



## **Deliverable Report**

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**Deliverable Title: OAM chiral writing and detection**

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**Name, title and organisation of the scientific representative of deliverable's lead beneficiary (task leader):**

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### **Deliverable table**

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<b>Deliverable name</b>	OAM chiral writing and detection
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**Subtask T3.2.1:** We will start our study using as model system a single mesoscopic spiral particle which is positioned in the focus of an OAM light beam. The spiral particle will be made out of photo-resist by means of holographic lithography using two counter-propagating Laguerre-Gauss (LG) beams (Philips Research will supply the technical know-how for this). The LG beams are to be normal to the substrate and have overlapping foci in the resist layer. If a  $l=\pm 1$  beam interferes with a  $l=0$  beam the resulting standing-wave pattern is a single spiral, whereas interference of  $l=\pm 1$  with  $l=\pm 1$  results in a double spiral; for symmetry reasons only the latter one leads to circular birefringence and is thus the only useful version. After photo-polymerization the unexposed resist is rinsed away so that a substrate with a free-standing spiral results. We plan to use a Fianium laser (a spectroscopically filtered white-light fiber laser) to study the OAM dependence of light scattering by the spiral. This source is tunable in the range 400-1000 nm. For variation of the OAM of the input beam (say  $l = -1, 0, +1$ ) we use a computer-generated fork hologram; we analyze in each case the OAM contents of the output beam and obtain thus the effect of the spiral-induced scattering on the output OAM intensity spectrum. We can also map the effect of this scattering on the phase relations between the OAM amplitude components, namely by using a superposition of OAM states as input beam. In this way we obtain the complete complex-valued scattering matrix. As an example of an OAM superposition, a HG01 beam is a phased superposition of LG01 and LG10 donut modes that travel with slightly different phase velocity along the spiral. As a result the nodal line of the HG01 mode is rotated. One may call this: OAM-type optical activity, as opposed to conventional optical activity which corresponds to rotation of linear polarization by passage through a circularly birefringent medium. This subtask is planned to be completed by milestone MS7, at mid-project time. *[Excerpt from GA-Annex I DoW]*

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Following this planning we have developed an interferometric photolithography set-up, using the standard SU-8 photo-resist. According to the manufacturer of this resist (Microchem) its spatial resolution depends largely on the pre- and post-exposure processing conditions; values of 50-100 nm are typical. The resist is spun as a thin layer on a substrate. The optical set-up uses a single-mode fiber-coupled laser, operating at around 400 nm; its output beam is divided at a beam splitter, then both beams are spatially modulated and then focused in a counter-propagating geometry onto the photo-resist layer. The spatial modulation is done using a spatial light modulator (SLM): we use a device from HoloEye (model Pluto) to perform the mode conversion from the fundamental fiber mode into the desired Laguerre-Gauss mode. The two beams are focused onto the substrate using long working distance, NA=0.55 objectives. For this type of counter-propagating interference lithography, optical path-length stability is of uttermost concern. Therefore, vibrations must be minimized in the relevant frequency range (down to 30 Hz). To accomplish this, we use a large, undamped optical table, on which we place a smaller 1.5x1.5 m breadboard, which is vibration isolated by automated pneumatic isolators. Interferometric measurements show that further active stabilization of the lithography setup is not required; this is essential because no working solution is known for counter-propagating lithography schemes. Solitary microspirals have thus been produced in the photo-resist layer covering the substrate. Since the pitch of the spiral particles has been designed as 200 nm, electron-beam microscopy (SEM) is needed to assess the quality of the lithographic fabrication process. We invoked the help of SEM experts for this diagnostic job; this involved the use of appropriate ultra-thin metallization layers.

The outcome of all this was rather disappointing; even 6 months after the nominal completion of the set-up we were not able to obtain consistent results. We feel that the prime reason for this is a lack



of effective resolution of the lithography. As a remedy we could spend more time to vary the SU-8 processing conditions; alternatively, we could explore other photo-resists. However, discussions with experts convinced us that either route would take at least several months more, without a guarantee for success. Therefore we have decided to give up on this interferometric lithography route to produce microspirals.

