Density of photonic states in cholesteric liquid crystals

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Density of photonic states $\rho(\omega)$, group v_g , and phase v_{ph} velocity of light, and the dispersion relation between wave vector k, and frequency $\omega(k)$ were determined in a cholesteric photonic crystal. A highly sensitive method (measurement of rotation of the plane of polarization of light) was used to determine $\rho(\omega)$ in samples of different quality. In high-quality samples a drastic increase in $\rho(\omega)$ near the boundaries of the stop band and oscillations related to Pendellösung beatings are observed. In low-quality samples photonic properties are strongly modified. The maximal value of $\rho(\omega)$ is substantially smaller, and density of photonic states increases near the selective reflection band without oscillations in $\rho(\omega)$. Peculiarities of $\rho(\omega)$, v_g , and $\omega(k)$ are discussed. Comparison of the experimental results with theory was performed.

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I. INTRODUCTION

The remarkable properties of photonic crystals leading to transformation of spectral and polarization properties of light attract attention both from the point of view of fundamental physics as well as practical applications [1-4]. A special place among photonic crystals is occupied by liquid crystals forming a variety of photonic structures. The simplest of them is the one-dimensional cholesteric liquid crystal (CLC). In cholesteric liquid crystals the preferred direction of long molecular axes (director of the liquid crystal) rotates forming a helical structure with period p. As a result of helical periodicity the solutions of Maxwell wave equations for CLC are Bloch waves with circular polarizations. Eigenwaves of one of the circular polarizations weakly interact with the cholesteric structure, while the interaction with the other polarization is strong [5,6]. In thick samples this interaction leads to formation of a forbidden photonic band in the frequency range $\omega_0/(1-\delta)^{1/2} > \omega > \omega_0/(1+\delta)^{1/2}$, where $\omega_0 = 2\pi c/\varepsilon_0^{1/2} p$, $\delta = (\varepsilon_{\parallel} - \varepsilon_{\perp})/(\varepsilon_{\parallel} + \varepsilon_{\perp})$ is the relative dielectric anisotropy, $\varepsilon_0 = (\varepsilon_{||} + \varepsilon_{\perp})/2$, and $\varepsilon_{||}$ and ε_{\perp} are the dielectric constants parallel and perpendicular to the director. In spite of numerous studies of liquid-crystalline photonic crystals, a number of characteristics of photonic structures remain open for investigation.

One of the most important characteristics of photonic crystals is the density of photonic states (DOS), i.e., the number of wave vectors k per unit frequency $\rho(\omega) = dk/d\omega$. Many optical properties are determined by DOS. In particular, according to Fermi's "golden rule" luminescence intensity is proportional to $\rho(\omega)$. For a large value of excitation an intensive stimulated emission appears in maxima of $\rho(\omega)$ at the boundaries of the band and lasing is observed at the corresponding frequencies [7-10]. Before the present work a large number of theoretical and experimental studies were performed aimed at the determination of spectral characteristics related to the presence of photonic bands. In particular, in luminescence spectra of cholesteric liquid crystals' spectral peculiarities were observed related to the photonic band [11,12]. In colloidal crystals by measurement of delay of light wave and interference the dispersion of light related to the photonic zone was measured [13–18]. Theory predicts nontrivial behavior of $\rho(\omega)$ near the boundaries of the

photonic band, and a nonmonotonic trend of DOS related to Pendellösung oscillations [10,19–22]. It is worth mentioning that Pendellösung oscillations are observed only in perfect samples. Small nonuniformities of the structure and thickness lead to disappearance of peculiarities related to Pendellösung oscillations, modification of the selective reflection bands, and to the smooth trend of reflection and rotation of the plane of polarization of light. A sufficient transformation should therefore also be expected in DOS and different related characteristics of the photonic crystal.

