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Light Scattering and Contrast in Thermally Addressed Liquid Crystal Displays

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The effects of structural order on the light scattering characteristics of liquid crystal display cells containing smectic *p*-*n*-octyl-*p*'-cyanobiphenyl have been investigated. The scattering characteristics of different structures in the cell due to surface treatments are qualitatively analyzed for their effects on the contrast ratio when used in the thermally addressed liquid crystal display (TALC). Of the four structures investigated, i.e., homeotropic, fan-like, uniaxial rods and spherulites, the spherulitic structure has been found to exhibit the scattering profile best suited for high contrast because of the near-zero scattering near the incident beam. Based on the light scattering theory for spherulites, optimization of structural parameters such as size and order of spherulites, and device design such as collecting angle of projection lens for maximum display contrast has been predicted. Methods to improve contrast have also been discussed.

INTRODUCTION

Thermally addressed liquid crystal display (TALC) was first described by Kahn.¹ In the application, the liquid crystal display cell must exhibit a high scattering written state and a weak scattering erased state. The erased state can be normally achieved by applying an electric field across the cell to induce a homeotropic alignment of the liquid crystals, resulting in a very weak scattering state. The written state can be obtained by thermal addressing in the absence of the applied electric field, which disrupts the homeotropic orientation and causes formation of new structural arrangements which scatter light strongly. The difference in the optical properties of these two states, combined with a suitable projection system, produces an image due to the difference in the intensity of light that passes through the projection lens.

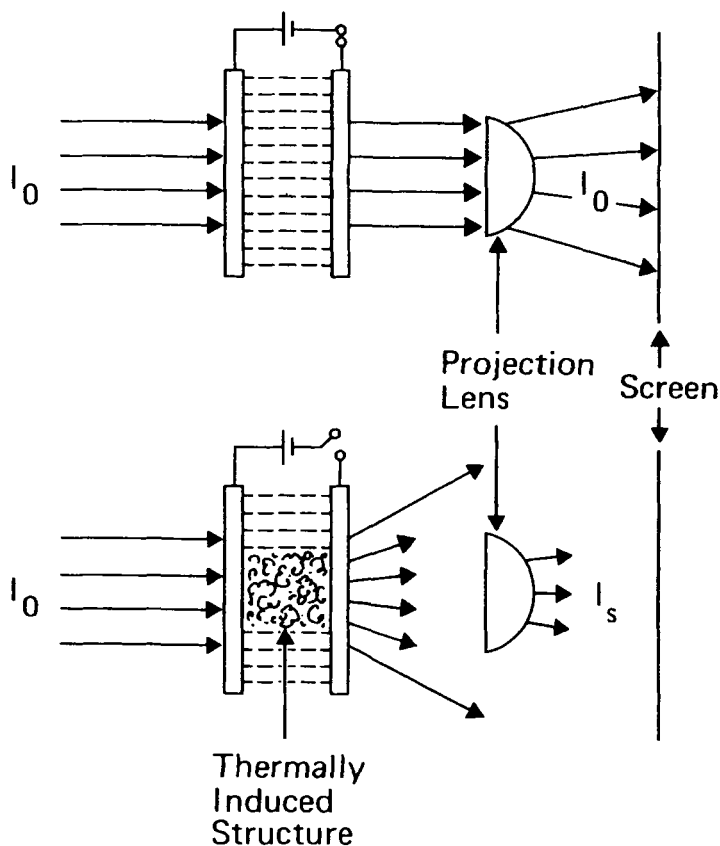


FIGURE 1 Schematic representation of TALC display.

A schematic representation of the display system is shown in Figure 1. The contrast is then strongly dependent upon characteristics of light scattering in the two states. Here, we want to emphasize "scattering characteristics" i.e., variations of scattering intensity with scattering angles, instead of absolute scattering intensity. This is because a strong scattering system with an inadequate scattering profile can yield poorer contrast than a weak scattering system with a proper scattering profile as will be described later.

In the TALC display, the scattering arises mainly from structural arrangements which cause fluctuations in optic axis orientation. In this work we discuss the different scattering characteristics from structural arrangements affected by surface treatments of liquid crystal cells containing smectic *p*-*n*-octyl-*p*'-cyanobiphenyl and their effects on contrast. Methods to improve contrast are enumerated. Although the discussion is centered on smectic liquid crystal cells, the structure-scattering relationships are invariant to the

phase of liquid crystals and, thus, can be applied to other TALC displays using either nematic or cholesteric liquid crystals.

