

## Time dependence of the magnetic grain concentration and secondary grain aggregation in ferronematic lyotropic liquid crystals subjected to magnetic field gradients

C. Y. Matuo\* and A. M. Figueiredo Neto

*Instituto de Física, Universidade de São Paulo, Caixa Postal 66318, 05315-970, São Paulo, SP, Brazil*

(Received 8 June 1998; revised manuscript received 15 March 1999)

The dynamical behavior of lyotropic nematic liquid crystals doped with ionic and surfacted magnetic fluids (*ferronematics*) is studied using a linear optical technique. The response of these mesophases to a combination of a static and a pulsed magnetic field is investigated by measuring the relaxation times as a function of the pulse width. A reversible modification of the magnetic grain concentration in the bulk of the samples and a secondary aggregation process due to the presence of a field gradient introduced by the pulsed field is discussed. [S1063-651X(99)04008-8]

PACS number(s): 61.30.Gd, 75.50.Mm, 78.20.Nv

### I. INTRODUCTION

Lyotropic ferronematic liquid crystals [1] are obtained by the doping of usual lyotropic nematic liquid crystals [2] with ferrofluids [3,4]. These ferrofluids, or magnetic complex fluids, are colloidal suspensions of small magnetic grains (typical dimension  $D_3 \sim 100 \text{ \AA}$ ) dispersed in a liquid carrier. In usual ferrofluids, the typical concentration of magnetic grains is about  $10^{16}$  grains/cm<sup>3</sup>, corresponding to the volume fraction of magnetic material  $\phi \sim 10^{-2}$ . Two different types of ferrofluids are available: surfacted and ionic ferrofluids. In the first case, the grains (usually Fe<sub>3</sub>O<sub>4</sub>) are coated with surfactant agents to prevent their flocculation [5]. In ionic ferrofluids, the magnetic grains (usually  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub>; MnFe<sub>2</sub>O<sub>3</sub>; CoFe<sub>2</sub>O<sub>3</sub>) are electrically charged to keep the colloid stable [6]. When a ferrofluid is highly diluted in a solvent, it became unstable and the grains flocculate.

It was theoretically predicted [1] and experimentally verified [7,8] that the doping of liquid crystals with ferrofluids (above a critical concentration) reduces the magnetic field required to orient the liquid crystal by  $10^3$  times. The doping of thermotropic liquid crystals with ferrofluids is a very delicate task, due to the low solubility of them in thermotropics. On the other hand, the doping of lyotropics with water base ferrofluids was done for the first time in 1979 by Liébert and Martinet [7] and since then this method has been used [9–11] to investigate the physical-chemical properties of lyotropics.

A serious problem that experimentalists usually face when they dope lyotropics with a small quantity of ferrofluids (usually 1  $\mu$ l of ferrofluid per 1 ml of the liquid crystal) is to control the stability of this doping with time *and* with the applied magnetic field. It is known that when a magnetic field gradient is applied in a ferronematic sample [12], the bulk concentration of magnetic grains varies. In ferrofluids, even a *uniform* magnetic field favors the formation of small chains. This process of chaining is one of the responsables

for the optical birefringence observed in these materials in the presence of a magnetic field [10]. In usual ferrofluids, this chaining process is reversible and, after the removal of the field, the grains became isolated again. The chaining itself does not change  $\phi$  in the sample subjected to a uniform magnetic field, but the grains cannot be considered isolated anymore. The cluster's hydrodynamic volume is a function of the field and the aggregate is now the basic unit that responds to the applied field. This aspect is essential when magnetic fluids are used to determine the local viscosity of a liquid by means of a "ferrofluid viscometer" [13]. This method is based upon the measurement of the induced birefringence as a function of the time, when a field is applied and after removed from a ferrofluid doped sample. The local viscosity ( $\eta$ ) is obtained from the measurement of the relaxation time ( $\tau_R$ ) of the induced birefringence, knowing the basic unit (usually assumed to be the isolated grain) hydrodynamic radius ( $R_H$ ). As the relevant parameter on this experiment is the hydrodynamic volume,  $\eta$  is proportional to  $R_H^3$ , and an uncertainty in  $R_H$  can introduce a large uncertainty in  $\eta$ .

To our knowledge, a systematic study of the effect of a small field gradient on  $\phi$  and  $R_H$ , particularly the time interval during which it is applied to a ferronematic sample, was not yet reported. In this work we use a linear optical technique to study the dynamical behavior of the magnetic grains (ionic and surfacted) in a ferronematic lyotropic liquid crystal subjected to spatially nonuniform and time-varying magnetic fields. The paper is organized as follows. The next section describes the dynamical model used to interpret the experimental results; after that, the experimental section brings the details of the setup and the samples used; finally, the results are presented and discussed, followed by the conclusion.

