Decay Distribution of Spontaneous Emission from an Assembly of Atoms in Photonic Crystals with Pseudogaps

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Spontaneous decay behaviors from an assembly of atoms (or molecules) in 3D photonic crystals (PC's) with *pseudo*gaps are investigated. Theoretically, a *lifetime distribution function* for these atoms or molecules is defined to reveal decay kinetics. 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 recent reported experiments.

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Since the pioneering work of Yablonovitch [1] and John [2], the spontaneous emission properties of atoms or molecules in the three-dimensional (3D) photonic crystals (PC's) are extensively investigated. Because of the difficulty in the fabrication of samples, the experimental studies in this topic have been achieved only in the 3D PC's with *pseudo*-photonic-band gaps (pseudo-PBG's). The inhibition effect of radiative decay in PC's was first reported ten years ago [3]. However, a later comprehensive experiment showed that the non-PC effects can account for a major fraction of the change in the radiative lifetime [4]. Recently, the wide lifetime distribution containing both inhibited and accelerated decay components was reported in the artificial opal sample [5], 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 [6], only a single decay lifetime was found and very little change in the decay lifetime was observed. Evidently, there exist substantial discrepancies in these experimental observations. Up to now, the clearly theoretical clarification for these experiments still remains an open question [7].

Theoretically, isotropic and anisotropic dispersion models near band edges have been extensively employed to study the spontaneous emission problems in the 3D PC's with *absolute*-PBG's [8–15]. Obviously, the dispersion models are not applicable to the 3D PC's with *pseudo*-PBG's, because the propagating modes in some directions overlap with *pseudo*-PBG's. Moreover, the position dependence of interaction between atoms and photons is omitted in these models, while it has been well known in the 1D inhomogeneous system [16–18]. To understand different experimental results, the conventional averaged lifetime over a few positions of atom was evaluated [19]. But, this attempt was a failure. Furthermore, it is assumed in the numerical calculations that the magnitudes $|\mathbf{E}_{n,\mathbf{k}}(\mathbf{r})|$ of the field modes are invariant under the operation of the lattice point group operator [19–21]. Recently, this assumption has been proven to be invalid and a new calculation method is presented [22].

In this Letter, we study the decay kinetics of the atoms (or molecules) in the PC's with *pseudo*-PBG's by considering the position-dependent interaction. Theoretically, a *lifetime distribution function* for an assembly of atoms or molecules is defined and numerically evaluated to reveal the dynamic decay processes. This can provide a theoretical clarification for the recent reported experiments.

For a two-level atom at $\mathbf{r}$ point in a PC, the Hamiltonian of the system reads as

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

In Eq. (1), b_i and $b_i^+ (i = 1, 2)$ are annihilation and creation operators of electron in the ground and excited states, respectively, $a_{n\mathbf{k}}$ and $a_{n\mathbf{k}}^+$ annihilation and creation operators of the photon; ω_0 is the atomic transition frequency; $\omega_{n\mathbf{k}}$ is the frequency of the electromagnetic (EM) eigenmode $\mathbf{E}_{n\mathbf{k}}(\mathbf{r}) = ic\nabla \times \mathbf{H}_{n\mathbf{k}}(\mathbf{r})/[\epsilon(\mathbf{r})\omega_{n\mathbf{k}}]$ in the PC's, which can be solved by the plane-wave expansion method [23]; $g_{n\mathbf{k}}(\mathbf{r})$ is the coupling coefficient, 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 $\mathbf{u}_d = u_d\hat{\mathbf{u}}$ is the dipole moment between two transition levels.

For the spontaneous emission, the state vector of the system can be expressed as

$$|\Psi(t)\rangle = C_2(t)|2,0\rangle + \sum_{n\mathbf{k}} C_{1,n\mathbf{k}}(t)|1, n\mathbf{k}\rangle, \tag{3}$$

with $C_2(t = 0) = 1$ at the initial moment; $|2,0\rangle$ and $|1, n\mathbf{k}\rangle$ represent the initial and final states of the system, respectively. Obviously, the Fourier spectrum $C_2(\omega) = \int_{-\infty}^{+\infty} C_2(t)e^{-i\omega t}\,dt$ includes all the evolution information of the excited state. According to the Green function method in time domain [24], the resulting expression for $C_2(\omega)$ reads

$$C_2(\omega) = \lim_{\eta\to 0^+} \times \frac{\Gamma(\mathbf{r},\omega) + 2\eta}{[\omega - \omega_0 - \Delta(\mathbf{r},\omega)]^2 + [\Gamma(\mathbf{r},\omega)/2 + \eta]^2} \tag{4}$$

with

$$\Gamma(\mathbf{r},\omega) = 2\pi \sum_{n\mathbf{k}} |g_{n\mathbf{k}}(\mathbf{r})|^2 \delta(\omega - \omega_{n\mathbf{k}}), \tag{5}$$

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

where P represents the integral principal value.

