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## Liquid Crystals

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# Chromaticity of polymer-dispersed cholesteric liquid crystals

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The colour producing properties of the polymer-dispersed cholesteric liquid crystal colour display have been measured. We show how the central wavelength of the reflection spectrum depends on the sample voltage, chiral concentration, temperature, and viewing angle. From the reflection spectra we calculate the chromaticity coordinates for two situations and display them on chromaticity diagrams. The results indicate that these new systems may be suitable for the use in multicolour displays.

## 1. Introduction

Recently, Crooker and Yang [1] have demonstrated that polymer-dispersed cholesteric liquid crystals can be used for application in electro-optic colour displays. In the polymer-dispersed cholesteric liquid crystal display, chiral nematic (i.e. cholesteric) materials with negative dielectric anisotropy and high chirality are dispersed in a non-mesogenic polymer. If the liquid crystal is confined to small, spherical cavities in the polymer, anchoring at the drop surfaces forces the liquid crystal into a complicated director configuration known as the spherulite model proposed by Frank and Pryce [2, 3]. Such cholesteric droplets appear colourless (see Figure 1 (*a*)). However, for materials with negative dielectric anisotropy, application of an electric field causes a reorientation of the cholesteric structure to a uniform orientation of the helix axis along the field direction. Consequently, a bright selective reflection appears (see figure 1 (*b*)) if the cholesteric pitch $p$ is in the range of the wavelength of visible light. In this configuration, the wavelength $\lambda_0$ of maximum reflectivity for light incident along the helix axis is given by

$$\lambda_0 = np, \tag{1}$$

where $n$ is a mean refractive index [4]. Compared to various other electro-optic effects in liquid crystals, the polymer-dispersed cholesteric liquid crystal display has the advantage that: (1) no polarizers are necessary to observe the selective reflection; (2) relaxation to the non-reflecting state occurs without special surface treatment; and (3) polymer-dispersed systems do not require a solid substrate, thereby allowing the production of large area flexible displays. The two latter properties have been impressively shown by polymer-dispersed nematic liquid crystals which have been extensively studied during the last decade [5–7].

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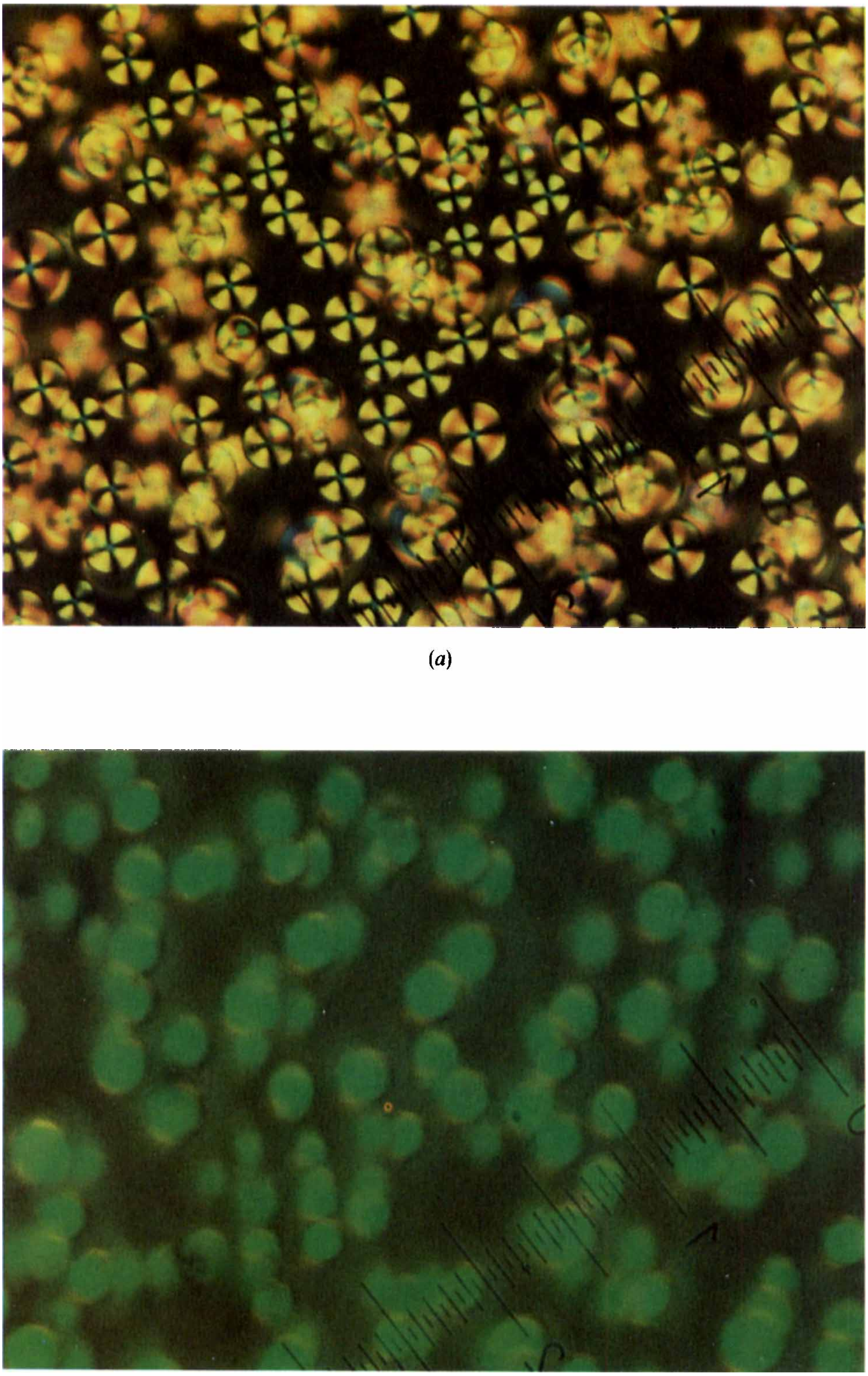


