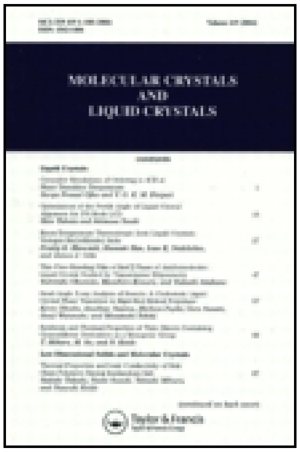

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An Investigation of Pre-Transitional Effects in a Thin Liquid Crystal Layer via the Excitation of ps Mixed Optic Modes

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AN INVESTIGATION OF PRE-TRANSITIONAL EFFECTS IN A THIN LIQUID CRYSTAL LAYER VIA THE EXCITATION OF ps MIXED OPTIC MODES

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Abstract The pre-transitional Kerr effect in K18 is examined using a novel technique. Mixed polarisation grating coupled guided modes are excited within the liquid crystal layer, these are probed for small changes in position using a differential technique which resolves induced birefringences to an accuracy of 5×10^{-8}. The experiment is performed at several different temperatures in order to characterize this pre-transitional behaviour, and from this, one of the Landau expansion coefficients can be found.

INTRODUCTION

Theory

The pre-transitional Kerr effect is exhibited by liquid crystal materials which are in their isotropic phase at a temperature which is close to the nematic/isotropic (N/I) phase transition temperature. When a field is applied to the material, an induced birefringence develops in the direction of the applied field. The linear variation of this induced birefringence with the square of the applied field characterizes the

Kerr response of the cell, the Kerr coefficient being:

$$K = \frac{\delta n}{\lambda E^2} \tag{1}$$

where λ is the wavelength of the incident radiation, E is the applied field, and δn is the induced birefringence. In many cases the Kerr response of a liquid crystalline material is an order of magnitude larger than that of other non mesomorphic Kerr liquids. For example nitrobenzene has a Kerr constant of 5×10^{-12} m V^{-2} whilst the liquid crystalline material p-[(p-methoxy benzylidene) amino]-benzonitrile, (MBAB), has a Kerr coefficient of 115×10^{-12} m V^{-2} at 0.3°C above its N/I phase transition [1]. This may be because in non liquid crystalline materials the Kerr effect is generally an electronic one, caused by the interaction of the external electric field with the electron wavefunctions of the sample. However in liquid crystalline materials the applied field induces order on the mesoscopic rather than the microscopic level, suppressing the 'director' fluctuations in the isotropic phase and causing some degree of nematic-like ordering. Indeed the formation of small nematic droplets within the isotropic fluid under an applied field is the basis for some light scattering experiments in the isotropic phase [2]. In general, the closer the temperature of the liquid crystal to the N/I phase transition, the more easily suppressible the fluctuations become - hence the rapid variation of the Kerr coefficient with temperature in liquid crystals.

The theory of Landau and De Gennes has been used to model pre-transitional behaviour in the isotropic phase. The brief summary of the theory which follows is taken from Vertogen and de Jeu [3]. The expression for the Helmholtz free energy of a nematic liquid crystal is given by the Landau expansion:

$$f = f_i + \frac{1}{3}AS^2 - \frac{2}{27}BS^3 + \frac{1}{9}CS^4 \tag{2}$$

where f_i is the Helmholtz free energy of the isotropic phase, S is the order parameter, and A, B, and C are the Landau expansion coefficients, all are functions of pressure and temperature. The additional free energy density f_E provided by an electric field is:

$$f_E = -\frac{1}{3}\epsilon_o \delta\epsilon_{ac} S E^2 \tag{3}$$

where $\delta\epsilon_{ac}$ is the low frequency dielectric anisotropy in the perfectly aligned phase. In the isotropic phase with the field applied, $S \ll 1$ so the high order terms in S can be neglected and minimising f with respect to S gives:

