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Optical properties and photonic density of states in one-dimensional and three-dimensional liquid-crystalline photonic crystals

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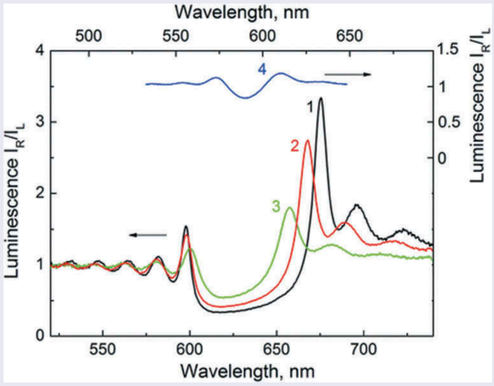
ABSTRACT
Complex optical investigations were performed in one-dimensional (cholesteric) and three-dimensional (Blue Phase II) liquid-crystalline photonic crystals. Spectra of optical transmission and luminescence in the range of the photonic stop band contain information about the local anisotropy, characteristics of the photonic stop band, photonic density of states. We determine the photonic density of states in one-dimensional and three-dimensional liquid-crystalline photonic crystals employing measurements of polarised luminescence. The width of the photonic band gap, density of states and the optical characteristics related to the density of states essentially change with temperature in one-dimensional cholesteric photonic crystal. Drastic transformation of the density of states was found at the transition from one-dimensional to three-dimensional (Blue Phase II) photonic crystal. The results of our investigations demonstrate the possibility to employ the applied method for various types of photonic structures.



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Cholesteric; blue phases; photonic density of states; luminescence; orientational order parameter

1. Introduction

Liquid crystals form several chiral one-dimensional and three-dimensional photonic-crystalline structures [1–4]. Cholesteric is the simplest structure in which the orientation of the long molecular axes rotates continuously in one direction and so a helicoidal structure is formed. Periodic spatial variation of the orientation of the ordinary n_o and extraordinary n_e refractive indices leads to formation of photonic band. Propagation of light and optical properties of the cholesteric photonic crystal depend in a prominent manner on the polarisation of light. When light propagates along the helicoidal axis the photonic band exists only for light of one circular polarisation. Light with the same handedness as the cholesteric helix undergoes selective reflection while the light wave with the opposite handedness propagates without reflection. The middle of the photonic band is located at $\lambda_0 = pn$ and the band has a width $\Delta\lambda = \lambda_0\delta$, where p is the helical pitch, $\delta = \left(n_e^2 - n_o^2\right)/\left(n_e^2 + n_o^2\right)$, $n^2 = \left(n_e^2 + n_o^2\right)/2$ [1]. In three-dimensional photonic crystals, so-called crystalline liquids [4] or Blue Phases (BPs) periodic variation of molecular orientation occurs in three directions in space. BPs are formed in a narrow temperature range between the cholesteric and isotropic phases. Substantial interest in BPs is related to their nontrivial optical properties and prospective applications [5–8].

One of the primary characteristics of photonic crystals (and in general media with spatially varying dielectric permittivity) is the photonic density of states (DOS) or the number of states per unit frequency, $\rho = dk/d\omega$. Many optical

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properties of photonic crystals are determined by DOS and development of methods for its determination is of substantial importance. Different studies were aimed at obtaining information about DOS in liquid-crystalline photonic crystals. DOS of cholesteric has been determined in the spectral range outside the photonic band from measurements of the rotation of the plane of polarisation of light [9]. Luminescence also contains rich information about DOS. Spectral properties of luminescence of guest molecules in cholesteric have been intensively investigated experimentally and theoretically in the last decades [10–18]. However, only recently DOS was determined outside and within the photonic band from luminescence measurements in cholesteric at room (ambient) temperature [19]. In spite of a long history of studies and considerable interest to Blue Phases, to the best of our knowledge, DOS in Blue Phases has not been measured. In this work, we determine the temperature dependence of DOS in cholesteric and DOS in BPII (simple cubic symmetry [4]) from measurements of polarised luminescence. Determination of DOS includes two stages. The first stage is the measurement of transmission spectra in the range of the photonic band and determination of a number of parameters which are necessary to obtain DOS from the luminescence. The second stage is the measurement of luminescence spectra and determination of DOS. We found an essential change of DOS with temperature in one-dimensional cholesteric and at the transition to the three-dimensional Blue Phase, and explain the difference of DOS on the basis of their structure.