STRUCTURE AND LIGHT SCATTERING

It is well known that surface treatments of a liquid crystal cell can affect the orientation of liquid crystals and, hence, result in structures of different orientation correlations. Depending on the size of the correlated structure and the optic axis orientation within the structure, the scattering of light has different characteristics. The different structures effected by surface treatments in smectic *p*-*n*-octyl-*p*'-cyanobiphenyl liquid crystal cells have been studied previously.² The findings are briefly described here to pave the ground for later discussions on the relationships between contrast and light scattering characteristics. The cell preparation methods were described in Ref. (2) and the description of sample cells is summarized in Table I. The cells were surface treated to induce perpendicular, parallel, or mixed (parallel orientation at one surface and perpendicular at the other surface) alignment of the liquid crystals with respect to the cell surface.

The structures of the smectic liquid crystals in Cells B, C, and D when viewed between crossed polaroids on a polarizing microscope are shown in Figure 2. Cell A appeared dark between crossed polaroids due to the homeotropic alignment and, therefore, is not shown in Figure 2. The structures found in Cell B (Figure 2a) are fan-like domains and those in Cell C are long uniaxially oriented rods with the long direction of the rods oriented parallel to the lapping direction. The structures in Cell D are two-dimensional spherulites. They are two-dimensional because the diameter of the spherulities is much larger than the thickness of the cell.

The light scattering patterns from these cells were obtained using a photographic scattering set up shown schematically in Figure 3. The scattering patterns were recorded in H_v and V_v mode of polarization; the subscript

TABLE I
Description of sample cells²

Sample Cell	Surface Treatment	Alignment	Thickness μm
A	polyfluorocarbon coated	perpendicular	13.0
B	SiO ₂ coated	parallel	13.0
C	lapped	parallel	13.0
D	combined ^a	mixed	13.0

^a One surface coated with SiO₂ and the other surface coated with polyfluorocarbon.

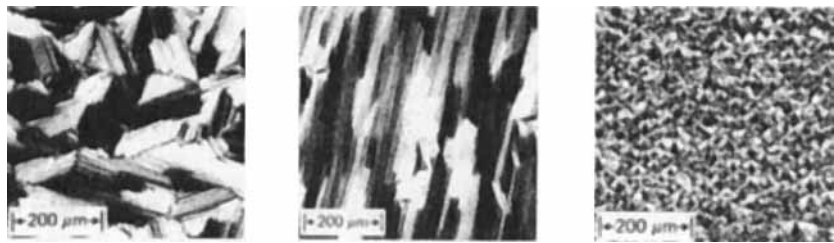


FIGURE 2 Photomicrographs of sample cells of various surface treatments between cross polaroids; (a) Cell B; (b) Cell C; (c) Cell D.

indicates the polarizer direction and the capital letter indicates that of the analyzer with V and H denoting vertical and horizontal directions, respectively. The patterns for Cells B, C, and D are shown in Figure 4. Again, the scattering patterns for Cell A are not shown because the scattering intensity from the cell was too weak to be recorded.

When polarizers are used, the scattering patterns exhibit distinct characteristics associated with different structures. In case no polarizer is used, as may be adopted for actual displays, the scattering patterns can be regarded as the superposition of H_v , H_h , V_v , and V_h patterns. For an isotropic scattering system H_v and V_h are the same, and so are V_v and H_h . Therefore, structure-contrast relationships can be analyzed by examining the H_v and V_v scattering characteristics separately which can otherwise be masked when no polarizer is used. The experimental scattering patterns shown in Figure 4 all agree well with the theoretically predicted scattering patterns from the corresponding structures;² i.e., four-fork pattern (Figure 4a) associated with fan-like structures, long streaks (Figure 4b) associated with uniaxially oriented rods and

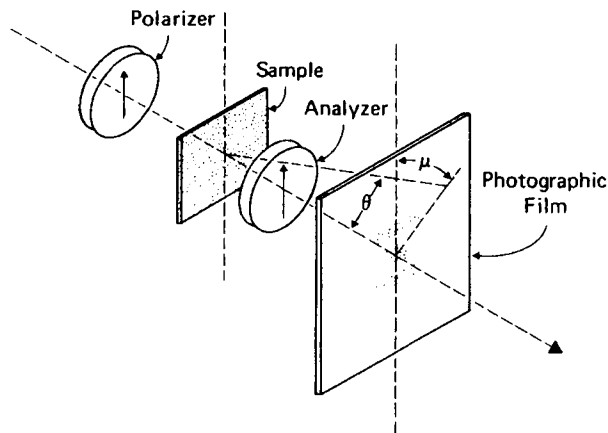


FIGURE 3 Schematic diagram of a photographic light scattering apparatus.

