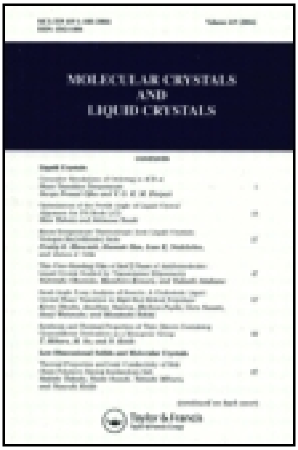

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Thermal Diffusivity Measurements in Lyotropic Ferronematics: Mode Mismatched Thermal Lens

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The mode mismatched thermal lens technique was used in the determination of thermal diffusivity of oriented lyotropic liquid crystal doped with ferrofluid. It was found that the behavior of the parallel thermal diffusivity as a function of ferrofluid content could not be explained in terms of the order parameter alone. Additional contribution coming from the ferrofluid itself seems to be necessary for the interpretation of the experimental data.

Keywords: thermal diffusivity; thermal lens; ferronematics

INTRODUCTION

There has been a growing interest in using photothermal techniques to measure optical and thermal properties of highly transparent materials. These methods are advantageous when compared with conventional thermometry because they allow the experiments to be performed without the difficulties of obtaining the steady state conditions. Thermal lens spectrometry is a transient technique that has been increasingly used to investigate the thermal transport properties of transparent materials. Thermal lens techniques are also non-contact, thus removing the requirement for perfect thermal contact between the measuring device and the sample. The thermal lens effect is created when the excitation laser beam passes through the sample and the absorbed energy is converted into heat, changing the optical path length, and producing a lens-like optical region in the sample. The propagation of a probe beam laser through the thermal lens will result in either a spread or a focusing of the beam center, depending on the sample properties. The sample optical absorption coefficient (A_e) and its thermal conductivity (k) mainly control the thermal lens signal intensity.

Earlier experiments using thermal lens spectrometry in liquid crystals used the so-called single beam configuration. Recently, the use of the mode mismatched thermal lens configuration [1,2] in the investigation of lyotropic liquid crystals was demonstrated [3]. The higher sensitivity of the technique allowed the determination of the thermal parameters of the sample without any addition of dyes destined to enhance optical absorption.

EXPERIMENTAL

The higher sensitivity for the mode-mismatched configuration is achieved when the sample is positioned at the waist of the excitation beam, where the power density is maximum, and at the confocal position of the probe beam. The experimental set up used an Argon ion laser as the excitation beam (λ_e=*514.5nm* and P_e=*10-100mW* at the sample) and a He-Ne laser as the probe beam (λ_p=*632.8nm* and P_p=*1mW* at the sample). A shutter controlled the exposure of the sample to the excitation beam. The output of a fast probe photodiode was coupled to a digital recorder, which was triggered by a second photodiode. Data were transferred from the recorder to the computer and stored in sequential files for further analysis. Each scan resulted in *1000* measured points. Moreover, the excitation beam spot size at the sample was $\omega_e=69\mu m$ and the probe beam spot size $\omega_p=154\mu m$.

Lyotropic nematic liquid crystals are mixtures of amphiphilic molecules and a solvent (usually water), under adequate temperature and concentration conditions. The basic units of these systems are anisotropic micelles. The mixtures investigated in this work were: Sample 1) potassium laurate (*29.4wt%*), decanol (*6.6wt%*), water (*64wt%*); Sample 2) Sample 1 doped with *0.08 wt%* of ferrofluid (Fe_3O_4 dispersed in water); Sample 3) Sample 1 doped with *0.34wt%* of ferrofluid (FF); Sample 4) Sample 1 (second preparation) doped with *0.29wt%* of FF; Sample 5) Sample 1 (second preparation) doped with *0.40wt%* of FF. Ferrofluid is usually added to enhance alignment of the micelles in the presence of magnetic field. The phase sequences of the samples were determined by optical microscopic and conoscopic observations, which showed that: Sample 1 is isotropic up to 15^0C, calamitic nematic from 15^0C to 50^0C and isotropic above 50^0C; the doped samples showed similar behavior with slight differences in the transition temperatures. The samples were encapsulated in quartz cells with thickness of $L=1.0\ mm$. The axes of the directors in the nematic phases were aligned for some hours in a magnetic field (*0.8T*) for orientations both parallel and perpendicular to the sidewalls. The measurements were performed at room temperature.

The thermal diffusivity (parallel and perpendicular to the director of the phase) was determined from the thermal lens measurements performed on the aligned samples. The experimentally observed time profile of the developing thermal lens, $I(t)$, was fitted to Eq. 1, and the thermal diffusivity, α, was obtained from the adjustable parameter t_c:

$$\frac{I(t)}{I(0)}=\left[1-\frac{\theta}{2}\tan^{-1}\left(\frac{2mV}{\left[(1+2m)^2+V^2\right](t_c/2t)+1+2m+V^2}\right)\right]^2 \qquad (1)$$

$I(0)$ is the signal intensity at $t=0$ (when the excitation beam is switched on), and

$$m=\left(\frac{\omega_p}{\omega_e}\right)^2, \qquad V=\frac{Z_1}{Z_C} \qquad (2)$$

Z_l is the position of the sample far from the probe beam waist and Z_C its confocal distance ($Z_C=\pi\omega_p^2/\lambda_p$). The parameters m and V depend on the experimental arrangement, and in this case were $m=5.0$ and $V=1.3$. Besides $I(0)$, θ and t_c are left as free parameters. They are related to the experimental and sample parameters through:

$$\theta=-\frac{A_e P_e L}{\lambda_p k}\frac{dn}{dT}, \qquad t_c=\frac{\omega_e^2}{4\alpha} \qquad (3)$$

where (dn/dT) is the temperature coefficient of the sample refractive index at λ_p.

