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Photonic band gap in the visible range in a three-dimensional solid state lattice

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Abstract. We report on the photonic band gap effect in the visible range in a three-dimensional dielectric lattice formed by closely packed silica spherical clusters and by interconnected cavities filled with various liquids. The spectral position and the spectral width of the optical ”stop-band” depend on the lattice period and on the relative sphere/cavity refraction index n. The stop band peak wavelength shows a linear dependence on n. Transmission characteristics of the lattice have been successfully simulated by numerical calculations within the framework of a quasicrystalline approximation.

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An idea of a photonic crystal which behaves with respect to photon waves as a dielectric crystal to electron waves, has been advanced by Yablonovitch [1] and John [2] and stimulated extensive studies in this field [3]. A number of basic issues like photon effective mass, Anderson localization of photons, modification of the photon density of states, inhibition and enhancement of spontaneous emission, coupled atom-field states, and others are currently being discussed in relation to three-dimensional dielectric lattices [4–6]. In spite of the significant progress in theoretical analysis of these problems, experimental studies are still at the preliminary stage. The photonic band gap effect has been observed in the radio frequency range [7,8], whereas the main field of the application of the new class of phenomena is expected to be optics and laser physics. Experiments in the optical range have been carried out for one-dimensional lattices (see Ref. 9 and refs. therein) in which an influence of spatial arrangement on photon localization and on the spontaneous emission rate have been observed. A breakthrough towards actual photonic crystals with the forbidden frequency gap corresponding to the optical range has been recently made by Tarhan et al. [9] who have demonstrated a formation of a pseudogap in the photon density of states in an ordered ensemble of spherical polystyrene particles. The pseudogap does manifest itself as a stop band in the transmission (reflection) spectrum of a purely refractive medium without dissipative losses. The stop bands arise due to diffraction of an optical wave on a threedimensional lattice made of dielectric particles.

Solid state photonic crystals exhibiting band gap in the optical range open up much more possibilities for research and applications. In the present communication we report on the photonic band gap effect in artificial opals made of closely packed SiO_2 spheres arranged in a cubic lattice.

Three-dimensional dielectric lattices have been developed from a sol of artificially grown monodisperse spherical SiO_2 particles. The regular structure arises due to close packing of monodisperse spheres. The lattice obtained after precipitation of the particles and subsequent drying is fragile because of weak bonding between silica globules. The lattice is hardened by hydrothermal treatment and annealing, and point contacts between the globules are converted into faceted ones. Under certain conditions voids form a regular sublattice that can be impregnated with liquid or solid inclusions. These structures are known as artificial opals and their properties and technology have been described previously [11].

Structures fabricated in such a manner consist of nearly spherical silica clusters of a size ranging from 0.2 to 0.3 μm arranged in a face-centered cubic (f.c.c.) lattice (Fig. 1). Each silica particle in its turn has an internal substructure [12]. Due to the internal substructure, the effective refraction index $n^*_{SiO_2}$ of silica particles ranges from the value $n^*_{SiO_2}$ = 1.45 inherent in a bulk silica down to $n^*_{SiO_2}$ = 1.26. All the lattices show a dip in the optical transmission spectrum with spectral position depending on the lattice period a_L and on the $n^*_{SiO_2}$ value. The nature of the spectrally selective transmission in a disperse medium without dissipation is nothing but multiple scattering and interference of light waves. It can be intuitively understood in terms of Bragg diffraction of optical waves. In other words, a formation of a pronounced stop band is indicative of a reduced density of

