Decay Kinetic Properties of Atoms in Photonic Crystals with Absolute Gaps

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Decay kinetic properties of a two-level atom near the band edges of photonic crystals (PCs) with absolute gaps are studied based on the Green's function expression for the evolution operator. The local coupling strength between the photons and an atom is evaluated by an exact numerical method. It is found that the decay behavior of an excited atom can be fundamentally changed by the variation of the atomic position: Weisskopf-Wigner and non-Weisskopf-Wigner decay phenomena occur at different atomic positions in the PCs as a result of a significant difference in the local coupling strength. Our finding implies that it is possible to engineer the luminescence spectrum by controlling the atomic position.

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Since the pioneering works of Yablonovitch and John [1], there has been a growing interest in both fabrication of photonic band gap (PBG) structures [2–5] and study of quantum electrodynamic (QED) behavior of atoms in photonic crystals (PCs) [6–14]. In the past 13 years, the isotropic and anisotropic dispersion models have been extensively employed to solve the QED problems in PCs. These two models predicted many novel quantum optics characteristics [6–13]. On the other hand, when the position-dependent interaction between photons and an atom is taken into account, the local density of states (LDOS) is more decisive for emission behavior of atoms in inhomogeneous media [15–19]. Very recently, based upon slow variation and nonsingularity near the band edges in both the DOS and LDOS of 3D PCs with absolute PBGs, Li *et al.* concluded that the Weisskopf-Wigner approximation (WWA) is universal for spontaneous emissions in 3D PCs [19]. This conclusion intimates that the predictions from the two dispersion models should be denied.

The study reported in this Letter has been inspired by the above-mentioned controversial results. We employed the Green function formalism of the evolution operator to investigate the dynamic decay of a two-level atom in 3D PCs with absolute PBGs. The local coupling strength between photons and an atom was evaluated by an exact numerical method. It is found that when the transition frequency ω_0 of a bare atom is inside a PBG and near the band edges, the population of the excited state exhibits envelope-damped Rabi oscillations and is rapidly trapped into a fractionalized steady state for these atomic positions with strong atom-photon interactions, while these phenomena disappear and the exponential decay law dominates the behavior of an excited atom for those atomic positions with weak atom-photon interactions. Similarly, when ω_0 is outside a PBG and near the band edges, the different positions of an atom can also lead to exponential or nonexponential decay of the excited state. Our results show that the variation of the atomic position can fundamentally change the atomic decay behavior. This sensitive position-dependent characteristic of the luminescence should lead to important applications not only in the PC field but also in other fields, such as single molecule spectroscopy [20].

We focus on the light emissions of an atom with two levels (denoted as $|1\rangle$ and $|2\rangle$) in perfect PCs without defects. When the atom is located at position $\mathbf{r}$ in a perfect PC, the Hamiltonian of the system in the rotating wave approach reads [13,14,18,19]

$$H = \hbar\omega_0 b_2^+ b_2 + \hbar\sum_{n\mathbf{k}} \omega_{n\mathbf{k}} a_{n\mathbf{k}}^+ a_{n\mathbf{k}} + \hbar\sum_{n\mathbf{k}} [g_{n\mathbf{k}}(\mathbf{r}) b_1^+ b_2 a_{n\mathbf{k}}^+ + g_{n\mathbf{k}}^*(\mathbf{r}) b_2^+ b_1 a_{n\mathbf{k}}], \tag{1}$$

where $b_{i=1,2}$ and $b_{i=1,2}^+$ ($a_{n\mathbf{k}}$ and $a_{n\mathbf{k}}^+$) are, respectively, the electronic (photonic) annihilation and creation operators, ω_0 is the transition frequency of a bare atom, $\omega_{n\mathbf{k}}$ is the frequency of the eigenmode of the electromagnetic field, $\mathbf{E}_{n\mathbf{k}}(\mathbf{r}) = ic\nabla \times \mathbf{H}_{n\mathbf{k}}(\mathbf{r})/[\epsilon(\mathbf{r})\omega_{n\mathbf{k}}]$ which can be obtained using the plane-wave expansion method [21], c is the speed of light in vacuum, $\epsilon(\mathbf{r})$ is the dielectric function in the PC, and the coupling coefficient $g_{n\mathbf{k}}(\mathbf{r})$ is given by

$$g_{n\mathbf{k}}(\mathbf{r}) = i\omega_0(2\epsilon_0\hbar\omega_{n\mathbf{k}}V)^{-1/2}\mathbf{E}_{n\mathbf{k}}(\mathbf{r}) \cdot \mathbf{u}_d, \tag{2}$$

where ϵ_0 is the permittivity of vacuum, V is the volume of the PC, and $\mathbf{u}_d$ is the transition dipole moment between the two atomic levels. In Eq. (1), the first two terms represent the noninteraction Hamiltonian H_0, and the third term is the interaction Hamiltonian H_I.

