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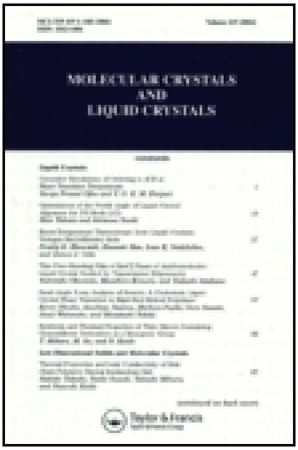

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Light Scattering in Monomeric and Polymeric Smectics

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Light Scattering in Monomeric and Polymeric Smectics

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(*Received May 27, 1986*)

An experimental method is proposed for assessing optical scattering in liquid crystals. Methods for generating dense, stored, scattering textures in both monomeric and polymeric smectics are described, and their optical properties are compared. The scattering textures in both types of material are very similar, and very high optical contrast ratios, 1000–2000, are obtained. A preliminary discussion of temperature effects is also included.

Keywords: light scattering, smectic phases, display devices, polymeric smectics

INTRODUCTION

Increasing use is being made of scattering effects in liquid crystals for both display and data storage purposes. In polymeric and smectic monomeric liquid crystals these scattering effects have the advantage of permanent, high resolution memory. In the majority of applications it is important to maximise the optical contrast ratio, so an understanding of the various factors that affect a measurement of contrast ratio is important. Similar understanding is necessary when optical contrast ratio is used as part of a material assessment programme. This paper discusses the various factors involved, using measurements

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on both a monomeric smectic A and on a polymeric smectic liquid crystal.

The analysis presented here is directly applicable to the use of LC cells in projection systems, though the principles discussed can be applied more generally. It is envisaged that the LC cell contains both clear and scattering regions, and that the projection system generates a bright image of the clear areas and a dark image of the scattering areas. The aim is to use the projection light with maximum efficiency, thus generating an image with maximum brightness and contrast. To achieve this at least three conditions must be satisfied. Firstly, the projection optics must collect all the light passing through clear regions of the LC cell. Secondly, the projection optics must efficiently reject all light deflected by the scattering regions of the cell. Thirdly, a technique must be used that generates such dense scattering textures in the LC that the light intensity passing undeflected through the scattering regions is minimised.

This paper discusses a simple experimental arrangement that permits convenient assessment of these factors, and presents results on the generation and measurement of scattering textures in both a monomeric smectic A LC and in a polymeric LC.

OPTICS

A schematic diagram of the measurement optics is shown in Figure 1.

The light source was a 3 mW HeNe laser operating cw at 633 nm. A considerable degree of attenuation, which was fixed throughout the experiments, was needed to avoid saturating the photodetector when the LC cells were in their clear states. The beam expander produced a parallel beam, 2 mm in diameter, to illuminate the LC cell. The cell was held in a transparent, thermostatically controlled box whose temperature could be set with 0.1°C accuracy between

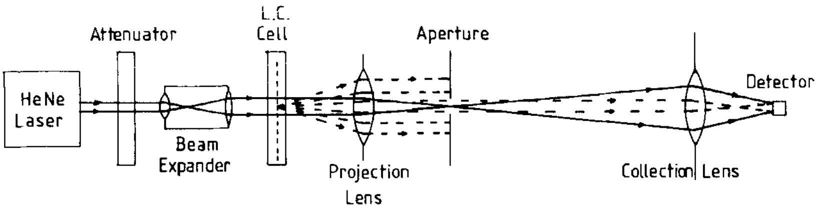

FIGURE 1 Schematic diagram of optical arrangement.

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20°C and 90°C. The projection lens, of 50 mm focal length, was positioned to generate an image of the cell at some 2 m distance. This ensured that the light scattered by the LC (dashed lines) and collected by the projection lens travelled in a parallel beam towards the detector. The projection lens also collected the non-scattered light (solid lines) and brought it to a focus in the plane of the aperture. This non-scattered light then diverged towards the collection lens which had a large enough aperture to collect and focus all the scattered and unscattered light. The final focal planes of these two components of light were not coincident, but when the photodetector (a 2 mm diameter Si photodiode) was placed between these two focal planes, all the light incident on the collection lens fell within the sensitive area of the detector. The aperture, which was selectable from 0.35 mm to 6 mm diameter, was the crucial part of the apparatus and was always placed accurately in the image plane of the light source. Then, provided the aperture diameter was greater than the image size of the source, all directly transmitted light passed unimpeded to the detector or screen. If the aperture size had been smaller than the source image, then the final image brightness would have been reduced. Using a laser source meant that very small apertures could be used. For a laser beam having the conventional Gaussian intensity profile the diameter, d, of the focused beam waist is given in Reference 1 by

