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Dependence of anisotropy for opal and inverse opal infiltrated with liquid crystal

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Recently the opal and inverse opal with a three-dimension regular structure have attracted much attention as pseudo type of photonic crystals from both fundamental and practical viewpoints, because novel concepts such as a photonic band gap (PBG) have been predicted, and various novel applications of the photonic crystals have also been proposed [1,2]. For many applications it is useful to obtain some degree of tunability of the photonic band structure through electo-optic effect. Therefore we have proposed to realize new functionality by the infiltration of liquid crystals into the interconnected nanoscale void of opal and inverse opal. The property of liquid crystals can be easily controlled by relatively low voltage. In earlier works a rather pessimistic conclusion regarding the efficacy of birefringent photonic crystals was drawn [3,4], but recently crystals have been suggested by both experimental and theoretical work [5,6].

In this study, we suppose that light incidents on the (1,1,1) director of a real spatial face centered cubic (fcc) structure of an opal and an inverse opal into which the various nematic liquid crystals are infiltrated, and illustrate the principle of the fully tunable electromagnetic device in detail. An opal and an inverse opal infiltrated with the liquid crystal we studied have not the complete PBGs but the PBGs only for the certain incident light wave, which may be trivial in the case of isotropic materials, but in the case of the anisotropic materials such as liquid crystals, even if they have the PBGs only for the certain incident light wave, we can acquire the significant informations in investigating them.

Following K.Busch and S.John's discussion [6], in order to determine the photonic band scheme of anisotropic crystals, we start with the wave equation satisfied by the magnetic field for a three-dimensional periodic array of scatters.

$$\nabla \times [\epsilon^{-1} \nabla \times \mathbf{H}(\mathbf{r})] = \frac{\omega^2}{c^2} \mathbf{H}(\mathbf{r}) \quad (1)$$

where $\nabla \cdot \mathbf{H}(\mathbf{r}) = 0$. Using the Bloch theorem, we may expand the magnetic field as

$$\mathbf{H}(\mathbf{r}) \equiv \mathbf{H}_\mathbf{k}(\mathbf{r}) = \sum_{\mathbf{G}} \mathbf{h}_\mathbf{G} e^{i(\mathbf{k}+\mathbf{G}) \cdot \mathbf{r}} = \sum_{\mathbf{G}} \sum_{\lambda=1}^2 h_{\mathbf{G}}^\lambda \hat{\mathbf{e}}_\mathbf{G}^\lambda e^{i(\mathbf{k}+\mathbf{G}) \cdot \mathbf{r}} \quad (2)$$

where λ label the two transverse polarizations for any plane wave such that $\hat{\mathbf{e}}_\mathbf{G}^{\lambda=1,2}$ and $\mathbf{k}+\mathbf{G}$ form an orthogonal triad. Because of the discrete translational symmetry of the lattice, the wave vector \mathbf{k} labeling the solutions may be restricted to lie in the first Brillouin zone (BZ). Consider the optimized opal consisting of backbone material with total volume fraction of 75.5% and the optimized inverse opal consisting of backbone material with total volume fraction of 24.5%. The former consists of an fcc lattice of the opal and possesses 24.5% air void sphere, and the latter consists of an fcc lattice of 75.5% closed packed air spheres and possesses 24.5% isotropic dielectric sphere. Suppose that their air spheres themselves are fully infiltrated with nematic liquid crystals now. The isotropic dielectric index for the backbone of the opal and inverse opal infiltrated with liquid crystals is $\epsilon_{\text{opal}} \approx 2.13$ and $\epsilon_{\text{inverse opal}} \approx 2.43$, respectively, while the anisotropic dielectric tensor of liquid crystals is

written as

$$\boldsymbol{\epsilon}_{LC} = \mathbf{O}(\theta, \phi) \text{diag}(\epsilon_{LC}^o, \epsilon_{LC}^o, \epsilon_{LC}^e) \mathbf{O}^T(\theta, \phi) \quad (3)$$

where $\mathbf{O}(\theta, \phi)$ is a rotation matrix which act on the corresponding diagonalized tensor. The angles θ and ϕ describe the orientation of the director $\hat{\mathbf{n}}$ with respect to the opal and inverse opal coordinate system. We choose photopolymer NOA61 as the inverse opal. For bulk nematic liquid crystal we may choose a coordinate system entries ϵ_{LC}^o - ordinary dielectric index, parallel and ϵ_{LC}^e - extraordinary dielectric index, perpendicular to its director $\hat{\mathbf{n}}$, respectively. For both the opal and the inverse opal, the coordinate system is fixed by the bulk backbone and the director $\hat{\mathbf{n}}$ can have different orientations with respect to this reference frame.