We denote $|I\rangle = |2,0\rangle$ and $|F_{nk}\rangle = |1, n\mathbf{k}\rangle$ as the initial and final states of the system. The state vector of the system evolves as $|\Psi(t)\rangle = C_e(t)|I\rangle + \sum_{nk} C_{nk}(t)|F_{nk}\rangle \equiv U(t)|I\rangle$ with initial conditions $C_e(0) = 1$ and $C_{nk}(0) = 0$. Here $U(t)$ is the evolution operator and $C_e(t) \equiv \langle I|U(t)|I\rangle$. From the Green function expression of $U(t)$, one can show [22]

$$C_e(t) = \frac{1}{2\pi i}\int_{-\infty}^{\infty} d\omega e^{-i\omega t}[G_{ii}^{-}(\omega) - G_{ii}^{+}(\omega)], \tag{3}$$

where $G_{ii}^{\pm}(\omega) = \lim_{\eta\to 0_+}\langle I \mid G(z = \omega \pm i\eta) \mid I\rangle$ with the resolvent $G(z) = (z - H/\hbar)^{-1}$. Note that $\langle F_{n\mathbf{k}} \mid H_I \mid I\rangle = g_{n\mathbf{k}}(\mathbf{r})$, $\langle F_{n'\mathbf{k}'} \mid H_I \mid F_{n\mathbf{k}}\rangle = 0$, and $\langle I \mid H_I \mid I\rangle = 0$. From the operator identity $(z - H_0/\hbar)G(z) = 1 + H_I G(z)/\hbar$ [23], we can analytically obtain

$$G_{ii}^{\pm}(\omega) = \frac{1}{\omega - \omega_0 - \Delta(\mathbf{r},\omega) \pm i[\Gamma(\mathbf{r},\omega)/2 + \eta]}, \tag{4}$$

with

$$\Gamma(\mathbf{r},\omega) = \frac{\alpha_0 c^3}{8\pi}\sum_n \int_{BZ} d\mathbf{k}\,\frac{\mid \mathbf{E}_{n\mathbf{k}}(\mathbf{r})\mid^2}{\omega_{n\mathbf{k}}}\,\delta(\omega - \omega_{n\mathbf{k}}), \tag{5}$$

$$\Delta(\mathbf{r},\omega) = \frac{1}{2\pi}\mathcal{P}\int_0^{\infty}\frac{\Gamma(\mathbf{r},\omega')}{\omega - \omega'}d\omega', \tag{6}$$

where $\alpha_0 = \omega_0^2 u_d^2/3\pi\epsilon_0\hbar c^3$ is the relative radiation linewidth of the atom in vacuum. In Eq. (5), the random orientation of $\mathbf{u}_d$ is considered. Inserting Eq. (4) into Eq. (3), we have

$$C_e(t) = \int_{-\infty}^{\infty} d\omega C_e(\mathbf{r},\omega)e^{-i\omega t}, \tag{7}$$

with

$$C_e(\mathbf{r},\omega) = \frac{1}{\pi}\lim_{\eta\to 0^+}\frac{\Gamma(\mathbf{r},\omega)/2 + \eta}{[\omega - \omega_0 - \Delta(\mathbf{r},\omega)]^2 + [\Gamma(\mathbf{r},\omega)/2 + \eta]^2}. \tag{8}$$

Thus, we see that $\Gamma(\mathbf{r},\omega)$ and $\Delta(\mathbf{r},\omega)$ represent the local coupling strength (LCS) and the level shift, respectively. When the mass renormalization contribution is taken into account, $\Delta(\mathbf{r},\omega)$ should be replaced by $[\Delta(\mathbf{r},\omega) - \Delta(\mathbf{r},0)]$ which is the Lamb shift [13,14,24].

