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Speckle observation of pulsed laser-induced dynamics in a guest–host smectic A liquid crystal system

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Using a pulsed, pump-probe experimental arrangement, we have investigated the speckle generated by a 100 μm path length cell of smectic A liquid crystal 4-cyano-4′-octylbiphenyl doped with 0·1 wt% dichroic dye. Upon irradiation, the guest–host system undergoes a phase transition from an initially transparent condition to a scattering state. We show that the statistical theory of speckle can be used to describe the evolution of scattering domains and estimate an rms refractive index variation of 0·0021 within the irradiated region for one case. Dynamic response as a function of molecular anchoring conditions was also investigated.

1. Introduction

Thermally induced scatter in smectic liquid crystals has been reported by several authors and has been utilized in laser-addressed liquid crystal displays, and other information storage media [1–8]. In these devices it is important that a stable scattering texture be realized. Recently, we described the use of dye-doped smectic A liquid crystal in a laser power limiting device and showed that the onset time for scattering and the permanence of the scattering state are both critically dependent upon irradiation conditions [9]. Fast response to a laser pulse threat and quick recovery is made possible in dye-doped smectic A liquid crystal by the existence of a transient scattering state. In order to tailor material properties to optimize device performance, however, it is important to understand the mechanism of the scattering process. In this paper, we describe the temporally resolved dynamics of laser-induced scattering in a guest-host smectic A liquid crystal. Reorientational effects in nematics have been successfully described using Kirchoff–Sommerfeld diffraction theory [10–12]. Here we show for the first time how the statistical theory of speckle can elucidate the nature of the scattering process.

2. Experiment

The mixture investigated consisted of the smectic A liquid crystal compound K-24 (4-cyano-4′-octylbiphenyl) and 0·1 wt% azo-type dichroic dye D2, both manufactured by Merck (Poole). This dye provides a strong absorption at 488 nm; the absorption coefficient of the guest-host mixture was measured as 57·6 cm^{-1} for the ordinary ray,

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whereas for undoped K-24 we measured 0·6 cm^{-1}. The ratio of dye absorbance in the homogeneous and homeotropic orientation was approximately 10 : 1.

The liquid crystal mixture was sandwiched between two quartz plates with a 100 μm fluid gap. In order to investigate dynamic response under different molecular anchoring conditions, each plate was partially coated with an organosilane film containing long (C_{18}) alkyl chains, as shown in figure 1. This material is known to impart strong, homeotropic anchoring [9], although the area of the cell with no coating also exhibited homeotropic orientation, since this is a minimum energy condition for a smectic A phase in this configuration [13, 14]. The high degree of uniaxial alignment in each region of the cell is depicted in the conoscopic isogyres of figure 2. Mylar strips of about 100 μm thickness were placed at the boundaries of the coated regions to maintain fluid segregation. The cell was translated via a micrometer screw orthogonal to the beam propagation direction, and the use of a single, segregated cell ensured the same focal conditions in each irradiated region. It will be shown that even though identical homeotropic alignment existed within each area of the cell, scattering dynamics were critically dependent upon anchoring strength.

