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Circular dichroism of cholesteric polymers and the orbital angular momentum of light

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We explore experimentally if light's orbital angular momentum (OAM) interacts with chiral nematic polymer films. Specifically, we measure the circular dichroism of such a material using light beams with different OAM. We investigate the case of strongly focused, nonparaxial light beams, where the spatial and polarization degrees of freedom are coupled. Within the experimental accuracy, we cannot find any influence of the OAM on the circular dichroism of cholesteric polymers.

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Molecular chirality is of very high importance in biology, chemistry, and materials science. Materials with molecular chirality can be investigated by optical means, because chiral molecules exhibit optical activity: Their interaction with light is sensitive to the circular polarization or helicity of the photons. This interaction is enantiomerically specific, thus giving information about the structure of chiral matter (see, e.g., [1]). One manifestation of optical activity is circular dichroism (CD), that is, light absorption sensitive to the handedness of the circular polarization. Optical activity is thus intrinsically linked to the *spin* angular momentum (SAM) of the photons. More recently, it was recognized that photons can additionally carry *orbital* angular momentum (OAM). Such photons appear naturally in Laguerre-Gaussian laser modes, where each photon carries $\ell\hbar$ of OAM (ℓ is the azimuthal mode index [2]). Does this additional degree of freedom play a role in CD?

If such an interaction were found, it is potentially useful for a broad range of research areas and applications, comparable to (spin-based) optical activity. Along this line, the electromagnetic interaction of OAM photons with atoms and molecules has received considerable attention. However, the situation is unclear: The interaction of OAM light with atoms and molecules has been studied in a number of theoretical papers, with controversial outcomes: Some predict that such an interaction should be observable within the electric dipole approximation [3,4], and some do not find such effects [5–8]. The latter outcome is supported by the only experimental investigation by Araoka *et al.* [9]. In that experiment, the authors tested whether the molecular CD of an optically active sample would be modified by using an OAM probe light, with negative results. Apparently, in a molecular system, the OAM does not participate in the same way in optical activity as the SAM does.

Since OAM is a spatial property, it makes sense to study if OAM has an effect in a system where the main contribution to optical activity does not stem from molecular chirality but from spatial resonances. Specifically, we address the structural chirality of a chiral nematic polymer, a polymerized variant of a cholesteric liquid crystal. In short-pitch chiral films, the alignment of the director has the same symmetry (helical) and periodicity (the optical wavelength) as the electric field vector in circularly polarized light (or as the wavefronts in OAM light). This results in (spin-based) optical activity effects orders of magnitude larger than in molecules. In this paper we investigate experimentally whether the OAM influences the CD of such a material.

We use a highly nonparaxial light beam so that the total angular momentum depends on the SAM and OAM in a nonseparable way [10]; this coupling emerges from spin-orbit interaction of light [11]. The coupling strength is proportional to $\theta_0^2/4$, where θ_0 is the (half-)aperture angle of the focused beam [12].

We can express the total angular momentum flux per unit length J_z, normalized to the energy flux per unit length E, as

$$\frac{J_z}{E} = \frac{\sigma_z + \ell}{\omega} + \frac{\sigma_z}{\omega}\left(\frac{4/\theta_0^2}{2p+\ell+1} + 1\right)^{-1},$$

where σ_z is the spin (± 1), ω the angular frequency, and ℓ and p the azimuthal and radial indices of the Laguerre-Gaussian mode. For low numerical aperture (NA) beams, the second term is negligible as in the case of Araoka [9]. We use a beam with NA = 0.55, which results in $\theta_0^2/4 = 0.1$; that is, contributions to the total angular momentum from terms involving both SAM and OAM are significant.[1]

Our cholesteric polymer films are based on chiral nematic mixtures [13–15] of a nematic reactive mesogen, 1, and a chiral nematic reactive mesogen, 2 [Figs. 1(a) and 1(b)]. Mesogens are monomers, which, if polymerized, show properties similar to those of liquid crystals. In our mixture, the wavelength of reflection can be adjusted by their mixing ratio. The materials are mixed at a 1:1 weight ratio to give reflection in the green part of the spectrum [Fig. 1(c)]. For photopolymerization, the isotropic photoinitiator Irgacure 369 (IRG369; Ciba) was added in a quantity of 1 wt%. To align the molecular layers at the bottom and top, a rubbed polyimide coating was used; to average out retardation effects, the rubbing directions were perpendicular with respect to each other. The thickness of the film was controlled by using 16-μm spacers.