Equation (8) clearly shows that the evolution spectrum of the upper level is of a non-Lorentzian shape. However, for a homogeneous medium, it can be approximately reduced to a Lorentzian spectrum. In this case, Eq. (5) leads to the relative local coupling strength (RLCS), $\alpha_f(\mathbf{r},\omega) \equiv \Gamma_f(\mathbf{r},\omega)/\omega_0 = 9\epsilon^{5/2}\alpha_0\omega/(2\epsilon + 1)^2\omega_0$, as given in Ref. [25]. In the visible regime, the magnitude of α_0 is of the order of 10^{-7}–10^{-6} for low excited states of atoms or molecules, 10^{-5} for high excited states, and at most 10^{-4}–10^{-3} for vibrational states of molecules, excitons, and shallow impurity states in semiconductors. Because the RLCS in a homogeneous medium is very small near ω_0, one can make a reasonable approximation to Eq. (8) by setting $\omega \simeq \omega_0$ in $\Gamma(\mathbf{r},\omega)$ and $\Delta(\mathbf{r},\omega)$. This leads to the famous exponential decay law (i.e., the WWA) with the radiative linewidth and Lamb shift given by $\Gamma_f(\mathbf{r},\omega_0)$ and $\Delta_f(\mathbf{r},\omega_0)$, respectively. Below, we show and explain that the WWA is in general not valid for spontaneous emissions of an atom in PCs with absolute PBGs.

The calculation of $\Gamma(\mathbf{r},\omega)$ in Eq. (5) involves an integral of the electromagnetic (EM) fields in the first Brillouin zone (FBZ). This is a time-consuming task. Thus, such an integral was often performed within an irreducible BZ [19,26], based on a linear tetrahedron method [27] and the belief that the eigenvectors of the EM field are invariant under the lattice point group operation. However, it has been shown that the latter belief is incorrect [28]. In the calculations presented in this work, a recently developed, exact numerical method [28] for the evaluation of integrals of vectorial fields in PCs was employed. In addition, a frequency cutoff $\omega_c \simeq 3\omega_0$ was made, which ensures the relative accuracy better than 10^{-3} for $\mid C_e(t)\mid^2$.

We carried out the calculations for two PC structures: (i) the diamond structure [21,29] consisting of dielectric spheres of the refractive index $n = 3.6$ in the air background with a filling fraction $f = 0.31$, whose two absolute PBGs span from $0.738(2\pi c/a)$ to $0.776(2\pi c/a)$ and from $0.990(2\pi c/a)$ to $1.028(2\pi c/a)$ (where a is the lattice constant) and (ii) the inverse-opal structure [5] consisting of air spheres in a medium with $n = 3.6$ and $f = 0.74$, whose absolute PBG ranges from $0.756(2\pi c/a)$ to $0.780(2\pi c/a)$. The FBZ was divided into 442 368 mesh points with the method of Ref. [30]. The eigenmodes were

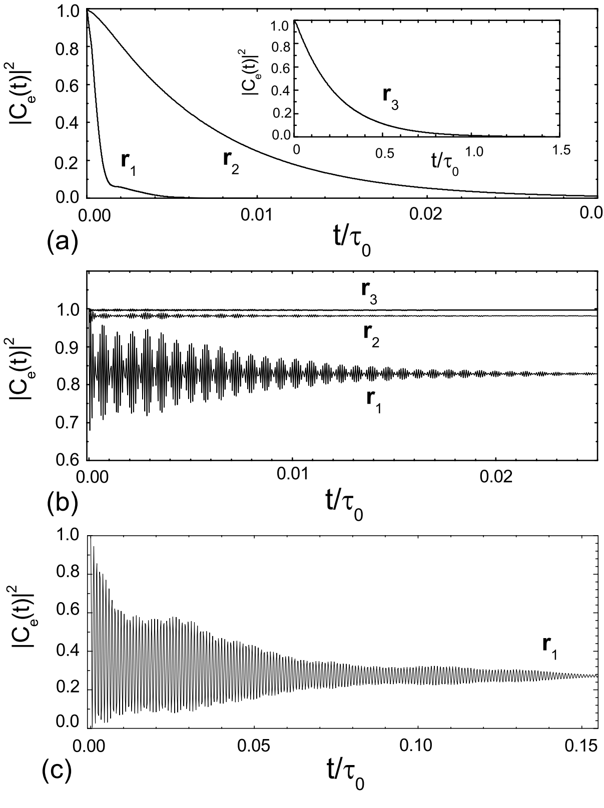

FIG. 1. Time evolution of the excited state population in the diamond structure for an atom at three different positions, $\mathbf{r}_1 = (0,0,0)a$, $\mathbf{r}_2 = (0.05,-0.125,0)a$, and $\mathbf{r}_3 = (0.5,0,0)a$. (a) For transition frequency $\omega_0 = 1.032(2\pi c/a)$ outside the gap and $\alpha_0 = 3 \times 10^{-5}$, (b) for $\omega_0 = 1.018(2\pi c/a)$ inside the gap and $\alpha_0 = 3 \times 10^{-5}$ (i.e., the case of nonresonant emission), and (c) for $\omega_0 = 1.018(2\pi c/a)$ and $\alpha_0 = 3 \times 10^{-4}$ (i.e., the case of resonant emission). τ_0 is the lifetime of the atom in free space.

solved by expanding the EM field with 965 and 4015 plane waves for the inverse-opal and diamond structures, respectively. The convergence accuracy of the eigenmodes was checked against calculations using the block iterative frequency domain method [31] with 32 768 plane waves and was found to be better than 1.2%.