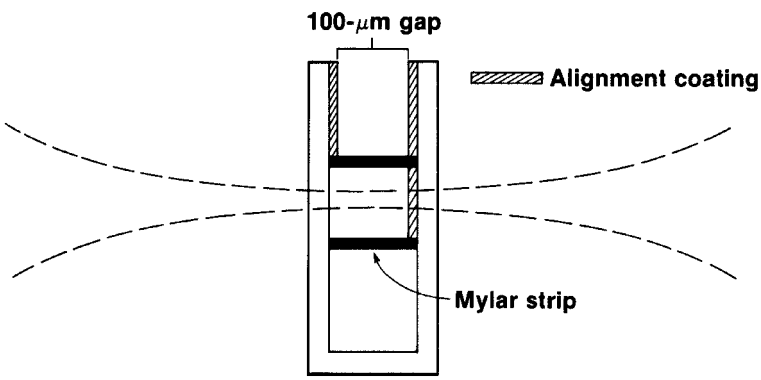
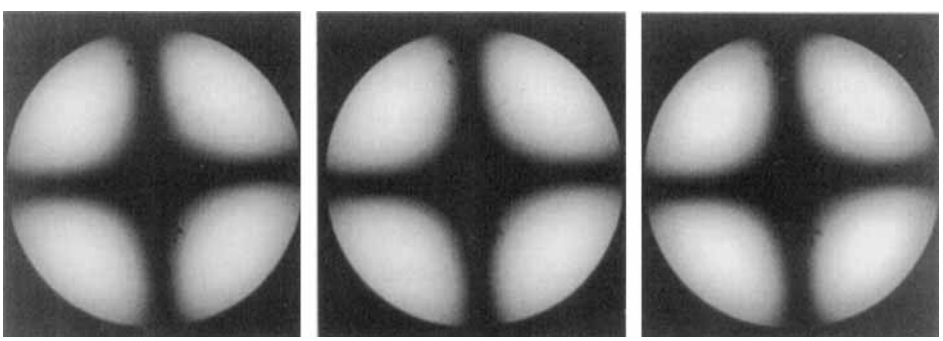


Figure 1. Cell geometry used to investigate guest–host system response to different molecular anchoring conditions. The use of a single, segregated cell ensured identical focal conditions in each region.



(*a*) (*b*) (*c*)

Figure 2. Conoscopic isogyres showing identical uniaxial alignment in each irradiated region; (*a*) alignment coating on both cell surfaces, (*b*) coating on one surface, (*c*) no alignment coating.

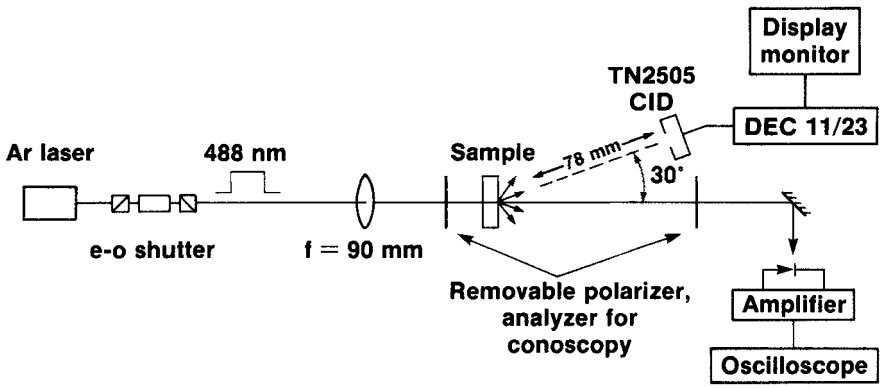
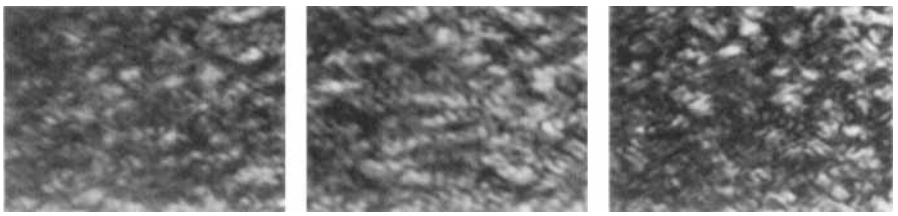


Figure 3. A pulsed argon laser was used as both pump and probe to characterize the spatio-temporal evolution of the scattered field.