Instead, we have explored another approach towards fabricating microspirals that has the flavour of self-assembly, namely the use of a cholesteric liquid crystal; in essence, this can be seen as an array of parallel microspirals. It is well known that such samples have extremely strong spin-optical activity due to their Bragg resonance. We guessed that such a cholesteric liquid crystal could show also OAM optical activity if illuminated by a strongly focused light beam (for a paraxial light beam this cross-effect should of course be absent). We have obtained such cholesteric liquid crystal films from Professor D.J. Broer, formerly with Philips Research and now with the Eindhoven University of Technology; he is a well known expert in polymer chemistry. Having thus escaped the fabrication problem we have used these samples to study the interaction with OAM light; this is close to our original goal and is already phrased in the original formulation of subtask T.3.2.1 (see above). Briefly, we have been looking for the OAM equivalents of optical rotation and optical dichroism.

(i) OAM optical rotation by a cholesteric liquid crystal.

In theory, there are analogous transformations of light's spin and orbital angular momentum [Allen and Padgett, *J. Mod. Opt.* **54**, 487 (2007)]; however, none have been observed experimentally yet. In particular, it is unknown if there exists for the orbital angular momentum of light an effect analogous to the spin angular momentum-based optical rotation; this would manifest itself as a rotation of the corresponding Hermite-Gauss mode. We have performed an experimental search for this effect in a cholesteric liquid crystal polymer, using strongly focussed, spin-orbit coupled light beam (NA=0.4) at a number of frequencies outside the Bragg resonance band. Within the experimental accuracy we found no effect; specifically, we found that the relative phase velocities of the orbital modes constituting the Hermite-Gauss mode agree to within  $10^{-5}$ . This makes OAM-sensitive interaction in a cholesteric system highly unlikely. We have published this result in *Opt. Express* [1]; it constitutes **D3.2a**

(ii) OAM optical dichroism of a cholesteric liquid crystal.

In a similar set-up we have looked for OAM dichroism, i.e. the absorptive conjugate of OAM optical rotation. In this case the light beam was again sharply focussed (this time NA=0.55) to provide spin-orbit coupling. The light frequency was tuned over the full visible spectrum, including the Bragg resonance band. This experiment allowed us to probe the effect of OAM on the dichroism. Again we found a null result, the differential absorption of  $l=+1$  and  $l=-1$  was in the  $10^{-4}$ - $10^{-5}$  range. We have published this result in *Phys. Rev. A* [2]; it constitutes **D3.2.b**

It follows from these two experiments that apparently OAM-sensitive interactions do not occur in cholesteric liquid crystals, at least not within the quoted experimental accuracy. A full theory for this is not available; we deal here with electronic dispersion of a system with mesoscopic spatial structure. It all depends how one averages the dielectric response of this medium. As an ad-hoc argument one may say that, in order to enable interaction with OAM, a hypothetical medium must show a spatially extended (over the light's beam waist) coherent response. Evidently, the intermolecular dipole-dipole interactions that successfully explain the physics of liquid crystals do not suffice for this. Our hypothetical medium will need to have chiral symmetry and show a



coherent nonlocal response on the scale of the beam waist, i.e. we need a chiral optical antenna. Therefore a demonstration of OAM-sensitive interaction should be easiest in the THz or RF range.

In conclusion, the original subtask T3.2.1 could not be achieved due to fabrication problems of the microspirals; we found a way out of this fabrication problem by using cholesteric liquid crystals. Although these seem to show related light scattering properties on a heuristic level, this is not brought out by experiment. We have well documented our findings in 2 papers and we feel that we have thus reasonably achieved the goal of T3.2.1 (keeping in mind that this subtask was risky and that a negative result can be as useful as a positive one).

[1] “Search for Hermite-Gauss mode rotation in cholesteric liquid crystals”, W. Löffler, M. P. van Exter, G. W. ‘t Hooft, G. Nienhuis, D. J. Broer, J. P. Woerdman, *Optics Express* **19**, 12978-12983 (2011). (D3.2a)

[2] “Circular dichroism of cholesteric polymers and the orbital angular momentum of light”, W. Löffler, D. J. Broer, J. P. Woerdman, *Physical Review A* **83**, 065801 (2011). (D3.2b)

# Search for Hermite-Gauss mode rotation in cholesteric liquid crystals

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**Abstract:** In theory, there are analogous transformations of light's spin and orbital angular momentum [Allen and Padgett, *J. Mod. Opt.* **54**, 487 (2007)]; however, none have been observed experimentally yet. In particular, it is unknown if there exists for the orbital angular momentum of light an effect analogous to the spin angular momentum-based optical rotation; this would manifest itself as a rotation of the corresponding Hermite-Gauss mode. Here we report an experimental search for this effect in a cholesteric liquid crystal polymer, using strongly focussed, spin-orbit coupled light. We find that the relative phase velocities of the orbital modes constituting the Hermite-Gauss mode agree to within  $10^{-5}$ .