2. Experimental methods

Measurements were performed on mixtures of nematic liquid crystal 7OCB (4-cyano-4′-n-heptyloxybiphenyl) from TCI Europe and chiral CB15 (4-cyano-4′-(2-methylbuthyl)biphenyl) from Synthon Chemicals doped by organic dyes. The mixtures form right-handed helical structures. As luminescent dyes, we used 4-diethylamino-4′-nitrostilbene (DEANS) and DCM (Synthon Chemicals) with concentration ~0.1–0.2%wt. The dipole moment of transition in the dyes is parallel to the molecular long axes. Composition of the mixture of nematic and cholesteric liquid crystals was chosen such that the selective reflection bands were in the range of the luminescence spectrum of the dye. Since the spectral position of selective reflection bands in cholesteric and BPII differ, in the measurements we used two types of samples with concentration of nematic 61%wt 7OCB (Sample 1) and 51.5%wt 7OCB (Sample 2). Sample 1 was employed to measure the temperature dependence of DOS in the cholesteric photonic crystal (the cholesteric – BP transition temperature was about 39.6°C). The spectral position of the photonic band in cholesteric in the employed mixture is practically independent of temperature. This enabled directly comparing DOS at different temperatures using the same sample. Sample 2 was used for measurements in BPII (the temperature interval of existence of BPII was about 0.1°C near the temperature 30.8°C).

The liquid crystals were capillary filled into 5-μm optical cells with planar surface alignment purchased from Instec Inc. Planar texture was formed in cholesteric with the helical axis perpendicular to the plane of the cell. In the BPII crystals, the [100] direction was oriented along the cell normal. Scheme of the experimental setup is shown in Figure 1. Samples were placed into an LTS120 hot stage (Linkam). During measurements after the change of temperature, the samples were kept at a constant temperature for a sufficient time to ensure achievement of the equilibrium state when spectra do not change. Luminescence was excited by a KLM-473–20 laser (FTI-Optronic) with emission wavelength 473 nm. The wavelength of the laser line was in the range of the absorption band of the dyes. Excitation was performed by light of left (nondiffracting) circular polarisation. The intensity of the laser light was kept low to avoid heating of the samples. Quality of the structure was controlled by optical microscope observations. Transmission and luminescence spectra were measured using an Altami LUM microscope equipped with a fibre optic spectrometer AvaSpec-2048 (Avantes). Spectra were measured in the direction along the normal of the cell, that is, along the cholesteric helix and the [100] direction in BPII.

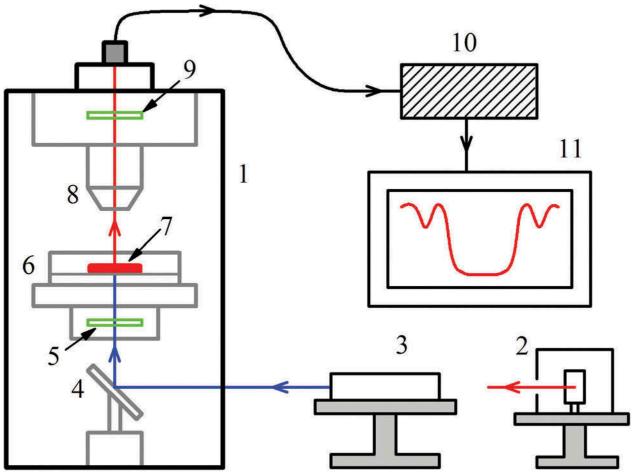

Figure 1. (Colour online) Schematic representation of the optical setup. 1 – optical microscope, 2 – lamp for measurements of transmission spectra, 3 – laser (λ = 473 nm) for excitation of luminescence, 4 – mirror, 5 – polariser, 6 – hot stage, 7 – sample, 8 – objective, 9 – analyser, 10 – spectrometer, 11 – personal computer.

