# Electromagnetic decay into a narrow resonance in an optical cavity

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The spontaneous electromagnetic decay of a two-level atom coupled to a narrow cavity resonance is investigated rigorously in terms of the (Hermitian) modes of the "universe" rather than the (dissipative) quasimodes of the cavity. Special attention is paid to the strong-coupling regime (atomic linewidth $\Gamma \gtrsim$ cavity resonance width $\gamma$), in which there are significant corrections to the golden rule. In particular, spontaneous decay is most rapid for *intermediate* values of the quality factor $Q$ of the cavity resonance. The photon line shape, the effect of several cavity resonances, and the competition of several transitions are also investigated. The additivity of partial rates does not hold, and in some circumstances, the addition of an extra decay channel may *reduce* the total decay rate. These results are relevant to optical processes observed in dielectric microspheres, and also to usual laser cavities.

## I. INTRODUCTION

Many optical phenomena involve the interaction of atoms (or molecules) with the electromagnetic field in a resonant cavity $C$, such as linear laser cavities. A parallel experimental program has recently been initiated on dielectric microspheres with radii in the range 5–50 $\mu$m, e.g., liquid droplets acting as optical cavities.[1–3] Roughly speaking, rays confined within the microsphere by total internal reflection at a near-glancing angle cause resonance when the circumference and the wavelength are in suitable ratios. Resonance-enhanced fluorescence,[1] stimulated Raman scattering,[2] and lasing[3] have all been reported.

In addition to potential applications, this experimental program is of fundamental interest for two reasons. First of all, thanks to the spherical symmetry, the electromagnetic modes are readily computable, not just for the resonant quasimodes but for all the modes of the universe, i.e., the microsphere cavity $C$ together with the outside environment $B$.[4] In contrast, the evaluation of even one quasimode for a linear cavity is quite cumbersome[5] and the consideration of all the modes of the universe, a more proper approach, has been restricted to a one-dimensional approximation.[6] For this reason, these microspheres provide excellent opportunities for confronting theory with experiment. Secondly, electromagnetic cavity modes of extremely high $Q(>10^{20})$ exist in the ideal situation of zero absorptive loss.[7] According to the golden rule,[8] the width (i.e., decay rate) $\Gamma$ of an excited atomic level is enhanced by a factor $K$ over the corresponding value $\Gamma^{(0)}$ in extendeded vacuum: $\Gamma \sim K\Gamma^{(0)}$, where $K \propto Q$. Thus one may anticipate situations where $\Gamma \gtrsim \gamma$, $\gamma=\omega/Q$ being the width of the cavity mode. Such nontrivial situations have never been thoroughly studied in the literature, since, "normally," the density of states in vacuum is a smooth function of frequency, i.e., effectively $\gamma \to \infty$. Such cases of special interest, $\Gamma \gtrsim \gamma$, characterized as strong coupling (since $\Gamma$ is proportional to the square of the dipole matrix element), or equivalently, as decay into a narrow cavity resonance, will be the main subject of this paper.

Dielectric microspheres of radius $a$ are generally described by the dimensionless size parameter $x=2\pi a/\lambda$, where $\lambda$ is the electromagnetic wavelength in vacuum. We shall take $x \sim 30$, scaling to other values wherever necessary. Other typical parameters might be $\Gamma \sim 300\Gamma^{(0)} \sim 3\times 10^{11}$ s$^{-1}$(1 ns lifetime in vacuum); the origin of the enhancement factor $\Gamma/\Gamma^{(0)}$ is well understood[4,8] and is also explained in Secs. II and III; the typical numerical value 300 is obtained from an evaluation of the density of states.[4] For an optical transition, say, $\lambda=0.5$ $\mu$m, a value $Q>10^4$ (Ref. 4) would make $\gamma=\omega/Q \lesssim \Gamma$, so that strong coupling is required in practice.

We shall stress that the golden rule, which underpins the vast majority of discussions of optical phenomena, is inadequate in the strong-coupling regime. For example, the golden rule suggests that for a transition exactly tuned to the cavity resonance, the spontaneous decay rate should be proportional to $Q$. This was first pointed out by Purcell some 40 years ago:[9] The density of states in the volume $V$ of the cavity is increased from the vacuum value $V\omega^2/\pi^2c^3$ to $D$ modes ($D$ being the degeneracy) in a frequency interval $\gamma=\omega/Q$, so that one expects an enhancement factor of order $(DQ/\omega)(V\omega^2/\pi^2c^3) \propto Q$. We shall show that this proportionality breaks down when $Q$ is large; in fact, for $Q$ larger than some $Q^*$, the spontaneous decay rate decreases with $Q$, and eventually goes as $Q^{-1}$. The photon spectrum also shows interesting features. This paper studies these and related issues for the spontaneous decay of a single atom coupled to a narrow resonance in an optical cavity. Although dielectric microspheres provide both the motivation and a clear-cut strong-coupling example, the formalism and results are, in principle, general. However, recent observation of cavity-enhanced decay of highly excited Rydberg atoms[10] belongs to the less interesting weak-coupling case.

The present problem is much simplified by separating the formalism into two largely independent parts. First of all, one must solve for the electromagnetic modes, leading to the local density of states $\rho(\omega,\mathbf{r})$ or a quantity $\mathcal{E}_{ij}(\omega,\mathbf{r})$ describing the vacuum fluctuation of the electric field,[4] as sketched in Sec. II. In almost all cases of interest, $\mathcal{E}_{ij}$ can be represented as a Lorentzian, which is then the input to the quantum-mechanical calculation of spontaneous processes in Sec. III, in which one no longer needs to know the details of the electromagnetic modes. Competition between different atomic lines (as encountered in fluorescence) and the influence of several nearby cavity modes (encountered for large dielectric microspheres with a large density of resonances[7]) are dealt with in Sec. IV. Comparison with works in the literature is given in Sec. V.

This paper is limited to the spontaneous decay of a single atom in an optical cavity. Stimulation by radiation with a narrow spectrum and the cooperative and coherent interaction of many atoms in the strong-coupling limit require a separate analysis. For comparison, the much simpler weak-coupling limit $\Gamma \ll \gamma$, previously treated within the realm of the golden rule,[8] will emerge as a limiting case.

## II. ELECTROMAGNETIC EIGENFUNCTIONS

Consider an atom with a lower level $a$ and an upper level $b$ at position $\mathbf{r}_0$ in a cavity. For $E1$ transitions, the relevant perturbation in the multipolar Hamiltonian is[11]

$$V=-\mathbf{P}\cdot\mathbf{E}(\mathbf{r}_0)\ , \tag{1}$$

where $\mathbf{P}$ is the electric dipole operator and $\mathbf{E}$ is the electric field. A factor $f=3n^2/(2n^2+1)$ should be inserted if the cavity consists of a dielectric of refractive index $n$.[8] The field $\mathbf{E}$ can be expanded in terms of a set of eigenfunctions $\mathbf{e}$ labeled by an index $s$, with frequency $\omega_s$,

$$\mathbf{E}(\mathbf{r})=-(i/\sqrt{2})\sum_s (a_s-a_s^\dagger)\mathbf{e}(s,\mathbf{r})\ , \tag{2}$$

where the annihilation and creation operators satisfy $[a_s,a_{s'}^\dagger]=\delta_{ss'}$ and the eigenfunctions are normalized to one quantum per mode:

$$[1/(4\pi)]\int_R d^3r\,\mathbf{e}(s,\mathbf{r})\cdot\mathbf{e}(s',\mathbf{r})=\hbar\omega_s\delta_{ss'}\ , \tag{3}$$

where the integral is over the universe $R$, say, a sphere of radius $\Lambda$. We shall concentrate on the quantity

$$\mathcal{E}_{ij}(\omega,\mathbf{r})=[1/(4\pi\hbar\omega)]\sum_s \delta(\omega-\omega_s)e_i(s,\mathbf{r})e_j(s,\mathbf{r})\ . \tag{4}$$