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**OCIS codes:** (160.1585) Chiral media; (030.4070) Modes; (350.4238) Nanophotonics and photonic crystals.

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## 1. Introduction

The circular polarization or *spin* angular momentum of light gives rise to circular birefringence in optically active media. It is well known that such media rotate the polarization of linear polarized light (i.e., optical rotation), which is a superposition of left- and right circularly polarized light. A beam of light can also have an *orbital* angular momentum (OAM) component of  $\ell\hbar$  per photon, this momentum is a spatial property and associated with an azimuthal component of the Poynting vector, best known to occur in Laguerre-Gauss laser modes. The analogue to linear polarization are here the Hermite-Gauss modes which are a superposition of opposite-handedness Laguerre-Gauss modes and carry no orbital angular momentum [1]. Allen and Padgett emphasized that the equivalent to optical (polarization) rotation is the (image-) rotation of the HG mode, which is a consequence of dispersion of the constituting LG modes of opposite handedness [2]. Such a situation can clearly be synthesized in an optical system which is not invariant under transverse translations, for instance by rotating an optical fiber bundle [2], or an image rotator. In stratified media, no experimental investigation of this issue has been done so far. For completeness we mention that the topic of our paper is related to the mechanical

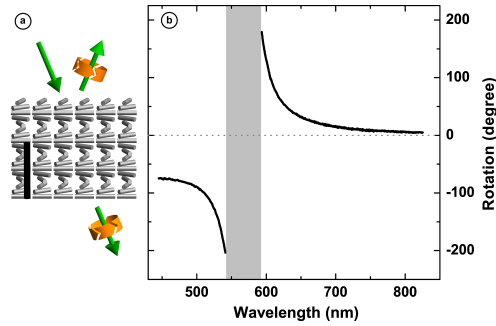


Fig. 1. Cholesteric liquid crystal. (a) Optical properties: If the wavelength of the incident light matches the pitch  $p_0$  of the cholesteric liquid crystal (black bar), the reflected (transmitted) light has the same (opposite) handedness as the CLC. (b) Experimentally measured optical rotation (in transmission, normal incidence) of linear polarized light, showing the expected dispersive behavior around the resonant reflection band (gray box).

Faraday effect [3] and to rotating media [4, 5].

Before addressing our experimental system we discuss the question: What is known about the interaction of OAM with matter in general? The first investigations along this line dealt with the mechanical angular momentum transfer of OAM light to astigmatic objects [6–8]. This has led to important developments in optical tweezers [6], and is well understood. The situation is quite different when we deal with the transfer of OAM to free atoms or molecules; this topic has been addressed in a number of theoretical papers [9–16] and one experimental paper [17]. Here, there is disagreement between the theory papers; some [12, 14] predict that effects of OAM of light on an atom or a molecule can be observed (in the linear regime and within the electric-dipole approximation) whereas others, including the only experimental paper [17], do not find such effects [10, 11, 13, 15]. One issue here is the discrepancy in the lateral dimension between the OAM mode on one side, and the atomic or molecular electron wavepacket on the other. This situation is different in the interaction of OAM light with macroscopic quantum objects, i.e., atomic and polaritonic condensates [18, 19], or, probably, using ultra-short wavelength OAM light and conventional atoms or molecules [20]. However, in none of these systems, the interaction is *per se* sensitive to the handedness of the OAM, which is key to find the OAM analogue to optical activity.

In the present paper we test experimentally for such behaviour, i.e., if the propagation depends on the handedness of the OAM, in a polymerized cholesteric liquid crystal (CLC). We chose this system because a CLC has a gigantic optical activity due to Bragg-like structural resonances (see, e.g., [21]). It is in particular interesting to consider interaction of a CLC with OAM light, since the helical alignment of the director in the cholesteric liquid crystal and the helical wavefronts in OAM light with  $\ell = \pm 1$  show similar symmetry.