The experimental set-up is shown in figure 3. An argon laser operating at $\lambda = 488$ nm and an electrooptic shutter provided a temporally square excitation pulse of 40 mW to the liquid crystal with a pulse width of 15 ms. The light was linearly polarized and focused to a $1/e^2$ spot diameter within the sample of 78 μm, as measured using a scanning slit (model 1080, Photon, Inc.). The output intensity of the argon laser was held constant by feedback control circuitry. The electrical bias of the shutter was adjusted to provide a small amount of light leakage (~ 3 mW), which was used to probe liquid crystal dynamics. This cw leakage beam was sufficiently reduced in intensity to eliminate potential alignment perturbations caused by residual heating, which was verified conoscopically before and after pump pulse irradiation. Upon removal of the pump, the liquid crystal was allowed to relax ballistically to the ordered state. The irradiation conditions used in this experiment were chosen to provide ~ 3 s for complete relaxation, during which a CID camera (model TN2505, CID Technologies) was used to take a snapshot of the scattered field of the probe at discrete time intervals past the excitation pulse. In this way, the dynamics of the liquid crystal in the relaxation regime were probed.

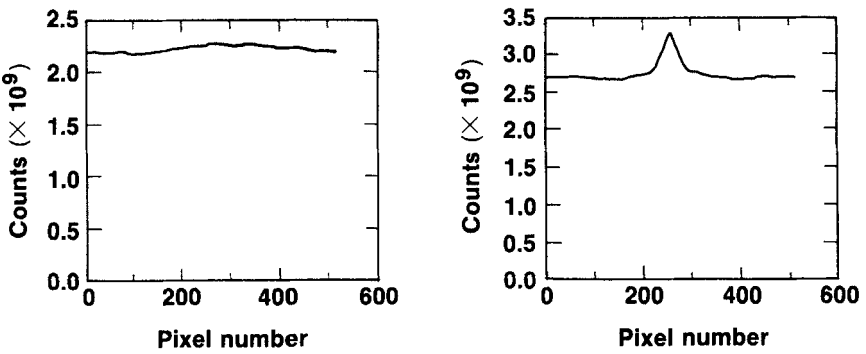
3. Results

The light scattered by the liquid crystal showed distinctive speckle features. The two images in figures 4 (*a*) and (*b*) show the scattered field produced by the liquid crystal at an early time past the foot of the excitation pulse (see figure 4 (*a*), $\Delta t = 100$ ms) and at a later time (see figure 4 (*b*), $\Delta t = 1{\cdot}5$ s). For comparison, the speckle from a mildly etched glass diffuser located in the same focal plane as the liquid crystal sample is shown in figure 4 (*c*). The speckle size in these figures is of the order of 0·5 mm. In order to verify that the liquid crystal speckle patterns produced by two different excitation pulses were statistically independent and not systematic in origin, spatial correlations of the liquid crystal scattered intensity were calculated as displayed in figure 5 (*a*) and (*b*). Figure 5 (*a*) is a cross correlation of two different speckle images produced by the liquid crystal with the same time delay (500 ms) past the foot of the pump pulse. The flatness of this curve indicates little correlation between these two images, and provides proof of the statistical independence of each probe snapshot. In contrast, a sharp peak and relatively flat baseline exists in the autocorrelation of the scattered field shown in figure 5 (*b*) and is characteristic of speckle. A lineout, however, taken through the centre of the CID images also reveals certain lens-like features in the liquid crystal speckle pattern.



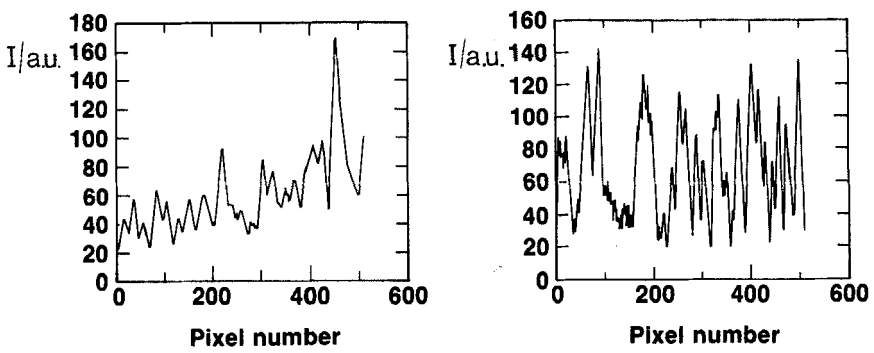
(*a*) (*b*) (*c*)

Figure 4. Scattered field of the probe beam showing liquid crystal speckle patterns produced at (*a*) 100 ms and (*b*) 1·5 s past the onset of the pump pulse. (*c*) Speckle produced by an etched glass diffuser located in the same focal plane as the liquid crystal.