### II. THEORY

We use the dynamical model proposed in Ref. [14]. The grains have a typical dimension  $D_3$ , magnetic moment  $\mu$ , and magnetization at saturation  $m_s$ . The liquid crystalline medium is characterized by an elastic constant  $K$  (in the one-constant approximation [15]). In the presence of a uniform magnetic field  $\mathbf{H}_1$ , the magnetic moment of the grains (as-

\*Permanent address: Depto. de Física, Centro de Ciências Físicas e Matemáticas, Universidade Federal de Santa Catarina, C.P. 476, 88040-900 Florianópolis, SC, Brazil.

sumed to be fixed in the crystalline structure of the grain) tend to align parallel to  $\mathbf{H}_1$ . If  $\theta(t)$  is the angle between  $\mu$  and  $\mathbf{H}_1$ , then the torque balance equation per unit area can be written as

$$\gamma D_1 \dot{\theta} = -\frac{K\theta}{D_2} - D_1 m_s H_1 \theta \phi, \quad (1)$$

where  $\gamma$  is the ferronematic viscosity;  $\dot{\theta} = d\theta/dt$ ; and  $D_1$  and  $D_2$  are typical dimensions [14] of rotation and distortion around the grains, respectively.  $\phi$  is assumed to be independent of the time and of the field  $H_1$ . Due to the chaining process (which we will call hereafter *primary*),  $D_1$  and  $D_2$  are functions of  $H_1$ , but time independent.

The solution of the differential equation (1) is

$$\theta(t) = \theta_0 \exp(-t/\tau), \quad (2)$$

where  $\theta_0$  is a constant and

$$\tau^{-1} = \frac{K}{\gamma D_1 D_2} + \frac{m_s}{\gamma} \phi H_1. \quad (3)$$

The distortion length  $D_2$  depends on the shape and shape anisotropy of the grain, and the local boundary conditions of the director at the grain's surface [16]. Here we assume that  $D_1 \sim D_2$  ( $\sim 2 R_H$ ) and Eq. (3) is written as

$$\tau^{-1} = \frac{K}{\gamma (D_1)^2} + \frac{m_s}{\gamma} \phi H_1. \quad (4)$$

For small values of  $\theta$ , the optical transmittance ( $I$ ) is proportional to  $\theta^2$  [17]:

$$I \propto \sin^2(2\theta) \propto \theta^2. \quad (5)$$

In this limit and taking into account Eq. (2),  $I$  can be written as

$$I \propto \exp(-2t/\tau). \quad (6)$$

Thus, if the transmittance decay is due to only one physical process, we expect that the transmittance as a function of time can be described by only one exponential function. In the case of more than one physical process,  $I(t)$  can be described by a sum of exponentials:

$$I_{exp2} = a_0 \exp\left(-\frac{2(t-t_0)}{\tau_1}\right) + b_0 \exp\left(-\frac{2(t-t_0)}{\tau_2}\right), \quad (7)$$

where  $a_0$  and  $b_0$  are constants,  $t_0$  is the initial time, and  $\tau_1$  and  $\tau_2$  are the characteristic response (relaxation) times.

### III. EXPERIMENTAL SECTION

#### A. Samples

The lyotropic liquid crystal used is a mixture of potassium laurate (28.74 wt.%), decanol (6.64 wt.%), and water (64.62 wt.%) in the calamitic nematic phase ( $N_c$ ). The phase sequence of this mixture as a function of the temperature is *isotropic* (12 °C)  $N_c$  (35 °C) *isotropic*. The experiments are performed at 22 °C. The ferrofluid doping is done with

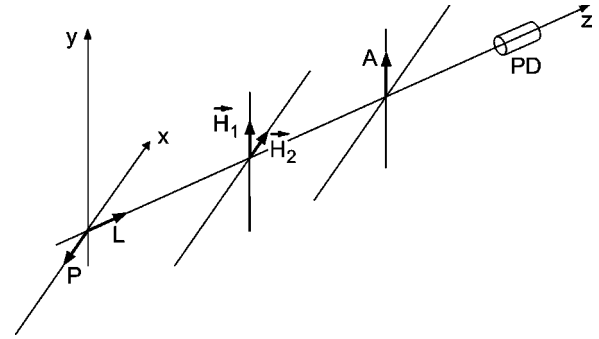


FIG. 1. Sketch of the setup. P, A, L, and PD are the polarizer, analyzer, laser beam direction, and photodetector, respectively.  $x$ ,  $y$ , and  $z$  are the laboratory frame axes.

both types of water-base ferrofluids (ionic and surfacted). The ionic ferrofluid (gently furnished by Dr. Bee from the Université Pierre et Marie Curie, Paris) has  $\gamma$ - $\text{Fe}_2\text{O}_3$  magnetic grains with a mean diameter of 95 Å and  $m_s = 442$  G. The surfacted ferrofluid (from Ferrofluidics Corporation) has  $\text{Fe}_3\text{O}_4$  magnetic grains, a mean diameter of 100 Å and  $m_s = 100$  G, double-coated with oleic acid.

The initial grain concentration in the samples is  $(7.9 \pm 0.9) \times 10^{13}$  grains/cm<sup>3</sup>, corresponding to  $\phi = (3.5 \pm 0.6) \times 10^{-5}$  and  $\phi = (4.1 \pm 0.7) \times 10^{-5}$  for the ionic and surfacted samples, respectively.

The ferronematics are encapsulated inside rectangular glass microslides (from *Vitro Dynamics*) with the following dimensions: 25 mm (length), 4 mm (width) and  $(400 \pm 40)$  μm (thickness).

