

Dynamics of the spontaneous emission of an atom into the photon-density-of-states gap: Solvable quantum-electrodynamical model

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We consider the process of spontaneous emission of a two-level atom embedded into a periodic layered dielectric structure. When the resonant atomic frequency lies within the photon-density-of-states gap, of width Γ , the atom emits light during a time interval $t\Gamma \ll 1$ until the spontaneous emission becomes fully inhibited. We discuss the enhancing of spontaneous emission by tuning the atomic resonance with a one-mode microcavity. In the $\lambda/4$ -shifted distributed feedback microcavities which possess the δ -function-like density of states, in the middle of the gap, the process of spontaneous emission has damped Rabi oscillation behavior; the damping depends on the width of the gap and the height of its walls. A profile of the density-of-states gap can be determined from the experimental data on the temporal behavior of the atomic spontaneous decay.

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The spontaneous emission is a fundamental and unavoidable consequence of the quantum nature of the interaction between an atom and an electromagnetic field [1]. The presence of a cavity [1,2] or reflecting walls [3] affects the process of spontaneous emission itself, namely, they may inhibit or enhance this process. Such phenomena have been observed in the atomic domain for very-high- Q cavities where by tuning a single-mode cavity on or off the atomic resonance, we respectively enhance or inhibit the spontaneous emission. It was also found that such atom-cavity coupling leads to a much more complex dynamics depending on the coherence of the cavity mode. This research area has also been extended to poorer cavities and is usually referred to as cavity quantum electrodynamics (CQED) [4,5].

We believe that Yablonovich was the first to suggest that if periodic dielectric structures have a photon band gap, then spontaneous emission should also be inhibited [6] by tuning the gap off the atomic frequency. He also proposed the use of such an effect to enhance the performance of quantum electronic devices. Recently, the improvement of semiconductor laser technology has allowed the creation of semiconductor microcavities [7,8] that are effective in the optical range of the spectrum, in contrast to the microwave cavities currently used by CQED researchers [9]. These optical microcavities (so-called λ cavities) consist of an active layer embedded into a periodic structure of alternating layers of different refractive indices. These structures can be employed to demonstrate the full quantum nature of the interaction of the cavity electromagnetic field with the active atoms in the optical region, and to allow the creation of a new generation of semiconductor lasers with extremely low threshold currents and possessing, perhaps, some micro-maser properties [10]. Such effects as the enhancing and

inhibition of spontaneous emission has been shown in these structures [7,8], but their dynamics has not been investigated in detail. Quantum-electrodynamical treatment of the behavior of an atom whose excited level lies near a photonic band edge was considered in Ref. [11]. It was reported that the excited atom experiences an anomalous Lamb shift and splits into a doublet [11]. In this paper we present a simple solvable model of the spontaneous emission dynamics of an atom embedded into a periodic dielectric structure, where the finite spectral width of the gap is accounted for.

The periodic layered structures are known to have a gap in the photon density of states (DOS) at frequencies which correspond to the total Bragg reflection [12]. The gap half-width is given by [12]

$$\Gamma = \omega_g (1/\pi)(\Delta n/n), \quad (1)$$

where $\omega_g = 0.5(\pi c/n_1 d_1) = 0.5(\pi c/n_2 d_2)$ is an angular frequency of the light corresponding to the center of the gap for the so-called quarter-wave stack, $n = (n_1 + n_2)/2$, $\Delta n = |n_1 - n_2|$, n_1 , n_2 , d_1 , d_2 are the refractive indices and the thicknesses of the alternating layers, respectively, and c is the velocity of light in a vacuum. For the usual parameters of the structures used in semiconductor microlasers (SML) $n \sim 3.5$, $\Delta n \sim 0.1$, $\omega_g \sim 10^{15} \text{ s}^{-1}$, and $\Gamma \sim 10^{12} \text{ s}^{-1}$. The light of frequency lying in the interval $(\omega_g - \Gamma, \omega_g + \Gamma)$ cannot propagate in the periodic structure under consideration. In other words, in this region of frequencies the photon DOS is null. This consideration is rigorous for one-dimensional consideration and can be used in the microlasers with metallic mirrors on the walls of the structure.

