Spontaneous Emission of an Atom in a Cavity with Nonorthogonal Eigenmodes

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The spontaneous emission of an excited atom in a lossy cavity with nonorthogonal eigenmodes is analyzed. The quantum Langevin formalism is used to describe the dynamics of the spontaneous decay. The analysis shows that the spontaneous decay is modified by the Q value and the effective mode volume factor of each cavity eigenmode. The effective mode volume is generalized for cavities with nonorthogonal modes, which can be a very significant modification in the microcavity regime. It is shown that the spontaneous decay is not enhanced by the excess noise factor as claimed by other analyses.

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The eigenmode nonorthogonality of damped resonators has attracted significant interest in the quantum optics field in the past few years. It was first predicted by Petermann [1] that there is an excess noise factor in the fundamental laser linewidth for the gained guided semiconductor lasers. This result was unusual in the sense that it contradicts the well-accepted one noise photon per mode for spontaneous emission statement. This difficulty was later resolved by realizing the fact that spontaneous emission in different modes is correlated [2]. This excess noise factor was subsequently generalized by Siegman [3,4] for lasers and amplifiers with nonorthogonal eigenmodes.

This excess noise factor has raised serious investigation both experimentally and theoretically. Several research groups have experimentally confirmed this excess noise factor in various laser systems [5–11]. Further theoretical progresses have also been made [12–21]. Conventionally, the cavity eigenmodes are often assumed to be orthogonal and each mode is quantized as a simple harmonic oscillator. While the orthogonality assumption is a good approximation in some cases, the cavity eigenmodes are in general nonorthogonal due to the lossy cavity boundary condition. The question of how to quantize nonorthogonal eigenmodes of lossy cavities has stimulated series of papers [22–32], where various quantization formalisms have been proposed with some providing the quantum theory of excess noise factor. All the formalisms are nevertheless not totally equivalent. One discrepancy in the derived results is the spontaneous decay rate for atoms in cavities with nonorthogonal eigenmodes.

Some predicted no excess noise factor enhancement in the spontaneous decay rate [14,26,32], while others predicted an enhancement by the excess noise factor or the square root of that [23,28–31]. The difference might arise from the subtle differences in various quantization approaches and the handling of the correlations among nonorthogonal modes. In the original Petermann's laser linewidth analysis, the free space spontaneous emission rate was used for excited atoms, likewise in Siegman's analysis. In both analyses, it is the spontaneous emission coupled to each cavity eigenmode that is enhanced by the excess noise factor rather than the spontaneous emission of each atom is enhanced. If the spontaneous emission is enhanced by the excess noise factor, then based on both analyses, the fundamental laser linewidth will be enhanced by the square of that, which is inconsistent with experimental observations.

The cavity modified spontaneous decay has been studied in the past [33–36]. It was first pointed out by Purcell [33] that the spontaneous decay rate can be enhanced by the cavity Q value and mode volume factor due to the modified cavity vacuum electromagnetic field. The conventional analysis, however, paid little attention to the eigenmode nonorthogonality. This analysis extends the Purcell's result to cavities with nonorthogonal modes and shows that the spontaneous decay is not enhanced by the excess noise factor.

Conventionally, electromagnetic radiation is quantized in terms of a set of orthonormal modes. $\hat{E} = \sum_k \sqrt{\hbar\omega_k/(2\epsilon_0)}\{\hat{a}_{e_k} e_k + \hat{a}^\dagger_{e_k} e^*_k\}$, where e_k is the power normalized plane wave mode with two orthogonal polarization states implicitly included in index k and $[\hat{a}_{e_k}, \hat{a}^\dagger_{e'_k}] = \delta_{kk'}$. When field operator $\hat{E}$ is referenced to a general cavity eigenmode basis, it can be expressed in a similar form [32], $\hat{E} = \sum_n \sqrt{\hbar\omega_n/(2\epsilon_0)}\{\hat{a}_n u_n + \hat{a}^\dagger_n u^*_n\}$, where u_n is the cavity eigenmode and $\hat{a}_n$ and $\hat{a}^\dagger_n$ are the new creation and annihilation operators assigned to mode u_n. For a lossy cavity, the modes $\{u_n\}$ are in general not orthogonal but instead biorthogonal to a set of adjoint modes $\{\phi_n\}$, i.e., $(\phi_n|u_m) \equiv \int \phi^*_n u_m d\mathbf{x} = \delta_{nm}$. Projecting the annihilation and creation parts of the above expressions by $\int_{\mathbf{x}} \phi^*_n$ and $\int_{\mathbf{x}} \phi_n$, respectively, we readily obtain the transformation [32]

