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Three-Dimensional Complete Photonic-Band-gap Structures in the Visible

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It is shown that for a simple face-centered-cubic lattice of spherical scatterers more than one complete photonic band gap (CPBG) can open and the relative gap width can be as large as 10% even if the host dielectric constant $\varepsilon_h = 1$. The key is to use the spheres made out of material having a Drude-like behavior of the dielectric function, and, to tune lattice spacing appropriately to a characteristic plasma wavelength. We argue that such structures can provide CPBG structures with tunable band gaps down to ultraviolet wavelengths.

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Photonic crystals are structures with a periodically modulated dielectric constant. In analogy to the case of an electron moving in a periodic potential, certain photon frequencies can become forbidden, independent of photon polarization and the direction of propagation—a complete photonic band gap (CPBG) [1,2]. As early as in 1972 photonic crystals with such a gap have been shown to offer the possibility of controlling the spontaneous emission of embedded atoms and molecules in volumes much greater than the emission wavelength [3] and, later on, to be an important ingredient in a variety of technological applications [4]. There is a common belief that, in the near future, photonic crystal systems will allow us to perform many functions with light that ordinary crystals do with electrons [4]. At the same time, photonic structures are of great promise to become a laboratory for testing fundamental processes involving interactions of radiation with matter in novel conditions. Unfortunately, technological difficulties in fabricating CPBG structures rapidly increase with decreasing wavelength for which a CPBG is required, mainly because of the simultaneous requirements on the dielectric contrast and the modulation (the total number and the length of periodicity steps). As yet no photonic crystals are available with CPBG at optical wavelengths and fabrication of photonic crystal with such a gap poses a significant technological challenge already in the near infrared [5,6]. In order to achieve a CPBG below the infrared wavelengths, the modulation is supposed to be on the scale of optical wavelengths or even shorter and, as for any CPBG structure, has to be achieved with roughly ten periodicity steps in each direction, a task currently beyond the reach of reactive ion and chemical etching techniques. Fortunately, such a modulation occurs naturally in colloidal crystals formed by monodisperse colloidal suspensions of microspheres. The latter are known to self-assemble into three-dimensional crystals with excellent long-range periodicity on the optical scale [7], removing the need for complex and costly microfabrication. Colloidal systems of microspheres crystallize either in a face-centered-cubic (fcc) or (for small sphere filling fraction) in a body-centered-cubic (bcc) lattice [7]. Let ε_h and ε_s be the host and sphere dielectric constant, respectively. If ε_s is less than ε_h, the so-called "air spheres" case, calculations show that a CPBG can open in a simple fcc structure [8], provided the dielectric contrast $\delta = \max(\varepsilon_h/\varepsilon_s, \varepsilon_s/\varepsilon_h) \gtrsim 8$ [9,10]. This puts a large constraint on the material choice which is yet to be overcome. It is even more difficult to fabricate crystals with a CPBG large enough for technological applications. If we define the relative gap width g_w as the gap width-to-midgap frequency ratio, $\triangle\omega/\omega_c$, practical crystals are expected to have g_w larger than 5%—to leave a margin for gap-edge distortions due to omnipresent impurities and yet to have a CPBG useful for applications. In order to achieve g_w larger than 5%, $\delta \gtrsim 12$ is required [9,10] which makes fabrication of photonic crystals with an operational CPBG at optical wavelength seemingly hypothetical [11]. The requirements on the dielectric contrast are less prohibitive (although still significant if an operational CPBG is required) for colloidal crystals with a diamond structure [1]. The latter, however, have yet to be fabricated.

Here we show (see Fig. 1) that three-dimensional structures of microspheres with a Drude-like dielectric function [12],

$$\varepsilon_s(\omega) = 1 - \omega_p^2/\omega^2, \tag{1}$$

where ω_p is called the plasma frequency, can exhibit CPBG ≈10% even if the host dielectric constant ε_h is as low as $\varepsilon_h = 1$ and a natural simple fcc structure of colloidal crystals is preserved. A Drude-like behavior can be found in certain metals and semiconductors [12,13] and, as shown recently, in new artificial structures [14]. Metals and semiconductors can be quite lossy at optical frequencies; however, their absorption can be negligible in a certain frequency window, in which case they behave as a conventional, although highly dispersive, dielectric. Typically, the plasma wavelength $\lambda_p = 2\pi\omega_p/c$, where c is the speed of light in vacuum, is closer to the short-wavelength edge of the nonabsorptive window, since for shorter wavelengths there is a higher probability to induce

