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Magnetic Platelets in a Nematic Liquid Crystal

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Results are presented for a theoretical and experimental investigation of magnetic platelets introduced into a nematic liquid crystal. The equilibrium positions of the platelets and nematic director as a function of applied field are calculated and compared with experiment.

I INTRODUCTION

Although fairly strong ($\gtrsim 1$ kG) magnetic fields are usually required to influence alignment of a nematic liquid crystal Brochard and de Gennes[1] have suggested that the nematic orientation could be coupled to much weaker magnetic fields if magnetic grains were incorporated into the nematic matrix. If the grains are non-spherical a non-random orientation may be imposed on the grains by the elastic properties of the nematic which could give rise to a net magnetic field. Such a system was called a "ferronematic." Brochard and de Gennes assumed the grains were needle shaped and performed a theoretical analysis of their behavior.

Such a system was prepared by Rault, Cladis and Burger[2] who suspended Fe_2O_3 needles in *p*-methoxybenzilidene-*p*-*n*-butylaniline (MBBA) and measured the magnetization for various temperatures and applied magnetic fields. However, no one has apparently observed the interactions of the individual magnetic particles nor compared their behavior quantitatively with theory.

We have prepared a ferronematic using Fe_3O_4 particles for the purpose of observing the individual grains. Our particles are platelets composed of many much smaller single domain grains. The form for the free energy and torques which are applicable for these particles are derived in Section II.

Section III gives the experimental observations we have made on the grains and comparison is made with the theory.

II TORQUE BALANCE ON A MAGNETIC DISK

If the Frank elastic constants may be set equal to a single constant, K, we may write the elastic free energy[3] of the nematic as

$$F = \tfrac{1}{2}K \int d\tau \frac{\partial n_j}{\partial x_i}\frac{\partial n_j}{\partial x_i} \tag{1}$$

We take the disk to have its symmetry axis along the z axis and at an angle θ with respect to n_0, the liquid crystal director far from the disk. Taking the center of the disk as the origin we may therefore express n_0 as

$$\mathbf{n}_0 = \mathbf{i}\sin\theta + \mathbf{k}\cos\theta \tag{2}$$

where $\mathbf{n} = \mathbf{n}_0$ as $r \to \infty$. Closer to the disk we would expect (Note Figure 1)

$$\mathbf{n} = \mathbf{i}\sin\alpha(r) + \mathbf{k}\cos\alpha(r) \tag{3}$$

with boundary conditions $\alpha(0) = 0$ and $\alpha(\infty) = \theta$. We assume $\mathbf{n}$ perpendicular to the disk at its surface as we find to be the case experimentally. Combining Eq's. 1 and 3 we have

$$F = \tfrac{1}{2}K \int (\nabla\alpha)^2 d\tau \tag{4}$$

For equilibrium we have, therefore,

$$\nabla^2\alpha = 0 \tag{5}$$

Surrounding the disk of radius a we would expect a series of nonintersecting surfaces on which the value of α would be constant. For the present symmetry neglecting edge effects we find a family of confocal ellipsoids,

$$\frac{x^2}{a^2+\psi} + \frac{y^2}{a^2+\psi} + \frac{z^2}{\psi} = 1 \tag{6}$$

where each value of ψ results in a different member of the family and a unique value of α:

$$\alpha = \alpha(\psi) \tag{7}$$

Using Eq. 5 we obtain

$$\frac{d^2\alpha}{d\psi^2}(\nabla\psi)^2 + \frac{d\alpha}{d\psi}\nabla^2\psi = 0 \tag{8}$$

Noting that $(d^2\alpha/d\psi^2)/(d\alpha/d\psi) = d[\ln(d\alpha/d\psi)]/d\psi$ we find Eq. 8 may be integrated to yield

$$\alpha = C_1 \int e^{-\int \nabla^2\psi/(\nabla\psi)^2 d\psi} d\psi + C_2 \tag{9}$$

with C_1 and C_2 determined by the boundary conditions. Solving Eq. 6 for ψ we find

$$\nabla^2\psi/(\nabla\psi)^2 = \left(\frac{1}{a^2+\psi} + \frac{1}{2\psi}\right) \tag{10}$$

