Spontaneous Emission in Chaotic Cavities

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The spontaneous emission rate $\Gamma$ of a two-level atom inside a chaotic cavity fluctuates strongly from one point to another because of fluctuations in the local density of modes. For a cavity with perfectly conducting walls and an opening containing $N$ wavechannels, the distribution of $\Gamma$ is given by $P(\Gamma) \propto \Gamma^{N/2-1}(\Gamma + \Gamma_0)^{-N-1}$, where $\Gamma_0$ is the free-space rate. For small $N$ the most probable value of $\Gamma$ is much smaller than the mean value $\Gamma_0$.

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The modification of the rate of spontaneous emission in a cavity has been a subject of extensive research \cite{1-8}. It was shown that the cavity can both enhance and inhibit the spontaneous emission at microwave and optical frequencies. The effect is due to a modification by the environment of the local density of modes at the position of the radiating atom. The efforts were concentrated on the fabrication of cavities of prescribed regular shape, the atoms being kept close to nodes or antinodes of the field patterns of the cavity modes.

What can be said if the shape of the cavity is not regular and the exact position of the atom is unknown? Irregular cavities have a complicated “chaotic” field pattern, and it becomes difficult to state whether the spontaneous emission rate $\Gamma$ of a particular atom is increased or decreased with respect to the free-space rate $\Gamma_0 = d^2\omega_0^3/3\pi\epsilon_0 \hbar c^3$ (corresponding to an electric dipole transition with moment $d$, frequency $\omega_0$). Nevertheless, a precise statement can be made about the statistical distribution of $\Gamma$. The distribution is universal, i.e. independent of the shape or size of the cavity, provided it is chaotic.

A chaotic cavity is large compared to the wavelength $\lambda_0 = 2\pi c/\omega_0$, and has a shape such that the light is scattered uniformly in phase space. (In a circular or cubic cavity, chaotic behavior may still occur because of diffuse boundary scattering or due to randomly placed scattering centers.) The only parameter which enters the distribution of $\Gamma/\Gamma_0$ is the strength of the coupling of the cavity modes to the outside world. We assume that the coupling is via a hole that is small compared to the size of the cavity and transmits a total of $N$ wavechannels.

For a hole of area $A$, $N \approx 2\pi A/\lambda_0^2$. Our result for the distribution of $\Gamma$ takes the universal form

$$P(\Gamma) \propto \frac{\Gamma^{N/2-1}}{(\Gamma + \Gamma_0)^{N+1}},$$

shown in Fig. 1 for several values of $N$. The distribution eventually becomes narrow and Gaussian for $N \gg 1$, while it is still broad and strongly non-Gaussian for $N$ as large as 10. The mean value of $\Gamma$ equals $\Gamma_0$ but the most probable value is smaller than $\Gamma_0$.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{fig1.png}
\caption{Probability distribution of the spontaneous emission rate $\Gamma$ (normalized by the free-space rate $\Gamma_0$), as given by Eq. (1) for several values of the number $N$ of wavechannels transmitted by the hole in the cavity. For $N \geq 2$ the distribution reaches its maximum at a rate $\Gamma = \Gamma_0(N-2)/(N+4)$ that is smaller than the mean value $\Gamma_0$. The variance of $\Gamma$ diverges for $N \leq 2$ and equals $4\Gamma_0^2/(N-2)$ for larger $N$. The dashed curve is the result (1) for a hole much smaller than a wavelength (transmittance $T = 0.1$).}
\end{figure}

As a possible experimental setup, one can imagine an array of cavities, each containing a few excited atoms, or a single cavity containing many excited atoms (widely separated so that they decay independently). The array of cavities might occur naturally in a porous material. Let $n(t)$ be the number of atoms that has not decayed by the time $t$. The fraction $n(t)/n(0)$ is the Laplace transform $\int_0^\infty \Gamma P(\Gamma) \exp(-\Gamma t) d\Gamma$ of the distribution (1), which is a confluent hypergeometric function. A time-
resolved measurement of the emitted intensity yields \( n(t) \) and thereby the probability distribution \( P(\Gamma) \). Fluctuations of the spontaneous emission rate give rise to an algebraic decay \( n(t) \propto t^{-N/2} \) for large \( t \), instead of the usual exponential decay \( \propto \exp(-\Gamma_0 t) \).

