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Ferronematics in Magnetic and Electric Fields

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We consider the effect of a magnetic field on a ferronematic. It is shown that first order Freedericksz transitions are possible in such systems as a consequence of elastic and diamagnetic anisotropies. Interestingly the possibility exists for a first order transition from a splay-bend distortion to a pure twist distortion or vice versa at a second threshold. A super-imposed electric field can also result in a first order Freedericksz transition.

Keywords: ferronematics, Freedericksz transition, tricritical behavior

INTRODUCTION

Ferronematics are nematic phases with preferentially aligned magnetic grains. Brochard and de Gennes¹ were the first to consider theoretically the behavior of ferronematics in magnetic fields. They assumed the magnetization of the medium to be large enough for diamagnetic effects to be ignored (magnetization 1 G) and got a second order Freedericksz transition. Such ferronematic phases were first prepared in the Laboratory with needle-like grains by Rault *et al.*² and later by others.^{3–5} Plate-like grains in the nematic phase have also been investigated.⁶ In these systems the magnetization appears to be very much smaller (of the order of 10^{-4} G). In addition, Shen and Amer⁵ have reported in a ferronematic, with magnetization \mathbf{M} perpendicular to the nematic director, the classical Freedericksz transition in the homeotropic geometry with applied field parallel to the magnetization. These results indicate that diamagnetic effects cannot be ignored. Further the full implications of elastic anisotropy have also not been worked out so far.

In this paper we consider the effects of both diamagnetic and elastic anisotropy in ferronematics in classical Freedericksz geometries. We find interestingly the transition to be first order below a critical magnetization or a critical ratio of the elastic constants and second order above it, thus exhibiting tricritical behavior. It should be mentioned that a first order transition is possible in the case of a twisted

nematic.⁷ Also more recently it has been shown^{8,9} that a classical Freedericksz transition can be made first order with the optical field of a laser.

In the homogeneous geometry we find at a higher field a first order transition from one type of distortion to another, in the presence elastic anisotropy. We have also worked out the consequences of an electric field acting along the imposed magnetic field. In this case also we find tricritical behavior.

THEORY

The free energy density for a ferronematic is given by:

$$F = \frac{k_1}{2} (\nabla \cdot \mathbf{n})^2 + \frac{k_2}{2} (\mathbf{n} \cdot \nabla \times \mathbf{n})^2 + \frac{k_3}{2} (\mathbf{n} \times (\nabla \times \mathbf{n}))^2 - \frac{\chi_a}{2} (\mathbf{n} \cdot \mathbf{H})^2 - \mathbf{M} \cdot \mathbf{H} - \frac{\epsilon_a}{8\pi} (\mathbf{n} \cdot \mathbf{E})^2 \quad (1)$$

where

\mathbf{M} = Magnetization

χ_a = Positive diamagnetic anisotropy

ϵ_a = Positive dielectric anisotropy

\mathbf{H} = Magnetic field

\mathbf{E} = Electric field

k_1, k_2, k_3 , are the splay, twist and bend elastic constants respectively.

We consider only ferronematics with \mathbf{M} along the director \mathbf{n} . Throughout this paper we assume both electric and magnetic fields to be parallel to undistorted \mathbf{n} . Further we assume the magnetic field to be antiparallel to the magnetization. The plates are at $z = 0$ and $z = d$ and are parallel to the x - y plane. We consider only fields very close to the threshold.

HOMEOTROPIC ALIGNMENT

In this case the distortion is given by $n = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta)$. This has a cylindrical symmetry about z-axis. In fact we get the familiar umbilic structure. Just above the threshold we can assume

$$\theta = \theta_m(r) \sin \pi z/d$$

$$\phi = \pm \tan^{-1} \frac{y}{x}$$

where d is the sample thickness.

Considering regions only far away from the core of the umbilic, i.e., $r \gg \xi$, we get the free energy density after averaging over sample thickness, as:

$$\bar{F} = F_o + \alpha/2 \theta_m^2 + \beta/4 \theta_m^4 + \gamma/6 \theta_m^6 \quad (2)$$

where

$$\alpha = (1/2)[k_3\pi^2/d^2 - mH + \chi_a H^2 + \epsilon_a E^2/4\pi]$$

$$\beta = (1/4) \left[\frac{(k_1 - k_3)\pi^2}{d^2} + \frac{mH}{4} - \chi_a H^2 + \epsilon_a E^2/4\pi \right]$$

$$\gamma = (1/144) \left[\frac{(k_3 - k_1)\pi^2}{d^2} + \chi_a H^2 - \epsilon_a E^2/4\pi - \frac{mH}{16} \right]$$

We notice \bar{F} to be very similar to the Landau free energy density. We have included the sixth power term to allow for negative values of β . Minimization of free energy density yields the equilibrium value of θ_m .

