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# Determination of the minimum concentration of ferrofluid required to orient nematic liquid crystals

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Lyotropic nematic mesophases doped with different concentrations of ferromagnetic grains are observed in a polarizing microscope. A minimum concentration of the grains is obtained above which a collective behavior of the nematic matrix is observed. The results are discussed in terms of the elastic continuum theory of liquid crystals.

Liquid crystals may be oriented[1] by electric and magnetic fields (**H**) or by shear stress. Since the anisotropic part of the diamagnetic susceptibility of these systems ($\Delta\chi$) is small[1] ($\sim 10^{-7}$ cgs units), high magnetic fields[2] ($\sim 10$ kG) are usually needed. In order to reduce the strength of $H$ in practical experiments ferromagnetic grains (ferrofluids) are introduced[2–7] in the liquid-crystalline system. A ferrofluid is a colloidal suspension of small ferromagnetic particles dispersed in a liquid medium.[8] It has the fluidity of a homogeneous liquid and high magnetic susceptibility. The magnetic particles, with typical dimensions of about $10^2$ Å, dispersed in a solvent, are coated[8] with a dispersive agent to prevent their aggregation. Nematic liquid crystals doped with ferrofluids have been called[2] "ferronematic" liquid crystals. With this doping, fields of about 10 G are enough to orient the mesophases.[5,6] The introduction of ferrofluids in liquid crystals serves the purpose not only of reducing the strength of $H$ necessary to orient the mesophases, but also allows the study of the physics of the liquid-crystalline system itself.[2,9]

Brochard and de Gennes[2] discussed in a very complete way the theory of magnetic suspensions in liquid crystals, concluding that the coupling between the ferrofluid particles and the liquid crystalline matrix is mainly of mechanical origin. They also predicted[2] that there is a minimum concentration ($C_m$) of ferrofluid above which the liquid-crystalline matrix is oriented by the magnetic grains. The knowledge of $C_m$ is important in practical experiments since the quantity of ferrofluid introduced in liquid crystals must be as little as possible, to preserve[7] the physical properties of the liquid-crystalline system. In this paper we report what we believe to be the first measurement of the minimum concentration of magnetic grains in a ferrofluid-doped lyotropic nematic liquid crystal for different sample thickness. The results are discussed in terms of the elastic continuum theory of liquid crystals.

The free energy ($F$) of a nematic liquid crystal in the presence of a magnetic field **H** is written as[1]

$$F=\tfrac{1}{2}K\int\left[(\mathrm{div}\,\mathbf{n})^2+(\mathrm{rot}\,\mathbf{n})^2-\frac{\Delta\chi}{K}(\mathbf{H}\cdot\mathbf{n})^2\right]dv\ , \qquad (1)$$

where $K$ is the Frank elastic constant[10] and **n** is the local director.

For nematic liquid crystal doped with magnetic grains in the presence of a small magnetic field (below the Freedericksz critical field $H_F$), the elastic term is by far[2] the predominant one at the free energy. Far from a single ferromagnetic grain ($r \gg L$, $L$ is the length of the grain), **n** is parallel to the rubbing direction $\mathbf{n}_0$. The distortion induced by the grain may be written as $\delta\mathbf{n}(r)=\mathbf{n}(r)-\mathbf{n}_0$. It can be shown from symmetry considerations[2] that for a cylindrical grain ($L/d\sim 10$, $d$ is its diameter)

$$\delta\mathbf{n}(r)=\frac{l(\cos\theta)}{r}(\mathbf{u}-\mathbf{n}_0\cos\theta) \qquad (2)$$

where **u** is a unit vector parallel to the axis of the grain and $\theta$ is the angle $(\mathbf{n}_0,\mathbf{u})$. The function $l(\cos\theta)$ has the dimensions of length and is called[2] the distortion amplitude. For a long rod grain, $l(\cos\theta=1)$ is comparable[2] to $L$. Considering a finite concentration of ferrofluid ($C$ represents grains per cm$^3$), the distortion effect of each grain is screened out[2] at distances larger than $\kappa^{-1}=(4\pi CL)^{-1/2}$. In a region of the nematic sample with linear dimensions $D$, **n** will follow the grains if $D\kappa>1$. Therefore, the minimum concentration of ferrofluid necessary to promote a collective behavior of the nematic matrix is roughly[2]

$$C_m\sim\frac{1}{LD^2}\ . \qquad (3)$$

For $D\kappa<1$, **n** is weakly perturbed inside the region of size $D$.

The lyotropic nematic liquid crystal used in this work is the mixture of potassium laurate (28.74 wt. %)/Decanol (6.64 wt. %)/$H_2O$ (64.62 wt. %). The measurements are performed at 23 °C. At this temperature, the lyotropic mixture has a calamitic nematic phase ($N_C$) as determined by conoscopic measurements of the birefringence. This phase is uniaxial and the director orients[7] parallel to the magnetic field. The magnetic particles are[7] grains of $Fe_3O_4$ with $L\sim 100$ Å, coated with oleic acid, dispersed in water. Nematic samples are sealed in flat glass microslides (thickness is $D$, width is 2mm, length is 2 cm) of different thickness (50,100,200 $\mu$m), for different ferrofluid concentrations ($C$). Before using, the microslides are cleaned with a continuum flow of water and dried in a stove.

