

## Spontaneous emission in a photonic crystal near the band edge: Field versus population dynamics

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We investigate the dynamical properties of the radiation field emitted from an excited two-level atom in a photonic crystal. If the transition frequency of the atom lies within a certain frequency range above the band edge, the emitted field consists of two components that show a different decay dynamics. In particular it is shown that one field component decreases faster than the atomic population with a decay constant depending on the distance from the atom. As a consequence, the decay rate of the electromagnetic field is spatially varying and, in general, can not be identified with the corresponding rate for the atomic population.

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Spontaneous emission results from the coupling of an atom or molecule to the vacuum of the electromagnetic field, and is determined by the local density of modes (LDM). Altering the mode structure, as, e.g., near reflecting surfaces or inside resonators, one can modify the emission dynamics and the Lamb shift [1]. Substantial deviations from the free-space LDM can be achieved in photonic crystals, which are artificially created periodic dielectric structures. For sufficiently large modulations of the dielectric constant these structures can possess gaps in the frequency spectrum [2]. It has been predicted that inside or in the vicinity of such gaps spontaneous emission can be reduced or even completely suppressed [2–5] and excited photon-atom bound states can exist [3–6]. Due to the pronounced frequency dependence of the LDM the spontaneous emission has, in general, a nonexponential character [3,7].

In contrast to the large number of theoretical papers on the subject only little experimental work has been published [8–10]. Due to the difficulty in fabricating three-dimensional photonic crystals with a pronounced band gap, unambiguous experimental proofs of several aspects of the atom-light interaction are still missing. In a recent paper, Petrov *et al.* reported the observation of a two-component fluorescence of dye molecules in a photonic crystal [8]. The decay rate of one of them was substantially reduced compared to the free-space value, that of the other was however enhanced. On average, the decay was changed just by a few per cent. On the other hand, a similar later experiment by Megens *et al.* [9] using a smaller wavelength did show only a single-component decay with a rate very close to the free-space value. The origin of the results obtained in Ref. [8] is still subject of some discussion [11,12]. To understand the different experimental results, an efficient formalism for the description of fluorescence from active materials in a photonic crystal was developed in Ref. [13], and accelerated and inhibited decay rates of atomic population predicted depending on the position and orientation of the dipole in the photonic crystal [14]. The purpose of the present paper is to show that an atom in a photonic crystal can emit radiation showing both accelerated and decelerated decays as compared to the atomic population. We show, furthermore, that the temporal behavior of the emitted field depends sensitively on the de-

tuning of the atomic transition frequency from the band edge and changes with the distance from the atom.

Let us consider a two-level atom in a photonic crystal. The excited state  $|1\rangle$  of the atom is radiatively coupled to the ground state  $|0\rangle$ , and the transition frequency  $\omega_0$  is assumed to be near the band edge  $\omega_c$ . The atom-field interaction is described in dipole and rotating-wave approximation by the Hamiltonian

$$\hat{H} = \hbar\omega_0|1\rangle\langle 1| + \sum_{\mathbf{k}} \hbar\omega_{\mathbf{k}}b_{\mathbf{k}}^{\dagger}b_{\mathbf{k}} + i\hbar \sum_{\mathbf{k}} g_{\mathbf{k}}(b_{\mathbf{k}}^{\dagger}|0\rangle\langle 1| - \text{H.c.}).$$

Here  $b_{\mathbf{k}}$  ( $b_{\mathbf{k}}^{\dagger}$ ) is the annihilation (creation) operator for the  $k$ th radiation mode in the photonic crystal with frequency  $\omega_{\mathbf{k}}$ .  $g_{\mathbf{k}} = \omega_0/\hbar \sqrt{(\hbar/2\epsilon_0\omega_{\mathbf{k}}V_0)}\mathbf{e}_{\mathbf{k}} \cdot \mathbf{d}$  is the atomic field coupling constant [3], where  $\mathbf{d}$  is the vector of the atomic dipole moment, and  $\mathbf{e}_{\mathbf{k}}$  are the polarization unit vectors. The state vector of the system is given by  $|\psi(t)\rangle = \alpha(t)e^{-i\omega_0 t}|1, \{0\}\rangle + \sum_{\mathbf{k}} \beta_{\mathbf{k}}(t)e^{-i\omega_{\mathbf{k}} t}|0, \{1_{\mathbf{k}}\}\rangle$ , with the atom initially in the excited state.  $|1, \{0\}\rangle$  describes the atom in the excited state with no photon in the reservoir modes, and  $|0, \{1_{\mathbf{k}}\}\rangle$  describes the atom in the ground state and a single photon in mode  $\mathbf{k}$ . The dispersion relation near one band edge can be expressed approximately by  $\omega_{\mathbf{k}} = \omega_c + A|\mathbf{k} - \mathbf{k}_n|^2$ , where  $A$  is a system dependent constant,  $\mathbf{k}_n$  is a finite collection of symmetry related points associated with the band edge. Solving the Schrödinger equation for amplitudes  $\alpha(t)$  and  $\beta_{\mathbf{k}}(t)$  and substituting the result into the expression for the field, one can easily obtain the field amplitude [15]. As shown in Ref. [6] the resulting radiation field consists of several components depending on the detuning  $\Delta = \omega_0 - \omega_c$  of the transition frequency  $\omega_0$ .

