Conservative form of the density of states of a photonic crystal with a pseudogap

Xiao-Dong Liu,[1,2] Yi-Quan Wang,[1] Bing-Ying Cheng,[1] and Dao-Zhong Zhang[1]
[1]*Optical Physics Laboratory, Institute of Physics, Chinese Academy of Sciences, Beijing 100080, China*
[2]*Department of Mechanical, Electric and Information Engineering, Dalian Nationality University, Dalian 116600, China*
(Received 3 February 2003; revised manuscript received 20 June 2003; published 18 September 2003)

We show that the total number of states in a photonic crystal in the entire allowed frequency regime will be conserved, and it is equal to that of its corresponding effective medium, i.e., if the density of states (DOS) has a valley(s) in some range(s) of frequencies, it must be compensated for by increases over some other range(s). This rule is of importance in developing a model pseudogap in order to describe the mean emission characteristics of the system when there is a collection of dependently emitting atoms or molecules with essentially random dipole orientations in a large volume and the spectrum of the active atoms is wide enough. This is because, with this rule, the states-conservative model always results in DOS-induced suppression, absolute enhancement, narrowing, spectrum splits, and redshift or blueshift of spontaneous-emission spectra.

DOI: 10.1103/PhysRevE.68.036610 PACS number(s): 42.70.Qs, 42.25.Fx, 42.50.Ct

I. INTRODUCTION

In recent years there has been great interest in spontaneous emission (SE) [1–6], Raman scattering [7,8], and other nonlinear effects [9–12] in photonic crystals (PC's). These research fields will lead to a multitude of potential applications in high-precision measurements, lasing without inversion, quantum computation, and quantum information theory. It is well known that spontaneous emission depends not only on the energy structure of an atom or molecule but also on the optical properties of the surrounding environment, more specifically, on the local density of states (DOS) of the radiation field [1–6]. Up to now, a few dispersion models near band edges have been extensively employed to study the SE problems in PC's with absolute photonic band gaps (PBG's) [2,3]. Obviously, to produce a full PBG typically requires a connected network of high-index material containing a periodic array of air voids or void regions made of a material with a dielectric constant that is sufficiently low relative to the first material; for example, a minimum dielectric ratio of 2.8 is required for a full PBG in an inverse opal structure. Clearly, PC's with complete PBG's are difficult to fabricate on the submicron length scale of the optical and near-infrared frequency range, although much progress has recently been made in fabricating three-dimensional (3D) PBG crystals at near-infrared wavelengths [13].

Recently, there has also been considerable experimental work on radiative emission from active materials embedded in optical PC's with photon propagation pseudogaps, i.e., materials that prohibit photon propagation in only certain directions [14–16]. In fact, many types of research do not require a complete photonic band gap but only a strong, or even not so strong, pseudogap. Thus the remaining problem is naturally how to develop a quantitative description and calculation of the interaction between an atom and the electromagnetic modes available in a PC.

Many studies show, as in Ref. [17], that the inhibition of spontaneous-emission rates (SER) for one range of transition frequencies tends to be accompanied by enhancement of the rates at other frequencies. It follows that any reduction in SER over some range of frequencies must, necessarily, be compensated by increases over some other range of transition frequencies. This is the sum rule for the modified SER, as called by Barnett and Loudon [17].

In this paper we demonstrate, for the first time to our knowledge, that the states-conservation rule exists over the whole frequency regime, and so we offer a states-conservation theorem within a modified frequency regime. We will also investigate applications of the rule and the theorem in expressing analytically the gap structure, especially the pseudogap, within the modified frequency regime.

II. CONSERVATIVE RULE OF THE DENSITY OF STATES

As is well known, the photon DOS in a three-dimensional PC is defined by [18]

$$\rho(\omega)=\frac{2V}{(2\pi)^3}\sum_n \int_{1\mathrm{BZ}} d^3k\ \delta(\omega-\omega_{n,\mathbf{k}}), \tag{1}$$

where V is the sample volume in three-dimensional space, $\omega_{n,\mathbf{k}}$ is the photon dispersion relation in the PC, and 1BZ means the first Brillouin zone of the PC. The factor 2 before V in Eq. (1) stands for the two polarizations that are degenerate in energy.