HOMOGENEOUS GEOMETRY

Here also we have both θ and ϕ distortions described by $\mathbf{n} = (\cos \theta \cos \phi, \cos \theta \sin \phi, \sin \theta)$. Just above the threshold we can assume θ and ϕ to be given by:

$$\theta = \theta_m \sin \pi z/d$$

$$\phi = \phi_m \sin \pi z/d$$

As before after averaging over the sample thickness we get

$$\bar{F} = F_o + \frac{\alpha_1}{2} \theta_m^2 + \frac{\alpha_2}{2} \phi_m^2 + \frac{\beta_1}{4} \theta_m^4 + \frac{\beta_2}{4} \phi_m^4 + \frac{\delta}{2} \theta_m^2 \phi_m^2$$

Here

$$\begin{aligned}\alpha_1 &= 1/2 \left[\frac{k_1 \pi^2}{d^2} - MH + \chi_a H^2 + \epsilon_a E^2/4\pi \right] \\ \alpha_2 &= (1/2) \left[\frac{k_2 \pi^2}{d^2} - MH + \chi_a H^2 + \epsilon_a E^2/4\pi \right] \\ \beta_1 &= (1/4) \left[\frac{(k_3 - k_1) \pi^2}{d^2} + MH/4 - \chi_a H^2 - \epsilon_a E^2/4\pi \right] \\ \beta_2 &= (1/4) \left[\frac{MH}{4} - \chi_a H^2 - \epsilon_a E^2/4\pi \right] \\ \delta &= (1/8) \left[\frac{(k_3 - 2k_2) \pi^2}{d^2} + 3 \left(\frac{MH}{2} - \chi_a H^2 - \epsilon_a E^2/4\pi \right) \right]\end{aligned}$$

This is similar to the generalized Lifshitz's expression for a two order parameter system.

When β_1 (or β_2) is negative, we will also have to add the sixth power term in θ (or ϕ). Coefficient of this term for θ and ϕ , respectively are:

$$\begin{aligned}\gamma_1/6 &= 1/96 \left[\frac{(k_1 - k_3) \pi^2}{d^2} - \frac{MH}{24} + 2/3 [\chi_a H^2 + \epsilon_a E^2/4\pi] \right] \\ \gamma_2/6 &= 1/96 [2/3 (\chi_a H^2 + \epsilon_a E^2/4\pi) - MH/24]\end{aligned}$$

RESULTS

Magnetic field effects

In the case of homeotropic geometry even in the one constant approximation we find the important result of β changing sign as \mathbf{M} varies. For a value of \mathbf{M} less than the critical value $M_c = 4\pi/d (k\chi_a/3)^{1/2}$, β is negative resulting in a first order transition. And for higher values, β is positive giving a second order transition. Therefore there exists a tricritical behavior at M_c . Figure 1 shows the variation of θ with H in the first order transition. It should be emphasized that the harmonic solution that we have assumed is not strictly valid when the sixth power term is considered. Hence our θ variation is only approximate. Figure 2 gives the variation of $\Delta\theta$, the jump at transition as a function of \mathbf{M} . The first order transition takes place at $\alpha = \beta^2/4\gamma$. Even when diamagnetic effects are ignored, we find tricritical behavior when elastic anisotropy is included. In this geometry we find the transition to be first order or second order depending upon whether $k_3 > 4k_1/3$ or $k_3 < 4k_1/3$. We find the Brochard-de Gennes solution of a second order transition only when

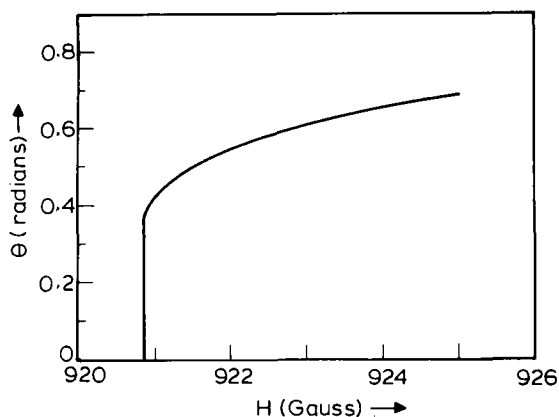


FIGURE 1 Onset of first order Freedericksz transition in the homeotropic geometry. $d = 20 \mu\text{m}$, $k_1 = k_2 = k_3 = 0.5 \times 10^{-6}$ dyne, $M = 1.8 \times 10^{-3}$ Gauss, $\chi_a = 0.5 \times 10^{-6}$ cgs units.

neither the diamagnetic nor elastic anisotropies are included. However, when one or both of them are present we have tricritical behavior.