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[1] In the experiment by Araoka *et al.* [9] the NA of the beam was apparently much lower than in our case.

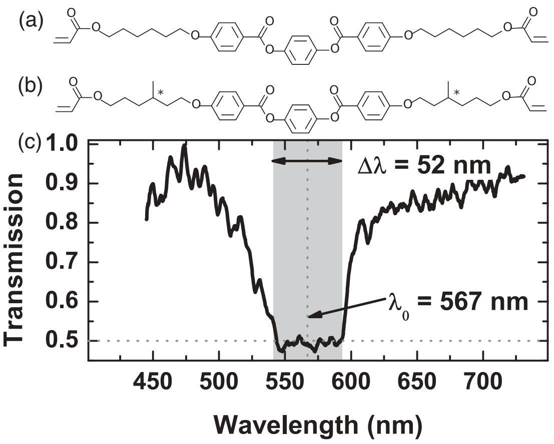

FIG. 1. (a, b) Structural formulas of the mesogens that we used. (c) Unpolarized transmission spectrum of the cholesteric polymer film. In the reflection band (gray box) the wavelength matches the pitch of the sample; there the reflected light's polarization has the same handedness as the cholesteric polymer, whereas the opposite polarization is transmitted.

The polymer was filled in the cell by using capillary forces and polymerized in its chiral nematic phase (at 85°C). Such a cholesteric polymer consists of helically oriented planes of parallel aligned molecules, and the pitch p_0 corresponds to a full 2π rotation of the director. This results in a Bragg-type reflection band which is sensitive to the circular polarization [16,17]: The polarization component with the same handedness as the director helix is reflected, while the other component interacts only weakly with the polymer and is transmitted. The polarization-averaged transmission spectrum (Fig. 1) therefore shows 50% transmission in the reflection band. From a nonchiral variant of the polymer, the ordinary and extraordinary refractive indices have been determined to be $n_o \approx 1.55$ and $n_e \approx 1.70$, respectively (at $\lambda = 500$ nm). From the full width of this band $\Delta\lambda = p_0 \Delta n$, where $\Delta n = |n_e - n_o|$, we can determine the pitch, $p_0 = 347$ nm; this agrees very well with the pitch determined from the reflection band center wavelength $\lambda_0 = \bar{n} p_0$, with the mean refractive index $\bar{n} = (n_o + n_e)/2$, $p_0 = 349$ nm.

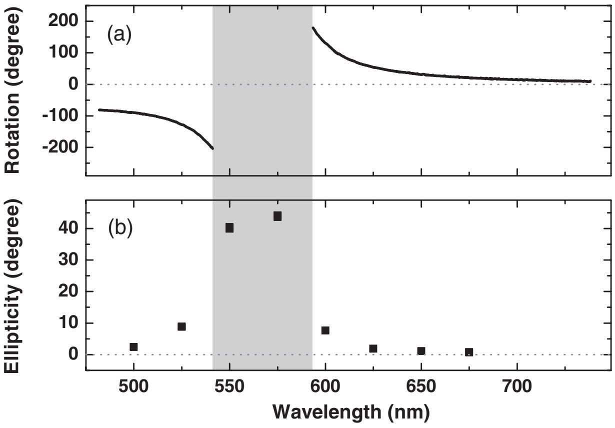

FIG. 2. Measured optical rotation (a) and circular dichroism (b) of the 16 μm thick cholesteric polymer. The optical rotation shows a dispersive behavior, while the CD shows that of a resonance. The CD is given as the ellipticity (circularly polarized light corresponds to an ellipticity of 45 deg). The gray box indicates the reflection band from Fig. 1.

