Magnetic-Field Dependence of the Order Parameter in a Nematic Single Crystal

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Results on the magnetic-field dependence of the order parameter in a nematic single crystal are reported for the first time: The induced birefringence of a nematic single crystal of 4'-*n*-heptyl-4-cyanobiphenyl was found to vary linearly with the magnetic field as predicted by the fluctuation theory.

In this Letter, we report the first measurements of the dependence of the order parameter on magnetic field in a nematic single crystal. These measurements confirm the validity of de Gennes' theory for orientation fluctuations.

The most fundamental idea which helps elucidate the behavior of a system near a critical point is the concept that the transition may be described by an order parameter S which represents a numerical measure of the amount and kind of ordering built up in the neighborhood of the critical point.[1] For example, in a single-domain ferromagnetic crystal, with an easy axis of magnetization along the z direction, a suitable order parameter is the statistically averaged z component of the magnetization $\vec{M}$. Ferromagnetic critical phenomena have been largely studied and the dependence of the order parameter on the temperature T or on the magnetic field $\vec{H}$ near the Curie point T_C has long been known: Whereas M is proportional to H above T_C in the paramagnetic phase, M has a $H^{1/\delta}$ variation (with $\delta = 4$ to 5) for $T \approx T_C$. In the nematic phase of a liquid crystal, the order parameter can be studied by measurement of the diamagnetic anisotropy χ_a, or the dielectric anisotropy ϵ_a, or the birefringence Δn.[2-7] The temperature dependence of S has been determined for various nematics in the last few years, but, to our knowledge, its dependence on the magnetic field has never been experimentally studied. However, knowing of the field dependence of S in the nematic phase and near the clearing point could be important elements for the general study of critical phenomena. The first results of this work are given in this Letter.

The effect of a magnetic field on the birefringence of a nematic liquid crystal has been discussed by de Gennes from the fluctuation theory for nematics.[8] It is well known that fluctuations of the alignment in nematics strongly influence the macroscopic properties of these media. Chatelain's experimental studies[9] had given a direct probe of the spontaneous fluctuations of the alignment in nematics. Such a phenomenon has been rigorously described by de Gennes in terms of small fluctuations of the director n parallel to the local optical axis. The effect of a magnetic field parallel to the z axis of a nematic single crystal is to decrease the magnitude of the fluctuations and consequently to increase the birefringence of the sample.

De Gennes[8] has shown that, in the approximation of the equality of the three elastic constants K_i, the effective anisotropy of the optical dielectric constant should be given by

$$\langle \epsilon_{\parallel} - \epsilon_{\perp} \rangle = \epsilon_a(T) + \frac{\epsilon_{a0} k_B T \chi_a^{1/2}}{2\pi K^{3/2}} |\vec{H}|, \qquad (1)$$

where ϵ_{a0} is the dielectric anisotropy for full alignment.[10]

Thus, the increase of the dielectric anisotropy induced by the magnetic field is expected to vary linearly with $|\vec{H}|$. The coefficient of $|\vec{H}|$ is small: taking $\epsilon_{a0} = 1$, $T = 300°K$, $\chi_a = 10^{-7}$ cgs, $K = 10^{-6}$ cgs the correction term is of the order 3×10^{-4} for $H = 10^5$ Oe, corresponding to a relative change in refractive-index anisotropy of the order 3×10^{-4}. A very sensitive polarimetry apparatus and high magnetic fields must be used to observe this effect and it explains why such a study had not yet been done.