According to the physical meaning of the DOS in a system, which is the number of allowed states per unit increase in ω, the total number of states in the system may be the integration of DOS over all the allowed frequency regime $\omega\in[0,\omega_M]$, where ω_M is large enough and far away from the modified regime of interest but not so large as to maintain the effective refractive index of the system n_{eff}, i.e., the average refractive index that is close to the value calculated according to Maxwell-Garnett theory is constant. This choice of ω_M is similar to that of John [1] and Bush [19], which is chosen to be approximately equal to the electron Compton frequency $\omega_M=mc^2$ that probes the relativistic aspects of the electron wave packet, and remains independent of its precise value in the scope of our analysis. Therefore, the total number of states in the system below the truncated frequency (or its responding wave vector) is

 ©2003 The American Physical Society

$$\int_0^{\omega_M}\rho(\omega)d\omega=\frac{2V}{(2\pi)^3}\sum_n\int_{1\mathrm{BZ}}d^3k\int_0^{\omega_M}d\omega\,\delta(\omega-\omega_{n,\mathbf{k}}). \tag{2}$$

It is of the utmost importance to note that, according to the equation

$$\int_{-\infty}^{+\infty}f(y)\,\delta(x-g(y))dx\equiv f(y), \tag{3}$$

no matter what form the dispersion curve $\omega_{n,\mathbf{k}}$ has, the following integration is always valid:

$$\int_0^{\omega_M}\rho(\omega)d\omega=\frac{2V}{(2\pi)^3}\sum_n\int_{1\mathrm{BZ}}d^3k=2MN. \tag{4}$$

Here, M is the truncated number of photonic bands corresponding to the truncated frequency ω_M, N is the number of unit cells, and the well-known relation in solid-state physics that the product of the volume of a unit cell and the volume of the first Brillouin zone equals $(2\pi)^3$ for all time is used. On the other hand, for its effective medium, the integral DOS truncated at frequency ω_M becomes

$$\frac{2V}{(2\pi)^3}\sum_n\int_{1\mathrm{BZ}}d^3k=\frac{Vn_{\mathrm{eff}}^3\omega_M^3}{3\pi^2}\equiv\int_0^{\omega_M}\rho_0(\omega)d\omega, \tag{5}$$

where $\rho_0(\omega)=Vn_{\mathrm{eff}}^3\omega^2\pi^{-2}$ is the density of states in the effective medium. In this paper, we let the light velocity in vacuum and Dirac constant $\hbar=c=1$. Therefore, we call Eq. (4) or Eq. (5) the rule of total states conservation, which says that the total number of states in a PC, in the entire allowed frequency regime, will always be conservative and will equal that of the corresponding effective medium. The states-conservation phenomenon of the PC DOS can always be seen in almost every theoretical outcome calculated according to realizable PC's [5] and in experimental results [16] as well.

III. APPLICATION

For the problem of the spontaneous emission of a specific active atom or molecule, model DOS's are generally invalid to account for quantum optical phenomena of this atom or molecule in 3D photonic crystals, and the following projected local density of states (PLDOS), which is proportional to the experimentally observable decay constant of an atom or a molecule [4], should be used [6]:

$$\rho_l(\mathbf{d},\mathbf{r}_0,\omega)=\sum_n\int_{1\mathrm{BZ}}d^3k\;\delta(\omega-\omega_{n,\mathbf{k}})|\mathbf{d}\cdot\mathbf{E}_{n,\mathbf{k}}(\mathbf{r}_0)|^2, \tag{6}$$