If $\Gamma(\mathbf{r},\omega)/\omega_0 \ll 1$ and $\Gamma(\mathbf{r},\omega) \neq 0$ near the transition frequency ω_0, Eq. (4) can be reasonably approximated to the famous Lorentzian form

$$C_2(\omega) \simeq \frac{\Gamma(\mathbf{r},\omega_0)}{[\omega - \omega_0 - \Delta(\mathbf{r},\omega_0)]^2 + \Gamma^2(\mathbf{r},\omega_0)/4}, \tag{7}$$

which leads to the Weisskopf-Wigner exponential decay law with a decay lifetime

$$\tau(\mathbf{r},\omega_0) = 1/\Gamma(\mathbf{r},\omega_0). \tag{8}$$

In fact, Eq. (5) gives the famous expression $\Gamma_0(\omega_0) = u_d^2 \omega_0^3 n / 3\pi\epsilon_0 \hbar c^3 = n\Gamma_v(\omega_0)$ [$\Gamma_v(\omega_0)$ corresponds to the value in vacuum] for the homogeneous bulk medium of refractive index n when the local field effect (LFE) is ignored. It should be mentioned that the LFE is important in determining the spontaneous emission lifetime (SELT) [25]. However, the recent experiments [5,6] were to study the inhibited and enhanced effects of the PC's on the spontaneous emission. Thus, in each of these two experiments, a reference sample was prepared for making a comparison in the lifetime. The experimental conclusions were drawn from this comparison. In other words, the lifetime ratio $\tilde{\tau}(\mathbf{r},\omega_0) = \tau(\mathbf{r},\omega_0)/\tau_f(\omega_0)$ [here $\tau_f(\omega_0)$ is the SELT in the reference sample] can reveal the effects of the PC's. Moreover, the host medium (in which atoms or molecules are embedded) of the reference sample in the experiments is the same as that in the PC. For this case, it is unnecessary to consider the modification of the LFE in the calculations of the lifetime ratio, as pointed out in Ref. [18].

It is shown from Eq. (8) that the assembly of atoms (or molecules) in the PC's with *pseudo*-PBG's should lead to a distribution of the decay lifetimes, rather than only a single lifetime decay. Thus, we define a lifetime distribution function as

$$\rho(\tilde{\tau}) = \sum_i W_i \delta(\tilde{\tau} - \tilde{\tau}(\mathbf{r}_i,\omega_0)), \tag{9}$$

where $\mathbf{r}_i$ denotes the position of the ith atom (molecule) and W_i is a weight factor. For the homogeneous distribution of the atoms (molecules) in space and the random orientation of the dipole moment, we have $W_i = 1$. The radiative intensity $I(\tilde{\tau})$ from these atoms (molecules) with the lifetime $\tilde{\tau}$ should be proportional to $\rho(\tilde{\tau})$.

Note that $|g_{n\mathbf{k}}(\mathbf{r} + \mathbf{R}_l)| = |g_{n\mathbf{k}}(\mathbf{r})|$ due to Bloch theorem. This makes it possible to calculate the distribution of the position-dependent decay lifetimes in a unit cell. The detailed numerical calculations involve the following steps: (i) We discretize the first Brillouin zone (BZ) into 256 000 grid points and a unit cell (UC) of the PC into 524 288 grid points; (ii) the eigenequation of magnetic field $\mathbf{H}_{n\mathbf{k}}(\mathbf{r})$ is solved by an expansion of 965 plane waves [23], and the convergency accuracy is better than 1%. Actually numerical calculations can be achieved in an irreducible BZ and an irreducible UC, due to the symmetry of the crystals (i.e., $|\mathbf{H}_{n\{\alpha|\mathbf{t}_0\}k}(\mathbf{r})| = |\mathbf{H}_{nk}(\{\alpha|\mathbf{t}_0\}^{-1}\mathbf{r})|$ [22,26], rather than $|\mathbf{H}_{n\{\alpha|\mathbf{t}_0\}\mathbf{k}}(\mathbf{r})| = |\mathbf{H}_{nk}(\mathbf{r})|$ [19–21]; here $\{\alpha|\mathbf{t}_0\}$ represents an operator of space group [27]); (iii) the lifetime distribution function is obtained by assuming random orientation of the atomic transition dipole moment. A given dye molecule has a fixed direction of the transition dipole moment, in contrast to an atom in which the transition dipole moment fluctuates on the time scale of the excited-state lifetime. But, the molecules in different positions should have different orientations of transition dipole moment; consequently, the assumption of the random orientation of the transition dipole moment still remains valid for an assembly of dye molecules.

