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Preliminary communication

Photopyroelectric investigation of the thermal conductivity anisotropy in oriented liquid crystals

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Photopyroelectric measurements of heat transport parallel or perpendicular to the director have been carried out for compounds of the 4,4′-dialkylazoxybenzene homologous series. A theoretical expression based on molecular length–width ratios predicts a thermal conductivity anisotropy ratio which is between 1·5 and 2 times larger than experimentally observed. However, the predicted order parameter temperature dependence seems to be consistent with the experimental data.

Although static thermal properties have been extensively investigated by DSC, AC-calorimetry or adiabatic scanning calorimetry [1], there is much less information on the dynamical thermal properties of liquid crystals. The thermal conductivity of aligned liquid crystals has been proved to be anisotropic, being a maximum (minimum) along (perpendicular to) the optical axis of the molecules. This anisotropy has previously been measured, e.g. by forced Rayleigh Scattering [2], by heat flux methods [3, 4] or by heat pulse methods [5].

Although thermal conductivity anisotropy in aligned liquid crystals is generally accepted, a quantitative explanation of this anisotropy is still lacking. Through different experiments, a correlation with the length–width anisotropy of the molecules seems to be established [2–5]. Molecules with a large length–width ratio tend to have a large thermal conductivity anisotropy. Recently, Bento and co-workers [6] proposed that a sample be considered as an array of thermal resistors of rod-like shape, and showed that the maximum anisotropy in the thermal conductivity, κ, obeys the relation $\kappa_\parallel/\kappa_\perp = \alpha_\parallel/\alpha_\perp \approx L/D$, with L and D the length and diameter of the cylinders, respectively. In fact, they adopted the results from a quasi-lattice model for the ratio between mass diffusion coefficients derived by Chu and Moroi [7]. The ratio between thermal diffusivities, and thus between thermal conductivities, for imperfect nematic order $S<1$ is expected to be:

$$\frac{\alpha_\parallel}{\alpha_\perp} = \frac{\kappa_\parallel}{\kappa_\perp} = \frac{(L/D)(1+2S)+(2-2S)}{(L/D)(1-S)+(2+S)}, \qquad (1)$$

where S is the scalar order parameter. In order to test this equation we determined the thermal conductivity anisotropy of members of a homologous series.

Most of the methods available for thermal conductivity investigations involve rather large temperature gradients over a large liquid crystal sample. Photothermal methods do not involve large temperature gradients and can be applied for very small samples; this makes them very attractive for liquid crystal studies.

We have measured the thermal conductivity by a photopyroelectric method [8, 9]. In this technique a (300 μm thick) pyroelectric transducer ($LiTaO_3$) is used to detect temperature variations at the back of the sample, which is sandwiched between a gold coated (at the sample side) quartz piece and the transducer. Temperature variations are caused by harmonically modulating the laser light intensity (at a wavelength of 633 nm), at a modulation frequency ω, which is absorbed by the gold coating and launches a thermal wave through the liquid crystal. Amplitude and phase, relative to the excitation, are measured with a two-phase lock-in amplifier.

For opaque samples (satisfied by the metallic coating), and for a large quartz–sample effusivity ratio, the complex amplitude of the pyroelectric current is given by [10]:

$$I(C,k) = \frac{I_0(1-R)p}{L_p\rho_p C_p(e_s e_p^{-1}+1)} \exp(-\sigma L) \qquad (2)$$

with I_0 the light intensity, R the reflectivity of the gold layer, e_s the sample effusivity and $\sigma = (1+i)(\omega/2\alpha_s)^{1/2}$,

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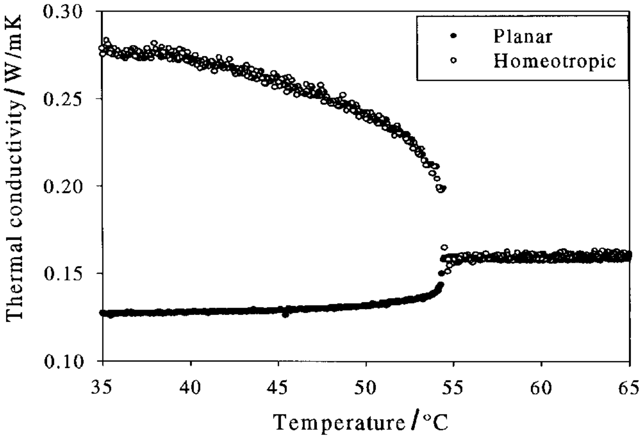

Figure 1. Temperature dependence of the thermal conductivity along and perpendicular to the molecular long axis of 6AB.