In the case with spherical symmetry, for example, $\mathbf{e}\propto\Lambda^{-1/2}$, while the frequency spacing is $O(\Lambda^{-1})$, so that $\sum_s \propto \Lambda d\omega_s$ and $\mathcal{E}_{ij}$ is independent of the quantization volume. Because of the normalization condition (4), $\mathcal{E}_{ij}$ is proportional to the frequency spectrum of $E_iE_j$ at $\mathbf{r}$ when there is half a quantum per mode, i.e., the vacuum fluctuations; in particular,

$$\langle \text{vac} \mid E_i(\mathbf{r},t)E_j(\mathbf{r},t') \mid \text{vac}\rangle =2\pi\hbar\int_0^\infty d\omega\,\mathcal{E}_{ij}(\omega,\mathbf{r})\omega e^{-i\omega(t-t')}\ . \tag{5}$$

The normalization of $\mathcal{E}_{ij}$ has been chosen so that in vacuum,

$$\mathcal{E}\equiv\sum_i \mathcal{E}_{ii}\to\rho_{\text{vac}}(\omega)=\omega^2/\pi^2c^3\ , \tag{6}$$

where $\rho_{\text{vac}}$ is the density of states per unit volume in extended vacuum. This choice is convenient because, as we shall see, $\mathcal{E}/\rho_{\text{vac}}$ will turn out to be the enhancement factor for spontaneous decay in the weak-coupling limit.

Since spontaneous emission is essentially "stimulation" by vacuum fluctuations, $\mathcal{E}_{ij}(\omega,\mathbf{r})$ is all we need. This function, in particular its trace $\mathcal{E}$, has been evaluated for dielectric microspheres.[4] It shows sharp resonances with $Q\sim10^4$ or more, and with magnitudes $\mathcal{E}/\rho_{\text{vac}}\sim300$ or more at the rim of the microsphere, where the internal fields are concentrated. The function $\mathcal{E}$ around each such resonance constitutes a complete description of a Fox-Li quasimode[5] for this system. These resonances, due to simple poles close to the real axis in the frequency plane, are adequately represented by Lorentzians.

## III. SPONTANEOUS EMISSION

### A. Formalism

We shall only consider the states $|b\rangle$ and $|as\rangle$, where the latter denotes the lower atomic state $|a\rangle$ with one photon in mode $s$, with amplitudes $C(t)$ and $D_s(t)$ in the interaction representation. Then in the rotating wave approximation, one obtains the usual Wigner-Weisskopf equations[12]

$$i\hbar\,dC/dt=\sum_s V_s^*D_s e^{i(\Omega-\omega_s)t}\ , \tag{7a}$$

$$i\hbar\,dD_s/dt=V_sCe^{-i(\Omega-\omega_s)t}\ , \tag{7b}$$

where $\hbar\Omega=E_b-E_a$ and

$$V_s=\langle as \mid V \mid b\rangle=(-i/\sqrt{2})\langle a \mid P_i \mid b\rangle e_i(s,\mathbf{r}_0)\ . \tag{8}$$

Eliminating $D_s$ then gives

$$dC(t)/dt=-(1/\hbar^2)\sum_a |V_s|^2\int_0^t dt'C(t') \times e^{i(\Omega-\omega_s)(t-t')}\ . \tag{9}$$

Use (8) for $|V_s|^2$, insert a factor $d\omega\,\delta(\omega-\omega_s)$ and assume that the atomic matrix element is isotropic,

$$\langle a \mid P_i \mid b\rangle\langle b \mid P_j \mid a\rangle\equiv M_{ij}=M\delta_{ij}\ , \tag{10}$$

then

$$\frac{dC(t)}{dt}=-\frac{2\pi}{\hbar}M\int_0^\infty d\omega\,\mathcal{E}(\omega)\omega\int_0^t dt'C(t')e^{i(\Omega-\omega)(t-t')}, \tag{11}$$

where $\mathcal{E}$ is understood to be evaluated at $\mathbf{r}_0$. We now assume that $\mathcal{E}$ is a narrow Lorentzian centered at $\omega_0$,

$$\mathcal{E}(\omega)\omega=\left[\frac{\omega_0^3}{\pi^2c^3}\right]K\frac{(\gamma/2)^2}{\xi^2+(\gamma/2)^2}\ , \tag{12}$$

where $\xi=\omega-\omega_0$. The first bracket is the value of $\mathcal{E}(\omega_0)\omega_0$ in vacuum, so $K$ is the cavity enhancement of $\mathcal{E}$ at the resonance peak, and is, as we shall see, the enhancement factor of the decay rate as well, at least in the weak-coupling limit. We expect the total strength of the resonance [i.e., the integral of (12) over $\xi$] to be independent of $Q$, so that $K \propto \gamma^{-1} \propto Q$. Then $C(t)$ satisfies

$$dC(t)/dt = \int_0^t dt' S(t-t')C(t') , \quad (13)$$

where the kernel $S$ is

$$S(t-t') = -\frac{K}{2\pi\tau_0}\int d\xi \frac{(\gamma/2)^2}{\xi^2+(\gamma/2)^2} e^{i(\Delta-\xi)(t-t')}$$
$$= -\frac{\gamma K}{4\tau_0}\exp\left[-\frac{\gamma}{2}+i\Delta\right](t-t') . \quad (14)$$

In (14) $\Delta$ is the detuning parameter and $\tau_0$ is the spontaneous lifetime in vacuum,

$$\Delta=\Omega-\omega_0 , \quad (15)$$

$$\frac{1}{\tau_0}=\frac{4M\Omega^3}{\hbar c^3} , \quad (16)$$

and we have assumed $\gamma$, $\Delta \ll \omega_0$, $\Omega$, so that the integral (14) can be evaluated by integrating over the whole real line.

The solution to (13), with initial condition $C(0)=1$, is

$$C(t)=(p_2-p_1)^{-1}(p_2e^{p_1t}-p_1e^{p_2t}) , \quad (17)$$

where $p_1,p_2$ are, in general, complex with negative real parts, and are the solutions to the secular equation

$$p\left[p+\frac{\gamma}{2}-i\Delta\right]+\frac{\gamma K}{4\tau_0}=0 . \quad (18)$$

Note that $\dot{C}(0)=0$, as is evident from (13).

### B. Weak-coupling limit

We expect (at least in the weak-coupling case) that the decay rate in an unbounded vacuum, $1/\tau_0$, is enhanced to $K/\tau_0$ due to cavity effects. The division into weak- and strong-coupling regimes will depend on the comparison of this rate with the rate of photon leakage out of the cavity, $\gamma$. Thus the weak-coupling case is defined by $K/\tau_0 \ll \gamma$, in which case we have

$$p_1 \approx -\frac{\gamma}{2}+i\Delta, \quad p_2 \approx -\frac{K}{2\tau_0}\frac{(\gamma/2)}{(\gamma/2)-i\Delta} . \quad (19)$$

Since $|p_1| \gg |p_2|$, for all but the shortest times (17) is dominated by the second term, and

$$|C(t)|^2 \approx e^{-\Gamma t} , \quad (20)$$

where

$$\Gamma=-2\,\mathrm{Re}p_2=\frac{K}{\tau_0}\frac{(\gamma/2)^2}{\Delta^2+(\gamma/2)^2} . \quad (21)$$

If the transition is tuned to the cavity resonance ($\Delta=0$), then $\Gamma=K\Gamma^{(0)}=K/\tau_0$. More generally, in view of (12) and (6), we may write

$$\Gamma/\Gamma^{(0)}=\mathcal{E}(\Omega)/\rho_{\mathrm{vac}}(\Omega) , \quad (22)$$

which is a precise statement of Purcell's classic argument[9] and agrees with an evaluation based on the golden rule.[8]

Nevertheless, (20) is incorrect at short times $t \ll \gamma^{-1}$, for which one has

$$C(t)\approx 1-(p_1p_2/2)t^2=1-(\gamma K/8\tau_0)t^2 . \quad (23)$$

The quadratic dependence on $t$, of course, follows from $\dot{C}(0)=0$.