We consider three different PC's of the fcc lattice consisting of spherical globules with 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 $f = 0.60$; (c) PC3 with $n = 2.6$, $n_b = 1.33$, and $f = 0.65$. PC1 and PC2 possess almost the same parameters as those of the samples in the experiments of Refs. [5,6], respectively. These three PC's possess *pseudo* gaps along the (111) direction.

Figure 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 $\Gamma(\mathbf{r}_1,\omega_0)/\Gamma(\mathbf{r}_2,\omega_0)$ 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.* [5]). Indeed, a very wide lifetime distribution is found. The relative width of the lifetime distribution (RWOLD) $T_{rw} = (\tau_{\max} - \tau_{\min})/\tau_{\min}$ for three different reference lifetimes is about 70%, which closes the measured value of about 120% in the experiment of Petrov *et al.* [5]. We also observe that the fraction of the accelerated or inhibited components in the whole distribution depends on the scale of the reference lifetime τ_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 [28], the spontaneous emission rate (SER) is $\Gamma_f = \Gamma_v[n - (n^2 - 1)^{1/2}]$ [29], where n is the refractive index and Γ_v is the SER in vacuum. On the other

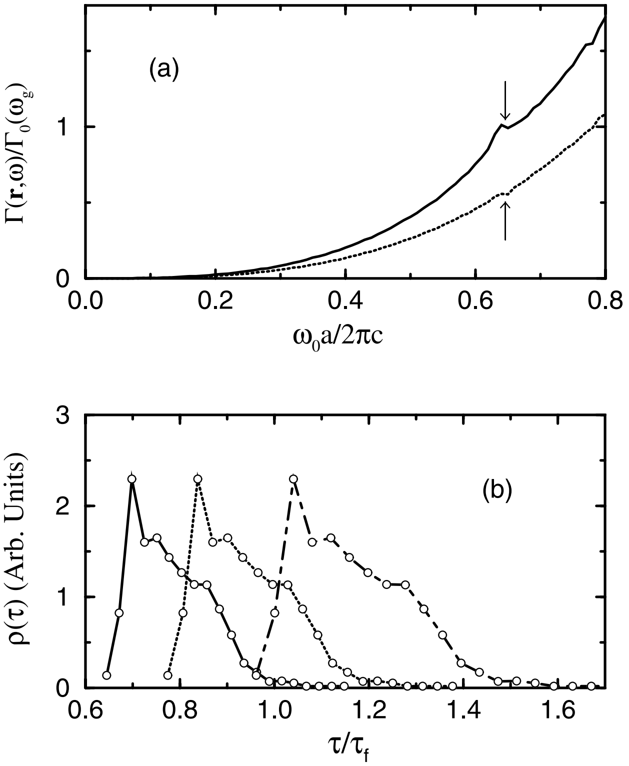

FIG. 1. (a) Radiative linewidth for two different atomic positions $\mathbf{r}_1 = (0.5, 0.5, 0.5)$ (solid line) and $\mathbf{r}_2 = (0.25, 0.25, 0.01)$ (dotted line) in the background medium of PC1. Center frequency ω_g of the pseudogap is indicated by arrows. (b) Decay lifetime distribution for the homogeneous dispersion of atoms over the background medium of PC1. The solid, dotted, and dashed lines correspond to $\tau_f = \tau_v$, $\tau_v/1.2$, and τ_v/n_b, respectively.

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 [5]. As a result, the value of Γ_f is between $0.385\Gamma_v$ and $1.49\Gamma_v$ for $n = n_b = 1.49$, 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. However, it is believed that the pure PC effects cannot cause these phenomena [7,19].

We now turn to study the PC2 sample. Figure 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.* [6]) 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. Figure 2(b) shows that 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 spherical surface leads to a very narrow lifetime distribution. In addition, when two atomic 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.

