Anisotropic Weak Localization of Light

Riccardo Sapienza,[1] Sushil Mujumdar,[1] Cecil Cheung,[2] A. G. Yodh,[2] and Diederik Wiersma[1,*]

[1]*European Laboratory for Nonlinear Spectroscopy and INFM, 50019 Sesto Fiorentino (Florence), Italy*[†]

[2]*Department of Physics and Astronomy, University of Pennsylvania, Philadelphia, Pennsylvania 19104, USA*

(Received 21 July 2003; published 23 January 2004)

We have observed angular anisotropy in weak localization of light from highly scattering, orientationally ordered, nematic liquid crystals. This demonstration of angular anisotropy in a multiple-scattering interference phenomenon was facilitated by a light scattering instrument with extraordinary angular resolution. The measured anisotropies were consistent with a simple model of coherent backscattering generalized for propagation-direction dependent mean free paths.

DOI: 10.1103/PhysRevLett.92.033903 PACS numbers: 42.25.Dd, 42.25.Hz, 42.70.Df, 61.30.Gd

Propagation of light waves in disordered, strongly scattering dielectrics is complex and full of surprises [1]. Perhaps the simplest form of transport in such dielectrics is light diffusion. The fundamental parameter in the light diffusion model is the transport mean free path, corresponding to the distance light travels in the medium before its direction is randomized. Surprisingly, interference effects can survive random multiple light scattering and lead to striking phenomena beyond diffusion theory [1–4]. The most robust of these interference phenomena is weak localization of light [2], which originates from the fundamental concept of reciprocity. In weak localization, interference leads to a net reduction of light transport similar to the weak localization phenomenon for electrons in disordered (semi)conductors. It is often seen as the precursor to Anderson (or strong) localization of light [3]. Weak localization of light can be detected since it is manifest as an enhancement of light intensity in the backscattering direction. This enhancement is called the cone of coherent backscattering. Since the first experimental observation of coherent backscattering from colloidal suspensions [2], the phenomenon has been successfully studied in strongly scattering powders [5,6], cold atom gases [7], two-dimensional random systems of rods [8], randomized laser materials [9], disordered liquid crystals [10,11], and even photonic crystals [12].

Liquid crystals in the nematic phase exhibit multiple light scattering, but, in contrast to most disordered materials, are orientationally ordered [13]. The simplest consequence of orientational anisotropy in multiple scattering is manifest in light diffusion. Theory predicts [14] and experiments confirm [15] that monodomain nematic liquid crystals are characterized by an anisotropic light diffusion constant. It is interesting to speculate about the possible survival of anisotropy effects in interference phenomena such as weak and strong localization. For weak localization, the width of the backscattering cone depends on the transport mean free path rather than the light diffusion constant. Mean free path anisotropy should therefore be manifest in angular anisotropy of the backscattering cone, even at very high orders of scattering. While pioneering experiments on coherent backscattering from monodomain liquid crystals have been performed [16], no measurements of the transport mean free path length or backscattering cone anisotropy have been reported. Thus, important questions remain open about how weak and strong localization are influenced by anisotropic multiple scattering.

In this Letter, we report the observation of anisotropic weak localization of light. We resolved the width of the coherent backscattering cone from an ordered nematic liquid crystal and found that it exhibits an angular anisotropy dependent on the direction of the nematic director. We introduce a simple model of coherent backscattering, generalized for an anisotropic transport mean free path, to account for the angular shape and width of the cone. We also determine the transport mean free paths in the two principal propagation directions.

The observation and study of weak localization from liquid crystals requires an extremely high angular resolution of at least 20 rad. To achieve such resolution we used the following scheme. The output of a single mode argon laser (2 W, 488 nm) was spatially filtered and collimated into an 8 cm diameter beam and reflected by a 15 cm wide beam splitter onto the sample. The backscattered light from the sample was collected through the beam splitter by a wide achromatic triplet lens (f 1250 mm) and monitored by a photomultiplier tube through a polarizer and a 10 m diameter pinhole placed exactly in the focal plane of the achromatic lens. All lenses were aligned perpendicular to the optical beams to avoid astigmatism, and the response of the setup was carefully checked to be isotropic. The measured noise level was always lower than 1% of the signal. An angular resolution of 10 rad was obtained, 2 orders of magnitude higher than previous liquid crystal work and 5 times better than the highest resolution reported in literature. This angular resolution enabled us to measure transport mean free paths larger than 1 mm, opening up the possibility to perform weak localization studies on complex fluids and biological tissue inaccessible to previous coherent backscattering instruments.

