Theory of the magnetohydrodynamic effect in nematic liquid crystals

A. N. Kuznetsov and T. P. Kulagina

Research Institute of Biological Tests of Chemical Compounds
(Submitted October 29, 1974)
Zh. Eksp. Teor. Fiz. 68, 1501-1505 (April 1975)

The behavior of nematic liquid crystals in a rotating magnetic field of arbitrary intensity is investigated within the framework of the swarm theory. The two-dimensional model of liquid crystals permits one, at least qualitatively, to explain the experimental results.

PACS numbers: 61.30.

Investigations of the behavior of nematic liquid crystals in a rotating magnetic field permits a study of their magnetic and hydrodynamic properties.

In Tsvetkov's first works,[1,2] devoted to the magnetohydrodynamic effect, it was observed that when the magnetic field is rotated relative to the sample the liquid-crystal medium rotates together with the field in such a way that the angle between the sample optical axis and the direction of the rotating magnetic field **H** remains constant in time. With increasing rotation frequency ω, the angle α increases in such a way that the values of $\sin 2\alpha$ are proportional to ω, whereas the summary moment M of the forces exerted by the magnetic field on the sample increases in direct proportion to ω (see the experimental plots in Fig. 1). At sufficiently high rotation frequencies, the moment of the forces reaches a maximum and then decreases with further increase of the frequency. In this case the liquid-crystal medium ceases to rotate uniformly with the field.

The theory proposed by Tsvetkov[1] for the effect is based on the swarm model, according to which the liquid-crystal medium constitutes an aggregate of mutually oriented molecules that are gathered into rather large group (swarms) by the intermolecular interaction. Anisotropic interactions between swarms are neglected. The swarms are axially symmetrical and are characterized, in particular, by an axially-symmetrical diamagnetic-susceptibility tensor. A constant magnetic field acting on the liquid-crystal sample orients the swarms in space, and in sufficiently strong (saturating) magnetic fields the sample becomes uniformly oriented. An investigation of the behavior of such a uniformly oriented sample in a rotating magnetic field, carried out by Tsvetkov,[1] has shown that the sample can have a rotation that is stationary in time only at frequencies ω lower than a certain critical value

$$\omega_{cr}=v(\chi_\parallel-\chi_\perp)H^2/2h. \qquad (1)$$

Here v is the volume of the swarm, $\chi_\parallel - \chi_\perp$ is the difference between the principal values of the diamagnetic susceptibilities, and h is a friction coefficient and depends on the shape and dimensions of the swarm, and also on the viscosity of the medium. The values of $\sin 2\alpha$ and M are determined in this case by the relations

$$\sin 2\alpha=\omega/\omega_{cr}, \quad M={}^1/_2 v(\chi_\parallel-\chi_\perp)H^2\frac{\omega}{\omega_{cr}}. \qquad (2)$$

If $\omega = \omega_{cr}$, then $\sin 2\alpha$ and M reach their maximum values

$$(\sin 2\alpha)_{max}=1, \quad M_{max}={}^1/_2 v(\chi_\parallel-\chi_\perp)H^2. \qquad (3)$$

At $\omega > \omega_{cr}$, the rotation should cease to be uniform and, as shown in [3], the time-averaged moment $\langle M\rangle$ of the forces should decrease with increasing rotating-field frequency like

$$\langle M\rangle=M_{max}\Big/\left[\frac{\omega}{\omega_{cr}}+\sqrt{\frac{\omega^2}{\omega_{cr}^2}-1}\right]. \qquad (4)$$

At sufficiently low rotation frequencies, relations (2) previously derived in [1] make it possible to explain the experimental dependences of $\sin 2\alpha$ and M on ω, but at frequencies corresponding to the maximum value of the moment, relations (2) and (3) do not account for the experimental results (see Fig. 1).