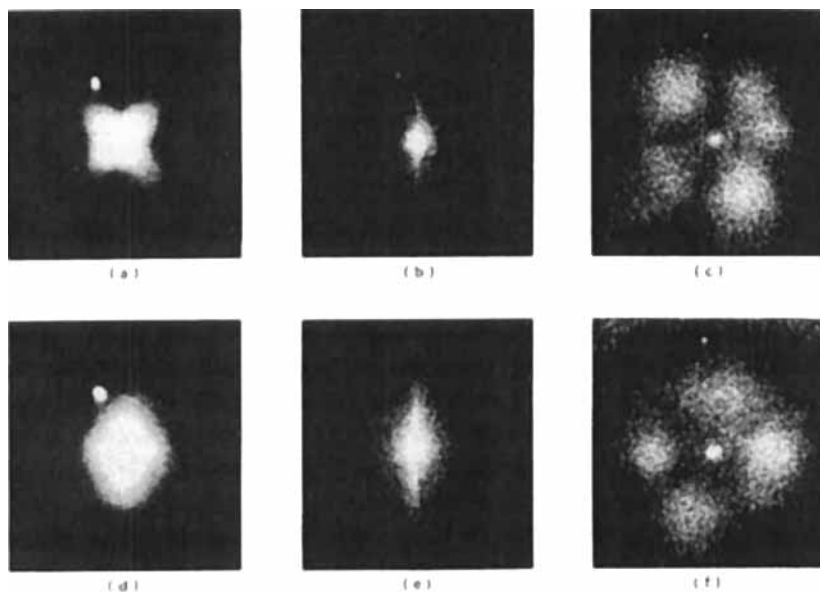


FIGURE 4 Light scattering patterns of sample cells of various surface treatments; (a) Cell B, H_v ; (b) Cell C, H_v ; (c) Cell D, H_v ; (d) Cell B, V_v ; (e) Cell C, V_v ; (f) Cell D, V_v .

clover-leaf patterns (Figure 4c) associated with spherulitic structures. By analyzing these different scattering characteristics, one can predict their effects on contrast ratio.

LIGHT SCATTERING AND CONTRAST

In the TALC display, as shown in Figure 1, a maximum contrast is achieved when the ratio of the collected intensity in the clear state I_0 with respect to the intensity in the written state I_s is the greatest. This means that in the scattering state, the light should be scattered away from the incident light so that it is not collected by the projection lens. Therefore, a highly scattering state which scatters light only at small angles θ (see Figure 3) will give poorer contrast than a weak scattering state which scatters light at large angles. Thus, the contrast is strongly dependent upon the scattering characteristics as well as the collecting angle of the projection lens.

We shall now analyze the scattering characteristics from the different cells. In cell A, the homeotropic alignment of the elongated molecules is parallel to the optical path of the incident light and the cross-section of the scattering domains is smaller than the wavelength of the light² and, hence, results in a very weak scattering state. This type of structure is also achieved in the erased

state, and is caused by electric field induced orientation. Therefore one would expect very low or zero contrast from this cell due to the lack of change in optical properties between the erased and written state.

The scattering intensity from fan-like structures in Cell B (Figure 4a and 4d) is strongest at $\theta = 0^\circ$ and decreases monotonically with increasing θ 's for both H_v and V_v . The rate of intensity decrease with θ is determined by the size of the scattering structure; the intensity decreases more rapidly for larger structures. Therefore for large fan-like structures as observed in Cell B, most of the scattering occurs near the incident beam as the patterns shown in Figure 4a and 4d cover only to $\theta \approx 5^\circ$. Therefore, if the collecting angle of the projection lens is greater than 5° , one can expect a very low contrast from the cell because most of the scattered light will be projected and there is only a small difference between I_0 and I_s . Even if the collecting angle of the projection lens is smaller than 5° , one still cannot expect a high contrast because the scattering intensity from fan-like structures is strongest at $\theta = 0^\circ$ resulting in a large value of I_s .

In Cell C the scattering structures are rod-like and the scattering intensity is strongest at $\theta = 0^\circ$. Based on the same reasons discussed above in the case of Cell B, a low contrast is expected from this cell. In addition, the azimuthal (μ direction in Figure 3) scattering intensity variation from this cell will be highly anisotropic due to the uniaxial orientation of the rod-like structures.² This is clearly shown in the scattering patterns shown in Figures 4b and 4e where the streaks are extended perpendicular to the orientation of the rods. The projected image from such anisotropic scattering will then have variable contrast depending on viewing angles.