(a)

(b)

Figure 1. The microscopic picture of a sample consisting of 19·1 per cent (by weight) CE2, 30·5 per cent ZLI 2806 and 50·4 per cent PVB ($T = 80{\cdot}0°C$). (a) $E_{rms} = 0$ (transmission, crossed polarizers), (b) $E_{rms} = 113{\cdot}5\ V/31{\cdot}2\ \mu m$ (reflection).

In contrast to the nematic polymer-dispersed liquid crystal displays, which allow switching between an opaque and a transparent state, polymer-dispersed cholesteric liquid crystal devices exhibit strong colour effects. This paper is devoted to the colorimetric properties of these devices. We have measured the dependence of the reflectivity spectrum on temperature, electric field strength, angle of light incidence, and chirality of the liquid crystal. We have also characterized the colour response of the display.

According to the CIE convention [8], the colour impression of a human standard observer can be described by the chromaticity coordinates $(x, y)$. From the reflected (or transmitted) intensity $\varphi(\lambda)$ of the respective object, the values

$$\left.\begin{aligned} X&=k\int_{380\,\mathrm{nm}}^{780\,\mathrm{nm}}\varphi(\lambda)\bar{x}(\lambda)\,d\lambda,\\ Y&=k\int_{380\,\mathrm{nm}}^{780\,\mathrm{nm}}\varphi(\lambda)\bar{y}(\lambda)\,d\lambda,\\ \text{and}\qquad Z&=k\int_{380\,\mathrm{nm}}^{780\,\mathrm{nm}}\varphi(\lambda)\bar{z}(\lambda)\,d\lambda \end{aligned}\right\} \quad (2)$$

can be calculated, where $\bar{x}(\lambda)$, $\bar{y}(\lambda)$ and $\bar{z}(\lambda)$ are the spectral tristimulus values [8]. By normalization, the chromaticity coordinates $(x, y)$ are obtained

$$x=X/(X+Y+Z), \qquad y=Y/(X+Y+Z). \quad (3)$$

Many colours in the visible region are obtainable by additive mixing of three colours, provided that the chromaticity coordinates of these three colours are close to those of monochromatic light at suitable wavelengths. In order to characterize our system with respect to possible applications, we have determined the chromaticity coordinates from the respective reflectivity spectra. Finally, we discuss the implications of our results for the use of polymer dispersed cholesteric liquid crystals in multicolour displays.

## 2. Experiment

The material under investigation consists of 50 per cent (by weight) of poly(vinylbutyral) (PVB; Aldrich, Germany), $x$ per cent of the chiral monomer CE2 (Merck, Poole, Great Britain) and $(50-x)$ per cent of the wide temperature range nematic mixture ZLI 2806 (Merck, Germany) exhibiting negative dielectric anisotropy. The concentration of the chiral component was varied between $x=15$ and $x=22$.