### C. Strong-coupling limit

On the other hand, for $K/\tau_0 \gg \gamma$, $\Delta^2/\gamma$,

$$p_{1,2}\approx -\frac{\gamma}{4}\pm\frac{i}{2}\left[\frac{\gamma K}{\tau_0}\right]^{1/2} . \quad (24)$$

The imaginary part gives rise to oscillations in $|C(t)|^2$, and the envelope decays at a rate

$$\Gamma=-2\,\mathrm{Re}p=\gamma/2 , \quad (25)$$

essentially independent of the detuning. The oscillatory behavior may be interpreted as the emission and reabsorption of one photon. The net decay rate is then determined by the rate of leakage of the photon, i.e., $\gamma$. Moreover, for $\Delta=0$, say we have (a) $\Gamma \propto K \propto Q$ in the weak-coupling limit [see (21)], while (b) $\Gamma \propto \gamma \propto Q^{-1}$ in the strong-coupling limit [see (25)], so the decay will be most rapid for intermediate $Q$. This point will be elaborated below.

### D. Time dependence and effective decay rate

To display numerical results, it is convenient to define the dimensionless quantities

$$K_0=K\gamma\tau_0, \quad R=K/\gamma\tau_0 \quad (26)$$

to describe the cavity resonance. The reason is that $K_0$ will be fixed for a given species of atoms as $Q$ is changed. For dielectric microspheres with $x\sim 30$, typical values would be $K\sim 300$, $\gamma \sim 300$ ns$^{-1}$, $\tau_0 \sim 1$ ns, so $K_0 \sim 10^5$. However, for different dielectric microspheres, $K_0$ scales roughly as $x^{-2}$, for the following reason. The quantity $\mathcal{E}$ is approximately equal to the density of states $\rho$, which satisfies[4]

$$V\int \rho\, d\omega \sim \cdots = D$$

when integrated over the resonance, where $\cdots$ is the total number of states, $V=(4\pi/3)a^3$ is the volume of the sphere, and $D=2l+1\sim 2x$ is the degeneracy, $l$ being the angular momentum. In the above estimate we have assumed that $\rho$ is everywhere the same order of magnitude as its spatial average over the whole microsphere, which is correct to a factor of about 3.[4] Thus

$$K_0 \propto K\gamma \propto \int \rho\, d\omega \sim \frac{D}{V} \propto \frac{x}{x^3}=x^{-2} .$$

The parameter $R$, on the other hand, varies as $R \propto Q^2$, and by using (21) with $\Delta=0$ we see that $R \ll 1$ ($R \gg 1$) will characterize the weak-coupling (strong-coupling) limit. $|C(t)|^2$ will only depend on $R$, the detuning in the dimensionless combination $\bar{\Delta}=K_0^{-1/2}\tau_0\Delta$, and time in the dimensionless combination $\bar{t}=K_0^{1/2}t/\tau_0$.

Figure 1 shows $|C(t)|^2$ versus $t$ for both the tuned ($\Delta=0$) and the detuned ($\Delta\neq 0$) cases. Exponential relaxation in the weak-coupling case ($R \ll 1$) is analogous to overdamping; underdamped oscillations occur for strong coupling ($R \gg 1$). However, the decay is most rapid for intermediate coupling ($R \sim 1$), analogous to critical damping. To be more explicit, consider the effective decay rate $\Gamma_e$,

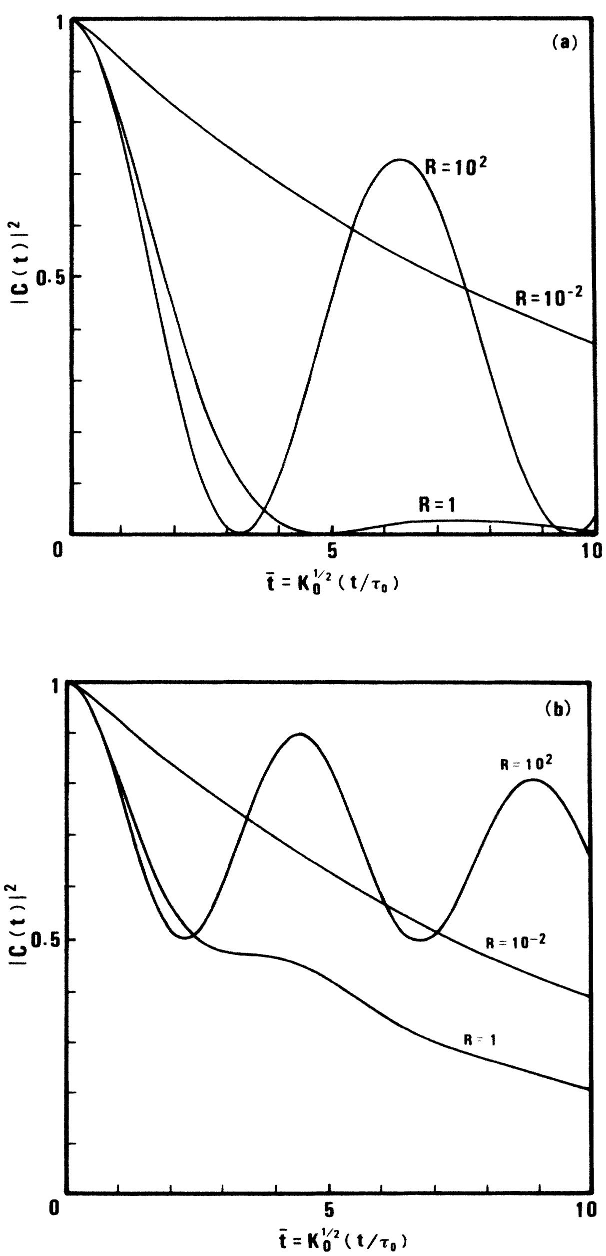


FIG. 1. Decay of $|C(t)|^2$. (a) Tuned case $\Delta=0$. (b) Detuned case $K_0^{-1/2}\tau_0\Delta=1$.

$$\Gamma_e^{-1}=\int_0^\infty dt\, |C(t)|^2 . \tag{27}$$

Again it is convenient to deal with the dimensionless combination $\bar{\Gamma}_e=K_0^{-1/2}\tau_0\Gamma_e$, and some arithmetic yields

$$\begin{aligned}\bar{\Gamma}_e^{-1}&=\frac{1}{(R^*)^{1/2}}\left[\left[\frac{R}{R^*}\right]^{1/2}+\left[\frac{R^*}{R}\right]^{1/2}\right]\\ &=\frac{1}{(R^*)^{1/2}}\left[\frac{Q}{Q^*}+\frac{Q^*}{Q}\right],\end{aligned} \tag{28}$$

where

$$R^*\equiv 1/(1+4\bar{\Delta}^2) , \tag{29}$$

and in (28) we have used the fact that $R\propto Q^2$. Thus, for fixed $\bar{\Delta}$, $\bar{\Gamma}_e$ has a maximum value of $(R^*)^{1/2}/2$ at $R=R^*$, decreases for both large and small $R$, and is symmetric under $R/R^*\to R^*/R$. Assuming typical parameters $K_0\sim 10^5$, $\omega_0\sim 2\pi c/0.5$ $\mu$m, and $\tau_0\sim 1$ ns gives $Q^*\sim 10^4/(1+4\bar{\Delta}^2)^{1/2}$. Thus the most rapid decay occurs for an *intermediate* $Q$ value; $Q^*\sim 10^4$ for exact tuning, less if the transition is detuned. This conclusion explains why very narrow cavity resonances (theoretical $Q>10^{20}$) found in numerical calculations[7] are not seen spectroscopically.

For relatively simple molecules, the fluorescence and Raman lines are so widely spaced that large detuning must be the rule rather than the exception. In this case we find from the weak-coupling result (21) that

$$\Gamma/\Gamma^{(0)}\sim\frac{1}{4\sqrt{R}}\frac{K_0^{3/2}}{(\Delta\tau_0)^2} , \tag{30}$$

which turns out to be correct up to a factor of 2 even for $R\sim 1$. Taking $K_0\sim 10^5$, $R\sim 1$, $\tau_0\sim 1$ ns, and $\Delta\sim 10^{12}$ s$^{-1}$ gives $\Gamma/\Gamma^{(0)}\sim 10$, showing significant cavity enhancement even when the atomic line and the cavity resonance have little overlap ($\Delta\gg\Gamma+\gamma$; in this case $\Gamma\sim 10^{10}$ s$^{-1}$, typically $\gamma\sim 3\times 10^{11}$ s$^{-1}$).