Figure 1 displays the decay behavior of an excited atom at three different positions in the diamond structure for ω_0 located (a) outside and (b) inside the second PBG. It is evident from Fig. 1(a) that when ω_0 is outside the PBG and near the gap edge, an excited atom at $\mathbf{r} = \mathbf{r}_1$ decays nonexponentially, while an excited atom at $\mathbf{r} = \mathbf{r}_2$ or $\mathbf{r} = \mathbf{r}_3$ decays exponentially to a good approximation. It is also clearly seen from Fig. 1(b) that when ω_0 is inside the PBG and near the gap edge, the population of the excited state of an atom at $\mathbf{r} = \mathbf{r}_1$ or $\mathbf{r} = \mathbf{r}_2$ exhibits Rabi oscillations with a damped envelope and is rapidly trapped into a fractionalized steady state, while this oscillatory and fractional trapping phenomenon is unobservable for an atom at $\mathbf{r} = \mathbf{r}_3$, and the WWA is valid. These results show that an atom at different positions in the PC can have fundamentally different radiation properties.

The above phenomena can be understood as follows. Equations (7) and (8) show that the emission decay of the excited state depends sensitively on the LCS. The LCS in the diamond structure is shown in Fig. 2 for the same three atomic positions as in Fig. 1. It is seen that the LCS for the three atomic positions exhibits drastic oscillations, and the difference in the peak values of the LCS near the band edges is of the order of 10^2 for different atomic positions. It is this giant difference in the LCS that leads to an essential difference in the emission behavior of an atom.

Furthermore, the frequency ω_m of a dressed-atom state is given by $\omega - \omega_0 = \Delta(\mathbf{r}, \omega)$ [11,22,32]. When there is a dressed-atom state inside a PBG, the evolution spectrum $C_e(\mathbf{r}, \omega)$ of the excited state can be expressed as

$$C_e(\mathbf{r}, \omega) = \begin{cases} \frac{1}{1-\Delta'(\mathbf{r},\omega_m)}\delta(\omega - \omega_m), & \text{(inside the PBG)}, \\ \frac{1}{\pi}\frac{\Gamma(\mathbf{r},\omega)/2}{[\omega-\omega_0-\Delta(\mathbf{r},\omega)]^2+\Gamma^2(\mathbf{r},\omega)/4}, & \text{(outside the PBG)}, \end{cases}$$

where $\Delta'(\mathbf{r}, \omega_m)$ is the first-order derivative of $\Delta(\mathbf{r}, \omega)$. The δ function inside the PBG leads to a localized field (LF), while the evolution spectrum outside the PBG corresponds to a propagating field (PF). The superposition of the LF and PF results in Rabi oscillations and fractional trapping behavior in the population of the excited state. However, when the LCS is small, as in the case of $\mathbf{r} = \mathbf{r}_3$, the presence of the PF does not bring about any observable effect in the excited-state population. It is worth pointing out that there are two types of emissions appearing outside the PBG. One is nonresonant emission in which no dressed-atom state may exist outside the PBG (i.e., no well-defined central frequency can be found in the PF). The anisotropic dispersion model predicted a similar "diffusion field." However, this field is extremely small and does not cause observable oscillations [13], as in the case of $\mathbf{r} = \mathbf{r}_3$ in Fig. 1(b). The other is resonant emission in which there exists another dressed-atom state outside the PBG. This corresponds to a splitting of the excited level, as predicted by the isotropic dispersion model [6]. This splitting can occur when $\alpha_0 \geq 3.0 \times 10^{-4}$ for an atom at $\mathbf{r} = \mathbf{r}_1$. Figure 1(c) shows the time evolution of

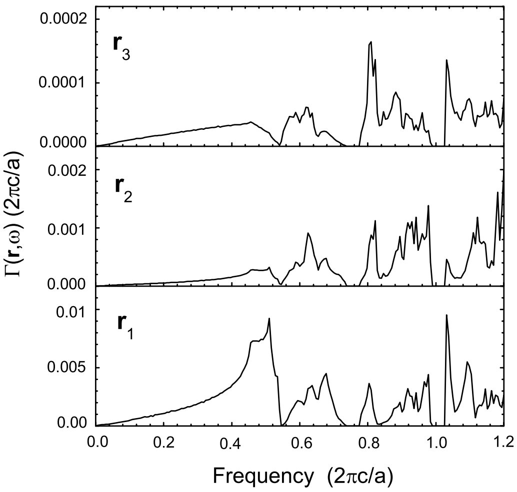

FIG. 2. Local coupling strength, $\Gamma(\mathbf{r}, \omega)$, in the diamond structure for an atom with $\alpha_0 = 3 \times 10^{-5}$ at the same three positions as in Fig. 1.