The scattering patterns (Figure 4c and 4f) from Cell D are characteristic of spherulitic scattering. The spherulitic scattering differs from the rod scattering in that there exist scattering maxima away from the incident light. The angle θ where the maximum scattering occurs is directly related to the average size of the spherulites. When the size of the spherulites is small, the scattering maximum is located at a larger angle θ , and vice versa. Theoretically, the H_v scattering intensity from a perfect spherulite is expected to be zero at $\theta = 0^\circ$.³ This is a consequence of the circular symmetry of the spherulite from which the scattered amplitude from one quadrant of the spherulite will exactly cancel that from another at $\theta = 0^\circ$. In real cases, such a perfect symmetry is not maintained in a spherulite as the optic axis may deviate from the perfect orientation due to the inclusion of amorphous or less ordered structures in the spherulite. Such orientation disorder contributes some scattering at zero scattering angle⁴⁻⁶ as evidenced in Figure 4c. Nevertheless, the excess scattering at angles close to incident beam represents a small fraction of the total scattering even for unpolarized incident light wherein the combinations of H_v and V_v scattering characteristic must be considered. Therefore, unlike the

scattering from rods where the maximum scattering occurs at $\theta = 0^\circ$, the spherulitic scattering is weak at very small θ 's and the maximum scattering occurs at an angle somewhere away from the incident beam. Therefore, if the collecting angle of the projection lens is smaller than the angle of maximum scattering, a low I_s and, hence, a high contrast will be achieved.

The comparison of light scattering characteristic of the four cells clearly shows the most favorable scattering structure to be spherulitic. Also a smaller collecting angle of the projection lens should give better contrast. A plot of the experimentally measured contrast vs collecting angle for the four cells is shown in Figure 5. The contrast were obtained by measuring the relative amounts of light that pass through the cell between the clear state and the scattered state. The detector is a Bendix PIN photodiode with a circular sensor of 1.2 cm in diameter. The collecting angle was varied by changing the distance between the sensor and the cell surface.

OPTIMIZATION OF CONTRAST BASED ON SCATTERING THEORY

Now that the favorable scattering structure for better contrast has been determined, one can carry out a detailed analysis of the scattering characteristics and their combination with the collecting angle of the projection lens for an optimum contrast. We make use of the light scattering theory for three-dimensional spherulites instead of the two-dimensional theory because in actual display applications the size of spherulites is smaller than the thickness of the cell and, hence, three-dimensional spherulites are formed ordinarily.

Light scattering from three-dimensional spherulites with anisotropic polarizabilities are expressed by:³

$$I_{H_v} = \frac{1}{4} K_1 V_s^2 \cos^2 \rho_2 \left\{ \left(\frac{3}{U^3} \right) (\alpha_t - \alpha_r) \left[\frac{\cos^2(\theta/2)}{\cos \theta} \right] \times \sin 2\mu (4 \sin U - U \cos U - 3 \text{Si } U) \right\}^2 \quad (1)$$

and

$$I_{V_v} = K_1 V_s^2 \cos^2 \rho_1 \left\{ \left(\frac{3}{U^3} \right) [(\alpha_t - \alpha_s)(2 \sin U - U \cos U - \text{Si } U) + (\alpha_r - \alpha_s)(\text{Si } U - \sin U) - (\alpha_t - \alpha_r)] \times \left[\frac{\cos^2(\theta/2)}{\cos \theta} \right] (4 \sin U - U \cos U - 3 \text{Si } U) \right\}^2 \quad (2)$$

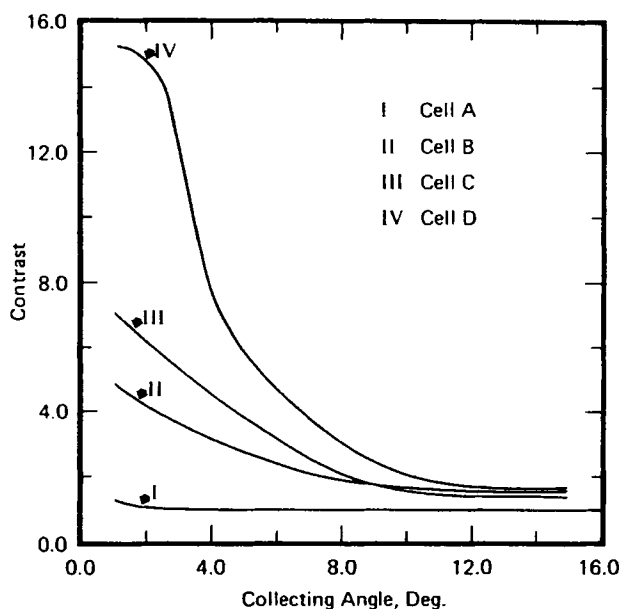


FIGURE 5 Measured contrast vs collecting angles for sample cells of various surface treatments.