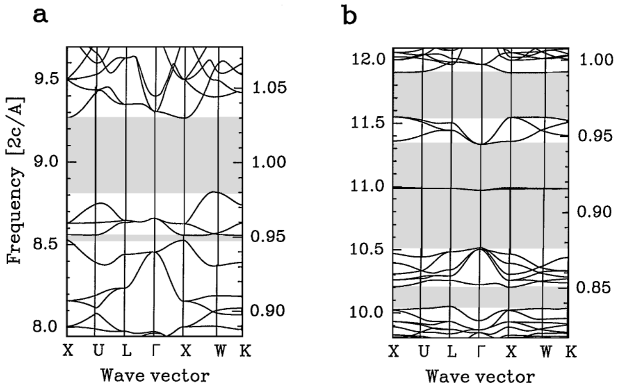

FIG. 1. Calculated photonic-band structure of a close-packed fcc lattice of spheres with a Drude-like dielectric function embedded in a host dielectric with the dielectric constant $\varepsilon_h = 1$. Optical frequency is plotted as a function of the wave vector, with the indices on the axis corresponding to special points of the Brillouin zone. Frequency is plotted in units of $2c/A$, with A the length of the conventional unit cell of a cubic lattice and c the speed of light in vacuum. Numerical values of frequency, when multiplied by the factor $1/(2^{3/2}\pi)$, yield those of r_s/λ, where r_s is the sphere radius and λ is the wavelength outside the spheres. For convenience, on the right y axes the ratio ω/ω_p is plotted. Shaded regions parallel to the wave vector axis in which there are no allowed frequencies indicate the existence of a CPBG. (a) is for $\omega_p = 9$ ($r_s/\lambda_p = 1.013$). Note a large CPBG ($g_w = 5.2\%$) with midgap frequency 9.033 and a second smaller complete band gap ($g_w = 0.5\%$) with midgap frequency 8.544. (b) shows the photonic band structure for $\omega_p = 12$ ($r_s/\lambda_p = 1.35$). Note four CPBG's with midgap frequencies at 10.130, 10.746, 11.172, and 11.728, with the respective gap widths 1.5%, 4.3%, 3.1%, and 3.9%. Two extremely narrow almost dispersionless bands slightly below $\omega = 11$ separate the second and the third CPBG.

electronic interband transitions. If this nonabsorptive window overlaps with the Drude-like behavior of the dielectric function around the plasma frequency, the following two features make the proposed photonic structures extremely interesting. First, since the host is assumed to be dielectric, one has $\varepsilon_h \geq 1$. Therefore $\varepsilon_s < 1 \leq \varepsilon_h$ and the proposed structure is analogous to a photonic crystal made of air spheres [15]. Second, because $\varepsilon_s(\omega_p) = 0$, the dielectric contrast δ becomes extremely large for frequencies around ω_p. The first property is necessary for opening a CPBG in a simple fcc structure, whereas the second property favors the opening of maximal CPBG's available for such structures [9,10]. An example of the material with the required properties is silver for wavelengths in the region 310–520 nm and zero crossing of Reε around 328 nm (see Figs. 3 and 5 of [13]).

Given a plasma frequency ω_p, we performed band-structure calculations, using a photonic analog [16] of the familiar Korringa-Kohn-Rostocker (KKR) method [17], for frequencies from $0.5\omega_p$ up to $1.1\omega_p$, assuming $\varepsilon_h =$ const in this frequency region. Compared to the plane-wave method, dispersion does not bring any difficulties to the KKR method and computational time is the same as without dispersion. In order to ensure convergence around 0.1%, spherical waves were included with the angular momentum up to $l_{\max} = 10$. Further discussion of convergence and errors can be found in [10].