Some minutes after filling a 50 or 100 $\mu$m microslide with the $N_C$ phase (without the magnetic field), an almost perfect planar texture is observed at the polarizing microscope. In the case of the 200-$\mu$m microslide, about 1 h is needed to observe large regions with a planar texture. To verify the direction of $\mathbf{n}$ of the $N_C$ phase in the microslides, x-ray diffraction experiments in transmission geometry[11] with the beam perpendicular to the biggest surface of the microslide are performed. The two typical first-order bands[7,11] are observed indicating that even without a particular surface treatment, $\mathbf{n}$ is oriented along the long axis (length) of the microslide. The existence of this rubbing direction $\mathbf{n}_0$ in microslides with the particular geometry used in this work can be understood taking into account that the microslide may be sketched as two half-cylindrical boundaries tied by two plane surfaces. At the plane surfaces $\mathbf{n}$ tends to orient parallel to the surfaces, without a particular direction. At the cylindrical boundaries, however, the condition of minimum free energy gives for the director the topological configuration of $\mathbf{n}$ parallel to the long axis of the half-cylinders. These fixed directions at the boundaries break the degeneracy of $\mathbf{n}$ at the bulk. This direction defines the rubbing direction $\mathbf{n}_0$.

*Evidence of the minimum concentration.* The samples are placed in a polarizing microscope (Wild, orthoplanpol) and a magnetic field of 100 G[12] ($<H_F$) is applied in the plane perpendicular to the light beam. The field may be parallel or at 45° of the polarizing light direction. A photoelectric cell coupled to a millivoltmeter measures the light intensity ($I$) transmitted by the sample. For $C \geq C_m$, the magnetic field orients the sample with $\mathbf{n} \| \mathbf{H}$. A planar texture[1] is obtained with the optical axis of the phase perpendicular to the light beam. $\Delta I$ measures the difference between the light intensity transmitted in both configurations of $\mathbf{H}$ parallel and at 45° of the incident polarizing light direction. For $C < C_m$, $\Delta I$ must be zero.

Figure 1 shows the values of $\Delta I$ as a function of the concentration of ferrofluid, of a ferrofluid-doped lyotropic nematic mesophase in a 200-$\mu$m-thick microslide. It is clearly shown the existence of a minimum concentration above which a collective behavior of the nematic matrix is observed. This result is independent of the applied magnetic field, for 10 G $< H < H_F$. For this sample thickness, $7\times10^8 < C_m < 10^9$ grains/cm$^3$ and expression (3) gives $C_m \sim 10^9$ grains/cm$^3$.

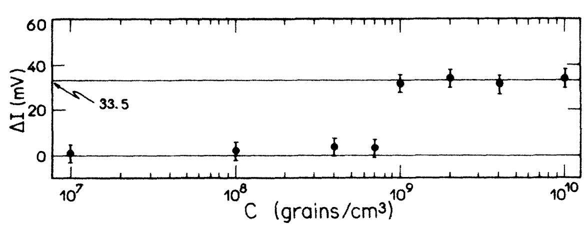


FIG. 1. Difference between the light intensity ($\Delta I$ in mV) transmitted by the ferrofluid-doped nematic sample in the configuration of $\mathbf{H}$ parallel and at 45° of the incident polarizing light direction as a function of the concentration of ferrofluid. $D = 200\ \mu$m.

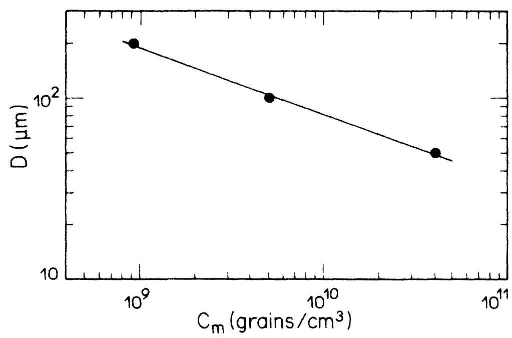


FIG. 2. Minimum concentration of ferrofluid ($C_m$) as a function of the sample thickness ($D$).

It is interesting to note that even in the nonperfectly planar regions of the sample doped with $C < C_m$, the texture is not modified by the action of the magnetic field ($H < H_F$—time of the experiment 24 h). These samples also presented the same x-ray diffraction patterns with the magnetic field in different orientations, in the plane of the biggest surface of the microslide. Samples doped with $C > C_m$ in the presence of $H$ ($< H_F$) present modifications of their optical textures in about 10 min. This fact indicates that even in the nonperfectly planar domains of the sample, the boundary conditions imposed by the glass surfaces are stronger than the field ($H < H_F$).

The dependence of $C_m$ with the sample thickness $D$ is also verified. Figure 2 shows the values of $C_m$ as a function of $D$. The smaller the sample thickness, the greater the minimum concentration of ferrofluid. Our results indicate that $C_m \sim D^a$ with $a \sim -2.5$. The values of $C_m$ for $D = 50$ and 100 $\mu$m, calculated with expression (3) are $10^{10}$ grains/cm$^3$ and $4\times10^{10}$ grains/cm$^3$, respectively.

Comparing our results with the values obtained with expression (3) one observes that (1) for $D = 50\ \mu$m, $C_m$ agrees very well with the theoretical value. However, for larger sample thickness ($D = 100$ and 200 $\mu$m), the experimental values of $C_m$ are smaller than the theoretical values; (2) the experimental dependence of $C_m$ with $D$ is stronger than theoretically predicted.

Expression (3), obtained considering long cylindrical ferromagnetic grains in a nematic matrix described by a unique elastic constant, gives a superior limit for $C_m$, mainly for increasing values of $D$. In actual ferrofluid-doped nematics, end effects of the grains interaction with the nematic matrix (neglected in the theory[2]) may play an important role in this problem.

As a final remark, the introduction of a small quantity of ferrofluid in the lyotropic nematic mesophase ($C = C_m$) does not modify the temperature transitions (accuracy $\pm 0.1\,^\circ$C) and the measured birefringence, compared with the original nematic phase.

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