$$S = \frac{\epsilon_o \delta\epsilon_{ac} E^2}{2a(T - T_c^*)} \tag{4}$$

where $a = A/(T - T_c^*)$ is the temperature independent Landau coefficient, and T_c^* is a temperature just below the N/I phase transition. The induced birefringence, δn is given by:

$$\delta n = \frac{\delta\epsilon_{max} S}{2\sqrt{\bar{\epsilon}}} \tag{5}$$

where $\bar{\epsilon}$ is the optical permittivity of the isotropic liquid crystal and $\delta\epsilon_{max}$ is the optical dielectric anisotropy in the perfectly aligned phase. Hence we have:

$$\frac{E^2}{\delta n} = \frac{4a\sqrt{\bar{\epsilon}}(T - T_c^*)}{\epsilon_o \delta\epsilon_{max} \delta\epsilon_{ac}} \tag{6}$$

From this expression, it is clear that the relationship between the reciprocal Kerr coefficient and temperature is linear. This relationship has been verified by a number of workers [1,4,5], although some liquid crystals do not follow this form [6,7]. When a linear relationship is established it is possible to find a value for a and hence the order parameter of the liquid crystal for a given temperature is determined.

Experimental Techniques

Numerous optical studies have been made of the pre-transitional behaviour in isotropic liquid crystalline materials. Many of these studies use polarising microscopy and ellipsometric techniques to determine the induced birefringence as a function of the field applied [4,6]. In this work, the effect is measured using diffraction grating coupling to optic modes excited within the liquid crystal layer. The liquid crystal forms the high index waveguiding medium between a silver coated diffraction grating and a glass top plate (see figure 1). Incident light may be diffracted by the grating and there is a range of angles of incidence, θ, for which the +1 diffracted beam undergoes total internal reflection at the liquid crystal top plate interface. In this regime, the Q of the system is high and the guided modes excited in the layer are strongly resonant. These resonances manifest themselves as sharp dips seen in the p polarised (transverse magnetic) or s polarised (transverse electric) reflectivity as the angle of incidence is scanned. Indeed the form of this reflectivity curve has been used in prism coupled systems to determine the director profiles of both uniform and non-uniform liquid crystal cells [8,9,10].

If the diffraction grating is now adjusted so that the grating grooves are neither parallel nor orthogonal to the plane of incidence (as in figure 1), then this broken symmetry gives rise to p to s polarisation conversion which is strongly

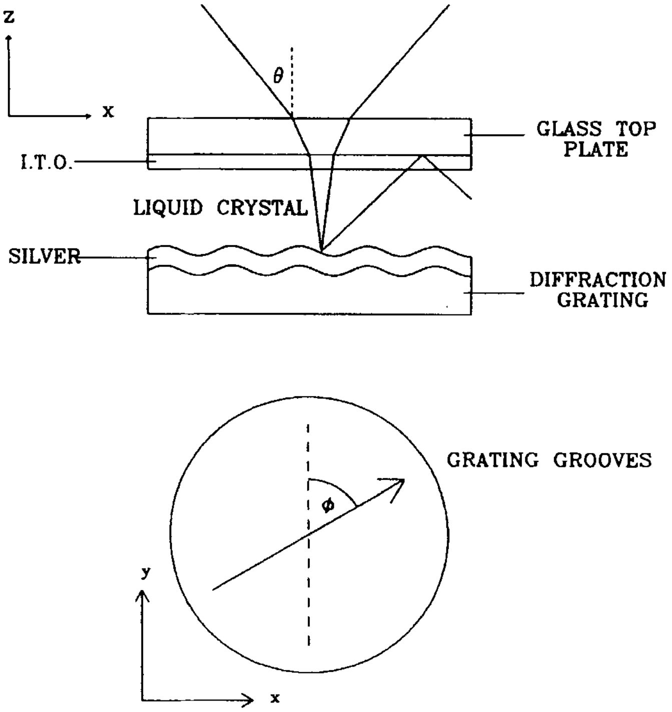

FIGURE 1 Diagrams of the experimental arrangement.

enhanced in the region of the guided modes. Thus with incident p polarised light and a crossed polariser in front of the detector, peaks are seen in the reflectivity as θ is scanned (see figure 2). These peaks have a half width at half maximum of between 0.025 and 0.04 degrees, depending on the uniformity of the thickness of the liquid crystal layer.