For Drude-like scatterers, the plasma wavelength sets a characteristic scale. Correspondingly, band gaps occur only for certain values of r_s/λ_p, where r_s is the sphere radius. The main issue in opening a CPBG is to tune r_s to the corresponding plasma wavelength λ_p to reach a certain threshold value of $r_s n_h/\lambda_p$ where n_h is the host refractive index, $n_h = \sqrt{\varepsilon_h}$. The largest CPBG's appear for the close-packed lattice (sphere filling fraction $f = 0.74$) in which case $r_s n_h/\lambda_p \gtrsim 0.9$ is required to open a CPBG. The midgap frequency is then close to ω_p. Since CPBG's increase with ε_h, it is interesting to discuss what happens under the worst circumstances when $\varepsilon_h = 1$. In the latter case the gap width g_w increases rapidly with r_s/λ_p ($g_w = 5.2\%$ for $r_s/\lambda_p \approx 1.013$) and reaches its maximum 9% for $r_s/\lambda_p \approx 1.13$. The resulting band structure for $r_s/\lambda_p \approx 1.013$ ($\omega_p = 9$ in units of $2c/A$, with A being the length of the conventional unit cell of a cubic lattice) is shown in Fig. 1a. The band structure bears clear resemblance to the band structure of purely dielectric fcc photonic crystals made of air spheres [8–10]. Note, however, one significant difference, namely, the opening of a narrow second CPBG (around $\omega = 8.54$ in Fig. 1a), between the corresponding fifth and sixth band of the purely dielectric fcc photonic crystal [8–10]. CPBG's open here at more than 2 times higher midgap frequencies, or, equivalently, for r_s/λ more than twice that for purely dielectric fcc structures [8–10].

It is important to realize that, thanks to the condition $r_s n_h/\lambda_p \gtrsim 0.9$ for opening a CPBG in the frequency region $0.6\omega_p \leq \omega \leq 1.1\omega_p$, absorption is still dominated by bulk properties, i.e., is negligible under our hypothesis, since the absorption induced by surface modes (plasmons) becomes relevant only for particle sizes much smaller than the wavelength [12]. One cannot get rid of absorption completely. Nevertheless moderate absorption was shown to cause only a slight perturbation of the band structure calculated in the absence of absorption [18].

As r_s/λ_p increases further, the CPBG's g_w decreases, however, even more than two CPBG's open in the spectrum, and the similarity of the band structure to that of a purely dielectric fcc photonic crystal is gradually lost. Figure 1b shows four CPBG's in the spectrum for $r_s/\lambda_p \approx 1.35$ ($\omega_p = 12$) and $n_h = 1$, all CPBG's opening below the plasma frequency. Quite remarkable is the appearance of an extremely narrow region of allowed frequencies (formed by two bands) slightly below $\omega = 11$ separating two CPBG's. Many of the applications involving photonic crystals rely heavily on the formation of such a band(s) as an impurity band due to impurities which act as small resonant cavities for photons [4]. Although the origin of the extremely narrow almost dispersionless bands "within a band gap" is different in our case, they still can perform many functions of the impurity band since they involve photons with extremely small group and phase velocities (less than $c/200$).

In real systems, a deviation from the ideal Drude behavior occurs at a proximity of the zero crossing of Reε at some λ_z. Let λ_p be the plasma wavelength extracted from the fit (1) to a material data. Then λ_z is redshifted compared to λ_p [13] and the band structure between λ_z and λ_p can be modified compared to the ideal Drude behavior (1). It is worthwhile to mention that the exact Drude dispersion (1) of ε_s is not necessary to reproduce the exceptional properties of metallodielectric photonic crystals; a reasonable $\varepsilon_s(\omega)$ going from negative values to zero is enough. Many of the above features (except for the extremely narrow almost dispersionless bands within a band gap which become wider) can be also reproduced for a constant and sufficiently small negative ε_s. For instance, let us take $\varepsilon_s = \varepsilon_s(\omega_c) \approx -0.0469$, corresponding to the Drude value of $\varepsilon_s(\omega)$ at the midgap frequency of the top CPBG in Fig. 1b. The corresponding band structure, as shown in Fig. 2, is distorted compared to Fig. 1b and its bottom half is qualitatively similar to that of Fig. 1a. Nonetheless, the band structure exhibits again four CPBG's. It is noteworthy that extreme negative values of ε_s do not help. For a perfect conductor, in which case $\varepsilon_s = -\infty$, no CPBG is found in the spectrum.