The fact that the most rapid decay occurs for intermediate values of $Q$ can be understood heuristically as follows. The decay involves two processes: (a) the emission of a photon at a rate $K\Gamma^{(0)}$, with $K\propto Q$, as discussed by Purcell[9] and (b) the leakage of the resultant photon out of the cavity, at a rate $\gamma\propto Q^{-1}$. The slower of the two processes is the rate-determining step, so that the observed decay rate goes as $Q$ for small values of $Q$ and as $Q^{-1}$ for large values of $Q$.

### E. Photon spectrum

The line shape will not be a single Lorentzian since the decay is not in general a pure exponential. The amplitude to find a photon in mode $s$ after the decay has occurred is

$$D_s(\infty)=(-i/\hbar)V_s\int_0^\infty dt\, C(t)e^{-i(\Omega-\omega_s)t} \tag{31}$$

and the intensity spectrum, normalized to $\int d\omega\, I(\omega)=1$, is

$$I(\omega)=\sum_s |D_s(\infty)|^2\delta(\omega_s-\omega)$$
$$=(2\pi M/\hbar)\mathcal{E}(\omega)\omega\,|\tilde{C}[i(\Omega-\omega)]|^2\,, \qquad (32)$$

where $\tilde{C}$ is the Laplace transform of $C$. Using (12) for $\mathcal{E}$ and (17) for $C(t)$ then gives

$$I=\frac{K}{2\pi\tau_0}\left[\frac{\gamma}{2}\right]^2\left[\left[\xi(\Delta-\xi)+\frac{\gamma K}{4\tau_0}\right]^2+\left[\frac{\gamma}{2}\right]^2(\Delta-\xi)^2\right]^{-1}, \qquad (33)$$

where $\xi=\omega-\omega_0$. Normally one expects that an oscillation $C(t)$ will give rise to a spectrum $|\tilde{C}[i(\Omega-\omega)]|^2$; the shift by $\Omega$ is due to the transformation between the interaction and Schrödinger representations. In this case we see from (32) that the actual spectrum is multiplied by the factor $\mathcal{E}(\omega)\omega$.

In the tuned case ($\Delta=0$), the denominator in (33) is proportional to

$$I^{-1}\propto\tfrac{1}{16}+\tfrac{1}{4}\left[-2+\frac{1}{R}\right]\bar{\xi}^2+\bar{\xi}^4\,, \qquad (34)$$

where $\bar{\xi}=K_0^{-1/2}\tau_0\xi$ represents the frequency shift from $\omega_0$ in dimensionless units. Thus, for $R<\frac{1}{2}$, $I$ consists of a single peak centered at $\bar{\xi}=0$, while for $R>\frac{1}{2}$, $I$ consists of two peaks. In fact, for very strong coupling $R\gg1$, the peaks occur at $\bar{\xi}=\pm\frac{1}{2}$, with widths $\delta\bar{\xi}\sim1/(2\sqrt{R})\rightarrow0$. These frequency peaks correspond to the oscillations shown in Fig. 1, described by the imaginary part of $p$ in (24). All these features are shown in Fig. 2(a). For relatively weak coupling, the width of the spectrum increases with $R$ or $Q$, while in the strong-coupling case the width of each peak decreases with $R$ or $Q$, which is just another way of seeing the behavior in (28). Note that for strong coupling ($R\gg1$) the frequency shift ($\bar{\xi}\sim\frac{1}{2}$ in dimensionless units) is large when compared to the width of the cavity mode itself ($\bar{\gamma}=K_0^{-1/2}\tau_0\gamma\sim1/\sqrt{R}$) and the width of the spectral line itself [$\delta\bar{\xi}\sim1/(2\sqrt{R})$]. Thus some care is necessary in interpreting the observed spectrum.

Figure 2(b) shows the spectrum for a detuned case. For weak coupling the spectrum is centered at the nominal transition frequency $\omega=\Omega$. As the coupling increases, the spectrum again shows a double-peak structure, with the peak farther from the cavity resonance being stronger.

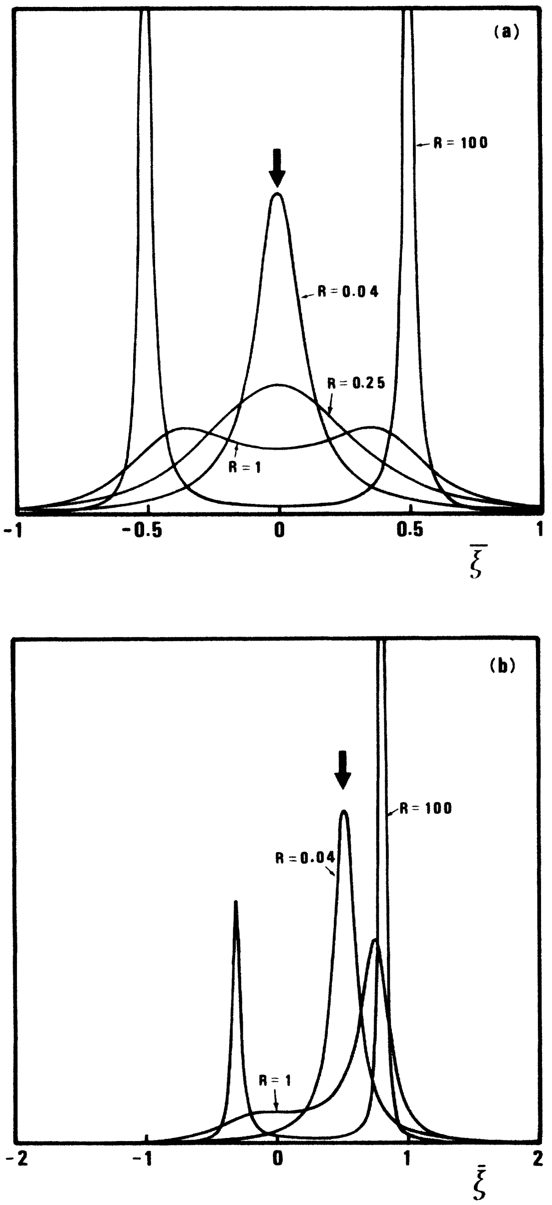


FIG. 2. Photon spectrum vs $\bar{\xi}=K_0^{-1/2}\tau_0(\omega-\omega_0)$. Arrow indicates nominal position of atomic line. Different curves are labeled by values of $R$. (a) Tuned case $\Delta=0$. (b) Detuned case $\bar{\Delta}=K_0^{-1/2}\tau_0\Delta=0.5$.

## IV. SEVERAL ATOMIC LINES OR CAVITY RESONANCES

### A. Effect of several cavity resonances

For weak coupling the effect of the cavity is local in frequency, so that $\Gamma$ depends only on the value of $\mathcal{E}$ at $\Omega$. However, in the strong-coupling case we may have to consider the effect of several resonances, especially for large dielectric microspheres with a high density of resonances.[7] (a) For large $K_0$, say, $K_0>100$, only a single resonance matters, the results of Sec. III apply, and $K_0$ can be scaled out in a simple way. (b) For intermediate $K_0$, say, $K_0\sim10$, several neighboring resonances have to be taken into account, and the situation is relatively complex. (c) For small $K_0$, say, $K_0<1$, many cavity resonances come in; then as far as the gross features of the spectrum are concerned, $\mathcal{E}$ can be replaced by its average value $\mathcal{E}_{av}$ over many resonances. For dielectric microspheres, these ranges correspond to the size parameter being (a) "small," say, $x<10^3$, (b) "intermediate," say, $x\sim3\times10^3$, (c) "large," say, $x>10^4$. These estimates of $x$ are obtained by using the typical value $K_0\sim10^5$ for $x\sim30$ and scaling by $K_0\propto x^{-2}$.