Much of the current attention in photonic crystals is given to the elaboration of the methods of calculation of DOS, group velocity of light v_g , dispersion $\omega(k)$, and their experimental determination. In the present work a method is proposed for determination of the DOS, v_{g} , and $\omega(k)$ in cholesteric photonic crystals from measurements of rotation of the plane of polarization of light. It was possible due to the unique structure of the cholesteric photonic crystal in which only one polarization strongly interacts with the periodic structure. DOS was determined in CLCs of two types: in perfect samples and in samples with distortion of chiral ordering. In perfect samples the photonic density of states increases essentially near the selective reflection band. Outside the band oscillations in $\rho(\omega)$ are observed, whose magnitude decreases upon going away from the band. We discuss the influence of the disorder on the measured characteristics. Spectral dependence of $\rho(\omega)$ transforms essentially in imperfect CLC. Absence of Pendellösung oscillations in selective reflection and transmission spectra leads to a smooth increase in $\rho(\omega)$ on approaching the photonic band. A comparison between $\rho(\omega)$, v_{α} , dispersion of $\omega(k)$ extracted from experimental data, and theoretical calculations shows good agreement.

II. DIFFERENT WAYS TO CALCULATE THE DOS

The conventional method of the calculation of $\rho(\omega)$ is the use of the complex transmission coefficient amplitude, $t(\omega) = X + iY = |t|e^{i\psi}$. The modulus of $|t(\omega)| = (X^2 + Y^2)^{1/2}$ determines the intensity of light transmitted through the photonic crystal $T = |t|^2$. The phase of the transmission function $\psi(\omega) = \arctan[Y(\omega)/X(\omega)]$ gives the phase accumulated when the pulse peak propagates through the photonic crystal. As ψ equals kL, where L is the thickness of the sample, $kL = \arctan(Y/X)$. Taking the first derivative with respect to ω ,

$$\frac{d}{d\omega}(kL) = \frac{d}{d\omega}\arctan(Y/X),\tag{1}$$

 $\rho(\omega)$ is written in the form [10,11,19,20,22]

$$\rho = \frac{1}{L} \frac{Y'X - X'Y}{X^2 + Y^2}.$$
 (2)

Equation (2) is valid for any type of photonic crystal and was used for calculation of $\rho(\omega)$ in different materials [10,11,19,20,22]. If the real and imaginary parts of $t(\omega)$ are known, $\rho(\omega)$ can be easily calculated. For cholesteric liquid crystals $t(\omega)$ was obtained in an analytical form [6]:

$$t(\omega) = \frac{\tau \beta_3 e^{i\tau L/2}}{\tau \beta_3 \cos \beta_3 L + i \left[\beta_3^2 + (\tau^2/4) - \kappa^2\right] \sin \beta_3 L}.$$
 (3)

In the above equation $\beta_3 = \kappa \sqrt{1 + (\tau/2\kappa)^2 - [(\tau/\kappa)^2 + \delta^2]^{1/2}}, \quad \kappa = \omega n_0/c, \quad n_0 = \sqrt{\varepsilon_0},$

and $\tau = 4\pi/p$. For a perfect sample with an integer number *m* of periods,

 $\tau L/2 = 2\pi m$ and $e^{i\tau L/2} = 1$. From (3) two equations for the real and imaginary parts of $t(\omega)$ read

$$X = \frac{\tau^2 \beta_3^2 \cos \beta_3 L}{\tau^2 \beta_3^2 + \kappa^4 \delta^2 \sin^2 \beta_3 L},\tag{4}$$

$$Y = -\frac{\tau \beta_3 (\beta_3^2 + \tau^2/4 - \kappa^2) \sin \beta_3 L}{\tau^2 \beta_3^2 + \kappa^4 \delta^2 \sin^2 \beta_3 L}.$$
 (5)

In such a way the DOS for CLC was calculated from Eqs. (2), (4), and (5) [10,11,22]. However, this way is not suitable for the experimental determination of DOS. The modulus of the transmission coefficient |t| can be extracted from measurements of transmitted intensity $T = |t|^2$. However, if the phase of the transmission function $\psi(\omega)$ remains unknown we cannot obtain information on important quantities such as $\rho(\omega)$, v_g , and the dispersion of photonic states.