Measurements were made from regions of samples with lateral size ~200 μm.

3. Results and discussion

Solid curve (1) in Figure 2 shows the transmission spectrum of cholesteric measured in light of right circular polarisation. The spectrum was normalised by the spectrum of the light source. The spectrum possesses the intensive band related to the Bragg reflection and subsidiary minima in the vicinity of band whose amplitude decreases away from the band. Presence of subsidiary minima in the spectrum indicates high quality of the sample [20–22].

The complex transmission coefficient t of a cholesteric slab of thickness L for normal incidence can be found from the analytical solution of Maxwell equations [20,21,23]:

$$t = \frac{\tau\beta_3 e^{i\tau L/2}}{\tau\beta_3 \cos\beta_3 L + i\left(\beta_3^2 + (\tau^2/4) - \kappa^2\right)\sin\beta_3 L} \tag{1}$$

In this equation $\beta_3 = \kappa\sqrt{1 + (\tau/2\kappa)^2 - \left[(\tau/\kappa)^2 + \delta^2\right]^{1/2}}$, $\kappa = 2\pi n/\lambda$, $\tau = 4\pi/p$. The transmission intensity is $T = |t|^2$. The boundaries of the photonic band are given by the condition $\beta_3 = 0$ and are located at $\lambda_{1,2} = \lambda_0\sqrt{1 \pm \delta}$. To calculate the spectrum as the first approximation we used the value of the refractive index n for 7OCB taken from [24]. For a precise description of the subsidiary minima, one has to take into account the dispersion of the refractive index. Its spectral dependence was taken from literature data for a similar compound [25]. The dashed curve 1 shows the transmission spectrum of cholesteric calculated according to (1) with parameters $\delta = 0.1029$, $n = 1.592$, $p = 400.3$ nm. As one can see, the calculation well agrees with the experimental spectrum. So the used values of δ, n and p are suitable for the description of optical properties of our photonic crystal. The vertical dash-dotted lines are the boundaries of the photonic band gap calculated from $\lambda_{1,2} = \lambda_0\sqrt{1 \pm \delta}$. The spectral width of the band in transmission spectrum is larger than the width of the photonic band which is due to the finite thickness of the samples. Figure 3 shows the transmission spectra of cholesteric at two temperatures: 25°C (curve 1) and 38°C (curve 2). On heating the width and the intensity of the selective reflection band decrease, which is related to the decrease of relative optical anisotropy δ. Squares in Figure 4(a) show the temperature dependence of the width of the photonic band calculated from the position of left and right boundaries of the photonic band gap. The width essentially decreases with temperature.

Presence of the photonic band strongly modifies the spectral intensity of spontaneous emission for the light of diffracting circular polarisation. Figure 5 shows relative luminescence intensity for right and left circularly polarised light I_R/I_L in cholesteric at different temperatures. Emission intensity strongly decreases inside the photonic band and has pronounced maxima on either side of the band gap. Subsidiary oscillations are present in luminescence