The condition for case (a) is obviously that the frequency shift $\delta\omega$ is small compared to the typical spacing $\omega_{sp}$

between cavity resonances. In the strong-coupling case, we have already shown that $\delta\omega = K_0^{1/2}/2\tau_0$. On the other hand, consider the total number of states in an interval $\omega_{sp}$,

$$\int_{\omega}^{\omega+\omega_{sp}} \rho\, d\omega \sim \left[ \frac{\omega^2}{\pi^2 c^3} \right] \omega_{sp} , \tag{35}$$

because in an average sense (as made precise by an asymptotic sum rule[4]) the density of states must be the same order of magnitude as in vacuum. Now assume that a certain fraction $f_3$ of the integral is due to resonances; by definition there is only one resonance in the interval $\omega_{sp}$, and since $\rho \sim \mathcal{E}$ (Ref. 4), by (12) we see that the left-hand side of (35) is $f_3^{-1}(\omega^2/\pi^2 c^3)(\pi/2)K\gamma$, so that

$$\omega_{sp} \sim \frac{\pi}{2f_3}\frac{K_0}{\tau_0} \sim 8\frac{K_0}{\tau_0} , \tag{36}$$

since $f_3 \sim 0.2$.[4,7] Thus $\delta\omega/\omega_{sp} \sim 1/(16\sqrt{K_0})$, so that for $K_0 > 100$, say [case (a)], $\delta\omega/\omega_{sp} < 0.01$ and neighboring resonances are too far to be relevant.

On the other hand, to replace $\mathcal{E}$ by its average $\mathcal{E}_{av}$ would require many cavity resonances within a frequency interval $\Gamma$ ($\Gamma \sim \Gamma^{(0)} \sim \tau_0^{-1}$ because $\mathcal{E}_{av}$ is roughly the same value as in vacuum), i.e., $\Gamma/\omega_{sp} \sim 1/(8K_0) \gg 1$, which is satisfied if $K_0 < 1$, say [case (c)].

### B. Competition between atomic lines

In practice, fluorescence spectra involve the competition between many atomic transitions. As a simple model, consider an atom with an upper level $b$ and two lower levels $a_1$, $a_2$ with allowed transitions $b \to a_1$ (frequency $\Omega_1$) and $b \to a_2$ (frequency $\Omega_2$), as illustrated in Fig. 3. The "normal" way of dealing with such a situation is to calculate the partial rates $\Gamma_{e1}, \Gamma_{e2}$. (The subscript $e$ serves as a reminder that even the partial rates may not be given by the golden rule, and that these are *effective* partial rates, as discussed in Sec. III.) Then one obtains the total rate $\Gamma'_e = \Gamma_{e1} + \Gamma_{e2}$ and the branching ratios $B'_\mu = \Gamma_{e\mu}/\Gamma'_e$, $\mu = 1,2$. The prime indicates that we expect the addition of partial rates to fail in general, so that these are only nominal values.

Denote the amplitudes to be in $|b\rangle$, $|a_1 s\rangle$, and

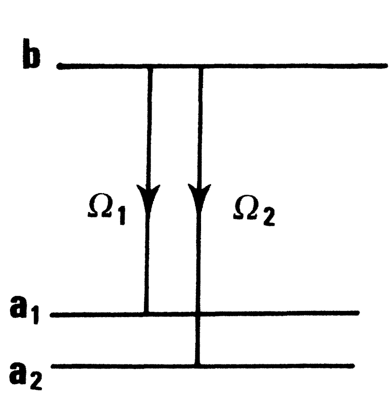


FIG. 3. Level scheme for two competing decay channels.

$|a_2 s\rangle$ as $C(t)$, $D_{1s}(t)$, and $D_{2s}(t)$ in the interaction representation, where, as before, $|a_1 s\rangle$ denotes the atom in state $|a_1\rangle$ together with one photon in mode $s$. We then obtain, in analogy to (11),

$$\frac{dC(t)}{dt} = -\frac{2\pi}{\hbar}\int d\omega\, \mathcal{E}(\omega)\omega \int_0^t dt' C(t') \times \left(M_1 e^{i(\Omega_1-\omega)(t-t')} + M_2 e^{i(\Omega_2-\omega)(t-t')}\right) , \tag{37}$$

where we have assumed

$$\langle a_\mu s \,|\, P_i \,|\, b \rangle \langle b \,|\, P_j \,|\, a_\mu s \rangle = M_\mu \delta_{ij}, \quad \mu = 1,2 . \tag{38}$$

Next assume the presence of two cavity resonances centered at $\omega_1$ and $\omega_2$,

$$\mathcal{E}(\omega)\omega = \sum_{\mu=1}^{2} \left[ \frac{\omega_\mu^3}{\pi^2 c^3} \right] K_\mu \frac{(\gamma_\mu/2)^2}{(\omega-\omega_\mu)^2 + (\gamma_\mu/2)^2} . \tag{39}$$

We shall illustrate only with some simple choices of parameters. First of all, the matrix elements will be chosen to be equal: $M_1 = M_2 = M$; the frequencies $\omega_1$, $\omega_2$ will be regarded as fairly close (since we expect $\omega_1, \omega_2$ to correspond to electronic transitions while their difference represents a vibrational shift), so that

$$4M\omega_1^3/\hbar c^3 \approx 4M\omega_2^3/\hbar c^3 = 1/\tau_0 . \tag{40}$$

Note that $1/\tau_0$ is now the *partial* rate for one of the transitions in vacuum, and the vacuum lifetime is $\tau_0/2$. Moreover, let the two cavity resonances have the same strength

$$K_1\gamma_1\tau_0 = K_2\gamma_2\tau_0 = K_0$$

so that the widths of the two resonances are now defined by the two remaining dimensionless parameters,

$$R_\mu = K_\mu/\gamma_\mu\tau_0, \quad \mu = 1,2 .$$

Furthermore, let the two atomic lines be exactly tuned: $\Omega_1 = \omega_1, \Omega_2 = \omega_2$. We put these into (37) and assume

$$|\Omega_1 - \Omega_2| \gg \gamma_1, \gamma_2$$

and ignore oscillating terms with frequencies of order $\Omega_1 - \Omega_2$. Mathematically, this means that the first (second) term in (39) multiplies only the first (second) term in the large parentheses in (37), while physically this means that, for example, a photon emitted in $b \to a_1 + \hbar\omega$ does not have the correct frequency to be absorbed in $a_2 + \hbar\omega \to b$, even as a virtual transition. Then some arithmetical calculation shows that $C(t)$ obeys (13), but with the kernel now being

$$S(t-t') = -\frac{1}{4\tau_0}\sum_{\mu=1}^{2} K_\mu\gamma_\mu e^{-\gamma_\mu(t-t')/2} , \tag{41}$$

which is just the sum of two terms as in (14) with $\Delta = 0$. The time evolution of $C(t)$ is readily solved in terms of its Laplace transform $\tilde{C}(p)$,

$$\tilde{C}(p)=\frac{(p+\gamma_1/2)(p+\gamma_2/2)}{p(p+\gamma_1/2)(p+\gamma_2/2)+\frac{K_0}{4\tau_0^2}[2p+(\gamma_1+\gamma_2)/2]}\,, \tag{42}$$

and the effective decay rate can be determined from

$$\Gamma_e^{-1}=\int_0^\infty dt\,|C(t)|^2=\int_{-\infty}^{\infty}\frac{d\xi}{2\pi}|\tilde{C}(-i\xi)|^2\,. \tag{43}$$

Some arithmetical calculation then gives, for $\overline{\Gamma}_e=K_0^{-1/2}\tau_0\Gamma_e$,

$$\overline{\Gamma}_e^{-1}=\int\frac{d\overline{\xi}}{2\pi}\frac{(\overline{\xi}^2+1/4R_1)(\overline{\xi}^2+1/4R_2)}{\overline{\xi}^2(\overline{\xi}^2-\alpha)^2+\beta^2(\overline{\xi}^2-1/4)^2}\,, \tag{44}$$

where

$$\alpha=\frac{1}{2}+\frac{1}{4\sqrt{R_1R_2}}\,,\quad \beta=\frac{1}{2}\left[\frac{1}{\sqrt{R_1}}+\frac{1}{\sqrt{R_2}}\right] \tag{45}$$

depend symmetrically on $R_1$ and $R_2$, and $\overline{\xi}$ has the same physical meaning as in (34).