The magnitude of these reflectivity maxima varies as the azimuthal angle of the grating is varied and can be maximised for each guided mode [11]. These features are very sensitive to changes in the dielectric tensor configuration and if a differential technique is used, similar to Cross et al.[12], then the birefringences induced by the applied field can be determined to an accuracy of 5×10^{-8}.

EXPERIMENTAL AND RESULTS

A 4.5 μm layer of K18 liquid crystal was confined between a 551.1 nm pitch diffrac-

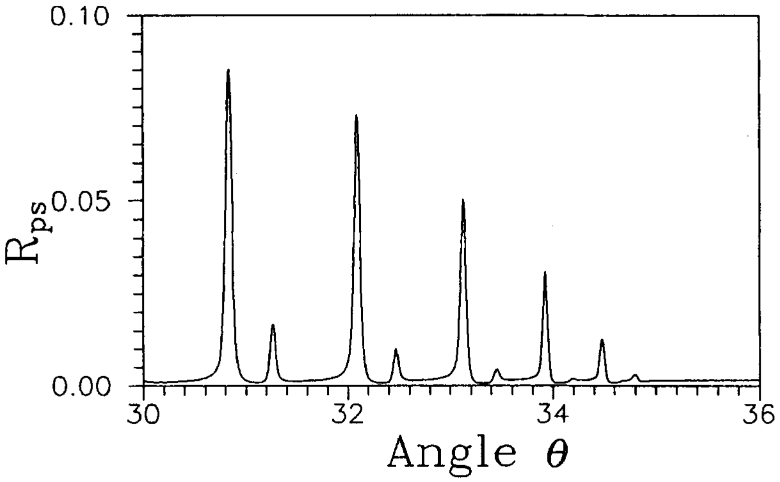

FIGURE 2 ps reflectivity as a function of angle.

tion grating coated with 100 nm of silver and a glass top plate which had been coated with 40 nm of indium tin oxide (ITO) in order to form the transparent top electrode. No aligning layers were present in the cell. The cell was housed in a lagged box which kept the temperature stable to ±0.1°C and hence the temperature was easily adjusted in the range T_c to T_c+4.5°C, where T_c is the nematic to isotropic phase transition temperature.

P polarised light of wavelength 632.8 nm incident on the glass top plate was used to couple to the resonant guided modes and a polariser was placed in front of the detector which only allowed s polarised light through. Scanning the reflectivity (termed R_{ps}) revealed the sharp peaks mentioned before, and the grating azimuthal angle, ϕ, was then adjusted in the x-y plane so that the grating grooves made an angle of 50° with respect to the y-axis. This maximised the reflectivity peak in the TM_5 mode.

A sinusoidal voltage of frequency 900 Hz was applied to the cell along the z direction between the ITO and silver electrodes, and the isotropic liquid crystal responded by developing an induced birefringence in phase with the applied field. The degree of birefringence induced by a given field depended on the temperature of the sample at the time and hence the experiment was performed at four different temperatures within the range previously quoted. This induced birefringence in turn affected the angular positions of the modes being studied, and hence, for a given angle a given modulation of the reflectivity signal, R_{ps}, was seen. This modulation had a frequency, 2ω, where ω is the frequency of the applied field. This is because the liquid crystal material under investigation has no effective permanent dipole moment and therefore responds to the magnitude of the field applied but is insensitive to its direction.