The liquid crystal *p*-pentyl-*p*′-cyanobiphenyl (5CB) was contained in a cylindrical cell of 8 cm diameter and 4 cm thickness which satisfied the requirement of having an optically thick sample. A monodomain nematic phase was obtained by heating the system well above the nematic-isotropic phase transition at 309 K and then cooling slowly overnight to 301 K in an external magnetic field of 0.5 T. The sample was left in the magnetic field for several days to assure good director alignment. Magnetic fields have been observed to modify multiple light scattering, leading to fascinating phenomena such as the photonic Hall effect [17], the breaking of time-reversal symmetry [18], and the elegant Hanle effect in coherent backscattering [19]. None of these effects play a role in our samples since the anisotropy is an intrinsic optical property of the nematic phase. Scattering in nematics is dominated by fluctuations of the otherwise aligned director and the magnetic field is used to obtain a monodomain phase. In addition, it suppresses long range orientational fluctuations in the sample that are larger than the magnetic coherence length . In our case the field strength is modest and 4.2 m, much larger than the wavelength, but orders of magnitude smaller than all sample dimensions, which assures that surface anchoring effects can be neglected.

In Fig. 1 we report the observed coherent backscattering cone for two monodomain cases and one polydomain case. The director in the two monodomain cases is in the sample plane in either the x or the y direction. The polarization is linear in the x direction and the polarization conserving channel is monitored. A clear angular anisotropy is visible that depends on the nematic director orientation. The middle panel in Fig. 1 shows the result for a polydomain nematic phase which was obtained by heating the sample into the isotropic phase and cooling down in zero field while vibrating the system. In a polydomain phase the scattering anisotropy still exists but only on length scales much smaller than the sample size. Hence, the anisotropy in the mean free path averages out over the whole sample volume and one expects to observe an isotropic coherent backscattering cone. We observe from Fig. 1(b) that the anisotropy indeed disappears in the polydomain nematic case.

In Fig. 2 we report the coherent backscattering cones as recorded in long linear scans in the two orthogonal scanning directions, after precise determination of the exact backscattering angle. We can observe that the coherent backscattering cone in the scattering plane parallel to the nematic director is narrower than the cone in the perpendicular plane. Note that the observed anisotropy cannot be due to polarization effects at the sample surface (such as birefringent internal reflection [20]) since we are comparing angular scans with the same polarization direction. The solid line in Fig. 2 was obtained from a simple coherent backscattering model generalized for an anisotropic system, described below. Figure 2 confirms

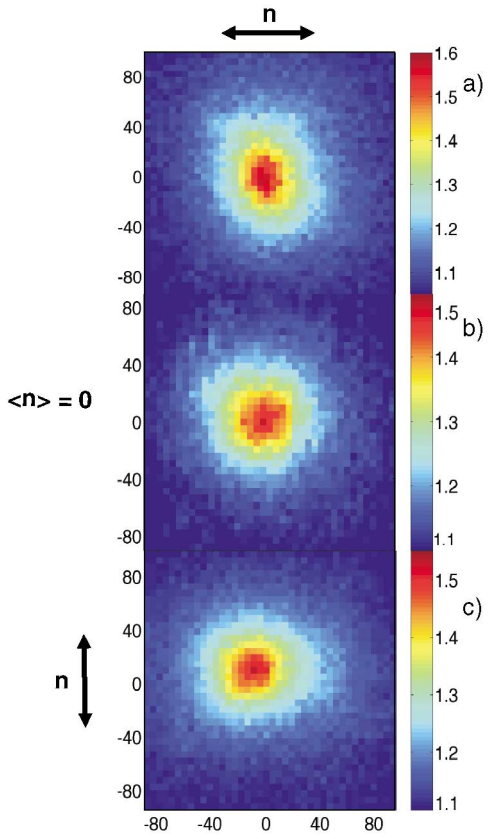

FIG. 1 (color). Coherent backscattering cones in polar color plots from nematic liquid crystal for three cases of the nematic director. Top: nematic director in x; bottom: director in y; middle: polydomain phase. The polarization is in the x direction in all cases.

that the experimental data correspond well to the classical theoretical cone shape.