Up to now, the disparity between theory and experiment was attributed to the dependence of the constants in (2) on the rotation frequency. This, however, cannot explain the disordering of the sample at $\alpha < 45°$, which was observed in [4] and was subsequently fully confirmed in [5,6]. The premature violation of the homogeneity of the sample can be attributed to the action of an unsaturated magnetic field on the liquid-crystal sample. In an unsaturated magnetic field, the ordering of the sample depends on the action of the projection of the rotating magnetic field on the swarm orientation axis. During the rotation, the angle between the swarm axis and the field direction increases, and this leads to a decrease of the projection of the magnetic field on the optical axis of the swarm and to additional disorientation of the sample.

The purpose of the present study was to investigate the behavior of a nematic liquid crystal in rotating magnetic fields of arbitrary intensity, within the framework of the swarm theory.[1)]

For simplicity, we confine the analysis to the model of a two-dimensional crystal, in which it is assumed that all the optical axes of the swarms of the liquid crystal lie in the same plane. The orientation of these axes in the sample is characterized by a dis-

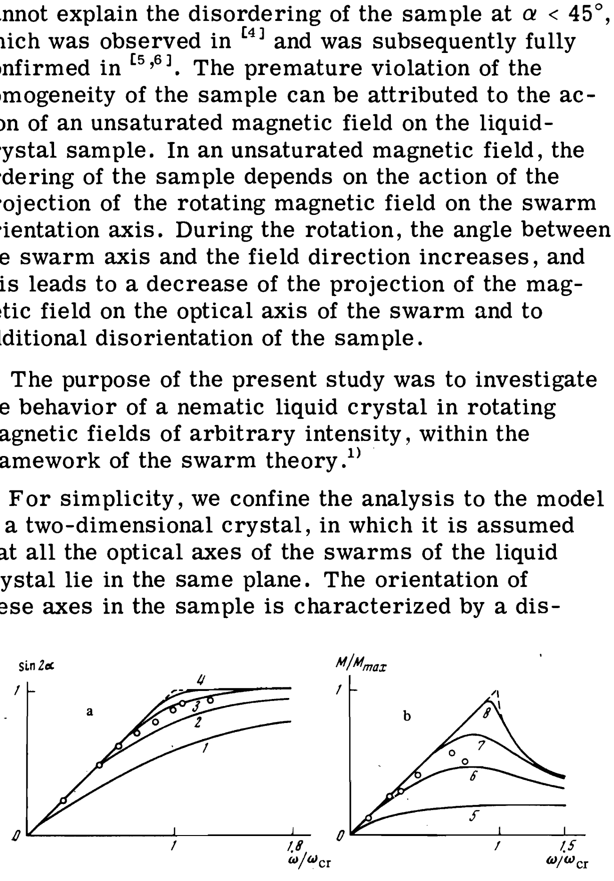

FIG. 1. Dependence of $\sin 2\alpha$ (Fig. a) and of M/M_{max} (Fig. b) on ω/ω_{cr} at various values of the parameter A: 1, 5) A = 1, 2, 6) A = 3; 3, 7) A = 10; 4, 8) A = 100; ○) experimental data from [4,5]: a) for para-azoxyphenetole at H = 3350 Oe; b) for para-azoxyanisole at H = 2950 Oe.

tribution function $W(\theta)$. The function $W(\theta, t)$ is given by the diffusion equation that describes the rotation of the swarm in an external field. In a rotating coordinate system tied to the field, the equation takes the form[7]

$$\frac{\partial W(\theta,t)}{\partial t}=\frac{\partial}{\partial\theta}\left[D\frac{\partial W(\theta,t)}{\partial\theta}-\left(\frac{M(\theta)}{h}+\omega\right)W(\theta,t)\right]. \tag{5}$$

Here $M(\theta) = -1/2\nu(\chi_{\parallel} - \chi_{\perp})H^2 \sin 2\theta$ is the moment of the forces exerted on the swarm by the rotating magnetic field, h is a friction coefficient and depends on the shape and dimensions of the swarm and on the viscosity of the medium, ω is the magnetic-field rotation frequency, and D is the coefficient of rotational diffusion of the swarm.