where $\mathbf{d}$ and $\mathbf{r}_0$ are, respectively, the direction unit vector of the dipole matrix element for the atomic or molecule transition and the position of the atom or molecule. However, if there is a collection of dependently emitting atoms or molecules with essentially random dipole orientations in a small volume around $\mathbf{r}_0$, in order to describe the mean emission characteristics of the system, we average over all solid angles. Hence the local density of states (LDOS) is defined as [6]

$$\rho_l(\mathbf{r}_0,\omega)=\sum_n\int_{1\mathrm{BZ}}d^3k\;\delta(\omega-\omega_{n,\mathbf{k}})|E_{n,\mathbf{k}}(\mathbf{r}_0)|^2. \tag{7}$$

The relation between the LDOS and the total DOS is given by the expression [6]

$$\rho(\omega)=\frac{1}{V_{\mathrm{WSC}}}\int_{\mathrm{WSC}}d\mathbf{r}\,\varepsilon(\mathbf{r})\rho_l(\mathbf{r},\omega), \tag{8}$$

where WSC and V_{WSC} stand for, respectively, the Wigner-Seitz unit cell and its volume, and $\varepsilon(\mathbf{r})$ denotes the position-dependent dielectric function of the PC. The above equation shows that for a small dielectric modulation in the photonic crystal, which implies a weak interaction between the dielectric and the electromagnetic field, the total DOS can provide a reasonable description of the field at any point in the photonic crystal [6]. Clearly, a crystal exhibiting a strong pseudogap or a full photonic band gap does not satisfy such a condition. Therefore, we can say that a model total DOS can be used in the problem of the spontaneous emission of a collection of dependently emitting atoms or molecules with essentially random dipole orientations in a small volume (much less than a unit cell of the PC) with a small dielectric modulation (a dielectric contrast less than 1.31 may be appropriate [5]), i.e., the photonic crystal has a photonic pseudogap.

Many authors have investigated the emission features while assuming forms of the DOS that exhibit only an inverse Lorentzian-like or inverse Gaussian pseudogap [20,21,6]. If we abide by the rule of total states conservation, the following forms will be appropriate:

$$\rho_L(\omega)=\rho_0(\omega)[1+aL(\omega,\omega_{c1},\Gamma_1)-bL(\omega,\omega_{c2},\Gamma_2)]. \tag{9}$$

or

$$\rho_G(\omega)=\rho_0(\omega)[1+aG(\omega,\omega_{c1},\Gamma_1)-bG(\omega,\omega_{c2},\Gamma_2)]. \tag{10}$$

For the situation when a peak, which we call the pseudoedge, and a pseudogap are expected, where $L(\omega,\omega_c,\Gamma)$ and $G(\omega,\omega_c,\Gamma)$ are, respectively, Lorentzian and Gaussian functions,

$$L(\omega,\omega_c,\Gamma)=\frac{\Gamma^2}{(\omega-\omega_c)^2+\Gamma^2}, \tag{11}$$

$$G(\omega,\omega_c,\Gamma)=\exp\left[-\left(\frac{\omega-\omega_c}{\Gamma}\right)^2\right]. \tag{12}$$

Parameters a, Γ_1, and ω_{c1}, respectively, describe the depth, width, and central frequency of the pseudogap; b, Γ_2, and ω_{c2}, respectively, are the parameters describing the height, width, and central frequency of the pseudoedge of the DOS.

