Theory of Spontaneous-Emission Line Shape in an Ideal Cavity

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The spontaneous-emission spectrum of an atom in an ideal cavity is calculated.

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A single atom is an ideal "laboratory" in which to study the interactions of matter and radiation. The most fundamentally quantum mechanical of these intereactions is responsible for spontaneous emission. The nature of spontaneous radiation in free space is well known, but this is not the case if the radiating atom is enclosed in a cavity.[1]

Recently Kleppner[2] has proposed a microwave cavity experiment very far from resonance. He has emphasized the possibility of greatly suppressing the spontaneous emission rate of a Rydberg atom whose preferred (also microwave) transition frequency falls in a spectral region where the cavity mode density is much lower than the free-space density of states. Greatly decreased natural linewidths are one expected consequence.

It is not generally realized that a fully quantum mechanical theory of natural linewidth under idealized cavity conditions has never been given. We will mention below the principal reasons for this. In this Letter we present what, to the best of our knowledge, are the first fully quantum mechanical predictions about the spectrum of spontaneous emission into a lossless cavity, by an atom with an isolated transition frequency. The atomic transition frequency may be arbitrarily near to, or many linewidths away from, a single, similarly isolated, cavity resonance line. We show not only that quantum theory makes strikingly different predictions from the corresponding semiclassical (nonquantized electromagnetic field) theory,[1,3] but also that the predictions of quantum theory do not always correspond to those of "everyday" (QED) in which the natural radiated line shape is Lorentzian and the population of the radiating level decays exponentially in time.

Several unusual complications arise in any study of this problem. The first concerns the meaning of the term spectrum, and the role of the atom's two-time dipole autocorrelation function $D(t, \tau) = \langle \hat{d}^{(-)}(t+\tau)\hat{d}^{(+)}(t)\rangle$. Its value at $\tau = 0$ gives the time dependence of the emission process. Under normal circumstances, at steady state, when D is a function only of τ, its Fourier transform is the emission spectrum.

In any ideal lossless case, however, this relation between correlation function and spectrum fails. This is the first complication that must be dealt with, and it arises because we are interested in precisely the situation where relaxation is absent, or is barely active. There is no time t after which $D(t, \tau)$ depends only on τ. Thus the dipole correlation function, and the emitted radiation, cannot be stationary. Until recently there was no acceptable definition of spectrum for nonstationary radiation. We will use here the "physical spectrum" of Eberly and Wódkiewicz,[4] which is based directly on an idealized analysis of spectral measurement.

A second complication arises from the strong but loss-free coupling of the atom and the cavity. This is what is responsible for any lengthening of radiative lifetime, and spectral narrowing. It is necessarily present even if the atomic frequency is far from resonance or below the cavity cutoff. A perturbative analysis of the radiation-matter coupling will not be accurate, despite the smallness of $e^2/\hbar c$. Therefore nonperturbative theoretical methods are necessarily required for a satisfactory analysis of the correlation function and the spectrum.

A third complication involves the type of behavior that is expected of the atom. It is clear that lifetime lengthening is accomplished only at the expense of changes in the character of the emission. That is, an extremely low density of radiation modes in the working region not only increases the atomic lifetime, but also changes qualitatively the nature of the radiation process. We have pointed out elsewhere[5] some of the unexpected aspects of an atom's response to quantized single-mode radiation.

With these three issues in mind, we now describe our calculations and our predictions regarding the spectrum. The Hamiltonian (in the rotating-wave approximation) governing the interaction of an isolated atomic transition and an isolated cavity mode is given by[3]

$$\hat{H} = \hbar\omega_{21}\hat{\sigma}_{22} + \hbar\omega_c \hat{a}_c{}^\dagger \hat{a}_c + \hbar\lambda(\hat{\sigma}_{21}\hat{a}_c + \hat{a}_c{}^\dagger \hat{\sigma}_{12}). \quad (1)$$

Here $\hat{\sigma}_{mn}$ is the operator of the atom that equals $|m\rangle\langle n|$ at $t=0$ (i.e., $\hat{\sigma}_{21}$ is the raising operator from level 1 to level 2, etc.) and $\hat{a}_c$ and $\hat{a}_c^\dagger$ are the usual radiation mode operators. All operators are understood to be in the Heisenberg picture. The coupling constant λ, assumed real for simplicity, depends on the transition dipole matrix element and mode polarization, cavity volume, etc., as usual: $\lambda = [2\pi\hbar\omega_c/V]^{1/2}\hat{\epsilon}\cdot\vec{d}_{21}$.

The dipole correlation $D(t, \tau)$ that is needed is proportional to $\langle\hat{\sigma}_{21}(t+\tau)\hat{\sigma}_{12}(t)\rangle$, and the atom's radiated power spectrum is given, apart from an arbitrary normalization factor, by[4]

$$\mathcal{S}(\omega) = 2\gamma \,\mathrm{Re}\int_0^T d\tau \exp[\gamma - i(\omega-\omega_c)]\,\tau \int_0^{T-\tau} dt' \exp[-2\gamma(T-t')]D(t', \tau).$$

Here T is the length of time the excited atom is in the cavity, i.e., $\mathcal{S}(\omega)=0$ for $T\le 0$. The parameter γ is the half-bandwidth of whatever spectrometer is being used to measure the spectrum (here assumed for simplicity to be a scanning Fabry-Perot interferometer).