*Region I.* If  $\Delta \leq \Delta_1 = \beta^{3/2}/\omega_c^{1/2}$ , the electric field has two components  $\mathbf{E}^{(1)} = \mathbf{E}_l^{(1)} + \mathbf{E}_d^{(1)}$ . Here  $\beta^{3/2} = (\omega_0 d)^2 \sum_n \sin^2 \theta_n / (8\pi\epsilon_0 \hbar A^{3/2})$ , and  $\theta_n$  is the angle between the dipole vector  $\mathbf{d}$  of the atom and  $\mathbf{k}_n$ .

$$\mathbf{E}_l^{(1)}(\mathbf{r}, t) = \mathbf{E}_0(\mathbf{r}) \frac{-i\pi}{F'(x_1)} e^{-i(\omega_0 - x_1)t - rP} \Theta(t - t_1), \quad (1)$$

$$\mathbf{E}_d^{(I)}(\mathbf{r}, t) = \mathbf{E}_0(\mathbf{r}) e^{i\phi} \int_{-\infty}^{\infty} dy \left\{ \frac{J(i(\Delta - x_1), y, t)}{iF'(x_1)} + \frac{\sqrt{i}}{\pi} \int_0^{\infty} dx K(x) J(x, y, t) \right\}. \quad (2)$$

$x_1$  is the real root of  $F(x) = x - \beta^{3/2}/(\sqrt{\omega_c} + \sqrt{x - \Delta}) = 0$  in the region  $\text{Re}(x) > \Delta$ ,  $P = \sqrt{(x_1 - \Delta)/A}$  and  $t_1 = r/(2AP)$ .  $F'(x)$  denotes the derivative of  $F(x)$ .  $\mathbf{E}_0(\mathbf{r}) \equiv (\omega_0/8irA\pi^2\epsilon_0)\sum_n e^{i\mathbf{k}_n \cdot \mathbf{r}} (\mathbf{d} - [\mathbf{k}_n(\mathbf{k}_n \cdot \mathbf{d})]/(\mathbf{k}_n^2))$ .  $\mathbf{E}_d^{(I)}$  is a stationary (i.e., nondecaying) component with a spatial localization length  $P^{-1}$ . The stationary field component reflects the formation of a bound atom-field state as predicted in Refs. [3–6]. The frequency of the localized field is  $\omega^{(I)} = \omega_0 - x_1$ , which is within band gap. The second field component  $\mathbf{E}_d^{(I)}$  has a diffusive character. The functions  $J(x, y, t)$  and  $K(x)$  in Eq. (2) are defined as  $J(x, y, t) = [i^{3/2}y + r/(2\sqrt{At})]e^{-y^2/(-xt + i(i^{3/2}y + r/(2\sqrt{At}))^2)}$  and  $K(x) = \beta^{3/2}\sqrt{x}(\omega_c - ix)/[(\Delta + ix)(\omega_c - ix) - \sqrt{\omega_c}\beta^{3/2}]^2 - i\beta^3x$  and  $\phi = -\omega_c t + r^2/(4At) + 3\pi/4$ . The decay of  $\mathbf{E}_d^{(I)}$  follows a power-law dependence on time and no unique decay rate can be associated with it. The field behavior is reflected in the dynamics of the atomic population. The amplitude  $\alpha(t)$  of the excited state in region I reads

$$\alpha^{(I)}(t) = \frac{e^{ix_1 t}}{iF'(x_1)} - \frac{\sqrt{i}}{\pi} \int_0^{\infty} dx K(x) e^{i\Delta t - xt}. \quad (3)$$