The experiments have been performed on 7CB (4'-*n*-heptyl-4-cyanobiphenyl) chosen because of its good chemical stability. The sample had a nematic isotropic phase transition at $T_c = 41.6°C$. It was sandwiched between two glass plates (24 mm long, 12 mm wide) separated by 150-μm-thick Mylar wedges. The long molecular axes were imposed to be parallel to the plates and parallel to each other in zero field (z direction) by an obliquely incident coating of SiO on the plates. Without any magnetic field, the sample exhibits a large birefringence which is strongly temperature dependent and thus the temperature stability must be better than 0.003°C. The high magnetic fields necessary for that experiment were ob-

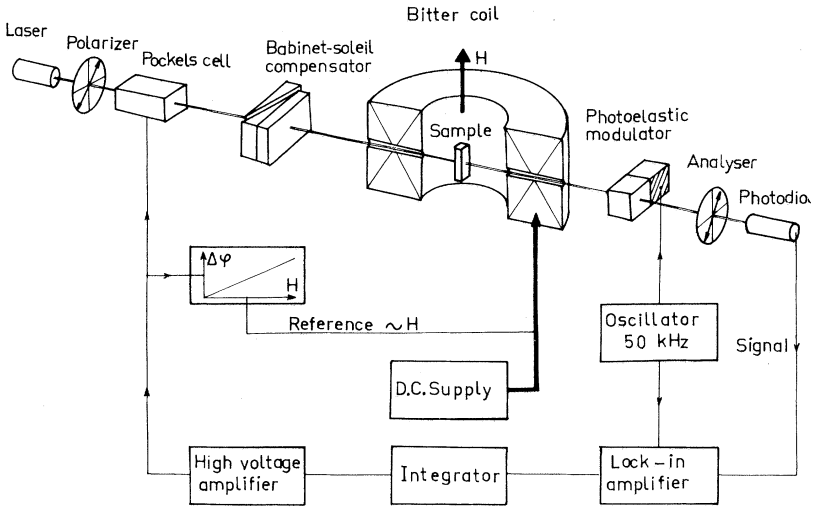

FIG. 1. Experimental apparatus for polarimetry measurements in high magnetic field.

tained with a Bitter coil.[14] The laser beam of the polarimetry apparatus passed through the coil perpendicular to the axis. The plane of the sample could be adjusted perpendicular to the laser beam direction. It could also rotate by steps of 1′ around the laser beam direction in order to set the orientation direction z of the molecules parallel to the direction of the magnetic field. The polarimetry apparatus used for these experiments was developed by G. Maret and M. v. Schickfus of the Max Plank Institut.[15] It can detect phase shifts as small as 10^{-4} rad. A schematic diagram of the experimental apparatus is given in Fig. 1.

The results of the measurements are directly given as a plot of the phase shift versus the magnetic field.

In Fig. 2 are shown the variations with the magnetic field (up to 120 000 Oe) of the phase shift $\Delta\varphi$ introduced by the sample at $T = 32.5°C$ (other measurements were made at different temperatures between 30 and 38°C giving equivalent results). Several curves have been recorded to show the reproducibility of the results. The direction of alignment of the molecules was adjusted parallel to the direction of the magnetic field ($\alpha = 0 \pm 2'$). A bad alignment of the sample in the magnetic

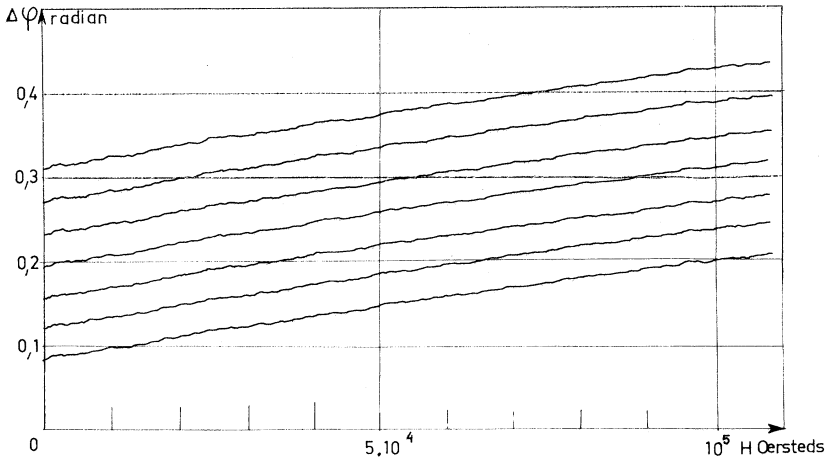

FIG. 2. Phase shift $\Delta\varphi$ vs magnetic field H for successive experiments at the same temperature (the origins have been shifted). The plot of $\Delta\varphi$ vs H and T was verified to be flat when no liquid crystal was placed between the two glass plates of the cell.