Samples were prepared by first mixing the components together with chloroform. Glass spheres with a diameter of 9·7 μm or 31·2 μm were added to establish the sample thickness. A glass slide coated with electrically conducting ITO layer was covered by the mixture and the chloroform allowed to evaporate for several hours. Then the sample was heated to 130°C in order to complete the evaporation of the solvent and the sample was covered by the second ITO-coated glass slide. In order to obtain drops of uniform size (7·5 ± 1·2 μm), the sample was cooled down from 130°C to 90°C at a constant cooling rate of 2°C $\mathrm{min}^{-1}$. The temperature of the sample was regulated using a pulse-width controlled PID thermostat (Instec/Apple IIe) with an accuracy better than ±0·01 K. For microscopic investigations, a Zeiss Universal microscope was used in reflection mode with crossed polarizers. A Jarrel Ash monochromator and photomultiplier were connected to the microscope tube in order to measure the reflected intensity spectrum.

For macroscopic studies, and for investigating the angular dependence of the selective reflection, a multichannel spectral analyser PR 702 (Photo Research) with a spectral resolution of approximately 1 nm was used. The optical head was adjusted to investigate the light reflected from a spot at the sample surface with approximately 1 mm diameter. 1 kHz alternating voltages were applied to the sample; reported voltages are in rms.

## 3. General properties and electro-optic characteristics

The samples all showed characteristic cholesteric droplets in the temperature range between about 67°C and about 105°C, with transition temperatures depending only slightly on the chirality of the sample. Application of AC electric fields above a threshold of $E_0 \approx 2$–$3\ \mathrm{V\,\mu m^{-1}}$ lead to the appearance of selective reflection colours. The intensity $I$ of the reflected light at the selective reflection wavelength increased continuously with increasing field strength until saturation (see figure 2). Due to the small slope of the $I$(V) characteristic, the intensity of the reflected light could be controlled by the field strength. A hysteresis of 0·4–0·7 V $\mathrm{\mu m^{-1}}$ was observed.

The reflection peaks show a spectral halfwidth of typically 40–50 nm (figure 2). No significant shift of the selective reflection wavelength with increasing voltage was observed within our experimental accuracy, which indicates that the electro-optic effect

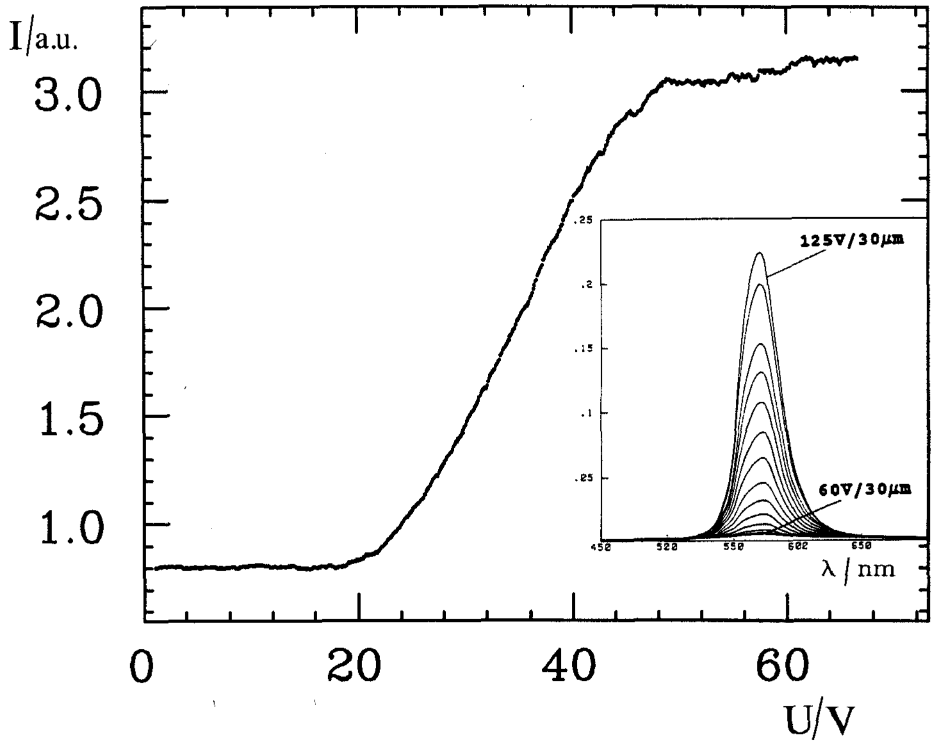


Figure 2. Intensity at $\lambda_0 = 452$ nm versus voltage for a sample consisting of 22·0 per cent CE2, 28·0 per cent ZLI 2806 and 50·0 per cent PVB ($T = 95{\cdot}0$°C, $d = 9{\cdot}7\ \mathrm{\mu m}$). Inset: reflectivity spectra at 5 V/30 µm increments for a sample consisting of 18·0 percent CE2, 31·9 per cent ZLI 2806 and 50·1 per cent PVB ($T = 90{\cdot}0$°C, $d = 30\ \mathrm{\mu m}$).

