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Effect of Electric Field on the Rheology in the Smectic and the Nematic Phases of Octyl Cyanobiphenyl

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Changes of the rheology under the application of an electric field, electrorheological (ER) effect, are studied in the smectic A (SmA) and the nematic (Ne) phases of 4-*n*-octyl-4'-cyanobiphenyl (8CB). In the SmA phase, the electric field induces a large decrease in the viscosity except just below the SmA-Ne transition point T_{AN}, where an increase in the viscosity is observed. While in the Ne phase, an increase in the viscosity, which is strongly dependent on temperature, is observed upon application of the electric field; near T_{AN} a peak in the viscosity is observed below 1 KV mm^{-1} followed by a gradual decrease to a constant value at higher electric field, whereas far from T_{AN} the viscosity increases monotonically with the electric field and saturates at high enough electric field. These results are discussed on the basis of the orientational change of the director and/or the smectic layer.

Keywords: Electrorheological effect; viscosity; nematic phase; smectic phase; phase transition

1. INTRODUCTION

The increase in the viscosity of fluids upon application of the electric field of a few kV mm^{-1} is known as electrorheological (ER) effect and was firstly studied on simple liquids in the first half of 1900's. A small viscosity change, typically a few percent, has been found in some polar liquids [1, 2]. A large ER effect, an increase in the viscosity of 2 $\sim$ 3 orders of the magnitude was found by Winslow [3] in 1947 in some colloidal suspensions consisting of water adsorbed particles and electrically non-conducting liquids. His novel study leads to recent extensive studies on this effect; the ER effect is also found in some non-aqueous suspensions containing semiconductive or ionic conductive particles [4, 5] and the effect has been explained by chain and/or column structures formed by an interaction between the dipoles induced at the particles [6–8].

The ER effect appears also in liquid crystals as shown in some reports on the effect of the electric field on the viscosity [9–12], but the systematic study of the ER effect has started more recently [13–17]. From the studies of ER effects in the nematic phase of poly (*n*-hexyl isocyanate) solution [13], MBBA (4-methoxybenzylidene-4′-*n*-butylaniline) [14] and 5CB (4-*n*-pentyl-4′-cyanobiphenyl) [15], it has been clarified that, if the electrohydrodynamic instability does not occur, the ER effect in the nematic phase is governed by an orientational change of the director, a unit vector specifying the orientation of the liquid crystal. In a Couette flow with the electric field being applied along the velocity gradient direction, i.e. perpendicular to the flow, the following characteristic properties have been observed [14, 15]. (1) In the absence of the electric field, a flow alignment of the director occurs with its direction slightly deviating from the velocity (flow) direction. (2) An increase in the viscosity is observed upon application of the electric field for liquid crystals having a positive dielectric anisotropy, since the electric field makes the director orient along the velocity gradient direction (perpendicular to the flow). (3) For those having a negative dielectric anisotropy the electric field makes the director orient along the velocity (flow) direction, leading to a decrease in the viscosity. (4) These orientational changes are determined by a balance of the viscous and the electric torques exerted on the director.

In the present study, investigations of the ER properties are made on the SmA and the Ne phases of 4-*n*-octyl-4′-cynanobiphenyl (8CB), which undergoes the phase transitions from the SmA to the Ne phase at 306.0 K and the SmA to the isotropic (Is) phase at 313.2 K. In MBBA [18] and 5CB [10], the Lesile coefficients α_2 and α_3 satisfy the relationship of $\alpha_2\alpha_3 > 0$, indicating that in the absence of the electric field there appears a flow alignment with its orientational angle given by $\theta = \tan^{-1}(\alpha_2/\alpha_3)$ [12, 19, 20]. While in Ne Phase of 8CB, a relationship of $\alpha_2\alpha_3 < 0$ holds near the SmA-Ne phase transition [21, 22], suggesting that there is no flow alignment in this temperature region [23]. A recent X-ray diffuse scattering experiment shows that in the temperature region where $\alpha_2\alpha_3 < 0$, the director orients perpendicular both to the velocity and the velocity gradient [24]. In addition, a large value of the Leslie coefficients α_1, which is not observed in nematic liquid crystals not undergoing the phase transition to the Sm phase, is reported near T_{AN} [21]. These characteristic flow properties in 8CB make us expect an appearance of ER effect which is different from MBBA and 5CB.

Furthermore, the ER effect in the SmA phase, which has not been clarified to the author's knowledge, is an interesting phenomenon to be studied.

