Nonequilibrium structures in the thin layers of ferronematics

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The onset of remagnetization of ferronematics (suspensions of stretched ferromagnetic colloidal particles in nematic solvent) filling a thin flat gap has been studied. We supposed that the average deviation of the particles axis from nematic director **n** is very small in each volume of the sample; both the initial nematic director $\mathbf{n}_0$ and the external magnetic field **H** being parallel to the plane of a gap contained by a sample. On the basis of a microscopic analysis and the Fokker-Planck equation for a ferromagnetic particle the macroscopic equation describing the kinetics of the suspension remagnetization has been derived. This equation includes an empirical relaxation time τ. With the help of this equation three cases have been considered. The first one concerns a suspension with zero initial magnetization and field **H** perpendicular to $\mathbf{n}_0$. The second one concerns a suspension with a frozen to a solvent nonzero magnetization (τ is greater than time of observation) and the field **H** directed such as in the first case. The third situation corresponds to nonzero initial magnetization $\mathbf{M}_0$ of a suspension, $\mathbf{M}_0$ being parallel to $\mathbf{n}_0$ and field **H** being antiparallel to $\mathbf{M}_0$. In the first and third cases remagnetization occurs as the second order phase transition only if H exceeds some critical value H_c and periodic structures can arise at the initial stage of this process if $H>H_{c1}>H_c$. In the second case ferronematic remagnetization is initiated by an infinitesimal magnetic field. The structures arising during this phase transition are homogeneous in both directions of $\mathbf{n}_0$ and **H**. [S1063-651X(98)02403-9]

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I. INTRODUCTION

The stable suspensions of nonspherical colloidal ferromagnetic particles in the nematic liquid crystals (ferronematics) attract considerable interest because their structure may be controlled by means of small magnetic fields, which is impossible for pure liquid crystals. Their physical properties have been investigated theoretically and experimentally, for example, in Refs. [1–7]. The equilibrium ferronematics Frederiks effect has been studied in Refs. [6,7]. The kinetics of such a phase transition has not yet been studied.

The main aim of the present research is the analysis of the initial stage of remagnetization in a ferronematic filling a thin flat gap. The analysis is based on the following assumptions.

First, all particles are identical and needlelike shaped. Their magnetic moments are constant at absolute values and are aligned along their symmetry axes.

Second, the interaction between ferromagnetic particles and the nematic solvent is very strong. As a result, all the particles are aligned along the nematic director **n** in each small volume element and average deviations of the particle axes from **n** are negligibly small.

Third, the particle concentration c is small enough to neglect their magnetic interaction with each other.

Fourth, the solvent molecules strongly interact with gap boundaries and on these surfaces vector **n** has a constant direction.

Finally, we shall neglect the interaction between the magnetic field and the solvent molecules.

It should also be noted that two limiting mechanisms exist for the ferroparticles reorientation in ferronematics under the presence of an external magnetic field. The first one is connected with the rotation of the particle with the nearest layer of liquid crystal. Ferroparticles with different orientations of magnetic moments turn in the field in different directions, therefore, this mechanism may lead to the occurrence of a variety of defects in a nematic matrix. The formation of such defects is very disadvantageous energetically, so the probability of this reorientation mechanism is very small.

In the second case each particle turns practically without any perturbation of the director field in the particle in the nearest vicinity. To do this, the particle needs to overcome a potential barrier corresponding to the perpendicular orientation of the particles and the director.

This mechanism of particle reorientation may be realized in practice if the thermal energy of the system is not negligibly small compared to the height of this barrier. We assume this method of particle reorientation.

II. EQUATION OF MAGNETIZATION CHANGE KINETICS

Our aim now is to derive the macroscopic kinetic equation for the absolute value of a ferronematic magnetization. Let us introduce a unit vector **e** aligned along the particle magnetic moment **m** and the distribution function $f(\mathbf{e})$ normalized to unity. The suspension magnetization **M** is

$$\mathbf{M}=mc\int \mathbf{e}f(\mathbf{e})d\mathbf{e}, \tag{1}$$

where m stands for the absolute value of the particle moment. The distribution function f may be determined from the Fokker-Planck equation [8]. Let us write this equation in the coordinate system connected with the nematic director:

$$\frac{\partial f}{\partial t}=\mathbf{K}\left(f\frac{D}{T}\mathbf{K}U\right)+\mathbf{K}D\mathbf{K}f, \tag{2}$$

$$\mathbf{K}=\left[\mathbf{e}\frac{\partial}{\partial\mathbf{e}}\right].$$

Here $U(\mathbf{e})$ is the particle potential energy and T is the absolute temperature in energetic units. In general, the rotational diffusivity D is a second rank tensor for a nematic system, its components are functions of $\mathbf{e}$. To simplify the calculations we suppose, as in [8], that D is a scalar constant. The order of value D will be determined below.

The potential energy U of the particle is defined as follows:

$$U=-(\alpha\mathbf{e})+U_n,\quad \alpha=\frac{m\mathbf{H}}{T},\tag{3}$$

where $\mathbf{H}$ is the magnetic field and $U_n(\mathbf{e})$ is the interaction energy between a particle and a nematic solvent. The explicit expression for U_n, as a rule, is unknown. Let φ be the angle between the particle axis and the director $\mathbf{n}$ and

$$U_n=-u_0w(\cos^2\varphi),\tag{4}$$

where $w(x)$ is a dimensionless function and u_0 is an amplitude of U_n. We assume that the interaction between a particle and a solvent is so strong that the probability of a particle to be aligned along $\mathbf{n}$ is much greater than that in other directions. In so doing, the function $w(x)$ is characterized by a sharp maximum at $x=1$ and minimum at $x=0$. Therefore, the strong inequalities

$$\delta u=u_0[w(1)-w(0)]\gg T,\quad \delta u\gg\alpha T\tag{5}$$

hold true. For the above mentioned reason, one may write (as was done in Refs. [9,10])

$$f\approx p_+\delta(\varphi-0)+p_-\delta(\varphi-\pi),\tag{6}$$

$$p_++p_-=1.$$

Here $p_\pm$ are the probabilities to find a particle magnetic moment aligned along the axes $\varphi=0$ and $\varphi=\pi$, respectively, and $\delta(x)$ is the delta function.

