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Light Transport in Opaque Liquid Crystal Structures

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An overview is given of recent work on multiple light scattering in opaque liquid phases and liquid crystal infiltrated macro porous structures. The nematic phase is relatively strongly scattering and for large enough samples permits to observe anisotropic light diffusion. Liquid crystal infiltrated macro porous glasses allow for temperature control over the light diffusion constant and, if infiltrated also with laser dye, for realizing temperature-tunable random laser action.

Keywords: light diffusion; multiple scattering; random laser; liquid crystals; dye

INTRODUCTION

Light scattering in liquid crystals is a widely studied topic. Mostly, however, experiments are limited to the single scattering regime: the incident light beam is scattered once by the liquid crystal. The nematic phase is quite opaque however, and for large enough samples this allows to enter the regime of multiple scattering in which the transport of the light through the liquid crystal can be described (in first approximation) as a diffusion process.

The study of multiple light scattering has become a very active field in physics in the last decade when it was found that various interesting interference effects can occur when light is multiply scattered by a disordered

dielectric structure[1]. An example of such an interference phenomenon is coherent backscattering or weak localization[2], which leads to an enhancement of the intensity in and closely around exact backscattering, due to interference between counter propagating light waves. Several of these interference effects that occur for light transport through disordered dielectric materials have there counterpart in electron transport. For instance, the Hall effect and magneto resistance have been observed for optical waves[3], and also other electron transport phenomena like Anderson localization (metal-insulator transitions) and universal conductance fluctuations have been observed for light waves[4, 5]. Important applications of multiple light scattering include medical imaging[6] and diffusing-wave spectroscopy (DWS)[7]. All these studies were concerned with isotropic random media.

The study of multiple light scattering in liquid crystal systems is interesting for various reasons. The partial ordering of the nematic phase leads to an anisotropic scattering function, which makes nematic liquid crystals fundamentally different from common random media. This anisotropy in the scattering cross section leads, for large enough samples, to an anisotropic diffusion process, and monodomain nematics are therefore ideal systems to study anisotropic multiple light scattering. Anisotropic light diffusion has recently been observed in cw experiments by Kao *et al.*[8] and later by us in time-resolved experiments[9]. Also coherent backscattering has been observed from large nematic systems[10], although the expected anisotropy in the backscattering cone has not been observed yet due to limited angular resolution in the experiments. A wide scale of inspiring theoretical work is available on multiple light scattering in liquid crystals[11], mostly developed by van Tiggelen and Stark.

Liquid crystal infiltrated macro porous glasses are even more strongly scattering than a pure liquid crystal sample in the nematic phase. At high temperatures the nematic phase goes over into the isotropic phase, which behaves as a normal liquid without birefringence. The refractive index of a liquid crystal is therefore strongly temperature dependent. This creates

interesting possibilities for manipulating the scattering properties of disordered materials after liquid crystal infiltration, either by changing the temperature or by applying an external field to align the nematic director. This feature is apparent, for instance, in polymer dispersed liquid crystal films that can be switched from opaque to transparent by an external field or by temperature[12]. Liquid crystals in confined geometries like porous glasses have been studied extensively in different contexts, using various experimental techniques like high-resolution ac calorimetry, NMR, magnetically induced birefringence, dielectric spectroscopy, and X-ray scattering[12, 13]. Light scattering from liquid crystals in confined geometries was studied, for instance, by Bellini et al[14], who measured, among other optical properties, the extinction of a laser beam through infiltrated silica aerogel at various temperatures. They observed a large increase of opacity below the nematic-isotropic transition temperature. In recent time-resolved experiments it was shown how liquid crystal infiltrated macro porous glasses can be used to control the diffusion constant for light waves via temperature[15]. The possibility to tune the scattering strength of a disordered sample (expressed in terms of the diffusion constant) has interesting applications for several multiple light scattering studies. One can use this principle to create a random laser source that can be brought above and below threshold via temperature[16]. In this article I will give an overview of various aspects of multiple light scattering in liquid crystal materials studied up to date.

