Mie resonances of dielectric spheres in face-centered cubic photonic crystals

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With use of plane waves as a basis for the band-structure calculation of a periodic assembly of highly refringent microspheres, it can be shown that resonance-mode frequencies of isolated dielectric spheres show up in the band structures. The strongly localized bands provided by the photonic-crystal analysis is compared with exact calculations made in spherical symmetry for an isolated microsphere. This comparison sheds some light on the effectiveness of the methods based on the description of mode coupling and, in particular, on the validity of tight-binding approaches of the description of photonic band structures. In addition, examining the effect of modifying the distance separating the spheres in the lattice, makes it easy to visualize the overlap between the modes of individual spheres. Thus quantitative information is provided on the geometry needed to feed energy into low-angular-momentum morphology-dependent resonances from a sharp source of the evanescent field and on the lifetime of these modes, when the resonances are disturbed by the proximity of a dielectric object of similar radius. © 2005 Optical Society of America

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1. INTRODUCTION

Understanding the optical properties of a photonic crystal requires the examination of Bragg multiple scattering in a lattice and a knowledge of the diffusion of waves by a particular element of the structure. When high-refractive-index contrasts are involved and structural elements are well separated, this two-stage process translates into the appearance of spectrally wide frequency gaps and strongly localized bands. Detecting the clear modal origin of a band provides valuable information on the overall optical response of a photonic crystal.

In solid-state physics, tight-binding approaches are used to understand the formation of a band from the hybridization of modes arising from isolated structures that are later assembled into a crystal. For electrons, the hybridization is simply the formation of a linear combination of atomic orbitals residing on each site, but the whole picture benefits from the strong localization of these orbitals, owing to the strong Coulomb attraction toward the atom nucleus. For photons, such a strong localization of modes on a structural element is not as easily obtained, so producing an efficient tight-binding representation of a complete photonic structure is not completely straightforward.

The often-quoted analogy between the electronic band structures of atomic crystals and the photonic dispersion in a periodic medium has driven many authors to propose photonic tight-binding schemes of various complexities. For two-dimensional photonic crystals, we should mention the work of Lidorikis et al., in which Mie resonances of isolated cylinders were the reference for localized “orbitals” while much information was still provided by more traditional plane-wave calculations. The tight-binding approach in acoustics and optics were guided by former results that, in particular, focused on the relationship between scattering resonances and the appearance of photonic band gaps. More in the spirit of detailed ab initio techniques, Wannier functions constructed from traditional band-structure calculations provided a systematic set of localized basis functions with many desirable properties. This particular basis was further considered recently as a universal tool for the description of bulk, defective, and even nonlinear photonic crystals.

This paper considers Mie resonances in the face-centered cubic assembly of dielectric spheres. The basic tools for this investigation are band-structure calculations, the associated densities of states, and the associated field maps. The main variable quantity will be the lattice parameter, and the radius of the spheres will be maintained as a reference constant. Proceeding in this way, our objective is to recognize, for large cell parameters, the resonance modes of the isolated sphere in the intricate spectrum of the crystal and, in so doing, point to the frequency ranges where a tight-binding description is meaningful. Additionally, useful information can be extracted from the observation of the dispersion change of these modes when the size of the crystal unit cell is tuned and the distance between the spheres is made to vary. The overlap between the external part of the modes, which controls the associated bandwidth, is also an important factor in determining the possibility of feeding the resonance modes with incident energy.

2. THEORY

In order to state the conventions and concepts that will be used later, we briefly describe the essential steps of Mie theory. The objective is the assessment of the resonance
electromagnetic modes for an isolated dielectric sphere and to provide a context for describing the localized states and their emergence from the band structure of a lattice of spheres.

A. Mie Resonances
The object under scrutiny is a homogeneous, isotropic, and optically linear material irradiated by an infinitely extended plane wave. Resonances are solutions of the sourceless Maxwell equations, which lead to a fully retarded vector Helmholtz equation. To account for the vector aspects of the problem, three instrumental vector retardation vector Helmholtz equation. Consequently, localized states will show up as flat bands in the band structure. As indicated by the following well-known expression, the density of states is related to the inverse of the group velocity:

$$D(\omega) = \sum_i \int_{E_i} \frac{dS}{\nabla E_i}.$$  

Then a localized mode can also be signaled by a sharp rise in the density of states in a very narrow spectral region. The localization of light on a specific site in the unit cell of the crystal structure produces an accumulation of modes of very closely distributed frequencies.