#### B. The setup

The experimental setup is sketched in Fig. 1. It consists in a polarized (along the  $x$  direction) HeNe cw laser beam (10 mW), two sources of magnetic fields, an analyzer (parallel to the  $y$  direction), and a photodetector connected to a computer. The laser beam direction is parallel to the  $z$  axis (normal to the biggest flat surface of the microslide) and its waist at the sample's position is about 1 mm. The magnetic field  $\mathbf{H}$  is a superposition of two independent fields. A static and homogeneous field [ $(\mathbf{H}_1$  oriented along the  $y$  axis) (Fig. 1)] generated by an electromagnet ( $10 \leq H_1 \leq 3000$  G). The poles of the electromagnet are of circular cross section (diameter is 5 in.) and the homogeneity of the field at the sample position is better than 1/1000. A second magnetic field  $\mathbf{H}_2$  (oriented along the  $x$  axis) is a pulsed field (strength is between 0 to 550 G) generated by two Helmholtz coils. This pulsed field gives rise to a field gradient symmetric with respect to the center ( $x=y=0$ ) of the sample. The measured magnetic field gradient from  $x=y=0$  towards the borders is from 2 to about 10 G/mm along the  $x$  direction and from 1 to 6 G/mm along the  $y$  direction, for  $H_2$  varying from 150 to 550 G.  $\mathbf{H}_1$  is oriented parallel to the longest axis of the sample holder and the direction of the analyzer. The strength and the time interval of the pulse (typically from 1 to 90 s) are controlled by a pulse generator (square wave). We checked that during the time of the experiment the temperature of the sample did not vary more than 0.3 °C due to the heating of the electromagnet and the Helmholtz coils.

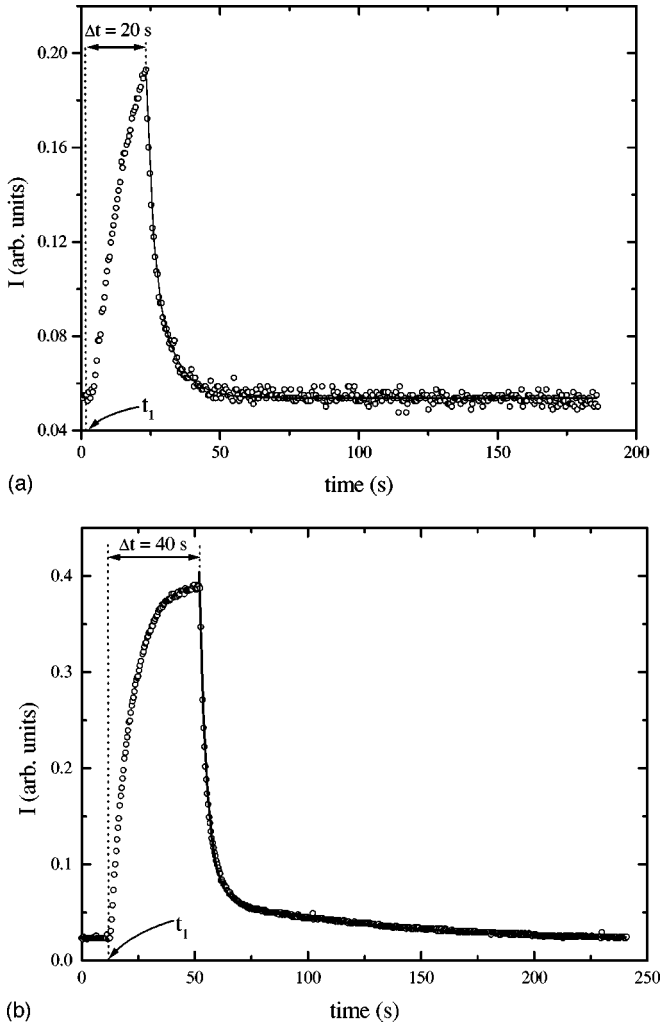


FIG. 2. Typical optical transmittance as a function of time. (a) Ionic ferronematic with  $c = (7.9 \pm 0.9) \times 10^{13}$  grains/cm<sup>3</sup> and  $\Delta t = 20$  s. The solid line is a fit with Eq. (6); (b) ionic ferronematic with  $c = (7.9 \pm 0.9) \times 10^{13}$  grains/cm<sup>3</sup> and  $\Delta t = 40$  s. The solid line is a fit with Eq. (7).

### C. Experimental procedure

The experimental procedure consists of measuring the transmittance of the sample, subjected to a magnetic field, as a function of time. Initially,  $\mathbf{H}_1$  ( $= 3000$  G, the highest field used in this experiment) is applied for about three hours for a complete alignment of the sample's director in the  $y$  direction. Modifications in  $\phi$  are not expected at this level. In this geometry, the light transmittance ( $I$ ) of the sample (between crossed polarizers), measured by the photodiode, is minimum. After that,  $\mathbf{H}_1$  is fixed at a given value and, keeping  $\mathbf{H}_1$  turned on, the magnetic field  $\mathbf{H}_2$  (strength of  $H_2/H_1 = 0.2$ ) is applied in a direction perpendicular to  $\mathbf{H}_1$  during a time interval  $\Delta t$  ( $1 < \Delta t < 90$  s). This pulse induces a grain's (and/or aggregate's) rotation, and the sample's optical axis tends to align in the direction defined by the total field  $\mathbf{H}_1 + \mathbf{H}_2$ . In this case, the maximum rotation angle of the sample's director is about 0.2 rad. This tilt gives an increase of the light transmittance. Besides this effect, as  $H_2$  introduces a field gradient in the sample, the grains (and/or aggregates) migrate from the center to the edges (due to the particular field geometry used) of the sample, modifying  $\phi$ .