ANISOTROPIC LIGHT DIFFUSION

The nematic phase of a liquid crystal is characterized by a global alignment of the molecules in a direction called the nematic director $\mathbf{n}(\mathbf{r})$, and an otherwise translational disorder. The strong opacity of the nematic phase comes about from local fluctuations in the nematic director[17] that elastically scatter light. Due to the partial order of the nematic phase, the

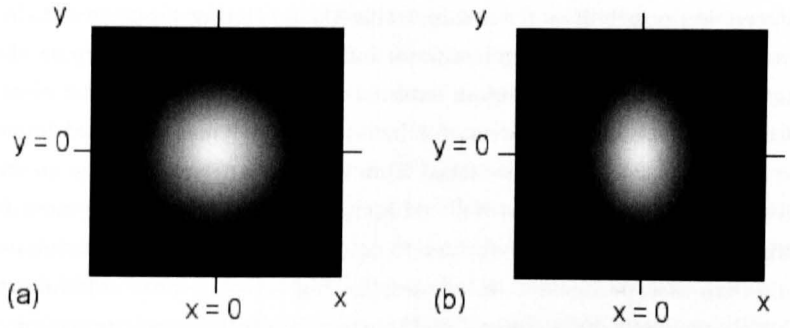

FIGURE 1 Calculated intensity distribution of the diffuse light on the output surface of an optically thick isotropic random sample (a) and of a liquid crystal in the nematic phase (b). In the latter case a magnetic field is applied in the y-direction to obtain a monodomain nematic phase. The anisotropic multiple scattering process becomes apparent as an anisotropy in the intensity distribution.

scattering cross section σ of these director fluctuations will be anisotropic and will depend on the propagation direction and polarization of the light relative to the nematic director[18]. A light wave propagating through a large and monodomain nematic phase will therefore perform an anisotropic random walk, of which both the average step length (called the transport mean free path ℓ) and velocity of propagation will be anisotropic. The transport mean free path ℓ will have the values $\ell_\perp$ and $\ell_\parallel$ respectively perpendicular and parallel to the nematic director.

The most direct manifestation of the anisotropy in multiple scattering of light through a monodomain liquid crystal can be seen in transmission. Fig. 1 shows the intensity distribution on the backside of a regular isotropic random sample and a nematic liquid crystal, illuminated from the front by a narrow laser beam. As a direct consequence of the anisotropy in the

transport mean free path, the random walk that the light performs in the nematic sample will make larger steps in the direction along the nematic director. This results in an oval shaped intensity distribution. This is the way that Kao et al.[8] observed anisotropic light diffusion in liquid crystals.

For isotropic media one can often use the diffusion approximation, which means that one describes the transport of the energy density of the light by a common diffusion equation with diffusion coefficient D. The diffusion constant can be related to the transport mean free path and transport velocity via: $D = 1/3v\ell$. For an anisotropic medium the diffusion equation reads:

$$\frac{\partial W(\mathbf{r},t)}{\partial t} = \nabla \cdot \mathbf{D}\nabla W(\mathbf{r},t) + S(\mathbf{r},t), \tag{1}$$

with $W(\mathbf{r},t)$ the energy density and $S(\mathbf{r},t)$ a source function. Choosing the nematic director along one of the coordinate axis, the distinct elements of the diffusion tensor will be $D_\perp$ and $D_\parallel$.

The anisotropy in the diffusion constant can be observed if one illuminates the sample with an ultra-short light pulse and monitor the temporal response of the diffuse transmission, either in the direction parallel or perpendicular to the nematic director. Fig. 2 shows the results of a time-resolved transmission experiment on the liquid crystal *p*-penthyl-*p*'-cyanobiphenyl (5CB) for planar and homeotropic alignment of the nematic director. The sample was oriented in the x-y plane and the laser pulse incident along z. The laser beam was narrow (1mm) and the transmitted diffuse light through the slab was recorded around $x = y = 0$. One can measure $D_\perp$ and $D_\parallel$ by orienting the nematic director either parallel (planar alignment) or perpendicular (homeotropic alignment) to the plane of the slab, using an external magnetic field (of strength B).

