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Spontaneous emission and lame shift in photonic crystals

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Abstract

Being motivated by the controversial results based on two dispersion models and Weisskopf–Wigner approximation (WWA), we introduce, for the first time to our knowledge, the position-dependent photon-atom interaction into the Green function method of the evolution operator and develop a universal theoretical approach to study spontaneous emission of atoms in photonic crystals (PCs). A position-sensitive generalized Lorentzian formalism (non-Lorentzian shape) for the decay of an excited atom in PCs is derived, and an exact numerical method for calculating the local coupling strength, proportional to the photonic local density of state (LDOS), is presented. For weak interaction PCs with pseudo gaps, the generalized Lorentzian formalism may be reduced to the famous Lorentzian spectrum. In this case, we introduce a lifetime distribution function for an assembly of atoms and find that it depends strongly on the atomic configuration in space, which clarifies successfully the tremendous discrepancy between different experiments. For the PCs with large full gaps, we find that the atomic position can fundamentally change the decay behavior of an excited atom: in strong interaction positions, the atomic decay is non-classical or exhibits an envelope-damped Rabi oscillation, while in weak interaction positions the WWA is valid. Recently, we also predicted giant Lamb shifts for hydrogen atoms in PCs, and revealed that in inhomogeneous electromagnetic environment, the dominant contribution to the Lamb shift comes from real photon emission, while the contribution from emission and reabsorption of virtual photon is negligible, in vast contrast with the case of free space where the virtual photon processes play a key role.

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1. Introduction

Since the pioneering works of Yablonovich and John [1], there has been a growing interest in both fabrication of photonic band gap (PBG) structures [2–5] and study of quantum electrodynamics (QED) behavior of atoms in photonic crystals (PCs) [6–14]. Since 1990, the isotropic and anisotropic dispersion models have been extensively employed to solve the QED problems in PCs. They predicted many novel quantum optics characteristics, such as the appearance of photon-atom bound states [6,7], the coexistence of the photon-atom bound states and propagating states in close vicinity to the band edges [6], the spectral splitting and oscillation behavior of spontaneous emission [8], enhanced quantum interference effects [9], the coherent control of spontaneous emission [10], non-Markovian effect [7,11,12], the existence of the diffusion field [13], etc. However, these two models only simulate the dispersion relation extremely close to the band edges, and furthermore ignore the strong space inhomogeneity of electromagnetic field in PCs. On the other hand, it has been recognized that the local density of states (LDOS) is more decisive for emission behavior of atoms in inhomogeneous media [15–20], when the position-dependent interaction between photons and atom is taken into account. Based upon slow variation and non-singularity near the band edges in both the DOS and LDOS of 3D PCs with absolute PBGs, the Weisskopf–Wigner approximation (WWA) was regarded as being universally valid for spontaneous emissions in 3D PCs [19]. The conclusion intimates that the predictions from the two dispersion models should be denied. This conducted to a very significant controversy on QED in photonic crystals. It is well known that the WWA is valid only when the coupling interaction between an atom and photons is weak. Based upon
the Green function method of the evolution operator, we recently developed and presented a general theory treatment on the spontaneous emission and Lamb shift of two-level atoms located in inhomogeneous electromagnetic environments [20–22], which is valid for the cases of both strong coupling interaction and strong space inhomogeneity. The validity conditions of both the dispersion models and WWA can be clearly revealed in this theoretical framework. For the PCs with large full gaps, we found that the atomic position can fundamentally change the decay behavior of an excited atom: in strong interaction positions, the atomic decay is non-classical or exhibits an envelope-damped Rabi oscillation, while in weak interaction positions the WWA is valid [21].

Lamb shift has been one of the central topics in QED. Naturally, strong suppression or enhancement of light emission by the PC environment is expected to modify the Lamb shift. However, very different predictions for the Lamb shift can be found in literature. The isotropic dispersion model [8] predicts an anomalous Lamb shift and level splitting for multi-level atoms. For two-level atoms, the anisotropic model [13] suggests that the Lamb shift should be much smaller than that in vacuum, while the pseudogap model [14] predicts a change of the Lamb shift of the order of 15% compared to its vacuum value. At last, a direct extension of the Lamb shift formulism for multi-level atoms in vacuum to the case of PCs suggests that the Lamb shift differs negligibly from its vacuum value [23]. Motivated by these controversial results, we derived a general equation of determining the Lamb shift of multi-level atoms in an inhomogeneous electromagnetic environment [24]. It is revealed that the dominant contribution to the Lamb shift comes from emission of real photon, while the contribution from emission and reabsorption of virtual photon is negligible. This is completely different from the conventional concept set up in 1947 [25], i.e. the virtual photon processes play a key role in Lamb shift of atoms in free space. The properties of the Lamb shift near the band gap are calculated numerically for an inverse opal PC. We find that the PC structure can lead to a giant Lamb shift, and the Lamb shift is sensitive to both the position of an atom in PCs and the transition frequency of the related excited level.