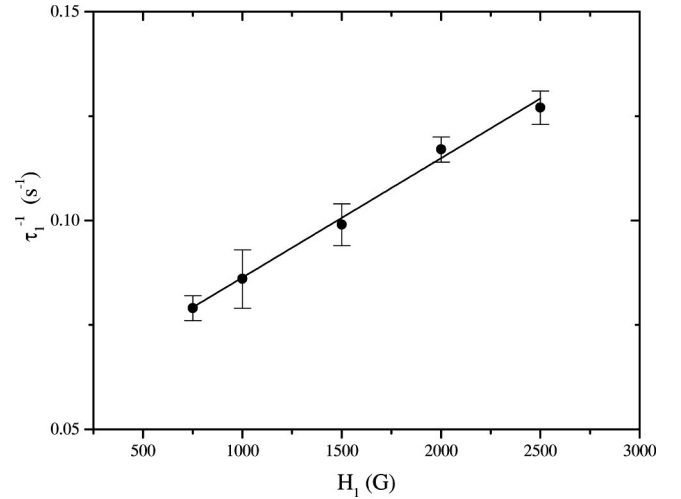


FIG. 3. Typical behavior of  $\tau_1^{-1}$  as a function of  $H_1$ . Ionic ferronematic sample of concentration  $c = (7.9 \pm 0.9) \times 10^{13}$  grains/cm<sup>3</sup>, with a pulse width of  $\Delta t = 20$  s. The solid line represents a linear fit with Eq. (4).

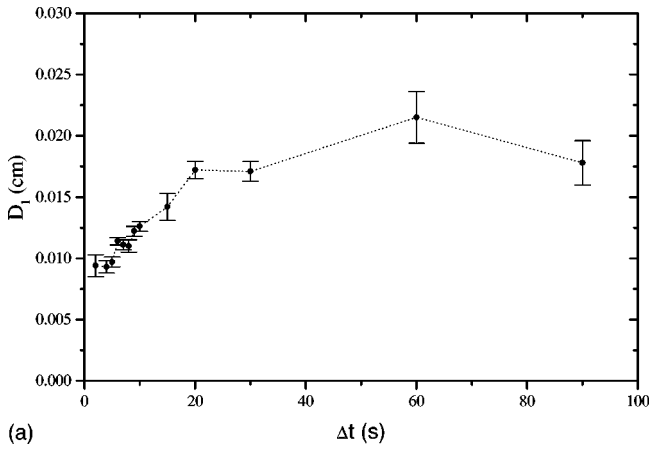
When the magnetic pulse ( $\mathbf{H}_2$ ) is turned off, the transmittance returns to its minimum value. Measurements of  $I(t)$  are performed, changing the strength of magnetic field  $\mathbf{H}_1$  ( $750 < H_1 < 2750$  G) and the field gradient, keeping the ratio  $H_2/H_1$  constant. Each value of  $\phi$  and  $D_1$  is obtained from a series of about six measurements of the relaxation times for different values of the fields. Each complete set of measurements takes about 30 min.

## IV. RESULTS AND DISCUSSION

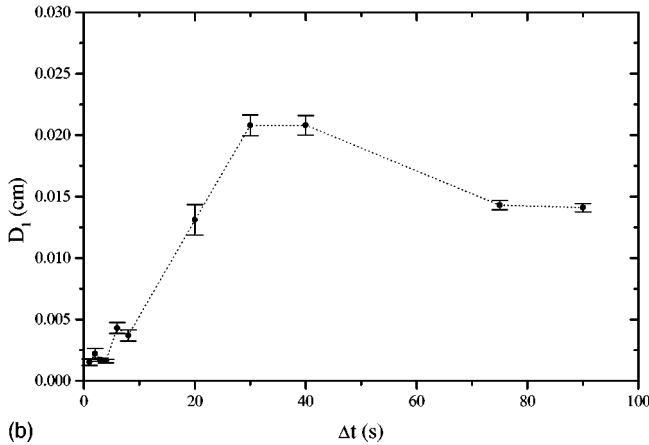
Hereafter, the nematic doped with ionic (or surfacted) ferrofluids will be called ionic (or surfacted) ferronematic. In the absence of  $H_2$ ,  $\phi$ ,  $D_1$ , and  $D_2$  are expected to be independent of the time, and the model described in Sec. II can be used to interpret the dynamical behavior of the sample.

Figure 2 shows two typical curves of  $I(t)$ . For  $t < t_1$ , only  $\mathbf{H}_1$  is present; at  $t_1$ ,  $\mathbf{H}_2$  is applied for an interval of time  $\Delta t$ . After this time interval,  $\mathbf{H}_2$  is switched off and the sample's director returns to the original orientation parallel to  $\mathbf{H}_1$ . In the case of Fig. 2(a),  $\Delta t$  ( $= 20$  s) is not enough to allow the complete orientation of the nematic director ( $\mathbf{n}$ ) parallel to  $\mathbf{H}$ . In Fig. 2(b) this complete orienting process ( $\mathbf{n} \parallel \mathbf{H}$ ) is achieved. Our experimental results show that it is not possible to fit the transmittance decrease (after  $\mathbf{H}_2$  is switched off) with only *one* exponential function in the case of  $\Delta t > 20$  s [for example, Fig. 2(b)]. In this case, Eq. (7) is used to obtain the characteristic reorienting (relaxation) times  $\tau_1$  and  $\tau_2$  for each pair ( $\Delta t$ ,  $H_1$ ). In the case of  $\Delta t < 20$  s, however, the decrease of  $I$  as a function of  $t$  can be fitted by a single exponential function [Eq. (6)]. This behavior is observed in both ionic and surfacted ferronematics.