$$d = \frac{4\lambda f}{\pi D} \tag{1}$$

where λ is the laser wavelength, f the focal length of the focusing lens, D the beam diameter at the lens. The beam diameters d and D are measured between the $1/e^2$ intensity points. In the present case with $D = 2$ mm, $\lambda = 633$ nm and $f = 50$ mm we obtained $d \div 20$ μm. Therefore, even with the smallest aperture of 0.35 mm diameter there was no significant attenuation of the non-scattered light.

LIQUID CRYSTAL CELLS

The LC cells consisted of two glass plates whose inner surfaces were coated with unpatterned transparent conductor (indium tin oxide). The plate separation of 14 ± 1 μm was achieved using glass fibre spacers in the peripheral seal only; the more normal technique of

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spreading spacers randomly over the whole cell area could not be used because of optical scattering from the fibres.

The monomeric LC used here was based on the commercial cyanobiphenyl mixture S2 (BDH Chemicals Ltd) whose phase transition temperatures are: smectic to nematic at 48.5°C, nematic to isotropic at 49.5°C. Homeotropic surface alignment was obtained using a chrome complex layer on both surfaces.

Several methods of obtaining dense scattering textures in monomeric smectics have been demonstrated. These include laser heating,[2] electrical pulse-heating through the ITO,[3] and dynamic scattering.[4] In our experience all three techniques are capable of generating dense, stable, scattering textures, so for convenience we used dynamic scattering here. The basic S2 mixture was doped with ~0.02% HMAB to enhance the AC conductivity. The scattering textures could then be generated by applying a 1 second burst of 100 V RMS, 100 Hz AC to the cell. The clear, homeotropic texture was generated by applying a similar burst of 10 kHz AC. The resulting textures were found to exhibit excellent uniformity and long-term stability.

The polymeric LC used here was a side-chain copolymer (known as PG 296) supplied by Hull University.[5] This has a polysiloxane backbone with equal proportions of two mesogenic side chains, randomly distributed, as shown in Figure 2. The particular sample of

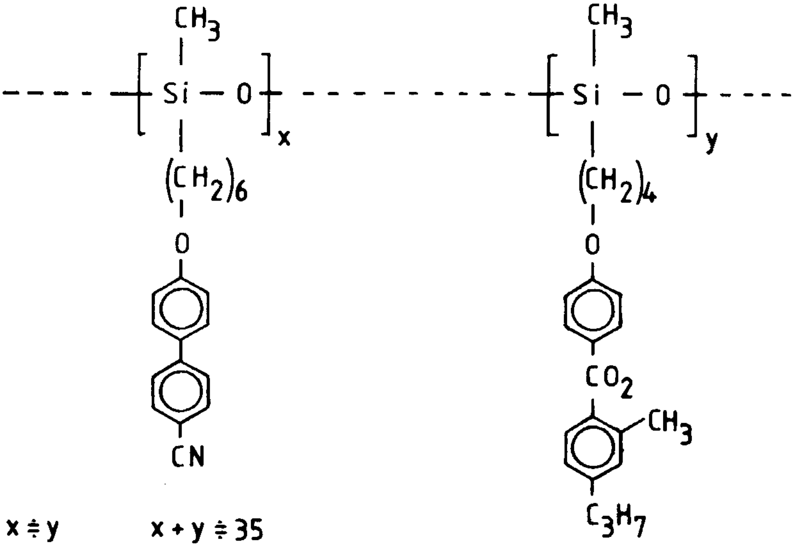

FIGURE 2 Chemical structure of PG 296 polymer.