A few of groups in the world have reported the experimental observations on suppression or enhancement of spontaneous emission of dye molecules [26–30] and quantum dots [31] in PCs. Due to the difficulty in the fabrication of samples, the experimental studies in this topic have been achieved only in the 3D PCs with pseudo PBG. The inhibition effect of radiative decay in PC’s was first reported in 1990 [26]. However, a later comprehensive experiment showed that the non-PC effects can account for a major fraction of the change in the radiative lifetime [27]. Eight years later, the wide lifetime distribution containing both inhibited and accelerated decay components was observed in the artificial opal sample [28], in which dye molecules homogeneously spread over the background solution. However, when the dye molecules are homogeneously embedded on a spherical layer inside silica globules in a similar system [29], only a single decay lifetime was found and a very little change in the decay lifetime was observed.

Evidently, the substantial discrepancies exist in these experimental observations [30]. To clearly understand these discrepancies, we for the first time introduced a lifetime distribution function for an assembly of atoms (or molecules) in 3D photonic crystals (PC’s) with pseudo gap [20]. Our calculations show that quite wide or narrow lifetime distributions can occur for different spread configurations of the atoms (or molecules). The pure PC effect may lead to coexistence of both accelerated and inhibited decay processes. These results provide theoretical clarification for substantial discrepancies in the reported experiments.

This paper is organized as follows: In Section 2, the theoretical framework based upon Green’s function method of evolution operator is described. In Section 3, numerical results on lifetime distribution for an assembly of atoms in the PCs with pseudo PBGs, dynamic decay processes for atoms in the PCs with full PBGs, and giant Lamb shift of atomic hydrogen in inverse-opal photonic crystals, are presented. Finally, a brief conclusion is drawn in Section 4.

2. Theoretical treatment on spontaneous emission and lamb shift in photonic crystals

In this section, we present the theoretical description on dynamic decay processes of excited atoms in PCs. Firstly we consider decay behavior of single atom. Then, we introduce a lifetime distribution function for an assembly of atoms. Finally, we derive a general equation of determining the Lamb shift.

2.1. Generalized Lorentzian spectrum formula

We consider spontaneous emission of multi-level atom located at the position \( r \) in PCs. Hamiltonian of the system can be presented in the form

\[
H = H_0 + H_{\text{int}} + H_{\text{ct}},
\]

with

\[
H_0 = \frac{p^2}{2m} + V_d + \hbar \sum_{nk} \omega_{nk} a_{nk}^\dagger a_{nk}, \quad H_{\text{int}} = \frac{e}{m} \mathbf{p} \cdot \mathbf{A}(r),
\]

\[
H_{\text{ct}} = (\delta m) m \mathbf{p}^2/2m,
\]

where the term \( H_0 \) stands for noninteracting Hamiltonian; the term \( H_{\text{int}} \) describes interaction between an atom and photons,

\[
\mathbf{A}(r) = \sum_{nk} (\hbar/2\omega_{nk})^{1/2} [\mathbf{E}_n(k, r) a_{nk}^\dagger + \text{H.C.}]
\]

being the quantized vector potential, the second-order term of the vector potential in \( H_{\text{int}} \) has been neglected; \( H_{\text{ct}} \) is a mass-renormalization counter-term for an electron of observable mass \( m \) [6,32]. The electromagnetic (EM) eigenmodes \( \{ \omega_{nk}, \mathbf{E}_{nk}(r) \} \) in PCs can be found by the plane-wave expansion method [33].

