Density of resonant states and a manifestation of photonic band structure in small clusters of spherical particles

Alexey Yamilov* and Hui Cao
Department of Physics and Astronomy, Northwestern University, Evanston, Illinois 60208, USA
(Received 25 October 2002; revised manuscript received 12 June 2003; published 25 August 2003)

We introduce a numerical recipe for calculating the density of the resonant states of the clusters of dielectric spheres. Using truncated multipole expansions (generalized multisphere Mie solution) we obtain the scattering matrix of the problem. By introducing an infinitesimal absorption in the spheres we express the dwell time of the electromagnetic wave in terms of the elements of the scattering matrix. Using the parameters in recent light localization experiments [Phys. Rev. Lett. **87**, 153901 (2001)], we demonstrate that the density of the resonant states, related to the dwell time, shows the formation of the photonic band structure in small clusters of dielectric spheres as the small as five particles. Density of resonant states of a cluster of 32 spheres exhibits a well defined structure similar to the density of electromagnetic states of the infinite photonic crystal. Our results suggest that, due to the formation of small ordered clusters, a significant modification of the density of electromagnetic states can occur in a random collection of monodisperse spheres.

DOI: 10.1103/PhysRevB.68.085111 PACS number(s): 42.70.Qs, 42.25.Bs, 42.68.Mj

The idea of employing photonic crystals, structures with a periodically modulated dielectric constant, to manipulate the density of states (DOS) of electromagnetic (EM) waves[1,2] led to an explosion of interest, both academic and practical.[3,4] In a photonic crystal, in a given crystallographic direction, light can propagate only for frequencies within photonic bands, described by a dispersion relation $\omega=\omega(\mathbf{k})$, similar to electron de Broglie waves in crystals. If the gaps between the bands overlap for all directions in the crystal, the photonic crystals possesses a complete photonic band gap.[5] For frequencies inside the complete gap, the density of EM states turns to zero, which leads to new phenomena such as the suppression of spontaneous emission, light localization, zero-threshold lasers, all-optical transistors and circuitry, and anomalous nonlinear properties.[1–4,6] For theoretical considerations, the size of the crystal is usually assumed infinite. In practice, however, one always deals with finite structures. Naturally, a question arises. How large should a photonic crystal be, in order to exhibit photonic band structure? In this paper, we show that the modification of the density of EM states may begin for a cluster of five spheres. This suggests that scattering of light by ordered aggregates of spheres can be important in the problem of light propagation in dense random media. Indeed, an ensemble of monodispersed resonant scatterers (e.g., spheres) is considered one of the most favorable[7] for satisfying Ioffe-Regel criterion for Anderson localization $kl_{\mathrm{scat}}\lesssim 1$,[6–11] where k is the wave number in the medium and l_{scat} is the scattering length. Closely packed face centered cubic (fcc) arrangement often appear in self-assembled structures and it is shown to have lowest free energy. This fact points to a high probability of formation of ordered clusters in a collection of spheres. Even though the complete photonic gap can never be realized in an fcc structure of dielectric spheres in air, for some parameters, the total density of EM states can be significantly suppressed in certain frequency regions. Our results suggest that such suppression can occur due to the presence of the clusters even without long range order.

Treatment of photonic structures, with all three dimensions of the order of the wavelength has been a challenge. When the distance between scatterers is comparable to their size, analytical methods, such as single scattering approximation, dipole approximation become invalid.[7] Numerical methods tailored for calculation of transmission through a slab[12,13] cannot be applied because it requires the lateral size of the slab to be larger than all characteristic lengths. The finite difference time domain (FDTD) method is used extensively in photonics design.[14] Obtaining the density of states with the FDTD-based ONYX method[13] requires a time-dependent solution. Combined with dense $\lambda/20$ spacial grids, it makes the problem computationally demanding in 3D, even for the smallest structure considered in this paper.

For small truncated photonic crystal structures, scattering language becomes appropriate. It has been proposed to use multipole expansions as a means of simplifying the numerical solution of the electromagnetic problem.[15–17] In Ref. 17, 2D Green's function was calculated in terms of multipole coefficients, which, in turn, allowed the calculation of the local DOS. In our paper we calculate the total density of resonant states (DORS) of the entire cluster in 3D, with the help of the generalized multisphere Mie (GMM) solution.[18] The obtained quantity corresponds to the local density of EM states integrated over the volume of all scatterers. First, we obtain the elements of the scattering matrix for the collection of dielectric spheres. Then, the density of resonant EM states of the cluster is expressed in terms of the scattering matrix. Within this approach, one only needs to evaluate vector-spherical-function expansion coefficients, there is no need, as in the FDTD method, to find EM fields at every spacial point. The method requires considerably less memory and computation time. An important class of photonic crystals, opals, can be treated in this framework as an example. We should note that the maximum number of particles, which can be treated numerically decreases with the increase of the size parameter $x=k\cdot r$ and refractive index. Here k denotes the wave number and r is the sphere radius.