(*a*) (*b*)

Figure 5. (*a*) Cross-correlation of two different liquid crystal speckle images with the same time delay (500 ms) past the excitation pulse. (*b*) Autocorrelation of a single liquid crystal speckle image.



(*a*) (*b*)

Figure 6. CID intensity lineouts of speckle generated by (*a*) the liquid crystal showing a strong, sloping background and low contrast speckle and (*b*) a mildly etched glass diffuser in the same focal plane as the liquid crystal showing high contrast, well-developed speckle.

In figure 6(*a*), the intensity profile of the liquid crystal speckle shows a strong background and relatively low contrast. In addition, the background contains a slight slope. The portion of this scan with higher background is that area of the CID camera closest to the beam propagation axis. The intensity lineout from the mildly etched diffuser, however, shows speckle with high contrast and uniform background, as seen in figure 6(*b*).

4. Discussion

Two key points can be made regarding the correlations indicated in figure 5. Firstly, the scattered intensity is in fact due to the dynamic interaction of the laser field with the guest–host mixture and is not a result of light scattering from other, fixed scatterers and second the evolution of scattering domains is a random process that can be described statistically. The intensity lineouts also suggest, however, that a deterministic, self-focusing effect accompanies the statistically random process that gives rise to speckle. The speckle patterns are not unlike what we would expect from a lens with roughly ground surfaces. In fact, in work reported elsewhere [9], we have observed self-focusing prior to the onset of scattering in this guest–host material. Also, when the excitation pulse width was extended to several hundred milliseconds, the formation of an intensity ring structure in the transmitted beam was observed. This diffraction ring pattern is well-known in nematic liquid crystals and has been reported as due to both thermal and orientational effects in these materials [11]. A lensing effect in the intermediate time regime investigated in this paper would therefore be expected, although further evidence is needed for verification.

The autocorrelations of the CID images have revealed several interesting features of the relaxation process. Figure 7 is a plot of the peak of the autocorrelation versus time delay for the regions of different anchoring strength. The peak of the autocorrelation is proportional to the mean square intensity within the solid angle subtended by the CID camera. Figure 7 indicates that the light scattered from each region of the cell showed the same general trend: increasing to a peak between 0·5 and 1 s and then gradually

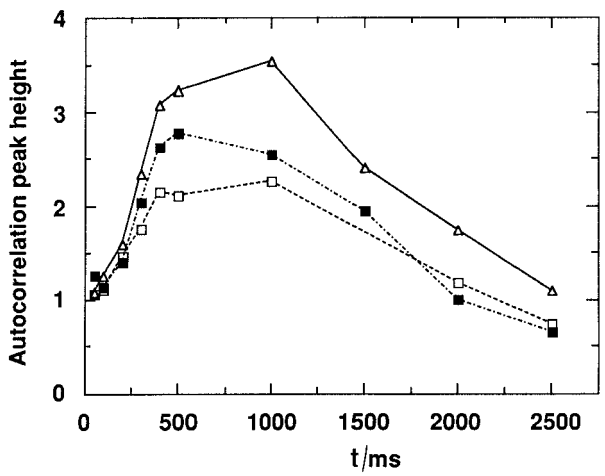


Figure 7. Peak of autocorrelation (proportional to mean square intensity) versus time delay for regions of the liquid crystal cell with different anchoring conditions. At the scattering angle investigated, the amount of light scattered is greater for weaker anchoring. □, two coatings; ■, one coating; △, no coating.