From the present study, following results are clarified. (1) In the SmA phase the viscosity decreases upon application of the electric field. (2) Near T_{AN} in the Ne phase, there appears a viscosity peak below 1 kV mm^{-1} when increasing the electric field, followed by a gradual decrease to a constant value at higher electric field. (3) Far from T_{AN} in the Ne phase, the viscosity increases monotonically with the electric field and saturates at high electric field, which is similar to the behavior in 5CB. These results are discussed on the basis of the orientational change of the director and/or the smectic layer.

2. EXPERIMENTAL

The liquid crystal 8CB was obtained from BDH (England) and was used without further purification. The SmA-Ne and the Ne-Is phase transition points, which are determined from the temperature dependence of the viscosity, are 306.0 K and 313.2 K, respectively. The rheology, shear stress vs. shear rate, was measured with a homemade viscometer of a concentric double cylinder type [25]. In the SmA phase, the measurements were made at shear rates lower than 35 s^{-1} to enable the measurements of the high viscosity in this phase, while in Ne phase, shear rates up to 600 s^{-1} was applied. High voltages were supplied to the gap (1 mm) between the inner and the outer cylinders to generate the electric field of a few kV mm^{-1} along the velocity gradient direction, i.e. perpendicular to the velocity (flow) direction. A high voltage source with a variable frequency ac power source (CVFT1-50HVP2, Tokyo Seiden Co., Ltd., Japan), working with an amplitude of up to 5 kV rms and a frequency from 10 Hz to 5 kHz, was used. In order to measure the temperature dependence of the ER effect, the temperature of the sample was controlled within ± 0.1 K using a heater and a thermocouple (chromel-constantan) attached on the outer cylinder. Throughout this article the amplitude of the ac electric field is expressed in rms.

3. RESULTS AND DISCUSSION

For understanding the relationship between the orientation of the director and the viscosity, three geometries (a), (b) and (c) are shown in Figure 1, where Miesowicz viscosities η_1, η_2, and η_3 are the viscosities with the director orienting along the z, x and y axes, respectively. The shear deformation

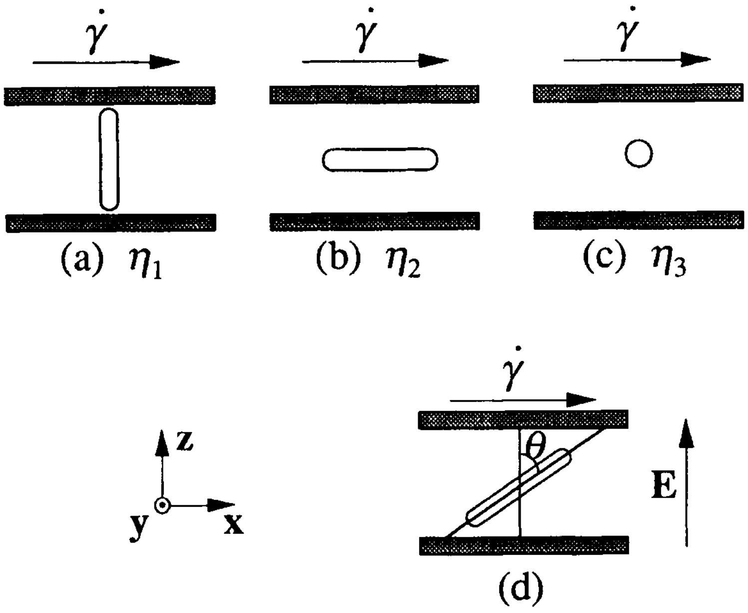

FIGURE 1 Relationship between the Miesowicz viscosities, $\eta_1 \sim \eta_3$, and the orientation of the director, (a) ~ (c). The velocity and the velocity gradient directions are along the x and the z axes, respectively, and the electric field is applied along the z axis. A coordinate system specifying the two-dimensional orientation of the director is given in (d).

and the electric field are applied along the x and the z axes, respectively. Figure 1-(d) is a coordinate system specifying the two-dimensional orientation of the director in the zx plane, which will be used for calculation of the ER effect in the Ne phase.

a. Temperature Dependence of the Viscosity Without Electric Field

Temperature dependence of the shear stress, which is proportional to the viscosity, measured at a constant shear rate is given in Figure 2. In the SmA phase the flow is non-Newtonian, and the shear stress decreases $2 \sim 3$ orders of magnitude when increasing the temperature from room temperature toward the SmA-Ne phase transition point T_{AN}. Such a temperature dependence is observed in other SmA liquid crystals [26].