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PG 296 incorporated a backbone of narrow polydispersity and mean chain length 35. The smectic to isotropic transition occurred with a wide biphasic region between about 68°C and 80°C. It should be emphasised that phase transition temperatures in polymeric LCs can be affected by many factors including chain length, polydispersity and purity, so it is not surprising that our measured transition temperatures differ from those quoted elsewhere for other PG 296 samples. A vacuum cell-filling technique at ~ 100°C was used which ensured good control and uniformity of cell spacing. Cells were prepared with either no surface alignment layer or with a chrome complex layer, but no significant effects due to surface alignment treatment were detected. Dense scattering textures were obtained using controlled cooling of the samples, as in Reference 6. The high temperature dynamic scattering effect of Reference 7 was not employed. It was found that by varying the cooling or annealing conditions a wide range of scattering densities could be achieved. Rapid cooling (i.e. from 90°C to 30°C in one or two minutes) produced only very weak scattering. Annealing at ~ 65°C for several hours, followed by slow cooling to 30°C produced much denser scattering, but the densest scattering was obtained with slow, continuous cooling from 90°C to 30°C over ~ 2 hours. All results quoted used this technique.

Figures 3 and 4 show photo-micrographs taken of typical scattering textures in the S2 and PG 296 cells respectively. Crossed polars were used to enhance the contrast. Rotation of the analyser, however, had no effect on the image contrast, indicating that both scattering textures were completely depolarising. Similarly, rotation of the sample between crossed polars confirmed that there were no macroscopic birefringence effects. It is seen that the microscopic structure of the two materials is strikingly similar. Close examination of these and

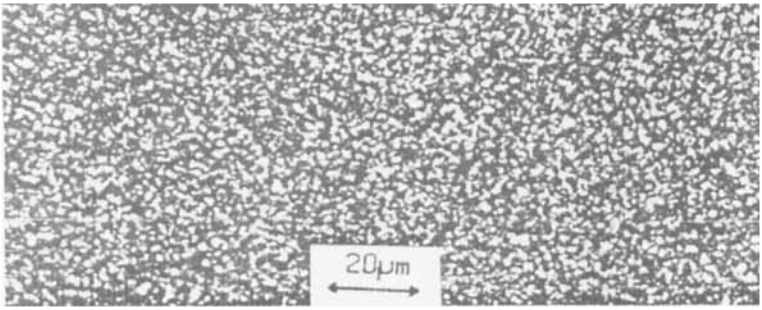

FIGURE 3 Photo-micrograph of scattering texture produced by dynamic scattering in S2; ×40 objective, crossed polars.

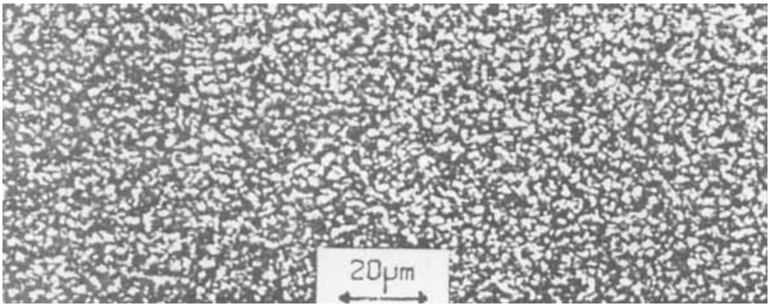

FIGURE 4 Photo-micrograph of scattering texture produced by controlled cooling in PG296; ×40 objective, crossed polars.

other pictures, however, suggests that the feature size in the polymer cell is marginally larger than in the S2 cell. These small differences are probably responsible for the differences between the optical data presented later, but should not be regarded as particularly significant. In this work little effort was expended to investigate the full range of scattering textures and densities obtainable. Doubtless in the case of the monomeric material by optimising the conductive dopant concentration, the electrical drive waveforms and the sample temperature at which scattering was generated, an increase in scattering density would be obtainable. Similarly, in the case of the polymer, closer control of the cooling conditions might also produce denser scattering.

To the unaided eye the two sample types appeared very similar, both being totally opaque. When placed in the measurement apparatus, however, a very weak component of unscattered laser light could just be observed producing a focused spot in the aperture plane.

RESULTS

Figure 5 shows graphs of detector output (in arbitrary units) plotted against aperture diameter in mm. The crosses represent data from the most densely scattering PG296 texture, and the stars were obtained from the densest S2 texture, both measured at about 25°C. The similarities between the two curves indicate the similarity of scattering in the two materials.