Now integrating Eq. 9 with the result of Eq. 10 we find

$$\alpha = \frac{2\theta}{\pi} \tan^{-1}(\sqrt{\psi}/a) \tag{11}$$

where from Eq. 6 we have

$$\psi = [r^2 - a^2 + \sqrt{(a^2 - r^2)^2 + 4a^2z^2}]/2 \tag{12}$$

Using Green's first theorem:

$$\oint \alpha\nabla\alpha \cdot d\mathbf{s} = \int [\alpha\nabla^2\alpha + (\nabla\alpha)^2] d\tau \tag{13}$$

where **ds** is an element of area enclosing the liquid crystal, together with Eq. 5 we have from Eq. 4

$$F = \tfrac{1}{2}K \oint \alpha\nabla\alpha \cdot d\mathbf{s} \tag{14}$$

The liquid crystal volume may be considered bound by the disk surface and a sphere of radius $r \gg a$. Integration over the former gives zero since $\alpha = 0$ at the disk. For $r \gg a$ from Eq. 12 $\psi \sim r^2$ and the arctangent may be expanded to give

$$\alpha \sim \theta(1 - 2a/\pi r) \tag{15}$$

Finally from Eq. 14 we have

$$F = 4\,Ka\theta^2 \tag{16}$$

and the resulting elastic torque, Γ_E, tending to reduce θ to zero is

$$\Gamma_E = 8\,Ka\theta \tag{17}$$

If the magnetic dipole moment of the disk is μ when a magnetic field, H, is applied there will be a magnetic torque Γ_M tending to make θ approach $\pi/2$ for H parallel to n_0:

$$\Gamma_M = \mu H \cos\theta \tag{18}$$

We assume μ lies in the plane of the disk. For equilibrium therefore we may equate the elastic and magnetic torques:

$$H = 8Ka\theta/\mu \cos\theta \tag{19}$$

When the magnetic field is removed and the disk returns to equilibrium a viscous torque, Γ_v, will act. For a disk Γ_v is of the form[4]

$$\Gamma_v = \frac{32\eta a^3}{3}\frac{d\theta}{dt} \tag{20}$$

where $d\theta/dt$ is the angular speed of the disk and η is the effective viscosity of the liquid crystal. From Eq. 17 and 20 we find

$$\theta = \theta_0 e^{-3Kt/4\eta a^2} \tag{21}$$

Experimentally we will be concerned with Eq's. 19 and 21.

III EXPERIMENT

Magnetic grains of Fe_3O_4 in a hydrocarbon base from Ferrofluidics Corp. were mixed with MBBA from Eastman Kodak. The grains are coated with an organic compound to retard coagulation. Originally the grains are quasi-spherical with a diameter of the order of 100A. When mixed with MBBA flocculation has occurred: A drop of ferrofluid is spread on the MBBA surface. The ferrofluid carrier diffuses rapidly into the MBBA leaving the grains in a thin coagulated layer at the surface. This thin layer is then broken apart and mixed with the MBBA substrate, each broken segment of the magnetic layer forming a platelet. The flocs we consider have a thickness to width ratio of about one to ten. We may approximate these platelets as disks for comparison with the previous section.

The MBBA is aligned homogeneously by the method of Chatelain[5] between two glass surfaces with a spacing of $\sim 10^{-2}$ cm. With no magnetic field applied the platelets when observed through a microscope are seen resting on end with θ equal to zero in accordance with the condition for minimum free energy, Eq. 16. Lecithin is used for homeotropic alignment, causing the broad side of the platelets to be seen. We conclude the director is perpendicular to the platelet surface at the platelet boundary. When a sample is freshly made certain regions can be seen to change from planar to perpendicular alignment. The platelets as expected are seen to turn 90° about a horizontal axis when the change occurs. If a magnetic field parallel to $\mathbf{n}_0$ is applied the platelets turn. For homogeneous alignment if the dipole moment is horizontal the equilibrium position will still present an edge view of the platelet although rotated

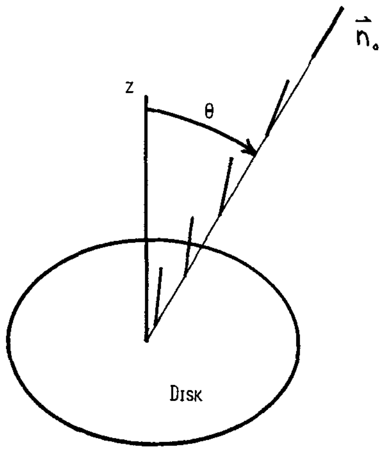

FIGURE 1 Director orientation as a function of distance from the disk.

