Abramowitz and I. Stegun (Dover, New York, 1964), p. 231. ¹⁰H. Morawitz, Phys. Rev. 187, 1792 (1969).

Propagation of Sound in Two-Dimensional He³⁺

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"Surface sound" has been observed for the first time, in He³ adsorbed on the free surface of liquid He⁴. Measurements of the surface-sound velocity and surface tension are used to obtain new values for the effective mass $M = (1.3 \pm 0.1)m_3$ and binding energy $\epsilon_0 / k_B = 2.28 \pm 0.03$ K of the adsorbed He³.

This Letter reports measurements of the velocity of surface sound on liquid He⁴, cooled below 100 mK, and covered with about 0.2 atomic layer of adsorbed He³. Earlier surface-tension studies¹⁻³ have shown that in such low concentrations the adsorbed He³ can be treated as a twodimensional Fermi gas of weakly interacting quasiparticles. Surface sound⁴ is a compressional, adiabatic, longitudinal wave in this almost ideal, two-dimensional system. The measurements allow one to check the theory governing the surfacesound velocity u_s , and to derive some of the propperties of the adsorbed He³ at low number density. These are the quasiparticle effective mass M_{\star} the binding energy to the surface relative to the bulk, ϵ_0 , and the effective interaction between the quasiparticles. The binding energy ϵ_0 has been calculated by a number of theorists⁵⁷ but no theoretical estimates for the effective mass of interaction have been published yet. The sign and magnitude of the interaction is of particular interest because of the possibility of observing twodimensional superfluidity in adsorbed He³.

The existence of surface sound was predicted by Andreev and Kompaneets.⁴ In their theory, because of the existence of surface excitations ---quantized capillary waves ("ripplons")⁸ and adsorbed He³ quasiparticles—the free surface of superfluid helium can transport mass, entropy, momentum, etc., and it obeys a set of hydrodynamic equations which are analogous to the two-fluid bulk equations. At low temperatures in pure He⁴, or in very dilute solutions of He³ in He⁴, the influence of the normal fluid in the bulk becomes negligible compared to that at the surface, and there are then two forms of small oscillations of the surface: capillary waves and what Andreev and Kompaneets called "surface second sound" (which we abbreviate to "surface sound"). The surface sound has velocity u_s given by

$$\nu_n u_s^2 = -\left(\frac{\partial\sigma}{\partial\ln N_s}\right)_s \tag{1}$$

where σ is the surface tension, $N_s = -(\partial \sigma / \partial \mu_s)_T$