Optical anisotropy of cubic photonic crystals under conditions of multiple-mode light propagation

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Abstract. Bragg reflection spectra of light are studied for opal-like photonic crystals made of polystyrene spheres. A resonant enhancement of reflectivity is observed in cross-polarization configuration of the analyzer and polarizer when varying the azimuthal orientation of a sample in respect to the incidence plane. The cross-polarization effect takes place at oblique incidence of light on the lateral (111) crystal plane with the plane of incidence being non-perpendicular to the inclined (11-1) crystal plane. The effect is shown to be due to the multiple Bragg diffraction of light when the resonant Bragg conditions are fulfilled at a certain angle of incidence and azimuth for the lateral and inclined crystal planes simultaneously.

1. Introduction
Opal-like structures considered as a typical example of three-dimensional (3D) photonic crystals (PhC) have received much attention during last decades both due to their non-trivial optical properties and potential applications in devices [1, 2]. A number of interesting and still not adequately explored phenomena occurring in the frequency range of low energy stop-bands are associated with the multiple Bragg diffraction (MBD) effects [3-6]. Most prominently, these phenomena are manifested in reflection spectra at large angles of incidence as a doublet structure of the reflectance contour [6-8].

As a rule, the simplest geometry of reflection is chosen so that the incidence plane of light is perpendicular to both the lateral (111) and inclined (11-1) crystal planes, which corresponds to the incidence plane azimuth $\phi = 0$ (see Fig.1). Such geometry allows one, in a rather simple way [5], to analyze separately the contribution to the reflection spectrum from the TE- and TM- electromagnetic modes propagating inside an opal-like PhC: these modes are excited in s- and p-polarization of incident light, respectively.

In this work, we discuss novel optical phenomena associated with resonant polarization mixing of electromagnetic modes in opal-like photonic crystals, which is the case, in spite of the high, fcc, symmetry of the sample, when the azimuth $\phi$ differs from $m\pi/3$ values ($m=0,1,\ldots,5$).

2. Experimental
The opal-like PhCs under study were prepared from monodispersive polystyrene spheres self-assembled to the films of about 5 μm in thickness. The particles were synthesized by styrene and
methacrylic acid emulsifier-free emulsion copolymerization. They were initiated by potassium persulfate, had a spherical shape, uniform surface and particle size distribution less than 2%. The Bragg reflection spectra were measured in s-s, p-p and p-s configurations of polarized light when incoming s- or p-polarized radiation was incident on the lateral reflecting surface (111) of the PhC film.

The azimuth angle \( \phi \) of the sample was measured from zero-value which corresponded to the perpendicular orientation of the incidence plane in respect to the inclined crystal planes (020) and (11-1). The sample was precisely oriented using measured Bragg reflection spectra under the MBD conditions [6].

![Figure 1. Geometry of Bragg reflection of light from an opal-like photonic crystal: the XY plane of the reference system coincides with the (111) lateral (reflecting) surface of the sample, the azimuth \( \phi \) of the incidence plane is reckoned from the XZ plane which is perpendicular to the inclined crystal planes (11-1) and (002), \( \theta \) is the angle of incidence.](image)

Figure 2 shows the s-s reflection spectra obtained at the azimuthal orientation \( \phi = 15^0 \) of the plane of incidence. In Fig. (a), the topogram of all the spectra measured is depicted where, together with the main reflection peak band, the reflectance deep is clearly seen crossing the peak band in the incidence angle range \( \sim 50^0 < \theta < 62^0 \). Some reflectance contours recorded at three angles of incidence are correlated with the topogram in Fig. (b).
Figure 2. (a) The topogram of measured reflectance in s-s configuration of polarization at the azimuth $\varphi = 15^0$ of the incidence plane; superimposed lines are calculated with the formulae (1) and (2) giving Bragg conditions for light reflection from the crystal planes (111) (black dashed line 1) and (11-1) (white solid line 2). (b) s-s reflectance spectra obtained at three different angles of incidence $\theta$, as indicated. $a_{00}$ is the smallest distance between the particle centers in the (111) plane, $\lambda$ is the wavelength of incident light.

A very surprising effect appears when measuring the same spectra in p-s cross-polarization configuration (see Fig. 3). Instead of expected absence of reflection signal, there exists a rather bright spot on the topogram (Fig.a). This spot is observed in the region where the deep in s-s reflection is registered (cf. Fig. 2), the maximum value of the p-s reflectance being about as high as 10%! It should be emphasized here that with decreasing azimuth $\varphi$ (up to $\varphi = 0$) the p-s reflection intensity tends to zero.