Recently, the propagation of microwaves through a collection of spherical scatterers was studied in Ref. 9, where

light localization has been observed. To illustrate our approach we will use the parameters from this reference. We demonstrate that in the considered system: (i) the signatures of the photonic band structure can appear for aggregates as small as five particles, (ii) in a cluster of 32 particles the density of (resonant) states can show strong resemblance to that of an infinite structure, with a pronounced depletion in the region of pseudogap, (iii) the photonic band structure of the finite clusters is tolerable to a certain degree of random displacements of the particles off their lattice sites, and (iv) the region with depleted density of states coincides with the region where the light localization in Ref. 9 was observed. The latter suggests that the localization may have been facilitated by the photonic bandgap effect due to short range ordering in the collection of spherical scatterers.

The cluster is an open system that does not support stable modes, quasistates of the EM field are leaky, and the density of the states cannot be specified. For the related problem of electrons[19] in a stochastic cavity with an opening the Wigner delay time

$$\hat{\tau}_w(E) = -i\hbar \frac{\partial \ln(\det \hat{\mathbf{S}})}{\partial E} = -i\hbar \,\mathrm{tr}\, \hat{\mathbf{S}}^+ \frac{\partial \hat{\mathbf{S}}}{\partial E}, \tag{1}$$

expressed in terms of scattering matrix $\hat{\mathbf{S}}$, can be shown to be proportional to the density of resonant modes[19–21]

$$\tau_w(E) \propto \sum_j \frac{\gamma_j/2}{(E-E_j)^2 + (\gamma_j/2)^2}, \tag{2}$$

where E_j and γ_j are the energy and the linewidth of the jth resonance. tr in Eq. (1) represents the trace operation. For an infinite system, the poles of the scattering matrix $\hat{\mathbf{S}}$ approach the real energy axis and the stationary modes are formed, γ_j becomes zero.[19] Then one can define a mode counting function $N(E_1,E_2) = \int_{E_1}^{E_2} \tau_w(E)dE$, which represents the integral density of states in the energy interval (E_1,E_2). Analytic property of the scattering matrix allows one to obtain a convenient expression for the delay time in Eq. (1).[7,19,22,23] Following Ref. 23 we introduce a weak absorption α in the scattering region, then perturbatively

$$\hat{\mathbf{S}}(E+i\alpha) \simeq \hat{\mathbf{S}}(E)\left(1 + i\alpha \hat{\mathbf{S}}^+ \frac{\partial \hat{\mathbf{S}}}{\partial E}\right). \tag{3}$$

This leads to a simple expression for the time delay matrix

$$\hat{\tau}_w(E) = -i\hbar \lim_{\alpha\to\infty} \mathrm{tr}\, \hat{\mathbf{S}}^+ \frac{\mathrm{Im}\, \hat{\mathbf{S}}(E+i\alpha)}{\alpha}, \tag{4}$$

which does not contain the energy derivative.

Implementing the above procedure for a chaotic cavity is straightforward.[23] However, it is not so for the ensemble of scatterers we would like to consider. At this point we return to the original expression in Eq. (1). For EM waves, one can define dwell time τ_d which closely follows EM version of the Wigner time, giving asymptotically the same result on the resonances.[7] Dwell time corresponding to the time spent inside the scatterers can be readily found in terms of scattering coefficients as follows:

$$\tau_d(\omega) = \frac{\sum_i^N \int_{S_i} W(\mathbf{r},\omega)dV}{c\sigma_{\mathrm{scat}}(\omega)}, \tag{5}$$

where $W(\mathbf{r},\omega) = n^2(\mathbf{r},\omega)E^2(\mathbf{r},\omega)/E_0^2$ is electromagnetic energy density normalized so that it is 1 for the incident plane wave (in vacuum), σ_{scat} is the total scattering cross section, and c is speed of light. The integral in Eq. (5) is taken over the volume of each scatterer S_i. The nominator of the above expression for τ_d has dimensionality of the volume, so that the total expression has a unit of r/c. When multiplied by the frequency ω the time given by Eq. (5) at the position of a resonance coincides with its quality factor Q. The expression for the dwell time can be understood physically as a ratio between the energy stored in a scatterer divided by outgoing energy current.

