Fragility of photonic band gaps in inverse-opal photonic crystals

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Inverse-opal techniques provide a promising routine of fabricating photonic crystals with a full band gap in the visible and infrared regimes. Numerical simulations of band structures of such systems by means of a supercell technique demonstrate that this band gap is extremely fragile to the nonuniformity in crystals. In the presence of disorder such as variations in the radii of air spheres and their positions, the band gap reduces significantly, and closes at a fluctuation magnitude as small as under 2% of the lattice constant. This imposes a severe requirement on the uniformity of the crystal lattice. The fragility can be attributed to the creation of this band gap at high-frequency bands (eight to nine bands) in inverse-opal crystals.

In recent years the fabrication of photonic crystals has attracted extensive interest,[1–3] as such artificial periodic structures may bring about some peculiar physical phenomena such as inhibition of spontaneous emission and localization of electromagnetic waves.[2–4] In addition, they possess possible applications in wide scientific and technical areas such as filters, optical switches, cavities, waveguide, design of low-threshold lasers, and high-efficient light emitting diodes.[1–3]

A three-dimensional (3D) photonic crystal with a full band gap in the visible and infrared regimes provides the most stirring potential in application. Recently, the fabrication of 3D photonic crystals of micrometer size have been demonstrated[5,6] using a layer-by-layer growth scheme[7] that employs state-of-the-art microlithography techniques, however it still remains a difficult and challenging task. Another routine that is in rapid progress is the self-arrangement of colloid, related artificial opals, and inverse-opal techniques.[8–13] Among them, the inverse-opal technique becomes an attractive candidate in the fabrication of optical photonic crystals. These crystals are composed of close-packed air spheres arranged in a face-centered-cubic (fcc) lattice embedded in a dielectric background. When the refractive index contrast is large enough, a full band gap opens at high-frequency bands.[14] Very recently, great progress has been made in this technique by several groups.[11–13]

As the crystals are of micrometer and submicrometer sizes, various kinds of nonuniformity inevitably occur in the fabrication process. A typical inverse-opal crystal is prepared as follows.[11] First, one should have a template assembled from a self-organizing system, for instance monodisperse silica or polystyrene colloidal crystal. Then sintering is used to create necks between spheres. This intersphere interconnection allows the precipitation of a desired background dielectric into the voids of the template by means of chemical reaction. Finally, the inverse-opal crystal is obtained by removing the original template material by calcination. In practice, nonuniformities occur at every step of the fabrication. For instance, the radius of spheres might vary even for monodisperse systems,[12] or the spheres might not array at exact lattice sites when they form colloidal crystal and when the template is sintered. In addition to these geometrical disorders, physical disorders such as incompleteness of precipitation and calcination can take place in the later steps of fabrication.

In this work, we will show via numerical calculations that the above geometrical nonuniformities, i.e., variations in the radii of spheres (size randomness) and their random displacements from lattice sites (site randomness), will greatly reduce the band gap of inverse-opal crystals. At a disorder magnitude as small as under two percent of the lattice constant, the band gap is closed even in the presence of a very high refractive index contrast. The high fragility of the band gap should impose a severe restriction on experimental efforts to control crystal uniformity.

The disorder in photonic crystals can be described by certain random parameters. In the case of site randomness, every sphere has the same radius r_0, while the x, y, and z components of the position of the ith sphere in the disordered crystal differ from those of the periodic case by γ_x, γ_y, and γ_z, respectively, where γ_x, γ_y, and γ_z are random variables uniformly distributed over the interval of $[-d_t, d_t]$. d_t denotes the strength of site randomness. For the case of size randomness with strength d_r, the spheres are arrayed in the original lattice sites, while the radius of the ith sphere is given by $r_i = r_0 + \gamma_r$, where γ_r is a random variable uniformly distributed over the interval $[-d_r, d_r]$. In reality, both kinds of disorder coexist with $d_t \neq 0$ and $d_r \neq 0$.

The electromagnetic problem in 3D perfect photonic crystals is solved with the use of the plane-wave expansion method.[15,16] The convergence for the ten lowest bands can be made better than 0.5% by adopting 343 plane waves. For a disordered crystal, a supercell technique[17–19] is employed with a cubic supercell composed of eight conventional fcc unit cells with 32 spheres and an expansion of 2197 plane waves, where the convergence is better than 1.0%. We have compared the results with those from a supercell with four conventional unit cells and found the same behavior.

We first investigate the band structure of a typical inverse-opal crystal. The result for a filling fraction of air spheres as $f = 0.78$ and a refractive index of the background dielectric as $n = 3.6$ is displayed in Fig. 1. It is clear that a full band gap opens between the eight and ninth photonic bands, with gap edges lying at X and W points, respectively.