Let us consider the dynamic process of spontaneous emission decay of a two-level atom (TLA) of resonant fre-

quency ω_a embedded into the medium with an effective photon DOS $\rho(\omega)$. For simplicity, let the atom be in the upper state at $t=0$. In this case the temporal evolution of the probability amplitude $C(t)$ of finding the atom in the same upper state is governed by the following equation [1,13]:

$$\frac{\partial C}{\partial t} = -g^2 \int_{-\infty}^{\infty} \rho(\omega) d\omega \int_0^t e^{-i(\omega-\omega_a)(t-t')} C(t') dt', \quad (2)$$

where g is a matrix element of optical transition (i.e., $g = \langle 2|H'|1 \rangle$, where H' is the interaction Hamiltonian). We assume $\hbar=1$ in Eq. (2). A Laplace transform of Eq. (2) results in

$$s\bar{C} - 1 = ig^2 \bar{C} \int_{-\infty}^{\infty} \frac{\rho(\omega) d\omega}{\omega - \omega_a - is}, \quad (3)$$

where

$$\bar{C}(s) = \int_0^{\infty} e^{-st} C(t) dt \quad (4)$$

is the Laplace transform of the probability amplitude. The radiation decay rate is given by

$$R(t) = -\frac{d}{dt} \ln |C(t)|^2. \quad (5)$$

We start by considering the case of uniform DOS ($\rho=\rho_0$). Equation (3) has the solution

$$\bar{C}(s) = (s + \gamma/2)^{-1}, \quad (6)$$

where $\gamma = 2\pi g^2 \rho_0$. This corresponds to an atomic exponential decay of

$$|C(t)|^2 = \exp(-\gamma t), \quad (7)$$

where γ is the radiation decay rate of TLA in a uniform dielectric medium.

Now we consider the radiative decay process of the atom with resonant frequency lying at the center of the photonic gap that is expected to lead to full inhibited spontaneous emission. At first we choose the following approximation for the DOS function:

$$\rho = \rho_0 \left[1 - \frac{\Gamma^2}{(\omega - \omega_0)^2 + \Gamma^2} \right], \quad (8)$$

where ρ_0 is a constant. It corresponds to a Lorentzian-like gap in the uniform background value of DOS $\rho=\rho_0$ with the frequency of the gap center ω_0 . At the atomic resonance $\omega_a = \omega_0$, the DOS is null, $\rho=0$, and the effective gap width is 2Γ . For such a density of states Eq. (3) can be solved:

$$\bar{C}(s) = \frac{s + \Gamma}{s(s + \Gamma) + (\gamma/2)s}. \quad (9)$$

The inverse Laplace transform gives for the temporal dependence of the probability amplitude,

$$C(t) = C_{\infty} + (1 - C_{\infty}) \exp[-(\Gamma + \gamma/2)t], \quad (10)$$

with a decay rate given by

$$R(t) = \frac{\gamma}{1 + C_{\infty} \{ \exp[(\Gamma + \gamma/2)t] - 1 \}}, \quad (11)$$

where $C_{\infty} = \Gamma / (\Gamma + \gamma/2)$. Although the exponent in the expression (10) contains a factor $(\Gamma + \gamma/2)$, the atom decays at the beginning with a rate γ . Indeed, Eq. (11) is reduced to simple one: $R \approx \gamma$ at $t \ll t_0 = (\Gamma + \gamma/2)^{-1}$ as in the case of free space ($\rho=\rho_0$) [see Eq. (7)]. During this stage, the atom decays without “feeling” the photon DOS gap, and the specific time of this process is determined by the frequency bandwidth of the gap Γ , the wall height ρ_0 , and the matrix element g . This process corresponds to the emission of high-frequency photons with energy higher than the gap energy and agrees well with the Heisenberg uncertainty principle. The process having exponential behavior (7) has a Lorentzian spectrum of the width $\sim \gamma^{-1}$, therefore part of the wings of this spectrum will lie in the continuum region of the DOS, and this part of the spectrum will be emitted. At $t \gg t_0$, the radiation decay rate becomes null and we get

$$|C|^2 = |C_{\infty}|^2 = \left| \frac{\Gamma}{\Gamma + \gamma/2} \right|^2. \quad (12)$$

During this stage of the atomic evolution, the TLA “sees” the gap and the spontaneous emission is inhibited completely. It is clearly seen that the atom with the resonant frequency lying at the center of the gap has a nonzero probability to emit the light ultimately, but it is inhibited by the presence of the gap. The result obtained, Eq. (10), contradicts the results following from the Weisskopf-Wigner theory of the spontaneous emission [1,13], but it is in agreement with the spirit of inhibited spontaneous decay. According to the former theory the atom cannot emit any light in our case of $\rho(\omega_a)=0$ and our model predicts just a reduced one. If the $|C_{\infty}|^2$ value is close to unity, then $C(t)$ is close to its initial value. It means that the atom can actually emit the light into the photon DOS gap only in the case when the gap width is comparable with or less than the rate of spontaneous emission to the free space, in full agreement with CQED.