The solid line is the theoretical curve as obtained by solving Eq. (1), using the boundary conditions of a slab geometry: $W(\mathbf{r},t) = 0$ at $z = -z_0$ and $z = L + z_0$, with L the physical thickness of the slab, and using the source function $S(\mathbf{r},t) = \delta(x)\delta(y)\delta(z-\ell_z)\delta(t)$. The distance z_0 is called the extrapolation length and depends on the refractive index mismatch

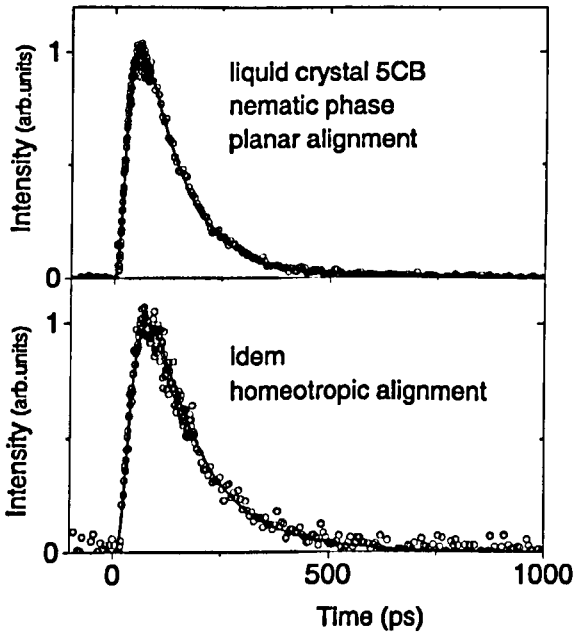

FIGURE 2 Time evolution of the transmission of a short probe pulse through liquid crystal 5CB in the nematic phase, in a planar alignment (upper) and a homeotropic alignment (lower). (T = 300 K, B = 0.5 T.) The solid lines are the theoretical curves as calculated from Eq. (2). The planar and homeotropic alignments of the nematic director allow to measure respectively $D_\perp$ and $D_\parallel$, demonstrating the anisotropy in the diffusion constant for light transport through an optically thick monodomain nematic. [From Wiersma et al, Phys. Rev. Lett. 83, 4321 (1999).]

between sample and surrounding medium[19]. We assume that the incident pulse is fully scattered at a depth ℓ_z and, for symmetry reasons, that the last scattering event takes place at $z = L - \ell_z$. The time evolution of the transmitted intensity is given by Fick's law ($I_{\rm tr} = -D_{zz}\nabla W(\mathbf{r},t)\mid_{z=L-\ell_z}$) and reads after solving Eq. (1):

$$I_{\rm tr} = \frac{I_0 \exp(-\Delta x^2/4D_{xx}t)\exp(-\Delta y^2/4D_{yy}t)}{\pi^{3/2}(4t)^{5/2}\sqrt{D_{xx}D_{yy}D_{zz}}} \times \sum_{n=-\infty}^{+\infty} A\exp(-A^2/4D_{zz}t) - B\exp(-B^2/4D_{zz}t), \quad (2)$$

with $A = (1-2n)(L+2z_0) - 2(z_0+\ell_z)$ and $B = (2n+1)(L+2z_0)$, and where Δx and Δy denote the shift of the incoming beam in x and y respectively. (Here $\Delta x = \Delta y = 0$.) Note that in the limit of long t, the transmitted intensity falls off as an exponential with time constant $\tau = (L+2z_0)^2/D_{zz}\pi^2$. For the planar alignment one finds: $D_{zz} = D_\perp = 3.62 \cdot 10^4$ m^2/s and for the homeotropic alignment: $D_{zz} = D_\parallel = 4.56 \cdot 10^4$ m^2/s, which leads to an anisotropy in the diffusion constant of: $D_\parallel/D_\perp = 1.26$.