By using the results of Refs. [9,10] and taking into account the strong inequalities (5), we obtain from Eq. (1) at $D=\text{const}$ the equations for $p_\pm$:

$$\begin{aligned}&\frac{dp_+}{dt}=-\frac{dp_-}{dt}=-(W_+p_+-W_-p_-)\\&=-(W_++W_-)p_++W_-;\\&W_\pm=c_\pm\exp[-(U_{\max}-U_\pm)/T],\\&c_\pm=\frac{D}{T}\gamma_\pm(\gamma_m/2\pi T)^{1/2}\sin\varphi_m,\\&\gamma_\pm\approx\frac{d^2U}{d\varphi^2}(\varphi=0,\pi),\\&\gamma_m\approx-\frac{d^2U}{d\varphi^2}\left(\varphi=\frac{\pi}{2}\right),\end{aligned}\tag{7}$$

where $U_{\max}$ is the maximum value of U, φ_m is the value of φ corresponding to $U_{\max}$, and U_+ and U_- have the meanings of two minima of the energy U ($U_+<U_-$). Taking into account the inequalities (5), we may use the following approximations:

$$U_+=-u_0w(1)-\alpha_{||}T,\quad U_-=-u_0w(1)+\alpha_{||}T,$$

$$U_{\max}=-u_0w(0)-\alpha_\perp T,$$

$$\varphi_m=\frac{\pi}{2},$$

where $\alpha_{||}$ and $\alpha_\perp$ are the components of vector α parallel and normal to the director $\mathbf{n}$. According to Eq. (6), the mean absolute value of the particle magnetic moment $\langle m\rangle$ may be introduced as follows:

$$\langle m\rangle=m(p_+-p_-).$$

Thus, we may obtain the expression for the ferronematic magnetization

$$M=c\langle m\rangle=cm(p_+-p_-).\tag{8}$$

It must be noted that according to our assumptions the directions of magnetization $\mathbf{M}$ and $\mathbf{n}$ coincide in each small volume of the system.

Let us denote by $p_\pm^0$ the equilibrium values of $p_\pm$ corresponding to the field $\mathbf{H}$. In the approximation (5) we get

$$p_\pm^0\approx\frac{1}{2}\frac{\exp(\pm\alpha_{||})}{\cosh(\alpha_{||})}.\tag{9}$$

The equilibrium magnetization M^0, taking Eqs. (8) and (9) into account, is

$$M^0=cm(p_+^0-p_-^0)=cm\ \tanh(\alpha_{||}).\tag{10}$$

Substituting Eqs. (9) and (10) into Eqs. (7) and (8), we come to the equation for the magnetization:

$$\begin{aligned}&\frac{dM}{dt}=-\frac{1}{\tau}(M-M^0),\\&\frac{1}{\tau}=W_++W_-\sim D\exp\left(-\frac{\delta u}{T}+\alpha_\perp\right)\cosh(\alpha_{||}).\end{aligned}\tag{11}$$

Let us estimate an order of magnitude of relaxation time τ. The estimation for the diffusion coefficient D is

$$D\sim\frac{T}{\eta l d_p^2},$$

where η is the Miesowicz shear viscosity and l and d_p are the length and diameter of a particle ($l\gg d_p$). For the values $l\sim10^{-5}$ sm, $d_p\sim10^{-6}$ sm, and $\eta\sim1$ ps [11] we have the estimation $\tau\sim[0.1\exp(\delta u/T-\alpha_\perp)/\cosh(\alpha_{||})]$sec.

Strictly speaking, the relation (7) is valid only for the constant potential barrier in the plane $\varphi=\pi/2$. In general, if $\alpha\neq0$ this does not hold because the height of barrier $U_{\max}$ in the plane $\varphi=\pi/2$ approximately equals $W(0)-T(\alpha_\perp\mathbf{e})$.

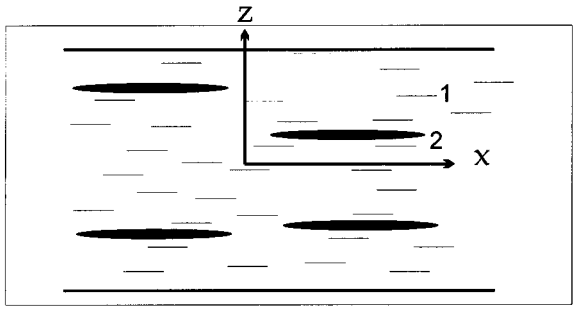

FIG. 1. Schematic representation of the studied system. (1) molecules of liquid crystal base; (2) ferromagnetic particles.

A strict derivation of equation similar to Eq. (7) for the systems characterized by an inconstant height of the potential barrier represents a very complicated problem. We do not know of any papers containing a constructional decision convenient for use in practice. However, it is clear that the main probability of flux during particle reorientation taking place in the neighborhood of a minimal point of the potential barrier. Therefore, in the first approximation $W_{\pm}$ may be estimated as if the height of the potential barrier was constant and equal to its height in a saddle point [in this case $-u_0\omega(0)-\alpha_{\perp}T$]. It is this approximation we have used. In this situation we obtain the upper estimate for the probability $W_{\pm}$ and lower estimate for the relaxation time τ. If α does not exceed unity, then our estimate does not lead to significant errors.