Detuning of the resonance frequency away from the center of the gap will spoil the inhibition of the spontaneous decay. In this case the density of states can be written as

$$\rho = \rho_0 \left[1 - \frac{\Gamma^2}{(\omega - \Delta_a - \omega_a)^2 + \Gamma^2} \right], \quad (13)$$

where Δ_a is detuning of the atomic line from the center of the gap. Using Eqs. (3) and (13), we obtain the Laplace transform $\bar{C}(s)$,

$$\bar{C} = \frac{s + \Gamma + i\Delta_a}{s^2 + s(\Gamma + \gamma/2 + i\Delta_a) + i\Delta_a\gamma/2}. \quad (14)$$

At small values of detuning $\Delta_a/\Gamma \ll 1$, the temporal behavior is determined by the expression

$$C(t) \approx C_{\infty} \exp[-i\Delta_a(1 - C_{\infty})t] + (1 - C_{\infty}) \exp[-(\Gamma + \gamma/2 + i\Delta_a C_{\infty})t]. \quad (15)$$

In this case we basically have the lossless free time evolution of the steady term, but a phase correction is introduced by the gap width through C_{∞} . Such terms in the

exponents of expression (15) result in an early oscillatory behavior of probability $|C(t)|^2$. However, at $t \rightarrow \infty$, we have $|C_\infty|^2$, as expected. For the opposite situation when $\Gamma + \gamma/2 \ll \Delta_a$, the tuning with the gap is poor but closer to the continuum, and we have

$$C(t) \approx \exp[-(\gamma/2 - ib\gamma)t], \quad (16)$$

where b is a coefficient of a magnitude of the order of unity. In this case the atom decays just like in free space, i.e., $|C(t)|^2 \approx \exp(-\gamma t)$.

We have to note that such a simple solution (10) corresponds to the case of no photons in the initial state. If the initial state consists of photons we should use another set of equations for the probability amplitudes [1]. This requires special consideration that is beyond the scope of this paper.

The semi-infinite periodic structures considered here are perfect reflectors with a unity reflectivity for frequencies lying in the gap of DOS. Therefore, structures consisting of distributed Bragg reflectors (DBR) as mirrors and the appropriate cavity length seem to be ideal for studying the process of the spontaneous decay and the coupling between an atom and a field in a lossless single-mode cavity [13]. It is known that in such a structure the TLA spontaneous emission has undamped oscillatory behavior that corresponds to the periodic exchange of energy between TLA and the single-mode electromagnetic field. Such an exchange is characterized by the so-called Rabi frequency in a strongly driven cavity [13] and by the zero-field Rabi flopping in QED empty cavities [14]. In the so-called $\lambda/4$ -shifted distributed feedback (DFB) structures there is an additional δ -function-like photon DOS inside the gap [15,16]. This δ function in the DOS means that there is a single mode of electromagnetic field supported by this cavity. Such structures are promising physical systems to investigate the localization of light and quantum electrodynamics of TLA in DFB single-mode microcavities [17]. In the simplest case when this δ -function level and atomic line lie exactly at the center of gap, the density of states can be written

$$\rho = \rho_1 \delta(\omega - \omega_a) + \rho_0 \left[1 - \frac{\Gamma^2}{(\omega - \omega_a)^2 + \Gamma^2} \right], \quad (17)$$

where ρ_1 is a constant. For this form of DOS, Eq. (4) has the solution

$$\bar{c} = \frac{s(s + \Gamma)}{s^3 + s^2(\Gamma + \gamma/2) + \kappa^2 s + \kappa^2 \Gamma}, \quad (18)$$

where $\kappa^2 = g^2 \rho_1$. Its role as a lossless single-mode cavity is clearly shown in the case when there are no walls of the gap and there is the only δ -function-like single-mode cavity DOS. In this case temporal solution is

$$C(t) = \cos(\kappa t), \quad (19)$$

the well-known result for the Rabi oscillation of TLA in the lossless cavity [13]. The CQED Rabi frequency is proportional to $N^{1/2}$, N the number of photons in the cavity, and as small as g for the zero-field Rabi frequency of an empty single-mode cavity.