We assume that an atom is excited initially, and it stays at the \( j \)th energy level without a photon in the EM field, and denote \( |j\rangle = |j, 1\rangle \) and \( |F_{nk}^+\rangle = |j, 1_{nk}\rangle \) (i.e. the atom is at the level \( j \) and the EM field has a photon in the state \( n k \)) as the initial
and final states of the system, respectively. The state vector of the system evolves in term of
\[ |\Psi(t)\rangle = U(t)|I\rangle = C_\varepsilon(t)|I\rangle + \sum_{j,k} C_{jk}(t)|F^j_k\rangle, \]
(2)

with the initial conditions \(C_{\varepsilon}(0) = 1\) and \(C_{nk}(0) = 0\), where \(U(t)\) is the evolution operator. Applying the Green’s function technique to the evolution operator, we obtain the Fourier transform \(C_\varepsilon(\omega)\) of \(C_\varepsilon(t)\) in the form \([34]\),
\[ C_\varepsilon(\omega) = \frac{1}{2\pi i} \left[ G^{\dagger}_{\varepsilon}(\omega) - G^+_{\varepsilon}(\omega) \right], \]
(3)

with
\[ G^{\dagger}_{\varepsilon}(\omega) = \lim_{\eta \to 0^+} \langle I | G(z = \omega \pm i\eta) | I \rangle, \]

where \(G(z)\) is defined by the operator identity \(G(z) = H + i\eta\) = 1. Projecting this operator identity onto the one-photon Hilbert space \([35]\) and noting that the nonvanishing matrix elements of \(H_{\text{int}}\) are \(\langle F^j_k | H_{\text{int}} | I \rangle\), we obtain the following analytic expression
\[ G^{\dagger}_{\varepsilon}(\omega) = \lim_{\eta \to 0^+} \frac{1}{\omega - p - \Delta(r, \omega) \pm i|\Gamma(r, \omega)/2 + \eta|}, \]
(4)

where
\[ \Gamma(r, \omega) = \sum_j \alpha_j g(r, \omega - \omega_j), \]
(5)
\[ \Delta(r, \omega) = \sum_j (\omega_j/2\pi)(\omega - \omega_j)\beta(r, \omega - \omega_j), \]
(6)

with
\[ g(r, \omega) = \frac{\pi^2 c^3}{\omega} \sum_{nk} |E_{nk}(r)|^2 \delta(\omega - \omega_{nk}), \]
(7)
\[ \beta(r, \omega - \omega_j) = \rho \int_{0}^{\omega_{\text{opt}}} \frac{g(r, \omega')}{(\omega - \omega_j - \omega')\omega'} d\omega'. \]
(8)

Here \(\omega_{\text{opt}} = mc^2/\eta\) is the relativistic limit of the photon energy \([25]\), \(\alpha_j = e^2|p_j|^2/3\pi m^2 c^2\) is the relative line width of the atomic radiation from the \(l\)-state to \(j\)-state in vacuum, and \(\rho\) stands for the principal value of the integral. In Eqs. (7) and (8), we have considered a random orientation of \(p_j\) and include the mass-renormalization contribution, respectively \([6,13,14,23,32]\). The function \(g(r, \omega)\) is the local spectral response function (LSRF) proportional to the photon local DOS.

Combining Eq. (3) with Eq. (4), we find that the decay of an excited atom can be seen
\[ C_\varepsilon(t) = \int_{-\infty}^{\infty} d\omega C_\varepsilon(\omega)e^{-i\omega t}, \]
(9)

with
\[ C_\varepsilon(\omega) = \frac{1}{\pi} \lim_{\eta \to 0^+} \frac{\Gamma(r, \omega)/2 + \eta}{|\omega - \omega_l - \Delta(r, \omega)|^2 + [\Gamma(r, \omega)/2 + \eta]^2}. \]
(10)

Eq. (10) clearly shows that the evolution spectrum of the upper level is of a non-Lorentzian shape. It provides a universal description for spontaneous emission of an atom in inhomogeneous media. In, we will show that in weak coupling limit, Eq. (10) can be reduced to pure exponential decay and the famous Lorentzian spectrum, i.e. the WWA. It can be seen from Eq. (10) that \(\Gamma(r, \omega)\) and \(\Delta(r, \omega)\) represent the local coupling strength (LCS) and the level shift, respectively.

