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Application of Cholesteryl Alkanoate Liquid Crystals for Light Polarization Control

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Chiral LC mixtures based on cholesterol esters with strong temperature dependence of the helical pitch were used for realization of the light-controlled adjustment of the selective reflection (SR) spectral position. Subtle variations of composition allowed obtaining mixtures with helical pitch rapidly decreasing with temperature and providing the fine tuning of the SR band with respect to the laser emission wavelength.

Control of light polarization plane in the spectral region of the SR band was investigated; potential applications in different aspects of optical signal processing, all-optical switching and photonics are discussed.

Keywords Chiral liquid crystal; nonlinear optics; photonic band gap; polarization plane control

1. Introduction

Chiral liquid crystals (CLC), which were the first ever observed liquid crystalline (LC) materials, have been experiencing a renewable interest in both science and application-related studies. Effective control of the helical pitch by external fields made CLC's promising for utilization in optical and electro-optic devices such as all-optical shutters and switches, high-speed modulators, or in adaptive laser-optics and lasing techniques [1–3]. If the spatial periodicity of the helical pitch is close to the wavelength of light, a strong distributed optical feedback occurs, giving birth to the unique effect of selective reflection of light (SR) that is characteristic of photonic band gap structures. Being extraordinary strong, the optical activity of CLC reveals also very large dispersion at the edge of the selective reflection band [4], which essentially enhances nonlinear response and may contribute substantially to the efficiency of diffraction or be the only nonlinear optical mechanism [5].

Helical pitch modulation should lead to corresponding changes in the dispersion of rotatory power for the wavelengths close to SR region. The period of CLC helix could be easily tuned by means of different external factors [4,6,7] that allows for

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expectations of many interesting applications. The current work is devoted to the investigation of the systems with temperature dependent position of the SR band from the standpoint of light polarization control.

Generally used in most applications cholesteric LC systems appeared to be, in fact, “induced cholesterics”, i.e., based on a nematic matrix containing a chiral dopant. In such systems, the selective reflection band is rather wide due to large birefringence of the aromatic matrix. Spectral position of the selective reflection band (wavelength of SR maximum, λ_{\max}) changes with temperature comparatively slowly (which allows obtaining temperature stability of different characteristics required in most cases). Strong temperature dependence of λ_{\max} ($\partial\lambda_{\max}/T$) in a specified range is very difficult to obtain with induced cholesterics.

In our work, we used cholesteric CLC systems based on cholesterol esters. In fact, mixtures of cholesterol derivatives with a substituent in 3 β -position (alkanoates, alkyl carbonates, halides) were the first CLC used for practical applications (temperature indication, thermography, etc.) [8,9]. By composing appropriate multi-component mixtures of cholesterol derivatives, one can obtain a wide variety of temperature dependences of the SR band with very large $\partial\lambda_{\max}/\partial T$ in a specified narrow temperature range. In addition, the selective reflection band in such systems can be very narrow (~ 20 nm and less). The general principles and ways of choosing the most suitable cholesteric composition had been established by early 1970-ies (probably the most detailed review can be found in [10]). So, in our experiments we re-visited these materials and used CLC based on cholesteryl alkanoates.

2. Experimental Methods

2.1. Chiral Liquid Crystalline Compositions

For our purposes, we required CLC mixtures that would exhibit strong temperature dependence of λ_{\max} close to the room temperature, with the given wavelength (corresponding to the testing laser radiation) falling onto the edge of the selective reflection band (long- or short-wavelength edge depending on the negative or positive sign of $\partial\lambda_{\max}/\partial T$). As a basic component of our mixtures, we used a formulation M1 comprising 60% cholesteryl nonanoate, 28% cholesteryl caprylate, and 12% cholesteryl caprylate. Two mixtures were made, optimized for two, at least, specified laser wavelengths (405 and 532 nm). Mixture #1 contained 44.9% M1, 29.9% cholesteryl valerate, and 25.2% cholesteryl oleyl carbonate, while Mixture #2 contained 55% M1, 35% cholesteryl valerate, 10% cholesteryl oleyl carbonate.

2.2. Experimental Techniques

Cells used in the experiments consisted of glass substrates, the inner surfaces of which were covered with rubbed PI for the establishment of a uniform planar texture; cell gap was established with Teflon spacers at approximately 10 μm . Liquid crystalline mixtures were introduced into cells in the isotropic state by capillary forces and cooled down to the room temperature in a natural way. The exact procedures of cell preparation were reported elsewhere [11,12].

