Coherent Backscattering of Light in a Nematic Liquid Crystal

H. K. M. Vithana, L. Asfaw, and D. L. Johnson
Physics Department, Kent State University, Kent, Ohio 44242
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The first observation of coherent backscattering (CB) in an optically anisotropic, homogeneous (liquid crystal) medium with long-range dielectric tensor fluctuations is reported. The results are compared with previous CB experiments in heterogeneous, centrosymmetric, isotropic scattering media and with theoretical predictions for isotropic, noncentrosymmetric media. Fundamental differences and analogies between these cases are observed and explained. Other liquid crystal phases of potential interest in the localization of electromagnetic modes are introduced briefly.

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There has been intense interest for nearly a decade in the survival of phase coherence in classical waves multiply scattered by disordered media [1]. Of particular interest is the prospect that strong enough random scattering may quench wave propagation and lead to localized normal modes in some range of frequencies. This phenomenon, called localization, was first studied by Anderson [2] who predicted that in disordered electronic media, electron Schrödinger waves may become localized in the presence of strong enough scattering potentials, leading to an insulating state of normally conducting materials. The electromagnetic analog of the Anderson insulating state is a disordered dielectric medium in which electromagnetic modes are localized [1].

Whereas the conditions for full localization are quite subtle and stringent [1] a precursor phenomenon known as weak localization, a manifestation of which is enhanced backscattering of coherent electromagnetic waves [3,4], has been observed in heterogeneous, isotropic, centrosymmetric, disordered, dielectric media [5–9], such as suspensions of polymer spheres in water or silica particles in air, and also in more exotic systems where scattering is quasi two dimensional [10]. Specifically, a cone of enhanced backscattering of angular width $\Delta\theta \sim \lambda/\langle l \rangle$ is observed, where $\langle l \rangle$ is the mean free path and λ the wavelength of a photon in the scattering medium [1]. This phenomenon is called coherent backscattering and is maximized for scattered light of the same circular polarization (helicity) as the incident beam [8–11]. In this helicity preserving channel the enhancement ratio of coherent backscattered light ($\theta = 0$) to diffuse scattering outside the cone ($\theta \gg \Delta\theta$) is 2:1 because in the exact backscattering direction the optical path length and scattering probability for time reversed multiple scattering paths are precisely equal; the time of flight in the scattering medium, even for the longest paths, is small compared with the relaxation times of the dielectric fluctuations in most realistic situations. Backscattered light of opposite helicity to the incident beam loses some coherence because, although the time reversed optical paths are the same, the scattering probability of the first and last scattering events differ. In this case $1 < r \equiv I(\theta = 0)/I(\theta \gg \Delta\theta) < 2$ is observed experimentally [5–9], consistent with theoretical predictions [4,11].

The purpose of this Letter is to report the first observations of coherent backscattering (CB) in a class of materials new to this area of physics, namely, liquid crystals. Specifically, CB in nematic liquid crystals was observed and the phenomenology was found to be somewhat different from that of previously studied systems. This difference is shown to result from the intrinsic optical anisotropy of liquid crystals and soft Goldstone modes [12] which profusely scatter light.

Figure 1 shows the geometry of the CB experiment. Unlike the case of isotropic media it is not convenient here to use circularly polarized light because the optical normal modes of a uniaxially anisotropic optical medium, such as a nematic liquid crystal, are linearly polarized as either ordinary ($\hat{\mathbf{p}} \perp$ optic axis) or extraordinary ($\hat{\mathbf{p}} \parallel$ principal plane, plane containing propagation vector, $\mathbf{k}$, and

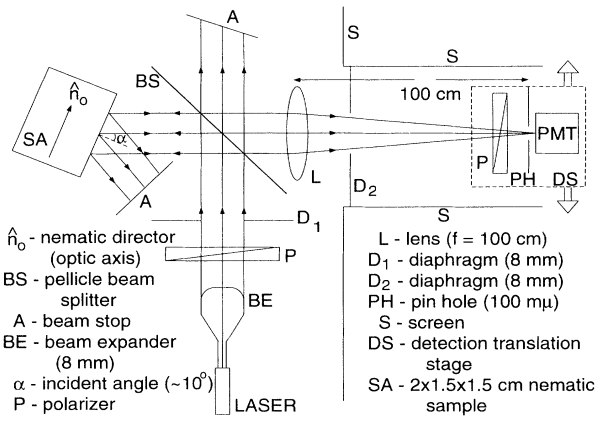

FIG. 1. Scattering geometry for CB experiments on liquid crystals. The angular resolution, Δ_R, is the pinhole diameter (100 μm) divided by the focal length of the lens (100 cm); therefore, $\Delta\theta = 10^{-4}$ rad which is less than but comparable with the width of the CB peak ($\sim 5 \times 10^{-4}$ rad). The 4 mW He-Ne laser, L, gave CB count rates of $\sim$1000 counts/sec (see Fig. 2). BS is a pellicle beam splitter that reflected (or passed) approximately one-half the light.