The calculation of \(g(r, \omega)\) in Eq. (7) involves an integral of the 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 (IBZ) \([19,36]\), based on a linear tetrahedron method \([37]\) 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 invalid, and an exact numerical method for the evaluation of integrals of vectorial fields in PCs has been established \([22]\). For calculating \(\beta(r, \omega - \omega_j)\), we make a reasonable approximation \([23,38]\): the dispersion function \(\varepsilon(r) - 1\) of a PC vanishes jump-wise at a certain higher optical frequency \(\omega_{\text{opt}}\), i.e. for \(\omega > \omega_{\text{opt}}\), the PC medium is approximately treated as a free space with \(\varepsilon(r) \approx 1\). We choose \(\omega_{\text{opt}}\) in such a way that our results are verified to be insensitive to perturbations. \(\omega_{\text{opt}}\) is found to be \(2\pi c(3.5)\) chosen in our calculations, where \(\alpha\) is the lattice constant of photonic crystals.

2.2. Lifetime distribution function for an assembly of atoms: weak coupling limit

We consider an assembly of two-level atoms that randomly spread in PCs with pseudo PBGs where the coupling interaction between atoms and photons is weak. In this case, one can make a reasonable approximation to Eq. (10) by setting \(\omega_{\text{opt}}\) in \(\Gamma(r, \omega)\) and \(\Delta(r, \omega)\). This leads to the famous exponential decay law (i.e. the WWA) with the radiative line width \(\Gamma(r, \omega)\) and Lamb shift \(\Delta(r, \omega)\).

\[ C_\varepsilon(t) = \exp(-i[\omega_l + \Delta(r, \omega_l)t - \Gamma(r, \omega_l)t/2]), \]
(11)
\[ C_\varepsilon(\omega) = \frac{1}{2\pi} \frac{\Gamma(r, \omega)}{[\omega - \omega_l - \Delta(r, \omega_l)]^2 + [\Gamma(r, \omega)/2]^2}. \]
(12)

For a two-level atom, we take \(j = g, e, l = e, \omega_l = 0\), and set \(\omega_0 = \omega_0 - \omega_g = \omega_e\). Note that \(\alpha_{\omega_g} = \alpha_\omega = 0\), then \(\Gamma(r, \omega_0 = \omega_0)\) and \(\Delta(r, \omega_e = \omega_0)\) can be written as
\[ \Gamma(r, \omega_0) = \frac{\alpha_\omega \pi^2 c^3}{\omega_0} \sum_{nk} |E_{nk}(r)|^2 \delta(\omega_0 - \omega_{nk}), \]
(13)
\[ \Delta(r, \omega_0) = \frac{1}{2\pi \rho} \int_{0}^{\omega_{\text{opt}}} \omega_0 \Gamma(r, \omega') d\omega'. \]
(14)

Eq. (12) shows that the population of the excited level decays by \(|C_\varepsilon(\omega)|^2 = \frac{\Gamma(r, \omega)}{\Gamma(r, \omega)}\). Thus, the lifetime of the excited state is \(\tau(r, \omega_0) = 1/\Gamma(r, \omega_0)\).
(15)

In fact, Eq. (13) can gives the famous expression \(\Gamma_{\omega_0}(\omega_0) = \alpha_\omega^2 \omega_0^3/3\pi \rho_0^2 c^5 = \eta \Gamma_{\omega_0}(\omega_0)\).
The radiative intensity $I(\tilde{r})$ from these atoms (molecules) with the lifetime $\tilde{r}$ should be proportional to $\rho(\tilde{r})$. Note that $|E_{ak}(\tilde{r} + \mathbf{R}_i)| = |E_{ak}(\mathbf{r})|$ for the PC’s, where $\mathbf{R}_i$ is a lattice vector due to Bloch theorem. This makes it possible to calculate the distribution of the position-dependent decay lifetimes in a unit cell.

2.3. Lamb shift

Eq. (10) shows the radiative correction to the bound level $b$ is determined by

$$\omega - \omega_b = \Delta(\mathbf{r}, \omega).$$

Inserting Eq. (6) into Eq. (17), we have

$$\omega - \omega_b = \sum_j \frac{\alpha_j}{2\pi} (\omega - \omega_j) \beta(\mathbf{r}, \omega - \omega_j).$$