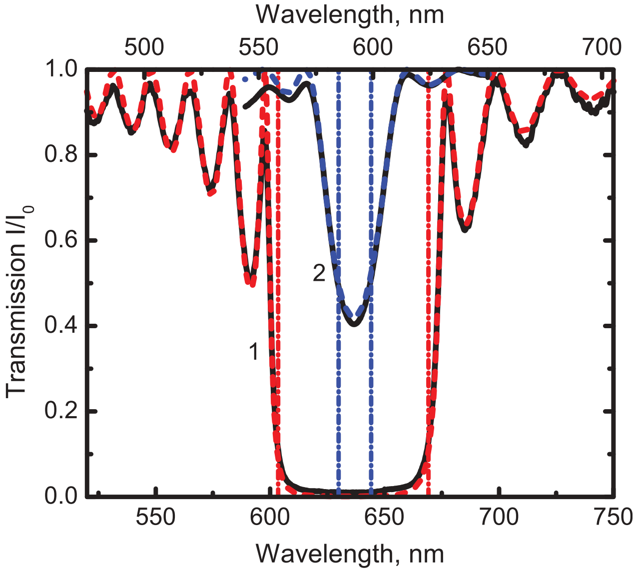

Figure 2. (Colour online) Solid curves show transmission spectra of cholesteric (1) and BPII (2). Dashed curves are the theoretical transmission spectra calculated according to Equation (1) with parameters δ= 0.1029, n= 1.592, p= 400.3 nm in cholesteric and δ= 0.0239, p= 377.6 nm, n= 1.567 in BPII. T = 25°C (cholesteric, Sample 1); T = 30.8°C (BPII, Sample 2). The vertical dash-dotted lines show the photonic band gap edges. Bottom axis in the Figure refers to the cholesteric, top axis to BPII.

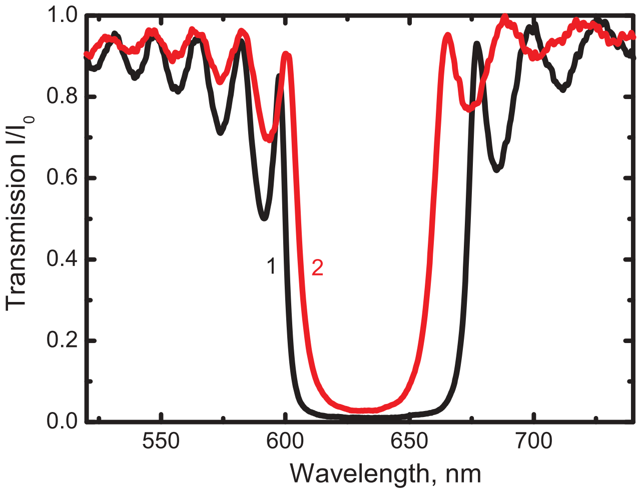

Figure 3. (Colour online) Transmission spectra of cholesteric at T = 25°C (1) and T = 38°C (2). Width and intensity of the photonic band decrease on heating (Sample 1).