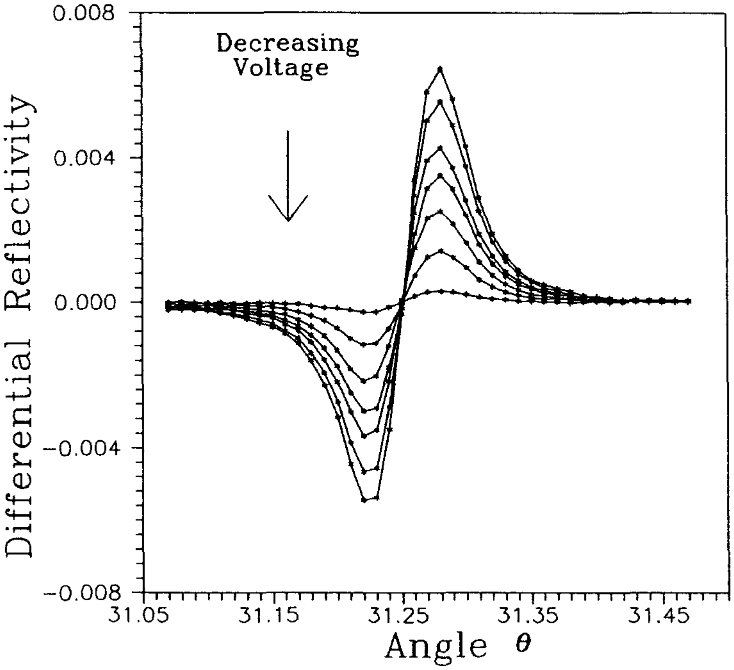

FIGURE 3 Differential reflectivity as a function of angle.

By using an appropriate lock-in amplifier to detect the 2ω signal, it was possible to obtain measurements of the differential reflectivity with θ. The TM_5 mode was studied in this way for applied root mean square (RMS) fields ranging from 0.02 V μm^{-1} to 0.2 V μm^{-1} (see figure 3). Note that the frequency was chosen to be low enough for the liquid crystal to respond in phase with the field. If the frequency is too high then the liquid crystal responds to the RMS field by developing a constant, unmodulated birefringence which is hence not detectable differentially.

The experimental results were then compared with the differential reflectivity obtained when a known shift (0.01°) is imposed on the zero field reflectivity data. In this way the angular shift in the mode position due to the applied field can be calculated. Then using the theory of Azzam and Bashara [13] the zero field R_{ps} data was modelled to obtain the thickness and real permittivity of the isotropic liquid crystal. From this modelling, the change in mode position due to the field could be directly related to the induced birefringence. Note that the theory referred to does not take into account the small perturbation of the mode positions due to the presence of the diffraction grating, however the error introduced by this is very small.

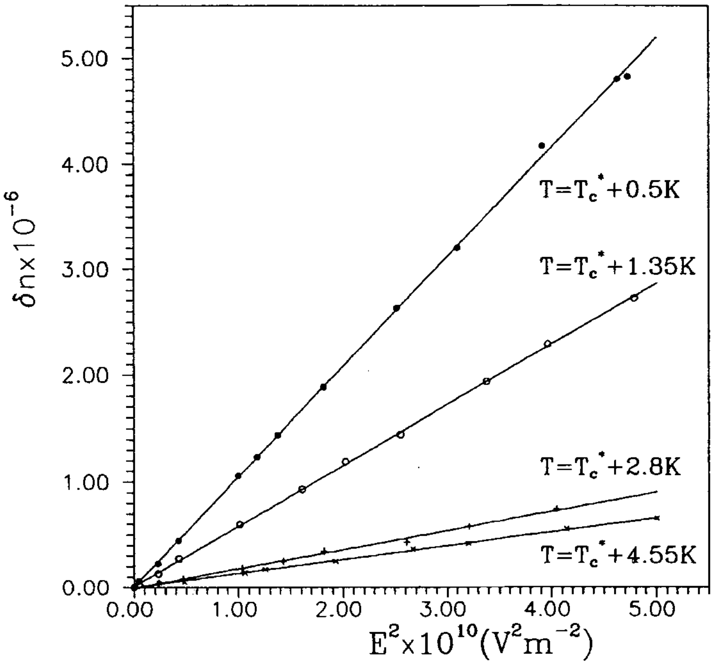

FIGURE 4 Linear variation of δn with E^2 for four different temperatures.