In the two dispersion models, $|E_{ak}(\tilde{r})|^2 (\nu^2/m^2)$, then Eq. (18) just gives the results described by Eq. (6a) of Ref. [6] provided we take $l=1$. For a two-level atom with $j=0, 1$, we note that $\alpha_{11} = 0$ due to $p_{11} \equiv 0$ ($p_{ij} = i(\omega - \omega_j)m_{ij}$), and Eq. (18) can be simplified to Eq. (4.9) of Ref. [14] by setting $l=1$ and $\omega_0$ as zero point of energy. In vacuum, $g(\mathbf{r}, \omega') = \omega'$, and by setting $\omega = \omega_0$ in the right-hand side of Eq. (18), we obtain

$$\Delta_0^0 = \frac{e^2}{6\pi^2 m_{0}^2 \epsilon_{0}^3} \sum_j -\omega_j |p_{ij}|^2 \beta(\mathbf{r}, -\omega_j),$$

where $\omega_j = \omega_j - \omega_0$ and

$$\beta(\mathbf{r}, -\omega_j) = -\ln(w_{\text{rad}}(w_{ij} + 1)) \approx -\ln(w_{\text{rad}}(w_{ij})).$$

Because $\beta(\mathbf{r}, -\omega_j)$ is a slowly varying function of $\omega_j$, it is reasonable to make the approximation, $\omega_j \approx \omega_j - \omega_0$, for $\beta(\mathbf{r}, -\omega_j)$ (see also Ref. [31]), with $\omega_0 > \omega_j$ being a weighted average of $\{\omega_j\}$. This approach implies that the dominant contributions to the Lamb shift come from the emission and reabsorption of virtual photons (corresponding to the transition processes from the $l$ level to higher levels), rather than emission of real photon (corresponding to transition processes from the $l$ level to lower levels). Noticing that $\sum_j \omega_j |p_{ij}|^2 = \hbar e^2 |\psi(0)|^2/2\epsilon_0$, where $\psi(0)$ is the wave function value at the center of an atom in the state $|l\rangle$, we finally obtain a standard nonrelativistic result,

$$\Delta_0^0 = \frac{e^2|\psi(0)|^2}{12\pi^2 m_{0}^2 \epsilon_{0}^3} \ln \frac{\omega_{\text{rad}}}{(\tilde{\delta} - \omega_0)}.$$

Thus, Eq. (18) gives a general result for a nonrelativistic radiative correction to a bound level of a multi-level atom in an inhomogeneous EM system.

When Eq. (18) is numerically solved for an actual PC structure, two different types of integrals for $\beta(\mathbf{r}, \omega - \omega_j)$ is encountered: the principal integral, when the integrand in Eq. (8) has a singularity, and the normal integral, otherwise. With this in hand, we find that the terms for $j < l$ and for $j > l$ in the right hand side of Eq. (18) contribute the principal and normal integrals near $\omega_j$, respectively. In order to show this clearly, we assume that $\omega = \tilde{\delta} + \omega_j$ is a solution of Eq. (18), and $[\delta] = \omega_{j+1}$, $[\omega] = \omega_{j-1}$, where $\omega_{j+1}$ is the closest to and higher than the frequency of the level $l$. For $j < l$, the integrand has a singularity due to $\tilde{\delta} + \omega_j - \omega_0 > 0$. But for $j > l$, the integrand has no singularity due to $\tilde{\delta} + \omega_j - \omega_0 < 0$.

In PCs, the LSRF $g(\mathbf{r}, \omega')$ displays dramatic fluctuations when the frequency $\omega'$ varies for a given position. Thus, the principal integral $\beta(\mathbf{r}, \omega') (\omega > 0)$ should be very sensitive to the value of $\omega$, and the contribution to the integral comes mainly from the region near the frequency $\omega$. However, for the normal integral $\beta(\mathbf{r}, \omega') (\omega > 0)$, the fluctuation in $g(\mathbf{r}, \omega')$ are smoothed out after integration, and $\beta(\mathbf{r}, \omega')$ is a slowly varying function of $\omega$, similar to the case of vacuum. These arguments have been confirmed in Ref. [24]. Therefore, the terms with $j > l$ in the right-hand side of Eq. (18) can be treated similar to the case of vacuum. If we consider $\omega - \omega_j \gg 1$, then the PCs do not bring about appreciable changes in those terms with $j < l$ compared to the case of vacuum. Therefore, Eq. (18) can be approximated as follows

$$\omega - \omega_l - \Delta_0^0 = \sum_{j < l} \frac{\alpha_j(\omega - \omega_j)}{2\pi} \beta(\mathbf{r}, -\omega_j) \int_{0}^{\omega_r} g(\mathbf{r}, \omega') - \omega' \frac{\omega}{(\omega - \omega_j - \omega')\omega'} d\omega'.