Strictly speaking the time measured by Eq. (5) is the time spent inside scatterers comprising the cluster. In general the multipole solution of the scattering problem would allow one to obtain the energy density even in between the scatterers. This, however, is not possible for the computer code[18] that we used in our numerical simulations. The dwell time given by Eq. (5) is nevertheless of direct physical importance for the random lasers with gain concentrated in the particles.[24]

The dwell time defined by Eq. (5) can be found by using the trick used in Eq. (4). Indeed $\int_{S_i} W(\mathbf{r},\omega)d\mathbf{r}$ can be found by introducing an infinitesimally small absorption in the refractive index of the spheres $n+i\kappa$[7]

$$\int_{S_i} W(\mathbf{r},\omega) \simeq \left(\frac{4\pi}{3} r_i^3\right) \frac{3}{8} n \lim_{\kappa\to 0} \frac{Q_{\mathrm{abs}\,i}}{x_i \kappa}, \tag{6}$$

where $Q_{\mathrm{abs}\,i} = \sigma_{\mathrm{abs}\,i}/(\pi r_i^2)$ is the absorption efficiency, and $x_i = 2\pi r_i/\lambda$ is size parameter of the ith sphere. This approach is similar to using the "nonunitary clock"[22] to measure the time spent by the light inside a one-dimensional chain. The problem of finding DORS for the finite sized cluster is now reduced to finding the absorption and scattering cross sections $\sigma_{\mathrm{abs}\,i}, \sigma_{\mathrm{scat}}$. This step can be done by evaluating the expansion series of GMM solution.[18] The obtained dwell times depends on the particular orientation of the cluster with respect to the incident plane wave. To obtain a measure of the total density of resonant states τ_d^{tot}, expression (5) has to be averaged over different orientations.

As was already mentioned we used the parameters of Ref. 9 to illustrate our approach. We considered the clusters of 5 and 32 spheres, with diameter 0.95 cm and refractive index $n=3.14$. For both clusters we assumed fcc ordering with the distance between nearest neighbors of 1.9 cm. Smaller cluster is comprised of three planar layers perpendicular to the 111 direction, with 1, 3, and 1 sphere in each layer as shown on Fig. 1(a). In the larger cluster, particles were arranged in 5 layers with 3, 7, 12, 7, and 3 spheres in each layer, with the same stacking as in the first cluster [Fig. 1(b)]. The system

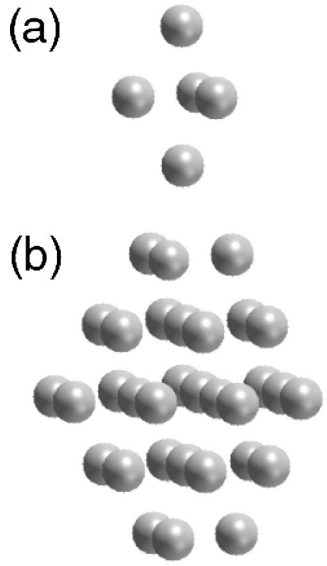

FIG. 1. The arrangement of spheres in (a) 5- and (b) 32-particle ordered clusters.

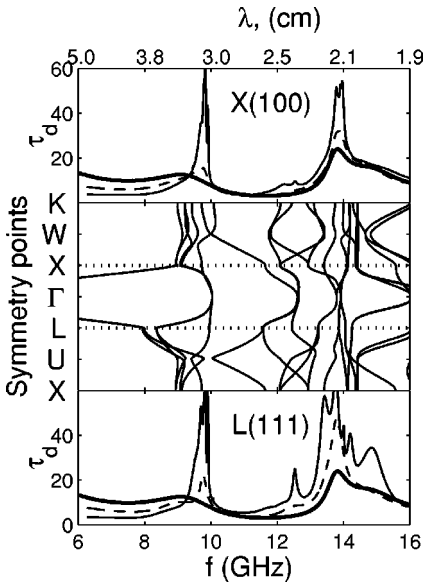

FIG. 2. The photonic crystal band structure (middle) is compared to the dwell times obtained for scattering from a single sphere (bold solid line), a cluster of five spheres (dashed line), and a cluster of 32 spheres (thin solid line). The upper graph corresponds to EM plane wave incoming in the 100 (X) direction, and the lower graph to the—111 (L) crystallographic direction.

considered here should be contrasted from the slab geometry (infinite planes), which has a quasi-1D nature.