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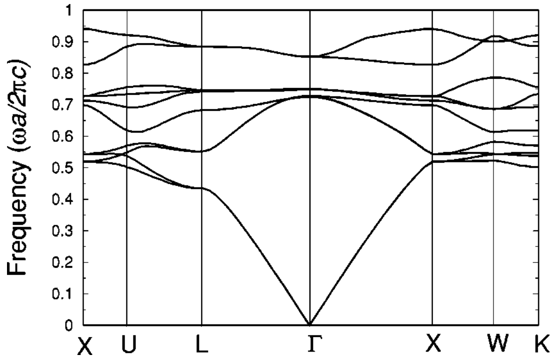

FIG. 1. Calculated band structures of an inverse-opal crystal along some important high-symmetry lines in the Brillouin zone. The crystal has a filling fraction of air spheres as $f=0.78$ and a refractive index of background dielectric as $n=3.6$.

The gap lies at a frequency of $0.786-0.827(2\pi c/a)$ with a normalized width of $\Delta\omega/\omega_g=5.1\%$. Here a is the fcc lattice constant, c is the light speed in the vacuum, and $\Delta\omega$ and ω_g are the gap width and midgap frequency, respectively. It should be noted that the inverse-opal crystal made of titania (TiO_2) (Ref. 12 and 13) does not possess a full band gap at eight to nine bands, as the refractive index of titania $n=2.8$ is not high enough.

As the band gap lies at high-frequency bands, it is expected that this gap will be sensitive to disorder. In fact, recent work on two-dimensional (2D) disordered photonic crystal[17] demonstrated that the higher band gap is far more sensitive to the site and size randomness than the ground band gap. Although this is apparent from a simple physical argument, our interest here is to investigate how fragile the gap in inverse-opal crystal could be in the presence of disorder. The quantitative answer to this question would serve as an important guide in the fabrication of such materials. The results for the density of states (DOS) in disordered inverse-opal crystals with $n=3.6$ and $f=0.78$ are displayed in Fig. 2 for the case of (b) size randomness with $d_r=0.05r_0$, (c) site randomness with $d_t=0.05r_0$, and (d) coexisting size and site randomness with $d_r=d_t=0.05r_0$. Note $0.05r_0=0.018a$, as $r_0=0.36a$ for a filling fraction of $f=0.78$. For these three cases, we obtain the DOS by solving Maxwell's equations at 4000 points inside the first Brillouin zone of a fcc unit cell. For clarity of comparison, we also plot the DOS of a perfect crystal in Fig. 2(a). The calculation points number about 10 000, and the DOS is normalized with respect to those of disordered crystals. The oscillation in the long-wavelength end of the four curves is due to the limited number of calculation points.

The band gap centered at about $0.805(2\pi c/a)$ is completely closed when $d_r=0.05r_0$ and $d_r=d_t=0.05r_0$. There remain significant dips in the DOS curves, a signature of pseudogaps. In the case of site randomness with $d_t=0.05r_0$, there still remains a greatly reduced band gap at $0.807-0.816(2\pi c/a)$ with a width of 1.0%. In contrast, the disorder affects the pseudogap centered at $0.53(2\pi c/a)$ only slightly. Another apparent characteristic is the flattening of high-frequency sharp peaks in the DOS of the perfect crystal by disorder. It seems that these peaks are created by long-range resonant scattering of lattices, and become wider in the case of random scattering.

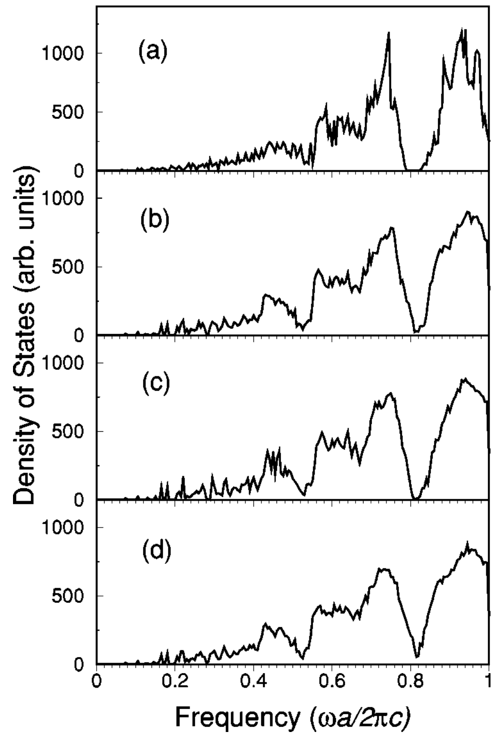

FIG. 2. Calculated density of states (DOS) in the inverse-opal crystal with $n=3.6$ and $f=0.78$ in the case of (a) no disorders, (b) size randomness with strength of $d_r=0.05r_0$, (c) site randomness with $d_t=0.05r_0$, and (d) coexisting size and site randomness with $d_r=d_t=0.05r_0$.