The integral (44) equals

$$\overline{\Gamma}_e=(\overline{\Gamma}_{e1}+\overline{\Gamma}_{e2})/[1+\overline{\Gamma}_{e1}\overline{\Gamma}_{e2}(1+\sqrt{R_1R_2})]\,, \tag{46}$$

where the denominator shows that the addition of partial rates fails, and, in fact, $\Gamma_e<\Gamma_e'=\Gamma_{e1}+\Gamma_{e2}$, e.g., for $R_1=1$, $R_2=100$, $\overline{\Gamma}_e=0.39$, and $\overline{\Gamma}_e'=0.60$. We may understand this phenomenon roughly as follows. Suppose $R_1\lesssim 1$ and $R_2\gg 1$, then the process $b\leftrightarrow a_2+\hbar\omega$ occurs rapidly, but does not contribute much to the decay. Now the system spends only a fraction of the time in $|b\rangle$, so that the process $b\rightarrow a_1+\hbar\omega$ is suppressed accordingly. In fact, for

$$R_2>(R_1+1+R_1^{-1})^2/R_1\,,$$

we would have $\Gamma_e<\Gamma_{e1}$, i.e., the addition of a decay channel may *lower* the overall decay rate. However, if one line is weakly coupled (e.g., $\overline{\Gamma}_{e2},R_2$ small) then additivity of partial rates approximately holds.

Here we must consider possible direct transitions between $|a_1\rangle$ and $|a_2\rangle$, neglected in the above treatment. Radiative transitions are forbidden because $|a_1\rangle$ and $|a_2\rangle$ must have the same parity (since they couple to $|b\rangle$ by $E1$). Higher multipolar transitions are further suppressed by the relatively small phase space and the absence of cavity enhancement. For nonradiative transitions (e.g., induced by collisions), we must distinguish two cases. (a) The strongly coupled transitions connects to the lower level ($R_2\gg 1$ in Fig. 3). We may represent the nonradiative processes by coupling the atom to a bath at temperature $T$. Then when the system oscillates to $|a_2s\rangle$, the bath will not significantly induce a transition to $|a_1s\rangle$, provided $\hbar(\Omega_2-\Omega_1)\gg k_BT$, which is likely in practice. The above analysis then remains approximately valid. (b) The strongly coupled transition connects to the upper level ($R_1\gg 1$ in Fig. 4). Then when the systems oscillates to $|a_1s\rangle$, the bath will rapidly induce a nonradiative transition to $|a_2s\rangle$, and the above analysis without the bath becomes incorrect. It will be interesting to investigate these nonradiative couplings in detail. These issues become unimportant if experiments could be performed on single isolated atoms.

It is also interesting to consider the branching ratios

$$B_\mu=\sum_s|D_{\mu s}(\infty)|^2 =\frac{2\pi M}{\hbar}\int d\omega[\mathcal{E}(\omega)\omega]_\mu|\tilde{C}[i(\Omega_\mu-\omega)]|^2\,, \tag{47}$$

where $[\;]_\mu$ indicates that only the $\mu$th term in (39) is to be kept. Some algebraic calculation then leads to

$$B_1=\frac{1}{8\pi\sqrt{R_1}}\int d\overline{\xi}\frac{\overline{\xi}^2+1/(4R_2)}{\overline{\xi}^2(\overline{\xi}^2-\alpha)^2+\beta^2(\overline{\xi}^2-1/4)^2} \tag{48}$$

and $B_2$ is obtained from (48) by interchanging $R_1$ and

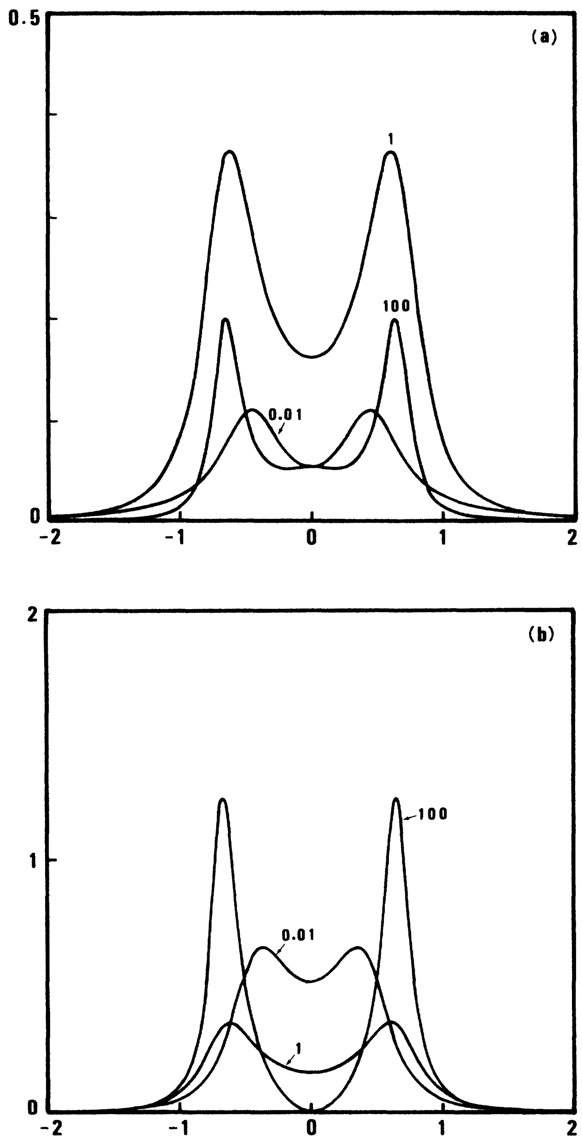


FIG. 4. Photon spectrum for the decay $b\rightarrow a_1+\hbar\omega$ vs $\overline{\xi}=K_0^{-1/2}\tau_0(\omega-\omega_0)$. (a) Fixed $R_2=1$ and various $R_1$ as shown on the curves. (b) Fixed $R_1=1$ and various $R_2$ as shown on the curves.

$R_2$. Somewhat surprisingly, this integral happens to equal the nominal branching ratio obtained by the additivity of rates

$$B_1 = B_1' \equiv \frac{\Gamma_{e1}}{\Gamma_{e1}+\Gamma_{e2}} , \tag{49}$$

where $\Gamma_{e\mu}$ are obtained by (28) with $R_\mu$ in place of $R$, and also setting $R^* = 1$ since we are considering a tuned case. We stress that the total rate does not satisfy additivity, even though the branching ratio does. In fact, the equality $B_1 = B_1'$ is merely an accident and does not hold in the general case with detuning or for cavity resonances with different strengths ($K_1\gamma_1 \neq K_2\gamma_2$). Nevertheless, (49) does confirm that a decay channel with *intermediate* $R$ would dominate when different channels compete.

The integrand in (48) is just the photon spectrum for the decay $b \to a_1 + \hbar\omega$, with $\bar{\xi} = K_0^{-1/2}\tau_0(\omega-\omega_1)$. Figure 4(a) shows the spectrum for fixed $R_2 = 1$ and various $R_1$. The spectrum is always double peaked even when this transition is weakly coupled ($R_1 \ll 1$), and there are no other cavity resonances in the vicinity. (Note that we have assumed $|\Omega_1 - \Omega_2|$ large.) In other words, when several competing channels coexist, the spectral lines cannot be interpreted independently. Figure 4(b) shows the same spectrum for fixed $R_1 = 1$ and different $R_2$, clearly demonstrating that "the other" decay channel can drastically affect the shape of the line under consideration. In these figures, the areas under the curves just give the branching ratios.