is only due to reorientation and that no electrostrictive effects occur. Microscopic observations show that the selective reflection originates from a region with its helix axis uniformly oriented along the field direction in the center of each droplet. The diameter of these regions grows with increasing field strength. This behaviour is in agreement with microscopic investigations on droplets with large pitch [3].

The colour of polymer-dispersed cholesteric liquid crystal samples can be controlled by the chirality of the liquid crystal. By varying the weight fraction $c$ of CE2 in the liquid crystal, we found (see figure 3) that the dependence of the cholesteric pitch $p$ (and thus, according to equation (1), the selective reflection wavelength) on chirality can be described by the relation

$$pc = \text{constant}, \qquad (4)$$

which is valid for many of the usual cholesteric systems [9], and especially for chiral–racemic mixtures.

The behaviour of the selective reflection wavelength for the particular material used in this study was strongly affected by the presence of a smectic A phase in the temperature range below the cholesteric phase. In the temperature region close to the clearing point, the cholesteric pitch depended only slightly on temperature; however, close to the smectic transition it diverged strongly (see figure 4). This behaviour is well-known for most conventional cholesteric systems [10]. As described in § 5, we used this strong temperature dependence to determine the chromaticity coordinates corresponding to selective reflection peaks throughout the visible wavelength range for the same sample.

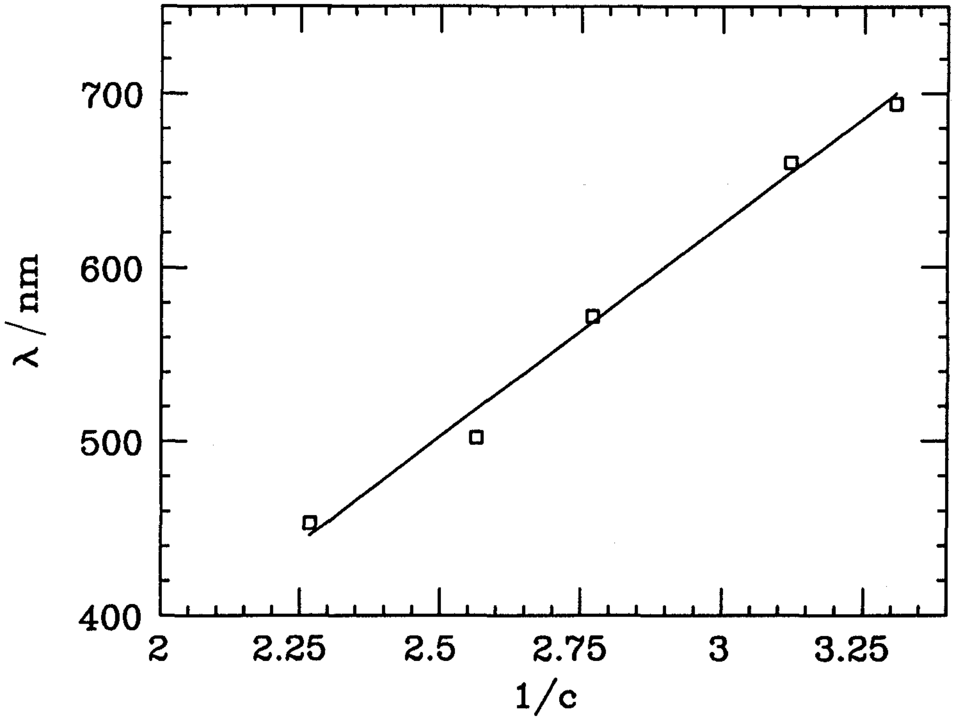


Figure 3. The influence of the chirality on the wavelength of selective reflection. Composition: 50 per cent PVB + 50 per cent momomer; $c$ = weight fraction of CE2 in the liquid crystal mixture CE2/ZLI 2806.