Anisotropy in coherent backscattering due to an anisotropic transport mean free path has been predicted numerically in Monte Carlo simulations [21]. However, no

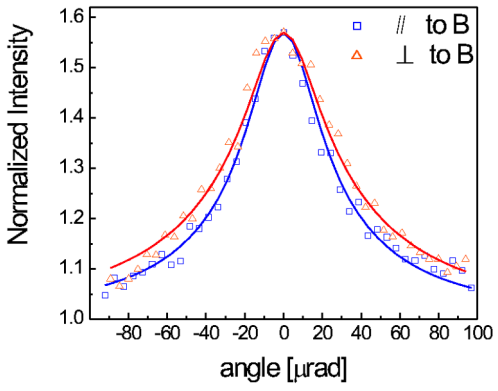

FIG. 2 (color online). Coherent backscattering from a monodomain nematic for both orthogonal scanning directions. Nematic director and polarization in the x direction.

exact theory on coherent backscattering from nematic liquid crystals is currently available. As a preliminary step in the direction of a full theory, we can generalize the standard coherent backscattering model for an anisotropic mean free path. The total normalized backscattered intensity due to multiple scattering is the sum of a weakly angular dependent diffuse background $_{\ell}$ and a contribution from interference between reciprocal light paths $_{c}$ [2]. The $_{\ell}$ term is completely flat in the angular range of our experiment and all interference effects are contained in $_{c}$. The enhancement factor E is defined as the enhancement of the intensity in the exact backscattering direction due to this interference. Its theoretical value is 2, since in exact backscattering $_{\ell}$ $_{c}$. In practice, one has to account for single scattering and stray light, which reduce the observed enhancement factor. The normalized backscattered intensity I is [6]

$$I \quad \frac{E \quad 1 \quad _{c})+ \quad _{\ell}}{_{\ell}}, \tag{1}$$

with E the experimentally obtained enhancement factor. We observe in Fig. 2 that the enhancement factor is 1.55 0.05, which is an excellent result considering the difficulties associated with resolving such a narrow backscattering cone [22].

We follow a standard approach for calculating $_{c}$ in which one solves Maxwell's equations for a random collection of point scatterers in a self-consistent way [2,9]. $_{c}$ can then be written as an integral over the distribution of distances between first and last scattering event times a cosine factor that accounts for the interference. This integral can be solved analytically for a sample in a slab geometry. In an anisotropic system such as a nematic liquid crystal, the distribution of distance becomes anisotropic, leading to an anisotropic backscattering cone. Assuming that the random walk in the three orthogonal propagation directions is uncoupled, we obtain for the interference contribution in the anisotropic case:

$$_{c} , \varphi \quad \frac{3}{2\ell^{3}\alpha u} \frac{+ u \ 1 \quad \exp \quad 2\alpha z_{0}}{u + \quad ^{2} + \quad ^{2}}, \tag{2}$$

where $k \ 1 \quad _{s}$, $u \quad 0.5\ell \ ^{1} 1 + \quad _{s} \ ^{1}$, $z_{0} \quad 2/3\ell$, $_{s} \quad \cos$, and $\quad k \sin$ with $\ell \quad \ell_{\perp} \sin\varphi + \ell_{\parallel} \cos\varphi$ and k the wave vector of the light in vacuum. Here is the scattering angle, φ is the angle of the nematic director $\mathbf{n}$ with respect to the scattering plane, and $\ell_{\parallel}$ and $\ell_{\perp}$ are the transport mean free paths parallel and perpendicular, respectively, to $\mathbf{n}$. The nematic director lies in the sample plane. Absorption by 5CB is negligible at our wavelength. Equation (2) describes an anisotropic backscattering cone of which the full width half-maximum W in the two orthogonal scattering planes is a measure of $\ell_{\parallel}$ and $\ell_{\perp}$:

$$W \varphi \quad 0 \quad \frac{0.7}{2\pi\ell_{\parallel}}, \qquad W\left(\varphi \quad \frac{\pi}{2}\right) \quad \frac{0.7}{2\pi\ell_{\perp}}. \tag{3}$$

In Fig. 3 we report a comparison between the data and the theoretical curves, plotted in a polar graph. The only fitting parameters are the enhancement factor E and the values of $\ell_{\parallel}$ and $\ell_{\perp}$. From the top panel (nematic director in x, extraordinary polarization) we find mean free paths $\ell_{\parallel} \quad 0.97$ mm and $\ell_{\perp} \quad 0.83$ mm, with a relative error of about 3% and an absolute error of 10%. To our knowledge, these are the first experimental data on the absolute values of the transport mean free paths in nematic liquid crystals. The resulting anisotropy is 1.17 0.05. For the bottom panel (director in y, ordinary polarization) we find $\ell_{\parallel} \quad 0.89$ mm, $\ell_{\perp} \quad 0.77$ mm, and $\ell_{\parallel}/\ell_{\perp} \quad 1.15$ 0.05. For the isotropic case in the middle panel, the fit gives $\ell_{\parallel}/\ell_{\perp} \quad 1.01 \quad 0.04$. These anisotropy values are in good agreement with available theories regarding anisotropic diffusion in nematics [14] that predict a mean free path anisotropy of about 1.18. For future studies it would be interesting to examine the magnetic field dependence of the mean free paths and anisotropy. The mean free paths are expected to get longer with increasing magnetic field due to a reduction of the magnetic coherence length, and the anisotropy is expected to become smaller.