There are three specific frequencies in the expression

(18), namely, γ , κ , and Γ . Therefore, let us introduce two parameters characterizing the problem: $\alpha = \gamma/2\Gamma$ and $z = \kappa/\Gamma$. Zero-field Rabi flopping (19) corresponds to $\alpha = 0$. The enhancing of the spontaneous decay by the QED coupling with a nearly empty cavity [14] can be seen by considering a good cavity (i.e., $z \ll 1$). Using Eq. (18), we obtain

$$C(t) = C_\infty \cos[\kappa t / (1 + \alpha)^{1/2}] + (1 - C_\infty) \exp[-(\Gamma + \gamma/2)t]. \quad (20)$$

Let us compare this expression with Eq. (10). We recognize the same decay behavior, and a lossless nutation of the steady value C_∞ in a continuous exchange of energy between the TLA and the cavity mode, just along the lines expected from CQED for enhanced spontaneous emission. At $\alpha \ll 1$ we have, according to Eq. (18),

$$C(t) = \left[1 - \frac{\alpha}{(1 + z^2)} \right] e^{-\gamma_1 t} \cos(\kappa_1 t) + \frac{\alpha z}{(1 + z^2)} e^{-\gamma_1 t} \sin(\kappa_1 t) + \frac{\alpha}{(1 + z^2)} \exp\{-[\Gamma + \gamma/2(1 + z^2)]t\}, \quad (21)$$

where $\gamma_1 = \gamma \kappa^2 / 4(\Gamma^2 + \kappa^2)$, $\kappa_1 = \kappa [1 - \gamma \Gamma / 4(\Gamma^2 + \kappa^2)]$. The terms discarded in Eqs. (19) and (20) are now clearly evident. The first two terms in this expression correspond to a CQED damped Rabi oscillation [18], where the oscillation damping parameter Γ is field dependent. Notwithstanding, we used the δ -function-like photon DOS, which corresponds to the lossless cavity; the process of spontaneous radiation is affected by such effective loss. The physical reason for this is as follows. The modulation sidebands of the atomic emission spectrum with frequencies $\omega \pm \kappa_1$ can leave the cavity because the photon DOS for them is not equal to zero. This spectrally selective effect has no atomic QED analog [14] and represents a difference that we intend to further analyze. On the other hand, the effective Rabi frequency change is mainly due to the gap width. The zero-field Rabi flopping ($\kappa \sim g$) becomes a quantum measure of the gap width by "noticing" that a lossless behavior is set if $g \ll \Gamma$, $z \ll 1$. The third term in Eq. (21) corresponds to the first stage of the process when the TLA does not feel the gap and therefore there is nonzero probability for the emitted photons to leave the cavity during this period, in a straightforward analogy of the filling time observed in the transient filtering of a Fabry-Pérot interferometer [19]. This is an effect of a new type of unavoidable transient loss in DFB cavities that however is much faster than the γ_1 rate, allowing for the observation of the Rabi oscillations.

The picture of TLA spontaneous emission in a perfect DFB cavity differs from the one in the single-mode lossless cavity usually considered in atomic CQED. The parameters determining this difference are the relative spontaneous rate $\alpha = \gamma/2\Gamma$ and the ratio of the Rabi frequency to the gap width $z = \kappa/\Gamma$. It is rather simple to estimate the z value for the typical experimental situations in

the following way. Bearing in mind that the photon DOS in the free semiconductor space is $\rho_0 = n^3 \omega^2 / \pi^2 c^3$ we have for the Rabi frequency $\kappa = (\rho_1 \gamma / 2 \pi \rho_0)^{1/2} \sim 4 \times 10^4 \text{ s}^{-1}$ for the free space decay rate $\Gamma \sim 10^9 \text{ s}^{-1}$. The corresponding optical power P inside the cavity can be estimated from the $N^{1/2}$ dependence of the Rabi frequency [1]. For $N = 100$ we have $P \sim 2 \text{ mW}$. Therefore the z and α values are much less than unity in the experimental situations and the difference between these two pictures is rather weak, although it takes place and can be enhanced by the Γ . We present in this paper the consideration of the temporal behavior of C only for two limiting cases $z \ll 1$ and $\alpha \ll 1$, where the expressions (21) and (20) have the same form. We shall discuss this behavior for arbitrary parameters z and α in a future paper.