*Region II.* If  $\Delta_1 \leq \Delta \leq \Delta_2 = \omega_c - \beta^{3/2}/[\frac{2}{3}\omega_c^{1/2} + (q_1 - q_2)^{1/3} - (q_1 + q_2)^{1/2}]$ , with  $q_1 = [(4\omega_c^3 - 20\omega_c^{3/2}\beta^{3/2})/27 + \beta^3]^{1/2}$  and  $q_2 = 10\omega_c^{3/2}/27 - \beta^{3/2}$ , the field has only a diffusive component  $\mathbf{E}^{(II)} = \mathbf{E}_d^{(II)}$ :

$$\mathbf{E}_d^{(II)}(\mathbf{r}, t) = \mathbf{E}_0(\mathbf{r}) e^{i\phi} \frac{\sqrt{i}}{\pi} \int_{-\infty}^{\infty} dy \int_0^{\infty} dx K(x) J(x, y, t). \quad (4)$$

In region II the Lamb shift moves all dresses frequencies exactly to the band edge,  $\omega^{(II)} \equiv \omega_c$ . The dynamics of the excited state population is again given by Eq. (3), however, without the first term.

*Region III.* Finally if  $\Delta \geq \Delta_2$ , the emitted field has two components  $\mathbf{E}^{(III)} = \mathbf{E}_d^{(III)} + \mathbf{E}_r^{(III)}$ .  $E_r$  has radiative character and is given by

$$\mathbf{E}_r^{(III)}(\mathbf{r}, t) = \mathbf{E}_0(\mathbf{r}) \frac{\pi}{G'(x_2)} e^{-i(\omega_1 + ix_2)t + irQ} \Theta(t - t_2). \quad (5)$$

Here  $x_2$  is the complex root of  $G(x) = x - i\beta^{3/2}/(\sqrt{\omega_c} - i\sqrt{ix + \Delta}) = 0$  in the region  $[\text{Re}(x) < 0 \text{ and } \text{Im}(x) < \Delta]$ . The imaginary part of  $x_2$  represents the modified Lamb shift, its real part characterizes the decay rate of the radiative field component,  $\omega^{(III)} = \omega_0 - \text{Im}[x_2]$ , and  $\Gamma_r = -\text{Re}[x_2]$ . The radiative component has a retardation time  $t_2 = r/[2A(\text{Im} + \text{Re})Q]$  with  $Q = \sqrt{(ix_2 + \Delta)/A}$ . The diffusion field has the form

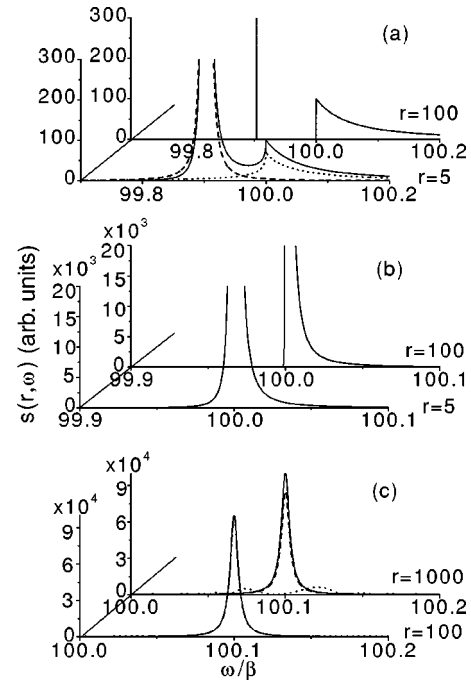


FIG. 1. Spontaneous emission spectrum for different distances from the atom and different detunings from band edge (top)  $\Delta = 0$ , i.e., region I, (middle)  $\Delta = 0.100002\beta$  region II, and (bottom)  $\Delta = 0.2\beta$ , i.e., region III near the band edge. The solid line represents the spectrum of the total field, the dashed and dotted lines show the spectrum of the individual components.  $r$  is measured in units of  $(\beta A)^{-1/2}$ . See text for details.

$$\mathbf{E}_d^{(III)}(\mathbf{r}, t) = \mathbf{E}_0(\mathbf{r}) e^{i\phi} \int_{-\infty}^{\infty} dy \left\{ \frac{J(i(\Delta - x_2), y, t)}{G'(x_2)} + \frac{\sqrt{i}}{\pi} \int_0^{\infty} dx K(x) J(x, y, t) \right\}. \quad (6)$$

Here the first term in the brackets is the dominant one. The complex amplitude of the excited state reads

$$\alpha^{(III)}(t) = \frac{e^{ix_2 t}}{G'(x_2)} - \frac{\sqrt{i}}{\pi} \int_0^{\infty} dx K(x) e^{i\Delta t - xt}. \quad (7)$$

The first term describes exponential decay with rate  $\Gamma_r$ , the second term describes a nonexponential decay, which is however only of relevance for very short times.