The anisotropy in the multiple scattering of light through a liquid crystal has a third consequence. Not only the spatial distribution of the light intensity on the backside of a sample will be anisotropic, also the angular distribution of the transmitted diffuse light will show an anisotropy. This anisotropy is still present after summing over all scattering angles in transmission. This means that the nematic phase behaves as a polarizer even for the diffuse transmission[20]. We made an attempt to observe this effect by measuring the total transmission through a monodomain nematic sample of 10 mm thickness and 37 mm diameter (disk shaped slab geometry), by illuminating the sample on its front interface with a narrow laser beam and collecting all the transmitted light with an integrating sphere. A polarizer was attached directly on the output surface of the sample. The input laser was vertically polarized and the nematic director was aligned vertically by an external magnetic field. We observed no difference in total transmission when rotating the polarizer on the output

surface of the sample, within our experimental error of 1%. This means that the predicted anisotropy disappears with the above configuration of the polarizer. The correct way to observe the effect would most likely be to measure at specific scattering angles and sum them individually, keeping the polarizer always perpendicular to the outgoing wavevector. This, however, was not possible in our experimental geometry.

INFILTRATED POROUS GLASS

In most disordered systems the opacity is a fixed number for a given sample. The opacity depends on the size and shape of the scattering elements and on their refractive index contrast and can therefore usually not be changed. However, by using porous glasses with liquid crystal infiltration we can obtain external control over the diffusion constant[15]. This method is based on an idea by Busch and John to infiltrate inverse Opal structures in order to obtain a tunable photonic crystal[21]. In that case an ordered dielectric structure is infiltrated with a medium (the liquid crystal) that has a refractive index which depends strongly on temperature and under certain conditions on an external field. The same principle can be applied to porous disordered structures, in which case the scattering strength of the single scattering elements, and thereby also the opacity of the whole system, will depend on temperature and/or on an external field.

An example of such a tunable disordered system is macro porous silica glass which has randomly oriented and interconnected pores, infiltrated with, for instance, the liquid crystal Octylcyanobiphenyl (8CB). Bulk 8CB has a smectic-A phase in the temperature range of 21.1-33.5°C, whereas a nematic phase is formed in the range of 33.5-40.8°C. In the nematic phase, 8CB is birefringent with refractive indices $n_o = 1.52$ and $n_e = 1.65$ at 36.2°C and 613 nm wavelength. The diffusion constant of this material can be characterized by time-resolved experiments as described before. In

Fig. 3 we see the measured diffusion constant for a wide range of temperatures. The light source was a cavity dumped, mode-locked dye laser operating at 613 nm wavelength and the time-resolved transmission was recorded with a Hamamatsu streak camera. Arrows indicate the phase transition temperatures of non-confined bulk 8CB. We can clearly see a strong dependence of the diffusion coefficient on temperature, especially in the temperature range of the nematic and isotropic phases. In the nematic phase the liquid crystal is birefringent, with ordinary and extraordinary refractive indices respectively n_o and n_e. Without an external field, the orientation of the nematic director will be random and different in every pore, so that a disordered birefringent medium is formed. The birefringence averages out on a macroscopic scale. In the isotropic phase also the local birefringence disappears, so that the refractive index contrast between liquid crystal and porous matrix changes considerably. This explains the strong decrease of D, when lowering the temperature into the nematic region.

The observed temperature dependence of the diffusion constant is very different from the typical phase behavior of bulk LC, due to the confinement in the porous silica. The porous silica host will force the liquid crystal molecules close to the pore walls in a certain direction. Due to this confinement both the smectic-A to nematic and the nematic to isotropic phase transitions are smeared out, which is in agreement with NMR, calorimetric, and spectroscopic studies of 8CB confined in random pores[23, 24, 25]. The smectic-A to nematic phase transition is so strongly smeared out that the diffusion constant increases smoothly with increasing temperature. Measuring the temperature dependence of the diffusion constant provides an alternative method to resolve phase transitions of liquid crystals in confined geometries. The advantage of measuring the diffusion constant compared to conventional transmission techniques becomes apparent at large optical thicknesses and very opaque samples where the coherent beam in transmission is below the detection limit and conventional optical experiments fail. For measurements of the diffusion constant, a