where I_{V_v} and I_{H_v} are the scattered intensities with parallel and crossed polarization; K_1 is a physical constant; V_s is the volume of the spherulite, $(\frac{4}{3})\pi R^3$, where R is its radius; $U = 4\pi(R/\lambda)\sin(\theta/2)$, where λ is the wavelength of the light in the scattering medium; α_t , α_r , and α_s are the polarizabilities of the spherulite for light polarized in the tangential and radial directions of the spherulites, and of the surroundings; and θ and μ are scattering angles as defined in Figure 3. The function $\text{Si } U$ is the sine integral $\int_0^U (\sin x/x)dx$, and $\cos \rho_1$ and $\cos \rho_2$ are defined as:

$$\cos \rho_1 = \frac{\cos \theta}{(\cos^2 \theta + \sin^2 \theta \cos^2 \mu)^{1/2}} \quad (3)$$

$$\cos \rho_2 = \frac{\cos \theta}{(\cos^2 \theta + \sin^2 \theta \sin^2 \mu)^{1/2}} \quad (4)$$

The scattering intensity I^θ inside a solid angle θ is then obtained from:

$$I^\theta = \int_{\theta'=0}^{\theta} \int_{\mu=0}^{2\pi} I_{sc}(\theta', \mu) \sin \theta' d\mu d\theta' \quad (5)$$

where I_{sc} is either I_{H_v} or I_{V_v} depending on the polarization condition. Calculated scattering intensities $I_{H_v}^\theta$ and $I_{V_v}^\theta$ of spherulites of $3 \mu\text{m}$ radius under

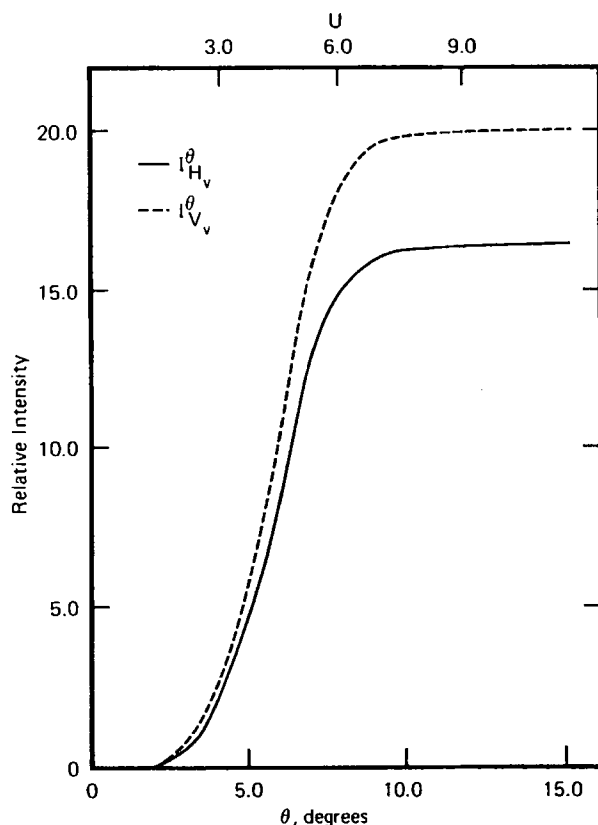


FIGURE 6 Calculated integrated scattering intensities within scattering angle for I_{H_v} and I_{V_v} from spherulites of 3 μ m in radius.

crossed and parallel polarization, respectively, are shown in Figure 6. The collecting angle of the projection lens, θ^* , is related to the scattering angle θ by the Snell's law given by $\sin \theta^* / \sin \theta' = n$, where n is the refractive index within the cell. As shown in Figure 6, the integrated scattering intensity for both H_v and V_v reaches an asymptote above $\theta = 9^\circ$. Assuming $n = 1.5$, then a projection lens with a collecting angle greater than 13.6° produces no images as all the scattered light is collected by the projection lens and no contrast can be obtained. The integrated scattered light decreases rapidly with smaller θ 's, and thus improves contrast. Based on the calculation, the best contrast for a cell containing 3 μ m spherulites is achieved when $\theta \approx 2.5^\circ$ or a projection lens collecting angle of 3.7° (for $n = 1.5$),