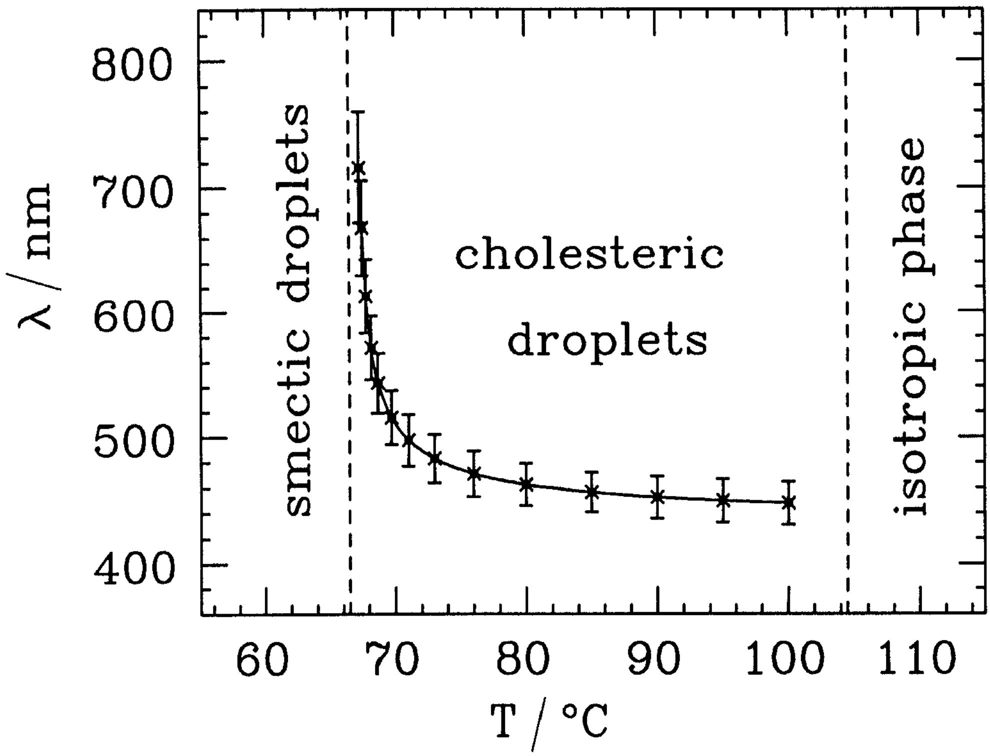


Figure 4. The dependence of the Bragg wavelength on temperature for a sample of 22·0 per cent CE2, 28·0 per cent ZLI 2806 and 50·0 per cent PVB ($E_{\mathrm{rms}} = 60\,\mathrm{V}/9{\cdot}7\,\mu\mathrm{m}$).

## 4. Angular dependence of the selective reflection

In analogy with Bragg's law, the angular dependence of the selective reflection wavelength can be described by the relation

$$\lambda = \lambda_0 \cos\theta' = \lambda_0 \sqrt{(1 - \sin^2\theta/n^2)}, \tag{5}$$

where $\lambda_0$ is the selective reflection wavelength given by equation (1), $\theta'$ is the angle of the incident and reflected beams with respect to the helix axis in the sample, and, since the electric field and the helix axis are both parallel to the surface normal, $\theta$ is the external angle of incidence and reflection with respect to the surface normal. We have measured the angular dependence of the reflectivity for different temperatures; the results are shown in figure 5. The solid lines are fits to equation (5), corresponding to a mean refractive index of $n = 1{\cdot}42 \pm 0{\cdot}14$. The latter value is smaller than expected, but within its error limits this fitting parameter is still close to the value of $n \approx 1{\cdot}5$ expected for the liquid crystal.

## 5. Chromaticity diagram

From the reflectivity spectra measured with the microscope for a sample consisting of 22 per cent CE 2, 28 per cent ZLI 2806 and 50 per cent PVB, we have calculated the chromaticity coordinates (see figure 6) according to equation (2) and (3) using the spectral tristimulus values given by the International Commission of Illuminance in the 1935 convention [8]. The chromaticity diagram (see figure 6) shows the chromaticity coordinates obtained for different temperatures, which vary due to the strong