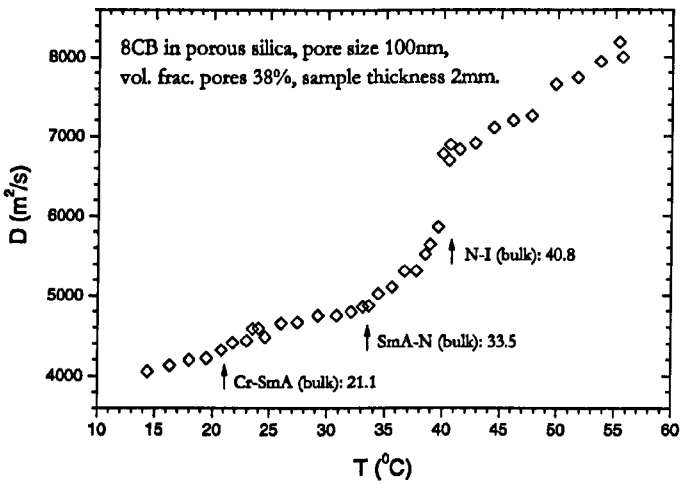

FIGURE 3 Temperature dependence of the diffusion constant of porous silica infiltrated with the liquid crystal 8CB. Pore size 100 nm, volume fraction of the pores 38 %, sample thickness 2 mm[22]. The arrows indicate the phase transition temperatures of a bulk (free) sample of 8CB. The phase sequence of 8CB is: crystalline (Cr) - Smectic A (SmA) - Nematic (N) - Isotropic (I). Note that even the nematic - isotropic phase transition is not sharp due to the geometrical confinement of the liquid crystal. We see that a considerable variation of the diffusion constant can be obtained by changing the temperature of the system. [From: Wiersma et al., Phys. Rev. B 64, 144208 (2001).]

large optical thickness of the sample is not a limiting factor but is even an advantage.

The strong dependence of the opacity on temperature is similar to a temperature dependent resistivity as often encountered in electronics[26]. The opacity decreases upon increasing temperature, so this system can be seen as the optical equivalent of a negative temperature coefficient (NTC) resistor. Having external control over the diffusion constant has important application in several multiple light scattering studies.

LIQUID CRYSTAL RANDOM LASER

So far we have discussed multiple light scattering in passive systems which scatter light without absorbing or amplifying it. Of particular interest are disordered materials in which multiple scattering is combined with optical amplification. This can be achieved by grinding an ordinary laser crystal like Titanium sapphire, or introducing scattering elements like microspheres or glass powder in a laser dye solution. If the laser crystal or dye is excited by an external laser source it will provide optical amplification inside the random system. A light beam at the appropriate wavelength will be both multiply scattered inside such medium and amplified. The overall gain depends on the volume of an amplifying random sample while the total losses depend on its surface. That means that there exists a critical volume above which the overall gain becomes larger than the total losses and the intensity diverges. This is a situation similar to laser action without an external cavity. The trapping mechanism that keeps the light waves long enough inside the system for the gain to become efficient is multiple scattering[27]. A disordered material with gain that is brought above threshold is now often referred to as a random laser[28].

The careful balance between gain and loss in a random laser can be expressed as a comparison between the actual excited volume of the sample and a critical volume V_{cr} or, in case of a slab geometry, as the comparison

between the actual thickness and a critical thickness L_{cr} defined as:

$$L_{cr} = \pi \ell_{amp}, \tag{3}$$

where ℓ_{amp} is the amplification length defined as the average distance (as measured in a straight line) between the start and end point of a random walk with amplification factor e^{+1}. The amplification length is given by

$$\ell_{amp} = \sqrt{\frac{\ell \ell_g}{3}}, \tag{4}$$

where ℓ_g is the gain length defined as $\ell_g = 1/\kappa_g$ with κ_g the gain coefficient inside the material, and ℓ the transport mean free path as defined before. When the critical thickness of the sample becomes smaller than its actual thickness, the overall gain becomes larger than the total losses and the system is above threshold. The emission properties of a random laser are somewhere in between those of a common light bulb and a regular laser. Random laser emission is reasonably narrow banded and can be pulsed. On the other hand, emission will occur in several random directions determined by the speckle pattern of the random scattering.