We next investigate the dependence of gap size on disorder in inverse-opal crystals. The results for three kinds of randomness are displayed in Fig. 3(a) for crystal with $n=3.6$ and $f=0.78$. The random strength is in units of sphere radius r_0. In the case of coexisting randomness, we set d_r

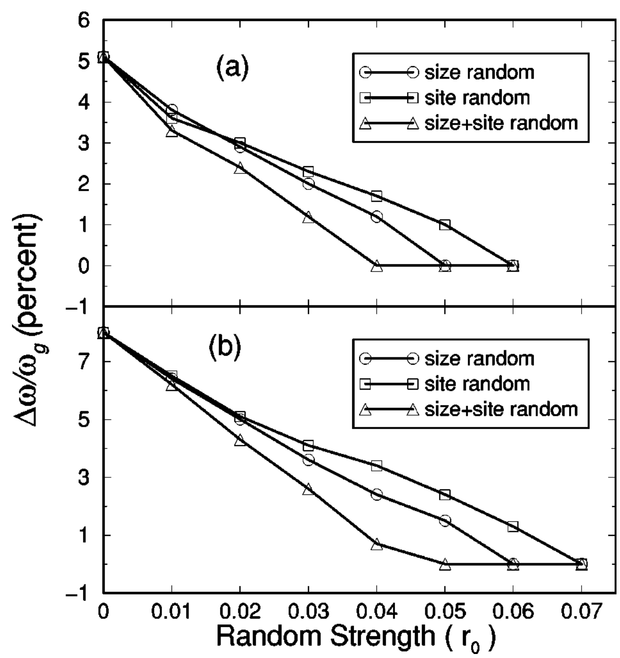

FIG. 3. Plots of dependence of band-gap size on the random strength of three kinds of geometrical disorder in an inverse-opal crystal with (a) $n=3.6$ and $f=0.78$; (b) $n=4.0$ and $f=0.78$.

$=d_t$. Naturally the coexisting randomness should reduce the band gap more than just either of the two randomnesses does. Indeed, this is verified by numerical calculations: the gap width decays fastest in the case of coexisting randomness. The band gap is closed for the site randomness at $d_t = 0.06r_0$, for the size randomness at $d_r = 0.05r_0$, and for the coexisting randomness at $d_r = d_t = 0.04r_0$. In addition, the gap size is reduced faster in the case of size randomness than site randomness. This means that the photonic band gap is more sensitive to the variations in the radii of spheres than the variations in their displacements. Similar behavior was found in 2D cases.[17] It can be argued qualitatively that varying the size of the sphere means changing the filling fraction, while the fraction does not change when displacing spheres from lattice sites. Thus, the size randomness reduces the gap size more significantly.

We now consider a practical material with a higher refractive index in optical frequency, germanium ($n = 4.0$). We have investigated the band gap of such an inverse-opal crystal with $f = 0.78$ in the case of various disorders with different random strengths. The results are plotted in Fig. 3(b), which looks quite similar to Fig. 3(a). A full band gap opens at $0.710 - 0.769(2\pi c/a)$ with a normalized width of 8.0%. This band gap almost decays linearly with respect to random strength, and is reduced to zero at site randomness $d_t = 0.07r_0$. For the case of size randomness, the band gap is closed at $d_r = 0.06r_0$, and it is closed at $d_r = d_t = 0.05r_0$ for the case of coexisting randomness.

Therefore, for such an inverse-opal structure, as two kinds of randomness generally coexist, the demand for high-quality geometrical uniformity is very severe: Nonuniformities as small as under 2% of the lattice constant will destroy the band gap completely.

One may expect that a further increase in the refractive index contrast will relax the severe requirement of geometrical uniformity, as the band gap of perfect crystals widens accordingly. At the same time, the midgap frequency falls, which means that the relative disorder strength decreases compared with the midgap wavelength. Therefore, the influence of disorder on the band gap should become smaller. However, such a naive conjecture is negated by realistic numerical calculations. Here we fix the random strength as $d_r = 0.05r_0$ for size randomness, $d_t = 0.05r_0$ for site randomness, and $d_r = d_t = 0.05r_0$ for coexisting randomness. Then we increase the refractive index of the background dielectric, and investigate the variation of the band gap. The results are displayed in Fig. 4, where the dependence of band-gap size on the refractive index for a perfect crystal is also plotted for comparison. The filling fraction of air spheres is $f = 0.78$ in all cases.