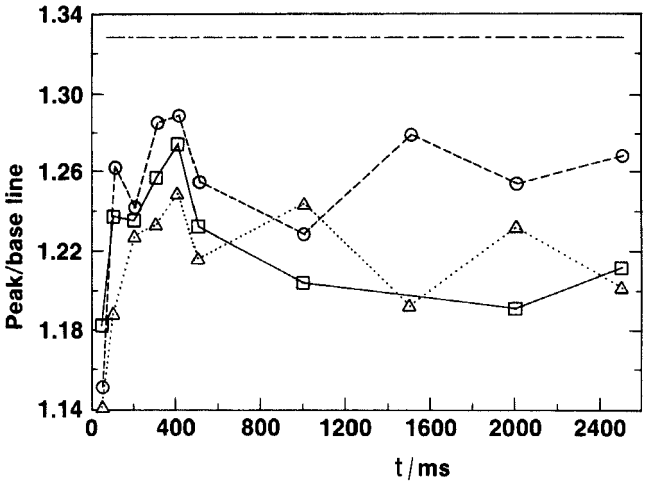


Figure 8. The autocorrelation peak-to-base-line ratio gives the relative magnitude of speckle versus lens-like components. Random molecular orientation in each region of the guest-host system is greatest at 400 ms past the excitation pulse. □, two coatings; ○, one coating; △, no coating; ———, mild diffuser.

decreasing as the system ballistically relaxed to the transparent, ordered state. Also note that the amount of light scattered progressively increased as anchoring strength was reduced. The relative magnitude of speckle and lens-like components of the scattered light has been investigated in figure 8 by plotting the ratio of the peak of the autocorrelation to its baseline. Speckle produced by a ground glass diffuser is caused by a random distribution of heights, or optical path length, on the diffuser surface. In the liquid crystal, this can be interpreted as a randomness in molecular orientation. For an ideal diffuser, the peak-to-baseline ratio is equal to two, whereas in the limit where the peak height equals the baseline, as with an ideal lens, a value of unity is obtained [15]. This ratio is insensitive to changes in the intensity of the light scattered by the liquid crystal and is a measure of the degree of order or disorder in the irradiated region. The plots in figure 8 show that the degree of disorder was greatest ~400 ms after the excitation pulse onset. Although the intensity of light measured at this scattering angle was greatest for the region with no coating (see figure 7), the degree of disorder was generally less than for areas of the cell with stronger anchoring. At longer time delays, the region with strongest anchoring uniformly approached a more ordered state, whereas regions with one or no coatings showed oscillatory behaviour between random and more ordered conditions. Oscillations between the isotropic and nematic phases in a dye-doped nematic film near the melting temperature have been reported by Simoni *et al.* [15] and more recently by Jánossy and Kósa [16]. Our experiment suggests that upon laser heating, periodic phase transitions may also occur in a guest–host smectic A liquid crystal, and shows that anchoring conditions play a critical role in the dynamics of molecular ordering during thermal relaxation of the system. In addition, for power limiting applications it is seen that device parameters may be tailored in order to enhance scattering efficiency.

From the statistical theory of speckle, the rms refractive index variation for the irradiated region of the guest–host mixture has been estimated. The table compares three different, quantitative features of speckle from earlier work [17] with data from the present experiment: (1) the ratio of the peak of the spatial autocorrelation, $R_u(0)$, to

Comparison of current data with earlier work on ground glass. From the statistical theory of speckle, the rms refractive index variation in the irradiated region of the liquid crystal has been estimated.

	Polarized			Unpolarized
	$\frac{R_u(0)}{R_u(\infty)}$	C_R	h_σ	$\frac{R_u(0)}{R_u(\infty)}$
Theory	2·00	1·00		0·707
Reference [17] Measured ground glass	1·88	0·94		
Current data Mild etch glass	1·33	0·58	0·28 μm	
Liquid crystal (No coating, $\Delta t = 50$ ms)	1·15	0·384	0·21 μm $\Rightarrow$ $\boxed{(\Delta n)_{rms} \approx 0{\cdot}0021}$	