Assuming the existence of a stationary distribution function $W(\theta)$ and taking its periodicity into account, we find that

$$W(\theta)=C_1\exp(-f(\theta))\left\{(e^{-2\pi CA}-1)\left[\int_0^\pi \exp(f(\theta))\,d\theta\right]^{-1}\cdot\times\int_0^\theta \exp(f(t))\,dt+1\right\}, \tag{6}$$

where

$$f(\theta)=2A(\sin^2\theta-C\theta),\quad C=\omega/2AD,\quad A={}^1/_4H^2v(\chi_\parallel-\chi_\perp)/Dh,$$

and the constant C_1 is determined from the normalization condition

$$\int_0^\pi W(\theta)\,d\theta=1.$$

It follows from (1) that $C = \omega/\omega_{cr}$.

Expression (6) is the sought solution of the problem. The mean value of the moment of the forces exerted in this case by the magnetic field on the swarm is given by

$$\langle M\rangle=\int_0^\pi M(\theta)W(\theta)\,d\theta \tag{7}$$

and can be obtained with the aid of expression (6) for the function $W(\theta)$.

We begin the analysis of the solution (6) with the case of saturating magnetic fields, corresponding to $A \to \infty$. Asymptotic estimates of the function $W(\theta)$, obtained by the Laplace method as $A \to \infty$ and at $C \le 1$, lead to the form $W(\theta) = \delta(\theta - \alpha)$, where $\alpha = (1/2)\sin^{-1}C$. This solution, as expected, agrees fully with the earlier results[1] and with those represented by relation (2). At $A \to \infty$ and $C > 1$ we have $W(\theta) = C_1C/(\sin 2\theta + C)$, which yields for $\langle M\rangle$, by virtue of (7), the expression (4) that follows from the theories proposed in [1,3].

At finite values of the parameter A, the orientation of the swarms differs significantly from that predicted by the theory[1] (see Fig. 2). It is seen from the figure that in this range of fields, as ω/ω_{cr} increases from zero to unity, the value of the angle α corresponding to the maximally probable swarm orientation differs more and more from the values of the angle α given by (2). This is accompanied by an additional broadening of the distribution function $W(\theta)$. This behavior of the function $W(\theta)$ agrees qualitatively with the sample disorientation process observed in the experiment of [4].

For a comparison with the experimental results, we have analyzed here the dependence of $\sin 2\alpha$ on ω/ω_{cr} at various values of the parameter A (see Fig. 1a). It is seen from Fig. 1a that when the parameter A changes from zero to ten the plot of $\sin 2\alpha$ against ω/ω_{cr} differs

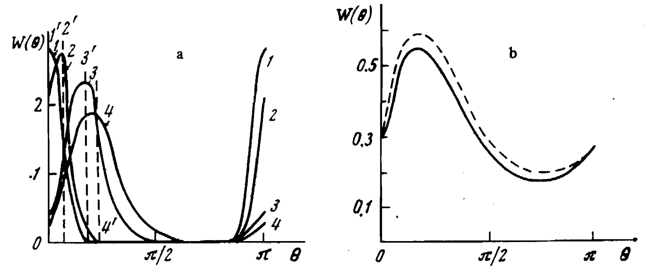

FIG. 2. Distribution function of the optical axes of the swarms relative to the direction of the magnetic field $W(\theta)$ at $C \leqslant 1$ (Fig. a) and $C > 1$ (Fig. b): at A = 10 (solid line) and as $A \to \infty$ (dashed line). In Fig. a, curves 1 and 1′ pertain to C = 0, 2 and 2′ to C = 0.2, 3 and 3′ to C = 0.4 and 4 and 4′ to C = 0.8; in Fig. b the curves pertain to C = 2.

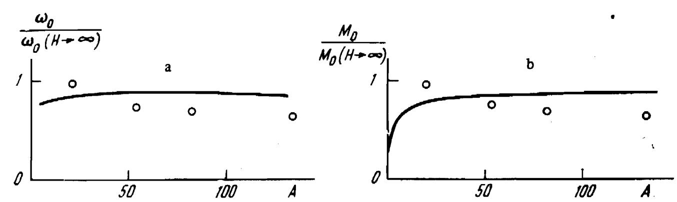

FIG. 3. Plots of ω_0/ω_0 $(H \to \infty)$ (Fig. a) and of M_0/M_0 $(H \to \infty)$ (Fig. b) against A; ○—experimental plots from [5] for para-azoxyanisole, with H changing from 2350 to 4100 Oe.

from the plot of relation (2), but it practically coincides with the latter when A is on the order of 100 (the dashed line shows that plot of $\sin 2\alpha$ vs. ω/ω_{cr} which follows from the theory[1]). At the same time, agreement with the data of [4] is observed already at A = 10.