$$\hat{a}_n = \sum_k (\phi_n|e_k)\hat{a}_{e_k}, \qquad \hat{a}^\dagger_n = \sum_k (\phi_n|e_k)^*\hat{a}^\dagger_{e_k}. \tag{1}$$

The commutation relation for $\hat{a}_n$ is

$$[\hat{a}_n, \hat{a}^\dagger_m] = \sum_{i,j} (\phi_n|e_i)(e_j|\phi_m)[\hat{a}_{e_i}, \hat{a}^\dagger_{e_j}] = (\phi_n|\phi_m), \tag{2}$$

where the closure relation $\sum_i |e_i)(e_i| = 1$ is used.

We now analyze the dynamics of an excited two-level atom in a lossy cavity. The quantities of interest are the atomic upper and lower population density operators $\hat{\sigma}_e \equiv |e\rangle\langle e|$ and $\hat{\sigma}_g \equiv |g\rangle\langle g|$, and dipole operator $\hat{\sigma} \equiv |g\rangle\langle e|$. The interaction between atom and field operators are introduced by the electric-dipole interaction Hamiltonian [32],

$$H_I = \sum_n \sqrt{\frac{\hbar\omega_n}{2\epsilon_0}}\{\hat{a}^\dagger_{u_n} u^*_n(\mathbf{x}) \cdot \hat{\sigma}(\mathbf{x}) e\vec{d} + \text{H.c.}\} \quad (3)$$

$$= \sum_n g_n \hbar \{\hat{a}^\dagger_{u_n} u^*_n(\mathbf{x})\hat{\sigma}(\mathbf{x}) + \text{H.c.}\}, \quad (4)$$

where $e\vec{d} = e\langle e|\vec{r}|g\rangle$ is the atomic dipole moment. The coupling factor $g_n = \sqrt{\omega_n/(2\epsilon_0\hbar)}\vec{\varepsilon}_n \cdot e\vec{d}$, where $\vec{\varepsilon}_n$ is the mode polarization vector. With the above interaction Hamiltonian, we have the following coupled quantum Langevin operator equations [32]

$$\frac{d}{dt}\hat{\sigma}_e(\mathbf{x}) = i\sum_n g_n\{\hat{a}^\dagger_n u^*_n \hat{\sigma}(\mathbf{x}) - \hat{\sigma}^\dagger(\mathbf{x})\hat{a}_n u_n\} \quad (5)$$

$$\frac{d}{dt}\hat{\sigma}_g(\mathbf{x}) = -i\sum_n g_n\{\hat{a}^\dagger_n u^*_n \hat{\sigma}(\mathbf{x}) - \hat{\sigma}^\dagger(\mathbf{x})\hat{a}_n u_n\} \quad (6)$$

$$\frac{d}{dt}\hat{\sigma}(\mathbf{x}) = i\sum_n g_n\{\hat{\sigma}_e(\mathbf{x}) - \hat{\sigma}_g(\mathbf{x})\}\hat{a}_n u_n \quad (7)$$

$$\frac{d}{dt}\hat{a}_n = -\gamma_n \hat{a}_n - i g_n \phi^*_n \hat{\sigma}(\mathbf{x}) + F_n, \quad (8)$$