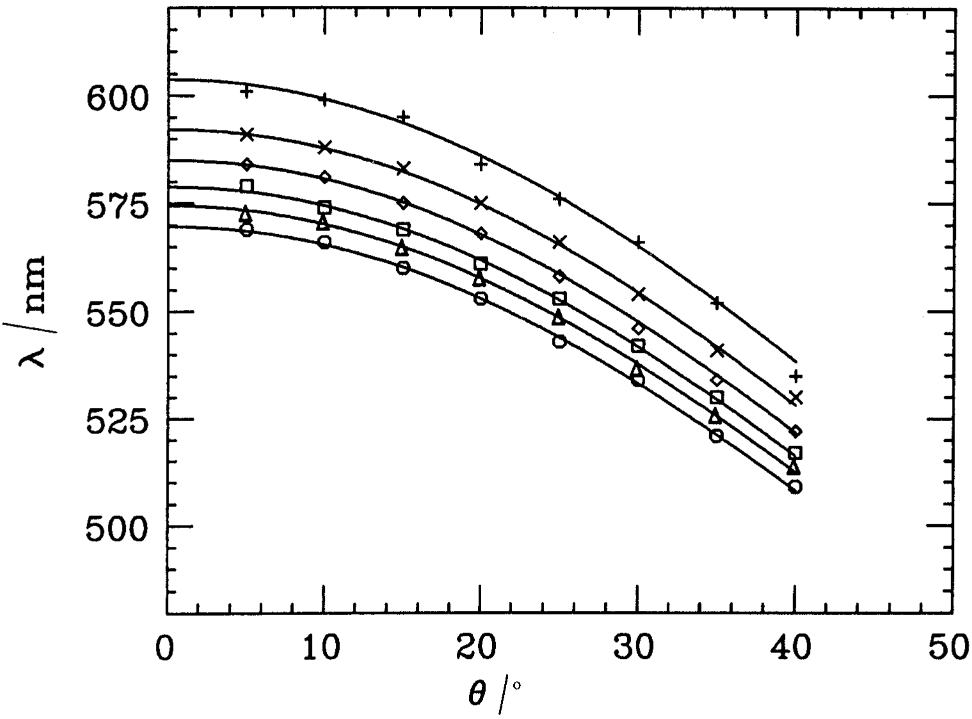


Figure 5. The angular dependence of the selective reflection for a polymer-dispersed cholesteric liquid crystal sample consisting of 18·0 per cent CE2, 31·9 per cent ZLI 2806 and 50·1 per cent PVB. Lines are fits to equation (5). +, 80°C; ×, 83°C; ◇, 86°C; □, 90°C; △, 95°C; ○, 100°C.