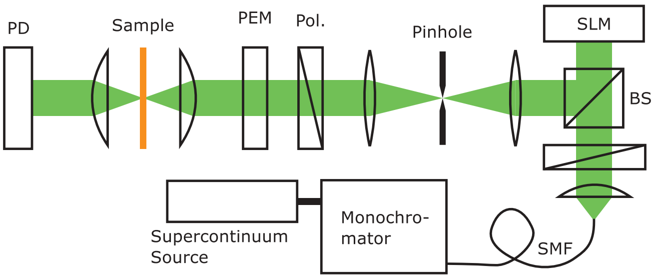

FIG. 3. (Color online) Scheme of the experiment. Pol., linear polarizer; PEM, photoelastic modulator; SLM, spatial light modulator; PD, photodiode; SMF, single-mode fiber. The PEM reference and the PD signal are connected to a lock-in amplifier to determine the circular dichroism.

We have characterized the optical activity of our cholesteric polymer using standard diagnostics. The (spin-based) optical rotation and CD of the sample are shown in Fig. 2. Conventional solutions of optically active molecules usually have a specific optical rotation of up to a few degrees per millimeter. Our cholesteric polymer by far exceeds this value; close to the reflection band, the optical rotation is about 2.3×10^4 deg/mm. In this band, the transmitted light is fully circularly polarized, and the optical rotation cannot be determined. Therefore, we have measured the CD, which can be done with a high precision at any spectral position. Figure 2(b) shows the ellipticity θ of the transmitted light; 45 deg corresponds to fully circularly polarized light.

For our experiment to study the effect of OAM on the CD, we use as a tunable light source, a supercontinuum source (Fianium SC1060), in combination with a monochromator (full width at half-maximum FWHM = 4 nm). After

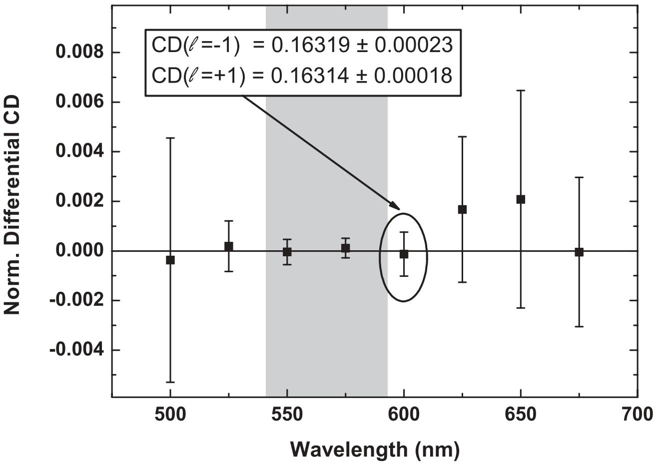

FIG. 4. OAM-differential circular dichroism (CD) of the cholesteric polymer. The measurement shows the normalized difference in CD for $\ell = -1$ and $\ell = +1$ OAM beams. The probe beam was focused using a 50×, NA = 0.55 objective. A nonvanishing differential CD would be an indication that the OAM influences CD. Inset: Measured CD signal at a wavelength of 600 nm, along with the uncertainty.

mode-filtering the output using a single-mode fiber, we synthesize the OAM mode by holographic beam shaping with a phase-only spatial light modulator (Fig. 3) in conjunction with a spatial filter (a pinhole in the Fourier plane of a lens). The polarization of this beam is controlled with a Glan-Taylor polarizer and a photoelastic modulator (PEM), which is set to modulate between left and right circular polarization. The sample is at the mutual focus between two microscopy objectives (Olympus MSPlan ULWD 50×, NA = 0.55; the backside aperture is filled by our light beam), forming a telescope. We record the transmitted light with a photodiode, this signal is fed to a lock-in amplifier which is phase locked to the PEM polarization modulation (50 kHz).