Figure 4 shows the induced birefringence as a function of the square of the RMS field for the four different temperatures at which the experiment was performed.

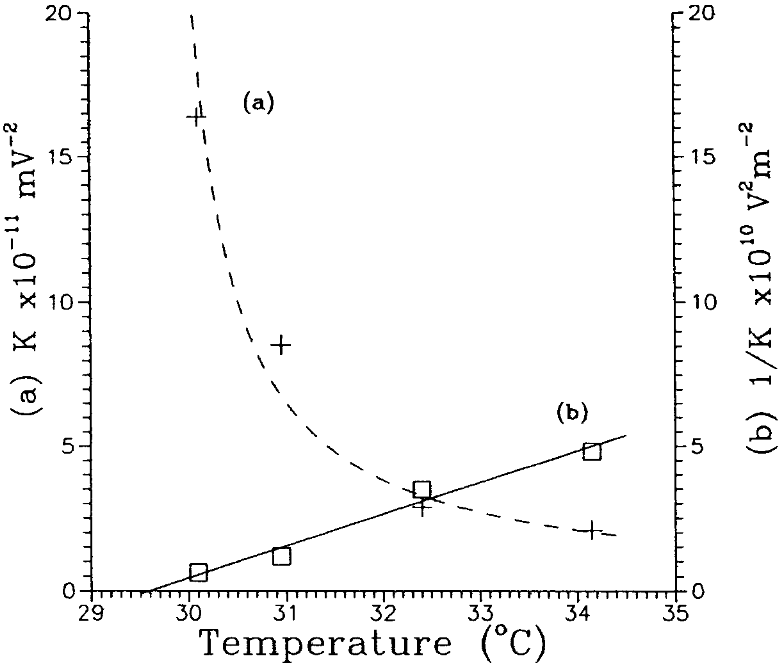

FIGURE 5 Variation of the Kerr coefficient with temperature.

The birefringence of the cell is clearly quadratic in field.

Figure 5 shows the dependence of the Kerr coefficient, K, and the reciprocal of the Kerr coefficient on the temperature, and it is clear that the relation predicted by Landau De Gennes theory is accurate for this material. The gradient of the $1/K$ versus T graph is 1.1×10^{10} V ^{2}m^{-1}, and this value, together with the value for $T_c^* = 29.6°$C also found from the graph, is used in equation (6) to determine the temperature independent parameter, a, for this material. Note that the value of $\bar{\epsilon}$ varies slightly with temperature so an average value of $\bar{\epsilon} = 2.486$ is used. The values for $\delta\epsilon_{max} = 0.520$ at $20°C$ and 632.8 nm and $\delta\epsilon_{ac} = 11.0$ at 1 KHz and 20°C were provided by Merck UK Ltd [14]. From these figures, a was found to be 139×10^3 J m^{-3} K^{-1}. This value compares favourably with n-(p-methoxy benzylidene)-p-butylaniline, MBBA, for which $a = 42 \times 10^3$ J m^{-3} K^{-1}. Using this figure in equation (4), S can be found for a given temperature and field. It is of the order 10^{-4}.

CONCLUSIONS

It is clear from the results that this technique is well suited to monitoring small changes induced in a liquid crystal cell by an a.c. electric field. The use of the ps mixed guided modes which are sharply defined features allows very good resolution, limited only at the moment by the signal to noise ratio of the detected signal. In addition the technique allows in situ measurement of the thickness and dielectric function of the material, to a fair degree of accuracy. The pre-transitional response of the material is adequately modelled by Landau de Gennes theory and using the theory, one Landau expansion coefficient, a, has been determined for for the material.

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