where γ_n is the decay rate of mode u_n and the atom is at position $\mathbf{x}$. F_n is the noise operator, where $[\hat{F}_n(t), \hat{F}^\dagger_n(t')] = 2\gamma_n[\hat{a}_n, \hat{a}^\dagger_n]\delta(t-t')$ to conserve the commutation relations for $\hat{a}_n$. The spontaneous decay of atom comes from two sources. One is from the interaction with the cavity-modified field, which is in turn damped by the reservoir through the lossy boundary condition as described by Eq. (8). The other one is the direct interaction with part of the reservoir field, e.g., those propagate perpendicular to the cavity axis. Here, we consider the case where the local field is significantly modified by cavity, therefore focusing on the first contribution. The resonant frequency spacing of cavity longitudinal modes is assumed to be much larger than the natural atomic linewidth; thus, only one longitudinal mode can significantly interact with the atom. On the other hand, there could be multiple transverse modes interacting with the atom because transverse mode frequency spacing is typically much smaller. For the simplicity of analysis, the dipole transition frequency is assumed to match to the resonant frequency of the lowest order mode $\hat{a}_0$; therefore, γ_0 is real. For other modes, the cavity decay rate γ_n will be a complex value, where the imaginary part accounts for the difference between atomic and resonant mode frequency.

The presence of adjoint mode ϕ_n in the amplitude operator rate Eq. (8) is derived from the interaction Hamiltonian

$$\frac{i}{\hbar}[H_I, \hat{a}_n] = i\sum_m g_m[\hat{a}^\dagger_m u^*_m(\mathbf{x})\hat{\sigma}(\mathbf{x}), \hat{a}_n] \quad (9)$$

$$= -i\sum_m g_m(\phi_n|\phi_m)u^*_m(\mathbf{x})\hat{\sigma}(\mathbf{x}) \quad (10)$$

$$= -i g_n \phi^*_n(\mathbf{x})\hat{\sigma}(\mathbf{x}), \quad (11)$$

where the approximation $\sum_m g_m(\phi_n|\phi_m) \simeq g_n \sum_m(\phi_n|\phi_m)$ is used because the summation is mainly contributed by the modes ϕ_m with frequencies close to that of ϕ_n.

We assume that the cavity decay rate is much faster than the atomic decay rate. The field amplitude $\hat{a}_n$ then adiabatically follows dipole moment,

$$\hat{a}_n = \frac{-i g_n \phi^*_n(\mathbf{x})\hat{\sigma}(\mathbf{x}) + F_n}{\gamma_n}. \quad (12)$$

Substituting this expression into Eq. (5), we have

$$\frac{d}{dt}\hat{\sigma}_e = -\hat{\sigma}^\dagger\hat{\sigma}\sum_n g^2_n \frac{\phi^*_n u_n + \text{c.c.}}{\gamma_n} + \sum_n g_n \frac{i F^\dagger_n u^*_n \hat{\sigma} + \text{H.c.}}{\gamma_n}. \quad (13)$$

Before taking reservoir average, we need to evaluate the second term of the above equation further to the order of g^2_n. From the dipole Eq. (7),

$$\hat{\sigma}_t = \hat{\sigma}_{t-\Delta t} + \int_{t-\Delta t}^{t}\sum_n i g_n(\hat{\sigma}_e - \hat{\sigma}_g)_{t'}\hat{a}_{nt'}u_n dt', \quad (14)$$

where time dependence is indicated in subscript. Using this expression

$$\sum_n g_n \frac{i u^*_n F^\dagger_{nt}\hat{\sigma}_t}{\gamma_n} = \sum_n g_n \frac{i u^*_n F^\dagger_{nt}\hat{\sigma}_{t-\Delta t}}{\gamma_n} - \sum_{nn'} g_n g_{n'} \frac{u^*_n u_{n'}}{\gamma_n} \times \int_{t-\Delta t}^{t}(\hat{\sigma}_e - \hat{\sigma}_g)_{t'}F^\dagger_{nt}\hat{a}_{n't'}dt'. \quad (15)$$