through an angle θ. If the dipole moment has a vertical component the platelet will tilt giving a view of the width of the platelet. For a platelet having a horizontal dipole moment and a radius, a, equal to 3.7×10^{-3} cm a magnetic field was applied which turned the platelet by $\theta = 70°$. The magnetic field was suddenly changed to zero and the subsequent values of θ were measured as a function of time. The results are shown in Figure 1. The solid line is the best fit for an exponential where we have

$$3K/4\eta a^2 = 1.6 \times 10^{-2}/\text{sec} \tag{22}$$

Taking $K = 5 \times 10^{-7}$ dynes[3] and $\eta = 0.3$ poise[6] we find $a = 8.8 \times 10^{-3}$ cm. Considering the approximations we are making in using one elastic constant and one viscosity constant and taking the shape of the platelet to be a disk an order of magnitude agreement is all we may legitimately expect. Also the surface of the platelet is not smooth and so does not give uniform coupling to the nematic as the theory of Section II assumes.

The angle of equilibrium for the platelet was measured as a function of magnetic field. The results are shown in Figure 2. The solid line is the fit to Eq. 19 where we have

$$8Ka/\mu = 7.6 \text{ Gauss} \tag{23}$$

Using again $K = 5 \times 10^{-7}$ dynes and $a = 4 \times 10^{-3}$ cm we find $\mu = 2 \times 10^{-9}$ erg/G. We conclude the grains are only slightly aligned in the platelet since uniform alignment would give a dipole moment several orders of magnitude larger. There should be platelet–platelet interaction observed for $R \sim a$ where R is the distance of separation of the platelets, and in fact the platelets are seen to both turn and coagulate.

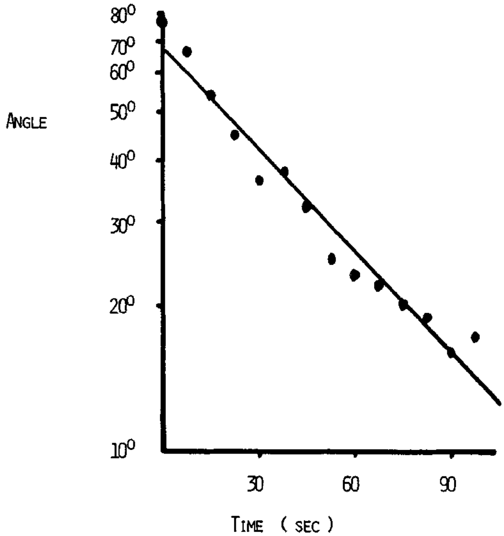

FIGURE 2 Orientation of a magnetic platelet as a function of time after the applied magnetic field is removed. The solid line is a best fit to an exponential.

Consider a platelet inclined at angle θ to n_0. When viewed through crossed polarizers with the initial polarizer parallel to $\mathbf{n}_0$ the field far from the grain should appear dark in a homogeneous sample. It is easily shown that the intensity around the grain should vary as

$$I = I_0 \sin^2[2(\theta - \alpha)]/\sin^2 2\theta \tag{24}$$

where I_0 is the intensity at $z = 0$ and α is given by Eq's. 11 and 12. Taking $\theta = 45°$ and considering only the intensity along the z axis ($x = y = 0$) Eq. 24 reduces to

$$I = I_0/(1 + z^2/a^2). \tag{25}$$

The intensity drops by a factor of two at a distance $z = a$ from the platelet. Qualitatively we see this to be so in Figure 4A, in which a platelet is positioned approximately as we have been considering.

If the director is perpendicular to the platelet surface in a homeotropically aligned nematic the edges of the platelet will cause an alteration of the nematic alignment. When observed between crossed polarizers light is seen at the edges of the platelet. When a horizontal magnetic field is applied the platelet turns around a vertical axis. The turning of the platelet disrupts the ordering of the nematic around the platelet allowing the transmitted light intensity to increase. Typically for a 50 μm diameter platelet with 100 G applied at 30° to μ the transmission of light doubles during the turning of the platelet. Another