The choice of these clusters was motivated for the following reasons. The shape of the clusters was chosen to be spherical. Such clusters have the smallest ''surface'' area (as compared, e.g., to the particles arranged in a planar layer), and it would correspond to the least free energy (the highest packing) configuration of the particles in a random medium. Moreover, to fully demonstrate the flexibility of our method we will consider the effect of disorder on the optical properties of the clusters. As mentioned above, to determine the density of resonant states one needs to average over angular orientations of the clusters. When the size of the cluster increases the angular size of a resonance should become smaller, which will require more orientations to be taken in the average. When performing the angular average for an ordered cluster one can take advantage of the symmetry of the cluster. For the chosen clusters nine symmetrically nonequivalent orientations (altogether 108 directions, accounting for symmetry) sufficed to get the convergence. Such symmetry reduction is not possible however in for disordered clusters, so one needs to take all 108 orientations into account. Since we also take average over 10 disorder realizations, the computation time limited the maximum size of the cluster to 32 spheres.

One characteristic property of a photonic crystal is spacial dispersion $\omega(\mathbf{k})$. On Fig. 2 we compare dispersion curves of the infinite photonic crystal, obtained using MIT photonic bands code,[25] to the dwell times for two clusters, described above, calculated for two different incident angles corresponding to X and L crystallographic directions. It can be easily seen that even for smaller cluster of five particles τ_d shows the formation of the resonant modes at the positions of photonic modes in the L direction. For the cluster of 32 particles, a well defined mode structure appears in both directions. However, it is more pronounced for the L direction, where six resonant states between 12 and 16 GHz can be traced to L modes of the infinite photonic crystal. The shift of modes can be ascribed to the coupling between different modes owing to the finite size of the system. Indeed, in the infinite structure the conservation of momentum of EM wave, due to periodicity, would decouple L modes, whereas the finite coupling in truncated crystal leads to the shift and widening of the resonances. Based on symmetry considerations, we can understand the stronger L modes as compared to X modes. The former are formed due to Bragg reflections from 3 or 5 (for the smaller and larger clusters, respectively) layers formed by adjacent (nearest) neighbors in the 111 direction, while in the X direction, Bragg planes are formed only by next-nearest neighbors, which would make them more susceptible to the truncation.

We note that our calculation scheme does not allow us to compare the relative values of the peaks. It gives only the contribution to DOS that comes from the scatterers. Nevertheless we can clearly identify the modes of the clusters with the modes of the infinite structure. Moreover this allows one to find the dispersion curves similar to Fig. 2(b) but in finite clusters.

τ_d^{tot} and angularly averaged σ_{scat} in the region of the first two single sphere Mie resonances are shown in Figs. 3(b), 3(c). For comparison we included DOS [Fig. 3(a)] calculated for the infinite crystal using the method described in Ref. 26. Judging from scattering cross section only, one cannot compare a finite cluster to the infinite structure, while τ_d^{tot} allows a straightforward comparison with DOS of the photonic crystal.

Figure 3(b) shows twofold decrease of σ_{scat} at Mie resonances in the cluster. This suppression of the scattering effi-

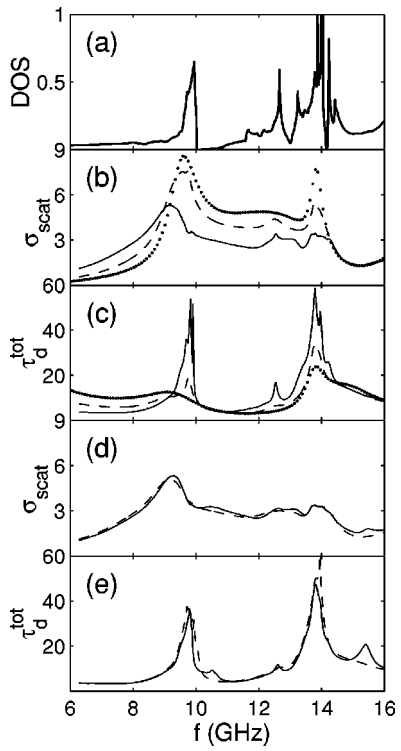

FIG. 3. (a) Density of EM states in the infinite photonic crystal. (b) Normalized scattering cross section for a single sphere (dotted), and clusters of five (dashed) and 32 (solid) spheres. (c) Normalized dwell time for the same as in (b). (d) Normalized scattering cross section for a cluster of 32 spheres, displaced by a half radius in a random direction off their lattice position (dashed line), and a cluster of 32 spheres, where 5 spheres were replaced by the smaller, 0.85 cm diameter, spheres (solid line). (e) Delay time for the same as in (d).