We are specifically interested in modes that develop their fields on the dielectric spheres, and this need can be reinforced by requesting a high contrast of the refractive indices. The localization of the fields can be easily understood if we refer to a variational picture of the mode formation, which reduces the photonic-crystal mode construction to an electromagnetic energy-minimization problem. The approximation made in choosing an incomplete basis set when approaching the electromagnetic modes by using tight-binding schemes is justified by the same arguments. The variational method, consisting of optimizing a linear combination of atomic orbitals with use of the energy variational principle, was introduced in 1929 by Sir John Lennard-Jones.

B. Localized Modes in Band Structures
Localized states can be recognized as modes for which the group velocity vanishes over a wide region of Bloch vectors. The significance of this is the immobility of the mode wave packet at a well-defined site in the structure. Consequently, localized states will show up as flat bands in the band structure. As indicated by the following well-known expression, the density of states is related to the inverse of the group velocity:

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The roots of transcendental Eq. (1) provide the frequencies of the resonance modes. The equations’ right-hand and left-hand sides are, respectively, related to the inside and the outside of the sphere. The outer radial part of the fields is described by a Hankel function that corresponds to a complex linear combination of Bessel spherical functions of the first and second kinds. Physically it conveys the existence of a field propagating away from the sphere. As a consequence, the mode lifetime is finite, and frequencies can assume only complex values. Therefore finding the resonance modes of the sphere amounts to extracting complex roots from Eq. (1). As far as we know, the use of numerical methods in this search cannot be avoided.

3. METHOD
In this paper electromagnetic coupling will be studied by assembling dielectric spheres into a face-centered cubic crystal structure. This structure generalizes a colloidal opal cubic structure by allowing the spheres to move away from each other. The first Brillouin zone of this highly isotropic structure has an external boundary that closely describes a sphere. In this structure each site has 12 nearest neighbors, all located at the same distance, which can be expressed as \(a/\sqrt{2}\) if the cubic lattice parameter is denoted \(a\).

In what follows, we lock in the value of two quantities: the radius of the sphere \(R\) and its refractive index \(n\). In the compact limit of this structure, the filling factor of the spheres reaches the value \(f=0.74\). For a crystal structure made more tenuous by increasing the lattice parameter \(a\), the lattice parameter is related to the filling factor by the simple relation

$$a = 2^{3/2} \sqrt{\frac{2\pi}{3f} R}.$$  

The refractive index is the second parameter that will be kept fixed. The recommendation of high index contrast made in Subsection 2.B suggests the choice of semiconducting material with a high index value, such as \(n=3.6\). It will then be easier to excite well-defined resonance modes of the dielectric spheres. By contrast, the cell pa-
rameter $a$ will be varied, starting from its compact lower limit $a_c=2\sqrt{2}R$. For this reference value of $a$, a strong interaction between the spheres is expected, so that tuning down the spheres’ influence on each other requires an increase in the lattice parameter. Consequently, flat bands corresponding to localization of electromagnetic modes will be given a better chance to emerge for large cell parameters and low filling fractions.

All the band structures have been calculated by the plane-wave method,\textsuperscript{10,13} which exploits the periodicity of the dielectric constant by the use of a Fourier series in the representation of the Bloch electromagnetic modes. The wave equation obeyed by the magnetic field is obtained from the Maxwell equations and leads to a Hermitian linear eigenvalue equation. For a given value of the Bloch wave vector in the Brillouin zone, eigenvalues provide the frequencies and hence the dispersion relations. For the low-lying bands under study, 749 was found to be an appropriate number of plane waves in the Fourier series.

4. RESULTS

We start with a large value of the lattice parameter $a$. In a crystal built by assembling distant dielectric spheres, Bloch linear combinations of the resonance modes of the sphere are expected to dominate the electromagnetic states. But these alone cannot rebuild the whole photonic crystal states. Indeed, while the filling factor of the structure is vanishing, the photonic crystal should start resembling empty states, leaving a continuum of long-wavelength unmodulated plane waves that are not included in the combination of localized morphology-dependent modes in the sphere. This creates a very strong difference from the linear combination of atomic orbitals techniques used in quantum chemistry: Photonic states are not all tightly bound, even when the distance between the spheres is made extremely large. Close to the Brillouin zone center, when the wave vector approaches the $\Gamma$ point the frequencies of two transverse branches vanish, as they do in the free-space vacuum, with the dispersion relation $\omega=kc$; such states cannot be easily represented by a linear combination of low-lying localized modes centered on the spherical sites. A tight-binding approach should then always include in the fundamental basis set a series of vacuum transverse plane waves in addition to the localized modes associated with weakly coupled structure elements.