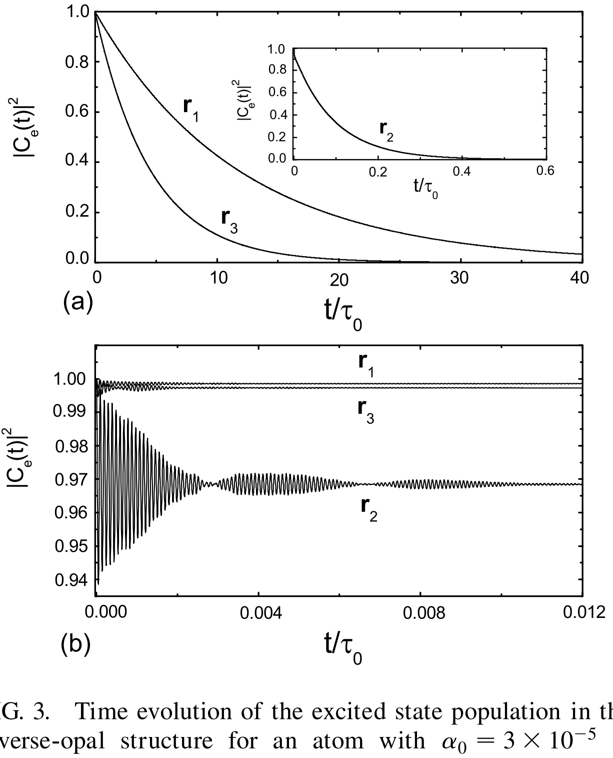

FIG. 3. Time evolution of the excited state population in the inverse-opal structure for an atom with $\alpha_0 = 3 \times 10^{-5}$ at three different positions, $\mathbf{r}_1 = (0, 0, 0)a$, $\mathbf{r}_2 = (0.34, 0, 0)a$, and $\mathbf{r}_3 = (0.24, 0.24, 0)a$. (a) For transition frequency $\omega_0 = 0.745(2\pi c/a)$ outside the gap and (b) for $\omega_0 = 0.762(2\pi c/a)$ inside the gap.

the excited atom population for resonant emission, in which the completely different oscillatory behavior occurs. Moreover, the resonant emission leads to a steady population of the upper level of about 0.27, much less than the value of 0.83 found in the nonresonant emission case.

Figure 3 shows the time evolution of the excited-state population in the inverse-opal structure for three different atomic positions. When the transition frequency $\omega_0 = 0.745(2\pi c/a)$ is outside the PBG, an excited atom at the three atomic positions exhibits the well defined exponential decay behavior [see Fig. 3(a)]. On the contrary, when $\omega_0 = 0.762(2\pi c/a)$ is inside the PBG, it is clearly seen that for an atom at $\mathbf{r} = \mathbf{r}_2$ [Fig. 3(b)] the population of the excited state exhibits Rabi oscillations and approaches a steady value of 0.9675 in the long-time limit. However, for an atom at $\mathbf{r} = \mathbf{r}_1$ or $\mathbf{r}_3$, the oscillatory magnitudes of the populations are extremely small, and the steady populations are 0.9985 and 0.9973, respectively. Thus, for an atom at $\mathbf{r} = \mathbf{r}_1$ or $\mathbf{r}_3$, the WWA is valid.

A possible experimental approach for observing the predictions in Figs. 1 and 3 is to randomly distribute a great number of atoms or molecules into the background medium of PCs. Atoms at different positions should then exhibit different decay properties. Finally, it is worth pointing out that stacking faults often occur in a fabricated inverse-opal PC. It has been shown that stacking faults can enlarge a PBG and introduce localized photon modes in the gap [33]. This can lead to an extremely strong atom-photon interaction for an atom lying in the defect region and can influence the light emissions of the atom profoundly. The study of this problem is in progress.

In summary, we have surveyed the spontaneous emissions of a two-level atom in PCs with absolute PBGs and found that the variation of the atomic position in a PC can have the decisive influence on the radiative behavior of an atom. It is anticipated that our study may provide a new way to reveal the interaction between photons and atoms (or molecules) in strong inhomogeneous electromagnetic systems, such as metal nanoparticle systems.

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