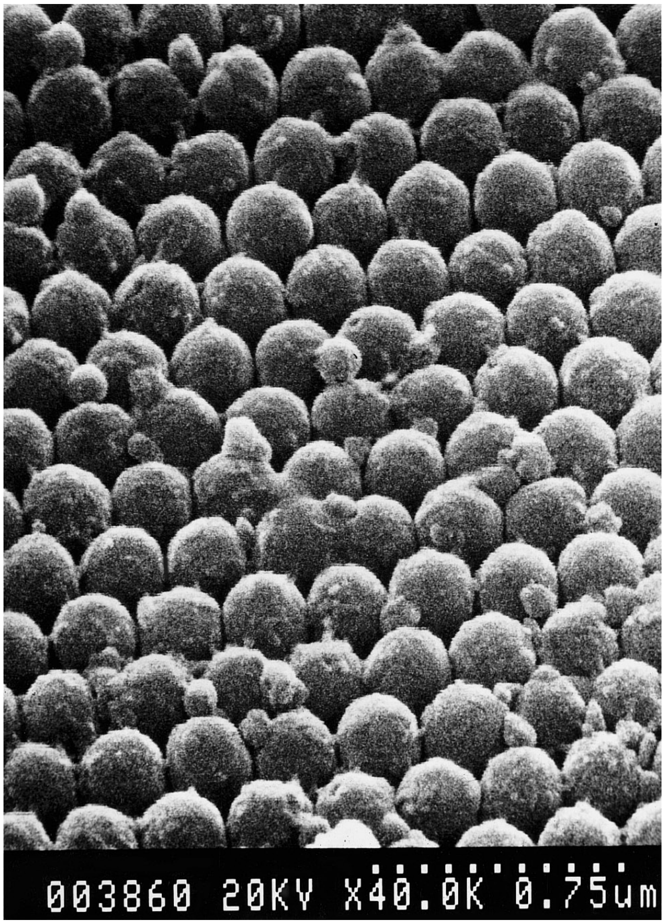

Fig. 1. A microphotograph of the opal surface

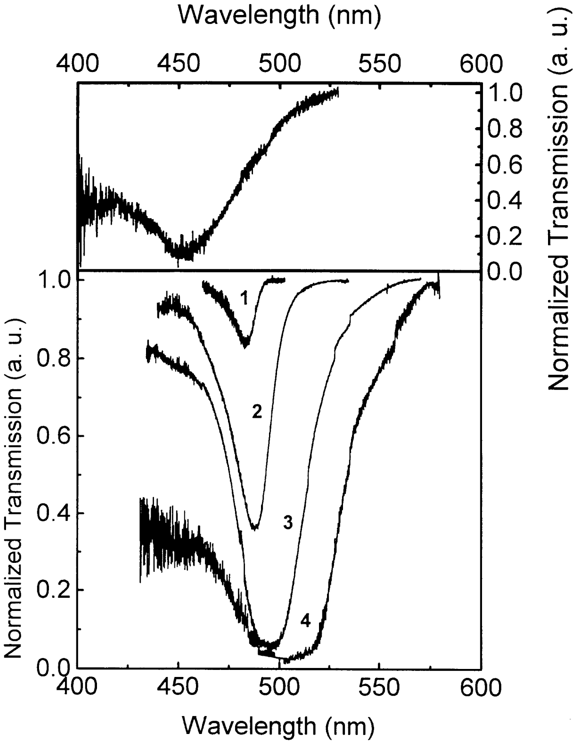

Fig. 2. Optical transmission spectra of the same sample with empty voids (*upper panel*) and impregnated with various fillers (*lower panel*): methanol (**1**), n = 1.3284, ethanol (**2**), n = 1.3614, cyclohexene (**3**), n = 1.4262, and toluene (**4**) n=1.4969

photon states inside the sample and thus can be classified as a photonic pseudogap phenomenon.

Intersphere voids form a network in which cavities of two types and channels connecting them can be distinguished. The voids can be impregnated with materials having refraction indices either higher or lower than those of the spheres. Therefore, it is possible to control the spectral position and contrast of the stop band. Figure 2 shows spectral characteristics of the same matrix filled with various liquids. With increasing the refraction index of the voids relative to that of the spheres the stop band peak wavelength λ_0 shifts linearly towards longer wavelengths (Fig. 3). Experimentally measured positions of the stop band peaks for a dry ($n_{\text{voids}} < n_{\text{spheres}}$) and impregnated sample ($n_{\text{voids}} > n_{\text{spheres}}$) fit the common straight line. This is probably a manifestation of the behavior known from the optics of scattering media: the scattering features from the two spatially complementary ensembles of particles, e.g., dense spheres in an empty space and, vice versa, hollow spherical voids in a dense medium, are basically equivalent. With an increase in the value of $n_{\text{voids}}/n_{\text{spheres}}$ a dip in the transmission spectrum becomes deeper and wider. The maximum transmission contrast reached in our experiments was about 10^3, the spectral width being 40 nm at the 10%-level (**curve 4** in Fig. 2).