We have used methods appropriate to Hamiltonian (1), especially those of Ackerhalt,[6] to solve the relevant Heisenberg equations exactly. We have obtained an expression for $D(t, \tau)$ that is exact to all orders of the radiation-matter coupling, for all values of t and τ, and for all values of the atom-cavity frequency mismatch $\Delta = \omega_{21}-\omega_c$, including exact resonance $\Delta=0$. As anticipated, D is not stationary: $D(t, \tau) \ne D(0, \tau)$. This is evident even in the simplest interesting case where (see Fig. 1)

$$D_{+,0}(t, \tau) = (2\nu_0)^{-2}\exp[i(\omega_c+\Delta/2)\tau]\{(\nu_0+\Delta/2)^2\exp(i\nu_0\tau) + (\nu_0-\Delta/2)^2\exp(-i\nu_0\tau) + 2\lambda^2\cos[\nu_0(2t-\tau)]\}.$$

Here the subscripts + and 0 indicate that the atom was excited (+) and the field was empty of photons (0) in the initial state; and $\nu_0 = [(\Delta/2)^2+\lambda^2]^{1/2}$. The corresponding expression for $D_{-,0}(t, \tau)$ vanishes identically, as should be expected. Note that the population of the upper level [which is proportional to $D_{+,0}(t, 0)$] does not decay exponentially but oscillates instead at the frequency ν_0. Both $D_{+,0}$ and $D_{-,0}$ would vanish in a semiclassical theory of the kind used in the earliest studies of cavity emission,[1] in which the radiation field was not quantized. We have also computed $D_{\pm,\alpha}(t, \tau)$ where α signifies the amplitude of any coherent radiation present in the cavity initially. In this way we can model the experimental possibility that there may not be exactly zero photons in the cavity when the atom enters. The radiation spectrum associated with $D_{+,\alpha}(t, \tau)$ depends in a complicated way on the various parameters involved, and we present graphical results below.

There are two limiting cases of general interest, which we can describe analytically first.

Case (1). Pure long-time vacuum spectrum, broadband detection, atom far from resonance [$\alpha=0$; $\Delta \gg \gamma, \lambda \gg T^{-1}$]:

$$\mathcal{S}(\omega) \to \gamma + 2[(\omega-\omega_{21})^2+\gamma^2]^{-1}. \tag{2}$$

This is the case Kleppner[2] has proposed to study, and if $\gamma < A/2$, where A is the free-space Einstein spontaneous-emission coefficient, our result agrees with his prediction of line narrowing.

Case (2). Pure long-time vacuum spectrum, atom near to resonance [$\alpha=0$; $\gamma, \lambda \gg \Delta \gg T^{-1}$]:

$$\mathcal{S}(\omega) \to \gamma + 2f(\gamma, \lambda)\{[(\omega-\omega_c-\lambda)^2+\gamma^2]^{-1} + [(\omega-\omega_c+\lambda)^2+\gamma^2]^{-1}\}. \tag{3}$$

In the broadband detection limit ($\gamma \gg \lambda$) we find $f=\frac{1}{2}$ and (3) is essentially identical with (2). However, in the narrow-band detection limit ($\gamma \ll \lambda$), $f=\frac{1}{4}$ and (3) shows a completely new feature. In this case there are two resolved peaks in the spectrum, at $\omega = \omega_c \pm \lambda$. We call this result *vacuum-field Rabi splitting*, where 2λ plays the role of Rabi frequency. It is reminiscent of the fluorescence line splitting predicted in the presence of an intense laser field by Mollow[7] and others and observed recently,[8] but it is not the same because it occurs here *in the absence of a cavity field.*

We will discuss in detail elsewhere situations in which the "expected" characteristics of the atom's fluorescence spectrum are either absent or overshadowed by special features of the QED cavity interaction. Here we mention briefly the two examples that may be of most interest in designing possible experiments: the vacuum-field Rabi splitting predicted above, and the influence of nonzero initial cavity excitation on line narrowing.

In Fig. 2 we show a series of predicted spectra, each one for a different value of the frequency mismatch Δ between the cavity and atom. As Δ increases, the vacuum Rabi splitting also in-

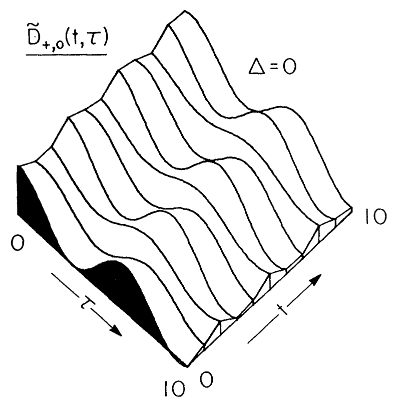

FIG. 1. A view of the dipole autocorrelation function $D_{+,0}(t,\tau)$ showing its dependence on t as well as τ. The tilde indicates that the rapid oscillation at ω_c has been eliminated.