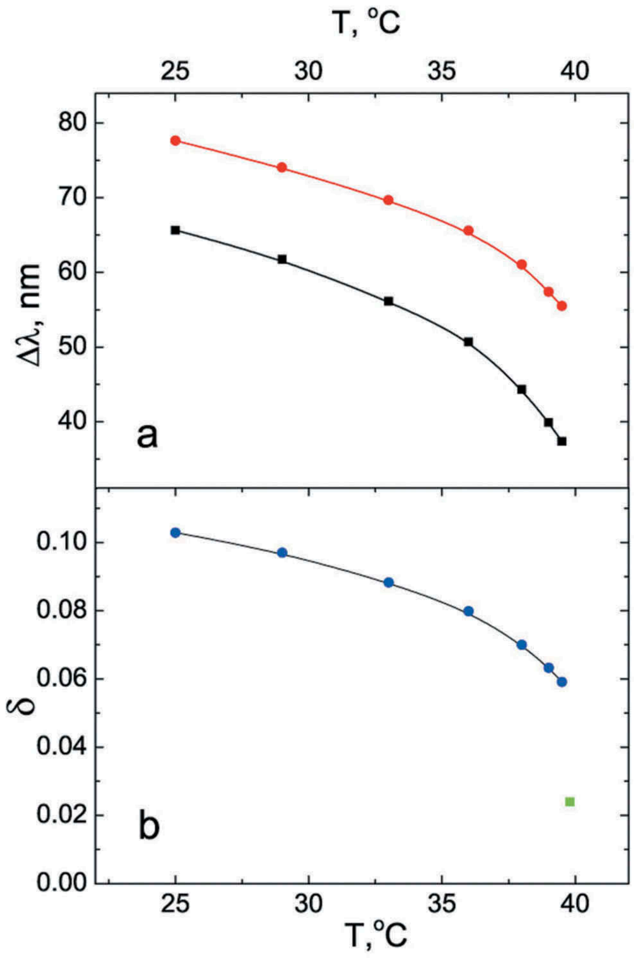

Figure 4. (Colour online) (a) Temperature dependence of the width of the photonic band gap (squares) and the distance between peaks in luminescence spectra (circles). The curves are guides to the eye. (b) Temperature dependence of the relative optical anisotropy δ in cholesteric (circles, Sample I) and in BPII (square, Sample 2). For the difference in the values of δ in cholesteric and BPII to be visible, the square (Sample 2) is shifted from 30.8°C to the melting temperature of cholesteric in Sample 1.

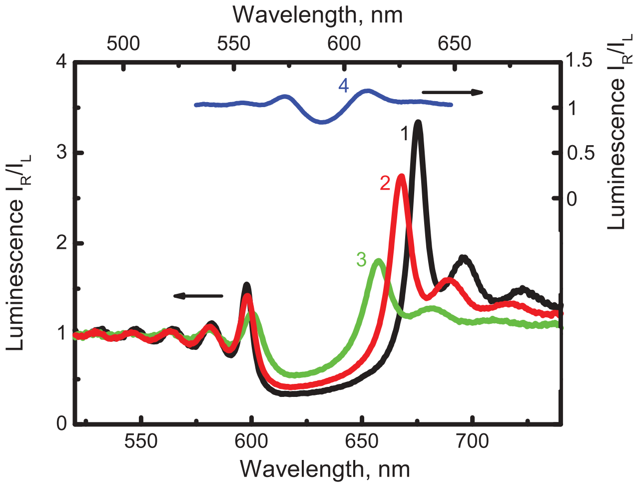

Figure 5. (Colour online) Relative intensity of luminescence of right and left circular polarisation I_R/I_L in the cholesteric phase (bottom and left axes) at T = 25°C (1), T = 33°C (2), T = 39°C (3), Sample 1 and in BPII (top and right axes) at 30.8°C (4), Sample 2.

as well as in the transmission spectra. The intensity of the peak at the long-wavelength edge of the photonic band is much larger than of the peak at the short-wavelength edge. The reason is related to the polarisation properties of light propagating in the cholesteric structure [11]. At the long-wavelength edge of the photonic band, the direction of polarisation of the eigenmode of light rotates nearly parallel to the local liquid crystal director, whereas at the short-wavelength edge it is nearly perpendicular to the local director [11]. So, the dipole moments of transition of the dye molecules in luminescence are oriented nearly along the director in the vicinity of the long-wavelength boundary of the gap and perpendicular to the director in the vicinity of the short-wavelength boundary. Emission probability which depends on the component of the transition dipole moment along the polarisation of light is, therefore, larger at the long-wavelength edge of the band. On heating, the intensity of the maxima of luminescence decreases and the dip in the spectra in the range of the photonic band becomes less pronounced (Figure 5). Decrease of the distance between main maxima in fluorescence spectra (circles in Figure 4(a)) correlates with the temperature dependence of the bandwidth and the relative optical anisotropy (Figure 4(b)).

To determine DOS we employ the approach based on the analysis of the polarisation properties of the eigenmodes of light in chiral liquid crystals [11]. DOS for the nondiffracting polarisation (i.e. for the structure without the photonic zone) in good approximation can be written as $\rho_0 = n/c$. For the light of diffracting polarisation the situation is more complex [11]. DOS of the photonic crystal can be determined from relative luminescence intensity I_R/I_L According to Fermi's Golden rule [11,19] the spontaneous emission is proportional to the density of photonic states and the probability of spontaneous light emission. The relative intensity of luminescence for diffracting (I_R) and non-diffracting (I_L) polarisation can be written as