We first discuss the properties of the spontaneously emitted radiation when the atomic transition lies in one of the three frequency regions and compare the field dynamics with that of the upper state population. For this we first consider the spectrum of the electromagnetic field at a particular distance  $r$  from the atom, defined via the Fourier transform of the electric field

$$S(\mathbf{r}, \omega) = \left| \frac{1}{2\pi} \int_0^{\infty} dt \mathbf{E}(\mathbf{r}, t) e^{i\omega t} \right|^2. \quad (8)$$

In Fig. 1 we have plotted  $s(r, \omega) = S(\mathbf{r}, \omega)/|\mathbf{E}_0(\mathbf{r})|^2$  for dif-

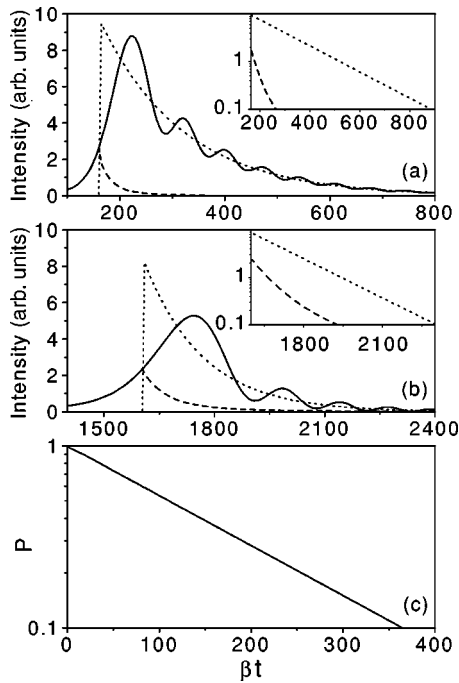


FIG. 2. Time evolution of intensity of total field (solid line), propagating (dotted line), and diffusion field (dashed line) for  $\omega_c = 100\beta$ ,  $\Delta = 0.2\beta$  and for different distance from atom  $r$  in units of  $(\beta A)^{-1/2}$ . (a)  $r=100$ ; (b)  $r=1000$ . The inset shows the same on a logarithmic scale. In the lowest graph the population decay is plotted, which is clearly exponential.

ferent values of  $r$  for the cases of  $\Delta \leq \Delta_1$ , i.e., in region I (top part),  $\Delta_1 \leq \Delta \leq \Delta_2$ , i.e., in region II (middle part), and  $\Delta \geq \Delta_2$ , region III (lower part). In region I one recognizes a singularity in the spectrum at the dressed transition frequency associated with the localized electric field. The wings of this line decrease rapidly with increasing distance  $r$  from the atom, which reflects the exponential localization of this field contribution, see Eq. (1). There is also a spectral contribution above the band edge corresponding to the diffusion field, which does not depend on the distance from the atom. The dashed line shows the spectrum of the localized field, the dotted line shows that of the diffusion field and the solid line the spectrum of the total field.

In region II there is only a diffusive contribution. No localized field exists and one peak in the spectrum occurs at  $\omega_c$ . The emission spectrum narrows as the distance from the atom increases. Finally in region III the spectrum is a narrow Lorentzian line at the dressed atomic frequency. Here dashed and dotted lines represent the spectra of the radiative and diffusive fields only and the full line represents the spectrum of the total field. Since the frequency lies in the propagation band, the entire energy of the emitted field can propagate out. Furthermore, the total emission spectrum does not depend on the distance from the atom. The spectrum of the radiative component is a narrow Lorentzian line, which has the same half width with the total spectrum. From Fig. 1 it can be seen that the peak of the radiative component decreases with the distance  $r$  from the atom. However, the part of the diffusive component increases with  $r$ . The spectrum of the diffusive

