

Viscoelastic relaxation of nematic liquid crystals in the region of the isotropic–nematic transitions

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(Submitted February 3, 1977)

Pis'ma Zh. Eksp. Teor. Fiz. **25**, No. 5, 266–269 (5 March 1977)

It is shown that in the region of the isotropic–nematic transition of MBBA and ZhK-440 the shear modulus and shear viscosity relax in the frequency range 70–300 kHz. The results are interpreted on the basis of the de Gennes theory of the relaxation of the short-range order parameter in the isotropic phase of nematic liquid crystals.

PACS numbers: 61.30. – v, 46.30.Jv

An investigation of the temperature dependence of the shear impedance by the method of torsional oscillations in^[1] the liquid-crystal MBBA and ZhK-440 has shown that the real R and imaginary X components of the impedance differ in magnitude ($R \neq X$) only in the region of the isotropic–nematic transition, while in the isotropic and nematic phases the shear modulus $G = (R^2 - X^2)/\rho$ (ρ is the density of the liquid crystal) is equal to zero. In the frequency range 70–300 kHz investigated by us, the shear modulus in the region of the isotropic–nematic transition depends on the frequency; the shear viscosity $\eta = 2RX/\omega\rho$ (ω is the cyclic frequency) in the isotropic phase near the temperature of the isotropic–nematic transition is lower than the static viscosity η_0 measured by the capillary method (Fig. 1). The dependence of the shear modulus and of the shear viscosity on the frequency points to a viscoelastic relaxation in the region of the isotropic–nematic transition.

In the investigated frequency region, the shear modulus is proportional to the square root of the frequency. This means that the relaxation frequency should be higher than the given frequency band. A characteristic of both of the investigated substances is that the shear modulus reaches a maximum at the bleaching point T_c and decreases to zero within 4–5°C away from the bleaching point.

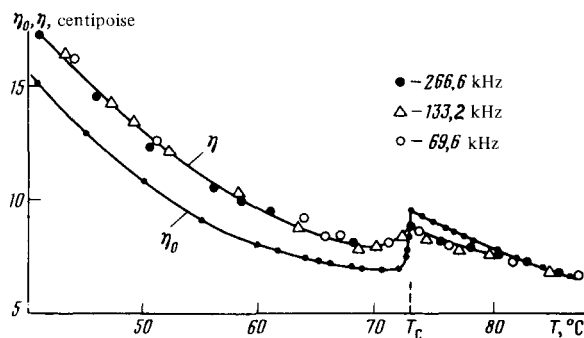


FIG. 1. Temperature dependence of the shear viscosity η and of the static viscosity η_0 of ZhK-440 at frequencies 69.6, 132.2, and 266.6 kHz.

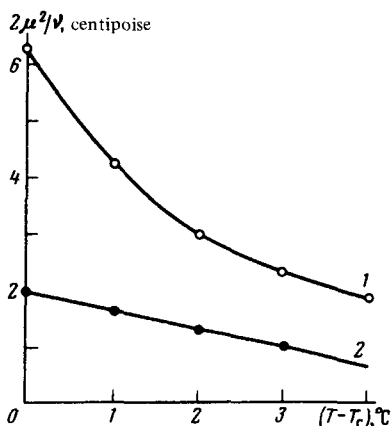


FIG. 2. Temperature dependence of $2\mu^2/\nu$ of MBBA (1) and of ZhK-440 (2).

The relaxation of the shear modulus $G(\omega)$ and of the shear viscosity $\eta(\omega)$ are described in the de Gennes statistical theory^[21] by the relations

$$G(\omega) = \frac{2\mu^2\Gamma}{\nu\left(1 + \frac{\Gamma^2}{\omega^2}\right)}, \quad (1)$$

$$\eta(\omega) = \eta_0 - \frac{2\mu^2}{\nu\left(1 + \frac{\Gamma^2}{\omega^2}\right)}, \quad (2)$$

where Γ is the frequency of the relaxation of the short-range order parameter, while η_0 , μ , and ν are quantities with the dimension of viscosity coefficients.