III. EQUATIONS OF NEMATODYNAMICS

Neglecting inertial effects, the standard equations of nematodynamics are [11]

$$\nabla\sigma=0, \quad \mathrm{div}\mathbf{v}=0, \tag{12}$$

$$[\mathbf{MH}]+[\mathbf{nh}]=\{\mathbf{n}[\gamma_1\mathbf{N}+\gamma_2(\mathbf{nA})]\},$$

$$\sigma_{ij}=-p\,\delta_{ij}+\alpha_1 n_i n_j A_{km} n_k n_m+\alpha_2 n_i N_j+\alpha_3 n_j N_i+\alpha_4 A_{ij}+\alpha_5 n_i n_k A_{kj}+\alpha_6 A_{ik} n_k n_j,$$

$$\mathbf{N}=\frac{d\mathbf{n}}{dt}+[\mathbf{n}\omega], \quad \omega=\frac{1}{2}\mathrm{rot}\mathbf{v}, \quad A_{ij}=\frac{1}{2}\left(\frac{\partial v_i}{\partial x_j}+\frac{\partial v_j}{\partial x_i}\right),$$

$$\mathbf{h}=K_1\nabla\mathrm{div}\mathbf{n}+K_2\{[\mathbf{n}\nabla(\mathbf{n}\mathrm{rot}\mathbf{n})]-2\mathrm{rot}\mathbf{n}(\mathbf{n}\mathrm{rot}\mathbf{n})\}+K_3\{\mathrm{rot}[\mathbf{n}[\mathbf{n}\mathrm{rot}\mathbf{n}]]-[\mathrm{rot}\mathbf{n}[\mathbf{n}\mathrm{rot}\mathbf{n}]]\},$$

$$\gamma_1=\alpha_3-\alpha_2, \quad \gamma_2=\alpha_6-\alpha_5, \quad i,j=x,y,z.$$

Here $\mathbf{v}$ is the velocity of a suspension, p is the pressure, $\alpha_1,\ldots,\alpha_6$ and K_1, K_2, K_3 are the Leslie and Frank coefficients of the nematic, and δ_{ij} is the Kroneeker symbol.

IV. THE INITIAL STAGE OF FREDERIKS TRANSITION IN DEMAGNETIZED FERRONEMATICS

We now consider a ferronematic inside a flat gap of thickness d (Fig. 1) and assume that the initial magnetization M_0 of the suspension is zero. Let us introduce a Cartesian coordinate system with the origin at the center of the gap. The axis Ox is parallel and axis Oz is normal to the boundary planes. We assume that the initial nematic director $\mathbf{n}_0$ is parallel to Ox and on the boundaries of a gap (i.e., $|z|=d/2$), and that the condition $n_x=1$ always holds. The constant magnetic field $\mathbf{H}$ aligned with the axis Oy is switched on at $t=0$. We assume that the dimensionless field is negligibly small ($\alpha\ll 1$), therefore $\tau(\alpha)\approx\tau(0)=\mathrm{const}$.

Experimental and theoretical investigations of the Frederiks effect kinetics in pure nematics [12–15] have shown that periodic nonequilibrium structures in the direction of axis Ox can occur in this process. Thus, we need to take into account the x and z dependencies of $\mathbf{n}$ and $\mathbf{v}$, but not the y dependence. The velocity $\mathbf{v}$ in this situation has only the component v_y [12–15].

Let us introduce the angle θ according to the formulas

$$n_x=\cos\theta, \quad n_y=\sin\theta.$$

Equations (11) and (12) become

$$\begin{aligned}\frac{\partial^2}{\partial t\partial x}[\theta(\alpha_3\sin^2\theta-\alpha_2\cos^2\theta)] &= \frac{\partial^2}{\partial x^2}\{v[2(\nu_1-\eta_4+\eta_a)\sin^2\theta\cos^2\theta \\ &+\eta_c\cos^4\theta+\eta_b\sin^4\theta]\}+\frac{\partial^2}{\partial z^2} \\ &\times[v(\eta_a\cos^2\theta+\eta_b\sin^2\theta)],\end{aligned} \tag{13}$$

$$\gamma_1\frac{\partial\theta}{\partial t}=\frac{\partial}{\partial x}[v(\alpha_3\sin^2\theta-\alpha_2\cos^2\theta)]+\frac{\partial^2}{\partial x^2}[\theta(K_1\sin^2\theta+K_3\cos^2\theta)]+K_2\frac{\partial^2\theta}{\partial z^2}+MH\cos\theta,$$

$$\frac{dM}{dt}=-\frac{\mu}{\tau}[M-\tanh(\alpha\sin\theta)],$$

$$\mu=mc, \quad \eta_a=\frac{1}{2}\alpha_4, \quad \eta_b=\frac{1}{2}(\alpha_3+\alpha_4+\alpha_6),$$

$$\eta_c=\frac{1}{2}(-\alpha_2+\alpha_4+\alpha_5), \quad \eta_4=\frac{1}{2}(\eta_b+\eta_c-\gamma_1),$$

$$\nu_1=\frac{1}{2}(\alpha_1+\alpha_4+\alpha_5+\alpha_6).$$

Here η_a, η_b, and η_c are the Miesowicz shear viscosities and η_1 is the elongated flow viscosity.