In the Ne phase, a gradual decrease in the viscosity is observed with increasing temperature, followed by a small increase of the viscosity near

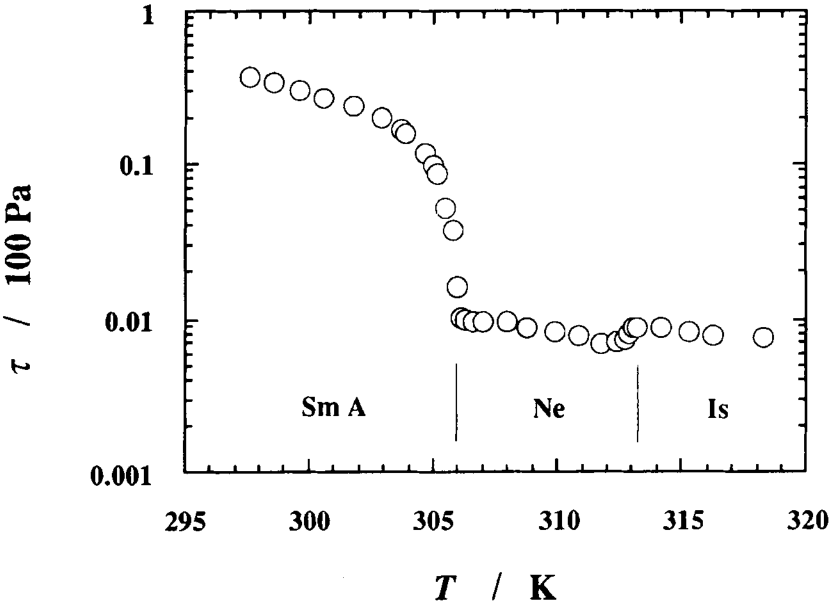

FIGURE 2 Temperature dependence of the shear stress τ measured at a shear rate of $32.95\ \mathrm{s}^{-1}$. The flow is non-Newtonian in the SmA phase and is Newtonian in the Ne and the Is phases. The phase transition temperatures T_{AN} and T_{NI} are 306.0 K and 313.2 K, respsectively.

the Ne-Is phase transition point T_{NI}. At T_{NI} the viscosity discontinuously jumps to the value in the Is phase. Our previous results on 5CB and MBBA show that the viscosity is somewhat larger than the Miesowicz viscosity η_2, indicating that a flow alignment occurs in these materials with the orientation of the director slightly deviating from the velocity direction [14, 15]. In 8CB the behavior is different form that in 5CB and MBBA. Owing to the presence of the Ne-SmA phase transition, η_2 in this material is reported to become so large near T_{AN}, making the observed viscosity in this temperature region smaller than η_2. In Figure 3 the observed viscosity in the Ne phase is given together with η_2 and η_3 reported by Kneppe *et al.* [21]. The observed viscosity is larger than η_2 just below T_{NI}, but gradually approaches to η_3 with decreasing the temperature toward T_{AN}. As to the director orientation in the Ne phase, the X-ray diffuse scattering study shows that the orientation of the director just below T_{NI} is near the velocity (x) direction, but changes its direction perpendicular both to the velocity and the velocity gradient directions, i.e. along the y axis, when lowering the temperature to T_{AN} [24]. Our result on the temperature dependence of the viscosity is