We proceed with the derivation of Eq. (1). We assume that the system is in the perturbative regime (3), so that the rate of spontaneous emission is given by the Fermi golden rule,

\[
\Gamma = \frac{2}{\hbar^2} \text{Im} \sum_{\mu} \langle f_{\mu}^R|\hat{\mathcal{E}}|0\rangle \langle 0|\hat{\mathcal{E}}^\dagger|f_{\mu}^L\rangle / (\omega_{\mu} - \omega_0 - i\gamma_\mu/2). \tag{2}
\]

Here \(|0\rangle\) is the initial state (excited atom + no photons) and \(|f_{\mu}^{L,R}\rangle\) is the final state (atom in the ground state + one photon in mode \( \mu \) with frequency \( \omega_{\mu} \), broadening \( \gamma_\mu \)). The index \( L \) or \( R \) refers to left and right eigenfunctions of the Maxwell equations in the open cavity, which form a biorthogonal set of modes. The conditions for the validity of perturbation theory will be discussed later.

Eq. (2) can be rewritten in terms of the local density of modes at the position \( \vec{r} \) of the atom,

\[
\Gamma = \frac{\pi \omega_0 q^2}{\hbar \varepsilon_0} \rho(\vec{r}, \omega_0), \tag{3}
\]

\[
\rho(\vec{r}, \omega) = \frac{1}{\pi} \text{Im} \sum_{\mu} \frac{E_{\mu}^{L*}(\vec{r}) E_{\mu}^R(\vec{r})}{\omega_{\mu} - \omega - i\gamma_\mu/2}, \tag{4}
\]

where \( E_{\mu}^{L,R} \) is the component along \( \vec{d} \) of the electric field in left or right mode \( \mu \). We consider an almost empty cavity without any dispersive or absorptive medium inside, in which case the distinction between the total and radiative density of modes (10) is irrelevant.

For a statistical description we study an ensemble of chaotic cavities with the same volume \( V \) and small variations in shape. The average density of modes \( \langle \rho(\vec{r}, \omega_0) \rangle \equiv \rho_0 = \omega_0^2/3\pi^2e^3 \) corresponds to the average rate \( \Gamma_0 \). Our aim is to find the probability distribution of \( \rho \). In Refs. (12) this distribution was obtained under the assumption that the broadening \( \gamma_\mu \) was the same for all modes and all cavities. In our problem, the broadening is different for each mode and each cavity, and the distribution turns out to be entirely different.

According to the universality hypothesis of chaotic systems, the statistical distribution of \( \rho \) can be described by the random-matrix theory of chaotic scattering (14). Starting point is the expression of the \( N \times N \) scattering matrix \( \mathcal{S} \) in terms of an \( M \times M \) real symmetric matrix \( H \) (representing the discretized Helmholtz operator of the closed cavity) and an \( M \times N \) coupling matrix \( W \),

\[
\mathcal{S}(\omega) = 1 - 2\pi i W^\dagger (\omega - H + i\pi WW^\dagger)^{-1}W. \tag{5}
\]

The matrix \( H \) is taken from the Gaussian orthogonal ensemble of random-matrix theory,

\[
P(H) \propto \exp\left[ -\left( \pi \rho_0 \right)^2 \text{tr} H^2/4M \right]. \tag{6}
\]

The limit \( M \to \infty \) is taken at the end of the calculation. The coupling matrix \( W \) has elements \( W_{mm} = (M/\rho_0 \pi)^{1/2} \delta_{mm}^{-1} \).

The local density of modes is obtained from a diagonal element of the Green function \( G(\omega) = (\omega - H + i\pi WW^\dagger)^{-1} \),

\[
\rho(\vec{r}_m, \omega) = -(M/\pi \omega) \text{Im} \text{tr} G_{mm}(\omega), \tag{7}
\]

where \( \vec{r}_m \) is the point in space associated with the index \( m \). Because of the orthogonal invariance of \( P(H) \), the distribution of \( \rho \) is independent of \( m \). Using Eq. (6), we can rewrite Eq. (7) in terms of the scattering matrix,

\[
\rho = \frac{M}{2\pi \omega} i \text{tr} S^\dagger \partial S/\partial H_{mm}. \tag{8}
\]

This representation of the local density of modes is the matrix analogue of the relationship (3) between the local density of electronic states and the functional derivative of the scattering matrix with respect to the local electrostatic potential, \( \rho(\vec{r}) = (i/2\pi)\text{tr} S^\dagger \partial S/\partial V(\vec{r}) \).