At the onset of ferronematic remagnetization the values of θ, v, and M/μ are small. We assume that $0<(H-H_c)\ll H_c$, where H_c is the critical field of the Frederiks phase transition. Therefore, we can write

$$\theta(x,z,t)=\cos qz\sum_k \theta_k\cos kx,$$

$$v(x,z,t)=\cos qz\sum_k v_k\sin kx, \tag{14}$$

$$M(x,z,t)=\cos qz\sum_k M_k\cos kx,\quad q=\frac{\pi}{d}.$$

Linearizing Eq. (13) over θ and v, taking Eq. (14) into account, we come to the following equation for the harmonic increment p_k:

$$\frac{d\theta_k}{dt}=\omega_k\theta_k+\frac{1}{\gamma_e}M_kH,$$

$$\frac{dM_k}{dt}=-\frac{1}{\tau}(M_k-mn\alpha\theta_k), \tag{15}$$

$$\omega_k=\frac{K_3k^2+K_2q^2}{\gamma_e},\quad \gamma_e=\gamma_1-\frac{(\alpha_2k)^2}{\eta_ck^2+\eta_aq^2}.$$

For all real nematics $\gamma_1\eta_c>\alpha_2{}^2$, therefore, $\gamma_e>0$.

The system (15) is valid provided that the characteristic values of θ_k and M_k are much more than their thermodynamical fluctuations. However, this condition fails for the onset of the Frederiks transition.

To account for the fluctuations of θ_k and M_k we introduce into Eq. (15) stochastic forces with zero average values:

$$\frac{d\theta_k}{dt}=\omega_k\theta_k+\frac{1}{\gamma_e}M_kH+\xi_k(t),$$

$$\frac{dM_k}{dt}=-\frac{1}{\tau}(M_k-mn\alpha\theta_k)+\zeta_k(t), \tag{16}$$

$$\langle\xi_k\rangle=0,\quad \langle\zeta_k\rangle=0.$$

From the fluctuation-dissipative theorem we have

$$\langle\xi_k(t)\xi_{k'}(t')\rangle=\frac{2T}{\gamma_eV}\delta(t-t')\delta_{kk'},$$

$$\langle\zeta_k(t)\zeta_{k'}(t')\rangle=\frac{2T\chi}{\tau V}\delta(t-t')\delta_{kk'}, \tag{17}$$

$$\langle\xi_k(t)\zeta_k(t')\rangle=0,\quad \chi=\frac{m^2c}{T}.$$

Here χ is the initial magnetic susceptibility of ferronematic.

Excluding M_k from Eq. (16), we obtain

$$\frac{d\theta_k(t)}{dt}+\omega_k\theta_k(t)-\frac{mn\alpha H}{\tau\gamma_e}\int_0^t\theta_k(t')\exp\left(-\frac{t-t'}{\tau}\right)=F(t), \tag{18}$$

$$F(t)=\frac{M_{k0}H}{\gamma_e}\exp\left(-\frac{t}{\tau}\right)+\frac{H}{\gamma_e}\int_0^t\exp\left(-\frac{t-t'}{\tau}\right)\zeta_k(t')dt'$$

$$+\xi_k(t),\quad M_{k0}=M_k(0).$$

Let us write θ_k in the following form:

$$\theta_k(t)=\Gamma(t)\theta_{k0}+\int_0^t\Gamma(t-t')F(t')dt',$$

$$\theta_{k0}=\theta_k(0),\quad \Gamma(0)=1,\quad \frac{d\Gamma}{dt}\Big|_{t=0}=-\omega_k \tag{19}$$

[the second condition for Γ follows from the system (15)].

Here Γ is the Green function. Substituting Eq. (19) into Eq. (18), we obtain

$$\Gamma(t)=\frac{\omega_k}{p_2-p_1}[\exp(p_1t)-\exp(p_2t)]+\exp(p_2t),$$

$$p_{1,2}=\frac{1}{2}\left[-\frac{1+\omega_k\tau}{\tau}\pm\sqrt{\left(\frac{1+\omega_k\tau}{\tau}\right)^2-4\frac{\omega_k\gamma_e-mn\alpha H}{\tau\gamma_e}}\right]. \tag{20}$$

The mean value $\langle\theta_{k0}\rangle$ (and therefore $\langle\theta_k\rangle$) is equal to zero. It is easy to show that $\langle F(t)\theta_{k0}\rangle=0$. Taking this into account we obtain the following equation for the second moment of θ_k:

$$\langle\theta_k(t)^2\rangle=\Gamma^2(t)\langle\theta_{k0}^2\rangle+\int_0^t\int_0^t\Gamma(t-t_1)\Gamma(t-t_2)$$

$$\times\langle F(t_1)F(t_2)\rangle dt_1dt_2. \tag{21}$$

The initial moments $\langle\theta_{k0}^2\rangle$ and $\langle M_{k0}^2\rangle$ are equal to the corresponding moments until the field H is switched on. They may be determined using standard thermodynamical considerations:

$$\langle\theta_{k0}^2\rangle=\frac{T}{(K_3k^2+K_2q^2)V},$$

$$\langle M_{k0}^2\rangle=\frac{T\chi}{V}. \tag{22}$$

Using Eqs. (17) and (18), one may obtain

$$\langle F(t_1)F(t_2)\rangle=\frac{H^2}{\gamma_e^2}\langle M_{k0}^2\rangle\exp\left(-\frac{t_1+t_2}{\tau}\right)$$

$$+2\frac{T\chi H^2}{V\gamma_e^2}\exp\left(-\frac{t_2-t_1}{\tau}\right)\Theta(t_2-t_1)$$

$$+\frac{2T}{V\gamma_e}\delta(t_2-t_1), \tag{23}$$

where $\Theta(x)$ is the theta function. Substituting Eqs. (20), (22), and (23) into Eq. (21), we come to the explicit expression for the second moment of $\theta_k(t)$.