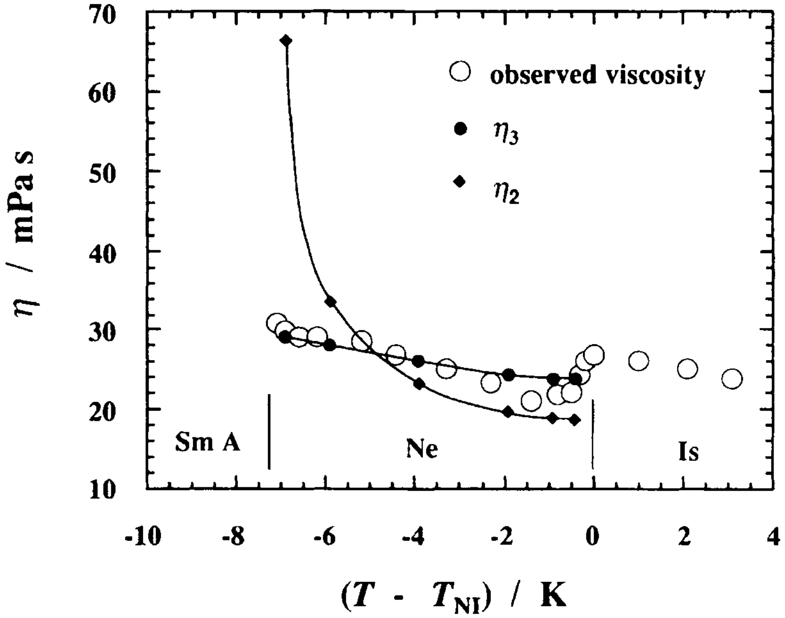

FIGURE 3 Temperature dependence of the viscosity observed at 32.95 s^{-1} and the Miesowicz viscosities η_2 and η_3 reported by Kneppe *et al.* (Ref. 21) in the Ne phase. The director orients nearly parallel to the velocity direction just below T_{NI} but changes its direction perpendicular to both the velocity and the velocity gradient directions when lowering the temperature toward T_{AN}.

consistently interpreted in terms of this orientational change of the director suggested by the X-ray study.

b. ER Effect in the SmA Phase

The change of the rheology, shear stress vs. shear rate, upon application of the electric field (200 Hz) is measured at 297.8 K. As Figure 4 shows, in the absence of the electric field the flow is non-Newtonian, which is evidenced by the non-linear relationship between the shear stress and the shear rate. When the electric field is applied, the viscosity decreases, accompanied by a change from a non-Newtonian to a Newtonian flow. In Figure 5 the electric field dependence of the induced shear stress $|\Delta\tau|$, i.e. the change of the shear stress by the application of the electric field, measured at a shear rate of 19.77 s^{-1} is depicted. The induced shear stress $|\Delta\tau|$ monotonically increases with the electric field and saturates at about 3 kV mm^{-1}, which is contrasted to the saturation in Ne phase where it occurs at about 1 kV mm^{-1}.

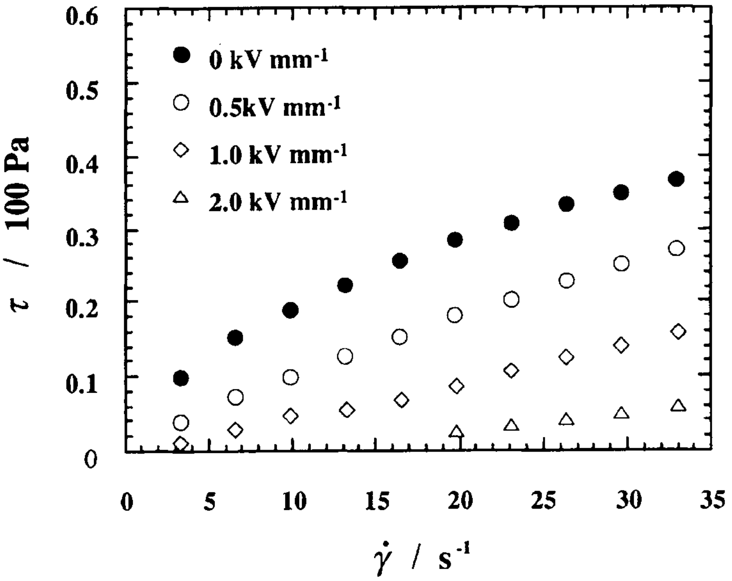

FIGURE 4 Change of the rheology, shear stress τ vs. shear rate $\dot{\gamma}$, upon application of an electric field (200 Hz) at 297.8 K. The shear stress, which is proportional to the viscosity, decreases with the electric field, accompanied by a change from non-Newtonian to Newtonian flow.