(21)

Eq. (21) shows that, compared to the case of vacuum, inhomogeneous EM systems lead to an additional contribution to the Lamb shift that comes mainly from the real photon processes, rather than the virtual photon processes, in contrast to the case of vacuum.

3. Application of the theory

In this section, we apply the theory addressed in section to study the spontaneous emission in actual PCs, and presented some numerical results.

3.1. Lifetime distribution in fcc PC structures

We consider two PC’s of the fcc lattice consisting of spherical globules with a refractive index $n$ in the background media with refractive index $n_b$: (a) PC1 with $n = 1.3$, $n_b = 1.49$, and filling fraction $f = 0.74$; (b) PC2 with $n = 1.45$, $n_b = 1.33$, and $f = 0.60$. The PC1 and PC2 possess almost the same parameters as those of the samples in experiments of
The value of $n=1.49$, close to $1.49 \Gamma_v$. As there exists no exact expression for the SER in the weak absorption case, thus, we prefer to adopt three different values of $\Gamma_f=1.00 \Gamma_v$, $1.20 \Gamma_v$, and $1.49 \Gamma_v$, in Fig. 1(b). In addition, the size inhomogeneity of the dielectric globules in the sample of Petrov et al. also causes some quantitative discrepancies between the theoretical evaluations and experimental results. Even so, our results have demonstrated that the pure PC effect may result in the wide distribution of lifetimes and the coexistence of both the accelerated and inhibited components in spontaneous decay kinetics.

We now turn to study PC2 sample. Fig. 2(a) displays RWOLD $T_{rw}$ at the center of the pseudo gap (i.e. at the probe frequency in the experiment of Megens et al. [29]) to be about 2% for the atoms (molecules) located on two different spherical surfaces inside the dielectric globules, and no appreciable PBG effects can be observed. This result agrees well with the experimental result of Megens et al. A very narrow RWOLD for different spherical surfaces manifests that the radial distribution of the lifetimes (RDOL) along any direction can very well reveal the decay kinetics for homogeneous spreading of the atoms (molecules) over the whole dielectric globules. Fig. 2(b) displays the RWOLD $T_{rw}$ inside the whole dielectric globules still remains large, up to about 38%. It is very interesting to note that the change in the lifetimes is lower than 4% for $0.4 \leq r/r_0 \leq 0.8$. The radius of the atomic layers of the samples in the experiment of Megens et al. is just $r/r_0=0.6$. Apparently, the special space configuration of the atoms (molecules) leads approximately to a single lifetime decay in the experiment of Megens et al., which is invalid for the homogeneous spreading of the atoms (molecules) over the whole dielectric globules. The symmetry of crystals leads to a very narrow lifetime distribution. In addition, when two atomic

![Graph](image_url)

**Fig. 1.** (a) Radiative line widths for two different atomic positions $r_1=(0.5, 0.5, 0.5)$ (solid line) and $r_2=(0.25, 0.25, 0.01)$ (dotted line) in the background medium of PC1. Center frequency $\omega_p$ of the pseudo gap is indicated by arrows. (b) Decay lifetime distribution for the homogeneous spread of atoms over the background medium of PC1. The solid, dotted, and dashed lines correspond to $\tau_1=\tau_2$, $\tau_1/1.2$, and $\tau_1/n$, respectively.

Refs. [28,29], respectively. These PC’s have pseudo gaps along the (111) direction.

**Fig. 1(a)** displays the radiative line width for two different positions of the atom (molecule) in the background medium of PC1. It can be seen that the ratio $\Gamma(r_1, \omega)/\Gamma(r_2, \omega)$ near the pseudo gap is high up to about 1.7. This implies that the lifetimes of the atoms (or molecules) possess a quite wide distribution. To clearly show this, we plot the lifetime distribution for homogeneous dispersion of the atoms (molecules) over the background medium in Fig. 1(b) with three different reference lifetimes, in which the frequency is at the lower band edge of the pseudo gap (i.e. at the probe frequency in the experiment of Petrov et al. [28]). Indeed, a very wide lifetime distribution is found. The relative width of the lifetime distribution (RWOLD) $T_{rw}=(\tau_{\text{max}}-\tau_{\text{min}})/\tau_{\text{min}}$ for three different reference lifetimes is about 70%, which closes the measured value of about 120% in the experiment of Petrov et al. [28]. We also observe that the fraction of the accelerated or inhibited components in the whole distribution depends on the scale of the reference lifetime $\tau_f$. In the case of atoms inside an optically thick film, if the localized modes can be reabsorbed and do not carry any energy away [40], the spontaneous emission rate (SER) is $I_f=I_v[n-(n^2-1)/2]$, where $n$ is the refractive index and $\Gamma_v$ is the SER in vacuum. On the other hand, if there is no absorption of emitted luminescence in the film, the SER is $\Gamma_f=\Gamma_v n$ [18]. In Petrov et al.’s experiment, a weak reabsorption of the fluorescence from the dye molecules exists in the reference film [28]. As a result, the value of $\Gamma_f$ is between $0.385 \Gamma_v$ and $1.49 \Gamma_v$ for