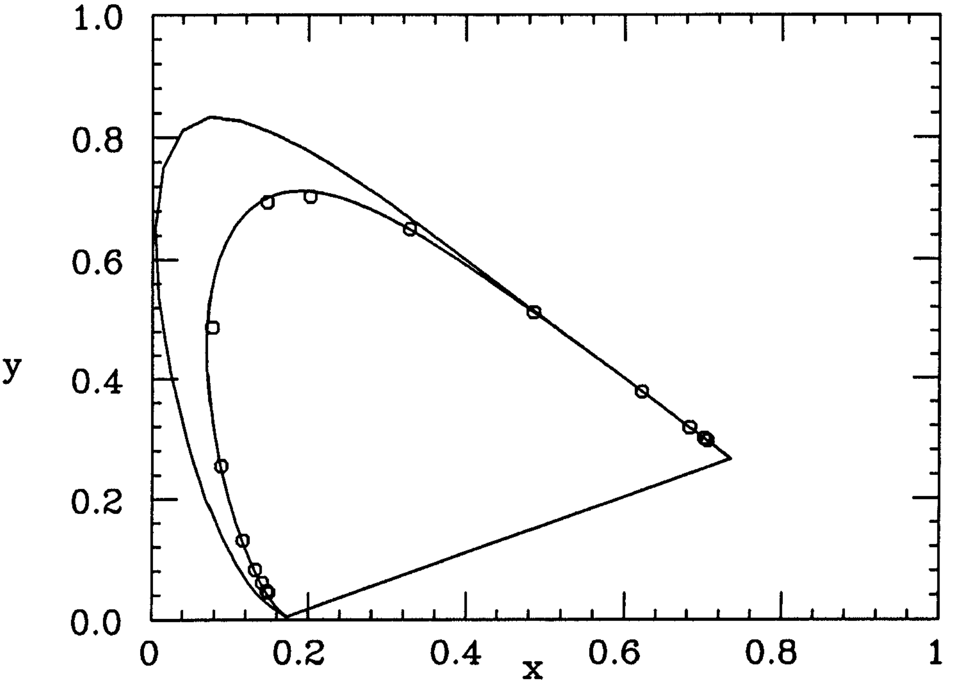
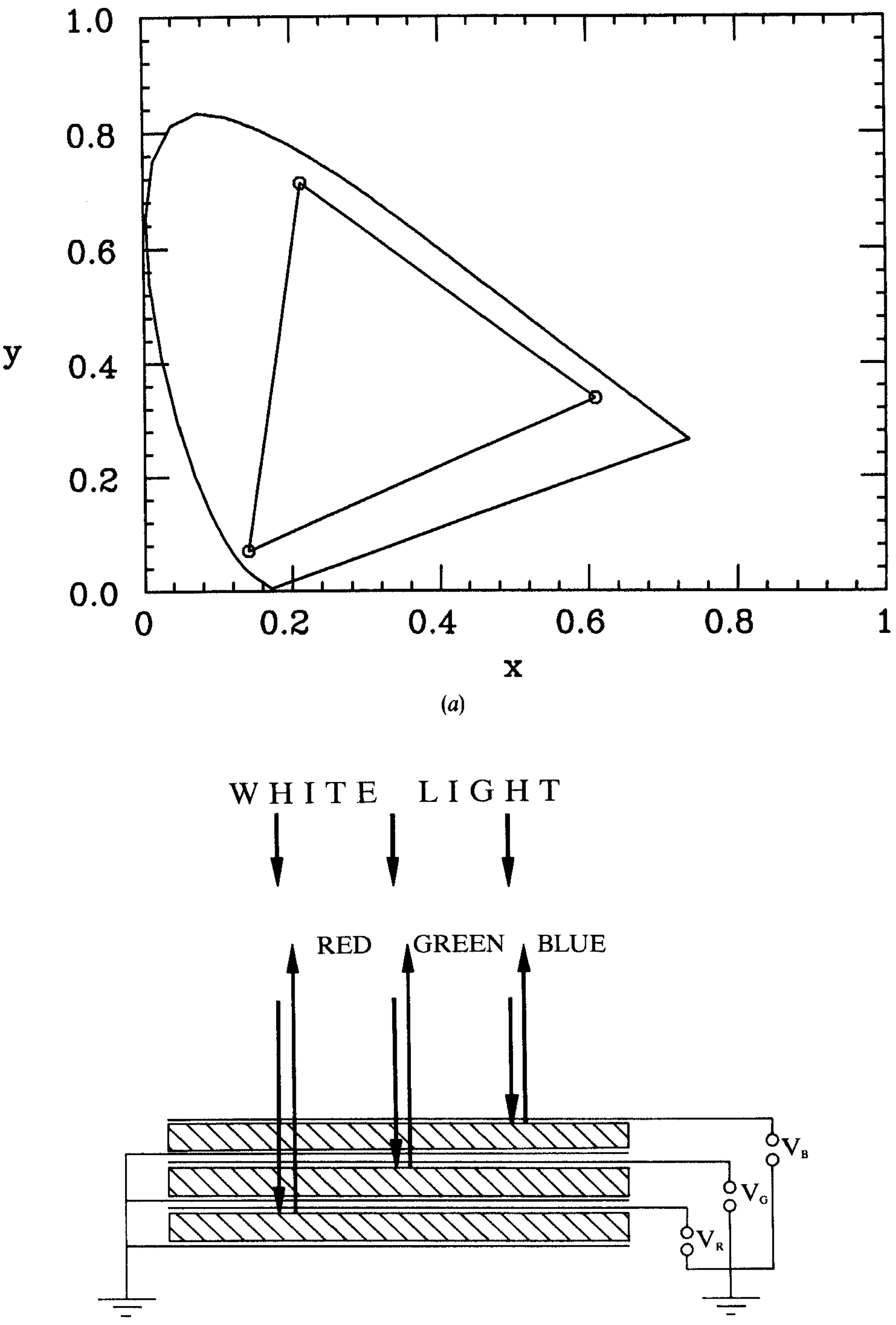


Figure 6. Chromaticity diagram representing the chromaticity coordinates for the sample containing 22 per cent CE2 at different temperatures (see figure 4).