When the transition is second order we get an umbilic with a continuous core, i.e., θ smoothly goes to zero as $r \rightarrow 0$. The core is very similar to the one discussed by Rapini.¹⁰ In the case of first order Freedericksz transition the core would not be continuous and we can expect a first order jump in θ to zero as $r \rightarrow 0$.

In the present problem the vanishing of α yields two values of H , viz., H_1 and H_2 and α is negative between H_1 & H_2 . The transition discussed above occurs at or below H_1 . Also we can expect a reverse transition to the undistorted state for $H \gg H_2$. However the nature of this transition is difficult to work out due to the large value of θ the system has, near this field. In all probability this will be a first order transition since $\alpha > 0$ and $\beta > 0$ for $H \gg H_2$.

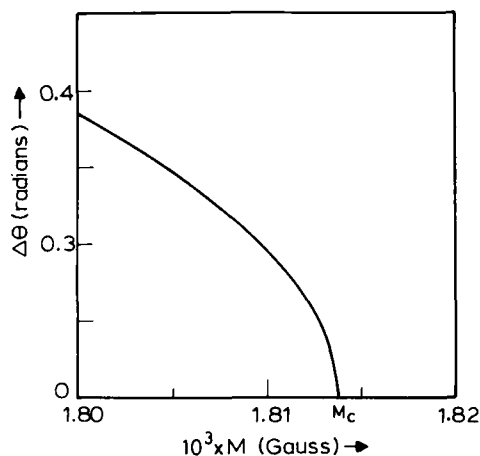


FIGURE 2 Jump $\Delta\theta$ at the transition as a function of M for the homeotropic geometry and for parameters given in Figure 1.

However, for $M < 2\pi/d(k\chi_a)^{1/2}$ the coefficient α is positive for all fields and we get a first order transition only when β is negative.

In the case of homogeneously aligned sample in the one constant approximation the free energy density goes over to the Lifshitz expression for two order parameters.¹¹ Lifshitz's theory predicts four possible states for the system:

$$\left. \begin{array}{ll} \text{(a)} & \theta = 0, \quad \phi = 0 \\ \text{(b)} & \theta = 0, \quad \phi \neq 0 \\ \text{(c)} & \theta \neq 0, \quad \phi = 0 \\ \text{(d)} & \theta \neq 0, \quad \phi \neq 0 \end{array} \right\} \begin{array}{l} \text{when } \alpha > 0 \text{ (undistorted state)} \\ \text{when } \alpha < 0, \quad \beta < |\delta| \\ \text{when } \alpha < 0, \quad \beta > |\delta| \end{array}$$

We always find only (b) and (c) solutions and never (d). Also (b) and (c) solution are equally energetic. This transition to either θ or ϕ alone can be first or second order, depending on M , just as in the case of homeotropic geometry. In this case it is possible to think of a new type of wall connecting these two distortions θ and ϕ . This will be quite different from the familiar Brochard walls which connect only degenerate θ or ϕ . These walls will be smooth when the transition is second order but will have discontinuities for a first order transition.

As before we again find interesting results even when diamagnetic terms are ignored. For example when $k_1 < k_2$ we always get solution (c) ($\theta \neq 0, \phi = 0$). This transition is first order for $k_3 < 4/3 k_1$ and second order for $k_3 > 4/3 k_1$. For $k_2 < k_1$ we always get solution (b) ($\phi \neq 0, \theta = 0$) whenever $k_3 \geq k_1$ and this transition is always second order. However one can even get solution (c) i.e., ($\phi = 0, \theta \neq 0$), when $k_2 < k_1$ provides k_3 is very much smaller than k_1 and the transition to this state is always first order.

When diamagnetic effects are also included we get solution (c) ($\phi = 0, \theta \neq 0$) for $k_1 < k_2$ and $k_3 \leq k_1$. This transition will be first order or second order depending upon the magnetization. However, one can even get solution (b) (i.e., $\phi \neq 0, \theta = 0$) for $k_1 < k_2$ provided k_3 is very much larger than k_1 . This will be first order. But diamagnetic effect does not drastically change the behavior when $k_2 < k_1$.

In this geometry we find another curious behavior. We have the possibility of a second threshold at which the system goes from solution (b) to solution (c) or vice versa under certain conditions. It goes from $\phi = 0, \theta \neq 0$ to $\phi \neq 0, \theta = 0$ through a first order transition when $k_1 < k_2$ and $k_1 < k_3$. Similarly it goes from $\theta = 0, \phi \neq 0$ to $\theta \neq 0, \phi = 0$ again through a first order transition when $k_1 > k_2$ and $k_1 > k_3$ (see Figure 3).