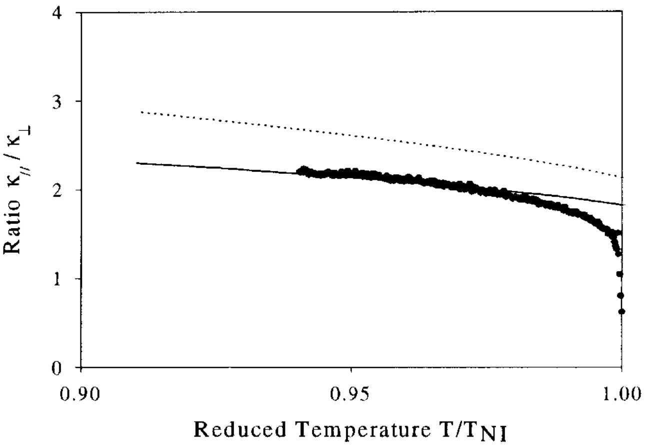

Figure 2. Temperature dependence of the thermal conductivity–anisotropy ratio in 6AB as a function of reduced temperature. Dotted line: predictions of equation (1) with a realistic L/D-ratio of 6; solid dots: experimental values; solid line: fit to the experimental data with $L/D=2{\cdot}7$.

where α_s is the sample thermal diffusivity, and p the pyroelectric coefficient of the transducer. L_P, ρ_P, C_P and e_P are, respectively, the transducer thickness, density, specific heat capacity and effusivity. This expression can be inverted to calculate the sample diffusivity and effusivity from the amplitude and phase of the signal. Sample independent parameters are determined in a calibration procedure.

Samples of thicknesses about 50 μm were used and homeotropic or planar alignment was achieved by surface treatments of the quartz piece and the pyroelectric transducer. Furthermore, a stabilizing DC-magnetic field of about 0·11 T in the proper direction was used to obtain good bulk alignment. In this way both $\kappa_\perp$ and $\kappa_\parallel$ could be measured.

We have investigated compounds of the 4,4′-dialkylazoxybenzene homologous series (nAB), (n=the number of carbon atoms in each terminal alkyl chain=6,7 and 8); these compounds have considerable nematic ranges. Figure 1 shows the thermal conductivity data for 6AB. The values in this figure are scaled to an estimated reference value of $0{\cdot}16\,\mathrm{W\,m^{-1}\,K^{-1}}$ at $T=65°\mathrm{C}$ in the isotropic phase. Variations of 50% of this value resulted only in changes of about 10% in the anisotropy ratio considered further. To compare our results for the thermal conductivity ratio with the predictions of equation (1), we used a Maier Saupe [11] approximation for the scalar order parameter S as a function of reduced temperature, which can be considered a good approximation of the real order parameter for the AB-series [12]. For a length–width anisotropy ratio of 6–7 (for the AB-series the width is about 5 Å, while the length varies from 30 to 37 Å), the thermal conductivity anisotropy ratio predicted by equation (1) is much larger than that experimentally measured. In fact, although the temperature dependence of the order parameter is reflected in the measurements, a L/D-ratio of 3·9 would be more appropriate for 6CB (figure 2). Furthermore, only a rather small increase in anisotropy was noticed for the higher members of the homologous series. The increased descrepancy between theory and experiment on approaching T_{NI} arises from growing deviations between the real and the theoretical (Maier–Saupe) order parameter used in equation 1.

From our results, we conclude that the thermal conductivity anisotropy in aligned liquid crystals cannot be explained in terms of real length–width anisotropies, but that introducing an effective length would be more appropriate. Apparently the thermal anisotropy is mainly carried by the rigid aromatic core anisotropy and not greatly by the length of the (flexible) alkyl chains.

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