A. Interpretation of the Band Structure

First we recall that the resolution of Eq. (1) provides the frequencies of the resonance modes of an isolated sphere. Here we consider a sphere made of a dielectric material with a refractive index of 3.6 placed at the origin of space in vacuum. Subsection 2.A shows that the modes depend on polarization. For $l=0$, the solutions are

[TE]: \[ \frac{\omega R}{2\pi c} = \frac{2k - 1}{4n}, \]  
[TM]: \[ \frac{\omega R}{2\pi c} = \frac{k}{2n}, \]  

and, with the selected refractive index, we obtain the following numerical values for the reduced frequencies: 0.069, 0.208, 0.347, ... in TE polarization and 0.139, 0.277, 0.416, ... in TM polarization. The $l=1$ modes are 0.132, 0.272, 0.412, ... in TE polarization and 0.186, 0.340, 0.481, ... in TM polarization. For $l=2$, we obtain for the TE and the TM polarizations 0.192, 0.333, 0.475, ... and 0.240, 0.393, ..., respectively.

The presence of the resonance modes of the isolated sphere is clearly noticeable in the band structure [Fig. 1(a)] for a large value of the cell parameter $a$. In this case, $a$ is taken to be 2 $\mu$m for which the spheres of radius $R = 0.3535$ $\mu$m have a quite small coupling. The calculated band structure reveals a set of flat bands that extend over a wide part of the Brillouin zone. The accumulation frequencies appear very clearly in the density of modes as shown in Fig. 2, which exhibits a series of narrow peaks indicating strong spectral and spatial localizations of light. In the same figure the frequencies of the reference isolated-sphere modes are also indicated.

The lowest band seen in Fig. 1(a), actually degenerated twice, is the acoustic band, whose frequency vanishes along with the wave vector $k$ in the long-wavelength limit $\Gamma$. In this limit, the field gradually becomes uniform. The slope of the dispersion relation gives both the group and the phase propagation velocities at wavelengths large enough to allow viewing the structure as a mean, uniform medium. At higher frequencies, the bands are dominantly associated with a resonance mode of the spheres. In Fig. 2

![Fig. 1. Photonic band structure of an assembly of spheres of radius $R=0.3535$ $\mu$m distributed according to a face-centered cubic lattice of cell parameter (a) $a=2$ $\mu$m and (b) $a=1$ $\mu$m. The refractive index of the spheres is set to 3.6. In units $2\pi/a$, $\Gamma=(0,0,0)$, $X=(0,1,0)$, $L=(1/2,1/2,1/2)$, and $U=(1/4,1,1/4)$.](image)
we can distinguish two types of peaks. The first set can be explained by a single resonance mode of the isolated sphere, as for the reduced frequency 0.2789, where we find a TE mode characterized by $k=2$ and $l=1$. The second set is seen as a group of crests as, for instance, those centered around 0.2005 or 0.2593. These are associated with the modes of different polarizations that coincide in frequency. In the face-centered cubic structure, these frequencies are redistributed over a perceivable spectral region. The bands, mixing the TM mode with $k=1$ and $l=1$ with the TE mode with $k=1$ and $l=2$, distribute themselves around the reduced frequency 0.2005.

The coupling of the two modes can be easily understood from the symmetry point of view. The separation of TE and TM modes is a natural consequence of the symmetry of the dielectric microsphere. The introduction of the cubic symmetry that follows from the setup of a lattice of spheres breaks the rotational invariance, so that spherical TE and TM modes with frequencies close to each other are allowed to hybridize. In one- or two-dimensional photonic crystals, enough symmetry is maintained to keep the exact independence of two distinct polarizations.

The periodic repetition of the spheres unavoidably influences the spectral location of the mode, besides introducing some tunneling-assisted broadening. The consequence is a frequency- and polarization-dependent shift that is apparent when one compares the frequencies of the sphere resonances with the location of their associated Van Hove singularities in the density of modes. This shift is, however, very difficult to assess from a band-structure calculation, as it is expected to be quite similar to the uncertainty that is due to the limited convergence of the plane-wave representation. In spite of this, however, the comparison we have made is very meaningful in view of the fact that the plane-wave basis is extremely different from the collection of eigenmodes of the sphere and still allows us to clearly recognize the localized sphere modes in the intricacy of the band structure.

**B. Interaction**

We now turn to the analysis of the coupling between the various sphere resonance modes in the face-centered cubic crystal. The third-lowest band in Fig. 1(a) is almost monochromatic across the whole Brillouin zone, indicating a mode strongly localized in the volume of the sphere. By comparison, the second band is seen to experience a significant group velocity in well-defined directions, expressing a delocalization of the associated mode.
The representation of the intensity of the magnetic field at the X point of the Brillouin zone actually confirms this interpretation. In Fig. 4 the field strength is represented on the (101) face of the crystal, which takes the shape of a square cell that contains spheres centered on its vertices and at its center. As expected, the map of the third mode [Fig. 4(b)] shows a strong localization of the field within the sphere volume, whereas for the second band [Fig. 4(a)] the field spreads over the sphere surface well into the surrounding vacuum.