The lifetime distributions of the atoms in PC3 are displayed in Fig. 3. Figure 3(a) shows that the observable RWOLDs T_{rw} of atoms outside dielectric spheres are high up to 600% $\sim$ 700%, for three different transition frequencies ω_0, respectively, at the lower, center, and upper edges of the *pseudo*-PBG's. Compared with the chosen reference lifetime τ_0, the decay process for most of the atoms outside dielectric globules is remarkably accelerated. However, Fig. 3(b) reveals that the spontaneous emission is substantially inhibited when atoms are embedded on the spherical surface of radius $r = 0.6r_0$ inside the dielectric

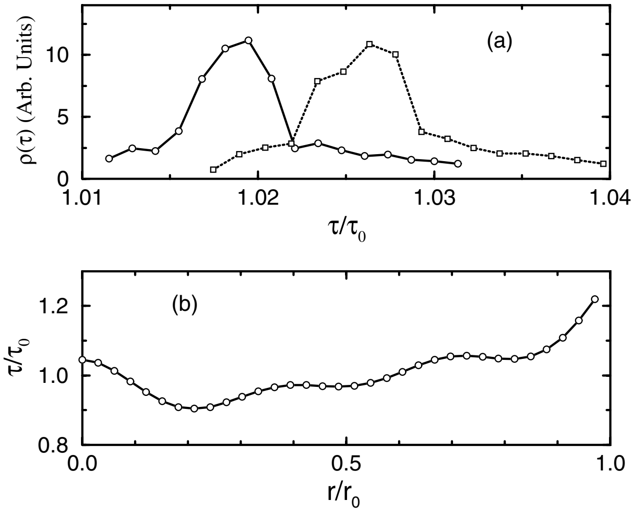

FIG. 2. (a) Lifetime distribution of atoms (molecules) located on two spherical surfaces with different radius r inside the dielectric globules of the radius being r_0 in the PC2, the solid line for $r = 0.6r_0$, and the dotted line for $r = 0.8r_0$. (b) Variation of the lifetime with the radius of the spherical surface along the x axes direction. τ_0 is the lifetime of the atoms in an infinite homogeneous medium with $n = 1.45$.

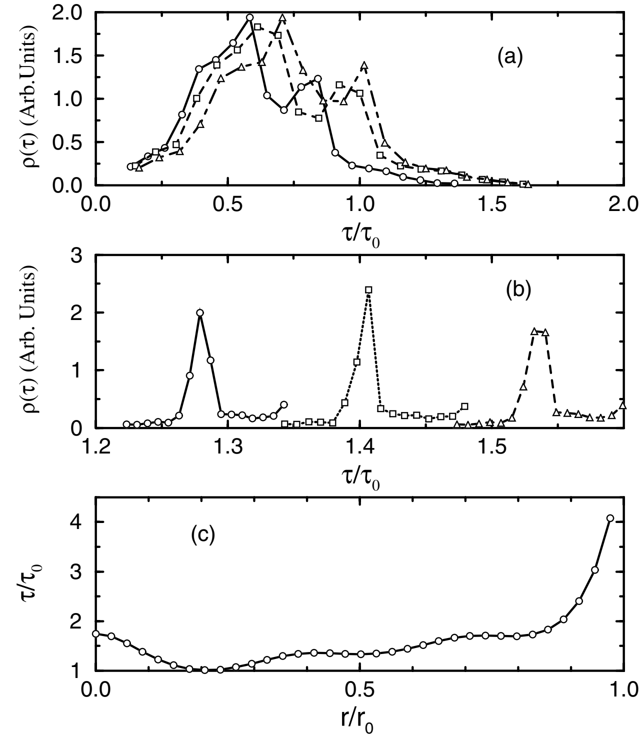

FIG. 3. Lifetime distribution of the atoms in PC3: (a) outside the dielectric spheres and (b) on the spherical surface with a radius of $r = 0.6r_0$ inside the dielectric globules. In (a) and (b), the solid, dotted, and dashed lines correspond to the transition frequency $\omega_0 = 0.387(2\pi c/a)$, $0.404(2\pi c/a)$, and $0.421(2\pi c/a)$, respectively. (c) Variation of the lifetime with the radius of the spherical surface along the x axes direction at $\omega_0 = 0.404(2\pi c/a)$. τ_0 is the lifetime of the atoms in an infinite homogeneous medium with $n = 2.6$.

spheres. We observe once again that the accelerated and inhibited decay processes of atoms simultaneously occurs. The observable RWOLDs T_{rw} for the atoms located on the spherical surface still remain narrower, less than 9%. Hence, the RDOL still approximately characterizes the decay kinetics of the atoms in the whole dielectric spheres. Figure 3(c) displays that the RWOLD inside the dielectric spheres is large, up to 400%. The results in Figs. 3(a)–3(c) demonstrate that the increase of index contrast rapidly broadens the lifetime distribution of atoms.