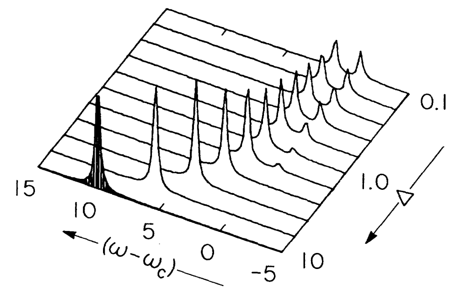

FIG. 2. A set of vacuum spectra, for which $T = 20\lambda^{-1}$, for values of atom-cavity detuning on a logarithmic scale from $\Delta = \frac{1}{10}$ (back line) to $\Delta = 10$ (front line). For small detuning the vacuum Rabi splitting is evident, and for large detuning the spectrum shows pure fluorescence. In between, one sees weak Rayleigh-type scattering at a position near $\omega = \omega_c$.

creases. At the same time one of the spectral peaks grows smaller. If the atom were in free space, this would be the Rayleigh scattering peak. In the limit, as Δ grows very large, only the "fluorescence" peak remains, and the spectrum has the character predicted by the approximate expression (2), consistent with the estimates of Kleppner, for example.

Figure 3 shows the effect, on the emission line shape, of any radiation that may already be in the cavity at the time the atom enters. The various spectra correspond to different values of the initial field strength. In the figure the cavity and the atomic transition are assumed exactly at resonance with each other. For small enough[9] values of initial field strength, vacuum-field Rabi splitting is evident. However, for larger values a transition region is evident where the vacuum-field splitting is obscured, before a different type of spectral shape emerges, a triplet of peaks. This triplet is, in fact, the exact analog of the intense-laser line splitting referred to above.[7,8] Spectra analogous to those shown in Fig. 3 have been calculated for an initially excited free-space atom.[10] They do not show doublet structure for weak initial fields.

In this short discussion we have (1) identified the complications that must be dealt with in a proper QED theory of the emission spectrum associated with atoms in ideal cavities; (2) constructed a moderately realistic quantum model for atomic emission in a cavity that includes the atom-cavity interaction and the spectral detection process; and (3) presented the new predictions of this theory for both the (necessarily nonstationary) dipole correlation function and the atomic emission spectrum.

Finally we emphasize the similarities as well as the strong qualitative differences between the predictions made here for spontaneous emission, using a consistent quantum theory of the atom-cavity interaction, and the predictions of "everyday" QED. There is a "familiar" domain (far from resonance) as shown in (2). Here the spectral emission line is indeed narrowed, and its width is limited only by the resolution of the spectrometer, in the ideal situation considered. However, there is another domain (near to resonance) in which atom-cavity line splitting is predicted by (3). And there is also a domain, predicted here for the first time, which reflects (see Fig. 3) the

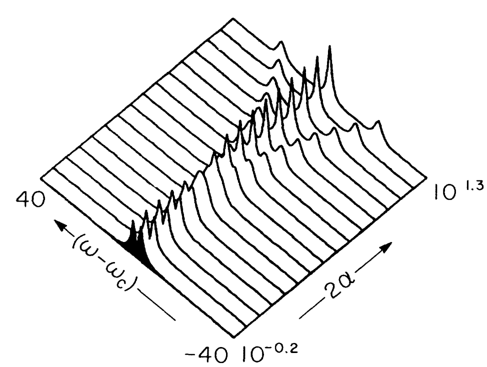

FIG. 3. A set of spectra, for which $T = 20\lambda^{-1}$, showing the influence of coherent radiation already present in the cavity at $t = 0$. The parameter 2α, which increases by the factor $10^{0.1}$ from spectrum to spectrum, is the effective Rabi frequency of the field initially in the cavity. The transition from two-peak vacuum Rabi splitting (small α) to three-peak ac Stark splitting (large α) is evident.

presence of any residual cavity excitation at $t = 0$. Such a prediction may possibly have relevance to actual experiments in which the cavity is not truly empty of photons at the outset of the observations. These two latter domains are joined by a region of complex atom-cavity coherence, which we will have to discuss elsewhere.

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[1]Emission in a cavity near resonance was the subject of early studies, both theoretical and experimental. See, for example, E. M. Purcell, Phys. Rev. 69, 681 (1946); N. Bloembergen and R. V. Pound, Phys. Rev. 95, 8 (1954); G. Feher, J. P. Gordon, E. Buehler, E. A. Gere, and C. D. Thurmond, Phys. Rev. 109, 221 (1958); A. Yariv, J. Appl. Phys. 31, 740 (1960). More recent work has appeared in whieh quantum features are emphasized. G. Barton, Proc. Roy. Soc. London, Ser. A 320, 251 (1970); P. Stehle, Phys. Rev. A 2, 102 (1970). However, neither of these papers is designed to deal with the present issue, namely the strong coupling of an isolated atomic transition with an isolated cavity resonance. Indeed, Barton's calculation (App. E) gives an infinite atomic linewidth in the small-cavity limit.

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