If the increment p_1 is positive, then the moment $\langle\theta_k^2\rangle$ increases with time, if negative then it decreases. The increment p_1 is more than zero provided that

$$mcH\alpha=cT\alpha^2>K_3k^2+K_2q^2. \tag{24}$$

Let

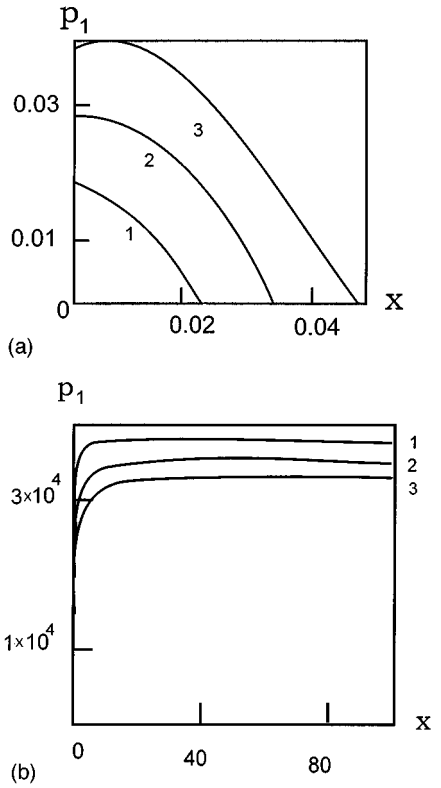

FIG. 2. Plots of p_1 ($\sec^{-1}$) vs $x=k^2/q^2$. (a) $\pi=10$ swx; $1-\alpha=0.008\,48\,4039$, $2-\alpha=0.008\,48\,4041$, $3-\alpha=0.008\,48\,4043$. (b) $\alpha=0.01$; $1-\tau=0.1$ sec, $2-\tau=10\,000$ sec, $3-\tau=20\,000$ sec. The system parameters are $c=10^{16}$ cm^{-3}, $T=300°$ Kk_B, $d=10^{-2}$ cm. The elastic and viscous characteristic of nematic are the same as for the PBG system [13]: $K_1=12.1\times10^{-7}$ dyn, $K_2=0.78\times10^{-7}$ dyn, $K_3=7.63\times10^{-7}$ dyn; $\eta_a=1.74P$, $\eta_b=0.37P$, $\eta_c=69.4P$, $\gamma_1=69.4\ P$.

$$\alpha_c=q\sqrt{\frac{K_2}{cT}}. \tag{25}$$

If $\alpha>\alpha_c$ then p_1 is positive for all $k^2<(cT\alpha^2-K_2q^2)/K_3$ and, therefore, the Frederiks transition occurs. This result for the critical field of Frederiks phenomenon corresponds to the results of equilibrium thermodynamical theory (see, for example, [11]).

In Fig. 2 the results are shown of calculation of p_1 as a function of k. If α is less than some critical $\alpha'>\alpha_c$, then $p_1(k)$ is a monotonically decreasing function. It means that the harmonic with $k=0$ has the maximum velocity of growth and the ferronematic will be homogeneous along the axis Ox during the Frederiks transition. But if $\alpha>\alpha'$, the function $p_1(k)$ have a maximum at $k_m\neq0$. Therefore the harmonics with $k=k_m$ has a maximum growth velocity and the nonequilibrium structures periodical along Ox will occur at the Frederiks transition. Earlier, such a conclusion was made in [12–15] for a pure molecular nematic with $\tau=0$. The plots of p_1 as function of τ are given in Fig. 3.

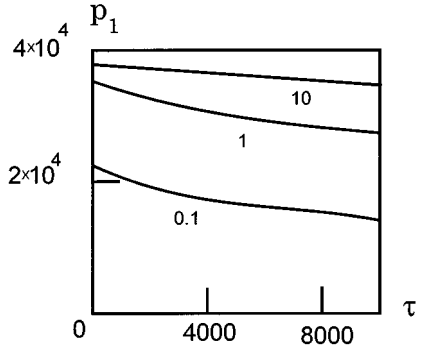

FIG. 3. Plot of p_1 ($\sec^{-1}$) as a function on τ (sec). The physical parameters of the system are the same as for Fig. 2. The figures near curves are equal to $(k/q)^2$.

V. THE INITIAL STAGE OF REMAGNETIZATION OF RIGIDLY MAGNETIZED FERRONEMATICS

We assume here that before the field **H** is switched on the ferronematic is magnetized along the Ox axis and the characteristic time τ is much greater than the time of the experiment. In this situation $M=\text{const}$ throughout the phase transition. At the initial stage of the process, Eqs. (13) reduce to

$$-\alpha_2\frac{\partial^2\theta}{\partial t\partial x}=\eta_c\frac{\partial^2 v}{\partial x^2}+\eta_a\frac{\partial^2 v}{\partial z^2},$$

$$\gamma_1\frac{\partial\theta}{\partial t}=-\alpha_2\frac{\partial v}{\partial x}+K_3\frac{\partial^2\theta}{\partial x^2}+K_2\frac{\partial^2\theta}{\partial z^2}+MH,$$

$$\theta\ll1,\quad M=\text{const}, \tag{26}$$

$$\theta(z,t=0),v(z,t=0)=0,\quad \theta(z=\pm d/2,t),$$

$$v(z=\pm d/2,t)=0.$$

We shall seek the solution of the problem (19) in the form

$$\theta=\sum_k w_k(z,t)\cos kx,\quad v=\sum_k u_k(z,t)\sin kx. \tag{27}$$

If the system is infinite in the direction of x then all harmonics with $k\neq0$ will dissipate. Thus the only harmonic with $k=0$ is of interest to us. In distinction to the preceding situation with zero initial magnetization, the small thermodynamical fluctuations of θ and M are not of principal importance for us now.