Quite usual arrangements were employed for the measurements. Selective reflection spectra were measured for cells with planar texture (layer thickness 10 μm) by means of a Hitachi-330 spectrophotometer. Spectral measurements for

determination of the light polarization states were carried out with the use of a home-made hot-stage, providing the temperature stabilization not worse than 0.2 K, and a CCD-based spectrometer, all being computer controlled (Fig. 1).

Measurements of the changes of the polarization state of the linearly polarized light passed through the cell comprising CLC layer were carried out within the spectral range from 425 to 600 nm. For determination of the polarization plane orientation (or the semimajor axis of the light polarization ellipse in the case of elliptically polarized light) the intensity of light is usually measured for four different orientations of the analyzer, which is rotated by the step of 45° [13]. In our case the analyzer was fixed with respect to the CCD-spectrometer because of polarization sensitivity of the latter and, instead, the pair 'polarizer – hot stage' was rotated, correspondingly (Fig. 1). The angle between transmission axes of analyzer and polarizer has been, thus, consecutively established as: $\theta_i = 0, 45, 90$ and 135° , and the sets of four intensity spectra, $I_i(\lambda)$, were recorded for each temperature ($I_1 \div I_4$ correspond, respectively, to analyzer orientation angles $\theta_1 \div \theta_4$).

2.3. Determination of Polarization Parameters

Spectral dependences of the Stokes parameters [14], $S_i(\lambda)$, were calculated for each light wavelength from the corresponding sets of the experimental data:

$$\begin{aligned} S_0 &= I_1 + I_3 = I_2 + I_4 = \frac{1}{2} \sum I_i \\ S_1 &= I_3 - I_1 \\ S_2 &= I_2 - I_4 \end{aligned} \quad (1)$$

The last parameter, S_3 , was not determined in the experiment directly but its modulus was calculated using the relationship $S_0^2 = \sum S_i^2$ that comes from the definition of the Stokes parameters.

The dependence of the angle between the biggest axis of the ellipse and OX (Fig. 2) on the light wavelength, $\psi(\lambda)$, was determined in terms of the Stokes

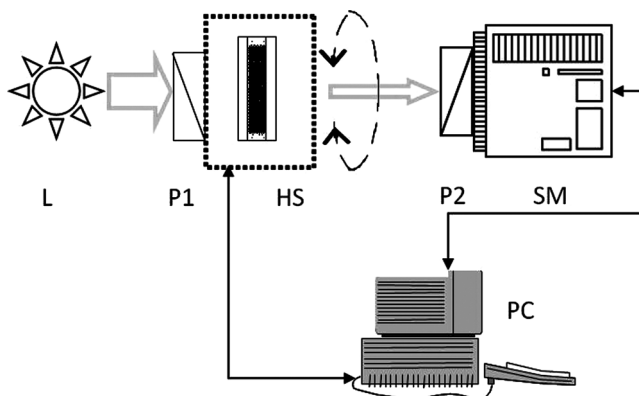


Figure 1. Scheme of experimental arrangement for spectral investigations of optical activity: L – light source, P1,2 – polarizers, HS – cell in hot stage, SM – spectrometer, PC – PC controlling unit.

parameters as $tg2\psi = S_2/S_1$ [14], while the ratio of the semiminor to the semimajor ellipse's axes (i.e., the ellipticity of transmitted light, $\varepsilon(\lambda) = \pm b/a$) could be calculated by virtue of the determined Stokes parameters via auxiliary angle χ : $tg\chi = \pm b/a$, where $\sin2\chi = S_3/\sqrt{S_1^2 + S_2^2 + S_3^2}$.

Based on the signs of the optical activity of CLC films for light wavelengths far beyond the selective reflection region, the angles of orientation of the semimajor axis of light polarization were calculated as function of the light wavelength taking account of the periodicity of tg function.

3. Results and Discussion

3.1. Temperature Dependence of the Selective Reflection

Selective reflection spectra for cells with planar texture of cholesteric LC mixtures were measured by means of the Hitachi-330 spectrophotometer at different temperatures (Fig. 3). The both mixtures exhibit decreasing of the helical pitch with rising temperature ($\partial\lambda_{\max}/\partial T < 0$).

Since the Mixture # 2 manifested stronger pitch vs. temperature dependence, all the following investigations were performed with this composition only. Temperature dependences of spectral positions of the maximum of the SR band as well as of its edges are presented in Figure 4.

It appeared that temperature increase by 10 K resulted in the shift of the selective reflection maximum by more than 60 nm while the width of the SR band remained almost unchanged.

