Comment on the Osipov-Terentjev Calculation of the Viscosity of Liquid Crystals

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The calculation of the viscosity of liquid crystals based on the rotational diffusion approximation recently reported by Osipov and Terentjev [1 a] is critically examined. It is shown that the main result of their treatment – the evaluation of the activational exponent – arises from the neglect of the second derivative of the distribution function with respect to the azimuthal angle in the Fokker-Planck equation. The correct asymptotic formula for the rotational viscosity coefficient is derived.

Key words: Liquid crystal, Viscosity coefficient, Rotational diffusion, Fokker-Planck equation, Energy barrier

Recently Osipov and Terentjev have published the results of their calculation of the viscosity of liquid crystals based on the rotational diffusion approximation [1]. The main result of their treatment is the evaluation of the activational exponent in the rotational viscosity coefficient γ_1, which arises from the surmounting of the mean-field potential barrier by particles. The purpose of this contribution is to demonstrate that the hydrodynamic flow of liquid crystals has nothing to do with this surmounting process and to point out an error in the calculations which results in an activational exponent. This is necessary now because reports on the "experimental confirmation" of the Osipov and Terentjev theory have begun to appear [2].

The rotational diffusion approach leads to the following expression for the rotational viscosity coefficient [3]:

$$\gamma_1 = \frac{c\,kT}{2D} \int_0^{\pi} W_0\, g\, \frac{\partial U}{\partial \vartheta} \sin\vartheta \, \mathrm{d}\vartheta\,, \tag{1}$$

where c is the particle number density, D the rotary diffusion coefficient, ϑ the angle between the particle axis and the director and $U(\vartheta)$ stands for the dimensionless ratio of the mean-field potential to the thermal energy kT. The particle orientation distribution function is represented in (1) by a product of the equilibrium Boltzmann distribution $W_0 \propto \exp(-U)$ and the function $g(\vartheta)$ which is a disturbance of the equilibrium distribution by a shear flow. The Fokker-Planck equation for the full distribution function reduces to the equation for g [3]:

$$\hat{\Lambda} g = \frac{\partial U}{\partial \vartheta}; \quad -\hat{\Lambda} = \frac{\partial^2}{\partial \vartheta^2} + \left(\operatorname{ctg}\vartheta - \frac{\partial U}{\partial \vartheta}\right)\frac{\partial}{\partial \vartheta} - \frac{m^2}{\sin^2\vartheta}. \tag{2}$$

While in the problem under consideration the value of m^2 is exactly equal to 1, I have deliberately written here the general form of this equation. The spectral properties of the operator $\hat{\Lambda}$ with $U = -q\cos^2\vartheta$ were first studied by Brown while solving the problem of magnetization relaxation of single-domain ferromagnetic particles. The relaxational time depends strongly on the dimensionless barrier height q and also on whether the magnetization takes place parallel ($m=0$) or perpendicular ($m=\pm 1$) to the easy magnetization axis. In the case $m=0$ (only this case was considered in [4]) Brown had found that the lowest eigenvalue of the operator $\hat{\Lambda}$ in the limit of large q decreases as

$$\lambda_1 = \frac{4}{\sqrt{\pi}}\, q^{3/2}\, e^{-q}\,.$$

Correspondingly the magnetization relaxation time $\tau \propto 1/\lambda_1$ increases exponentially. This dramatic behavior is due to the fact that the magnetization of the particle cannot reverse its direction without passing over an energy barrier that is very high in comparison with the thermal energy kT. A similar result has later been obtained both numerically [5] and analytically [6] in connection with the problem of the dielectric relaxation in nematic liquid crystals.

There is no over-barrier passage in case of transverse relaxation ($m=\pm 1$ in the operator $\hat{\Lambda}$). This process is simply a turn of the liquid crystal molecule or of the magnetization vector of the ferromagnetic particle to the nearest direction of the symmetry axis. Contrary to the preceding case, the higher the barrier is, the faster goes this process – all the eigenvalues of the operator $\hat{\Lambda}$ in the case $m=\pm 1$ are directly proportional to q in the limit of large q [5, 6].

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Now let us look at the Osipov-Terentjev solution of the problem (1), (2). They have dropped the last term in the operator $\hat{\Lambda}$, believing its being small. This is just the same as to put $m=0$. Consequently they have got the exponential behavior of the rotational viscosity since $\gamma_1 \propto 1/\lambda_1$. Meanwhile the correct solution ($m^2=1$) of (2) in the limit of large q can be written straightforwardly. It has the form

$$g=\vartheta \quad \text{for} \quad \vartheta<\pi/2; \qquad g=\vartheta-\pi \quad \text{for} \quad \vartheta>\pi/2. \quad (3)$$

Though (3) does not contain m explicitly, only $m^2=1$ provides the compensation of infinity which arises in (2) at $\vartheta=0$ and $\vartheta=\pi$. Substituting $g(\vartheta)$ into (1) and evaluating the integral with asymptotic accuracy one gets

$$\gamma_1=\frac{ckT}{D}\left(1-\frac{1}{2q}+\ldots\right).$$

This result is clearly a saturation effect. The only exponential temperature dependence can be expected in the rotary diffusion coefficient D, but its origin is not due to the passage by particles over a mean-field potential barrier, and its calculation is beyond the framework of the theory under consideration.

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