The matrix \( S^\dagger \partial S/\partial H_{mm} \) is closely related to the matrix

\[
Q = -i S^\dagger \partial S/\partial \omega, \tag{9}
\]

known as the Wigner-Smith time-delay matrix (16). Namely, in view of Eq. (5) we have

\[
i \text{tr} S^\dagger \partial S/\partial H_{mm} = (AA^\dagger)_{mm}, \quad Q = A^\dagger A, \tag{10}
\]

where \( A = (2\pi)^{1/2} GW \). Since \( A \) is an \( M \times N \) matrix, the product \( AA^\dagger \) has \( M \times N \) zero eigenvalues. The remaining \( N \) nonzero eigenvalues are the same as the eigenvalues of \( Q \), which are the so-called proper delay times (17) \( \tau_1, \ldots, \tau_N \). Their statistical distribution is known (18).

\[
P(\tau_1, \ldots, \tau_N) \propto \prod_{i<j} |\tau_i - \tau_j| \prod_{k} \tau_k^{-3N/2-1} e^{-\pi \rho_0 V/\tau_k}. \tag{11}
\]

For the local density of modes (8), this implies that

\[
\rho = \frac{M}{2\pi \rho_0} \sum_{j=1}^N \tau_j^2 u_j^2, \tag{12}
\]

where \( u_j \) is the \( j \)-th element of the eigenvector of \( AA^\dagger \) corresponding to the eigenvalue \( \tau_j \). In the limit \( M \to \infty \), the distribution of the vector \( \vec{u} \) is Gaussian, \( P(\vec{u}) \propto \exp(-\frac{1}{2} M |\vec{u}|^2) \).

Eq. (12), together with the distribution (11) of the \( \tau_j \)'s and the Gaussian distribution of the \( u_j \)'s, completely determines the distribution of \( \rho \) and hence of \( \Gamma \). We replace the integration over all \( \tau_j \)'s by the integration over all elements of an arbitrary real \( N \times \)
$N$ matrix $B$ such that the $\gamma_j$’s are eigenvalues of $(BB^\dagger)^{-1}$. The matrix $B$ has distribution \[ P(B) \propto \exp(-\pi \rho_0 V \text{tr} BB^\dagger)(\det BB^\dagger)^{(N+1)/2}. \] Using the dimensionless variable $x = \rho_0 = \Gamma_0 / \rho_0$ and properly rescaling $\vec{u}, B$, the integral for the distribution becomes \[ P(x) \propto \int d\vec{u} \int dB e^{-\text{tr} BB^\dagger - |\vec{u}|^2} \times \det(BB^\dagger)^{N+1} \delta(x - |B^{-1}\vec{u}|^2). \] (13)

We first compute the distribution of the vector $\vec{v} = B^{-1}\vec{u}$, which is given by Eq. (13) with the delta function replaced by $\delta(\vec{v} - B^{-1}\vec{u})$. The result is $P(\vec{v}) \propto (1 + |\vec{v}|^2)^{-N-1}$. Due to rotational invariance of the Gaussian distribution for $\vec{u}$, the distributions of $x$ and $|\vec{v}|^2$ are the same. Hence $P(x) = \int d\vec{v} P(\vec{v}) \delta(x - |\vec{v}|^2) \propto x^{N/2-1}(1 + x)^{-N-1}$. This is the result \[ \text{(a)} \] announced in the introduction and plotted in Fig. 1. It decreases monotonically for $N \leq 2$, and has a maximum at non-zero $\Gamma$ for larger $N$.