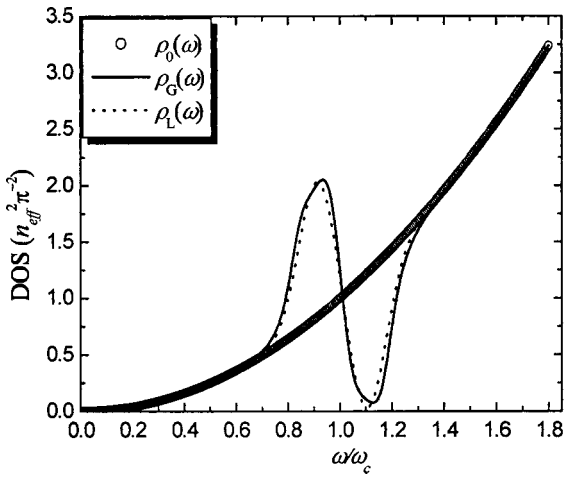

FIG. 1. States-conservative model DOS's: Gaussian-like $\rho_G(\omega)$ described by Eq. (10) with $b=1.491$, $\omega_{c1,2}=1\pm0.1$, $\Gamma=0.1$, $a=1$ (solid line), Lorentzian-like $\rho_L(\omega)$ described by Eq. (9) with $b=1.488$, $\omega_{c1,2}=1\pm0.1$, $\Gamma=0.1$, $a=1$ (dotted line), and $\rho_0(\omega)$ of effective medium (open circles).

In the equations above, a and b are dimensionless, and $\Gamma_{1,2}$ and $\omega_{c1,2}$ are in units of ω_0 that is the central frequency of the DOS's modified region of interest, not the central frequency of the valley or the peak of the DOS. Both of these DOS's have the features that they approach the free space DOS far away from ω_c, i.e., $\rho(0)=0$, $\rho(\omega\gg\omega_c)=n_{\mathrm{eff}}^3\omega^2\pi^{-2}$. The parameters in Eqs. (9) and (10) should be determined by the condition

$$\int_0^\infty [\rho(\omega)-\rho_0(\omega)]d\omega=0. \tag{13}$$

For example, $b=1.488a$ when $\omega_{c1,2}=1\pm0.1$, $a<1.06$, and $\Gamma=0.1$ for a Lorentzian-like DOS $\rho_L(\omega)$ in Eq. (9); $b=1.491a$ when $\omega_{c1,2}=1\pm0.1$, $a<1.025$, and $\Gamma=0.1$ for a Gaussian-like DOS $\rho_G(\omega)$ in Eq. (10). Here, it must be noted that the model DOS's in this paper have a strong pseudogap in order to illustrate our points boldly. A typical bandwidth of fluorescence spectra of most efficient organic fluorescence materials is $\Gamma=0.1$, although Γ is much smaller for most single atoms and molecules. Figure 1 shows these two model DOS's using $a=1$.

From Fig. 1, we can see that both forms of the DOS that exhibits an inversed Lorentzian pseudogap and a Gaussian pseudogap are similar to each other. It must be noted that $L(\omega,\omega_c,\Gamma)$ can be used in some cases for simplicity, but this is not square integrable, so we adopt the Gaussian-like pseudogaps because they cannot lead to any qualitative distinctions.

Among several models of SE of a two-level atom, we adopt the theory developed by Woldeyohannes *et al.* [18], which is a theory for a three-level atom, and let the two lower levels $|1\rangle$ and $|0\rangle$ be equal to one $|1\rangle$. We also adopt the authors' symbol system [18] to describe our two-level atom. That is, $|2\rangle$ is the excited level, $|1\rangle$ is the ground state, and γ_{21} is the radiation linewidth, which can be as wide as 10%, like that of the photoluminescence spectrum or Raman spectrum. The transition frequency that has considered the Lamb shift is represented by ω_{c1} and is in units of ω_0. Then the emission spectrum will be

$$S(\omega)\propto\frac{\gamma_{21}^2 z^2(\omega)}{(\omega-\omega_{21})^2+\gamma_{21}^2}, \tag{14}$$

where $z(\omega)$ is the "atom form factor" expressed in terms of the photon DOS $\rho(\omega)$. Given a model DOS $\rho(\omega)$, we can deduce

$$z(\omega)=\frac{dk(\omega)}{d\omega}=(6\pi^2)^{1/3}\rho(\omega)\left[\int_0^\omega\rho(\omega')d\omega'\right]^{2/3}. \tag{15}$$

For simplicity, we study the enhancement and suppression of SE of a two-level atom in a PC with the model DOS expressed by Eq. (10) using the parameter $b=1.488$ for $\omega_{c1,2}=1\pm0.1$, $a=1$, and $\Gamma=0.1$ (see Fig. 1). It should be noted that the central frequency of the pseudogap is, more accurately, $1.105\omega_0$, which is numerically the zero point of the DOS.