The cavity eigenmode noise operator F_{nt} is not correlated to $\hat{\sigma}_{t-\Delta t}$ operator. The reservoir average of $F^\dagger_{nt}\hat{\sigma}_{t-\Delta t}$ is zero and can be removed. Use Eq. (12) for $\hat{a}_{n't'}$,

$$\langle F^\dagger_{nt}\hat{a}_{n't'}\rangle = \left\langle\frac{-i g_{n'}\phi^*_{n'}F^\dagger_{nt}\hat{\sigma}_{t'}}{\gamma_{n'}}\right\rangle + \left\langle\frac{F^\dagger_{nt}F_{n't'}}{\gamma_{n'}}\right\rangle. \quad (16)$$

Again, the reservoir average of $F^\dagger_{nt}\hat{\sigma}_{t'}$ is zero because F_{nt} is not correlated to $\hat{\sigma}_{t'}$. The noise operators $F^\dagger_{nt}$ and $F_{n't'}$ are $\delta(t-t')$ correlated, $\langle F^\dagger_{nt}F_{n't'}\rangle = \langle F^\dagger_n F_{n'}\rangle\delta(t-t')$, where the diffusion coefficient $\langle F^\dagger_n F_{n'}\rangle = (\gamma_n + \gamma_{n'})\langle\hat{a}^\dagger_n\hat{a}_{n'}\rangle$ from fluctuation dissipation theorem.

With these results, the reservoir average of Eq. (13) becomes

$$\frac{d}{dt}\langle\hat{\sigma}_e\rangle = -\left\{\sum_n g_n^2 \frac{\phi_n^* u_n}{\gamma_n}\langle\hat{\sigma}_e\rangle + \text{H.c.}\right\} - \left\{\sum_{nn'} g_n g_{n'} u_n^* u_{n'} \frac{\gamma_n + \gamma_{n'}}{2\gamma_n\gamma_{n'}}\langle\hat{\sigma}_e - \hat{\sigma}_g\rangle\langle\hat{a}_n^\dagger \hat{a}_{n'}\rangle + \text{H.c.}\right\}. \tag{17}$$

The above equation is the essential result. The first term describes the spontaneous decay. The second term describes the stimulated decay due to cavity field. This result is similar to a previous derivation [29] except the additional derived stimulated decay term. When the cavity modes are orthogonal, i.e., $\phi_n = u_n$ and $\langle\hat{a}_n^\dagger\hat{a}_{n'}\rangle = \langle\hat{n}\rangle\delta_{nn'}$, where $\hat{n}$ is the photon number operator, this equation reduces to a conventional form

$$\frac{d}{dt}\langle\hat{\sigma}_e\rangle = -\sum_n 2g_n^2\frac{u_n^* u_n}{\gamma_n}\langle\hat{\sigma}_e\rangle - \sum_n 2g_n^2\frac{u_n^* u_n}{\gamma_n}\langle\hat{\sigma}_e - \hat{\sigma}_g\rangle\langle\hat{n}\rangle. \tag{18}$$

We see that the one noise photon per mode for spontaneous emission is recovered. The $u_n^* u_n$ term is in fact the effective volume factor $1/V_{\text{eff}} \equiv \sum_n u_n^* u_n / \int u_n^* u_n$ in the classic cavity enhanced spontaneous decay literatures [33–37]. The expression $\text{Re}\{\sum_n \phi_n^* u_n\}$ in Eq. (17) is therefore a generalized effective mode volume factor for cavities with nonorthogonal modes, $1/V_{\text{eff}} \equiv \text{Re}\{\sum_n \phi_n^* u_n\}$. This factor is an important modification in the microcavity regime, where only one or few modes contribute to the spontaneous decay.