Superimposed on the data points are solid curves of the form

$$I(D) = I_o + SD^2 \tag{2}$$

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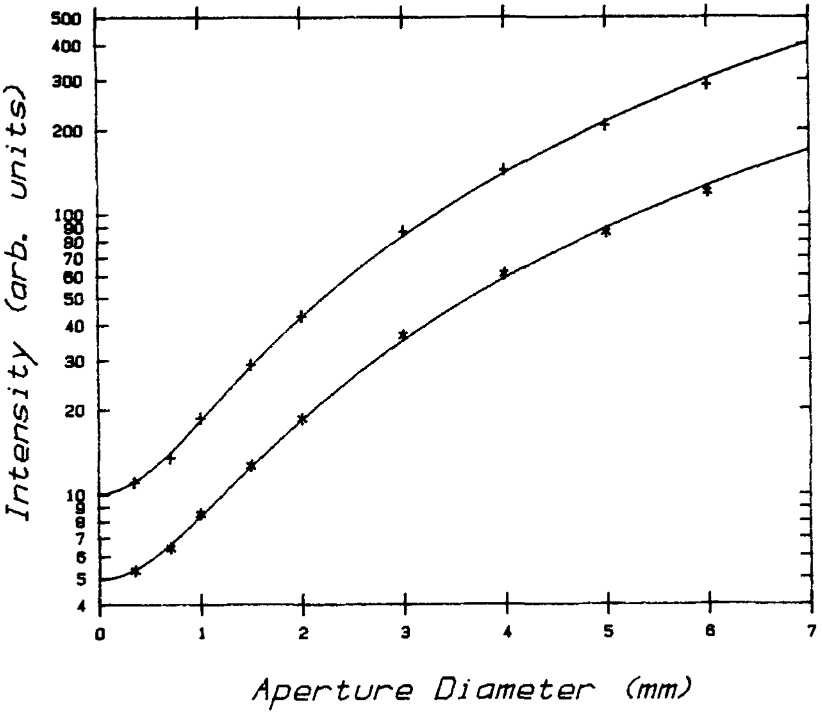

FIGURE 5 Graphs of transmitted intensity versus aperture diameter for PG 296 (+) and S2 (*); solid curves from equations (3) and (4).

where I(D) is the detector output for aperture diameter D, I_o represents unscattered laser light, and S is proportional to the scattered intensity per unit area of aperture. The plotted curves are a least squares fit to the fractional error of each data point, equal weighting being given to all points.

For the polymer cell

$$I(D) = 10.05 + 8.07\, D^2 \tag{3}$$

and for the monomer cell

$$I(D) = 4.93 + 3.34\, D^2 \tag{4}$$

These figures indicate that, not only was there more unscattered light from the polymer cell, but also the forward scattered intensity was greater.

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The fitting formula used here implicity assumes that the angular distribution of scattered light is isotropic. Over the very small range of scattering angles used here (~ 3.5° with the largest aperture) departures from isotropic are likely to be small. At greater scattering angles, however, the limitations of the formula would become apparent for two principal reasons. Firstly, the fundamental LC scattering might not be isotropic. Secondly, the scattering takes place in a medium of high mean refractive index, i.e. $\bar{n} \sim 1.5$, so the angular scattering profile is considerably modified by refraction and reflection loss at the glass to air interface.

These data were converted into "contrast ratio" figures by dividing these intensity figures into the intensity transmitted by the clear cells. The clear state was obtained in the monomeric cell by inducing a homeotropic texture by applying a burst of 100 V RMS at 10 kHz. The clear state for the polymer was taken as the isotropic phase at 85°C. The two clear states were indistinguishable. Figure 6 shows the contrast ratio figures for both cells, the solid curves being derived from the fitted formulae above.

A preliminary investigation was made of the variation of scattering texture with temperature, particularly with the S2 cell. Initially the cell was made densely scattering at room temperature. Using the 0.35 mm aperture, measurements of transmitted intensity were made as the temperature was slowly raised and lowered, always remaining in the smectic phase. It was found that in general the transmitted intensity increased as the temperature increased. This change was accurately reversible provided the cell temperature did not exceed 41°C, the contrast ratio reducing by about 35% over this range.

Microscope examination showed no detectable change in the LC structure over this range, so it is suggested that the variation of birefringence, Δn, was responsible. At temperatures between 41°C and 47°C small irreversible changes in transmitted intensity occurred, but the resulting scattering textures remained long-term stable. It is believed that realignment of the surface layers of LC by the homeotropic alignment treatment caused these small changes. Between 47°C and 48.5°C (the smectic to nematic phase transition) the surface alignment established a clear, homeotropic texture throughout the cell.