Inserting Eqs. (2) into Eqs. (1) results in an infinite matrix eigenvalue problem

$$\sum_{\mathbf{G}} \sum_{\lambda=1}^2 \mathbf{H}_{\mathbf{G}\mathbf{G}}^{\lambda\lambda} h_{\mathbf{G}}^{\lambda} = \frac{\omega^2}{c^2} h_{\mathbf{G}}^{\lambda} \quad (4)$$

where

$$\mathbf{H}_{\mathbf{G}\mathbf{G}} = [\mathbf{k} + \mathbf{G}] [\mathbf{k} + \mathbf{G}]^T \begin{bmatrix} \hat{\mathbf{e}}_2 \cdot \mathbf{\epsilon}_{\mathbf{G}-\mathbf{G}}^{-1} \cdot \hat{\mathbf{e}}_2 & -\hat{\mathbf{e}}_2 \cdot \mathbf{\epsilon}_{\mathbf{G}-\mathbf{G}}^{-1} \cdot \hat{\mathbf{e}}_1 \\ -\hat{\mathbf{e}}_1 \cdot \mathbf{\epsilon}_{\mathbf{G}-\mathbf{G}}^{-1} \cdot \hat{\mathbf{e}}_2 & \hat{\mathbf{e}}_1 \cdot \mathbf{\epsilon}_{\mathbf{G}-\mathbf{G}}^{-1} \cdot \hat{\mathbf{e}}_1 \end{bmatrix} \quad (5)$$

and $\mathbf{\epsilon}_{\mathbf{G}-\mathbf{G}}$ is the Fourier coefficient of $\mathbf{\epsilon}(\mathbf{r})$.

For numerical purpose Eq. (5) is truncated by retaining only a finite number of reciprocal lattice vector [7]. In the case of birefringent or biaxial dielectric materials, the dielectric tensor $\mathbf{\epsilon}(\mathbf{r})$ in Eq. (2) is real and symmetric. We executed calculations at $N = 245$. Eigenfrequencies calculated with Ho, Chan and Soukoulis 's method for $N = 245$ and $N = 567$, respectively, differ at most by 0.1% of their absolute value thanks to the high convergence of the HCS method. In addition, it has been reported that eigenfrequencies calculated with Ho, Chan and Soukoulis 's method for $N = 531$ and $N = 1219$, respectively, are converged to approximately 0.01% [6]. Therefore we can acquire the accurate calculational result at $N = 245$.

Their phenomena are shown by investigating at $\pi/a(1,1,1)$ point in the reciprocal lattice that is parallel to the director of light, one of several L points that constitute the BZ. The L point we mention below indicates $\pi/a(1,1,1)$ point in the reciprocal lattice. As is well known, at L point of photonic band schemes of the fcc structure for the isotropic material, first and second band degenerates, similarly third and fourth band also degenerates but there exists the gap between second and third band. In the case of anisotropic materials, however, their