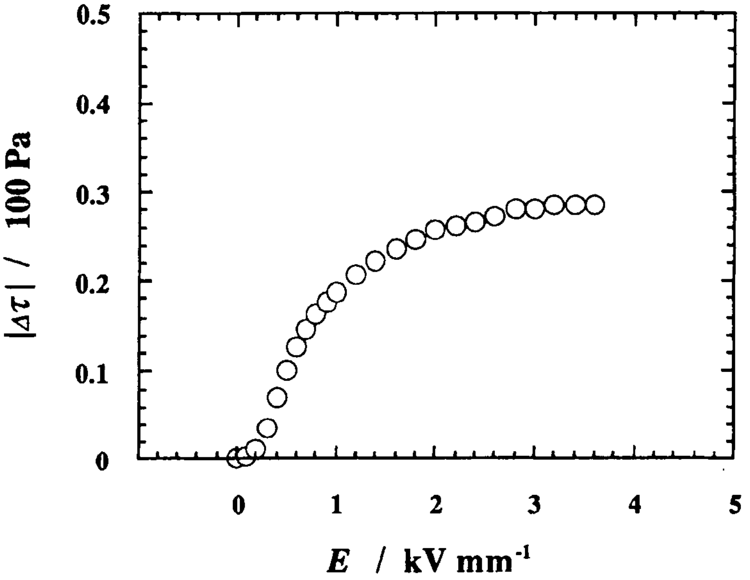

FIGURE 5 Electric field (200 Hz) dependence of the induced shear stress $|\Delta\tau|$ at 297.8 K in the SmA phase, measured at a shear rate of 19.77 s^{-1}. $|\Delta\tau|$ increases monotonically with the electric field and saturates around 3 kV mm^{-1}.

On the other hand, the electric field induces an increase in the shear stress just below T_{AN}. In the measurements of the shear stress, a large fluctuation in the observed data was recognized at electric fields higher than $2\,kV\,mm^{-1}$.

The viscosity in the SmA phase has not been thoroughly understood, which makes it difficult to understand the mechanism of ER effect in this phase. But characteristic flow properties indicated for the three geometries (a), (b) and (c) in Figure 1 are suggestive [12], (a) In the case that the normal to the Sm layer is parallel to the velocity gradient, the flow is Newtonian with the viscosity being comparable with that in the Ne phase. In this geometry, unless the thickness of the sample is kept constant with an accuracy better than c.a.100A, there would appear dilated or compressed regions along the flow, leading to a flow instability. (b) In the case that the normal to the Sm layer is parallel to the velocity direction, the viscosity is about 100 times larger than that in the cases (a) and (c), and the fluid is characterized by a plug flow, where the parabolic shape of the velocity profile, which is characteristic to the Poiseuille flow, does not appear. (c) In the case that the normal to the Sm layer is perpendicular to both the velocity and the velocity gradient directions, the flow is a Bingham plastic type with a relationship of $\tau = \alpha_4/2 \cdot \dot{\gamma} + \tau_0$. Here τ, α_4, $\dot{\gamma}$ and τ_0 are shear stress, Leslie coefficient, shear rate and yield stress, respectively.

The X-ray diffuse scattering study shows that the director has a geometry (c) just below T_{AN}, followed by a coexistence of the geometries (a) and (c) at lower temperatures [24]. Referring to the characteristic flow properties described above and the X-ray's result, we can understand the ER effect at room temperature as follows. When the electric field is not applied, the flow is characterized by coexistence of the geometries (a) and (c). The application of the electric field makes a change of the geometry to (a), i.e the normal to the smectic layer is made to orient parallel to the velocity gradient direction, leading to the decrease in the viscosity with the flow characterized by a Newtonian fluids. The field induced increase of the shear stress just below T_{AN} can be attributed to the change of the geometry from (c) to (a). The geometry (a), which is assumed as an orientation under the electric field, is ascertained by the fact that extrapolation of the Miesowicz viscosity η_1 in the nematic phase to the value at room temperature is almost coincide with the observed value. The positive dielectric anisotropy of this material [27], which makes the director orient parallel to the electric field (the velocity gradient direction), and the large fluctuation of the observed values of the shear stress also support this geometry.

c. ER Effect in the Ne Phase

c-1. ER Effect Near T_{AN}

The change of the rheology, shear stress vs. shear rate, upon application of the electric field is measured at 306.2 K, just above T_{AN}, and the result is given in Figure 6. In the absence of the electric field, the shear stress linearly increases with the shear rate, showing that the flow is Newtonian. When the electric field is applied, a changes of the rheology is observed with an increase in the viscosity and a change of the flow from a Newtonian to a non-Newtonian fluid. It should be noted that the viscosity increases at low field, shows a maximum and becomes smaller at high fields. The electric field dependence of the shear stress τ obtained at a shear rate of 329.5 s^{-1} is depicted in Figure 7. The viscosity increases steeply at low field below c.a.0.3 kV mm^{-1}, followed by a viscosity peak around 0.7 kV mm^{-1} at medium field and a gradual decrease to a saturated value at high enough field.

