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Decay of an emitting dipole between two parallel mirrors*

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A classical treatment is presented for the modification of the emission rate of an atom between two parallel mirrors. For the case of two perfectly reflecting mirrors, the results are identical with previous treatments of this problem using a quantum mechanical approach. The purely classical treatment has the advantage of being able to treat nonperfectly reflecting mirrors.

Recently, Milonni and Knight,[1] Stehle,[2] and Philpott[3] have discussed the modification of the emission rate of an excited atom owing to the presence of perfectly reflecting mirrors using a quantum mechanical approach. In a series of articles[4-6] we have discussed the same effect for a single mirror using the purely classical theory developed by Sommerfeld[7] for radio wave propagation near the earth's surface. The latter method has the advantage of being able to treat real mirrors, i.e., those with absorptive parts to their dielectric constants. In the present paper, we extend this method to treat an emitting atom between two parallel and infinite mirrors. Milonni and Knight[1] in their quantum mechanical treatment of this problem, find (within the Wigner–Weisskopf approximation) that the rate of decay of an atom, with transition dipole perpendicular to perfectly reflecting mirrors, is given by

$$b^{\perp} = \frac{3\pi b^{\circ}}{k_1 L}\left(\frac{1}{2} + \sum_{j=1}^{[k_1 L/\pi]} \frac{1-j^2\pi^2}{k_1^2 L^2}\cos^2\frac{j\pi d}{L}\right) . \qquad (1)$$

Here b° is the decay rate in the absence of the mirrors, L is the distance between the mirrors, d is the distance betteen the emitter and one of the mirrors, $[k_1 L/\pi]$ is the greatest integer part of $k_1 L/\pi$, and $k_1 (= 2\pi n_1/\lambda)$ is the propagation constant at emission wavelength λ in the dielectric medium (refractive index n_1) between the two mirrors. This notation differs slightly from that of Milonni and Knight, mainly in our use of d as a variable instead of the position of the atom relative to the midpoint between the two mirrors (their z_0).

If this system (Fig. 1) is treated clasically by assuming the atom to be an oscillating dipole, we find that, for real mirrors with dielectric constants ϵ_2 and ϵ_3, a dielectric spacer of dielectric constant ϵ_1, and a luminescent state with quantum yield q,

$$b^{\perp} = b^{\circ}(1-q) + \tfrac{3}{2} q b^{\circ}\,\mathrm{Im}\int_0^{\infty} d\tau\,\tau^3 F(\tau)/l \ , \qquad (2)$$

where

$$F(\tau) = \frac{[1+R_{1,2}\exp(-2lk_1 d)][1+R_{1,3}\exp(-2lk_1 s)]}{[1-R_{1,2}R_{1,3}\exp(-2lk_1 L)]} \ , \qquad (3)$$

with

$$l \equiv -i(1-\tau^2)^{1/2} \ , \qquad (4)$$

$$m \equiv -i(\epsilon_2/\epsilon_1 - \tau^2)^{1/2} \ , \qquad (5)$$

and

$$n \equiv -i(\epsilon_3/\epsilon_1 - \tau^2)^{1/2} \ . \qquad (6)$$

The branches of the square roots in the definitions of l, m, and n have been chosen to keep their real parts positive for all positive values of τ. The quantities $R_{1,2}$ and $R_{1,3}$ are the reflectivities of the two interfaces and are given by[6]

$$R_{1,2} = (\epsilon_2 l - \epsilon_1 m)/(\epsilon_2 l + \epsilon_1 m) \ , \qquad (7)$$

and

$$R_{1,3} = (\epsilon_3 l - \epsilon_1 n)/(\epsilon_3 l + \epsilon_1 n) \ . \qquad (8)$$

(See Ref. 5, 6, or 8 for more general remarks on the details of the classical approach.)

In the limit that both mirrors are perfectly reflecting, ϵ_2 and $\epsilon_3 \to \pm\infty (R_{1,2} = R_{1,3} = 1)$, we find

$$b^{\perp} = b^{\circ}(1-q) + 3q \lim_{\delta\to 0} \times \mathrm{Im}\int_{0+i\delta}^{1+i\delta} du(1-u^2)\,\frac{\cos(k_1 ud)\cos(k_1 us)}{\sin(k_1 uL)} \ . \qquad (9)$$

Evaluating the integral by finding the residues at the poles $u = j\pi/k_1 L$, $j = 0, 1, 2, \ldots, [k_1 L/\pi]$, (the Fabry–Perot modes), we find

$$b^{\perp} = b^{\circ}(1-q) + \frac{3\pi q b^{\circ}}{k_1 L}\left(\frac{1}{2} + \sum_{j=1}^{[k_1 L/\pi]} \frac{1-j^2\pi^2}{k_1^2 L^2}\cos^2\frac{j\pi d}{L}\right), \qquad (10)$$

in agreement with Milonni and Knight[1] (who take $q = 1$) but without any recourse to quantum mechanics.