Figure 3. (a) The topogram of measured reflectance in p-s cross-polarization configuration at the azimuth $\varphi = 15^0$ of the incidence plane; superimposed lines are calculated with the formulae (1) and (2) giving Bragg conditions for light reflection from the crystal planes (111) (white dotted line 1) and (11-1) (white solid line 2). (b) p-s reflectance spectra obtained at three different angles of incidence $\theta$, as indicated. $a_{00}$ is the smallest distance between the particle centers in the (111) plane, $\lambda$ is the wavelength of incident light.
3. Discussion

In accordance with the technique developed in [6], based on comprehensive analysis of Bragg reflection spectra at $\phi = 0$, we carefully measured the parameter values of the PhC sample under study. The mean distance between neighboring particles placed in the lateral plane is $a_{00} = 279$ nm, the average dielectric constant is $\varepsilon_0 = 2.185$, the coefficient of uniaxial compression along [111] direction is $\eta = 0.94$. These values allow us to describe in a good approximation [7] the spectral positions $\lambda_{111}$ and $\lambda_{11-1}$ of the main features appearing in reflection spectra using the following Bragg-type formulae:

$$\lambda_{111} = \sqrt{8/3}a_{00}\eta\sqrt{\varepsilon_0 - \sin^2 \theta},$$

$$\lambda_{11-1} = \frac{\sqrt{6}a_{00}\eta}{1 + 2\eta^2}\left(\eta\sqrt{2}\sin \theta \cos \varphi + \sqrt{\varepsilon_0 - \sin^2 \theta}\right),$$

where the indices 111 and 11-1 differ the wavelengths $\lambda$ are referred to Bragg diffraction from the corresponding crystal planes. As follows from above formulae, only $\lambda_{11-1}$ depends on the azimuth $\varphi$ of the incidence plane, $\lambda_{111}$ gives the s-s reflection peak band position as a function of the angle of incidence $\theta$, whereas $\lambda_{11-1}$ describes the behavior of the s-s reflection deep.

Figure 2 demonstrates clearly that the spectral positions of the reflectance maxima and deeps are fitted well by Eqs. (1) and (2), respectively. This allows us to consider unambiguously the spectral deep observed at the 15° azimuth in the vicinity of the interception point

$$\lambda_\phi = \frac{4\sqrt{3}a_{00}}{4 - \eta^2}\sin \theta_\phi \cos \varphi,$$

for the theoretical curves (1) and (2) as being due to the MBD effect. Because the enhancement of reflectivity in cross-polarization configuration (see Fig. 3) takes place also in the vicinity of this interception point, we can ascribe such enhancement to the MBD effect, as well.

![Figure 4](image_url)

**Figure 4.** The eigenmode energy spectra (a,b) of the opal-like PhC as compared with the reflection spectrum for p-s cross-polarization at the angle of incidence $\theta = 54^\circ$ (a), 56° (b,c) and at the azimuthal angle $\varphi = 0^\circ$ (a), 15° (b,c). At $\varphi = 0^\circ$ the p-s signal disappears practically. On the panel (c), the experimental and theoretical data are represented by points and the solid line, respectively.
When calculating energy and reflectance spectra we took into account 3 reciprocal lattice vectors $G_{111}$, $G_{11-1}$ and $G_{020}$ which form the Bloch mode amplitude in the vicinity of the $\lambda* \pi$ point (3). Fig. 4(a) shows that at $\varphi = 0^\circ$ inside the crystal there exist the electromagnetic mode which can be classified as TE- and TM-modes. It worth to note that the TM5 and TE6 curves intersect at ~$\lambda*$ However when $\varphi \neq 0^\circ$ (in our case $\varphi = 15^\circ$, see Fig. 4(b)) the anti-crossing region arises around the point $\lambda*$, and polarization mixing between all the modes 1-6 takes place. Such kind of mixing is more essential in the vicinity of the crossing point. Therefore the maximum of the cross-polarization signal should be located near this point. As shows the panel (c) the calculated curve is well fitted to the experimental data. As follows from our theoretical calculations, the FWHM of the p-s reflectance contour is determined by the lifetime $\tau = \Gamma^{-1}$ of the electromagnetic state. We have obtained the value $\Gamma = 50$ meV which allowed us to describe theoretically well all our experimental data including the reflectivity contours in p-p and s-s configurations.

It should be noted that in spite of a weak uniaxial anisotropy of the sample under study the observed cross-polarization effect does not associate with such kind of anisotropy and is, in fact, the intrinsic property of an ideally cubic photonic crystal with the opal-like structure. There exist a number of techniques for preparing artificial opals which allow one to make samples with certain optical and other properties (see, for example, [9] and references therein). Unfortunately, preparing opal-like PhCs with ideal cubic symmetry meets, as a rule, specific technological difficulties which can lead to a number of additional experimentally observed anisotropy effects (as it was pointed, in particular, in [9-11]). The cross-polarization effect analysed in our work is described adequately in the framework of the dynamical Bragg diffraction theory [6, 8]. Making use of this theory we present for the first time detailed comparison between the experimental and theoretical cross-polarization spectra and established the characteristic lifetime ~ 0.1 ps of the electromagnetic state in the vicinity of the stop-band, which corresponds to the measured $\Gamma$ value.

4. Conclusion
The results presented in this work demonstrate that in spite of high (nearly cubic) symmetry of opal-like photonic crystals their optical properties exhibit strong anisotropy which becomes very much more pronounced when the multiple Bragg diffraction conditions are fulfilled. In such a case, the resonant polarization mixing of diffraction processes governed by two or more systems of crystal planes takes place. So, for some directions of propagation, the TE- or TM-classification of electromagnetic modes fails because other eigenmodes are excited with polarization states different from ones of strongly defined TE- and TM-modes. As a result, at p- or s-input polarization, reflected light becomes elliptically polarized, which appears experimentally as a resonant cross-polarization effect. It should be concluded that this effect can be considered as a new direct manifestation of the multiple Bragg diffraction in 3D photonic crystals.

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