Based upon the lifetime distribution function, we have studied the decay kinetics of the spontaneous emission from an assembly of the atoms in the PC's with *pseudo*-PBG's. It is shown that the decay kinetics strongly depends 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 PC's. 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 recent reported experiments.

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[1] E. Yablonovitch, Phys. Rev. Lett. **58**, 2059 (1987).
[2] S. John, Phys. Rev. Lett. **58**, 2486 (1987).
[3] J. Martorell and N. M. Lawandy, Phys. Rev. Lett. **65**, 1877 (1990).
[4] B. Y. Tong *et al.,* J. Opt. Soc. Am. B **10**, 356 (1993).
[5] E. P. Petrov *et al.,* Phys. Rev. Lett. **81**, 77 (1998).
[6] M. Megens *et al.,* Phys. Rev. A **59**, 4727 (1999).
[7] M. Megens *et al.,* Phys. Rev. Lett. **83**, 5401 (1999); E. P. Petrov *et al.,* Phys. Rev. Lett. **83**, 5402 (1999).
[8] S. John and J. Wang, Phys. Rev. Lett. **64**, 2418 (1990); Phys. Rev. B **43**, 12 772 (1991).
[9] S. Bay, P. Lambropoulos, and K. Molmer, Phys. Rev. Lett. **79**, 2654 (1997); Phys. Rev. A **55**, 1485 (1997).
[10] S. John and T. Quang, Phys. Rev. A **50**, 1764 (1994); T. Quang and S. John, Phys. Rev. A **56**, 4273 (1997).
[11] S. Y. Zhu, H. Chen, and H. Huang, Phys. Rev. Lett. **79**, 205 (1997).
[12] T. Quang *et al.,* Phys. Rev. Lett. **79**, 5238 (1997).
[13] A. G. Kofman, G. Kurizki, and B. Sherman, J. Mod. Opt. **41**, 353 (1994).
[14] S. John and T. Quang, Phys. Rev. Lett. **74**, 3419 (1995); **76**, 1320 (1996).
[15] S. Y. Zhu *et al.,* Phys. Rev. Lett. **84**, 2136 (2000); Y. Yang and S. Y. Zhu, Phys. Rev. A **62**, 013805 (2000).
[16] I. Alvarado-Rpdriguez, P. Halevi, and A. S. Sanchez, Phys. Rev. E **63**, 056613 (2001).
[17] J. P. Dowling and C. M. Bowden, Phys. Rev. A **46**, 612 (1992).
[18] H. P. Urbach and G. L. J. A. Rikken, Phys. Rev. A **57**, 3913 (1998).
[19] Zhi-Yuan Li and Zhao-Qing Zhan, Phys. Rev. B **63**, 125 106 (2001); (private communication).
[20] Z. Y. Li, L. L. Lin, and Z. Q. Zhang, Phys. Rev. Lett. **84**, 4341 (2000); Z. Y. Li and Y. Xia, Phys. Rev. A **63**, 043817 (2001).
[21] T. Suzuki and P. K. L. Yu, J. Opt. Soc. Am. B **12**, 570 (1995).
[22] X. H. Wang, B. Y. Gu, and R. Z. Wang (to be published).
[23] K. M. Ho, C. T. Chan, and C. M. Soukoulis, Phys. Rev. Lett. **65**, 3152 (1990); K. M. Leung and Y. F. Liu, *ibid.* **65**, 2646 (1990); Z. Zhang and S. Satpathy, *ibid.* **65**, 2656 (1990); K. Busch and S. John, Phys. Rev. E **58**, 3896 (1998).
[24] C. Cohen-Tannoudji, J. Dupont-Poc, and G. Grynberg, *Atom-Photon Interactions: Basic Processes and Application* (John Wiley & Sons, New York, 1992); P. Lambropoulos, G. M. Nikolopoulos, T. R. Nielsen, and S. Bay, Rep. Prog. Phys. **63**, 455 (2000).
[25] R. J. Glauber and M. Lewenstein, Phys. Rev. A **43**, 467 (1991); G. L. J. A. Rikken and Y. A. R. R. Kessener, Phys. Rev. Lett. **74**, 880 (1995).
[26] R. Z. Wang, X. H. Wang, B. Y. Gu, and G. Z. Yang (unpublished).
[27] J. Callaway, *Quantum Theory of the Solid State* (Academic Press, Inc., New York, 1991).
[28] E. Yablonovitch, T. J. Gmitter, and R. Bhat, Phys. Rev. Lett. **61**, 2546 (1988).
[29] H. Khosravi and R. Loudon, Proc. R. Soc. London A **436**, 373 (1992).