A similar regularity is observed also, for the summary moment calculated with the aid of (7). Here the $M(\omega)$ curves, just as in the experiments [1,5], are bell-shaped (Fig. 1b). The proximity $M(\omega)$ to its limiting value can be characterized by the ratios of M_0 and of the corresponding value of ω_0 at a given field H to their maximum values $M_0(H \to \infty)$ and $\omega_0(H \to \infty)$ at a value of H corresponding to saturation (Fig. 3). It is seen from Fig. 3 that the experimental values of $M_0/M_0(H\to\infty)$ and $\omega_0/\omega_0(H \to \infty)$ obtained from the data of [5] are indeed smaller than unity, as follows from the present theory. However, whereas the theory calls for these ratios to tend to unity with increasing H, this tendency is not observed under the experimental conditions. The discrepancy can be explained by assuming that in this case other factors, particularly perhaps also those noted in [1], play a role in this case in addition to the effect of incomplete saturation. In the experiments analyzed above they used magnetic fields of intensity from 1700 to 4650 Oe. These fields are usually regarded as strongly saturating.[8-10] An increased role of the incomplete saturation should be observed in weaker fields.

To obtain more accurate quantitative estimates we must consider a three-dimensional model of the liquid crystal. The two-dimensional model proposed in this paper allows us to describe the behavior of a three-dimensional liquid crystal if the swarms are sufficiently long cylinders with their long axes perpendicular to the plane of rotation of the magnetic field. Tsvetkov[1] has shown that at sufficiently low rotation frequencies ω the swarms can be regarded as spherical, and at frequencies close to ω_{cr} they should assume a cylindrical shape, so that at critical frequencies our theory can be regarded as more appropriate for the experimental situation; this is all the more so, since at large rotation frequencies of the magnetic field the two-dimensional model explains the uniform distribution, observed in [6], of the swarm orientation in the plane of the magnetic field.

The authors are sincerely grateful to N. N. Korst for a number of useful discussions and for interest in the work.

[1)]This model does not make it possible to investigate the spatial structure of a liquid-crystal sample, which was investigated in [11] for the case of a rotating magnetic field within the framework of the continual theory.

[1]V. N. Tsvetkov, Zh. Eksp. Teor. Fiz. **9**, 602 (1939).
[2]W. Zwetkoff, Acta Physicochimica U.R.S.S., **11**, 537 (1939).
[3]H. Gasparoux and J. Prost, J. de Phys., **7**, 953 (1971).
[4]A. N. Kuznetsov and V. A. Radtsig, Zhur. Strukt. Khim. **13**, 802 (1972).
[5]V. N. Tsvetkov, I. P. Kolomiets, E. I. Ryumtsev, and F. M. Aliev, Dokl. Akad. Nauk SSSR **209**, 1074 (1973).
[6]J. W. Emsley, J. C. Lindon, G. R. Luckhurst, and D. Shaw, Chem. Phys. Lett. **19**, 345 (1973).
[7]S. Chandrasekhar, Rev. Mod. Phys. **15**, 1 (1943).
[8]A. Soupe, Angew. Chem., Internat. Edit. **7**, 97,(1968).
[9]C. H. Massen, J. A. Pouls and R. D. Spense, Adv. in Chemist. **63**, 72 (1967).
[10]W. Maier, G. Barth and H. E. Wiehl, Zs. Electrochem. **58**, 674 (1954).
[11]E. I. Kats, Zh. Eksp. Teor. Fiz. **65**, 324 (1973) [Sov. Phys.-JETP **38**, 158 (1974)].

Translated by J. G. Adashko
163