![Graph](image_url)

**Fig. 2.** (a) Lifetime distribution of atoms (molecules) located on two spherical surfaces with different radius $r$ inside the dielectric globules of the radius being $r_0$ in the PC2, the solid line for $r=0.6 \ r_0$ and the dotted line for $r=0.8 \ r_0$. (b) Variation of the lifetime with the radius of the spherical surface along the $x$-axes direction. $r_0$ is the lifetime of the atoms in an infinite homogeneous medium with $n=1.45$. 
layers of $r/r_0 = 0.2$ and 0.9 are simultaneously embedded in the dielectric globules, the spontaneous decay kinetics containing both the accelerated and inhibited components can also be observed in PC2.

3.2. Non-classical decay in diamond structure

We now exam the decay behavior of an excited atom in the diamond structure [33,41] consisting of dielectric spheres of the refractive index $n = 3.6$ in the air background with a filling fraction $f = 0.31$, whose absolute PBGs span from 0.738 to 0.776 and from 0.990 to 1.028 $(2\pi c/\lambda)$ (where $\lambda$ is the lattice constant). The FBZ was divided into 442368 mesh points with the method of Ref. [42]. The eigenmodes were solved by expanding the EM field with 4015 plane waves. The convergence accuracy of the iterative frequency domain method [43] with 32,768 plane waves, and was found to be better than 1.2%.

Fig. 3 displays the decay behavior of an excited atom at three different positions in the diamond structure for $\omega_0$ located (a) outside and (b) inside the second PBG. It is evident from Fig. 3 (a) that when $\omega_0$ is outside the PBG and near the gap edge, an excited atom at $r = r_1$ decays non-exponentially, while an excited atom at $r = r_2$ or $r = r_3$ decays exponentially to a good approximation. It is also clearly seen from Fig. 3(b) that when $\omega_0$ is inside the PBG and near the gap edge, the population of the excited state of an atom at $r = r_1$ or $r = r_2$ 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 $r = r_3$, 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. Eq. (10) shows that the emission decay of the excited state depends sensitively on the LCS. The LCS in the diamond structure is shown in Fig. 4 for the same three atomic positions as in Fig. 3. 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 $\omega_m$ of a dressed-atom state is given by $\omega - \omega_0 = \Delta(r, \omega)$ [11,34,44]. When there is a dressed-atom state inside a PBG, the evolution spectrum $C_e(\omega)$ of the excited state can be expressed as

$$C_e(\omega) = \begin{cases} 1 - \Delta'(r, \omega_0) \delta(\omega - \omega_m), & \text{inside the PBG}, \\ \frac{1}{\pi} \frac{T(r,\omega)/2}{(\omega - \omega_0 - \Delta(r,\omega))^2 + T^2(r,\omega)/4}, & \text{outside the PBG}, \end{cases}$$

where $\Delta'(r, \omega_0)$ is the first derivative of $\Delta(r, \omega)$. The $\delta$ 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 diamond structure for an atom at $r = r_1$, $r = r_2$, and $r = r_3$. The oscillation frequency $\omega_0 = 3 \times 10^{-5}$ at the same three positions as in Fig. 3.
the population of the excited state. However, when the LCS is small, as in the case of \( r = r_3 \), 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 non-resonant emission in which no dressed-atom state may exist outside the PBG (i.e., no well-defined central frequency can be found in the PBG). 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 \( r = r_3 \) in Fig. 3(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 \( \omega_0 \geq 3.0 \times 10^{-4} \) for an atom at \( r = r_3 \). Fig. 3(c) shows the time evolution of the excited atom population for resonant emission, in which a 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 non-resonant emission case.