The spectral position and the amplitude of the stop band depends also on the angle between the direction of propagation of light and crystal axes. Optical properties of different samples with variable crystal orientation filled with the same liquid confirmed the f.c.c. structure of the lattices [13].

As the cavities can be impregnated with various liquids, it is of principal importance to examine the effect or the photonic band gap on the spontaneous emission of the molecules resonant to the pseudogap. The first results providing an angular-dependent dip in the emission spectrum of the dye molecules and a deviation in the fluorescence kinetics will be published in the forthcoming paper [14].

An appearance of localized states and a formation of dips in transmission spectra of photonic crystals originate from interference of waves that experience multiple scattering by a partially ordered system of mesoscopic particles. To calculate optical characteristics of such a system, a technique based on the statistical multiple wave scattering theory (MWST) can be used. This technique is widely used presently in studies of the interaction of electromagnetic radiation with randomly inhomogeneous media (see, e.g., Ref. 15). In Ref. 16 a technique based on MWST has been proposed which provides computation of coherent transmission and reflection of a multilayer scattering medium consisting of the periodically alternating continuos and dense disperse layers. When the thickness of continuos layers approaches zero, the structure of such a system becomes similar to that of photonic crystals if the packing of particles in disperse monolayers is extremely high.

In order to describe the spectral features of transmission and reflection of a multilayer scattering medium consisting of a system of correlated scatterers, one should account for cooperative interference effects, namely coherent rescatter-

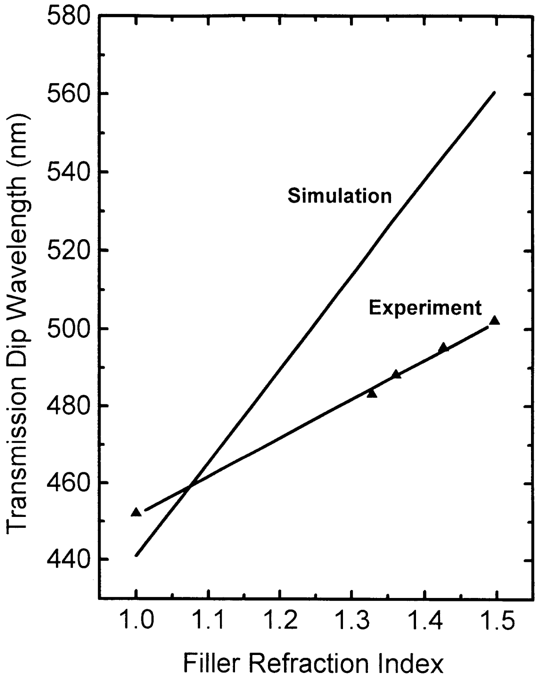

Fig. 3. Stop band peak dependence on the refraction index of the filler (calculated and measured)

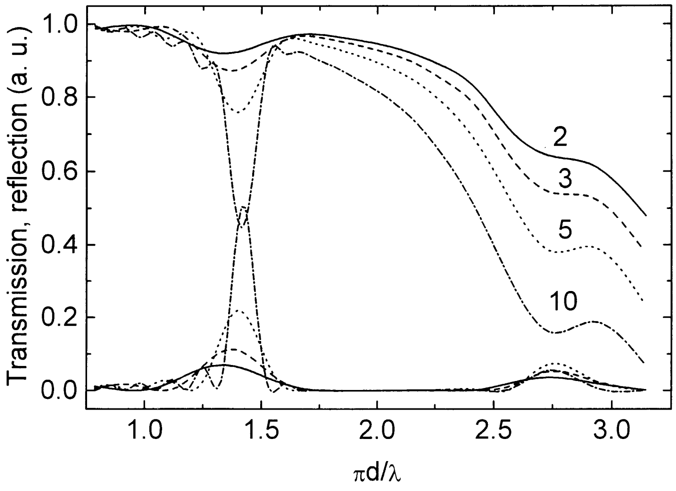

Fig. 4. Optical transmission and reflection spectra for a set of sequentional layers consisting of a closely packed spheres with cubic symmetry as a function of a dimensionless parameter $\pi d/\lambda$, where d is the sphere diameter and λ is the photon wavelength. Numbers at the curves indicate the number of the layers. Refraction index of spheres versus voids is 1.26