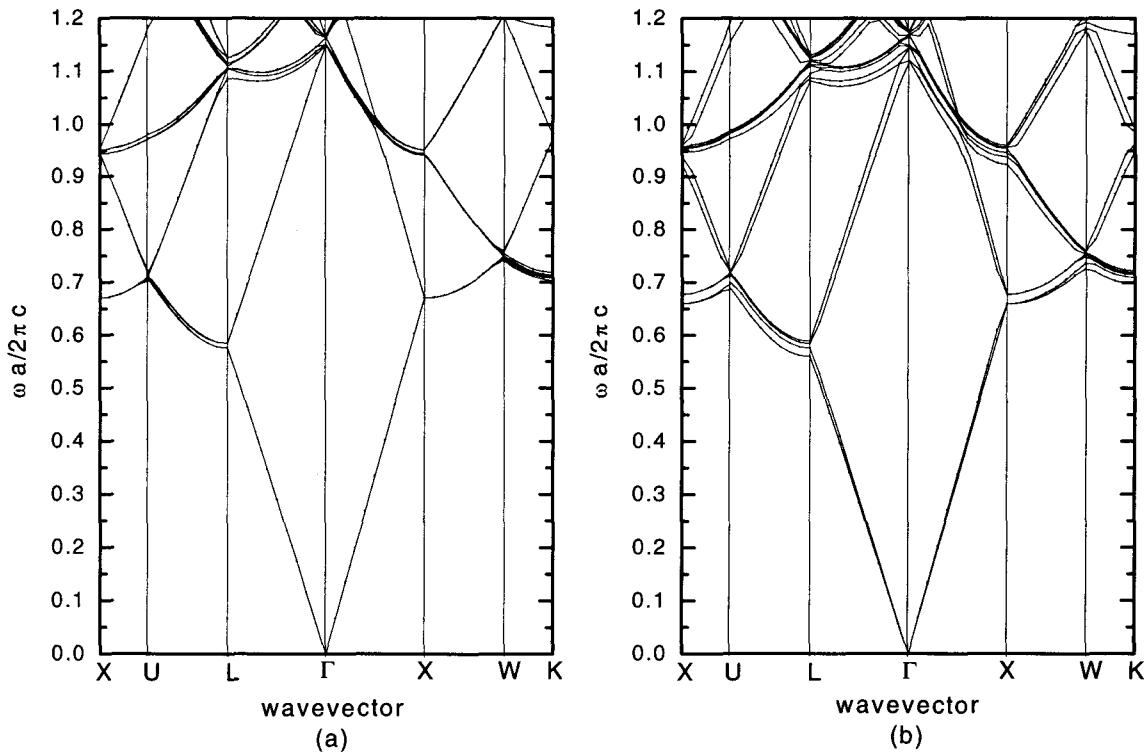


FIG.1. (a) Band structure of the opal ($n_{\text{opal}} = 1.46$) infiltrated with liquid crystal ($n_{\text{LC}} = (n_{\text{LC}}^e + 2n_{\text{LC}}^0)/3 = 1.584$, $n_{\text{LC}}^e = 1.706$, $n_{\text{LC}}^0 = 1.522$) when each director of the nematic axis turns at random, that is, isotropic material. (b) Band structure of the opal ($n_{\text{opal}} = 1.46$) infiltrated with liquid crystal ($n_{\text{LC}}^e = 1.706$, $n_{\text{LC}}^0 = 1.522$) when the director of the nematic axis turns at $\phi = \pi/4$ and $\theta = -\pi/2 + \cos^{-1}(1/\sqrt{3})$, that is, it is perpendicular to (1,1,1) director of the light. X, U, L, Γ , W and K point are $2\pi/a(1,0,0)$, $2\pi/a(1,0,0)$, $2\pi/a(1/2,1/2,1/2)$, $(0,0,0)$, $2\pi/a(1,1/2,0)$ and $2\pi/a(3/4,3/4,0)$, respectively in the reciprocal lattice.