In the TALC display where unpolarized light is being used and the absorption by liquid crystals are negligible, then transmitted intensity I_t is given by:

$$I_t = I_0 - (I_{H_v}^\infty + I_{V_v}^\infty) \quad (6)$$

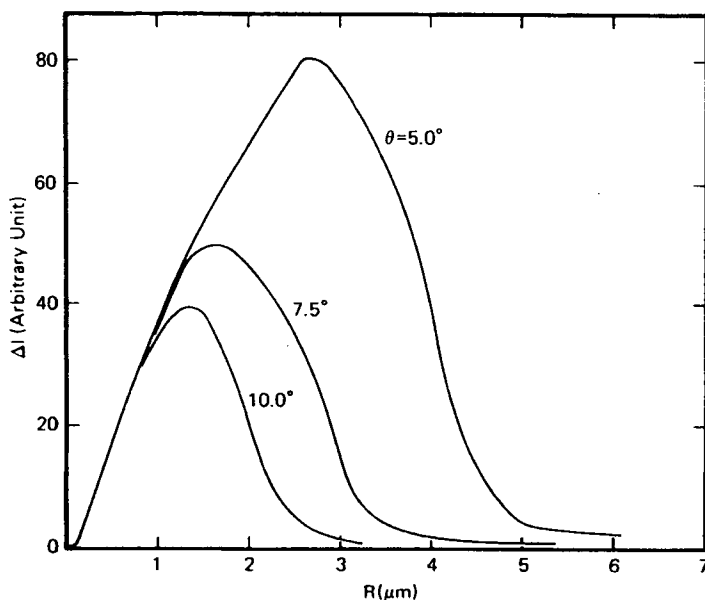


FIGURE 7 Calculated I in arbitrary unit versus radius of spherulites.

where I_0 is the incident light intensity. The superscript ∞ represents an integration from $\theta = 0^\circ$ to $\theta = 180^\circ$. The total light intensity collected by a projection lens I_s is then

$$I_s = I_t + I_{H_v}^\theta + I_{V_v}^\theta \quad (7)$$

By substituting Eq. (6) into Eq. (7),

$$I_s = I_0 - \Delta I \quad (8)$$

where

$$\Delta I = (I_{H_v}^\infty - I_{H_v}^\theta) + (I_{V_v}^\infty - I_{V_v}^\theta) \quad (9)$$

In order to obtain maximum contrast, I_s should be minimized, i.e., ΔI should be maximized. Calculated value of ΔI for various combinations of R and θ are shown in Figure 7. The calculation shows that the spherulite radius should not be smaller than $0.5 \mu\text{m}$ since ΔI decreases rapidly for all θ 's when R is less than $0.5 \mu\text{m}$. It also indicates that the projection lens with a smaller collecting angle has better contrast latitude for structural size variations as judged from the width of the curves in Figure 7. The analysis also provides optimization of material requirements and device design for maximum contrast. If the projection lens has a collecting angle of $\theta^* = 7.5^\circ$ assuming the refraction index of the cell is about 1.5, then the radius of the spherulites in the cell should be in

the range of $1 \sim 4 \mu\text{m}$ from the curve for $\theta = 5^\circ$ shown in Figure 7. For spherulites of optimum size Eqs. (1) and (2) also predict that materials with larger polarizability anisotropy, i.e., $(\alpha_r - \alpha_t)$, should give better contrast due to the stronger scattering intensities.

METHODS TO IMPROVE CONTRAST

Based on the analysis of scattering characteristics for various structures in the smectic liquid crystals, it is obvious that in order to obtain high contrast the scattering structure should be spherulitic, and the collecting angle of the projection lens should be optimized according to the size of the spherulites. Usually, the size of the spherulites is large and, hence, requires a very small collecting angle of the projection lens and, thus, reducing the brightness of the display. Therefore, methods to reduce the size of spherulites must be investigated.

We have found that by applying a small electric field, either a.c. or d.c. during writing, i.e. when the liquid crystals are heated to the nematic or isotropic phase and then cooled to the smectic phase, the size of the spherulites can be altered. The required voltage for size reduction is in the range of $1 \sim 20 \text{ V}$ compared to $100 \sim 200 \text{ V}$ required for obtaining the clear state. Above 20 V , a partial loss of spherulitic structure was observed due to the occurrence of homeotropic alignment of the liquid crystals. The size reduction caused by a 10 V d.c. in cell D is shown in Figure 8. It appears that the applied voltage not only reduces the size but also narrows the size distribution of the spherulites which is also advantageous. This is because any distribution of sizes, when compared with a monodisperse spherulitic system, shifts the scattering profile towards lower angles.^{7,8}

The extent of the size reduction was found to depend on the applied voltage. A largest change in size was observed at a small voltage, ca. 1 V . Further higher voltages caused only a gradual decrease in size. The changes in scattering pattern due to the applied voltage are shown in Figure 9 for unpolarized incident beam. Figure 9a represents the scattering pattern from the cell when no voltage was applied. The label at the upper part of the picture indicates the angle at which the maximum scattering occurs. It should be noted that this angle is double the θ defined earlier. It can be seen (Figure 9b) that when voltage of 1 V is applied, the maximum scattering angle is shifted significantly. Due to the spherulitic scattering characteristics, little scattering is noted near the incident beam and the strongest scattering appears as a band (superposition of H_v and V_v) with its locations inversely related to the size of the spherulites. With increasing applied voltage, the band is further shifted