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optic axis) waves. Therefore, linearly polarized light in a geometry that couples to the sample as either a pure ordinary (O) or extraordinary (E) mode was used and both the polarized (EE and OO) and depolarized (EO) channels were studied. Note that because of the use of linearly polarized incident light the presence of single backscattering events cannot be avoided, as they can in the isotropic case, by imposition of a $\lambda/4$ plate in front of the sample [8]. Consequently the ideal backscattering ratio cannot always be expected to be 2:1 as it would be for the helicity preserving channel in centrosymmetric isotropic media; see, however, below.

To achieve single mode coupling to the sample it was necessary to align the optic axis (nematic director) at the surface so that, depending on its polarization, the incident beam can excite either an O or an E mode. This was accomplished by unidirectional rubbing of a thin polyimide layer deposited on the glass substrate bounding the incident and opposite faces of the sample. A weak magnetic field was also used to align the average bulk director without significantly suppressing fluctuations [12] or destroying time reversal invariance [11]. Clearly there are many scattering geometries which are degenerate for isotropic systems but not for nematics. Only two are required to reveal one of the fundamental differences between these two classes of scattering media. A second fundamental difference, which cannot be resolved by these data, is that dielectric fluctuations fall off algebraically (R^{-1}) in the nematic [12] but exponentially in isotropic systems studied thus far.

Results for two geometries are presented in Fig. 2. In Fig. 2(a) the incident and backscattered light are both extraordinary modes (EE) while in Fig. 2(b) the incident beam is extraordinary and the backscattered light is ordinary (EO). The EE data are qualitatively similar to helicity preserving circularly polarized CB data [8] from isotropic, centrosymmetric media. The angular width of the CB peak ($\Delta\theta \sim 5\times10^{-4}$ rad) would imply a mean free path of the order of a millimeter or so ($\langle l\rangle \sim \lambda/\Delta\theta$). Although the mean free path in a liquid crystal is anisotropic, turbidity measurements on thick films [13] suggest that an average value of this order is expected in a typical nematic. So Fig. 2(a) is more or less as naively expected, albeit we do not know what the ratio r should be on theoretical grounds because of the possibility of single backscattering events (see below).

In addition to the two geometries of Fig. 2, data were collected on three other geometries. With reference to Fig. 1 as a top view, these geometries are referred to as OOV, EEV, and OOH, where V and H refer to the director (optic axis) orientation as vertical or horizontal, respectively. In this notation the geometries associated with the data in Figs. 2(a) and 2(b), respectively, are EEH and EOH. In all four of the polarization preserving geometries (EE or OO) the incoherent background scattering was essentially the same, 1080 ± 50 counts/sec, whereas the EOH geometry background was 640 ± 20 counts/sec. This difference can be explained by invoking two assumptions: (1) Single backscattering from bulk nematic director fluctuations, which is allowed in the EEH and EOH but not the other three geometries [12], is negligible, and (2) there exist polarized single backscattering events which contribute approximately 440 counts/sec (1080 − 640 counts/sec) in the EE and OO geometries and which arise from sources other than director fluctuations, such as cell interfaces or dust particles in the sample. The nematic sample was used as purchased and was composed of the British Drug House nematic mixture E48, a solution of several compounds. Single backscattering events due to director fluctuations are expected to be rare because of the inverse square law dependence of the nematic scattering cross section on the transferred wave vector [12]. An alternative to the above interpretation is that there is less phase space available for depolarized than for polarized incoherent backscattering, whereas all EE and OO geometries have equal phase space for incoherent backscattering. The latter makes the former seem highly unlikely and the theory of nematic light scattering [12] has no suggestion of such behavior. There have been no multiple scattering calculations done based on nematic light scattering theory. Photon diffusion in a nematic is probably only moderately anisotropic [13].