Coupling Eq. (27) with Eq. (26), we have

$$\alpha_2 k\frac{\partial w_k}{\partial t}=-\eta_c k^2 u_k+\eta_a\frac{\partial^2 u_k}{\partial z^2},$$

$$\gamma_1\frac{\partial w_k}{\partial t}=\alpha_2 k u_k-K_3 k^2 w_k+K_2\frac{\partial^2 w_k}{\partial z^2},\quad k\neq 0,$$

$$\gamma_1\frac{\partial w_0}{\partial t}=K_2\frac{\partial^2 w_0}{\partial z^2}+MH, \tag{28}$$

$$w_k(z,0),\ \ u_k(z,0)=0,\quad w_k\left(\pm\frac{d}{2},t\right),$$

$$u_k\left(\pm\frac{d}{2},t\right)=0,\ \text{for all}\ \ k.$$

The solution of Eq. (28) is

$$w_k=0,\ \ k\neq 0,$$

$$w_0=\frac{4MHd^2}{K_2}\sum_{n=1,3,\ldots,}^{\infty}\frac{1}{(\pi n)^3}\left[1-\exp\left(-\frac{K_2}{\gamma_1}q_n^2 t\right)\right]\times\sin\left[q_n\left(z+\frac{d}{2}\right)\right], \tag{29}$$

$$q_n=\frac{\pi n}{d}.$$

As follows from Eq. (29), the distortion of nematic structure can occur now in the infinitesimal magnetic field (the same conclusion was arrived at the analyses [6,7] of the equilibrium state of ferronematics) and ferronematic has a homogeneous structure in the gap plane during remagnetization.

VI. THE FIELD H ANTIPARALLEL TO INITIAL MAGNETIZATION

Let at the initial moment the nematic director $\mathbf{n}_0$ and ferronematic magnetization $\mathbf{M}_0$ be aligned with the axis Ox. At $t=0$ the external magnetic field $\mathbf{H}$ antiparallel to $\mathbf{M}_0$ is switched on. Our aim again is the investigation of the initial stage of the $\mathbf{n}$ reorientation when the angle θ between $\mathbf{n}$ and $\mathbf{n}_0$ is very small. We assume that M_0 is much more than its thermodynamical fluctuations and neglect them.

During reorientation $\mathbf{n}$ may rotate in a sufficiently complex manner and may be unhomogeneously dependent on radius vector $\mathbf{r}$. But if θ is small then the movement of $\mathbf{n}$ may be interpreted as a superposition of its rotation in plane (x,y) (with structures periodical along the axis Ox and axis Oy) and rotation in plane (x,z). Let us study these types of movement of $\mathbf{n}$ separately.

A. Rotation of n in plane (x,y). Structures periodical along axis Ox

The suspension velocity $\mathbf{v}$ now must be aligned with axis Oy and periodical along Ox. The nematodynamic equations (12) in this situation are

$$\frac{\partial}{\partial t}\frac{\partial}{\partial x}[\theta(\alpha_3\sin^2\theta-\alpha_2\cos^2\theta)]=\frac{\partial^2}{\partial x^2}\{v[2(\nu_1-\eta_4+\eta_a)\sin^2\theta\cos^2\theta+\eta_c\cos^4\theta+\eta_b\sin^2\theta]\}+\frac{\partial^2}{\partial z^2}[v(\eta_a\cos^2\theta+\eta_b\sin^2\theta)], \tag{30}$$

$$\gamma_1\frac{\partial\theta}{\partial t}=\frac{\partial}{\partial x}[v(\alpha_3\sin^2\theta-\alpha_2\cos^2\theta)]+\frac{\partial^2}{\partial x^2}[\theta(K_1\sin^2\theta+K_3\cos^2\theta)]+K_2\frac{\partial^2\theta}{\partial z^2}+MH\sin\theta,$$

$$\alpha_2=\frac{1}{2}(\eta_b-\eta_c-\gamma_1),\quad \alpha_3=\frac{1}{2}(\eta_b-\eta_c+\gamma_1),$$

$$\eta_4=\frac{1}{2}(\eta_b+\eta_c-\gamma_1).$$

For this situation in the linear approximation by H, Eq. (11) is

$$\frac{\partial M}{\partial t}=-\frac{1}{\tau}(M+\chi H\cos\theta). \tag{31}$$

Here we take into account the sign of $\mathbf{H}$ projection into axis Ox.