We now apply the result to calculate the spontaneous decay rate of an atom inside a cavity with nonorthogonal eigenmodes. The cavity is formed by two symmetric spherical mirrors located at $z = 0$ and $-L$ with a Gaussian reflectivity profile and negative focal length. This cavity is chosen mainly because analytical expressions of eigenmodes are available. The atom is at the center of the cavity $z = -L/2$. This unstable cavity has non-power orthogonal Hermite-Gaussian eigenmodes. For simplicity, let us consider only the lowest order mode. The contribution from each high order mode adds linearly to the total decay rate but with smaller magnitude due to resonant frequency offset and higher cavity decay rate. The lowest order transverse mode at the middle of cavity is [38],

$$u_{T,0} = \left(\frac{2}{\pi w^2}\right)^{1/2}\exp\left(-i\frac{\pi r^2}{\tilde{q}\lambda}\right), \tag{19}$$

where $r^2 = x^2 + y^2$ and $1/\tilde{q} \equiv 1/R - i\lambda/\pi w^2$. The adjoint mode is

$$\phi_{T,0} = -i\frac{(2\pi)^{1/2}w}{\tilde{q}^*\lambda}\exp\left(i\frac{\pi r^2}{\tilde{q}^*\lambda}\right). \tag{20}$$

To be complete, we should also include the longitudinal mode function. The longitudinal eigenmode and adjoint mode, with the forward and backward propagation spelled out, are [26]

$$u_{L,n} = \frac{1}{p}\begin{pmatrix} e^{(ik_n+\gamma_z)z} \\ re^{(-ik_n-\gamma_z)z}\end{pmatrix} \quad \text{and} \quad \phi_{L,n} = \frac{p}{2L}\begin{pmatrix} e^{(ik_n-\gamma_z)z} \\ \frac{1}{r}e^{(-ik_n+\gamma_z)z}\end{pmatrix}, \tag{21}$$

where $r = -e^{-\gamma_z L}$, k_n is the resonant wave vector value and p is the power normalization constant. It is straightforward to find the spontaneous decay rate for an atom at the center of the cavity,

$$g_0^2\frac{\phi_0 u_0^* + \text{c.c.}}{\gamma_0} = \frac{e^2 d^2 \omega_0^3}{3\pi\epsilon_0\hbar c^3}\frac{\omega_0}{2\gamma_0}\frac{6\pi c^3}{\omega_0^3 V_{\text{eff}}}, \tag{22}$$

where $V_{\text{eff}} = L\pi w^2/2$ is the effective mode volume. The first factor on the right-hand side is the free space decay rate. The second factor is the cavity quality factor Q. The third factor depends on the cavity geometry and is proportional to the λ^3 to V_{eff} ratio. This result is similar to the previous cavity-modified spontaneous decay study [34] except that V_{eff} is now obtained from the generalized effective mode volume expression.

One may try to express the effective mode volume in terms of the transverse excess noise factor K_n. At the cavity symmetric plane, the transverse adjoint mode is related to the eigenmode by $\phi_n = \sqrt{K_n}e^{-i\theta_n}u_n^*$. For the lowest order mode, $\phi_0 = (1 - i\pi w^2/\lambda R)u_0^* = \sqrt{K_0}e^{-i\theta_0}u_0^*$, where $K_0 = 1 + (\pi w^2/\lambda R)^2$ and $\theta_0 = \tan^{-1}(\sqrt{K_0 - 1})$. The effective mode volume factor is rewritten as

$$\frac{1}{V_{\text{eff}}} = \text{Re}\left\{\frac{1}{L}\sqrt{K_0}e^{i\theta}u_0^2\right\} \tag{23}$$

$$= \text{Re}\left\{\frac{2}{L\pi w^2}\sqrt{K_0}e^{i\theta}e^{-2(r^2/w^2)\sqrt{K_0}e^{i\theta}}\right\}. \tag{24}$$