ciency can be attributed to the hybridization of the single particle resonances[27] due to multiple scattering. The dwell time is closely related to the nearfield[7] inside the clusters, where formation of the Bragg standing modes leads to substantial modification of the spectrum [Fig. 3(c)]. For a 32-sphere cluster one can see a strong resemblance between the density of the resonant modes expressed by τ_d^{tot} and the DOS in the photonic crystal [Fig. 3(a)]. It is interesting to compare our results to the 1D case considered in Ref. 28. For a stack of periodically arranged quantum wells it was argued that with an increase in the system size, the subradiant EM modes formed stable modes of the photonic pass band with large lifetimes, similar to our result for dwell times in the finite 3D clusters.

To assess the sensitivity of our results to disorder in the cluster, we performed the calculations of σ_{scat} and τ_d^{tot}, Figs. 3(d), 3(e), for the cluster of 32 spheres displaced by the half of the particle radius in random directions (dashed lines) off their initial positions and the cluster of 32 spheres where five spheres are replaced by defect spheres of a different diameter (0.85 cm) (solid lines). One can see that these two types of disorder had different effects on the spectra. While positional (topological) disorder led only to smearing of only sharp resonances, the defect spheres also introduced a number of new peaks. The later effect can be related to the new resonances introduced by the defect spheres. The effect of the topological disorder on photonic bandgaps was studied in (finite but comprised of large number of particles) 2D (Refs. 29,30) and infinite 3D photonic crystals.[31,32] Our conclusion on stability of the band structure to the topological disorder seems to be in line with Refs. 29,31. However, contrary to our case of small clusters, in large systems there may exist long-range on-average periodicity.

It is worth noting that fcc opal structures are not usually considered as a good candidate for potential applications connected to complete photonic bandgap.[33] However, as it can be seen from our example, for some parameters, DOS can be significantly suppressed in a wide spectral region. Figure 3 also suggests that such structures can be tolerant to strong topological disorder.

As was already mentioned in the Introduction, our results are relevant for the problem of light propagation in a dense random media made of particles with a narrow size distribution. Topological disorder does not prevent the occurrence of ordered clusters. In Ref. 9, random displacement of scatterers off their lattice positions did not have a significant effect on the density of EM states. Our calculations performed for the clusters with similar random displacements [Figs. 3(d), 3(e)], confirm small effects on the formation of the photonic band structure. This also suggests that the light localization observed in Ref. 9 may have been facilitated by depletion of the density of electromagnetic states due to the presence of the pseudogap.

The presence of clusters may significantly reduce the scattering length l_{scat}. Indeed, the scattering inside the clusters is dominated by the Bragg mechanism, while l_{Bragg} can be as small as a few lattice constants for high index contrast structures. Furthermore, in Ref. 34 it was shown that the periodicity of a photonic crystal may also exhibit itself in coherent back-scattering effects. The modification of coherent back scattering due to the presence of ordered clusters is an interesting problem that deserves a detailed consideration.

In conclusion, we calculated the scattering cross section and density of resonant states in small 3D ordered aggregates of dielectric spheres within the framework of the rigorous GMM solution. In contrast with previous studies we made no assumptions about the strength, size, or separation between scatterers. All multiple scattering effects were automatically retained in the solution. The results suggest that the photonic band structure of the infinite crystal can show up for clusters as small as five particles. The density of resonant states is significantly perturbed compared to single particle case. In the considered example, a cluster of 32 particles the density of resonant states can show a close resemblance to the DOS of the infinite system.

A.Y. gratefully acknowledges numerous insightful discussions and comments by A. A. Chabanov, A. A. Lisyansky, and L. I. Deych. This work was supported by the National Science Foundation under Grant No. DMR-0093949. H.C. acknowledges the support from the David and Lucile Packard Foundation and Alfred P. Sloan Foundation.