ing on particles and interference of scattered waves. Cooperative effects in a single monolayer are usually considered in a quasicrystalline approximation [17, 18]. On the assumption of the statistical independence of the individual monolayers it is possible at first to find the scattering amplitude of a single monolayer taking into account the multiple rescattering of the particles within the layer and then to account for the irradiation between the different monolayers of the sample under consideration. The computational procedure is described in Ref. 13. Here we omit a description of the technique and provide the results of the numerical analysis only.

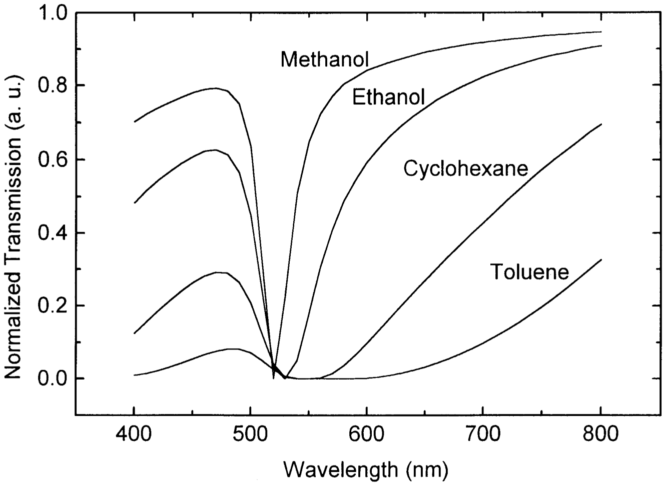

Fig. 5. Calculated transmission spectra of the multilayered close-packed system consisting of the spherical particles. The particle diameter is d = 0.2 μm, the particle refraction index is n_{eff} = 1.26, the number of the layers is N = 300. The refractive index of the medium embedded in the voids between the particles is n_{fil} = 1.0 (**1**), 1.3284 (**2**), 1.3614 (**3**), 1.4262 (**4**), and 1.4969 (**5**)

Numerical calculations have been made for a layered system consisting of close packed monolayers of spherical particles with hollow voids and with voids filled with another material. The density of particle arrangement within a given monolayer was close to the maximal one. The interlayer spacing defined as the distance between the two planes plotted through the centers of the particles in two neighboring monolayers was taken to be equal to the particle diameter. The spectral dependence of the coherent transmission and reflection of the system is presented in Fig. 4 for a set of the monolayer numbers. It is seen that a dip in the transmission spectrum and the corresponding maximum in the reflection spectrum appear which are enchanced with growing number of the layers.

Transmission spectra for the systems with the filled voids are shown in Fig. 5. With growing refraction index of the embedded material, the dip is getting deeper and shifts towards long-wave range. The computed spectral position of the dip for the structures similar to that examined experimentally is presented in Fig. 3. One can see that the calculated and the measured dependencies of the dip minima versus the refraction index of the filler are qualitatively the same. The theory gives the sharper growth of the dip peak wavelength versus refractive index of the filler. The possible reason for this discrepancy might be connected with a partial impregnation of the silica globules by the filler resulting in a deviation of their effective refraction index.

In conclusion, we have demonstrated a formation of the photonic frequency stop band in a three-dimensional dielectric lattice consisting of the two sublattices of the purely reactive media without dissipative losses. The numerical simulations based on the quasicrystalline approximation show a reasonable agreement of the calculated and the measured spectral features. The results obtained can be interpreted in terms of the renormalization of the photon density of states and the formation of a photonic pseudogap. The further progress towards solid-state photonic crystals can be achieved by enhancement of the relative refraction to get the true band gap in all directions. On the next step embedding of light absorbing and emitting species like rare-earth ions, organic molecules and semiconductor quantum dots will provide experimenters an opportunity to test the basic effects that are of great theoretical and practical importance.

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