As has been shown in the experimental investigation [16], either enhancement or inhibition of the spontaneous decay of a molecule with a wide spontaneous-emission spectrum can be observed in a photonic crystal, depending on the mutual position of the photonic band gap/pseudogap and fluorescence spectrum. Usually, ω_{21} is situated around ω_{c1}, and δ_λ is the detuning frequency of ω_{21} from the frequency ω_{21}, i.e., $\delta_\lambda=\omega_{21}-\omega_{c1}$.

Figures 2(a) and 2(b) show the plots of spontaneous-emission spectra $S(\omega)$ of two kinds of two-level atoms in a PC with this model DOS for the cases of $\delta_\lambda=-0.1$ and 0, respectively. Each of them is accompanied by a standard Lorentzian spectrum centered at its corresponding radiation frequency $1+\delta_\lambda$. From Fig. 2, one can see that significant enhancement and suppression do indeed occur at the frequencies that correspond to the DOS peak and valley, respectively, with complete suppression at the zero-DOS frequency, which is in agreement with the Fermi golden rule. Especially if the radiation transition frequency $\omega_{c1,2}=1+\delta_\lambda$ is situated near the low-frequency side of this model pseudogap, both significant enhancement and suppression result in a dark line at the zero-DOS frequency ω_{c1}, with a clear redshift of the maximum frequency and a significant narrowing of the emission spectrum. Similarly, if the DOS has a pseudoedge situated at the high-frequency side of its pseudogap, and the radiation transition frequency $\omega_{c1,2}=1+\delta_\lambda$ is also situated near the high-frequency side, they will result in a clear blueshift. The redshift has just been detected by Koenderink *et al.* [16] but the blueshift has not yet been detected.

IV. DISCUSSION

As pointed out by Li *et al.* [22], although the DOS of a PC exhibits significant oscillation over a wide range, on average it follows that of the effective medium, i.e., $\rho(\omega)\sim\omega^2$ in three dimensions, and this square DOS will persist