$$\frac{I_R}{I_L}=\frac{\rho}{\rho_0}\frac{\left\langle|d_1|^2\right\rangle}{\left\langle|d_2|^2\right\rangle} \tag{2}$$

where $\left\langle|d_1|^2\right\rangle$ and $\left\langle|d_2|^2\right\rangle$ are the averaged-squared values of the transition dipole moment component along the polarisation of light for the diffracting and nondiffracting polarisation [11]. $\left\langle|d_1|^2\right\rangle$ and $\left\langle|d_2|^2\right\rangle$

depend on the orientational order parameter S for the dipole moment of transition in luminescence and on the ellipticities $f_{1,2}$ of the polarisation eigenstates [11]

$$\left\langle |d_{1,2}|^2 \right\rangle = \frac{2}{3}\frac{f_{1,2}^2 - 1/2}{f_{1,2}^2 + 1} S + 1/3 \tag{3}$$

where

$$f_{1,2} = \frac{1 - \delta - (\beta/\kappa)^2 - (\tau/2\kappa)^2}{\beta\tau/\kappa^2}$$

In the above equations for light wave of the diffracting polarisation $\beta = \beta_3$, for light of the nondiffracting polarisation $\beta = \kappa\sqrt{1 + (\tau/2\kappa)^2 + \left[(\tau/\kappa)^2 + \delta^2\right]^{1/2}}$. Inside the photonic band, f_1 is a complex value and $\mathrm{Im}(f_1)$ has to be substituted in (3) instead of f_1. In the range of the photonic band $\left\langle |d_1|^2 \right\rangle$ has pronounced peculiarities, adopting maximum and minimum values at the long-wavelength and at the short-wavelength edge of the band, while $\left\langle |d_2|^2 \right\rangle$ weakly depends on the wavelength.

Luminescence spectra of the cholesteric photonic crystal (Figure 5) and Equations (2 & 3) were used to determine the DOS. In the calculation, we use material parameters found from transmission spectra and the order parameter S obtained from independent measurements. Determination of the order parameter in cholesteric is a nontrivial task [26]. To estimate S we employ measurements of polarised luminescence in nematic [27]. For these measurements, nematic 7OCB doped by the fluorescent dye was used. Luminescence was excited by left circularly polarised light. The intensity of luminescence with polarisation parallel $I_{||}$ and perpendicular $I_{\perp}$ to the nematic director was measured. The order parameter S was obtained from $S = \frac{r' - 1}{r' + 2}$ [11,27] using the ratio $r' = \frac{n_o I_{||}}{n_e I_{\perp}}$ for luminescence in the spectral range of the photonic band gap of cholesteric. The employed method is preferable over measurements of absorption frequently used to determine S in nematic because in our experiment the order parameter is related to the ordering of transition dipole moments in luminescence. The order parameter enters the expressions used to determine DOS (2,3). The obtained spectra of DOS in the cholesteric photonic crystal at different temperatures are shown in Figure 6. Far from the photonic band the relative value of DOS ρ/ρ_0 is close to unity. It is known that in an infinite structure the DOS diverges at the gap edges. For a finite sample (Figure 6), two main maxima of DOS are located near the photonic band gap. These resonances in DOS on cooling become sharper and stronger. Their spectral position correlates with the change of the width and the position of the edges of the photonic band. The distance between maxima and the gap magnitude decrease with temperature. On heating, the dip of DOS in the range of the photonic band also decreases. The magnitude of maxima of DOS near the two edges of the photonic band is comparable in contrast to the relative intensity of peaks in luminescence (Figure 5). The main peculiarities of DOS are consistent with the theoretical DOS of perfect cholesteric photonic crystals [9,28–30].