In the linear approximation by θ and v Eqs. (30) and (31) become

$$-\alpha_2\frac{\partial}{\partial t}\frac{\partial}{\partial x}\theta=\eta_c\frac{\partial^2 v}{\partial x^2}+\eta_a\frac{\partial^2 v}{\partial z^2},$$

$$\gamma_1\frac{\partial\theta}{\partial t}=\alpha_2\frac{\partial v}{\partial x}+K_3\frac{\partial^2\theta}{\partial x^2}+K_2\frac{\partial^2\theta}{\partial z^2}+MH\theta \tag{32}$$

$$\frac{\partial M}{\partial t}=-\frac{1}{\tau}(M+\chi H).$$

Solving the last equation, we obtain

$$M=-\chi H+(M_0+\chi H)\exp\left(-\frac{t}{\tau}\right). \tag{33}$$

Substituting Eq. (33) into the first and second equations of Eq. (32), we come to the following system:

$$-\alpha_2\frac{\partial}{\partial t}\frac{\partial}{\partial x}\theta=\eta_c\frac{\partial^2 v}{\partial x^2}+\eta_a\frac{\partial^2 v}{\partial z^2},$$

$$\gamma_1\frac{\partial\theta_1}{\partial t}=\alpha_2\frac{\partial v}{\partial x}+K_3\frac{\partial^2\theta}{\partial x^2}+K_2\frac{\partial^2\theta}{\partial z^2}+H(M_0+\chi H)\theta\exp\left(-\frac{t}{\tau}\right)-\chi H^2\theta. \tag{34}$$

Below we assume that $(H-H_c)/H_c\ll 1$, where H_c is the critical field of reorientation [1,11]. We seek the solution of Eq. (34) in the form

$$\theta=\sum_k \theta_k(t)\cos kx\cos qz, \quad v=\sum_k v_k(t)\sin kx\cos qz. \tag{35}$$

Substituting Eq. (35) into Eq. (34), one obtains

$$\alpha_2 k\frac{\partial\theta_k}{\partial t}=-(\eta_c k^2+\eta_a q^2)v_k,$$

$$\gamma_1\frac{\partial\theta_k}{\partial t}=-\alpha_2 k v_k-(K_3k^2+K_2q^2)\theta_k+H(M_0+\chi H)\exp\left(-\frac{t}{\tau}\right)\theta_k-\chi H^2\theta_k. \tag{36}$$

Excluding v from Eq. (36), we come to

$$\frac{\partial\theta_k}{\partial t}=\omega_k(t)\theta_k, \tag{37}$$

$$\omega_k=\left[H\tau(M_0+\chi H)\exp\left(-\frac{t}{\tau}\right)-\chi H^2-K_2q^2-K_3k^2\right]\beta_k^{-1},$$

$$\beta_k^{-1}=\frac{\eta_c k^2+\eta_a q^2}{(\gamma_1\eta_c-\alpha_2^2)k^2+\eta_a\gamma_1 q^2}.$$

It must be noted that for all real systems $\beta_k>0$.

Until this time the fluctuations of θ_k were not taken into account. To account for them let us introduce into Eq. (37) the random force $\xi_k(t)$ and rewrite this equation as follows:

$$\beta_k\frac{\partial\theta_k}{\partial t}=\beta_k\omega_k\theta_k+\xi_k. \tag{38}$$

The correlation relations for $\langle\xi_k\rangle$ are given in Eq. (17).

Using Eqs. (38) and (17), we obtain

$$\langle\theta_k^2(t)\rangle=\left(\langle\theta_{0k}^2\rangle+\frac{4T}{\beta_k V}\int_0^t\exp[-2g_k(t')]dt'\right)\exp[2g(t)],$$

$$g_k(s)=\int_0^s\omega_k(s')ds'=\frac{G(s)-K_3k^2s}{\beta_k}, \tag{39}$$

$$G(s)=H\tau(M_0+\chi H)\left[1-\exp\left(-\frac{s}{\tau}\right)\right]-(\chi H^2+K_2q^2)s.$$

The initial value $\langle\theta_{0k}^2\rangle$ of the moment $\langle\theta_k^2\rangle$ may be determined according to thermodynamical theory of fluctuations. Using the standard results of this theory, we get the following expression:

$$\langle\theta_{0k}^2\rangle=\frac{2T}{V}(K_3k^2+K_2q^2+M_0H_0)^{-1}, \tag{40}$$

where $\mathbf{H}_0$ is the magnetic field up to the moment $t=0$ when the field $\mathbf{H}$ is switched on (we assume that $\mathbf{H}_0$ is parallel to $\mathbf{M}_0$).

At $t/\tau\ll 1$,

$$g_k\approx\frac{1}{\beta_k}(M_0H-K_2q^2-K_3k^2)t. \tag{41}$$

If $h=M_0H-K_2q^2>0$ then g_k is positive for all $k<\sqrt{h/K^3}$. As follows from Eq. (39), the corresponding moment $\langle\theta_k^2\rangle$ increases. So the nematic reorientation will occur if

$$H>H_c=\frac{K_2q^2}{M_0} \tag{42}$$

and H_c is the critical field of this ferronematic remagnetization. This result is in agreement with the conclusions of Refs. [1,11].

The value $k=k_c$ corresponding to the maximum of positive g_k, is

$$k_c^2=\frac{1}{2a}(-b+\sqrt{b^2+4\varphi a}),$$

$$a=\frac{\eta_c(\gamma_1\eta_c-\alpha_2^2)}{\gamma_1\eta_a^2q^4}\frac{K_3t}{G(t)}, \quad b=\frac{2\eta_c}{\eta_a q^2}\frac{K_3t}{G(t)}, \tag{43}$$

$$\varphi=\frac{\alpha_2^2}{\gamma_1\eta_a q^2}-\frac{K_3t}{G(t)}.$$

Physically admissible values of $k_c^2>0$ correspond to $\varphi>0$ or, in other words,

$$H\tau(M_0+\chi H)\left[1-\exp\left(-\frac{t}{\tau}\right)\right]-(\chi H^2+K_2q^2)t>\frac{\eta_a\gamma_1}{\alpha_2^2}K_3q^2t. \tag{44}$$

At $t/\tau \ll 1$ the inequality (44) holds when

$$H > H_{c1} = H_c\left(1 + \frac{\eta_a \gamma_1}{\alpha_2^2}\frac{K_3}{K_2}\right). \tag{45}$$

Therefore if $H > H_{c1}$ then g_k as a function from k has a maximum at $k = k_c$ and nonequilibrium structures periodical along the axis Ox will arise in this ferronematic at the onset of its remagnetization.