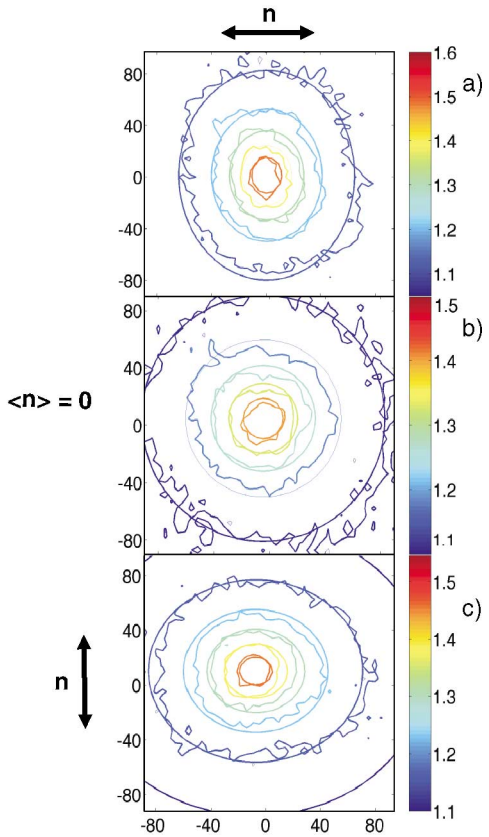

FIG. 3 (color). Comparison between data and theoretical curves to determine the anisotropy in the mean free path.

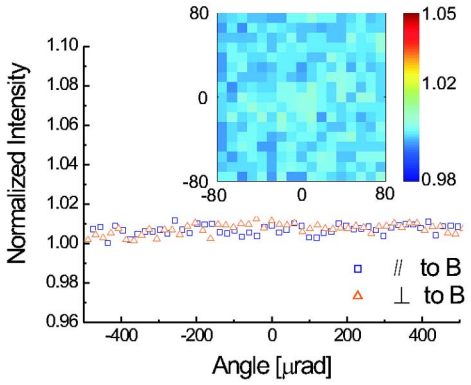

FIG. 4 (color online). Backscattered intensity in the crossed polarization channel. No enhancement is observed within the noise level of our data (1%).

Our accurate apparatus also enabled us to resolve a discrepancy in the literature about multiple scattering from liquid crystalline nematics. The liquid crystal birefringence leads to a substantial phase difference between the ordinary and extraordinary modes of propagation (much more than 2π over the length scale of the mean free path) and hence there should be no backscattering enhancement in the polarization reversing channel [11,16]. While the experiment by Vithana *et al.* [16] observed no cone in this channel, the experiment by Kuzmin *et al.* [11] revealed the presence of a 3%–5% backscattering enhancement for reversing polarization. In Fig. 4 we show the backscattered signals in the polarization reversing channel from the monodomain nematic phase. The setup was aligned in the polarization conserving channel and great care was taken to maintain optical alignment while changing polarization channels. We confirmed there was no residual backscattering cone in the polarization reversing channels (ordinary-extraordinary and extraordinary-ordinary) within the noise level of our experiment of 1%.

In summary, we have investigated weak localization in anisotropic systems, in particular, nematic liquid crystals. We observed an angular anisotropy associated with an anisotropic transport mean free path in a high-order multiple-scattering interference phenomenon. Interesting future studies could involve strongly scattering anisotropic systems, for example, anisotropic mesoporous semiconductors. It is as yet completely unknown how anisotropy influences the Anderson localization of light.

We thank B. van Tiggelen, R. Righini, M. Colocci, A. Lagendijk, and T. Lubensky for discussions and acknowledge support from the NSF (Grants No. DMR 0203378 and No. MRSEC 00-79909), the INFM (projects RANDS and Photonic), MIUR–Cofin 2002, and the ICTP (TRIL Grant SM).

*Electronic address: wiersma@lens.unifi.it
†URL: www.complexphotonics.org

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