At present, the widely used measurements for the characterization of quantities related to the imaginary part of $t(\omega)$ are the interference of the light reflected from two surfaces of the sample and time-resolved interferometry [16–18,23–25]. Here we apply a highly sensitive technique, rotation of the plane polarization of light, for determination of phase delay and extraction of the characteristics of the band structure in cholesteric photonic crystal. Actually, the problem is resolved as full information about $X(\omega)$ and $Y(\omega)$ is not necessary to determine $\rho(\omega)$, but only the arctan of their ratio (1). Notice that $X(\omega)$ and $Y(\omega)$ have the same denominator [(4) and (5)], so the ratio Y/X is simplified:

$$Y/X = -\frac{\left(\beta_3^2 + \tau^2/4 - \kappa^2\right)}{\tau\beta_3} \tan\beta_3 L.$$
 (6)

Remarkably, precisely the same ratio is contained in the expression for rotation of the plane of polarization of light [26],

$$\varphi = \frac{1}{2} \left\{ (\tau - \beta_1)L - \arctan\left[\frac{\left(\beta_3^2 + \tau^2/4 - \kappa^2\right)}{\tau\beta_3} \tan\beta_3 L\right] \right\},\tag{7}$$

where $\beta_1 = \kappa \sqrt{1 + (\tau/2\kappa)^2 + [(\tau/\kappa)^2 + \delta^2]^{1/2}}$. The derivative of the first term in (7) with respect to ω equals with good accuracy $-n_0 L/2c$. So DOS can be determined from $d\varphi(\omega)/d\omega$:

$$\rho(\omega) = \frac{n_0}{c} + \frac{2}{L} \frac{d\varphi}{d\omega}.$$
(8)

This result allows to obtain $\rho(\omega)$ from the measurement of rotation of the plane of polarization of light. From $\varphi(\omega)$ the real part of the refractive index related to the existence of the pseudogap can be extracted. $T(\omega)$ and $\psi(\omega)$ give the full information about $t(\omega)$ and the complex refraction index [17],

$$n_{\rm eff} = \operatorname{Re}(n_{\rm eff}) + i\operatorname{Im}(n_{\rm eff}) = \frac{c}{\omega L} \left(\psi - i\frac{1}{2}\ln T\right).$$
(9)

Notice that Eq. (8) outside the photonic stop band can be simply obtained in the assumption that the index of refraction and the wave vector are real quantities. Refractive index nfor circular polarization strongly interacting with the periodic structure of CLC can be represented as a part independent of the photonic structure n_0 and additive Δn related to the presence of the periodic structure $n = n_0 + \Delta n$. Change of the refractive index Δn can be expressed via the phase difference of the light amplitude $\Delta \psi$ connected with the photonic band [14],

$$\Delta n = \frac{\Delta \psi \lambda}{2\pi L}.$$
 (10)

Delay in $\Delta \psi$ leads to rotation of the plane of polarization of light φ which is related with $\Delta \psi$ by a simple equation, $\varphi = \Delta \psi/2$. From (10) it follows that

$$\Delta n = \frac{\varphi \lambda}{\pi L}.$$
(11)

The wave vector $k = 2\pi n/\lambda$ can be written as

$$k = \frac{\omega n_0}{c} + \frac{2\varphi}{L}.$$
 (12)

The dispersion relation is split in two parts. Taking the derivative we get Eq. (8) for DOS.

III. DETERMINATION OF DOS, GROUP VELOCITY, DISPERSION IN CLC

Measurements were performed on CLC formed by a mixture of esters of alkyl derivatives of benzoic acid with chiral dopant VIH-16 (VIC, Vilnius University, Lithuania). The mixture has the cholesteric phase with left-hand helix at room temperature. Measurements of transmission and rotation spectra were performed on samples of two types. Samples of the first type were prepared in an optical cell whose surfaces were coated with a polyamide film. The director of the liquid crystal orients near the surface along the direction of rubbing which is parallel on two surfaces of the cell. In samples of the second type of cell surfaces were not treated by aligning coating. The preferred orientation of the director parallel to the plane of the cell was achieved by shearing the glass plates of the cell with respect to each other. Measurements of spectra were performed employing a charge-coupled device (CCD) spectrometer. Rotation of the plane of light polarization was



FIG. 1. Transmission spectrum in nonpolarized light (dashed curve) and rotation of the plane of polarization of light (points) for a perfect sample with aligning coating on the cell surfaces. Oscillations in transmission and rotation of the plane of polarization of light are related to Pendellösung beatings and demonstrate high quality of the sample. T = 23.5 °C, sample thickness $L = 5.8 \,\mu$ m.

obtained from spectra recorded with different orientation of the analyzer. Very close to the stop band the polarization of transmitted light strongly deviated from linear. This did not allow to measure rotation of the plane of polarization of light very close to, and inside, the stop band.