We test for a possible influence of the OAM on CD by comparing the CD signal of light with different OAM, $\ell = +1$ or $\ell = -1$, at various spectral positions, within and outside of the cholesteric reflection band. To isolate an OAM-induced effect, we introduce the normalized differential CD ($\Delta = V_{\mathrm{ac}}/V_{\mathrm{dc}}$ is the measured polarization-differential transmitted intensity normalized by the dc voltage):

$$\delta_{\mathrm{OAM}} = \frac{\Delta_{\ell=+1} - \Delta_{\ell=-1}}{\Delta_{\ell=+1} + \Delta_{\ell=-1}}$$

An influence of the OAM on CD would be demonstrated if $\delta_{\mathrm{OAM}} \neq 0$. Figure 4 shows the main experimental result. We conclude that the OAM has no effect here. For completeness we mention that the analogous experiment with a collimated beam, not using the microscopic telescope, shows equally a vanishing influence of OAM on the CD. We were able to determine the CD with a relative uncertainty of 0.1%; this is a significant improvement compared to the experiment by Araoka *et al.* [9], with an uncertainty of about 2%.

In conclusion, we did not observe interaction of light's OAM with a highly optically active cholesteric polymer in the nonparaxial regime. This has two implications. First, OAM light apparently does not interact with the spatial resonances in a cholesteric polymer. Second, this demonstrates that the conversion of SAM to OAM, as occurs at a high-NA lens, cannot be reversed: A lens cannot simply convert light's OAM to SAM. This agrees with recent theoretical results [11].

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[1] L. D. Barron, *Molecular Light Scattering and Optical Activity* (Cambridge University Press, Cambridge, 2004).

[2] L. Allen, M. W. Beijersbergen, R. J. C. Spreeuw, and J. P. Woerdman, Phys. Rev. A **45**, 8185 (1992).

[3] A. Alexandrescu, D. Cojoc, and E. Di Fabrizio, Phys. Rev. Lett. **96**, 243001 (2006).

[4] R. Jáuregui, Phys. Rev. A **70**, 033415 (2004).

[5] L. C. Dávila Romero, D. L. Andrews, and M. Babiker, J. Opt. B **4**, S66 (2002).

[6] D. L. Andrews, L. C. Dávila Romero, and M. Babiker (2003), e-print arXiv:physics/0305002.

[7] D. L. Andrews, L. C. Dávila Romero, and M. Babiker, Opt. Commun. **237**, 133 (2004).

[8] M. Babiker, C. R. Bennett, D. L. Andrews, and L. C. Dávila Romero, Phys. Rev. Lett. **89**, 143601 (2002).

[9] F. Araoka, T. Verbiest, K. Clays, and A. Persoons, Phys. Rev. A **71**, 055401 (2005).

[10] S. M. Barnett and L. Allen, Opt. Commun. **110**, 670 (1994).

[11] K. Y. Bliokh, M. A. Alonso, E. A. Ostrovskaya, and A. Aiello, Phys. Rev. A **82**, 063825 (2010).

[12] T. A. Nieminen, A. B. Stilgoe, N. R. Heckenberg, and H. Rubinsztein-Dunlop, J. Opt. A **10**, 115005 (2008).

[13] D. J. Broer and I. Heynderickx, Macromolecules **23**, 2474 (1990).

[14] R. A. M. Hikmet, J. Lub, and D. J. Broer, Adv. Mater. **3**, 392 (1991).

[15] J. Lub, D. J. Broer, R. A. M. Hikmet, and K. G. J. Nierop, Liq. Cryst. **18**, 319 (1995).

[16] W. D. St. John, W. J. Fritz, Z. J. Lu, and D.-K. Yang, Phys. Rev. E **51**, 1191 (1995).

[17] D. W. Berreman and T. J. Scheffer, Phys. Rev. Lett. **25**, 577 (1970).