3.2. Control of Light Polarization Plane Rotation

Polarized spectra $I_\lambda(\lambda)$ were recorded for the cell with the mixture # 2 in the temperature range $21 \div 33.5^\circ\text{C}$ with the step 0.5°C . The Stokes parameters were determined and, calculated from these, the angles of polarization rotation in dependence on the light wavelength and temperature are presented in Figure 5.

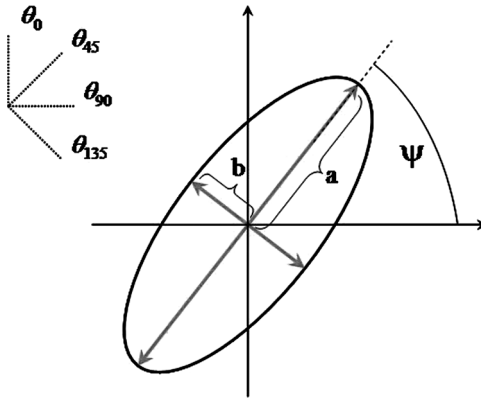


Figure 2. To determination of the light polarization parameters: θ_i are angles of the respective orientation of the analyzer; ψ is azimuth of light polarization ellipse orientation, a , b are the semimajor and the semiminor axes of light polarization ellipse.

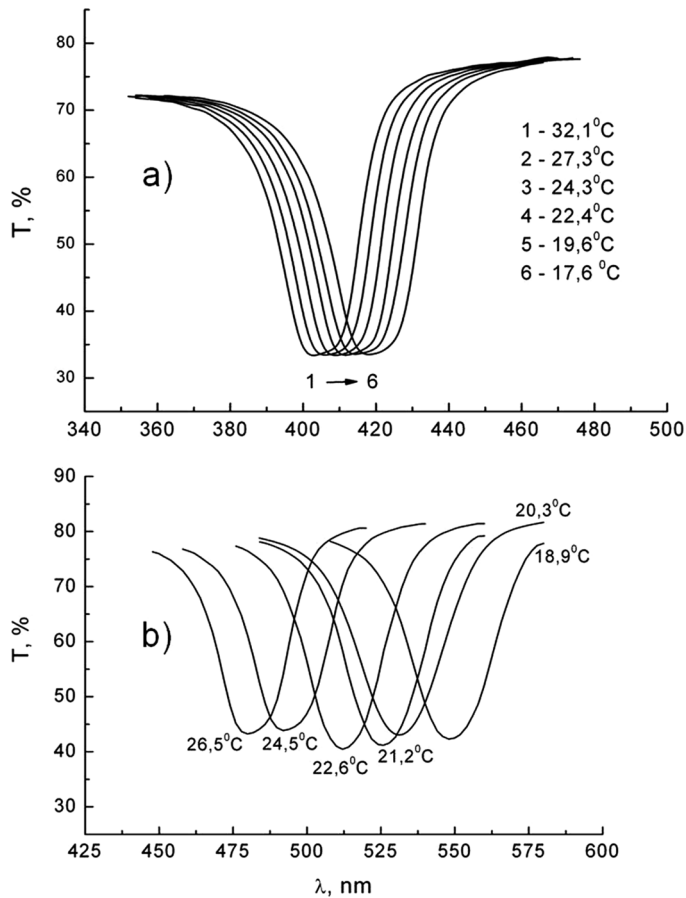


Figure 3. Selective reflection spectra for mixtures #1 (a) and #2 (b) at different temperatures.

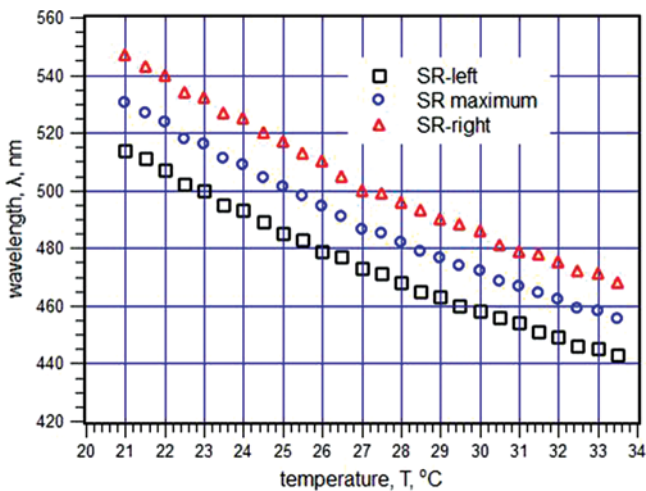


Figure 4. Temperature dependences of the maximum and the edges of SR band spectra for the mixture # 2. (Figure appears in color online.)