In a stratified multilayer CLC (see inset Fig. 1), the optically anisotropic molecules are helically arranged. In every plane perpendicular to the substrate, the directors are aligned (due to rubbing of the substrates), and the director of adjacent LC planes is rotated a little due to molecular chirality, finally forming a helical orientation of the molecules. If the pitch of the director-helix is of the order of the optical wavelength, a polarization-selective Bragg-type reflection band appears [22]: The portion of the incoming light where the polarization handedness matches that of the CLC is reflected while the other is transmitted. These polarization-dependent optical properties can be explained on a macroscopic level using spatial dispersion, i.e., non-vanishing imaginary antisymmetric part (gyration) of the effective dielectric tensor [23]. This is well known in molecular optical activity, which can be seen as a consequence



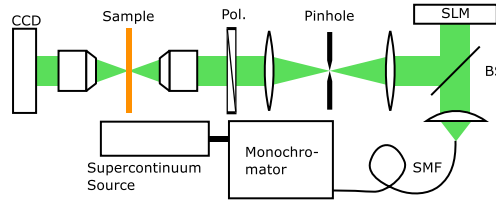


Fig. 2. Scheme of the experiment. Pol.: linear polarizers; PEM: photo-elastic modulator; SLM: spatial light modulator; PD: photo diode; SMF: single-mode fiber. The CCD is used to record the profile of the transmitted mode.

of molecular spatial dispersion. In optically active media, these atomic-scale inhomogeneities have chiral structure and give rise to a dielectric tensor with non-zero gyration. Spatial dispersion is also well known in photonic structures with inhomogeneities on the scale of the optical wavelength, e.g., photonic crystals (see, e.g., [24]). In this case, spatial dispersion does not originate from coherent oscillation of electrons in the unit cell, which has a size of order  $\lambda$ , but from multiple reflection, retardation, and interference of light. Well-known effects in this category in photonic crystals are the superprism effect [25] and negative refraction [26], and eventually, the optical activity of cholesteric liquid crystals like the one used here.

In analogy to optical rotation measurements [2], we measure the relative phase delay of OAM light with different handedness after passing through the CLC sample. This can be done simply by observing the rotation of the nodal line of a  $HG_{1,0}$  Hermite-Gaussian beam [2], since this orientation  $\phi$  depends on the relative phase of its components in the LG mode decomposition:

$$\cos(\phi)|HG_{1,0}\rangle + \sin(\phi)|HG_{0,1}\rangle \hat{=} e^{i\phi}|LG_{-1,0}\rangle + e^{-i\phi}|LG_{+1,0}\rangle \quad (1)$$

If we introduce in a HG beam a (hypothetical) medium with different effective refractive indices  $n_{+\ell}$  and  $n_{-\ell}$  of the  $LG_{-\ell,0}$  and  $LG_{+\ell,0}$  modes ( $n_{+\ell} \neq n_{-\ell}$  for  $\ell = 1$ ), i.e., providing intermodal dispersion of these modes, the HG mode pattern is rotated. This is the closest possible analogue to conventional linear optical activity.

## 2. Experimental setting

Light from a supercontinuum source (Fianium SC400, Fig. 2) is filtered spectrally using a monochromator, and then filtered spatially by coupling to a single-mode fiber. The Laguerre-Gaussian mode is synthesized using a phase-only spatial light modulator (SLM, Hamamatsu X10468) in conjunction with a spatial filter (a pinhole in the Fourier plane of a lens). We use linearly polarized light under normal incidence. The cholesteric sample is at the mutual focus between two microscopy objectives (20x, NA = 0.4) forming a telescope, and the transmitted light is recorded with a CCD (in the absence of loss, transmitted and reflected light are directly related and mode rotation would appear simultaneously in both). Both the SLM and the CCD are mounted on a strong solid support to avoid angular drifts. We determine the orientation  $\phi$  of the nodal line by computer analysis of the CCD mode images (Fig. 3), using the following technique: Since the nodal line of the HG mode picture is approximately vertical, we determine for each CCD row the horizontal node position by fitting a parabola to the horizontal intensity cross-section. The final step is linear regression of these horizontal node locations, from which the orientation of the nodal line, as well as its uncertainty, can be determined.