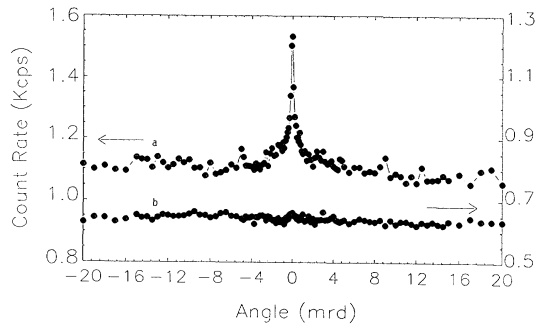

FIG. 2. Results of CB experiments in the (a) EEH and (b) EOH geometries (see text).

With the above assumptions the ratios of peak-to-background scattering after subtracting 440 counts/sec from all of the EE and OO data are $r(EEH) = 1.68$, $r(EOH) = 1.00 \pm 0.05$, $r(OOV) = 1.64$, $r(EEV) = 1.38$, and $r(OOH) = 1.50$. Except for the variations in their ratios all four of the EE and OO geometries have backscattering profiles identical, within experimental uncertainty, to that of Fig. 2(a). The observed variations in r are a result of a few points near the peaks. Our resolution is not good enough to resolve these extremely narrow peaks. The r values should not be taken as more than best estimates, for the given resolution, which attempt to account for single backscattering and spurious backscattering. It is clear, however, from the sharpness of the CB peaks, that this experiment is in the so-called meso-

scopic regime [14], i.e., $\lambda \ll \langle l \rangle \ll L \ll L_i$ where $L \sim 1$ cm is the sample size, $\langle l \rangle \sim 1$ mm is the photon mean free path, and L_i is the inelastic scattering or absorption length. If $L \ll L_i$ were not true the CB peaks would be severely truncated [8]. This is not entirely surprising because the high temperature isotropic phase of E48 is transparent and colorless and the relevant nematic normal mode relaxation times are relatively long, $\tau(q) = \eta/Kq^2 > \sim 10^{-5}$ sec, where η and K are the viscosity and the director curvature elasticity coefficients, respectively [12]. Furthermore, fast relaxations correspond to large q director modes which decrease in thermal amplitude as q^{-2} so the dominate scattering modes are much slower than 10^{-5} sec.

The experimental results for *EO* scattering (*EOH*) are quite different as shown in Fig. 2(b). Here, unlike the analogous isotropic case (*RL* or *LR* scattering), which exhibits a reduced but substantial CB peak, *no residual coherence in the backscattered light is found* [$r(EOH) = 1.00 \pm 0.05$]. To understand this result requires a direct comparison with CB in isotropic, centrosymmetric, and noncentrosymmetric media.

For centrosymmetric, isotropic scattering media the various legs of the time reversed multiple scattering paths are labeled *R* or *L* for right or left circular polarization in Fig. 3(a), which illustrates a *helicity reversing* multiple scattering channel. Consideration of an appropriately modified scattering sequence reveals that *helicity preserving* channels (*RR* and *LL*) have identical optical paths and probabilities for time reversed scattering and thus $r = 2$ [11]. The helicity reversed channels (*RL* and *LR*) also have identical time reversed optical paths because the indices of refraction of *R* and *L* polarized light are equal in centrosymmetric media; however, the probability of the time reversed paths, while the same for the interior part of a scattering loop (that is, after the first and before the last scattering event), differ at the first and last scattering events as is clear from consideration of Fig. 3(a). This results in a partial destruction of coherence [11].

In noncentrosymmetric (non-parity-conserving) media, coherence is in double jeopardy for helicity reversed scattering because not only are the probabilities of time reversed channels unequal but the optical paths are also unequal, even at $\theta = 0$. Specifically, the optical path difference is $|n_R - n_L|\langle l \rangle$ where $n_{R,L}$ are the indices of refraction for *R* and *L* polarized light and $\langle l \rangle$ is the average difference in geometric path. l is defined in Fig. 3 and is a stochastic variable; hence, if $\langle l \rangle \gg \lambda/|n_R - n_L|$ one can expect only incoherent backscattering. This CB problem has been treated theoretically [11] but not experimentally. In Fig. 3(b) *R* and *L* of Fig. 3(a) are replaced by *E* and *O*, as appropriate for a nematic scattering medium. Figure 3(b) illustrates a *depolarizing* multiple scattering channel. Consideration of an appropriately modified scattering sequence reveals that *polarization preserving* channels (*EE* and *OO*) have identical optical paths and