The existence of two characteristic times in lyotropic nematic liquid crystals (without ferrofluid) was previously observed [18]. These characteristic times had been associated with the two physical processes of the sample's orientation, one in the bulk (with  $\tau \sim 10^2$  s) and another at the surface (with  $\tau \sim 10^5$  s). This large characteristic time [18] refers to a gliding anchoring of  $\mathbf{n}$  onto the glass surfaces of the sample



(a)

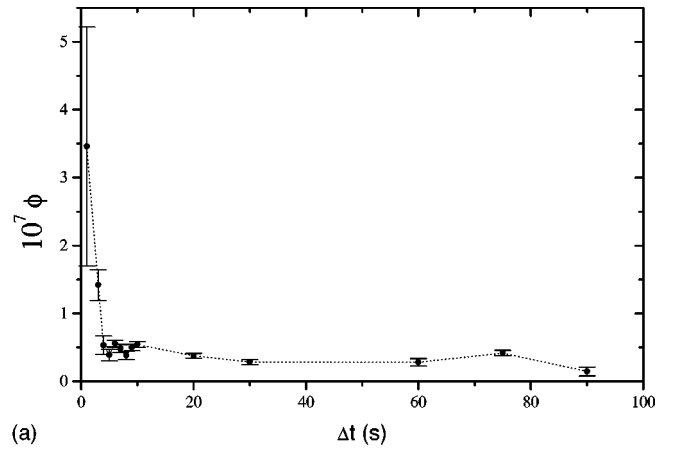


(b)

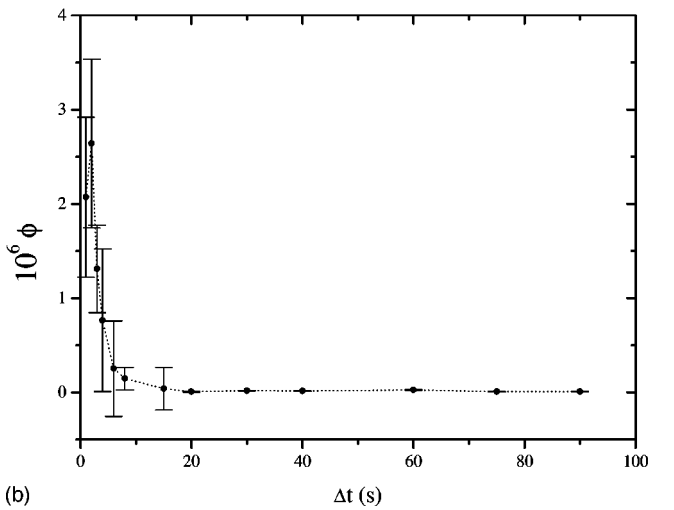
FIG. 4. Rotation (distortion) characteristic dimension  $D_1$  as a function of  $\Delta t$ . (a) Ionic ferronematic sample with a concentration  $c = (7.9 \pm 0.9) \times 10^{13}$  grains/cm<sup>3</sup>. The dotted line is only an eye guide. (b) Surfacted ferronematic sample with a concentration  $c = (7.9 \pm 0.9) \times 10^{13}$  grains/cm<sup>3</sup>. The dotted line is only a guide to the eye.

holder. Our results (for  $\Delta t > 20$  s) will be interpreted in this context. We will associate the smaller time ( $\tau_1$ ) to the process occurring in the sample's bulk and the larger one ( $\tau_2$ ) to the surface gliding of the director in the glass-liquid crystal interface. For values of  $\Delta t$  below  $\sim 20$  s (for all the values of  $H$  investigated), only the bulk of the sample experiences a significant variation in the director's orientation.

Figure 3 shows a typical result of  $\tau_1^{-1}$  as a function of  $H_1$  for the ionic ferronematic sample (surfacted ferronematic samples give similar results). The errors are evaluated taking into account not only the fitting procedure, but also the reproducibility of the experiment. The linear behavior predicted by Eq. (4) is observed with all the values of  $\Delta t$  investigated. These results show that the response of ferronematic liquid crystals to applied magnetic fields is different from the conventional lyotropic nematics. The nematic phase without the doping presents characteristic relaxation times proportional to  $H^{-2}$  [18]. Using the known values of  $m_s$ , the lyotropic nematic elastic constant  $K(10^{-6}$  dyn) [19] and the measured viscosity [ $\gamma = (0.58 \pm 0.02)$  poise at 22 °C] [20], the dependences of  $D_1$  and  $\phi$  with  $\Delta t$  are obtained [Eq. (4)]. Comparing the results of  $D_1$  versus  $\Delta t$  (Fig. 4) and  $\phi$  versus  $\Delta t$  (Fig. 5), obtained with both ferronematics, we note that the qualitative behavior is the same,  $D_1$  increases in the be-



(a)



(b)

FIG. 5. Volumetric fraction  $\phi$  as a function of  $\Delta t$ . (a) Ionic ferronematic sample of concentration  $c = (7.9 \pm 0.9) \times 10^{13}$  grains/cm<sup>3</sup>. The dotted line is a guide to the eye. (b) Surfacted ferronematic sample of concentration  $c = (7.9 \pm 0.9) \times 10^{13}$  grains/cm<sup>3</sup>. The dotted line is a guide to the eye.

ginning of the process when the field gradient is applied and after it tends to be independent of  $\Delta t$ ;  $\phi$  decreases in the beginning of the process when the field gradient is applied and after it tends to be independent of  $\Delta t$ .