The quantity $2\mu^2/\nu$ tends to $\eta_0 - \eta(\omega)$ as $\omega \rightarrow \infty$. This quantity, calculated from (1) and (2) for both substances, decreases with increasing temperature, (Fig. 2).

The temperature dependence of the relaxation time τ , calculated from the frequency dependence of the shear modulus $G(\omega)$ for MBBA and ZhK-440, is shown in Fig. 3. The relaxation time decreases with increasing temperature

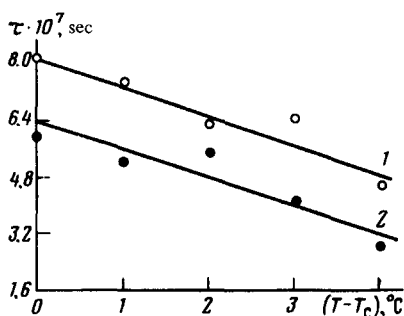


FIG. 3. Temperature dependence of the relaxation time τ of MBBA (1) and of ZhK-440 (2).

TABLE I.

Substance	$\xi(T)$ at $T - T_c = 0^\circ\text{C}$	$\xi(T)$ at $T - T_c = 4^\circ\text{C}$
MBBA	250\AA	170\AA
Zhk-440	200\AA	140\AA

from 0.81×10^{-6} sec at $(T - T_c) = 0^\circ\text{C}$ to 0.46×10^{-6} sec at $(T - T_c) = 4^\circ\text{C}$ in MBBA, and from 0.59×10^{-6} sec to 0.28×10^{-6} sec, respectively for ZhK-440. The relaxation frequency Γ is close in magnitude to the investigated frequency band. Thus, the viscoelastic relaxation times are longer than the bulk-viscosity relaxation time (10^{-8} sec).^[3]

This conclusion agrees with the theory of the relaxation of the short-range order parameter in the isotropic phase, developed by de Gennes for nematic liquid crystals.^[2] de Gennes introduces the short-range order in order to describe the dimensions $\xi(T)$ of the anisotropic molecule groups in the isotropic liquid, and defines it by means of the anisotropic part of the diamagnetic-susceptibility tensor. The temperature dependence of the dimensions of the anisotropic groups is estimated by the relation

$$\xi(T) = (L/\Gamma\nu)^{1/2}, \quad (3)$$

where L is the elasticity coefficient, equal to 2.3×10^{-6} dyn for MBBA.^[4]

Using our experimental data and the data of^[5], we have found that $\xi(T)$ decreases with increasing temperature.

We used for ZhK-440 the same values of L and ν as for MBBA.

In the nematic phase, the shear viscosity $\eta(\omega)$ is larger than the static viscosity η_0 of ZhK-440 and lower than the static viscosity of MBBA. This is due to the difference in the orientation of the MBBA and ZhK-440 by the glass surface. Our optical observations have shown that the MBBA molecules are oriented perpendicular to the surface of the glass, while the ZhK-440 molecules are oriented parallel. The depth of penetration $\delta = (\eta/\omega\rho)^{1/2}$ into the liquid crystal, of the shear ultrasonic wave radiated by the glass rod, in the frequency range 70–300 kHz, is of the same order as the distance over which there appears the orienting action of the wall. The MBBA molecules in the sound field are directed along the shear viscosity gradient, while the ZhK-440 molecules are perpendicular to both the flow and to the viscosity gradient. According to^[6], the shear viscosity of ZhK-440 corresponds to the maximum value of the viscosity coefficient, while the shear viscosity of MBBA is close to the minimum.

In the employed capillary viscosimeter, the distance over which the orienting action of the wall manifests itself is much less than the diameter 0.73 mm of the capillary, and at a flow velocity on the order of 20 sec^{-1} the molecules are not fully oriented in the flow direction. It is possible therefore that the statistical viscosity η_0 takes on an intermediate value between the maximum and minimum values of the viscosity coefficients.

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