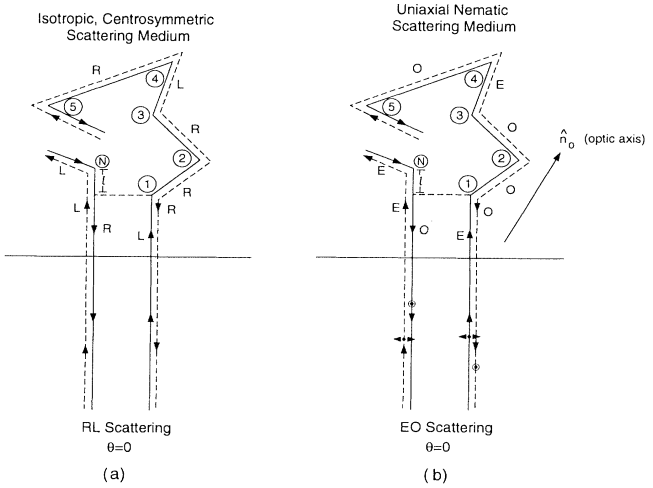

FIG. 3. Schematic of time reversed multiple scattering paths for (a) helicity reversing and (b) polarization reversing channels in scattering media as indicated in the figure. The dashed and solid lines are pairs of time reversed scattering paths. *N* represents the last scattering event; events 6 through $N-1$ are not shown. The polarization states are the same for both paths except for the incoming and outgoing legs. ⊙ and ←●→ signify polarization in (b).

probabilities for time reversed scattering; thus $r = 2$, analogous to the helicity preserving case, if single backscattering events are ignored (see below). The major qualitative difference between these experiments on nematics and previous experiments on centrosymmetric isotropic media can be seen by comparing the results of the *EO* geometry [Fig. 2(b)] and analogous *RL* or *LR* geometries. From Fig. 2(b), $r(EOH) = 1.00 \pm 0.05$, i.e., a complete loss of coherence was found, whereas $r_{LR} \sim 1.3$ in the analogous centrosymmetric, isotropic case [8]. As pointed out above $1 < r_{RL} = r_{LR} < 2$ because the asymmetry of the scattering events only partially destroys coherence. An analogous asymmetry exists for *EO* (or *OE*) scattering in nematics as a glance at Fig. 3(b) indicates. Here, however, not only are the time reversed scattering probabilities different but the optical path for time reversed scattering differs by an amount $(n_e - n_o)l$, where n_e and n_o are the extraordinary and ordinary indices of refraction and l is the Euclidian length shown in Fig. 3. This is exactly analogous to the noncentrosymmetric, helicity reversing case discussed above. Again l is a stochastic variable; specifically, $\langle l \rangle$ is the mean free path of the incident light and is known in this case to be much larger than a wavelength [13]. Hence the phase of time reversed *EO* or *OE* backscattered light is completely diffused resulting in $r_{EO} = r_{OE} = 1$ as observed. This result is an extreme example (by analogy) of the predicted optical path length related reduction in coherence of the helicity reversed channel in parity nonconserving (noncentrosymmetric) media [11]. It is the first experimental confirmation of the underlying physics of that prediction,

namely that $n_R \neq n_L$ leads to incoherent phases of time reversed channels. Here the condition $n_e \neq n_o$ has the same effect, and since $\langle l \rangle (n_e - n_o) \gg \lambda$ phase coherence is completely destroyed and $r_{EO} = 1$.

In conclusion, we note that there are a variety of liquid crystal phases that are interesting in the context of multiple scattering phenomena. Anisotropic and chiral twisted phases such as the cholesteric and blue phases or the chiral smectic-C phase come to mind as particularly interesting [12]. Unfortunately the refractive index variations are not large ($n_e - n_o \sim 0.1$–0.3) so strong localization may not be feasible. However, there is a one-dimensional photonic band gap for light of appropriate helicity and wavelength range in a cholesteric which would be interesting in this regard. We have observed helicity conserving CB in a disordered (disclination filled) cholesteric for a wavelength outside the gap; $\Delta\theta$ was larger than for the nematic but strong speckles were present due presumably to slow time averaging. Diffusive wave spectroscopy experiments in liquid crystals should also prove interesting [15]. Finally, we note that heterogeneous structures involving liquid crystals, such as polymer dispersed liquid crystals [16], or liquid crystals in porous media, are excellent candidates for a variety of interesting multiple scattering experiments.

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