Figures 4(a) and 5(a) show the pulse width dependence of  $D_1$  and  $\phi$ , respectively, in the case of the ionic ferronematic. The values of these parameters obtained in our experimental conditions are *mean values* within the laser spot. The errors are determined from error propagation, taking into account also the reproducibility of the experiment. Figure 4(a) shows the variation of  $D_1$  as a function of  $\Delta t$ . The characteristic length of rotation (distortion)  $D_1$  increases from  $\sim 1 \times 10^{-2}$  up to  $\sim 2 \times 10^{-2}$  cm with increasing  $\Delta t$ . This increase of  $D_1$  is an indication that the grains tend to agglomerate in the presence of the field gradient, since it measures the typical dimension of a distorted volume around the grain. Besides this effect, the field gradient promotes a modification on  $\phi$  with time. Initially, in the absence of the magnetic field gradient, the ferronematic has  $\phi = (3.5 \pm 0.6) \times 10^{-5}$ . In the presence of the field gradient, the

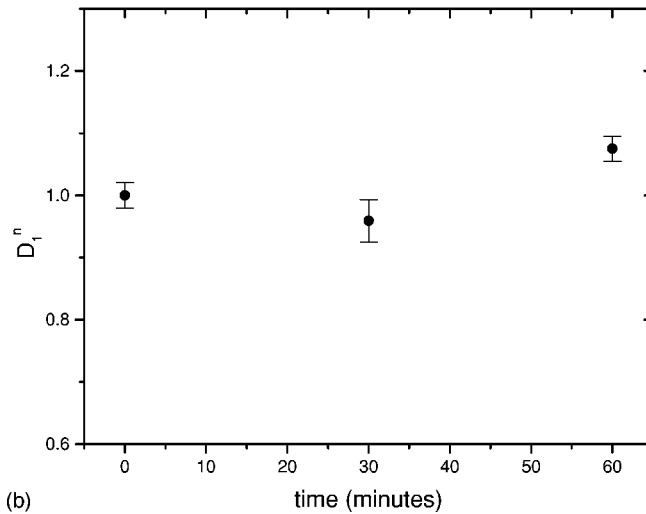
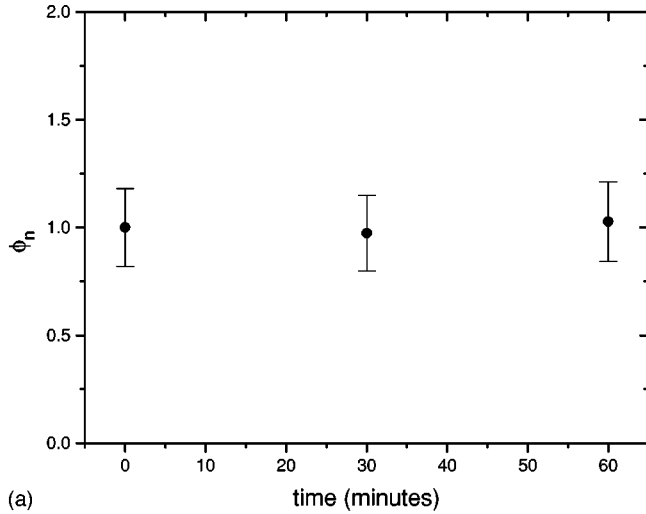


FIG. 6. (a) Normalized volumetric fraction  $\phi$  of three consecutive sets of measurements with the same sample as a function of time. Each set takes 30 min to be performed. Surfacted ferronematic sample,  $\Delta t = 20$  s. (b) Normalized  $D_1$  of three consecutive sets of measurements with the same sample as a function of time. The same set of measurements as (a).

values of  $\phi$  decrease from  $3.5 \times 10^{-7}$  ( $\Delta t = 1$  s) to values around  $3 \times 10^{-8}$  ( $\Delta t = 90$  s). This result is due to the migration of magnetic grains from the center of the sample to the borders of the sample holder. This method of changing the grains' concentration in a ferronematic sample has been used [12] to measure the effect of a surface field on the value of the splay-bend elastic constant.