For a random laser, having control over the diffusion constant has important consequences. The lasing threshold depends on the diffusion constant D of the random material because ℓ in Eq.[4] is proportional to D. (See text above Eq.[1].) This means that if we have a temperature-dependent diffusion constant, we are able to bring the random laser above and below threshold by changing its temperature. In addition, one could temperature-control the bandwidth of the random laser emission this way. A temperature dependent diffusion constant is precisely what can be achieved with a liquid crystal infiltrated porous glass, as explained before. If we add a tiny amount of laser dye to the liquid crystal we can introduce the optical gain required for random laser action.

Fig.4 shows the emission spectrum of a sample made by sintering SK11 glass powder (Schott) and infiltrating with liquid crystal 7CB + DCM laser dye (Lambdachrome 6500) at various temperatures. The sample was excited with a frequency-doubled Q-switched Nd:YAG laser, operating at

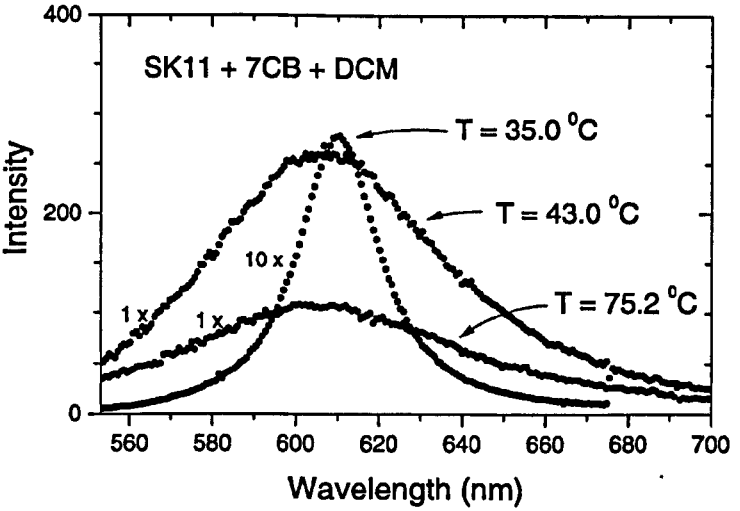

FIGURE 4 Emission spectra of sintered SK11 infiltrated with 7CB + DCM at various temperatures. Concentration of DCM in liquid crystal: 4.1 mmol/l, volume fraction liquid crystal in sintered glass 0.26. Sample thickness 1.3 mm, excitation beam diameter on sample 1.9 mm. Excitation pulse energy 8 mJ, pulse duration 14 ns. The spectrum at 35 °C has been scaled down by a factor of ten. By changing the temperature one observes a considerable difference in emission bandwidth. At 35 °C the system is above threshold which gives rise to a narrowing of the emission spectrum and a strong increase in intensity. [From: D.S. Wiersma and S. Cavalieri, Nature, to be published.]

10 Hz repetition rate. The emission spectrum was recorded by collimating the diffuse emission from either the front or the rear sample surface onto the input slit of a single grating spectrometer equipped with a cooled and gated optical multichannel analyzer to provide single shot spectra.

At 35 ^{0}C the sample is in the nematic phase while at 43 ^{0}C and 75.2 ^{0}C, the sample is in the isotropic phase. We see that indeed there is a strong effect of temperature on emission spectrum. In the nematic phase the diffusion constant becomes small enough (and therefore the scattering becomes strong enough) for the random laser to go above threshold. The system behaves as a light source of which the spectral width can be controlled via temperature. The fact that random laser sources can be made extremely small (tens of microns) and the possibility to work with different spectral tuning curves allows for interesting application as sources in active displays and temperature sensitive screens. The tunable random laser can be designed to have its threshold behavior at very specific temperatures, which opens up applications in remote temperature sensing especially in the temperature regime of biological processes.

CONCLUSIONS

We have discussed various aspects of multiple light scattering in liquid crystal systems. The nematic liquid crystal phase can give rise to anisotropic light diffusion if the sample size is large enough to allow multiple light scattering and the nematic phase is monodomain. Liquid crystals inside porous structures can scatter light even more strongly. The various liquid crystal phases with their different refractive indices can be used to obtain a material in which the light diffusion constant can be tuned externally via temperature. This opens up interesting possibilities for instance for a random laser source. The random laser can be externally controlled via temperature this way, creating a source of which the emission bandwidth is temperature dependent.

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