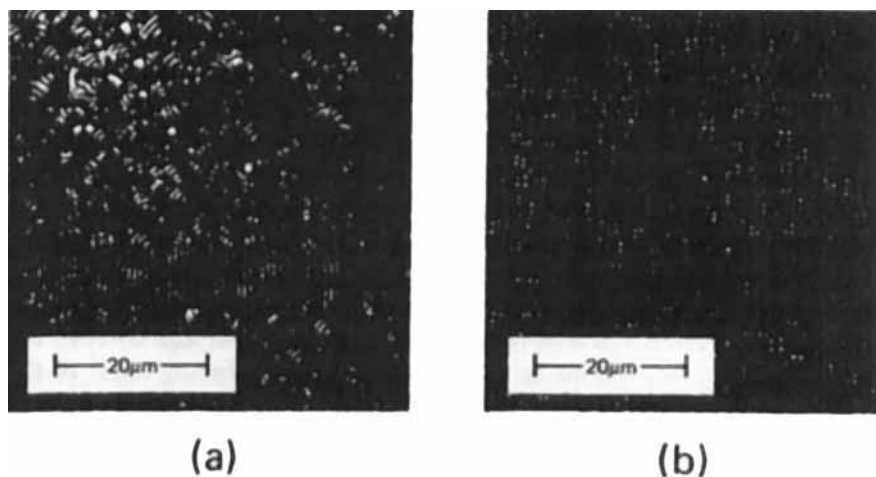


FIGURE 8 Photomicrographs showing the effect of applied voltage in the size of spherulites; (a) no applied voltage; (b) 10V applied voltage.

outward, but the extent of shift is gradual due to the gradual change in size as mentioned above.

The size of spherulites can also be controlled by the cell thickness. It was found that the spherulite size in a wedge-shaped cell of the same surface treatments as Cell D increases with cell thickness.² It was also reported² that the smaller spherulites in a thinner cell is more ordered than the larger ones found in the thicker cells. The disorder in spherulites contributes scattering near $\theta = 0^\circ$,⁵ which causes a reduction in contrast. Therefore a thinner cell is desirable for obtaining smaller spherulites of better ordering. However, the overall scattering intensity from a thinner cell is weaker than that from a thicker cell because the overall scattering intensity increases with increasing scattering volume. Optimization of scattering characteristics and intensity must then be considered when the size of spherulites is to be controlled by adjusting cell thicknesses.

The disorder in spherulites is strongly dependent on the rate at which the liquid crystals are cooled to the smectic phase.² When the liquid crystals are cooled rapidly from the nematic phase or isotropic melt to the smectic phase, the resultant spherulites are highly disordered. It has been shown² that in a rapidly quenched thick cell, the disorder becomes so large that the spherulitic aggregates of liquid crystalline domains turn into randomly oriented aggregates which scatter light strongest at $\theta = 0^\circ$. Therefore, slowing down the cooling rate by thermal biasing of the cell around the temperature of smectic phase transition and controlled heating to avoid superheating of the liquid crystals will improve the contrast.