The same linear dependence of  $\tau_1^{-1}$  with  $H_1$  is observed in the case of surfacted ferronematics. Figures 4(b) and 5(b) show  $D_1$  versus  $\Delta t$  and  $\phi$  versus  $\Delta t$ . Surfacted ferronematics present larger values of  $\tau_1$  than the ionic ferronematic (from 7 to 10 times larger, depending on the strength of  $H_1$ ). The errors are obtained following the same procedure described before. To check this grain's migration process we measured  $\phi$  at different positions of the sample in the plane perpendicular to the laser beam. With a micrometric positioner we change the place of the incident beam in the sample. In a perimeter of 1 mm far from the center of the

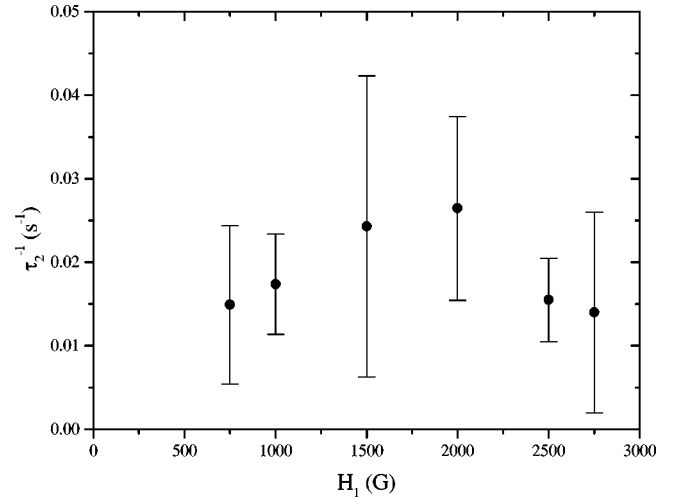


FIG. 7. Typical behavior of  $\tau_2^{-1}$  as a function of  $H_1$ . Ionic ferronematic sample of concentration  $c = (7.9 \pm 0.9) \times 10^{13}$  grains/cm<sup>3</sup>, with a pulse of  $\Delta t = 20$  s.

sample ( $x = y = 0$ ), we measured an increase in  $\phi$  of about 30% with respect to the center ( $\Delta t = 20$  s). To verify if after the switching off of  $H_2$  the grains diffuse back into the bulk of the cell and the secondary aggregation is reversible, we measured consecutively a series of three  $\phi$  and  $D_1$ , at the same sample position, as a function of time ( $\Delta t = 20$  s). Each complete set of measurements which allows the determination of  $\phi$  and  $D_1$  takes about 30 min. Figures 6(a) and 6(b) show the normalized volume fraction of magnetic material [ $\phi_n(t) = \phi(t)/\phi(0)$ , where  $\phi(0)$  is the first value of  $\phi$  obtained], and the normalized  $D_1$  [ $D_1^n(t) = D_1(t)/D_1(0)$ , where  $D_1(0)$  is the first value of  $D_1$  obtained], respectively, as a function of time. An inspection of Fig. 6(a) indicates that, at least for the field gradients and fields used in our experiment, the grain's migration process is *reversible*, i.e., after the removal of the gradient the sample behaves like a freshly made one.  $\phi$  obtained depends essentially on  $\Delta t$  and consecutive measurements give the same  $\phi$ . The same conclusion seems to be applicable to  $D_1$ , which, despite the experimental points, showed a larger dispersion [Fig. 6(b)] compared to the  $\phi$  results [Fig. 6(a)].

Comparing the results of  $\phi$  versus  $\Delta t$  of both ferronematics [Fig. 5] we note that, for the smallest  $\Delta t$ , the surfacted ferronematic presents  $\phi$  about 10 times larger than the ionic ferronematic. It is interesting to note that the initial value of  $\phi$  (ferronematic in the absence of a magnetic field gradient) is  $10^2$  times (ionic ferronematic) and 10 times (surfacted ferronematic) larger than the values measured even with  $\Delta t = 1$  s. Figure 4 indicates that when the ferronematic sample (both, ionic and surfacted) is subjected to a magnetic field gradient, the chaining process is much more intense in the beginning, and after tends to stabilize. After about 20 s in the field gradient,  $\phi$  and  $D_1$  are almost independent of  $\Delta t$ .

As discussed before, in usual ( $\phi \sim 10^{-2}$ ) ionic ferrofluids, the magnetic grains have a surface charge density to prevent their aggregation. In the surfacted ferrofluids, however, the magnetic grains are coated with surfactant agents, and the repulsion between them is mainly steric. The basic units of the lyotropic liquid crystals are micelles (aggregates of am-

phiphilic molecules). Besides the micelles, isolated amphiphilic molecules exist in the bulk of the nematic phase. It was previously observed in lyotropic nematics [18,21] that a flat surface can stabilize a lamellar layer on it. This process was described as a surface-screening effect [21] where the amphiphilic molecules form a carpet over the glass interface. As the grains in surfacted ferrofluids are already coated with molecules having the same characteristics of the lyotropic amphiphilic molecules, the grains could also be surrounded by an extra layer constituted by amphiphilic molecules of the lyotropic. In this case, the effective size of the grains could increase and their mobility in the nematic matrix should be smaller. It could explain why  $D_1$  and the typical relaxation times are bigger in surfacted ferronematics than in ionic ferronematics.