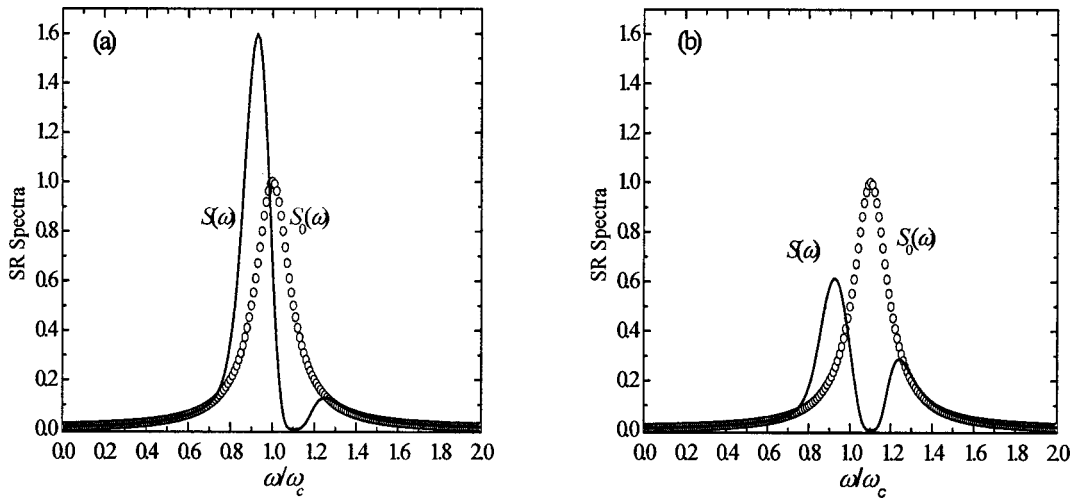

FIG. 2. Plots of SE spectra $S(\omega)$ of two kinds of two-level atoms in a PC with the model DOS described by $\rho_G(\omega)$ of Fig. 1. In (a), $\delta_\lambda = -0.1$, and in (b), $\delta_\lambda = 0$. Each of them is accompanied by a standard Lorentzian spectrum $S_0(\omega)$ centered at its corresponding radiation frequency $1+\delta_\lambda$.

better in higher-energy regions. From Fig. 8 of the paper of Li *et al.* [4], we can observe that the parabolic curves of the DOS of the corresponding effective media with an average dielectric constant depart slightly from the average curves of the DOS calculated for their special inverse-opal structure. This is due not to the error of our theory and Li's main idea but to the positive departure from Maxwell-Garnett effective medium theory at high volume fraction of the dispersed scatterers. It occurs because the DOS is proportional to n_{eff}^3 in three dimensions, and a slight departure of n_{eff} can lead to a significant change of the average DOS. On the other hand, one can obtain an exact value of the average dielectric constant of the medium through fitting the average curves of the DOS calculated for some special dielectric structure.

In fact, in any actual PC there are a number of defects that may reduce all resonances present having over the SE. But if these unexpected random imperfections that plague any experimental structure are much tinier than the radiation wavelength, their effects are small on the band structure and may tend to smooth the change of the band microstructure and therefore smooth the change of the decay rate and emission spectrum. This smoothing effect is expected to be similar to the effect of absorption in the sample [23,24] and not-so-accurate spectrum measurement, which has been observed in almost every experiment reported and agrees with a wide-band SE model and slowly varying photonic band-structure model.

Theoretical enhancement and suppression of spontaneous emission are significant, but they are not very easy to observe because these results reflect the effects from throughout the whole PC and are the average of all directions, although, strictly speaking, enhancement and suppression of spontaneous emission depend on local DOS. This can be detected by using integral-sphere technology. In a given direction, the SE spectrum is usually different from the others, i.e., angle-dependent just as most of the reported experiments detected [16]. Therefore, it is not surprising that a possible phenomenon may appear wherein the emission is enhanced in some frequencies according to the above calculations, and relatively suppressed emission occurs in these frequencies in some directions. Perhaps there are some directions in which the detected spectra may be representative of the angle-averaged total emission spectrum.

V. CONCLUSION

In conclusion, we have shown that the total number of states in a PC in the entire allowed frequency regime will be conserved and will be equal to that in the corresponding effective medium. From the conservation rule, we have deduced the theorem of states conservation within a modified frequency regime in PC's with pseudogaps, which says that if a valley of the DOS appears in some range of frequencies, it must necessarily be compensated for by increases over some other range of frequencies. This theorem has an application in developing a model DOS for a pseudogap of a PC because not every study in the literature gives a correct pseudogap model of the DOS. Assuming a states-conservative DOS, we have naturally obtained a DOS-induced suppression and enhancement along with a redshift or blueshift of spontaneous-emission spectra. Here, the enhancement of the spontaneous-emission spectrum is an absolute one relative to the emission intensity in a vacuum at corresponding frequencies of the pseudoedge, but not relative to that at the frequencies of the pseudogap. The absolute enhancement and redshift or blueshift of spontaneous-emission spectra are the only new effects predicted by our states-conservative DOS model. Surely, as mentioned above, our analyses should be used in the problem of the spontaneous emission of a collection of dependently emitting atoms or molecules with essentially random dipole orientations in a small volume of a photonic crystal with a small dielectric modulation, i.e., the photonic crystal has a photonic pseudogap.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge fruitful discussions with Professor B. Y. Gu, Professor L. A. Wu, and Dr. J. L. Wu at the Institute of Physics, Chinese Academy of Sciences. This work is supported by the National Key Basic Research Special Foundation of China under Grant No. 2001CB610402 and the National Nature Sciences Foundation of China under Grant No. 60078007.