We see that $1/V_{\text{eff}}$ depends on the atomic position r and is a complicated function of $\sqrt{K_0}$ due to the phase term θ_0, which is an inverse trigonometric function of $\sqrt{K_0 - 1}$. However, at $r = 0$, $1/V_{\text{eff}}$ is reduced to $\text{Re}\{(2/L\pi w^2) \times \sqrt{K_0}e^{i\theta_0}\} = 2/(L\pi w^2)$ and is independent of K_0. In fact, it can be shown that $\text{Re}\{\phi_n^* u_n\}$ is independent of K_n at $r = 0$ for all higher order complex Hermite-Gaussian modes. This is different from the previous analysis [23,28–31] where the spontaneous decay is claimed to be enhanced by K or $\sqrt{K}$ for an atom at the center of the cavity.

In general, the excess noise factor K increases as the geometrical magnification M of the unstable cavity increases, e.g., $K \simeq (M^2 - 1)^4/M^4$ for the lowest order mode [38]. The cavity $Q \simeq 1/\ln M$. The pro-K enhancement prediction $\gamma_a \simeq Q\sqrt{K}$ or $\simeq QK$ is then an increasing function of M. However, as M increases, the atom sees more of the external reservoir and the cavity-modified spontaneous decay rate should reduce to free space value. This discrepancy also occurs for cavities with longitudinal nonorthogonal modes. One can consider a symmetric stable cavity with finite mirror reflectivity R. The excess noise factor is $K = (1 - R)^2/R(\ln R)^2$ and $Q \simeq -1/\ln R$. The value for $Q\sqrt{K}$ or QK monotonically increases as mirror reflectivity R decreases. This again contradicts the expected free space decay rate as R approaches zero. On the other hand, the derived decay rate $\sum_n 2g_n^2 \,\mathrm{Re}\{\phi_n^* u_n/(\gamma_n + i\Delta\omega_n)\}$, where $\Delta\omega_n$ is the mode frequency detuning from atomic resonance, can be reduced to free space value when γ_n approaches infinity. Let us consider a cubic cavity with length L and mirror reflectivity R. The x, y, and z components of cavity eigenmodes are similar to the nonorthogonal longitudinal modes in Eq. (21). The frequency detuning $\Delta\omega_n = n\Delta\omega_{ax}$, where $\omega_{ax} = \pi c/L$ is the axial mode frequency spacing. When R is large and L is small, i.e., $\gamma_n \ll \omega_{ax}$, the decay rate is mainly dominated by one mode. When R is small, the mode decay rate γ_n is large. We need to sum up the contributions from all modes for the spontaneous decay because $\omega_{ax} \ll \gamma_n$. The summation $\sum_n$ can be approximated by $\int \rho(\omega) d\omega$, where the mode density $\rho(\omega) = L^3\omega^2/(\pi^2 c^3)$. The summation becomes

$$\int \frac{e^2 d^2 \omega_0^3}{3\pi\epsilon_0 \hbar c^3} \mathrm{Re}\left\{\frac{1}{\gamma_n + i\Delta\omega_n} \frac{3\pi c^3}{\omega_0^2 L^3}\right\} \frac{L^3\omega^2}{\pi^2 c^3} d\omega = 3\frac{e^2 d^2 \omega_0^3}{3\pi\epsilon_0 \hbar c^3}. \tag{25}$$

When we take into account the atomic random polarization direction, the factor of 3 is reduced to 1 and the free space spontaneous decay rate is recovered. The same calculation can also be used to obtain the free space decay rate for L approaching infinity.

In summary, a quantum rate equation for an atom in a lossy resonator is derived. The cavity eigenmode nonorthogonality is explicitly addressed in the derivation. The effective mode volume in the conventional cavity-modified spontaneous decay rate expression is generalized for cavities with nonorthogonal modes. By considering a simple example, it is shown that the spontaneous decay rate depends only on the cavity Q value and the generalized V_{eff} and is independent of the excess noise factor. A physical argument is provided to support this independence of excess noise factor for spontaneous decay. The reduction to the free space value at the limit of infinitesimally small mirror reflectivity and infinitely large cavity is also shown.

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