We note that the idea of using highly dispersive metallic and semiconductor components for photonic structures is not new [14,18–25]. Indeed, as early as in 1980, periodic systems with metallic scatterers were studied in the long-wavelength limit as a candidate for an efficient selective surface for solar energy purposes [19]. Nevertheless, calculations using the plane-wave method [1] have often been restricted to an extremely low filling fraction f of metallic components, namely, $f \leq 1\%$ [20]. Also the main interest was in microwave [14,22] or even in radio frequency applications [23].

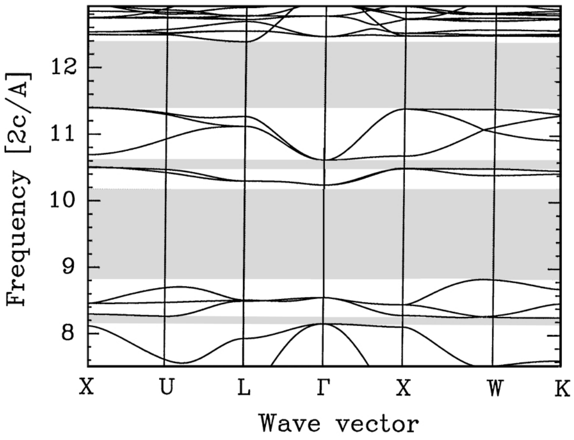

FIG. 2. Calculated photonic-band structure of a close-packed fcc lattice of spheres with a constant negative dielectric constant $\varepsilon_s = \varepsilon_s(\omega_c) \approx -0.0469$, corresponding to the Drude value of $\varepsilon_s(\omega)$ at the midgap frequency of the top CPBG in Fig. 1b.

Surprisingly enough, no systematic search has been made to look for CPBG's in a simple fcc structure of spheres with a Drude-like behavior of the dielectric function. We have shown that monodisperse colloidal suspensions of such spheres can provide three-dimensional CPBG structures with sizable band gaps $\approx 10\%$. This provides a margin large enough for gap-edge distortions due to omnipresent imperfections and impurities to allow both technological and experimental applications involving the proposed structures. Contrary to some previous statements [11,25], there is no need to change the natural simple fcc structure of colloidal systems to achieve a significant CPBG. In addition, the proposed photonic structures can have more than one CPBG and, for r_s/λ_p large enough (≈ 1.3), an extremely narrow almost dispersionless bands within a band gap appear. Dielectric contrast is no longer an issue, since CPBG's open in the spectrum already for a host dielectric constant $\varepsilon_h = 1$ and a CPBG with $g_w \approx 5\%$ can be achieved for sphere filling fractions from $f = 0.55$ till $f = 0.74$ (close-packed case). Moreover, it can be shown that a coating of spheres by an insulator or semiconductor (see Ref. [26] for silica coated silver microspheres) does not destroy the exceptional properties of simple fcc structures of noncoated metallic spheres [27].

The main experimental problem in fabricating the proposed photonic structures is to synthesize large enough spheres to reach the threshold value $r_s n_h/\lambda_p \gtrsim 0.9$. (Surprisingly enough, the same threshold value r_s/λ_p is also required to open a CPBG in a two-dimensional close-packed square lattice of metallic rods embedded in a dielectric [28].) Thus the main task is opposite to that using etching techniques which aim at ever shorter length scales. However, a method to produce monodisperse gold colloids of several hundred nm radius and larger has been developed [29]. Recent results on the fabrication of such spheres from silver, i.e., material with a Drude-like behavior of the dielectric function are promising [30]. The only remaining problem is to control the size polydispersity of spheres and reduce it below 5% to trigger crystallization [7].

The region of plasma frequencies of conventional materials ranges from the near infrared to the ultraviolet [12]. However, in a recent interesting paper [14] it has been shown that a whole new class of artificial materials can be fabricated in which the plasma frequency may be reduced by up to 6 orders of magnitude compared to conventional materials, down to GHz frequencies. Correspondingly, the proposed structures can provide CPBG structures from the GHz up to ultraviolet frequencies. Last but not least, for spheres immersed in a liquid, by applying an electric field one can switch in ms from an fcc colloidal crystal to a body-centered-tetragonal (bct) crystal: a so-called martensitic transition [31]. Therefore, the proposed structures are also promising candidates for the CPBG structures with tunable band gaps.

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