A progressive change from Fig. 1(a) to Fig. 1(b), which corresponds to a reduction of the lattice parameter \( a \) by a factor of 2, illustrates the delocalization of the fields when the spheres’ electromagnetic modes start interacting strongly. The influence of the modification of the lattice parameter is more noticeable in Fig. 3. Indeed, it highlights two elements. The third band is much less dispersive than the second band. Moreover, the figure shows that the field is more strongly localized as the cell parameter \( a \) becomes higher than 1.25 \( \mu \text{m} \) in our case. This is noticed clearly starting from the separation of the curves associated with the third band. This last element makes it possible to evaluate quantitatively the zone for which a tight-binding approach is valid. For strong coupling, the presence of the sphere resonance modes is not completely obvious when one is considering the crystal band structure. The influence of localized modes on those associated with the neighboring sites is so strong that the range of vanishing group velocities is now considerably reduced. The distribution of intensities of the magnetic field confirms the delocalization at least for the spread mode associated with the second band, shown in Fig. 5(a). The confined mode associated with the third band [Fig. 5(b)] is essentially unchanged: At this distance of approach, this highly confined mode does not yet overlap its replicas of the neighboring sites of the lattice.

5. CONCLUSION

A system of spheres distributed on the nodes of a crystal lattice, separated by vacuum, can be considered unrealistic except in the limit of a compact structure. However, one should realize that the region separating the spheres could be filled with a mechanically stiff material of very low refractive index. Such materials exist: Among polymers, for instance, polytetrafluoro-ethylene is known to exhibit a refractive index that does not exceed 1.35. Also, by high-current electroanodization, very light porous silicon can be produced with a refractive index that approaches that of vacuum. So if in the present work the widely-separated-spheres model has been used as a theoretical tool, the conclusions derived for this system can also suggest more practical investigations if the fabrication technology can put such low-index materials to work.

Photonic crystal made from the periodic repetition of structural elements that support individual localized modes is an important class of inhomogeneous dielectric structures. The description of such crystals could refer to an instructive analogy with molecular crystals, except that it is not possible to avoid the presence of extended states that are maintained in the interstitial regions if the photonic-crystal filling fraction is tuned down to zero. Such structures, with a weak overlap between the localized modes supported by the loosely assembled elements, are interesting from many points of view; one of the most interesting aspects of these photonic materials is the fact that they give rise to dispersion relations that saturate the energy at a constant level in a very wide range of Bloch wave vectors. This creates a case in which the group velocity of the propagated waves becomes interestingly small for technological applications. This particular behavior corresponds to a special propagation mechanism in which the energy uses a hopping mechanism to transfer from one localized mode on a structural element to a mode on a neighboring one and, from site to site, across the whole lattice. This mechanism is reminiscent of the propagation of electromagnetic excitation on chains of distant metallic nanoparticles, with notable differences. For instance, the resonant localized modes do not rely on material dispersion but rather on a high-index contrast at the outer surface to provide the necessary confinement. Another important point is that these dielectric resonators can avoid conductive dissipation, which is still one of the main limitations of the lifetime of metallic chain excitation.

The calculation of the band structure and the density of modes of a crystal characterized by a large cell parameter and a strongly localizing element defines a new type of
photonic structure, which could be designated as molecular photonic crystals. The present detailed study of a molecular photonic crystal that assembles simple spheres on a face-centered cubic lattice has underlined the emergence of two distinct types of modes. The first one arises from the Bloch combination of a single mode per site, with a frequency well separated from other resonances. This kind of mode gives rise to a very sharp isolated structure in the density of modes. The spherical dielectric element provides such a state, but many other geometries could certainly be examined as other candidates. The second case involves a coupling between modes of two different polarizations and angular momenta but that are degenerate or nearly degenerate in the density of modes. These modes provide groups of closely related bands characterized by low group velocity and high spectral sharpness.

Finally, a comparison between the isolated sphere and the loosely coupled lattice of spheres provides data on the interaction distance between the spheres. By studying the broadening of the bands associated with a linear combination of excitations localized on spherical structural elements, one can assess the distance at which neighboring elements in the crystal start interacting, which means allowing for the modes’ overlap. This information is highly valuable not only for designing the molecular photonic crystals mentioned above but also for discussing questions related to the transfer of energy into a localized mode such as the whispering gallery mode.

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