[1] S. John and J. Wang, Phys. Rev. Lett. **64**, 2418 (1990).
[2] S. Y. Zhu, Y. P. Yang, H. Chen, H. Zheng, and M. S. Zubairy, Phys. Rev. Lett. **84**, 2136 (2000).
[3] H. Z. Zhang, S. H. Tang, P. Dong, and J. He, Phys. Rev. A **65**, 063802 (2002).
[4] Z. Y. Li, L. L. Lin, and Z. Q. Zhang, Phys. Rev. Lett. **84**, 4341 (2000).
[5] X. H. Wang, R. Z. Wang, B. Y. Gu, and G. Z. Yang, Phys. Rev. Lett. **88**, 093902 (2002).
[6] N. Vats, S. John, and K. Bush, Phys. Rev. A **65**, 043808 (2002).
[7] H. G. Winful and V. Perlin, Phys. Rev. Lett. **84**, 3586 (2000).
[8] S. V. Gaponenko, Phys. Rev. B **65**, 140303 (2002).
[9] S. F. Mingaleev and Y. S. Kivshar, Phys. Rev. Lett. **86**, 5474 (2001).
[10] C. M. Bowden and A. M. Zheltikov, J. Opt. Soc. Am. B **19**, 2046 (2002).
[11] M. Soljacic, S. G. Johnson, S. H. Fan, M. Ibanescu, E. Ippen, and J. D. Joannopoulos, J. Opt. Soc. Am. B **19**, 2052 (2002).
[12] J. Martorell, J. Opt. Soc. Am. B **19**, 2075 (2002).
[13] S. Noda, K. Tomoda, N. Yamamoto, and A. Chutinan, Science **289**, 604 (2000).
[14] J. Martorell and N. M. Lawandy, Phys. Rev. Lett. **65**, 1877 (1990).
[15] E. P. Petrov, V. N. Bogomolov, I. I. Kalosha, and S. V. Gaponenko, Phys. Rev. Lett. **81**, 77 (1998); M. Megens, J. E. G. J. Wijnhoven, A. Lagendijk, and W. L. Vos, Phys. Rev. A **59**, 4727 (1999); H. P. Schriemer, H. M. van Driel, A. F. Koenderink, and W. L. Vos, *ibid.* **63**, 011801 (2000); S. G. Romanov, T. Maka, C. M. S. Torres, M. Muller, and R. Zentel, J. Appl. Phys. **91**, 9426 (2002).
[16] A. F. Koenderink, L. Bechger, H. P. Schriemer, Ad Lagendijk, and W. L. Vos, Phys. Rev. Lett. **88**, 143903 (2002).
[17] S. M. Barnett and R. Loudon, Phys. Rev. Lett. **77**, 2444 (1996).
[18] M. Woldeyohannes, S. John, and V. I. Rupasov, Phys. Rev. A **63**, 013814 (2000).
[19] K. Busch, N. Vats, S. John, and B. Sanders, Phys. Rev. E **62**, 4251 (2000).
[20] G. M. Nikolopoulos and P. Lambropoulos, Phys. Rev. A **61**, 053812 (2000).
[21] P. Lambropoulos, G. M. Nikolopoulos, T. R. Nielsen, and S. Bay, Rep. Prog. Phys. **63**, 455 (2000).
[22] Z. Y. Li and Y. N. Xia, Phys. Rev. A **63**, 043817 (2001).
[23] H. T. Dung, L. Knoll, and D. G. Welsch, Phys. Rev. A **67**, 021801 (2003).
[24] Z. L. Wang, C. T. Chan, W. Y. Zhang, Z. Chen, N. B. Ming, and P. Sheng, Phys. Rev. E **67**, 016612 (2003).