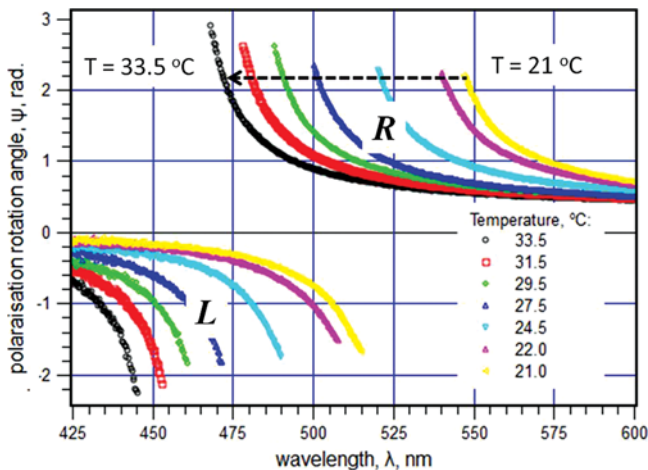


Figure 5. Angle of polarization plane rotation for the mixture #2 (change of the orientation of the semimajor axis of the polarization ellipse) measured at different temperatures: *L* (*R*) – leftwing (rightwing) edge of the SR band.

It is seen that the increase of the cell temperature by approximately 4 K results in the change of polarization plane orientation greater than $\pi/2$ for the light with wavelength falling at the leftwing edge of the selective reflection band (with light wavelength shorter than that of selective reflection band), while for the rightwing edge this polarization rotation appeared to be a bit smaller.

Comparison of the peculiarities of optical properties of CLC's that reveal different spectral position of the SR band is convenient to organize with respect to the reduced wavelength, $\lambda' = \lambda/\lambda_{SR}$ (λ_{SR} is the wavelength of the maximum of the selective reflection band).

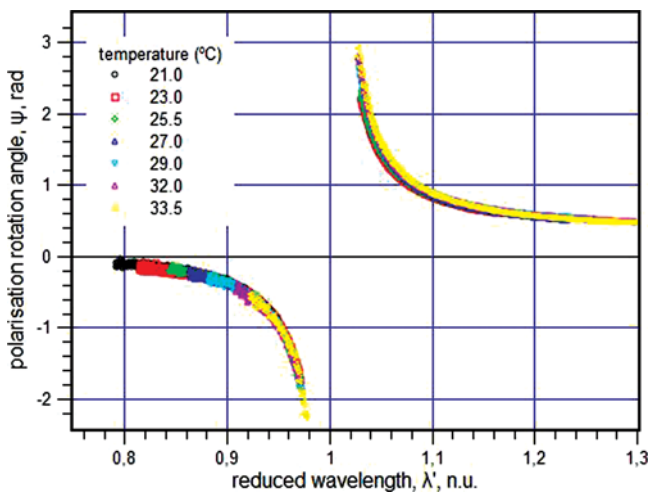


Figure 6. Angles of polarization plane rotation vs reduced wavelength, $\lambda' = \lambda/\lambda_{SR}$, measured at different temperatures of the mixture #2.

As it is seen from the Figure 6 the ability of the studied CLC compositions to rotate light polarization plane does not depend on the sample temperature that points out that the LC order parameter of the mixture practically appears to be constant within the experimental temperature interval.

4. Conclusions and Perspectives

It was shown that optical activity of thin films of multi-component compositions of the cholesterol esters depended on the period of the helical twisting only, which, in turn, exhibited strong temperature dependence. Thus, variation of the cell temperature by a few degrees could result in substantial rotation of polarization plane for the testing light with the given wavelength that could find application in the polarization switching.

Strong dispersion of optical activity at the edge of the selective reflection band contributes substantially to or even may be the only mechanism of optical nonlinearity [5]. One of the particularly promising peculiarities of the use of chiral LC for different aspects of optical signal processing and photonics, is the possibility to make optical read-out spectrally independent from the pumping by a proper adjustment of the helical pitch. Since the initial helical pitch depends on the chemical composition of the system it could be useful in controlling optical signals of very high intensity, for instance, in the IR range. Though cautiously keeping in mind constraints imposed by rather slow mechanisms guiding LC parameters, such systems could be considered for a possible application for all-optical wavelength conversion, all-optical switching or incoherent-to-coherent light transformation.

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