COMBINED EFFECT OF ELECTRIC AND MAGNETIC FIELDS

In view of many interesting effects associated with Freedericksz transition in combined electric and magnetic fields,¹² it is worth looking at it in ferronematics also. We have considered the effects of an electric field applied parallel to the magnetic field. For simplicity we have assumed $k_1 = k_2 = k_3$. Up to a critical value of the electric field $E_c = 2\pi/d(k\pi/3\epsilon_a)^{1/2}$ the transition will continue to be second order. But at higher fields the transition will be first order thus exhibiting tricritical behavior as in the case of M . Figure 4 illustrates this feature.

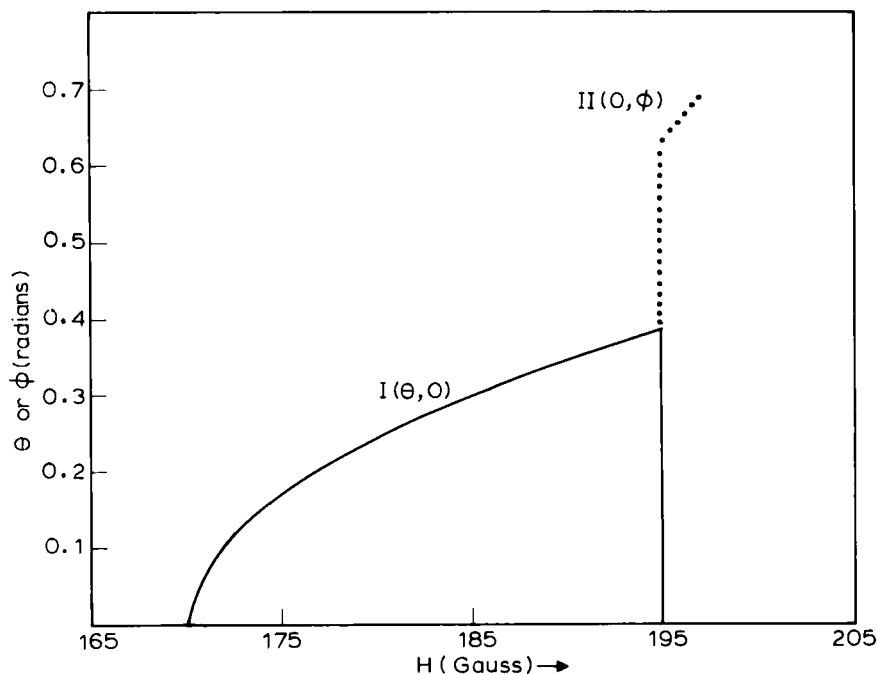


FIGURE 3 The classical second order Fredericksz transition with the second instability in the homogeneous geometry. The second transition is of first order and it is from splay-bend to a twist configuration. $d = 20 \mu\text{m}$, $k_1 = 0.55 \times 10^{-6}$ dyne, $k_2 = 0.6 \times 10^{-6}$ dyne, $k_3 = 1.5 \times 10^{-6}$ dyne, $M = 8 \times 10^{-3}$ Gauss, $\chi_a = 10^{-7}$ cgs units.

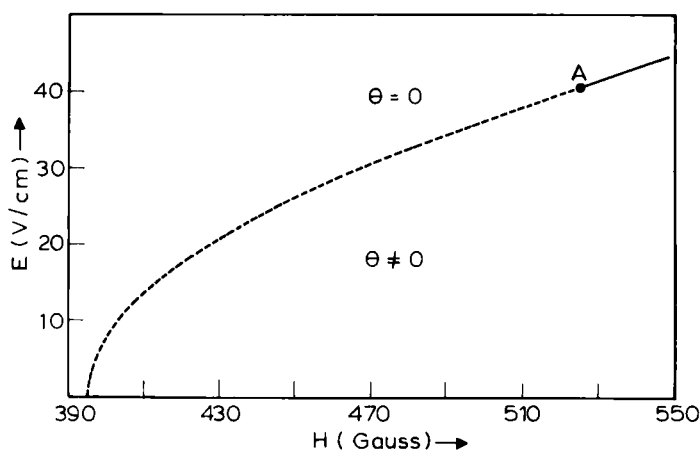


FIGURE 4 Tricritical behavior in parallel electric and magnetic fields in the homeotropic geometry. $d = 5 \mu\text{m}$, $k_1 = k_2 = k_3 = 10^{-6}$ dyne, $M = 0.1$ Gauss, $\epsilon_a = 0.1$ cgs units. The dashed line represents the second order transition and the full line the first order transition.

NEMATICS WITH NEGATIVE DIAMAGNETIC ANISOTROPY

Here one always gets a second order Freedericksz transition in the one constant approximation. Also we find the elastic anisotropy to result in a first order transition from θ to ϕ or ϕ to θ at a second threshold.

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