Our sample is a cholesteric liquid crystal polymer where the pitch  $p_0$  (a full  $2\pi$  rotation of the director, see Fig. 1) is of the order of the optical wavelength [27, 28], in our case, around 570 nm. The sample consists of a mixture of one chiral and one achiral reactive mesogen, and



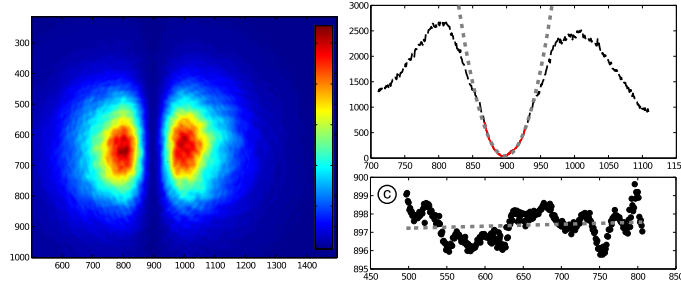


Fig. 3. Demonstration of the fit algorithm used to determine the nodal line orientation of the Hermite-Gaussian type mode: (a) 2D intensity profile; (b) example fit (intensity as a function of CCD column) of a horizontal cross section of this profile; and (c) linear regression (as a function of the CCD row) to find the global angle of this nodal line.

a photoinitiator for photopolymerization. The molecular layers are aligned at the bottom and top substrate by a rubbed polyimide coating, while the thickness of the film was controlled by using 16  $\mu\text{m}$  spacers. Photopolymerization was initiated in the chiral nematic phase at 85  $^{\circ}\text{C}$ . In the Bragg-type reflection band [22, 29] around  $\lambda_0 = \bar{n}p_0$  (full width  $\Delta\lambda = p_0\Delta n$ , where  $\Delta n = |n_e - n_o|$ ), the circular polarization component, which has the same handedness as the CLC, is reflected, while the other component is transmitted (see Fig. 1).  $\bar{n} = (n_o + n_e)/2$  is the average refractive index; for a non-chiral variant of the LC, the refractive indices at  $\lambda = 500\text{ nm}$  have been measured to be  $n_o \approx 1.55$  and  $n_e \approx 1.70$ . Fig. 1b shows the typical dispersion-like optical rotation of the sample with a reflection band at 550-590 nm. The optical rotation is very large: Close to the reflection band, the optical rotation is around  $2.3 \times 10^4$  degrees/mm, this is very much larger than what is possible in samples with molecular optical activity only. This allows us to use a thin sample and microscopy objectives for focussing.

In strongly focussed light beams, the spin and orbital angular momentum are coupled via spin-orbit interaction. More precisely, in a Laguerre-Gaussian beam, the total angular momentum flux per unit length  $J_z$ , normalized to the energy flux per unit length  $E$  is given by [30]:

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

where  $\sigma_z$  is the spin AM,  $\omega$  the wave number,  $\ell$  and  $p$  the azimuthal and radial index of the Laguerre-Gaussian mode, and  $\theta_0$  is the half aperture angle of the focussed beam. Eq. (2) follows directly from that in [30] using  $z_R = \pi w_0^2/\lambda$  and  $w_0 = \lambda/(\pi\theta)$ , where  $w_0$  and  $z_R$  are the Gaussian beam width and Rayleigh range, respectively. The second term in Eq. (2) is the consequence of spin-orbit coupling, and  $\theta_0^2/4$  is a measure of its strength [31]. We use a beam with  $NA = 0.4$ , this results in  $\theta_0^2/4 = 0.05$ . That is, about 5% of the total angular momentum stems from terms where both the SAM and OAM contribute in a non-separable way.