(*a*)

(*b*)

Figure 7 (*a*) Chromaticity coordinates for three samples with different chiralities [*c* per cent CE2, (50-*c*) per cent ZLI 2806 and 50 per cent PVB] at $T=90°\mathrm{C}$, $E_{\mathrm{rms}} \approx 6\,\mathrm{V}\,\mu\mathrm{m}^{-1}$. The chiral concentration is $c = 15{\cdot}1$ per cent (red), 19·6 per cent (green) and 22·0 per cent (blue), respectively. (*b*) Multicolour display using three polymer-dispersed cholesteric liquid crystal sheets with different colours.

temperature dependence of the Bragg wavelength. The lines connecting the experimental values correspond to gaussian line shapes with a half-width of 50 nm ($\sigma = 30{\cdot}03$ nm).

Note that the experimental values enclose a large part of the area of the chromaticity diagram. This indicates that a large part of the visible spectrum can be obtained by additive colour mixing. If light from two sources exhibiting the spectra A and B are mixed, the corresponding values ($X_A$, $Y_A$, $Z_A$) and ($X_B$, $Y_B$, $Z_B$) in equation (2) have to be added and the resulting chromaticity coordinate ($x$, $y$) given by equation (3) is an arithmetic average of the chromaticity coordinates ($x_A$, $y_A$) and ($x_B$, $y_B$) of the two sources. As a consequence, all chromaticity coordinates on a straight line between ($x_A$, $y_A$) and ($x_B$, $y_B$) are accessible if the intensities of the two sources can be controlled separately. This principle is well-known and widely used in multicolour displays (such as TV screens) where a large triangle in the chromaticity diagram is accessible by adjusting the intensities of three basic colours.

In order to use polymer-dispersed cholesterric liquid crystal displays for this purpose, the three basic wavelengths should be obtained by variation of the chirality rather than by temperature. Figure 7 shows the measured chromaticity coordinates for three mixtures with different concentration of the chiral compound CE2 at the same temperature. We find that a large triangle within the chromaticity diagram is accessible by additive colour mixing. Note that a single polymer-dispersed cholesteric liquid crystal sample is transparent for all wavelengths not contained in the relatively narrow selective reflection peak. Therefore, in principle, three polymer-dispersed films, controlled by three separate voltages, can be stacked in order to give a multicolour display which works in reflection.

A multicolour, nematic polymer-dispersed liquid crystal display has been proposed by Doane *et al.* [11]. The authors suggest sending three white light beams through different colour filters, adjusting their intensities by voltage control of the transparency of a polymer-dispersed liquid crystal device, and recombining the three beams in order to get full colour projection. In contrast to this device, a polymer-dispersed cholesteric liquid crystal display would require neither beam splitting nor use of colour filters. Each polymer-dispersed cholesteric liquid crystal layer is a wavelength selective reflector with controllable reflectivity.

## 6. Conclusions

We have shown that polymer-dispersed cholesteric liquid crystals are well suited for multi-colour displays. Due to the narrow spectral half-width of the selective reflection peaks, their chromaticity coordinates are very close to those of monochromatic light. As a consequence, a large area of the chromaticity diagram is accessible by additive colour mixing of three colours. The respective wavelengths of maximum reflectivity can be adjusted by variation of the chirality. In principle, this variation can be done without affecting the transition temperatures by changing the ratio of two enantiomers in a chiral–racemic mixture. Due to the small slope of the electro-optic characteristics, the intensity of selective reflection of a polymer-dispersed cholesteric liquid crystal sample can be controlled by the applied voltage. On the other hand, due to this characteristic these displays are not favourable for multiplexing.

We have also found that the selective reflection wavelength decreases with increasing angle of observation with respect to the surface normal, as expected from Bragg's law. This behaviour is certainly a disadvantage for conventional displays, but the opportunity to control the colour by the angle of light incidence may be useful for special applications.

Investigations on the dynamic behaviour of polymer-dispersed cholesteric liquid crystal displays are published elsewhere [12]. The shortest switching times reported so far have been a few tenths of a second [1, 12]. It has been shown [12] that high voltages can speed up the switch-on time but may also stabilize a field-induced texture which decays rather slowly when the voltage is turned off. Small droplet sizes hasten both the switch-on and the switch-off times. However, it should be noted that the droplet size has to be no less than the order of the cholesteric pitch (0·3–0·5 μm) in order to obtain selective reflection. With decreasing drop diameter, the maximum reflectivity is expected to decrease while the half-width is expected to increase. Therefore, compromises in the drop size have to be found in order to realize both bright colours and sufficiently short switching times.

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