The above discussion with two (or, more generally, a few) lower levels is only relevant to simple molecules. For complex molecules, e.g., dyes, one must consider a large number of lower levels $a_1, a_2, \ldots$ forming a continuum. Then (37) becomes

$$\frac{dC(t)}{dt} = -\frac{2\pi}{\hbar}\int d\omega\, \mathcal{E}(\omega)\omega \times \int d\Omega \int_0^t dt' C(t') \times M(\Omega) e^{i(\Omega-\omega)(t-t')} , \tag{50}$$

where, assuming isotropic matrix elements,

$$\sum_\mu \langle a_\mu | P_i | b\rangle\langle b | P_j | a_\mu\rangle \delta(\Omega-\Omega_\mu) \equiv M(\Omega)\delta_{ij} . \tag{51}$$

Then the kernel $S$ is

$$S(t-t') = -\frac{2\pi}{\hbar}\int d\omega \int d\Omega\, \mathcal{E}(\omega)\omega M(\Omega) e^{i(\Omega-\omega)(t-t')} , \tag{52}$$

and since $M(\Omega)$ may be assumed to be broad, $S(t-t')$ is significant only for $t-t' \to 0$, so that (13) becomes Markovian,

$$\frac{dC(t)}{dt} \approx -\left[\frac{\Gamma}{2} + i\delta\right] C(t) , \tag{53}$$

$$\frac{\Gamma}{2} + i\delta = \frac{2\pi}{\hbar}\int d\omega \int d\Omega \frac{\mathcal{E}(\omega)\omega M(\Omega)}{i(\omega-\Omega+i\epsilon)} . \tag{54}$$

The important point is then that $S(t-t')$ has a broad frequency spectrum even if $\mathcal{E}(\omega)$ is narrow, so that the usual results—an exponential relaxation rate $\Gamma$ plus a frequency shift $\delta$—are obtained. Therefore the strong-coupling case does not arise for a continuous fluorescence spectrum.

### C. Background in cavity resonance

Even for a fairly sharp resonance, $\mathcal{E}(\omega)$ contains some background in addition to (12). Mathematically, the background can be represented by a second, broad Lorentzian, in the manner of (39). It can readily be shown that for a *single* atomic decay $b \to a + \hbar\omega$ into the sum of two Lorentzians, the time dependence is again given by (13) and (41), so the results of Sec. IV B can be directly applied to this case.

The second Lorentzian (i.e., background) is broad, so it belongs to weak coupling ($R_2 \ll 1$), and the additivity of rates apply. The rate for the background alone, $\Gamma_{e2}$, depends only on the background contribution to $\mathcal{E}(\omega)$, typically two orders of magnitude below the resonance contribution, and hence negligible on resonance. However, the background sets a lower limit of $\sim 1/\tau_0$ to the total decay rate.

## V. DISCUSSION

There are several possible approaches to the problem of electromagnetic interaction when the electromagnetic modes are affected by the environment (a cavity, or nearby walls) and are no longer plane waves. One could first solve the atom together with the plane waves, obtaining the usual vacuum decay rate, and then consider the effect of the environment on the emitted photon. This intuitive approach, often adopted in elementary discussions, assumes that the atom does not "know" about the environment, only the emitted photon does. The observed enhancement[10] as well as suppression[13] of electromagnetic decay for highly excited Rydberg atoms placed in tiny cavities point to the inadequacy of such a point of view.

More correctly, one must solve for the electromagnetic field together with the cavity or walls in the first instance, and then couple the atom to the normal modes of this combined system. This approach has been extensively discussed using linear-response theory[14] or electromagnetic eigenfunctions,[11,15] mainly for nearby surfaces, but also for cavity effects.[6,16] The approach in Ref. 16 is similar to the present work, but only the weak-coupling case is discussed there. The work in Ref. 6 is strictly one dimensional and again restricted to second-order perturbation theory, i.e., the adoption of the golden rule in some guise. To our knowledge, there has been no theoretical work pertaining to the strong-coupling limit.

A third approach uses the discrete (dissipative) quasimodes of the cavity rather than the continuous (Hermitian) modes of the universe,[17] in terms of which some of the results given here are known. The equivalence is sketched in Appendix A. The quasimode approach is usually justified in the following way. Let the central system $C$ (here the cavity) be described by a Hamiltonian $H_C$ and the bath $B$ (the rest of the universe) be described by a Hamiltonian $H_B$, with the two subsystems coupled by an interaction term $H_I$;

$$H = H_C + H_B + H_I \ .$$

The effect of $H_I$ is then eliminated and expressed as (a) a dissipation and (b) a fluctuating force acting on $C$.[18] There appears to be a little-noticed difficulty in applying such arguments to problems of the present type, which may be characterized in general as involving leaky cavities.[19] For example, the degrees of freedom of $C$ are $\mathbf{E}(\mathbf{r},t)$, $r<a$, while those of $B$ are $\mathbf{E}(\mathbf{r},t)$, $r>a$. The two are coupled not by a term in the Hamiltonian but by boundary conditions. The Hilbert space of wave functions satisfying suitable boundary conditions, hence the subspaces for $C$ and $B$, "know" about each other through the conditions at the interface. Formally, the Hilbert space of the universe, $\mathcal{H}$, then is not given by the direct product $\mathcal{H}_c \otimes \mathcal{H}_B$. This problem has prompted extensive studies of models "strings" in which subsystems are coupled by boundary conditions,[19,20] but to our knowledge a satisfactory basis for the quasimode approach is still lacking.

We may regard the present work in another light. Usual derivations of the golden rule pertain to a broad density of states, $\gamma \to \infty$. The opposite case of one nondissipative electromagnetic mode, i.e., $\gamma$ strictly equal to zero, has also been studied in detail,[21] again with emphasis on the inapplicability of the golden rule. The present work then provides an interpolation between these extremes. Similarly, departure from the golden rule for electromagnetic decay into a continuum ($\gamma \to \infty$) and into discrete modes ($\gamma \to 0$) has been studied for the $2p-1s$ transition in hydrogen;[22] the present formalism of narrow resonances ($\infty > \gamma > 0$) in principle provides an interpolation between the two.

It should also be noticed that whenever the kernel $S(t-t')$ is concentrated at small values of $t-t'$ [as in the "usual" case of broad $\mathcal{E}(\omega)$, or in (52)], then the formalism becomes identical to that of Weisskopf and Wigner.[12]

We must also discuss the application of the present formalism to the usual linear laser cavities, which belong to the strong-coupling case $R \gtrsim 1$, i.e., $\Gamma \gtrsim \gamma$, as is evident from the fact that laser linewidths are determined more by the cavity loss characteristics ($\gamma$) than by the atomic line shape ($\Gamma$). Moreover, the atomic line usually covers many cavity resonances, i.e., $K_0 \lesssim 1$. These estimates are given in Appendix B. Strong coupling is even more crucial for stimulated processes, especially at gain threshold (formally $\gamma \to 0$, so that $R \propto 1/\gamma \to \infty$). For this reason, the extension of this work to stimulation by electromagnetic waves of narrow bandwidth $\gamma$ can be very interesting.

However, laser cavities suffer from three drawbacks in the present context. First of all, there seems to be little experimental work on spontaneous processes, for which the theory, as developed here, is simplest. Secondly, many cavity resonances are involved, complicating the analysis. Finally, the structure of the electromagnetic modes is less well known. For example, an important parameter is the fraction $f_3$ of modes which are resonant, the rest $(1-f_3)$ being background.[4] In dielectric microspheres, the fraction $1-f_3$ corresponds to nearly radial rays which do not suffer total internal reflection, while the fraction $f_3$ corresponds to tangential rays; $f_3 \sim 0.2-0.4$, and is accurately computable for any given experimental situation. In linear laser cavities, the fraction $1-f_3$ describes rays at a large angle relative to the laser axis, which therefore escape; the fraction $f_3$ describes rays nearly parallel to the laser axis, which are therefore well confined and hence resonating. The relative importance of these two classes of modes is, to our knowledge, not well studied. One-dimensional models[6] are likely to be unrealistic because there are no off-axis rays. Thus experiments dedicated to spontaneous processes as well as a detailed calculation of electromagnetic modes are both necessary before the results obtained here can be applied in detail to laser cavities.