### 3. Results and discussion

Finally, Fig. 4 shows our experimental result: Within our experimental accuracy of around 0.05 degree, we cannot find a rotation of the nodal line of the Hermite-Gaussian mode in the non-paraxial regime while passing through the cholesteric liquid crystal. This accuracy is good enough to exclude that the spin-orbit mixed components (5%) of the light mediate Hermite-Gaussian mode rotation via optical rotation (up to 200 degrees, Fig. 1). Similar results were obtained in the paraxial case (not shown). Our experiment tested for intermodal dispersion of OAM modes with different handedness, which is the closest analogy of optical rotation for

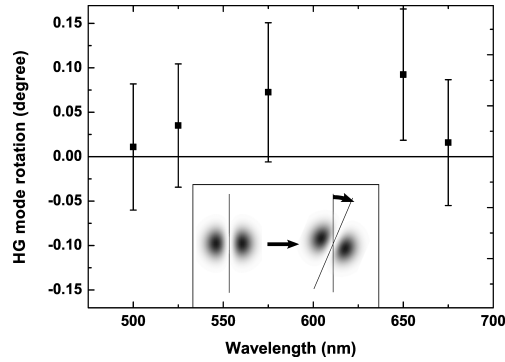


Fig. 4. Measurement of the  $HG_{1,0}$ -mode rotation angle (inset) at different wavelengths for focussed probe light (NA=0.4).

OAM. In terms of refractive-index difference, our results give an upper limit of  $|n_{+\ell} - n_{-\ell}| < 8 \cdot 10^{-6}$  for  $\ell = 1$ . Probably this limit could be lowered by using a specially designed superposition of higher-order OAM modes, however, our data analysis method works best for the  $\ell = \pm 1$  case.

One might believe that this result is to be expected in our case: To rotate the plane-wave decomposition of a focused HG mode, we require  $\vec{k}_{out} = R \cdot \vec{k}_{in}$ , where  $R$  is a rotation matrix which rotates the wave vector  $\vec{k}_{in}$  around the geometric beam axis. If the medium is translation invariant in the sample plane, in linear optics, such a  $R$  can not be constructed. However, this translation symmetry holds only within an effective refractive index model, but not on the atomic scale. Even if the wavelength is much larger than the microscopic length scale of the system (e.g., the crystal lattice constant), the optical properties are influenced by the microscopic structure. This is well known in crystal optics: Within the dipole approximation, cubic crystals should be isotropic, however, many show birefringence due to spatial dispersion. This effect has been predicted by Lorentz in 1878, firstly discovered in [32], and became relevant in UV lithography today [33]. It is not clear yet if such structure could allow for OAM-sensitive scattering. Compared to conventional crystals, in our helically-arranged cholesteric liquid crystal polymer, the transverse symmetry is broken on a larger, supramolecular scale. Our results, however, make a OAM-sensitive interaction in a cholesteric system highly unlikely.

It appears, to make a medium react on OAM, that a spatially extended (comparable to the light's beam waist) coherent response of the medium is required; and evidently the intermolecular dipole-dipole interactions, which explain successfully the physics of liquid crystals, do not suffice. Similarly, the storage of spatial phase information in a classical gas of cold atoms [34] is entirely different from vortex creation in a BEC atomic condensate using OAM light [18]. In the first case, only the local phase information is stored (similar to holography), and only in the latter case, a single quantum object carries the OAM information previously carried by the light. A hypothetical medium with OAM-sensitive interaction will need to have a chiral symmetry and give a coherent non-local response on the scale of the beam waist, i.e., a kind of chiral optical antenna. The construction of a medium from such building blocks is even more challenging than the development of metamaterials since the coherence length must be considerably larger here. Therefore, a demonstration should be easiest in the THz or radio-frequency range.

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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 ( $\ell$  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  $\theta_0$  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  $\sigma_z$  is the spin ( $\pm 1$ ),  $\omega$  the angular frequency, and  $\ell$  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.<sup>1</sup>

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- $\mu$ m spacers.

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

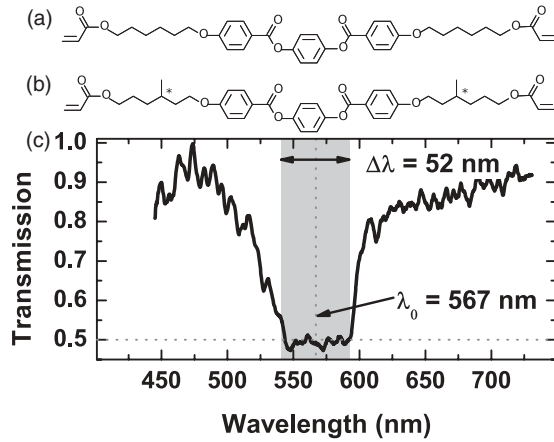


FIG. 1. (a, b) Structural formulas of the mesogens that we used. (c) Unpolarized transmission spectrum of the cholesteric polymer film. In the reflection band (gray box) the wavelength matches the pitch of the sample; there the reflected light's polarization has the same handedness as the cholesteric 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  $2\pi$  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