Figure 1 shows the transmission spectrum of CLC (the dashed curve) and rotation of the plane of polarization of light (points) for a sample in the cell with aligning coating. The plateau in the minimum of the transmission spectrum is close to 0.5 since one circular polarization is nearly totally reflected, and the other one propagates freely. Apart from the reflection band, subsidiary maxima and minima (Pendellösung oscillations) are visible. Pendellösung oscillations manifest themselves also in the peculiarities of spectral dependence of rotation of the plane of polarization of light. They were predicted by theory [6] and observed in high-quality samples [26]. Further, the CLC whose spectrum is given in Fig. 1 will be called a perfect sample. Transmission and rotation spectra for a sample without aligning coating (Fig. 2) substantially differ from the case of the perfect sample. Band half width is larger, and the spectrum changes smoothly in the long-wavelength and short-wavelength regions. The CLC sample whose spectrum is given in Fig. 2 we shall further call an imperfect sample. Magnitude of the rotation of the plane of polarization of light in imperfect samples increases with approaching the band but peculiarities related to Pendellösung oscillations are not observed. It is worth noting that the spectrum of selective reflection of CLC without aligning coating can differ substantially depending on the method of preparing the sample. However, its basic properties remain the same: reflection in a broader spectral range and smooth trend of rotation of the plane of polarization of light. Such experimental spectral dependence of rotation of the plane of polarization of light is usually given in the literature [2,6].

The measurements performed enable us to extract DOS from the spectra of rotation of the plane of polarization of light using Eq. (8). The solid curve in Fig. 3(a) shows



FIG. 2. Transmission spectrum in nonpolarized light (dashed curve) and rotation of the plane of polarization of light (points) for an imperfect sample without aligning coating on the cell surfaces. Pendellösung oscillations in the spectra are absent. T = 26 °C, sample thickness $L = 7.75 \,\mu$ m.



FIG. 3. Normalized DOS of perfect (a) and imperfect (b) samples of cholesteric photonic crystal (solid curves) obtained from rotation of the plane of polarization of light in Figs. 1 and 2. The dashed line in (a) is the theoretical spectrum of DOS calculated using Eqs. (2), (4), and (5) with material parameters ($n_0^2 = 2.491$, p = 362 nm, $\delta =$ 0.0638) corresponding to transmission spectra of Fig. 1. Oscillations in $\rho(\omega)$ [Fig. 3(a)] are related to the finite size of the photonic crystal. Experimentally obtained $\rho(\omega)$ is in agreement with calculated DOS. Two vertical lines show the edges of the forbidden photonic band.

the dependence $\rho(\omega)$ for the perfect sample normalized by $\rho_0 = n_0/c$. DOS is maximal near the boundaries of the band gap. Outside the stop band DOS is a strongly oscillating function which is related to interference. The amplitude of DOS oscillations increases near the stop band. Experimentally determined DOS can be compared with results of theoretical calculations. We used the analytical solution [Eqs. (4) and (5)]following from the Maxwell equations. The dashed curve in Fig. 3(a) shows $\rho(\omega)$ calculated using the material parameters obtained from comparison of the transmission spectra with theoretical dependence $I/I_0 = (T+1)/2$. Here T is the transmission coefficient for light with diffracting circular polarization, $T(\omega) = X^2(\omega) + Y^2(\omega)$. From Fig. 3(a) it can be seen that theoretical equations well describe the peculiarities of the DOS. Maxima in DOS correspond to regions of maximal transparency of the photonic crystal. For samples of infinite thickness $\rho(\omega) = 0$ inside the band, and diverges at the boundaries of the photonic band [11] (Van Hove singularities). For a finite photonic crystal the maxima are shifted from the band edges; DOS is nonsymmetrical with respect to the midgap frequency ω_0 . The short-wavelength maximum of $\rho(\omega)$ is larger than that at the long-wavelength edge. Theoretical $\rho(\omega)$ also reproduces the above described behavior. In the photonic band gap the wave vector is a complex value and the field decays exponentially. Inside the pseudogap the noise in $\varphi(\omega)$ increases and we could not determine DOS. However, calculations show that even in the center of the band the normalized DOS $\rho(\omega)/\rho_0$ in our sample is rather essential (~ 0.3 of the regular value 1).