$$C_R = \sigma / \langle u \rangle$$

$$C_R = \left[\frac{R(0)}{R(\infty)} - 1 \right]^{1/2}$$

its baseline, $R_u(\infty)$; (2) the contrast ratio of the speckle, C_r; and (3) the rms height variation or diffuser roughness, h_σ. As previously noted for an ideal diffuser (many π radians differential phase), the peak of the autocorrelation is twice that of its baseline, with a contrast ratio of unity. In earlier experimental work by George and co-workers [17], values approaching the theoretical limit were obtained for ground glass. In the current experiment, somewhat lower values were obtained on etched glass due to the mildness of the etch. From the theory described in [17], an rms height variation on the etched surface of 0·28 μm was calculated. Using the same approach, we have determined an effective rms roughness of 0·21 μm for the liquid crystal. Under the assumption that the interaction takes place over the full 100 μm path length of the cell, an rms refractive index variation of 0·0021 is obtained. This calculation is for the region with no alignment coating and a time delay of 50 ms.

5. Conclusion

We have shown that the molecular reorientation which leads to scattering in this guest–host smectic A system involves both statistically random and deterministic processes. As such, the interaction region can be modelled as a time varying diffuser with lens-like features. This is in contrast to the guest–host nematic, in which only a self-focusing effect has been observed [18]. From the diffuser model we have estimated the rms refractive index variation for this interaction, and have shown that the strength of molecular anchoring plays a critical role in scattering dynamics. Further application of the theory of speckle can reveal the scale and temporal behaviour of scattering domains and interaction volume.

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References

[1] KAHN, F. J., 1973, *Appl. Phys. Lett.*, **22,** 111.
[2] DEWEY, A. G., JACOBS, J. T., HUTH, B. G., SINCERBOX, G. T., SPROKEL, G. J., JULIANA, A., and KOEPCKE, R. W., 1977, *SID 77 Digest*, p. 108.
[3] ARMITAGE, D., 1981, *J. appl. Phys.*, **52,** 1294.
[4] MORI, K., MATSUMURA, S., and KOBAYASHI, S., 1986, *Molec. Crystals liq. Crystals Lett.*, **4,** 63.
[5] HUGHES, A. J., and DALEY, R., 1987, *Molec. Crystals liq. Crystals*, **148,** 163.
[6] NAKAMURA, K., NANATAKI, H., and ARIMOTO, M., 1990, *J. appl. Phys.*, **67,** 996.
[7] NAWA, N., and NUO, D., 1991, *Jap. J. appl. Phys.*, **30,** 1038.
[8] BIRECKI, H., NABERHUIS, S., and KAHN, F. J., 1983, *Proc. S.P.I.E. Int. Soc. Opt. Engng*, **420,** 194.
[9] MARSHALL, K. L., GUARDALBEN, M. J., and SCHMID, A. W., 1990, Eye protection employing opto-optical and thermo-optical effects in smectic liquid crystals, Naval Air Development Center Final Report, Contract N62269-87-C-0264.
[10] KHOO, I. C., HOU, J. Y., LIU, T. H., YAN, P. Y., MICHAEL, R. R., and FINN, G. M., 1987, *J. opt. Soc. Am.* B, **4,** 886.
[11] BLOISI, F., VICARI, L., SIMONI, F., CIPPARRONE, G., and UMETON, C., 1988, *J. opt. Soc. Am.* B, **5,** 2462.
[12] WU, J. J., CHEN, S.-H., FAN, J. Y., and ONG, G. S., 1990, *J. Opt. Soc. Am.* B, **7,** 1147.
[13] DE GENNES, P. G., 1975, *The Physics of Liquid Crystals* (Clarendon Press).
[14] GRAY, G. W., and GOODBY, J. W. G., 1984, *Smectic Liquid Crystals* (Leonard Hill).
[15] SIMONI, F., CIPPARRONE, G., UMETON, C., and KHOO, I. C., 1988, *Optics Lett.*, **13,** 886.
[16] JANOSSY, I., and KOSA, T., 1991, *Molec. Crystal liq. Crystals*, **207,** 189.
[17] GEORGE, N., JAIN, A., and MELVILLE, R. D. S., JR., 1975, *Appl. Phys.*, **7,** 157.
[18] JANOSSY, I., and LLOYD, A. D., 1991, *Molec. Crystals liq. Crystals*, **203,** 77.