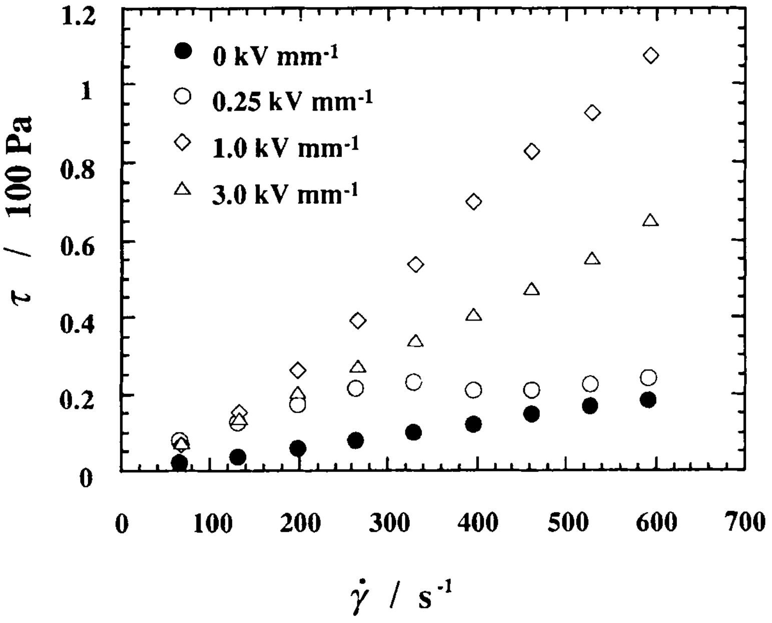

FIGURE 6 Change of the rheology upon application of the electric field (200 Hz) at 306.2 K, just above T_{AN} in the Ne phase. The shear stress at high field becomes smaller than that at medium field.

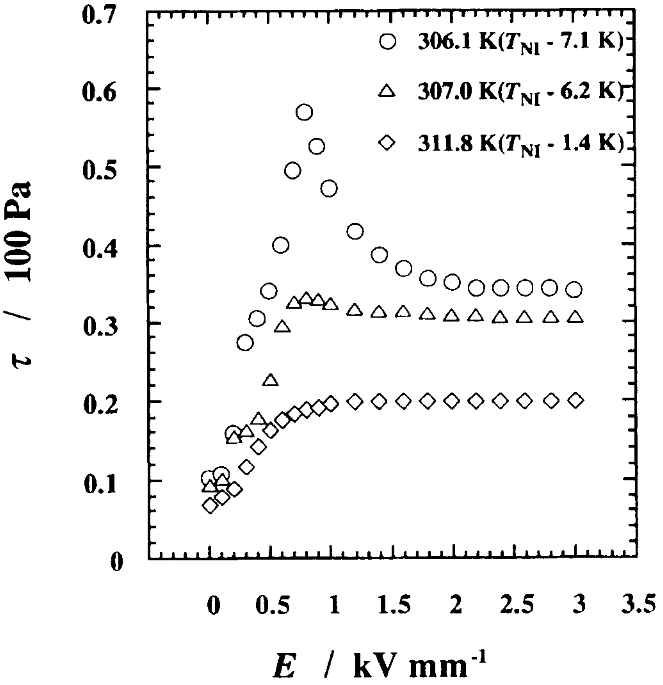

FIGURE 7 Electric field (200 Hz) dependence of the shear stress τ measured at a shear rate of 329.5 s^{-1}. Near T_{AN} there appears a peak in τ, followed by a gradual decrease to a constant value at high fields. Far from T_{AN}, τ increases monotonically and saturates at high fields.

If an electric field above a certain amplitude is applied along the z axis, the director would orient in the zx plane. Under such a consideration, the ER effect near T_{AN} is calculated on the basis of the Leslie-Ericksen theory [28, 29]. We use the coordinate system given in Figure 1-(d), where the orientation of the director is specified by θ. The viscosity η is determined by

$$2\eta(\theta) = 2\alpha_1\cos^2\theta\sin^2\theta + (\alpha_3 + \alpha_6)\sin^2\theta + (\alpha_5 - \alpha_2)\cos^2\theta + \alpha_4, \qquad (1)$$

where $\alpha_1 \sim \alpha_6$ are the Leslie coefficients [20, 23]. The first term in the left hand side, $2\alpha_1\cos^2\theta\sin^2\theta$, is frequently neglected owing to the small value of α_1 compared to other Leslie coefficients [20], but this term is left in Eq. (1) since α_1 in 8CB is reported to become large near the Ne-SmA phase transition [21]. The flow alignment angle is determined from Eq. (2), which is derived from a balance of the electric and the viscous torques exerted on the director [15, 20, 23].

$$(\alpha_3\sin^2\theta - \alpha_2\cos^2\theta)\dot{\gamma} - 1/2\varepsilon_a E_0{}^2\sin 2\theta = 0, \tag{2}$$

where ε_a and E_0 are the dielectric anisotropy and the amplitude of the electric field, respectively.