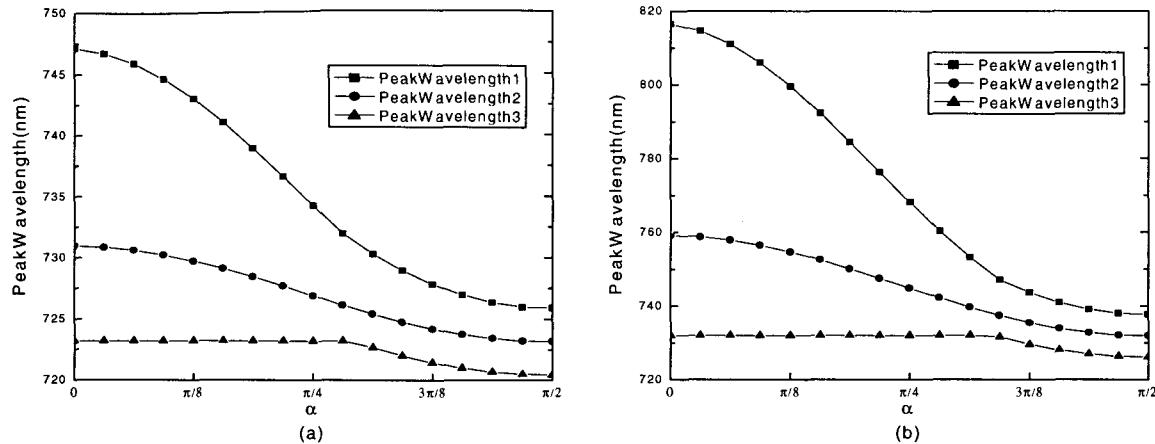


FIG.2. Dependence of the reflective PeakWavelength1, 2 and 3 for (a) the opal ($n=1.46$) infiltrated with liquid crystal ($n_{LC}^e = 1.706, n_{LC}^o = 1.522$) and (b) the opal infiltrated with liquid crystal ($n_{LC}^e = 2.223, n_{LC}^o = 1.590$) on the orientation of the nematic director $\hat{n}(\phi, \alpha)$ for fixed angle $\phi = \pi/4$ at L, $\pi/a(1,1,1)$ point in the reciprocal lattice.

degeneracies disappear and new gaps between first and second band and between third and fourth band are created as shown in Fig.1, that is, there exist three reflective peak wavelengths for anisotropic materials although there exists only one reflective peak wavelength for isotropic materials. We define peak wavelengths reflected from band gaps between first and second band, between second and third band and between third and fourth band as PeakWavelength1, PeakWavelength2 and PeakWavelength3, respectively. The peak wavelength is defined as the center point of the band gap.

We evaluate the peak wavelength of the opal and the inverse opal infiltrated with liquid crystals with various indices of refractive for $\phi = \pi/4$ and α , where $\alpha = \theta + \pi/2 - \cos^{-1}(1/\sqrt{3})$ ranges from 0 to $\pi/2$ in terms of spherical coordinates. For $\alpha = 0$ the nematic axis is perpendicular to the director of light and the anisotropy is the biggest at L point, while for $\alpha = \pi/2$ the nematic axis is parallel and the anisotropy disappears at L point. We investigate the opal infiltrated with two kinds of liquid crystals with $n_{LC}^e > n_{LC}^o$. One is the liquid crystal whose principal indices of refraction are $n_{LC}^e = 1.706$ and $n_{LC}^o = 1.522$ corresponding to those of 5CB, the other is the liquid crystal whose principal indices of refraction are $n_{LC}^e = 2.223$ and $n_{LC}^o = 1.590$ [8], and choose $a = 300\sqrt{2}$ (nm).

In Fig.2 we display the dependence of PeakWavelength on α ranging from 0 to $\pi/2$. For the opal infiltrated with the former liquid crystal in Fig 2. (a) and for the opal infiltrated with the latter liquid crystal in Fig. 2 (b). In Fig.2 (a) and (b) although all of PeakWavelength1, PeakWavelength2 and PeakWavelength3 monotonously decrease as the angle α approaches to $\pi/2$, scales of the whole changes of PeakWavelengths in Fig.2 (a) extremely differ from those in Fig.2 (b). That is, the scales of the latter are much bigger than those of the former.

Similarly the same calculation was executed on the inverse opal. The results on the inverse opal infiltrated with the former liquid crystal and on the inverse opal infiltrated with the latter liquid crystal are shown in Fig 3. (a) and Fig 3. (b), respectively. As evident in the figures, the larger the anisotropy of the liquid crystal, the bigger the scale of the whole PeakWavelengths. Therefore the liquid crystal with larger anisotropy exhibits larger, the bigger the scale of the change of PeakWavelengths regardless of the opal or the inverse opal.