RESULTS AND DISCUSSION

Figure 1 shows the experimental data and the adjusted curve for Sample 3 with the micelles aligned perpendicular to the sidewalls. As it can be seen, the signal intensity decreases with time, resulting in a positive value for θ. This implies $(dn/dT)<0$, which is characteristic of liquids. The best fit values for this case are: $I(0)=3.40V$, $\theta=0.28$, $t_c=8.52\ ms$. The experiment was repeated at least five times for each sample and orientation. Table I shows the average values of the determined thermal diffusivity for each case.

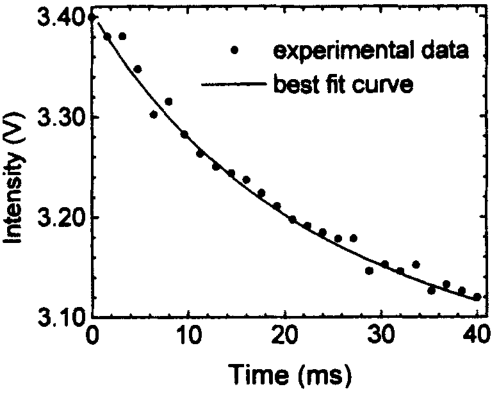

FIGURE 1 Experimental data and the adjusted curve for Sample 3 oriented perpendicular to the sidewalls.

TABLE I Thermal diffusivities for both orientations and the ratio $\alpha_{//} / \alpha_{\perp}$.

Sample	FF content	$\alpha_{//}$ (10^{-3} cm^2/s)	$\alpha_{\perp}$ (10^{-3} cm^2/s)	$\alpha_{//} / \alpha_{\perp}$
1	-	1.69 ± 0.05	1.40 ± 0.03	1.21
2	0.08 wt%	1.40 ± 0.09	1.35 ± 0.13	1.04
3	0.34 wt%	1.25 ± 0.09	1.44 ± 0.14	0.85
4	0.29 wt%	1.58 ±0.04	1.54 ± 0.05	1.03
5	0.40 wt%	1.67 ± 0.10	1.75 ± 0.04	0.95

As it can be seen from Table I, the parallel thermal diffusivity decreases with the addition of ferrofluid for low concentrations. The same trend was found in measurements in lyotropic liquid crystal doped with ferrofluid using mirage effect [4]. Figure 2 shows the thermal diffusivity behavior as a function of ferrofluid concentration using data from Table I.

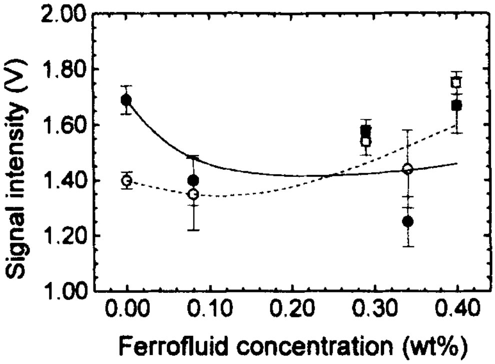

FIGURE 2 Parallel (solid symbols) and perpendicular (open symbols) thermal diffusivities as a function of ferrofluid concentration (circles: 1st preparation; squares: 2nd preparation). Lines are intended to be a guide to the eye.

As mentioned above, the role of ferrofluid in the liquid crystal is to enhance alignment of the micelles in the presence of magnetic field. This means that the order parameter increases with ferrofluid content. In Ref. 3 it was shown that the thermal anisotropy, defined as the ratio of the parallel to the perpendicular thermal diffusivities, could be explained in terms of the geometry of the micelles. This model was deduced by considering that the phonon mean free path is limited by the micelles dimensions. In this case, the interfaces between micelles and the solvent play the dominant role in thermal conductivity or resistivity. By using this simple model in order to explain the thermal behavior of the doped samples one concludes that the thermal anisotropy should increase with the order parameter. The experimental data indicate, however, the contrary.

Now, if we turn our attention to the thermal diffusivity itself, instead of the thermal anisotropy, we found that the model above does not explain its behavior as a function of the ferrofluid content. When the order parameter increases, the parallel thermal diffusivity would be expected to follow the same trend [5]. However, both mirage and thermal lens measurements unambiguously show its decrease, at least for low concentrations of ferrofluid. It seems, therefore, that one must consider the influence of the ferrofluid in the liquid crystal thermal diffusivities not only through a micelle orientation mechanism, but also by taking into account the possible contribution of the ferrofluid particles to the thermal resistance. Nonetheless, additional experimental data for this liquid crystal are needed in order to discriminate among the possible distinct contributions to the heat conduction.

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