The band-gap size for the perfect crystal grows from 5.1% at $n = 3.6$ to 13.1% at $n = 5.6$, an increase of over two and a half times. In the presence of either size randomness or site randomness, the gap size still increases remarkably when the refractive index is increased, although it is greatly reduced when compared with that of the perfect crystal. However, for a practical situation where size and site randomness coexist, the band gap is closed completely, irrespective of a very high refractive index contrast in the crystal. This surprising characteristic can be understood qualitatively as follows. The growing of the refractive index will result in band-

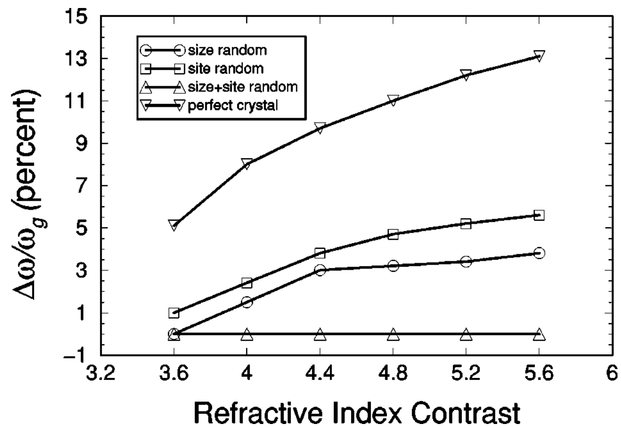

FIG. 4. Dependence of band-gap size on the refractive index contrast in a perfect inverse-opal crystal and disordered crystals with three kinds of geometrical randomness. The filling fraction of air spheres is $f = 0.78$ in all cases.

gap widening and downshifting of the midgap frequency. However, at the same time, the scattering strength due to both kinds of randomness also increases. In addition, it is seen from Fig. 4 that the band-gap growth in the perfect crystal as well as in the disordered crystal with either size or site randomness all show a saturation behavior. It is thus expected that the size reduction in the presence of coexisting randomness should overwhelm the size increase caused by refractive index growth. Therefore, the band gap keeps closed at all refractive index contrasts.

Some words should be said concerning another routine of fabricating a photonic crystal at the optical frequency, i.e., microlithography, such as electron-beam lithography and x-ray lithography. In the scheme of layer-by-layer growth,[5–7] the crystal opens a large full band gap between the second and third photonic bands, a ground band gap. Although significant nonuniformities will occur in the growth process with current techniques for submicrometer-sized structures, the band gap is fairly robust to such nonuniformities according to recent numerical simulations.[18,19] There are two key points: One is the opening of a band gap at ground photonic bands, the other is the large size of the band gap. Therefore, from the viewpoint of the tolerance of band gaps to geometrical nonuniformities, the microfabrication routine might be superior to the inverse-opal technique.

The above results are obtained from a finite-sized system. When system size is increased, more localized states will appear deeper inside the band gap and, therefore, reduce the gap width. For an infinite system, a real band gap could be much smaller. We estimate the real gap in an infinite system by using the well-known Saxon-Hunter theorem for electronic systems.[20] For simplicity, we consider the case of size randomness. The real gap can be estimated by measuring the overlap of two band gaps in two extreme cases: one for a pure lattice of radius $r_0 + d_r$ and the other for a pure lattice of radius $r_0 - d_r$. When $n = 3.6$ and $f = 0.78$, we find that the real gap closes at $d_r \approx 0.004a$, about five times smaller than the value of $0.02a$ predicted by our numerical calculations using 32 spheres. In reality, the system size could be much larger but finite. Therefore, the upper limit of disorder strength will lie somewhere between $0.02a$ and $0.004a$. This implies an even much more stringent condition, if not impossible, in the sample frabrication.

In summary, we have investigated the band gap in inverse-opal photonic crystals in the presence of geometrical nonuniformities by numerical simulations with the use of the plane-wave expansion method combined with a supercell technique. It is found that this band gap is extremely fragile to the nonuniformity in crystals. In the presence of both size and site randomness, the band gap reduces significantly and closes at a fluctuation magnitude as small as under 2% of the lattice constant. This imposes a severe demand on the high-quality lattice uniformity. Such fragility can be attributed to the creation of the band gap at high frequency bands (eight to nine bands) in inverse-opal crystals. It is expected that the presence of other physical irregularities can further restrict the opening of a full band gap.

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