Normalized DOS of an imperfect sample was determined using $\varphi(\omega)$ from Fig. 2 and is shown in Fig. 3(b). As can be seen, $\rho(\omega)$ changes dramatically with respect to the perfect sample. Oscillations in $\rho(\omega)$ are absent. The maxima of $\rho(\omega)$ are broadened and the distance between them is larger than between the main maxima in the perfect sample. DOS increases smoothly on approaching the pseudogap and then decreases near it [Fig. 3(b)]. The absolute value of $\rho(\omega)$ in the maxima is significantly smaller than in the perfect sample.

We will now consider the group v_g and phase v_{ph} velocity of light. The group velocity can be determined from a simple relation $v_g = d\omega/dk = 1/\rho$. In Fig. 4 the solid curve shows $v_{e}(\omega)$ obtained from the experimental spectral dependence of DOS in the perfect sample. In an infinite sample the group velocity decreases near the band gap and becomes zero at the edge. In a finite sample the main transformation of v_{g} is in the region close to the pseudogap. The group velocity $v_g(\omega)$ reaches minima in a narrow range near the pseudogap and then increases as the edges are approached (Fig. 4). We should note here that in a finite sample the group velocity in the pseudogap can exceed the velocity of light in vacuum, c. For our sample the calculated group velocity in the middle of the pseudogap is superluminal ($v_g \approx 2.05c$). Note that this value for v_{e} does not violate Einstein causality [27,28]. The energy velocity and velocity of the forward front of the light pulse remain subluminal.

In an infinite sample the dispersion curve $\omega(k)$ has a smooth dependence on k and is tangent to the band edge on the boundary of the Brillouin zone. In the finite sample dispersion curves $\omega(k)$ obtained using Eq. (12) are shown in



FIG. 4. Plot of the dimensionless group velocity of light v_g/c (solid curve) for the perfect sample of finite thickness. The extremes of the group velocity appear in the finite sample on either sides of the stop band. Dimensionless phase velocity of light v_{ph}/c for the sample of finite thickness (dashed curve). Two vertical lines show the edges of the stop band. $L = 5.8 \,\mu$ m.

Fig. 5 (points). Contrary to the infinite sample they exhibit pronounced peculiarities related to the finite layered structure. Theoretical dependence $\omega(k)$ calculated using parameters (n_0, p, δ) obtained from the transition spectrum of Fig. 1 appears as a solid line. The theoretical curve reproduces the experimental peculiarities in $\omega(k)$. Essentially smooth variation of v_{ph} as compared to v_g is related to the fact that v_{ph} is determined by the absolute value of the refractive index. Low precision of the measurements of $\varphi(\omega)$ close and within the pseudogap could not allow us to determine $\omega(k)$ in these regions. Since in the finite sample $\omega(k)$ must be a continuous curve, from the experimental data we can evaluate the lower limit of v_g in the middle of the pseudogap $v_g > \Delta \omega / \Delta k$, where $\Delta \omega$ is the difference between frequencies of the measured points on either side of the pseudogap, that are nearest to



FIG. 5. Experimental dispersion of light $\omega(k)$ for the perfect sample of finite thickness (points). $\omega = 2\pi c/\lambda$. Theoretical dispersion (solid lines). Two horizontal lines show the edges of the forbidden photonic band. The dashed line shows the dispersion for the medium without the photonic band. $L = 5.8 \,\mu$ m.

the pseudogap; Δk is the difference in wave vectors of these points. We obtain $v_g > 1.24c$, that is, group velocity v_g in the pseudogap becomes greater than the speed of light in vacuum.

In conclusion, a method based on the measurements of rotation of the plane of polarization of light is derived for determination of $\rho(\omega)$, v_g , and $\omega(k)$ in CLCs. The method is applied for determination of DOS in perfect and imperfect CLCs. It is shown that DOS depends substantially on the quality of the sample. In perfect samples the experimentally determined DOS demonstrates peculiarities predicted by theory: maxima of DOS near the boundaries of the photonic band and oscillations related to Pendellösung beatings. In imperfect

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samples DOS increases monotonically on approaching the band; the spectral dependence of DOS in these samples depends on the method of preparation. We present results on the group v_g and phase v_{ph} velocity of light and dispersion of light $\omega(k)$ that allow a better understanding of the propagation of light in cholesteric photonic crystals.

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