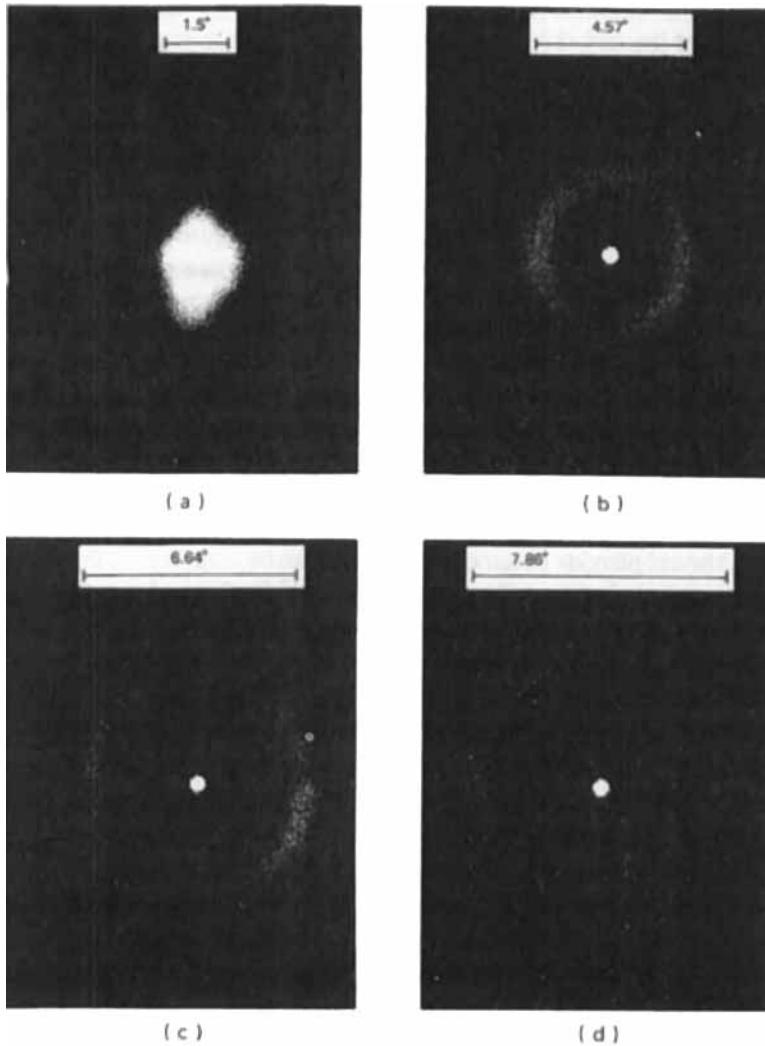


FIGURE 9 Changes in light scattering pattern for unpolarized incident beam due to applied voltages; (a) no voltages; (b) 1V; (c) 5V; (d) 10V.

Since contrast is affected by the volume of thermally induced scattering structures in the cell, it is essential that the thermal absorption is high. Usually, the liquid crystals used in TALC display absorb weakly in the wavelength region of the heating laser, either a GaAs or HeNe laser. An improvement in the thermal absorption can be achieved by coating the cell surface with an absorbing dielectric layer. In this method, the heat absorption occurs at the

surface and a temperature gradient exists both in the lateral and thickness directions of the cell. The temperature gradient can cause the formation of spherulites of non-uniform size and ordering which reduces not only the contrast but also the resolution. Incorporation of a dye which absorbs in the wavelength region of the laser can solve these problems as well as greatly increase the contrast due to the uniform absorption throughout the volume.⁹ In addition, the dye may act as a nucleating agent which increases the number of spherulites and, hence, reduces the size of the volume filling spherulites. Therefore, incorporation of an absorbing dye which can be mixed uniformly with the liquid crystals without destroying the liquid crystalline behavior of the material should improve the contrast as well as reduce the energy requirement for the writing laser beam.

In a scattering system I_{V_v} is usually more intense than I_{H_v} . This is because I_{H_v} comes only from orientation fluctuations while I_{V_v} arises from both orientation and density fluctuations. The liquid crystal cells studied in this work showed very strong I_{H_v} suggesting that the scattering is largely due to orientation fluctuations.² Since contrast is the ratio of I_0/I_s , elimination of I_{H_v} in I_s should provide better contrast. This can be achieved by using parallel polarizer and analyzer to allow only I_{V_v} in I_s .¹⁰ Such a polarization filtering to improve contrast works with parallel polarization only for the black-on-white display (written spot is dark). Under cross polarization condition, little light is transmitted in the clear state. Hence, when the I_{H_v} is strong, cross polarization filtering should improve the contrast for white on black display (written spot is bright).

CONCLUSION

Of the various structures observed in TALC display cells, we have concluded, on the basis of the characteristics of light scattering from these structures that the most desirable structure for obtaining high contrast is spherulitic. Based on the structure-scattering relationships of spherulites, optimization of structure parameters such as the size and order of spherulites, and the device design such as the collecting angle of the projection lens has been deduced.

Methods to improve contrast have been devised from both materials and device aspects. In the material areas, these include: (1) formation of spherulitic structure of liquid crystalline material in combination with the surface treatments; (2) large polarizability anisotropy of liquid crystals; (3) incorporation of a dye which absorbs strongly in the wavelength region of the thermally addressing laser. From the device aspects the most important consideration is to optimize (1) the collecting angle of the projection lens with respect to the scattering characteristics of the cell. Other methods include:

(2) size control of spherulities by applying an electric field; (3) optimization of cell thickness to obtain proper size and order of spherulites without losing scattering intensities; (4) thermal biasing and controlled heating of the cell to reduce cooling rate; and (5) contrast increase by polarization filtering.

The experiments and discussions are all based on the transmission projection. In the reflective projection systems the reflective properties of the cell surfaces become important because the path length of light through different interfaces, e.g., air-glass, glass-liquid crystals, etc. is doubled as compared with that of the transmission projection system. However, the scattering characteristics of the liquid crystal cell are not changed other than the possible effect of multiple scattering. Therefore the discussions presented in this work are also applicable to the reflective projection systems.

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