*Electronic address: a-yamilov@northwestern.edu

[1] E. Yablonovitch, Phys. Rev. Lett. **58**, 2059 (1987).
[2] S. John, Phys. Rev. Lett. **58**, 2486 (1987).
[3] *Photonic Crystals: The Road from Theory to Practice*, edited by S. G. Johnson and J. D. Joannopoulos (Kluwer Academic, 2002).
[4] An extensive bibliography on photonic crystals is maintained by Yu. Vlasov at URL http://www.pbglink.com/
[5] K.M. Ho, C.T. Chan, and C.M. Soukoulis, Phys. Rev. Lett. **65**, 3152 (1990).
[6] *Photonic Crystals and Light Localization in the 21st Century*, edited by C. M. Soukoulis (Academic, Boston, 2001).
[7] A. Lagendijk and B.A. van Tiggelen, Phys. Rep. **270**, 143 (1996).
[8] *Introduction to Wave Scattering, Localization, and Mesoscopic Phenomena*, edited by P. Sheng (Academic, San Diego, 1995).
[9] A.A. Chabanov and A.Z. Genack, Phys. Rev. Lett. **87**, 153901 (2001).
[10] A.A. Chabanov, M. Stoytchev, and A.Z. Genack, Nature (London) **404**, 850 (2000).
[11] D.S. Wiersma, P. Bartolini, A. Lagendijk, and R. Righini, Nature (London) **390**, 671 (1997).
[12] N. Stefanou, V. Yannopapas, and A. Modinos, Comput. Phys. Commun. **132**, 189 (2000).
[13] J. Pendry and A. Mackinnon, photonics website http://www.sst.ph.ic.ac.uk/photonics/
[14] A. Taflove and S. C. Hagness, *Computational Electrodynamics*, 2nd ed. (Artech House, Boston, 2000).
[15] D. Felbacq, G. Tayeb, and D. Maystre, J. Opt. Soc. Am. A **11**, 2526 (1994); V. Twersky, J. Acoust. Soc. Am. **24**, 42 (1992); S.C. Lee, J. Quant. Spectrosc. Radiat. Transf. **48**, 119 (1992); L.M. Li, and Z.Q. Zhang, Phys. Rev. B **58**, 9587 (1998).
[16] F.J.G. de Abajo, Phys. Rev. Lett. **82**, 2776 (1999).
[17] A.A. Asatryan, K. Busch, R.C. McPhedran, L.C. Botten, C.M. de Sterke, and N.A. Nicorovici, Phys. Rev. E **63**, 046612 (2001).
[18] Y.l. Xu, Appl. Opt. **36**, 9496 (1997), see also references in the online manual for multisphere scattering code at http://www.astro.ufl.edu/~xu/
[19] Y.V. Fyodorov and H.-J. Sommers, J. Math. Phys. **38**, 1918 (1997).
[20] E. Akkermans, A. Auerbach, J. Avron, and B. Shapiro, Phys. Rev. Lett. **66**, 76 (1991); E. Doron and U. Smilansky, *ibid.* **68**, 1255 (1992).
[21] G. Iannaccone, Phys. Rev. B **51**, 4727 (1995).
[22] S.A. Ramakrishna and N. Kumar, Eur. Phys. J. B **23**, 515 (2001).
[23] C.W.J. Beenakker and P.W. Brouwer, Physica E (Amsterdam) **9**, 463 (2001).
[24] H. Cao, in *Progress in Optics*, edited by E. Wolf (North Holand, Amsterdam, 2003), Vol. 45.
[25] S.G. Johnson and J.D. Joannopoulos, Opt. Express **8**, 173 (2001); http://ab-initio.mit.edu/mpb/
[26] K. Busch and S. John, Phys. Rev. E **58**, 3896 (1998).
[27] A. Moroz, J. Phys.: Condens. Matter **11**, 2503 (1999).
[28] L.I. Deych and A.A. Lisyansky, Phys. Rev. B **62**, 4242 (2000).
[29] M.M. Sigalas, C.M. Soukoulis, C.-T. Chan, and D. Turner, Phys. Rev. B **53**, 8340 (1996).
[30] A.A. Asatryan, P.A. Robinson, R.C.M.L.C. Botten, N.A. Nicorovici, and C.M. de Sterke, Phys. Rev. E **62**, 5711 (2000).
[31] S. Fan, P.R. Villeneuve, and J.D. Joannopoulos, J. Appl. Phys. **78**, 1415 (1995).
[32] Z.Y. Li and Z.Q. Zhang, Phys. Rev. B **62**, 1516 (2000).
[33] K.M. Leung and Y.F. Liu, Phys. Rev. Lett. **65**, 2646 (1990).
[34] J. Huang, N. Eradat, M.E. Raikh, Z. Vardeny, A.A. Zakhidov, and R.H. Baughman, Phys. Rev. Lett. **86**, 4815 (2001).