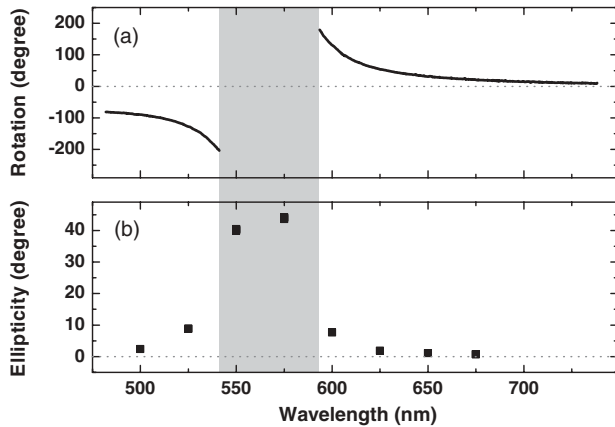


FIG. 2. Measured optical rotation (a) and circular dichroism (b) of the 16  $\mu\text{m}$  thick cholesteric polymer. The optical rotation shows a dispersive behavior, while the CD shows that of a resonance. The CD is given as the ellipticity (circularly polarized light corresponds to an ellipticity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

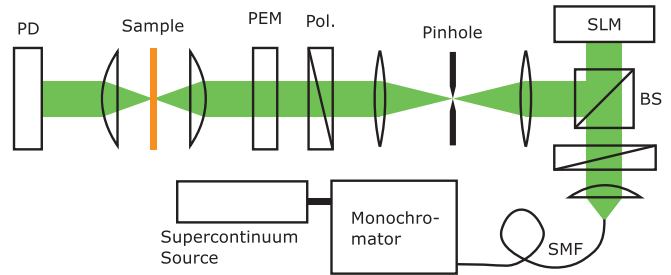


FIG. 3. (Color online) Scheme of the experiment. Pol., linear polarizer; PEM, photoelastic modulator; SLM, spatial light modulator; PD, photodiode; SMF, single-mode fiber. The PEM reference and the PD signal are connected to a lock-in amplifier to determine the circular dichroism.

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.

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  $2.3 \times 10^4$  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  $\theta$  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

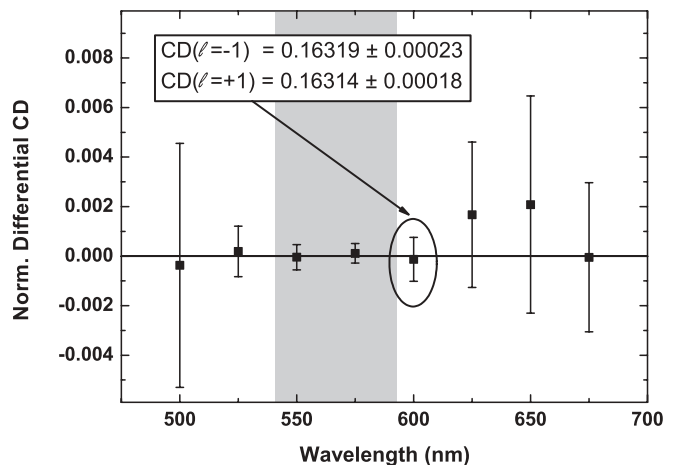


FIG. 4. OAM-differential circular dichroism (CD) of the cholesteric polymer. The measurement shows the normalized difference in CD for  $\ell = -1$  and  $\ell = +1$  OAM beams. The probe beam was focused using a 50 $\times$ , NA = 0.55 objective. A nonvanishing differential CD would be an indication 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 $\times$ , 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 ( $\Delta = V_{ac}/V_{dc}$  is the measured polarization-differential transmitted intensity normalized by the dc voltage):

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

An influence of the OAM on CD would be demonstrated if  $\delta_{\text{OAM}} \neq 0$ . 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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