If $\alpha_2\alpha_3 < 0$ and $E = 0$, Eq. (2) shows that there is no solution to give the flow alignment angle. In the Ne phase of 8CB, $\alpha_3 > 0$ and $\alpha_2 < 0$ near the Ne-SmA phase transition point T_{AN}, indicating that the flow alignment is not possible in this temperature region unless the electric field is applied. If the electric field is applied with its amplitude larger than a certain value, we can get a solution in Eq. (2) and expect an alignment of the director in the zx plane.

In 8CB, temperature dependences of the Miesowicz viscosities, η_1, η_2 and η_3, and the rotational viscosity γ_1, are reported [21, 22, 30], of which the data by Kneppe *et al.* [21] are used to obtain the Leslie coefficients using the well known relations. Inserting the Leslie coefficients thus obtained and the reported dielectric anisotropy [27] into Eqs. (1) and (2), we calculate the electric field dependence of the shear stress τ at various temperatures (Fig. 8). Here the result is given as function of $(T - T_{\mathrm{NI}})$ since there is a difference in the phase transition point between the reported [21] and our results. As this figure shows, the calculated shear stress above medium electric field is qualitatively in good agreement with the experimental one (Fig. 7), If we consider that the Leslie coefficients near T_{AN} include uncertainty of at least 10% [21, 30]. The fact that the peak appears also in the calculated shear stress indicates that the large α_1, which might be due to the Ne to SmA phase transition, is responsible for it; the increase in the amplitude of the electric field makes a change of the alignment angle θ from a some value to 0° passing through 45°, at which the first term in Eq. (1) has a maximum to give rise to the peak in the shear stress. The behavior at high field, the gradual decrease of the viscosity to the saturated one, can be attributed to the orientational change of the director toward the velocity gradient direction ($\theta = 0$).

The mechanism of the ER effect at low field, where the alignment of the director at a definite angle is not possible, is complex. The X-ray diffuse scattering study suggests that the director orients along the y axis, perpendicular both to the velocity and the velocity gradient directions [24]. The increase of the viscosity in the low field region, thus, is qualitatively understood to occur as a result of the orientational change of the director from the y axis to the zx plane.

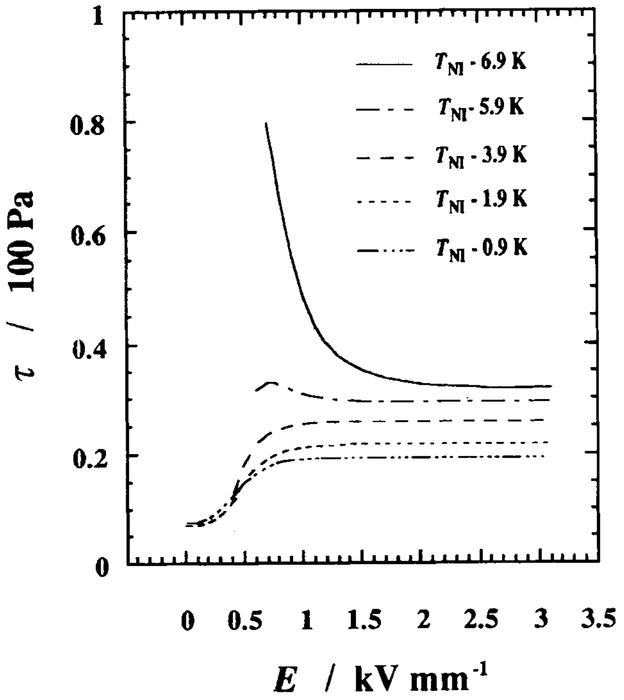

FIGURE 8 Calculated result of the electric field dependence of the shear stress τ at various temperatures. The results is qualitatively in good agreement with experimental results given in Figure 7.

c-2. ER Effect Far from T_{AN}

In this temperature region, the flow changes from Newtonian to non Newtonian and then to Newtonian with increase in the amplitude of the electric field as shown in Figure 9. The electric field dependence of the shear stress at 311.8 K is given in Figure 7; the shear stress increases monotonically with the electric field and saturates at high enough field. This behavior is similar to that in 5CB [15] and can be interpreted as caused by an orientational change of the director from the direction near the velocity to that along the velocity gradient (perpendicular to the flow) upon application of the electric field.