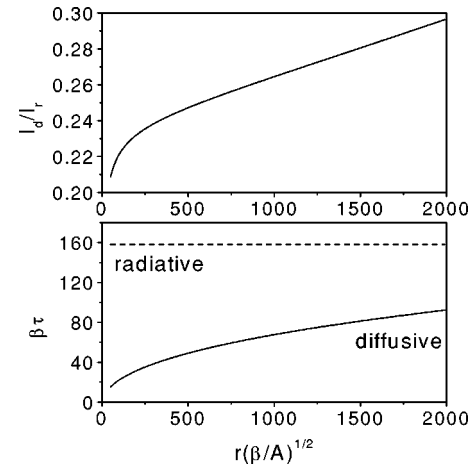


FIG. 3. Spatial dependence of relative intensity of diffusive to radiative field (upper curve) and decay times (lower curve). The solid line corresponds to the diffusive field component; the dashed line to the radiative.

component has several maxima with a lower cutoff at the band edge. The position of its most dominant peak changes with the distance from the atom and comes closer to the frequency of the radiative component.

We now analyze the dynamics of the intensity of the field in region III, where both diffusive and radiative components contribute to the transport of energy away from the atom. If the transition frequency lies deep in the propagation band, the diffusion field is negligible. However, if the detuning from the band edge is not too large, both field components need to be taken into account. In Fig. 2 we have plotted the total field intensity as well as the intensities of the diffusive and radiative parts as functions of time for two different distances from the atom. After the retardation time of the radiative field  $t_2$ , the intensity of this component decreases exponentially with a single decay constant  $\Gamma_r$ . The decay of the diffusion field, on the other hand, resembles a power-law dependence. If, as in the example shown, the magnitudes of the diffusive and radiative parts are comparable, the total intensity shows oscillations due to interference of the two components.

Another interesting feature of the decay of the diffusion field is that it slows down with increasing distance from the atom. This is illustrated in Fig. 3, where we have shown the relative intensities of the diffusive and radiative field components as well as the decay times as functions of the distance from the atom. For small values of  $r$  the initial decay of the diffusion field is much faster than that of the radiative component. With growing distance from the atom, the diffusion decay approaches, however, more and more that of the radiative component. At the same time the intensity of the diffusive field increases relative to the radiative component. This can be interpreted as a partial transfer of energy from the radiative field component to the diffusive field component. In fact, one finds that Eq. (5) for  $\mathbf{E}_r^{(III)}$  can be rewritten in the form

$$\begin{aligned} \mathbf{E}_r^{(\text{III})}(\mathbf{r}, t) = & \frac{\pi \mathbf{E}_0(\mathbf{r})}{G'(x_2)} \Theta\left(t - \frac{r}{v_f}\right) \\ & \times \exp\left\{-i[\omega - \text{Im}(x_2)]\left(t - \frac{r}{v_p}\right)\right. \\ & \left.+ \text{Re}(x_2)\left(t - \frac{r}{v_e}\right)\right\} \end{aligned} \quad (9)$$

with  $v_p = [\omega_1 - \text{Im}(x_2)]/\text{Re}(Q)$ , the energy velocity  $v_e = \text{Re}(x_2)/\text{Im}(Q)$ , and the front velocity  $v_f = 2A(\text{Re} + \text{Im})(Q)$ . It can be proven analytically that the energy velocity is larger than the front velocity  $v_e > v_f$ . This means, during the propagation of the radiative field component, its energy is partly transferred into the diffusion field.

Finally we discuss how the presence of different decay times associated with the diffusive and radiative field components reflects itself in the dynamics of the atomic population. As can be seen from the lower plot in Fig. 2, the excited state amplitude of an atom in region III decays after some initial time according to an exponential law with a decay rate equal to that of the radiative field  $\Gamma_r$ . The exponential decay of  $\alpha^{(\text{III})}(t)$  is described by the first term in Eq. (7), which is dominant after some initial time period. Thus we conclude that the coexistence of field components with different char-

acteristic decay times does not correspond to a similar behavior of the atomic population.

In summary, we have investigated the behavior of the radiation field emitted by a two-level atom in a photonic crystal. The emission spectrum and decay properties strongly depend on the distance from the atom and the position of the atomic transition frequency relative to the band edge. Under certain conditions, namely, if the bare atomic transition frequency is in region III not too far from the band edge, there can be two field components with different characteristic decay times and spectra. As a consequence the field intensity shows an oscillatory decay. The spectrum of the total field, as defined in Eq. (8), is however Lorentzian and the atomic population decays always exponential with a single decay constant. The detection of field components with decelerated and accelerated decays in Ref. [8] does, however, not necessarily imply a modified spontaneous decay of the atomic excitation.

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