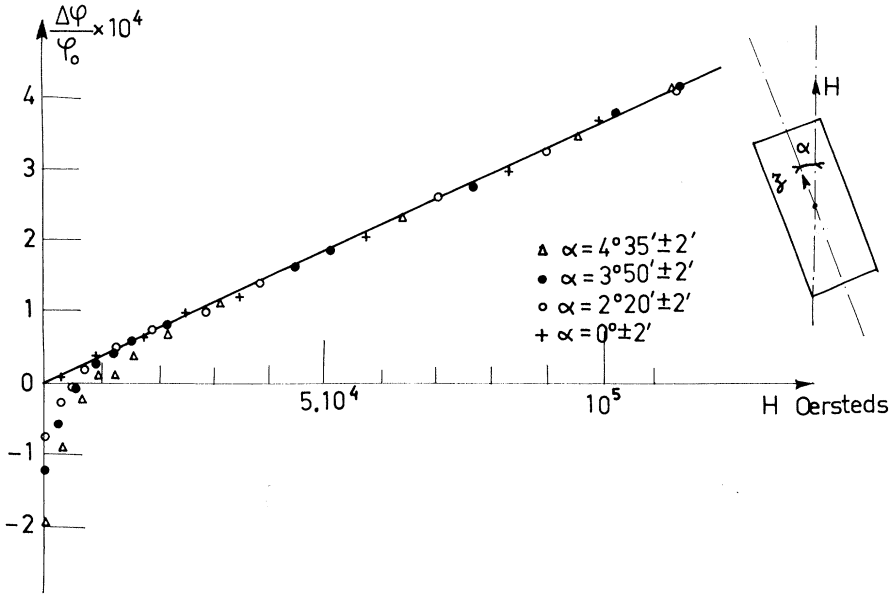

FIG. 3. Relative phase shift $\Delta\varphi/\varphi_0$ vs magnetic field for different values of the angle α between the optical axis of the sample and the direction of the magentic field. For $\alpha = 0$ and $H = 0$ the phase shift introduced by the sample is $\varphi_0 = 300$ rad.

field induces a parasitic phenomenon (Fig. 3) which only appears in low fields (up to 25 000 Oe for $\alpha = 4°$).

Moreover, the sample was rotated through a full 180° about the direction of the magnetic field and the sign of $\Delta\varphi$ was found to be the same as for $\alpha = 0$. This demonstrates the dependence of the birefringence on the absolute value of the magnetic field and confirms the usual symmetry for a nematic liquid crystal.

Thus, in high fields for $\alpha \neq 0$, and in the whole range (0–120 000 Oe) for $\alpha = 0$, $\Delta\varphi$, and consequently Δn, is found to vary linearly with $|\vec{H}|$ as predicted by the theory. The agreement is also rather good in a quantitative way: Our measurements give for the relative phase shift between 0 and 120 000 Oe, $\Delta\varphi/\varphi = 4.2 \times 10^{-4}$. From (1), one gets, with $\chi_a = 1.47 \times 10^{-7}$ cgs,[16] $K \approx 10^{-6}$ cgs, $T = 305°K$: $\Delta(\langle\epsilon_{\parallel} - \epsilon_{\perp}\rangle)/\langle\epsilon_{\parallel} - \epsilon_{\perp}\rangle = 3 \times 10^{-4}$.

Thus, it has been proved that the birefringence of a nematic single crystal varies linearly with the magnetic field. This result fills an important lacuna in the study of the order parameter and provides a new important element for the comprehension of critical phenomena in general.

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