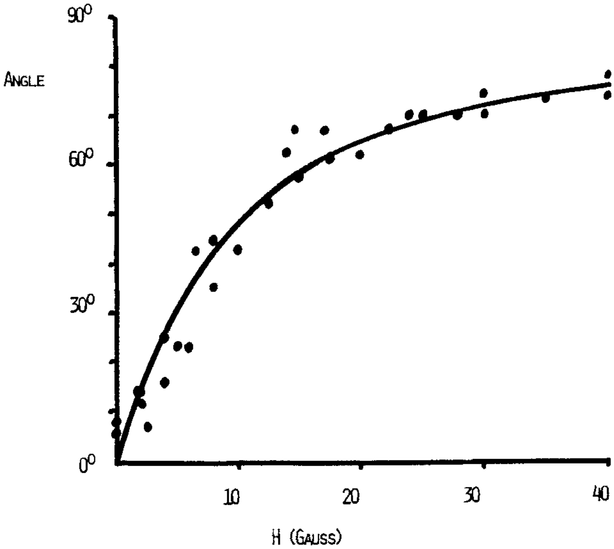

FIGURE 3 Equilibrium orientation as a function of applied magnetic field. The solid line is the fit for Eq. 19.

way of increasing the transmission is by turning the sample while keeping the platelets fixed in orientation. Figure 4B shows a sample rotated with a period of about ten seconds in a 50 G field. Figure 4C shows the sample not being rotated and with the field removed.

The platelets may be transported by several ways in addition to a coupling to the fluid motion observed in other colloids. Due to their magnetic moment

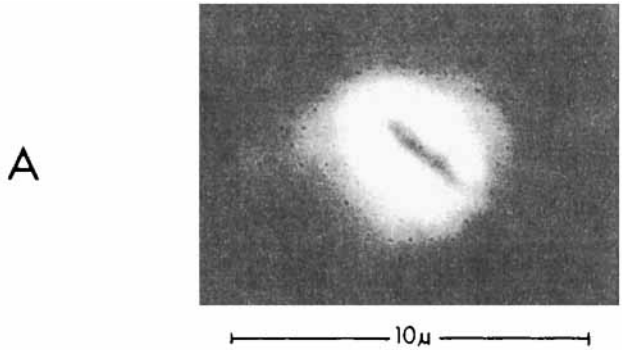

FIGURE 4A Photomicrograph showing an edge view of a platelet in MBBA for homogeneous alignment observed between crossed polarizers oriented parallel and perpendicular to n_0. The white area corresponds to the distortion of the director near the platelet.

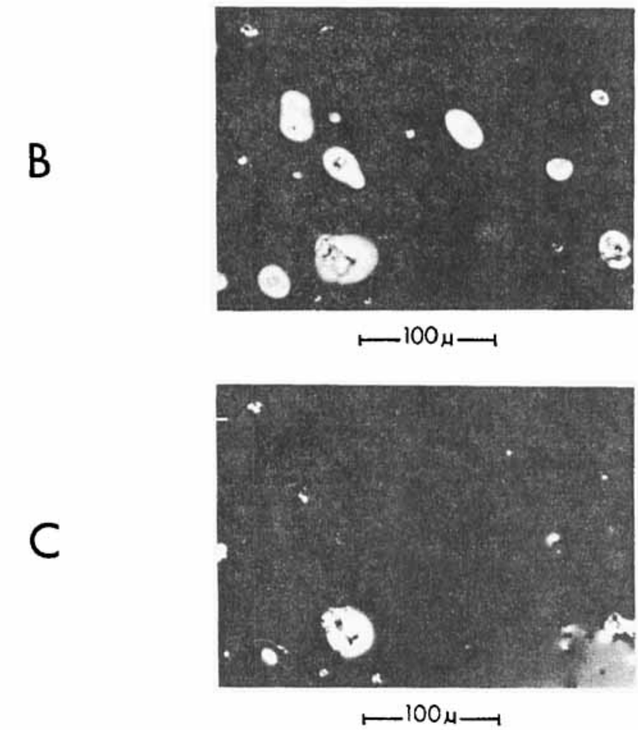

FIGURE 4B and 4C Photomicrographs of platelets in MBBA for homeotropic alignment observed through crossed polarizers. In 4B the MBBA sample is rotated in a stationary magnetic field. In 4C the MBBA sample is stationary and the field removed.

a gradient of applied magnetic field causes transport. There can be inter-platelet interaction causing transport and coagulation. There may be platelet interaction with a point disinclination causing the platelet to move to the disinclination point. The platelets may become attached to moving disinclination lines. Each of these means of transport has been observed. Finally, we expect the platelets to move to regions where **n** is perpendicular to the applied field. Since magnetic needles should move to regions where **n** is parallel to the field a means of separation of particles is possible due to their shape.

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