These results clearly show that the tunability of the photonic crystal can be realized by the opal and the inverse opal infiltrated with liquid crystal.

By the way, so far it was not clarified which reflection peak among PeakWavelength1, PeakWavelength2 and PeakWavelength3 is the strongest. In the experimental study, the existence of several reflection peaks and the stepwise change (hop) of the peak have been observed. Though detailed behavior is not clear, following discussion

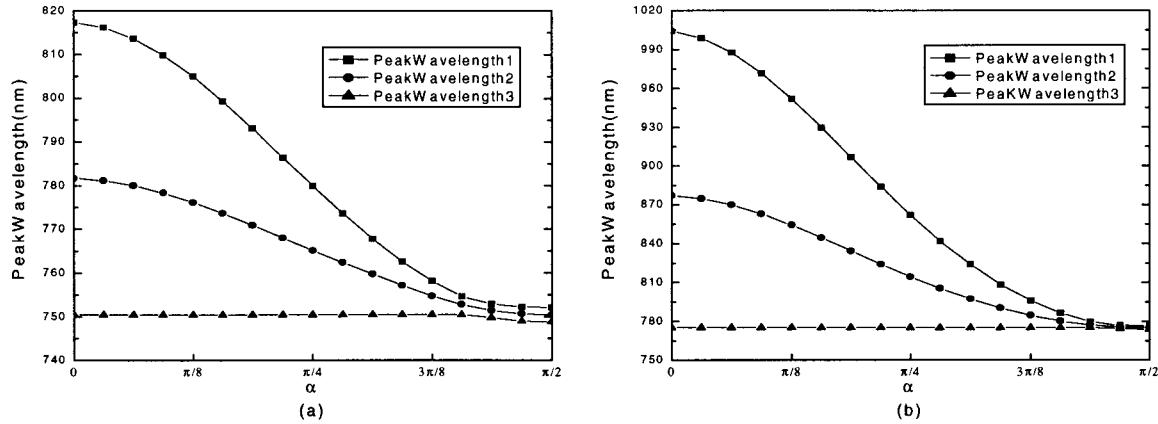


FIG.3. Dependence of the reflective PeakWavelength1, 2 and 3 for (a) the inverse opal ($n=1.56$) infiltrated with liquid crystal ($n_{LC}^e=1.706, n_{LC}^o=1.522$) and (b) the inverse opal infiltrated with liquid crystal ($n_{LC}^e=2.223, n_{LC}^o=1.590$) on the orientation of the nematic director $\hat{n}(\phi, \alpha)$ for fixed angle $\phi=\pi/4$ at L, $\pi/a(1,1,1)$ point in the reciprocal lattice.

is possible by interpreting that the PeakWavelength reflected from the biggest band gap dominates and exhibits the strongest intensity. For example, in Fig.2 (a), PeakWavelength1 possesses the biggest band gap at $\alpha=0$, while PeakWavelength2 does by $\alpha=\pi/2$. Because the material becomes more isotropic and the band gaps corresponding to PeakWavelength1 and 3 tend to zero as the angle α approach to $\pi/2$. Therefore the hop from PeakWavelength1 to PeakWavelength2 may be observed in measuring the reflective PeakWavelength. Moreover, the anisotropy of the liquid crystal may cause the complete PBG in the photonic crystals of the fcc structure. Generally there exist degeneracies in the wavevectors along X- U- L and X- W- K in them [7]. As shown in Fig.1, however, their degeneracies disappear and gaps generate. In conventional fcc photonic crystals which possess high dielectric contrasts, therefore, there may exist the complete PBG under the influence of the liquid crystal.

In conclusion, we have solved the problem of the light propagation for the director of real spatial (1,1,1) on a fcc structure to the opal and the inverse opal infiltrated with various liquid crystals. We demonstrate theoretically that when an optically birefringent nematic liquid crystal is infiltrated into the void regions of an opal and an inverse opal, degeneracies seen in the photonic band scheme of these opals disappear, resulting in the appearance of several new peaks in the reflection spectrum. Tunability of the reflection peak upon application of electric field is discussed as functions of its anisotropy of the liquid crystals.

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