Comment on "Observation of higher-order diffraction features in self-assembled photonic crystals"

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The purpose of this Comment is to show that we find that the conclusions presented in a paper by Nair and Vijaya [Phys. Rev. A **76**, 053805 (2007)], concerning the perfect periodic ordering of self-assembled photonic crystals, are not supported and contradict previous studies of this matter.

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By comparing measured optical spectra and the calculated photonic band structure of colloidal crystals, the authors of Ref. [1] concluded that the "high-energy features are due to the perfect periodic ordering present in the photonic crystals with less defects and disorder." Nevertheless, the theoretical and experimental results previously published in Ref. [2] support the opposite conclusion, that disorder plays a crucial role in determining the shape of measured spectra, especially in the high-energy range.

In Ref. [2], we have predicted rigorously an abruptly fluctuating reflectance for perfectly ordered colloidal crystals for $a>\lambda$ (where a is the lattice constant of the fcc lattice conventional cubic cell) as the reflectance spectrum in Fig. 1(c) of Ref. [2] shows. We explicitly pointed out that none of the spectra reported so far had shown any of these fine features, but much smoother ones, just as shown in the measured spectra presented in Ref. [1]. In particular, the measured reflectance (R) and transmittance (T) spectra shown in Fig. 1(a) of Ref. [1] prove the presence of extinction since conservation of energy is not satisfied ($R+T<1$). The sum of reflection and transmission is less than 60% at the pseudo-band-gap position, which implies that 40% of the intensity is lost. We have found that this loss is due to the diffusely scattered light produced by disorder in the crystalline structure, which removes energy from the specularly reflected and forwardly transmitted beams. Such losses, simulated with a generic extinction in our model, are extraordinarily amplified for $a>\lambda$. This extinction amplification effect is responsible for the shape of the actual reflectance and transmittance spectra and can be simulated in a theoretical calculation by adding an imaginary part ε_i to the dielectric constant of the spheres. We have obtained excellent agreement between theory and the experimental results of three different research groups by means of this approach, as was published in Ref. [2]. This modeling of disorder effects is also supported by Ref. [3], where it is found that the imaginary part ε_i depends on the number of layers of the crystal slab, as Fig. 5 in Ref. [3] shows. This is a clear indication that the extinction observed in experiments is not only due to absorption within the material of the spheres since, in that case, a constant ε_i should be expected as a function of the number of layers. Furthermore, the extinction coefficient for the materials of common use in colloidal crystals is negligible for photon energies in the near-infrared and visible spectra, as the optical properties of polystyrene in Ref. [4] show. The importance of considering the effect of imperfections when analyzing the optical response of a crystal made of colloidal particles has also been demonstrated in Refs. [5–7] for the case of reflectance peaks corresponding to the photonic pseudogap that opens between the lower-energy bands at the L point. Thus, not even for the much more robust first Bragg peak can extinction be neglected.

If we compare the values of ε_i used in Figs. 1(c) (almost perfect crystal, $\varepsilon_i=0.0001$) and 2(c) (imperfect crystal, compatible with measurements, $\varepsilon_i=0.04$) of Ref. [2], we can conclude that the degree of disorder must still be largely reduced in order to get the high reflectance peaks predicted for perfect crystals. This is also the case for the samples studied in Ref. [1], as can be readily seen by comparing Figs. 2(a) and 2(b) of Ref. [1] and Fig. 3(a) of Ref. [2]. From this comparison, an imaginary dielectric constant of around 0.10 can be assigned to the measured reflectance spectra in Ref. [1]. Hence, we have conclusive evidence that there is still a long way to go to achieve true defect-free colloidal crystals.

As a final remark, we should point out that the comparison between the photonic band structure calculated for a perfect, infinite lattice and the experimental results attained for crystals made of a finite number of sphere layers, whose quantity varies typically between one and a few tens of layers, must be done keeping in mind that we are comparing two very different structures. This comparison is based on the assumption that the optical response of real lattices when illuminated by an incident plane wave can be described using the photon modes calculated for the ideal structures without any input or output signal (i.e., the structure is infinite). Although this approximation can be used to explain some spectral features present in large crystals, its validity is limited when we are dealing with slabs composed of a few layers, for which the calculation of their spectral response in reflection and transmission is essential. A very good example of

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the misleading results that arise from a direct comparison between the spectra and the photonic band structure have recently been reported by García-Santamaría *et al.* [8] Moreover, we have proved that the fine features observed in the reflectance and transmittance experimental spectra measured from the highest quality samples so far attained can be rigorously related to multipolar resonances of the sphere ensemble when excited by an external incident plane wave. The effect of extinction is that of smoothing those resonances, only a few robust ones surviving the effect of imperfections. For very large crystals, one could assume that the propagation modes are more closely described by those of the infinite lattice. However, from the analysis of the experimental results of several groups, we can conclude that extinction caused by imperfections makes the response of a real lattice made of six compact sphere layers in the $a > \lambda$ range indistinguishable from the response of thicker ones. Note that this is not the case for the lower-energy range, for which the effect of extinction is less dramatic and the gradual effect of defects on intensity and width of the peak associated with the pseudogap has been studied in crystals whose thickness is comprised between a few and several tens of sphere layers.

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