Observations on polymer-filled cells were less conclusive. Significant variations in transmitted intensity occurred over the entire smectic range investigated (20°C to 65°C) but it was less easy to distinguish reversible and irreversible changes because of the very long time-constants observed. It appeared that at each temperature there was

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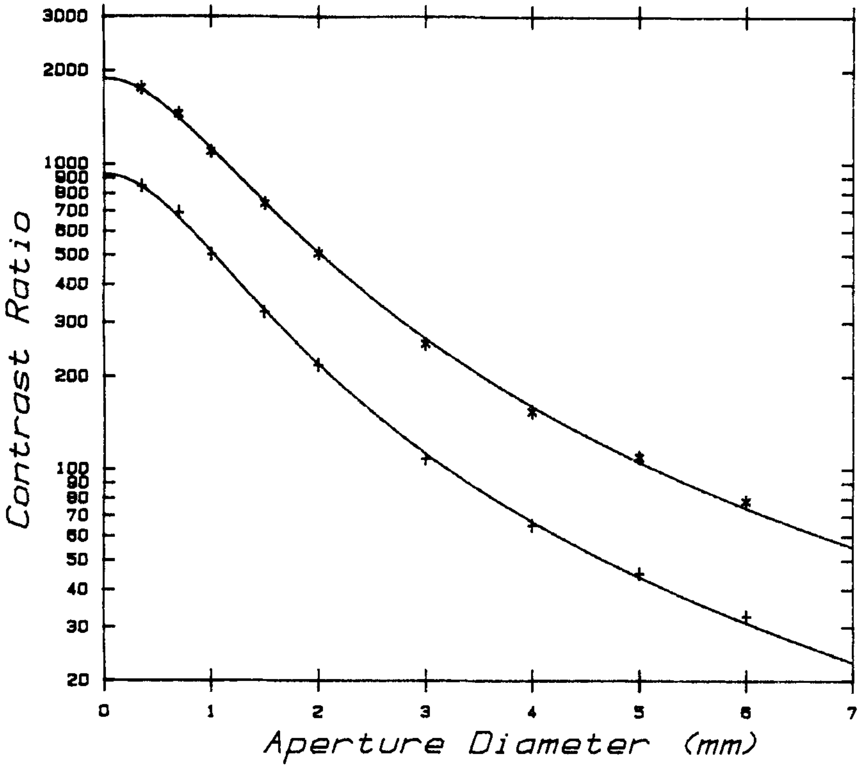

FIGURE 6 Graphs of contrast ratio versus aperture diameter derived from Figure 5 data for PG296 (+) and S2 (*).

an optimum structure of minimum thermodynamic energy, and that the rate of change of structure depended upon how close the instantaneous structure was to optimum, and upon the viscosity at that temperature. A contribution to transmitted intensity variations from the temperature dependence of birefringence could also be discerned.

CONCLUSIONS

The first conclusion from this work is that, contrary to the claims in Reference 6, monomeric and polymeric LCs can exhibit very similar light scattering properties, provided sensible techniques are used to generate the scattering textures. This similarity is not unexpected since the photo-micrographs indicate that the microscopic structures of both materials are similar, as are the molecular structures that give

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rise to the birefringence. The slight differences in light intensity and contrast ratios obtained are probably not of great significance.

The second conclusion is that very high contrast ratios, > 1000:1, can be obtained provided the appropriate optical arrangement is used. With laser sources the full contrast range can be exploited, but with more conventional sources such as arc or filament lamps the maximum contrast will be reduced. The critical factor is the size of the image of the source in the aperture plane, since the aperture diameter must be set marginally larger than this value to obtain maximum screen brightness. From Figure 6 it can be seen that even with a 5 mm diameter aperture the contrast ratio can be greater than 100:1 with the S2 cell.

Finally, the effects of temperature are twofold. There is a narrow band of temperatures near the upper phase transition temperature where the scattering textures are unstable. This temperature range is probably both material- and cell-dependent, but with the S2 material and homeotropic alignment used here the unstable range was less than 2° wide. This range was not determined for the polymer cell, partly because of the wide biphasic temperature band. At lower temperatures with both materials excellent long-term stability was found, coupled with a small reversible variation attributed to changes of birefringence with temperature.

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