This calculation holds for the so-called orthogonal symmetry class (symmetry index $\beta = 1$), relevant for optical systems with time-reversal symmetry. The local density of states for systems with broken time-reversal symmetry (unitary class, $\beta = 2$) or with broken spin-rotational symmetry (symplectic class, $\beta = 4$) is relevant in condensed matter physics. We have repeated our calculations for $\beta = 2, 4$ and found $P_N^{(\beta)} = P_{\beta N}^{(1)}(x)$, with $P(1)(x)$ given by Eq. (1).

So far we have assumed that the hole in the cavity fully transmits at least one wavechannel, so that the transmittance $T$ of the hole (the ratio of the transmitted and incident power) is $\geq 1$. If the hole is smaller than a wavelength, then $T$ becomes $< 1$. The scattering matrix $S(T)$ of the cavity coupled by a hole with transmittance $T < 1$ can be expressed in terms of the scattering matrix $S|_{T=1}$,

\[ S(T) = \frac{S|_{T=1} + \sqrt{1-T}}{1 + S|_{T=1}\sqrt{1-T}}. \] (14)

To find the distribution of the local density of modes, we start from Eq. (8) with $S$ replaced by $S(T)$, repeat similar steps and average over $S|_{T=1} = e^{i\phi}$ at the end. The result is

\[ P(x) = \frac{2}{\pi^2 \sqrt{4T}} \int_0^\pi d\phi \times \frac{\sqrt{2 - T + 2\sqrt{1-T}\cos \phi}}{1 + x(2 - T + 2\sqrt{1-T}\cos \phi)/T^2}, \] (15)

plotted also in Fig. 1 (dashed line, for $T = 0.1$). It decreases monotonically for any $T < 1$.

The variance $\langle (\Gamma - \Gamma_0)^2 \rangle$ diverges if $N \leq 2$ but the divergency is removed when we take into account the condition of applicability of the Fermi golden rule \[ \text{(b)}. \] The perturbative treatment is valid as long as the decay rate $\Gamma$ of the excited atom remains smaller than the width $\gamma_\mu$ of the cavity modes contributing to the decay. Estimating the width of the main contributing mode as $1/\rho_0 = \Gamma_0 / \rho_0 V$, we get a condition $\Gamma \ll (\Gamma_0 / \rho_0 V)^{1/2}$. Therefore, any divergent contribution of the large-$\Gamma$ tail should be cutoff at $\Gamma \simeq (\Gamma_0 / \rho_0 V)^{1/2}$. The weight of the tail is negligibly small provided $1/\rho_0 V = \rho_0 V/9\pi \epsilon_0 \hbar c \ll 1$. To estimate this parameter, we write \[ \delta = \epsilon_0 a_B (a_B \text{ is the Bohr radius}), \] $\omega_0 = 2\epsilon c / \lambda_0$, $V = L^3$. Then $\Gamma_0 / \rho_0 V \simeq 3.21 \times 10^2 L^3 / \lambda_0$ is close to 1 for $z = 0.17$, $L = 0.53 \text{ mm}$, $\lambda_0 = 530 \text{ nm}$.

We can get large room for applicability of Eqs. (1), (15) by going to weaker (possibly magnetic) dipoles, smaller cavities, or larger (possibly microwave) wavelengths.

We conclude with a comparison with previous work on the local density of states in chaotic cavities \[ (1)\ldots. \] That work was motivated by different physical applications (Knight shift in NMR or optical absorption). Our application is in a sense dual to that of Ref. \[ (1) \], where complicated electronic states interact with simple radiation states. Instead, we have the simplest possible electronic system—a two level atom—and a complicated structure of radiation modes. In Refs. \[ (1)\ldots \] it was assumed that the cavity was coupled to the outside via a tunnel barrier of large area. In this case statistical fluctuations in the broadening of the levels $\gamma_\mu$ (from level to level and from cavity to cavity) can be ignored. In the case of a relatively small opening, considered here, fluctuations of the $\gamma_\mu$’s are essential. The resulting distribution \[ (a) \] of the local density of modes turns out to be very simple, compared with the result of Ref. \[ (1) \], (involving a five-fold integral in the case of unbroken time-reversal symmetry). We obtained our result within the framework of random-matrix theory. It would be interesting to see if it can be reproduced using the supersymmetry technique of Refs. \[ (1)\ldots \].

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