To summarize, spontaneous decay in the strong-coupling regime shows many unusual features: nonexponential decay, maximum decay rate for *intermediate* $Q$ values, nonadditivity of partial rates, and the possible *reduction* of the total decay rate upon the addition of an extra decay channel. The strong-coupling limit is experimentally accessible and the design of experiments to directly observe some of these interesting features should be contemplated. For example, a single transition between two levels in general shows up as double peaks in the photon spectrum. Analogous phenomena for stimulated processes will be relevant to lasers, and are under investigation.

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## APPENDIX A

In this appendix we sketch the connection to the quasimode approach. Consider a "two-state" system

$$\begin{bmatrix} 1 \\ 0 \end{bmatrix} = | b \rangle, \quad \begin{bmatrix} 0 \\ 1 \end{bmatrix} = | as \rangle \ ,$$

where $s$ indicates a photon in a single quasimode $s$. Let the energies be $\Delta$ and 0 (i.e., frequencies are measured from the center of the quasimode $\omega_0$); then taking $\hbar=1$, the Hamiltonian is

$$H = \begin{bmatrix} \Delta & \sigma \\ \sigma & -i\gamma/2 \end{bmatrix} , \tag{A1}$$

where the diagonal term $-i\gamma/2$ represents the dissipation of $| as \rangle$ and $\sigma$ represents coupling between $| b \rangle$ and $| as \rangle$. The energy eigenvalue $E$ satisfies

$$E^2+\left[\frac{i\gamma}{2}-\Delta\right]E-(\sigma^2+\frac{i\gamma}{2}\Delta)=0\ . \tag{A2}$$

On the other hand, the time dependence of the upper state is given in the interaction representation by $C(t)\propto e^{pt}$ with $p$ given by (18), or in Schrödinger representation by $e^{(p-i\Delta)t}$ so that $E=\Delta+ip$. Inserting this into (18) shows that $E$ satisfies

$$E^2+\left[\frac{i\gamma}{2}-\Delta\right]E-\left[\frac{K_0}{4\tau_0}+\frac{i\gamma}{2}\Delta\right]=0\ , \tag{A3}$$

so that the two descriptions give the same time dependence if we identify

$$\sigma^2=K_0/4\tau_0\ . \tag{A4}$$

Apart from the fact that such a comparison is necessary to identify parameters such as $\sigma$ in terms of fundamental quantities, the quasimode description is also handicapped by not distinguishing photons in the quasimode, so that one is unable to discuss the photon spectrum in a simple way.

## APPENDIX B

Here we estimate the parameters describing a linear cavity. Consider a laser cavity with length $L$, a waist area, say, $4\lambda^2$ (an optimistic estimate), and a fractional loss $\epsilon$ per reflection (including absorptive losses). Then

$$\gamma\sim\epsilon c/L\ . \tag{B1}$$

The density of states at the waist is

$$\rho(4\lambda^2 L)\sim\int\rho\,dV\equiv\rho^C\ , \tag{B2}$$

so that the integral over one resonance is

$$\int_{\text{res}}\rho\,d\omega\sim\frac{1}{4\lambda^2L}\int_{\text{res}}\rho^C d\omega\sim\frac{1}{4\lambda^2L} \tag{B3}$$

assuming no degeneracy. On the other hand $\rho$, or $\mathcal{E}$, is parametrized as (12), so

$$\int_{\text{res}}\rho\,d\omega\sim\left[\frac{\omega^2}{\pi^2c^3}\right]\left[\frac{\pi}{2}\right]K\gamma\ , \tag{B4}$$

giving

$$K\gamma\sim\frac{1}{8\pi}\frac{c}{L}\ . \tag{B5}$$

We then obtain

$$K_0\sim\frac{1}{8\pi}\frac{c\tau_0}{L},\quad R\sim\frac{1}{8\pi^2\epsilon^2}\frac{L}{c\tau_0}\ . \tag{B6}$$

For typical parameters, $c\tau_0\sim L$, $\epsilon\sim10^{-3}-10^{-2}$, we would have $K_0\sim1/(8\pi)\ll1$, $R\sim10^2-10^4\gg1$.


[1]R. E. Benner, P. W. Barber, J. F. Owen, and R. K. Chang, Phys. Rev. Lett. **44**, 475 (1980); H.-M. Tzeng, M. B. Long, and R. K. Chang, SPIE J. **573**, 80 (1985); H.-M. Tzeng, K. F. Walls, M. B. Long, and R. K. Chang, Opt. Lett. **9**, 273 (1984).

[2]J. B. Snow, S.-X. Qian, and R. K. Chang, Opt. Lett. **10**, 37 (1985); S.-X. Qian, J. B. Snow, and R. K. Chang, *ibid.*, **10**, 499 (1985); S.-X. Qian and R. K. Chang, Phys. Rev. Lett. **56**, 926 (1986).

[3]H.-M. Tzeng, K. F. Wall, M. B. Long, and R. K. Chang, Opt. Lett. **9**, 499 (1984); S.-X. Qian, J. B. Snow, H.-M. Tzeng, and R. K. Chang, Science **231**, 486 (1986).

[4]S. C. Ching, H. M. Lai, and K. Young, J. Opt. Soc. Am. B (to be published).

[5]A. G. Fox and T. Li, Bell Syst. Techn. J. **40**, 453 (1961).

[6]R. Lang, M.O. Scully, and W. E. Lamb, Phys. Rev. A **7**, 1788 (1973). J. C. Penaforte and B. Baseia, *ibid.* **30**, 1401 (1984).

[7]S. C. Hill and R. E. Benner, J. Opt. Soc. Am. B **3**, 1509 (1986).

[8]S. C. Ching, H. M. Lai, and K. Young, J. Opt. Soc. Am. B (to be published).

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

[10]S. Haroche, P. Goy, J. M. Raimond, C. Fabre, and M. Gross, Philos. Trans. R. Soc. London, Ser. A **307**, 659 (1982); P. Goy, J. M. Raimond, M. Gross, and S. Haroche, Phys. Rev. Lett. **50**, 1903 (1983).

[11]E. Power and T. Thiunamachandran, Phys. Rev. A **25**, 2473 (1982).

[12]V. Weisskopf and E. Wigner, Z. Phys. **63**, 54 (1930); W. Heitler, *The Quantum Theory of Radiation,* 3rd ed. (Oxford, Clarendon, 1954).

[13]D. Kleppner, Phys. Rev. Lett. **47**, 233 (1981); R. G. Hulet, E. S. Hilfer, and D. Kleppner, *ibid.* **55**, 2137 (1985).

[14]J. M. Wylie and J. E. Sipe, Phys. Rev. A **30**, 1185 (1984); **32**, 2030 (1985); A. D. McLachlan, Mol. Phys. **6**, 423 (1963); **7**, 381 (1963); G. S. Agarwal, Phys. Rev. A **11**, 230 (1975); **11**, 243 (1975); **11**, 253 (1975).

[15]M. J. Mehl and W. L. Schaich, Surf. Sci. **99**, 553 (1980); G. Barton, J. Phys. B **7**, 2134 (1974).

[16]P. Stehl, Phys. Rev. A **2**, 102 (1970).

[17]S. Sachdev, Phys. Rev. A **29**, 2627 (1984); G. S. Agarwal and R. R. Puri, *ibid.* **33**, 1757 (1986).

[18]W. H. Louisell and L. R. Walker, Phys. Rev. **137**, B204 (1965); I. R. Senitszky, *ibid.* **119**, 670 (1960); **124**, 642 (1961); **131**, 2827 (1963); M. Lax, *ibid.* **145**, 110 (1966); H. Mori, Prog. Theor. Phys. **33**, 424 (1965); R. Kubo, Rep. Prog. Phys. **29**, 255 (1966).

[19]H. M. Lai, P. T. Leung, and K. Young, Phys. Lett. A **119**, 337 (1987).

[20]H. Dekker, Phys. Lett. A **104**, 72 (1984); **105**, 395 (1984); **105**, 401 (1984); Phys. Rev. A **31**, 1607 (1985).

[21]E. T. Jaynes and F. W. Cummings, Proc. IEEE **51**, 89 (1963).

[22]M. Hillery, Phys. Rev. A **24**, 933 (1981).