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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}.

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

References and links

1. M. J. Padgett and J. Courtial, "Poincaré-sphere equivalent for light beams containing orbital angular momentum," Opt. Lett. **24**, 430–432 (1999).
2. L. Allen and M. Padgett, "Equivalent geometric transformations for spin and orbital angular momentum of light," J. Mod. Opt. **54**, 487–491 (2007).
3. G. Nienhuis, J. P. Woerdman, and I. Kuščer, "Magnetic and mechanical faraday effects," Phys. Rev. A **46**, 7079–7092 (1992).
4. M. Padgett, G. Whyte, J. Girkin, A. Wright, L. Allen, P. Öhberg, and S. M. Barnett, "Polarization and image rotation induced by a rotating dielectric rod: an optical angular momentum interpretation," Opt. Lett. **31**, 2205–2207 (2006).
5. J. Leach, A. J. Wright, J. B. Götte, J. M. Girkin, L. Allen, S. Franke-Arnold, S. M. Barnett, and M. J. Padgett, "'aether drag' and moving images," Phys. Rev. Lett. **100**, 153902 (2008).
6. H. He, M. E. J. Friese, N. R. Heckenberg, and H. Rubinsztein-Dunlop, "Direct observation of transfer of angular momentum to absorptive particles from a laser beam with a phase singularity," Phys. Rev. Lett. **75**, 826–829 (1995).
7. N. B. Simpson, K. Dholakia, L. Allen, and M. J. Padgett, "Mechanical equivalence of spin and orbital angular momentum of light: an optical spanner," Opt. Lett. **22**, 52–54 (1997).
8. M. W. Beijersbergen, L. Allen, H. E. L. O. van der Veen, and J. P. Woerdman, "Astigmatic laser mode converters and transfer of orbital angular momentum," Opt. Commun. **96**, 123–132 (1993).
9. S. J. van Enk and G. Nienhuis, "Commutation rules and eigenvalues of spin and orbital angular momentum of radiation fields," J. Mod. Opt. **41**, 963–977 (1994).
10. M. Babiker, C. R. Bennett, D. L. Andrews, and L. C. Dávila Romero, "Orbital angular momentum exchange in the interaction of twisted light with molecules," Phys. Rev. Lett. **89**, 143601 (2002).
11. L. C. Dávila Romero, D. L. Andrews, and M. Babiker, "A quantum electrodynamics framework for the nonlinear optics of twisted beams," J. Opt. B **4**, S66 (2002).
12. R. Jáuregui, "Rotational effects of twisted light on atoms beyond the paraxial approximation," Phys. Rev. A **70**, 033415 (2004).

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13. D. L. Andrews, L. C. Dávila Romero, and M. Babiker, "On optical vortex interactions with chiral matter," Opt. Commun. **237**, 133–139 (2004).
14. A. Alexandrescu, D. Cojoc, and E. D. Fabrizio, "Mechanism of angular momentum exchange between molecules and laguerre-gaussian beams," Phys. Rev. Lett. **96**, 243001 (2006).
15. R. Grinter, "Photon angular momentum: selection rules and multipolar transition moments," J. Phys. B **41**, 095001 (2008).
16. V. Klimov, D. Bloch, M. Ducloy, and J. R. R. Leite, "Detecting photons in the dark region of Laguerre-Gauss beams," Opt. Express **17**, 9718–9723 (2009).
17. F. Araoka, T. Verbiest, K. Clays, and A. Persoons, "Interactions of twisted light with chiral molecules: an experimental investigation," Phys. Rev. A **71**, 055401 (2005).
18. M. F. Andersen, C. Ryu, P. Cladé, V. Natarajan, A. Vaziri, K. Helmerson, and W. D. Phillips, "Quantized rotation of atoms from photons with orbital angular momentum," Phys. Rev. Lett. **97**, 170406 (2006).
19. D. Sanvitto, F. M. Marchetti, M. H. Szymanska, G. Tosi, M. Baudisch, F. P. Laussy, D. N. Krizhanovskii, M. S. Skolnick, L. Marrucci, A. Lemaitre, J. Bloch, C. Tejedor, and L. Vina, "Persistent currents and quantized vortices in a polariton superfluid," Nat. Phys. **6**, 527–533 (2010).
20. A. Picón, J. Mompart, J. R. V. de Aldana, L. Plaja, G. F. Calvo, and L. Roso, "Photoionization with orbital angular momentum beams," Opt. Express **18**, 3660–3671 (2010).
21. I. Khoo, *Liquid Crystals*, Wiley series in pure and applied optics (Wiley-Interscience, 2007).
22. W. D. St. John, W. J. Fritz, Z. J. Lu, and D.-K. Yang, "Bragg reflection from cholesteric liquid crystals," Phys. Rev. E **51**, 1191–1198 (1995).
23. L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon Press, 1975), Vol. 8.
24. M. Notomi, "Manipulating light with strongly modulated photonic crystals," Rep. Prog. Phys. **73**, 096501 (2010).
25. H. Kosaka, T. Kawashima, A. Tomita, M. Notomi, T. Tamamura, T. Sato, and S. Kawakami, "Superprism phenomena in photonic crystals," Phys. Rev. B **58**, R10096–R10099 (1998).
26. E. Cubukcu, K. Aydin, E. Ozbay, S. Foteinopoulou, and C. M. Soukoulis, "Electromagnetic waves: negative refraction by photonic crystals," Nature **423**, 604–605 (2003).
27. D. J. Broer and I. Heynderickx, "Three-dimensionally ordered polymer networks with a helicoidal structure," Macromolecules **23**, 2474–2477 (1990).
28. R. A. M. Hikmet, J. Lub, and D. J. Broer, "Anisotropic networks formed by photopolymerization of liquid-crystalline molecules," Adv. Mater. **3**, 392–394 (1991).
29. D. W. Berreman and T. J. Scheffer, "Bragg reflection of light from single-domain cholesteric liquid-crystal films," Phys. Rev. Lett. **25**, 577–581 (1970).
30. S. M. Barnett and L. Allen, "Orbital angular momentum and nonparaxial light beams," Opt. Commun. **110**, 670–678 (1994).
31. T. A. Nieminen, A. B. Stilgoe, N. R. Heckenberg, and H. Rubinsztein-Dunlop, "Angular momentum of a strongly focused gaussian beam," J. Opt. A **10**, 115005 (2008).
32. J. Pastrnak and K. Vedam, "Optical anisotropy of silicon single crystals," Phys. Rev. B **3**, 2567–2571 (1971).
33. A. G. Serebryakov and F. Bociort, "Spatial dispersion of crystals as a critical problem for deep uv lithography," J. Opt. Technol. **70**, 566–569 (2003).
34. J. W. R. Tabosa and D. V. Petrov, "Optical pumping of orbital angular momentum of light in cold cesium atoms," Phys. Rev. Lett. **83**, 4967–4970 (1999).