Let k_m be the value of k corresponding to the maximum $\langle \theta_k^2 \rangle$. It is easy to show from Eqs. (39), (40), and (43) that $k_m = 0$ at $t = 0$ and $k_m \to k_c$ with the increasing t. As k_c is not a constant (but a decreasing function of t), the period of harmonic θ_k with a maximum amplitude will decrease at the onset of reorientation with the subsequent increase.

B. Rotation of n in (x,y) plane. Structures periodical along axis Oy

In this case the solvent velocity is directed along axis Ox. Instead of Eq. (30) the nematodynamic equations (12) are now

$$\begin{aligned}\frac{\partial}{\partial t}\frac{\partial}{\partial y}[\theta(\alpha_3\cos^2\theta - \alpha_2\sin^2\theta)] &= \frac{\partial^2}{\partial y^2}\{v[2(\nu_1 - \eta_4 + \eta_a)\sin^2\theta\cos^2\theta \\ &+ \eta_c\sin^4\theta + \eta_b\cos^2\theta]\} + \frac{\partial^2\theta}{\partial z^2} \\ &\times [v(\eta_a\sin^2\theta + \eta_b\cos^2\theta)],\end{aligned} \tag{46}$$

$$\gamma_1\frac{\partial\theta}{\partial t} = \frac{\partial}{\partial y}[v(\alpha_3\cos^2\theta - \alpha_2\sin^2\theta)] + \frac{\partial^2}{\partial y^2}[\theta(K_1\cos^2\theta + K_3\sin^2\theta)] + K_2\frac{\partial^2\theta}{\partial z^2} + MH\sin\theta.$$

Repeating the reasonings of Sec. VI A we obtain the following expressions instead of Eqs. (39) and (40):

$$\theta = \sum_k \theta_k \cos ky \cos qz,$$

$$\langle \theta_k^2(t) \rangle = \left(\langle \theta_{0k}^2 \rangle' + \frac{4T}{\beta_k' V}\int_0^t \exp[-2g_k'(t')]dt'\right) \times \exp[2g_k'(t)], \tag{47}$$

$$g_k'(s) = \frac{G(s) - K_1k^2 s}{\beta_k'},$$

$$\beta_k' = \gamma_1 - \frac{\alpha_2^2 k^2}{\eta_b(k^2 + q^2)},$$

and

$$\langle \theta_{0k}^2 \rangle' = \frac{T}{V}(K_1k^2 + K_2q^2 + M_0H_0)^{-1}. \tag{48}$$

Here $G(s)$ is the same function as in Eq. (39). Instead of Eq. (43) we obtain now

$$(k_c')^2 = \frac{1}{2a'}[-b' + \sqrt{(b')^2 + 4\varphi' a'}],$$

$$a' = \frac{\gamma_1\eta_b - \alpha_3^2}{\gamma_1\eta_b^2 q^4}\frac{K_1 t}{G(t)}, \quad b' = \frac{K_1 t}{q^2 G(t)}, \tag{49}$$

$$\varphi' = \frac{\alpha_3^2}{\gamma_1\eta_b q^2} - \frac{K_1 t}{G(t)}.$$

At $t/\tau \ll 1$ Eq. (49) has admissible solutions if

$$H > H_{c2} = H_c\left(1 + \frac{\eta_b\gamma_1}{\alpha_3^2}\frac{K_3}{K_2}\right).$$

If $H > H_{c2}$ and $H_{c1} > H_{c2}$ then $g_{k_c'}' > g_k$ and nonequilibrium structures periodic along Oy will arise at the onset of ferronematic remagnetization. If $H > H_{c1}$ and $H_{c2} > H_{c1}$, these structures will be periodical along Oy. It is necessary to note that these structures cannot occur at times $t \gg \tau$, so it is very difficult to observe them in pure molecular nematics where τ, as a rule, is very small.

The rotation of n in the plane (x,z) may be studied just as was done in Sec. VI A or VI B. The analysis shows that in this case $\langle \theta_k^2 \rangle$ is much less than that in Eqs. (41) and (47).

VII. CONCLUSIONS

If the initial nematic director $\mathbf{n}_0$ and external magnetic field $\mathbf{H}$ are parallel to the plane of a gap filled by the ferronematic under the condition of $\mathbf{H} \perp \mathbf{n}_0$, then ferronematic magnetization occurs as second phase transition (the Frederiks effect) provided that the magnetic field is larger than the critical one, and nonequilibrium structures periodic along $\mathbf{n}_0$ can occur at the onset of a Frederiks transition. The growth of these structures decreases with an increase of the characteristic time τ of the suspension remagnetization. If τ is large enough, these structures cannot arise. Similar structures cannot occur in rigidly magnetized ferronematics. If nonzero initial magnetization $\mathbf{M}_0$ is frozen in the solvent with the magnetic field $\mathbf{H}$ perpendicular to $\mathbf{M}_0$ but parallel to the switched on gap plane, then the ferronematic remagnetization is induced by an infinitesimal field and the solvent structure will be homogeneous during this process.

If the ferronematic has a nonzero initial magnetization $\mathbf{M}_0$ parallel to the gap plane and the field $\mathbf{H}$ antiparallel to $\mathbf{M}_0$ is switched on, then nonequilibrium structures may occur in the ferronematic at the onset of its remagnetization provided that the field exceeds the critical one.

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