3.3. Giant lamb shift in an inverse-opal structure

We now apply Eq. (21) to study the Lamb shift for a hydrogen atom in the inverse-opal PC created by air spheres in a medium with \( h = 3.6 \) and \( f = 0.74 \). The numerical results are displayed in Fig. 5. First, we obtain an interesting result that the PCs environment has no effect on the 2s state due to \( \alpha_{2s1s} = 0 \); this result coincides with the prediction obtained earlier from the isotropic dispersion model [6]. However, for the 2p state, we have \( \Delta_{2p} = 0 \) and \( \alpha_{2p1s} \approx 4 \times 10^{-7} \). Numerical results for the Lamb shift of the 2p state are presented in Fig. 5. We find no level splitting, which differs our finding from the prediction of the isotropic model [6]. In addition, the Lamb shift depends strongly on not only the transition frequency but also on the atomic space position, different from dispersion models [6,13,14]. The similar properties can also be found for the 3s, 3p, and 3d states.

Analyzing the results presented in Fig. 5, we notice that the Lamb shift can take very large positive or negative values and, therefore, it can be termed as a giant Lamb shift. Comparing the results for the PC with those for vacuum, we find that the Lamb shift may be enhanced in the PC by one or two orders of magnitude. Furthermore, it is significant to point out that the giant Lamb shift may occur for the transition frequency being either near or far away from the photonic band gap. The above-mentioned results are in contrast to the predictions based on dramatically simplified models [6,13,14,23]. In Ref. [23], a PBG structure was simply treated as an averaged homogeneous medium. This smooths out the contribution to the Lamb shift from real photon processes that play a key role in inhomogeneous systems. In the isotropic model [6], \( g(r, \omega) \sim (\omega - \omega_c)^{-1/2}/\omega \), that gives an infinite interaction between atom and photons at the band edge \( \omega = \omega_c \), leading to the level splitting and anomalous Lamb shift. In the anisotropic model [13], \( g(r, \omega) \sim (\omega - \omega_c)^{-1/2}/\omega \), that leads to coupling interaction near the band edge being smaller than that in vacuum where \( g(r, \omega) = \omega; \) it predicts much smaller Lamb shift than that in vacuum. In pseudogap model [14], \( g(r, \omega) \sim 1 - \exp((\omega - \omega_c)^2/\sigma^2) \), that gives rise to small values of the Lamb shift near a pseudogap. Clearly, these models lose the main physical characteristics of the LSRF \( g(r, \omega) \) in realistic PCs that may result in the giant Lamb shift and other significant effects.

Based upon the position-dependent Lamb shift, we can suggest a possible experimental approach for verifying our theoretical predictions: if assembly of atoms spreads randomly in PCs, the atoms at different positions have different values of the Lamb shift. Then the l-state levels of many atoms should form a l-state mini-band, similar to the velocity-dependent Doppler effect in atomic/molecular gases. This mini-band should be experimentally observable through the emission spectrum of these atoms.

4. Summary and conclusions

In this work, we have introduced the position-dependent photon-atom interaction into the Green function method of the evolution operator and have developed a universal theoretical approach to analyze spontaneous emission of atoms in photonic crystals that is valid for other inhomogeneous electromagnetic environments. We have applied our theory to study the decay behaviors of atoms in several types of PC structures. Firstly, we have investigated the decay kinetics of the spontaneous emission from an assembly of the atoms in the PC’s with pseudo PBG by using the lifetime distribution function. We have shown that the decay kinetics depends strongly on the spread configurations of the atoms (molecules) in the PC’s. The concept of the single-averaged-lifetime of spontaneous emission remains generally invalid for the PCs.
The pure PC effect may cause the coexistence of both the accelerated and inhibited decay processes. These results provide a theoretical clarification for the substantial discrepancies among the earlier reported experimental results.

Secondly, 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 a decisive influence on the radiative behavior of an atom: in strong interaction positions, the atomic decay is non-classical or exhibits an envelope-damped Rabi oscillation, while in weak interaction positions the WWA is valid. It is anticipated that our study may open a new way to reveal the interaction between photons and atoms (or molecules) in strong inhomogeneous electromagnetic systems, such as metal nanoparticle systems.

Finally, we have developed a general formalism for calculating the Lamb shift for multi-level atoms. We have revealed that the real photon processes play a key role in inhomogeneous dielectric structures. Our numerical results for systems, such as metal nanoparticle systems.

We believe our results provide a deeper insight into the theory of spontaneous emission in PCs and they may be useful for many applications such as the development of thresholdless lasers.

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