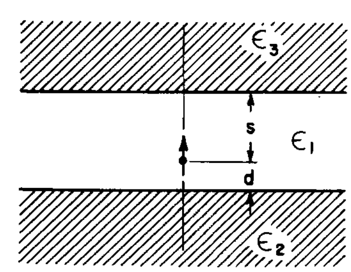

FIG. 1. Geometry of the two mirror problem. The quantities d and s are the distances between the dipole and the two mirrors. The mirrors are parallel, infinite in extent, and separated by a distance $L = d + s$.

The general result for the case of the dipole oriented parallel to the mirrors is a bit more complicated and, for brevity, will not be given here (but is available on request). However, for perfectly reflecting mirrors we find

$$b^{\parallel} = b^{\circ}(1-q) + \frac{3\pi q b^{\circ}}{2k_1 L} \sum_{j=1}^{[k_1 L/\pi]} \frac{1+j^2\pi^2}{k_1^2 L^2} \sin^2 \frac{j\pi d}{L}, \qquad (11)$$

in agreement with Milonni and Knight but, again, with no recourse to quantum mechanics.

The utility of Eqs. (10) and (11) would be restricted to highly reflecting mirrors and large values of L and d, while the general expression Eq. (2) is not so restricted. For example, for the hypothetical case of a Drude free electron gas metal $[\epsilon = 1 - (\omega_p/\omega)^2$, where ω_p is the plasma frequency], numerical calculations using Eq. (2) ($k_1 L = 0.4$, $k_1 d = 0.2$, and $\epsilon_1 = 2.25$) show that for $\epsilon_2 = \epsilon_3 = -10000$, $b^{\perp}$ is about 10% larger than the perfect mirror result from Eq. (10). For a more realistic value $\epsilon_2 = \epsilon_3 = -25$, $b^{\perp}$ is about three times larger than the perfect mirror result. From earlier comparisons of the classical theory with experimental results for single mirror systems,[4,5,8] we may reasonably conclude that Eq. (2) and the corresponding expression for the parallel case will offer a good description of the lifetime behavior for mirror separations which are only a small fraction of the emission wavelength.

The effect of mirrors on the lifetime of an emitting atom or molecule is of current interest for a variety of reasons,[1-6,8-11] including the obvious relevance of the two mirror problem to microptical lasers. We have shown in the present paper that the classical theory is capable of reproducing the results of the quantum mechanical theory and can be used for nonperfectly reflecting mirrors. However, Barton[11] has pointed out differences between the classical and a *full* quantum mechanical treatment; for example, the quantum mechanical treatment assigns shifts and widths to each quantum level rather than to each spectral line as in the classical approach.

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[1]P. W. Milonni and P. L. Knight, Opt. Commun. **9**, 119 (1973).
[2]P. Stehle, Phys. Rev. A **2**, 102 (1970).
[3]M. Philpott, Chem. Phys. Lett. **19**, 435 (1973).
[4]R. Chance, A. Prock, and R. Silbey, J. Chem. Phys. **60**, 2184 (1974).
[5]R. Chance, A. Prock, and R. Sibley, J. Chem. Phys. **60**, 2744 (1974).
[6]R. Chance, A. Prock, and R. Silbey, "Comments on the Classical Theory of Energy Transfer," J. Chem. Phys. (to be published).
[7]A. Sommerfeld, Ann. Phys. (Leipz.) **28**, 665 (1909); **81**, 1135 (1926).
[8]K. H. Tews, Ann. Phys. (Leipz.) **29**, 97 (1973); Thesis, Marburg/Lahn (1972); J. Lumin., in press.
[9]H. Morawitz, Phys. Rev. **187**, 1792 (1969).
[10]H. Morawitz and M. Philpott, Phys. Rev. B **12**, 4863 (1974).
[11]G. Barton, Proc. R. Soc. A **320**, 251 (1970); G. Barton (unpublished results).