The values of  $\tau_2$ , obtained [Eq. (7)] when  $\Delta t \geq 20$  s with both ferronematics, are of the order of  $10^2$  s, practically independent of  $\Delta t$ , and do *not* follow the behavior predicted by Eq. (4) (Fig. 7). The errors are evaluated as discussed before. This characteristic time is three orders of magnitude smaller than the time associated with the director gliding process in nondoped lyotropic nematic samples [18]. In this context, the ferrofluid doping and the migration of magnetic grains from the bulk to the interface (due to the field gradient) could modify the anchoring properties of the liquid crystal. Ionic and surfacted ferronematics present  $50 < \tau_2 < 150$  s, and  $100 < \tau_2 < 300$  s, respectively. Assuming that these times are associated with the gliding of the director in the interface solid-liquid crystal, these results indicate that the anchoring of surfacted ferronematics is stronger than ionic ferronematics.

## V. CONCLUSIONS

It was verified that both types of ferronematic liquid crystals (ionic and surfacted) in the presence of a magnetic field presents only one response time, associated with the bulk orientation of the director, when  $\Delta t < 20$  s. With  $\Delta t > 20$  s, in both doping cases, two response times are observed. One of them, associated with a bulk process, scales as  $H^{-1}$  and, another one, almost independent of  $H$ , is associated with a surface process. The presence of a small magnetic field gradient (during a time interval  $\Delta t$ ) reduces the volume fraction of magnetic material  $\phi$  in the sample, the greater  $\Delta t$ , the smaller  $\phi$  in the center of the sample. For the same  $\Delta t$ ,  $\phi$  is bigger in the surfacted ferronematic than in the ionic one. The ionic ferronematic's relaxation times are smaller than the surfacted ones. It was verified also that the distortion length  $D_1$  increases in both types of ferronematics as a function of  $\Delta t$ . This result is interpreted as an indication of a secondary chaining process (since a primary one is due to the presence of the uniform field) due to the presence of the magnetic field gradient. After the removal of the small field gradient, the grains diffuse back to the bulk of the sample and the aggregation process occurring during  $\Delta t$  is undone.

## ACKNOWLEDGMENTS

We are indebted to Professor J. C. Bacri and Dr. A. Bee from Jussieu (Paris) for helpful discussions and for kindly supplying the ionic ferrofluids. Fundação de Amparo à Pesquisa do Estado de São Paulo (FAPESP), Fundação Vitae, COFECUB-USP, and PRONEX are acknowledged for financial support. Dr. A. Roy is acknowledged for helpful discussions.

- 
- [1] F. Brochard and P. G. de Gennes, *J. Phys. (Paris)* **31**, 691 (1970).
- [2] P. G. de Gennes and J. Prost, *The Physics of Liquid Crystals*, 2nd ed. (Clarendon Press, Oxford, 1993).
- [3] R. E. Rosensweig, *Ferrohydrodynamics* (Cambridge University Press, Cambridge, 1985).
- [4] S. W. Charles and J. Popplewell, in *Ferromagnetic Materials*, edited by E. P. Wohlfarth (North-Holland Publishing Company, Amsterdam, 1980), Vol. 2.
- [5] R. Massart, US Patent No. 4329241 (May 1982).
- [6] R. Massart, *IEEE Trans. Magn.* **MAG-17**, 1247 (1981).
- [7] L. Liébert and A. Martinet, *J. Phys. (France) Lett.* **40**, L-363 (1979).
- [8] A. M. Figueiredo Neto and M. M. F. Saba, *Phys. Rev. A* **34**, 3483 (1986).
- [9] A. M. Figueiredo Neto, Y. Galerne, A. M. Levelut, and L. Liébert, in *Physics of Complex and Supermolecular Fluids*, edited by S. Safran and N. A. Clark, EXXON Monograph Series (Wiley, New York, 1987), p. 347.
- [10] C. Y. Matuo, F. A. Tourinho, and A. M. Figueiredo Neto, *J. Magn. Magn. Mater.* **122**, 53 (1993).
- [11] A. M. Figueiredo Neto, in *Phase Transitions in Complex Fluids*, edited by P. Tolédano and A. M. Figueiredo Neto (World Scientific, Singapore, 1998), p. 175.
- [12] S. Fontanini, A. L. Alexe-Ionescu, G. Barbero, and A. M. Figueiredo Neto, *J. Chem. Phys.* **106**, 6187 (1997).
- [13] J. C. Bacri, J. Dumas, D. Gorse, R. Perzinski, and D. Salin, *J. Phys. (France) Lett.* **46**, L-1199 (1985).
- [14] J. C. Bacri and A. M. Figueiredo Neto, *Phys. Rev. E* **50**, 3860 (1994).
- [15] E. B. Priestley, in *Introduction to Liquid Crystals*, edited by E. B. Priestley, P. I. Wojtowicz, and P. Sheng (Plenum Press, New York, 1979).
- [16] S. U. Burylov and Y. L. Raikher, *J. Magn. Magn. Mater.* **122**, 62 (1993).
- [17] M. Born and E. Wolf, *Principles of Optics*, 6th ed. (Pergamon Press, London, 1980).
- [18] E. A. Oliveira, A. M. Figueiredo Neto, and G. Durand, *Phys. Rev. A* **44**, R825 (1991).
- [19] T. Kroin, A. J. Palangana, and A. M. Figueiredo Neto, *Phys. Rev. A* **39**, 5373 (1989).
- [20] The viscosity of the ferronematic is measured using an Ostwald viscosimeter.
- [21] S. Fontanini, A. Strigazzi, G. Barbero, M. C. Salvadori, and A. M. Figueiredo Neto, *Liq. Cryst.* **24**, 793 (1998).