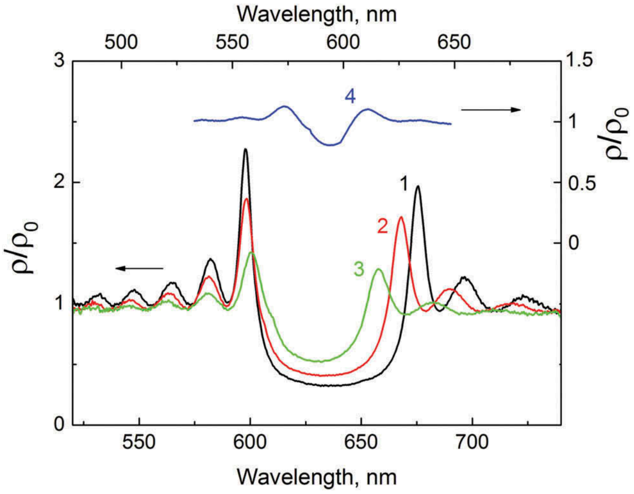

Figure 6. (Colour online) Photonic density of states in cholesteric at T = 25°C (1), T = 33°C (2), T = 39°C (3), Sample 1 (bottom and left axes) and in BPII at 30.8°C (4), Sample 2 (top and right axes) determined from luminescence spectra using Equations (2 & 3).

Now we will discuss the peculiarities of the three-dimensional periodic BPII. Solid curve 2 in Figure 2 shows the transmission spectrum of BPII measured in light of right circular polarisation. As in the one-dimensional photonic crystal selective reflection and photonic band exist only for one (namely, right) circular polarisation. The relative spectral position of [100] reflection in BPII with respect to selective reflection in cholesteric in the same sample $\lambda_{\mathrm{BPII}}/\lambda_{\mathrm{Ch}} \approx 1.16$ agrees with results of earlier measurements [31]. The dashed curve is the theoretical transmission spectrum calculated according to Equation (1) with the magnitude of the relative optical anisotropy δ=0.0239. This is approximately 2.5 times smaller than in cholesteric near the transition to the Blue Phase. The width of the photonic band is also essentially less than in the cholesteric phase. A small value of the relative optical anisotropy δ is in qualitative agreement with theory and the experimental data determined from rotation of the plane of polarisation of light and integral intensity of reflections [32–34]. Solid curve 4 in Figure 5 shows relative luminescence intensity in right and left circularly polarised light I_R/I_L in BPII. At present, the values of S connected with different Fourier harmonics of Blue Phases are not known. There are no available methods for the direct measurements of their values. We remind that BPII has cubic symmetry [4] with local anisotropy. We evaluated the magnitude of the local order parameter in BPII using the value of δ in cholesteric, in BPII and the local order parameter S in cholesteric. The orientational order parameter S is approximately proportional to the optical anisotropy δ [35,36], which allows to evaluate the order parameter related with [100] reflection of BPII (about 0.069) and to obtain the density of photonic states in BPII (Figure 6) from luminescence using Equation (3). At the transition to the Blue Phase, the density of states changes radically. The dip in the region of the photonic band and the subsidiary maxima become small (Figure 6). These transformations are due to the three-dimensional structure of Blue Phases and a small value of the relative optical anisotropy δ. In BPII several Fourier harmonics of the dielectric anisotropy tensor exist in contrast to cholesteric where the modulation of the dielectric anisotropy is related only with the cholesteric helical axis. Narrow temperature window of the Blue Phase (~0.1°C) did not allow us to determine the temperature dependence of DOS in BPII. We regard this as a subject of future investigations.

4. Summary

Optical transmission and luminescence spectra were measured in the range of the photonic band of one-dimensional and three-dimensional liquid-crystalline photonic crystals. We determined the temperature dependence of the density of states in a cholesteric photonic crystal, density of states in Blue Phase II from measurements of polarised luminescence. Maxima of the density of states is located near the boundaries of the photonic band. Substantial transformation of the density of states in cholesteric with temperature is observed which is connected with the decrease of local optical anisotropy and order parameter. The transition to the three-dimensional BPII photonic crystal leads to a cardinal change of the density of states. Our results demonstrate the applicability of the employed method for investigation of different photonic structures.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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