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

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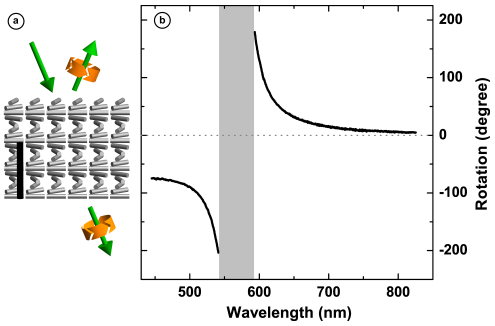

Fig. 1. Cholesteric liquid crystal. (a) Optical properties: If the wavelength of the incident light matches the pitch p_0 of the cholesteric liquid crystal (black bar), the reflected (transmitted) light has the same (opposite) handedness as the CLC. (b) Experimentally measured optical rotation (in transmission, 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

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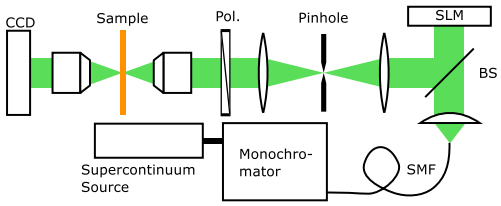

Fig. 2. Scheme of the experiment. Pol.: linear polarizers; PEM: photo-elastic modulator; SLM: spatial light modulator; PD: photo diode; SMF: single-mode fiber. The CCD is used to record the profile of the transmitted mode.

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 λ, 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 ϕ depends on the relative phase of its components in the LG mode decomposition:

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

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 polarizated 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 ϕ 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 2π 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

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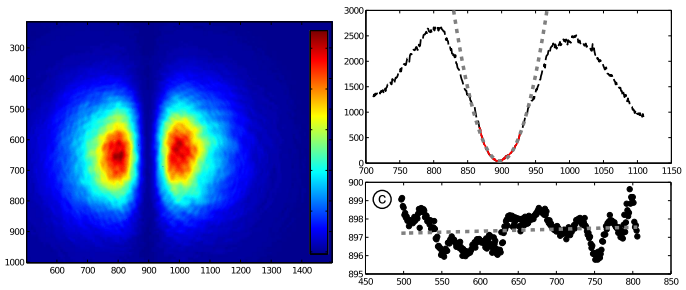

Fig. 3. Demonstration of the fit algorithm used to determine the nodal line orientation of the Hermite-Gauss type mode: (a) 2D intensity profile; (b) example fit (intensity as a function of CCD column) of a horizontal cross section of this profile; and (c) linear regression (as a function of the CCD 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 μm spacers. Photopolymerization was initiated in the chiral nematic phase at 85 $^{\circ}C$. 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 $\lambda = 500\,nm$ 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 2.3×10^4 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]:

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

where σ_z is the spin AM, ω the wave number, ℓ and p the azimuthal and radial index of the Laguerre-Gaussian mode, and θ_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

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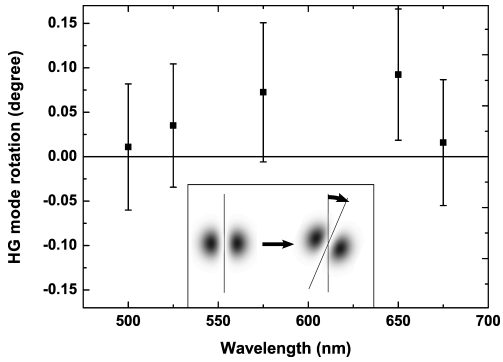

Fig. 4. Measurement of the $HG_{1,0}$-mode rotation angle (inset) at different wavelengths for focussed probe light (NA=0.4).

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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