For other types of DOS profiles, the specific computation implied by Eq. (3) for an arbitrary $\rho(\omega)$ is rather cumbersome and often impractical. There are a few solvable cases where they show a similar behavior as previously discussed. For the rectangular well

$$\rho = \rho_0 [\Theta(\omega - \omega_g - \Gamma) + \Theta(\omega_g - \omega - \Gamma)] \quad (22)$$

(Θ is the unit step function), it is possible to find the exact solutions $C(t)$ for the two limiting cases in the time domains: $(\Gamma + \gamma/2)t \ll 1$ and $(\Gamma + \gamma/2)t \gg 1$. In the former case the atom decays again exponentially without feeling the gap and we have expression (7) for the $|C(t)|^2$. In the latter case

$$C(t) = \text{const} = \frac{\pi \Gamma / 2}{\pi \Gamma / 2 + \gamma / 2} . \quad (23)$$

In addition, it is possible to show that for this rectangular gap form the approximate solution of Eq. (3) will be

$$C(t) = \frac{\pi \Gamma / 2}{\pi \Gamma / 2 + \gamma / 2} + \frac{\gamma / 2}{\pi \Gamma / 2 + \gamma / 2} \exp[-(\pi \Gamma / 2 + \gamma / 2)t] . \quad (24)$$

In this case the width of the gap is effectively larger $\pi/2$ times in comparison to the Lorentzian-like gap.

Therefore, it is important to emphasize that the process of light emission by the atom into the gap of the photon DOS depends mainly on the width of the gap and the height of its walls and slightly on the profile itself. Such a behavior is expected for the other type of gap as well, and that the spontaneous decay process will be described with similar expressions to the ones given above, Eq. (11), with some smooth function of Γ instead of just Γ in the case of Lorentzian-like well, Eq. (11). Such expected behavior allows for approximations that would simplify the analytical approach.

A rather more important related problem is given by the inverse problem of determining such parameters as the gap width and depth (wall height) and its profile from the experimental data. This is possible by obtaining the information on the temporal behavior of spontaneous emission. Indeed, we can change the integration order in the expression (3) and rewrite this equation, for simplicity

on resonance, in the form

$$\frac{\partial C}{\partial t} = -g^2 \int_0^t \rho(t-\tau) C(\tau) d\tau , \quad (25)$$

where $\rho(t) = \int_{-\infty}^{\infty} \rho(\omega) e^{-i\omega t} d\omega$ is the response function of the structure. By representing the DOS function $\rho(\omega)$ in the form $\rho(\omega) = \rho_0 [1 - \delta\rho(\omega)]$, where $\delta\rho(\omega)$ is the gap profile, then the Laplace transform of Eq. (28) will give the following expression for the Laplace transform of the response function $\overline{\delta\rho}(s)$:

$$\overline{\delta\rho}(s) = \frac{2\pi}{\gamma \{s + \gamma/2 - [\overline{C}(s)]^{-1}\}} . \quad (26)$$

Therefore, by knowing the experimental $C(t)$ behavior it is possible to get $\overline{\delta\rho}(s)$, and after subsequent inverse Laplace and Fourier transforms to obtain the $\delta\rho(\omega)$ spectrum, ultimately determining the gap profile.

In conclusion, we have considered in frames of simple exactly solvable models the dynamical process of spontaneous emission of the two-level atom embedded into periodic layered dielectric structures. We obtain explicit expressions for the behavior of inhibited and enhanced spontaneous emission and their dependence on the gap characteristics, especially on its width. We also draw the appropriate comparisons with its atomic CQED analogs. When a resonant frequency of the atom lies at the center of the gap of the photon density of states, it emits light during the time period $t < t_0$, where $t_0 \sim 1/\Gamma$ (Γ is the gap width) until reaching the expected inhibited spontaneous emission. The enhancement and strongly driven single-mode cavities are studied in the $\lambda/4$ -shifted DBR microcavities which possess the δ -function-like density of states in the middle of the gap. The process of spontaneous emission has damped Rabi oscillation behavior. This damping depends on the width of the gap and the height of its walls and exhibits the spectral coupling of the cavity and atomic dynamics. Results we obtained are valid to some extent for any type of structure having the gap in the photon density of states. Therefore, the results are not restricted only by the case of the one-dimensional problem. Open cavities typical for all type lasers do not usually have the gap in the photon DOS, although DOS may have some features. Spontaneous emission in open resonators is the item of special consideration that is beyond the scope of this paper. We have also shown that the profile of the DOS gap can be determined from the experimental data on the temporal behavior of the atom spontaneous decay.

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