As obvious from Figure 7, the change of the shear stress saturates at high enough fields even near T_{AN}, indicating that if a sufficiently high electric field is applied the director orients along the velocity gradient direction in the whole temperature region of the Ne phase. In Figure 10, temperature

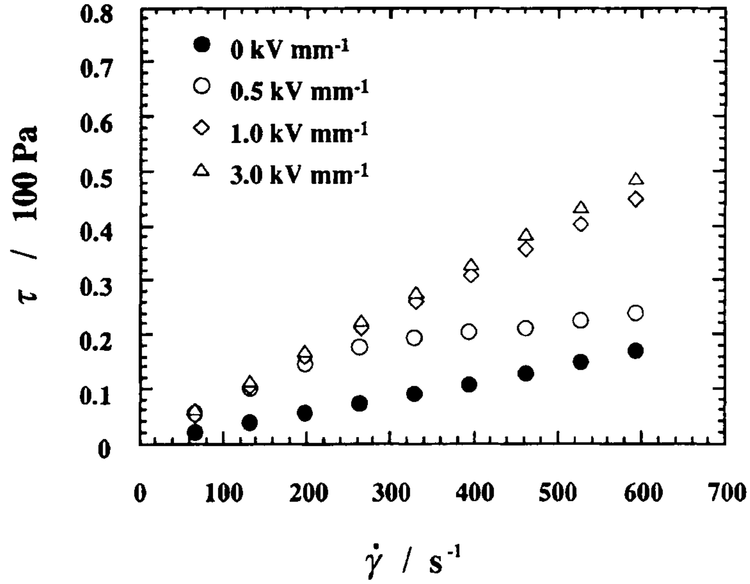

FIGURE 9 Change of the rheology upon application of the electric field (200 Hz) at 308.9 K, far from T_{AN} in the Ne phase. The shear stress increases with the electric field, accompanied by a change from Newtonian to non-Newtonian and again to Newtonian flow.

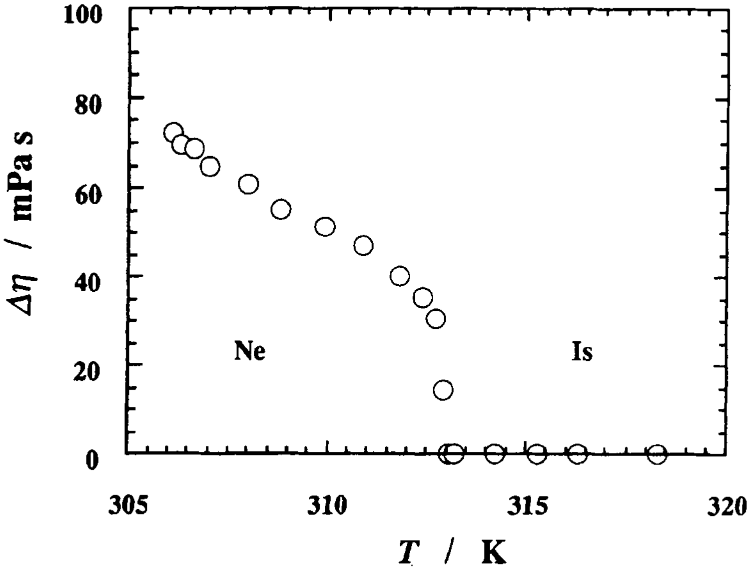

FIGURE 10 Temperature dependence of the viscosity change $\Delta\eta$, which is induced by the electric field (3 kV mm^{-1}, 200 Hz), measured at a shear rate of 32.95 s^{-1} in the Ne and the Is phases. Change of the order parameter as a function of temperature is responsible for this temperature dependence.

dependence of the viscosity change induced by the electric field of 3 kV mm^{-1} is depicted. The temperature dependence is similar to that in 5CB [15], implying that it is due to the temperature dependence of the order parameter in the Ne phase [31].

d. ER Effect in the Is Phase

In the Is phase, no distinct rheological change under the application of the electric field is observed as evidenced by the result of the shear stress vs shear rate measurements with and without the electric field (Fig. 11).

4. SUMMARY AND CONCLUSIONS

Effects of the electric field on the rheology are investigated in SmA, Ne and Is phases of 8CB. As shown in Figure 12 the shear stress, which is proportional to the viscosity, decreases in the SmA phase except just below T_{AN},

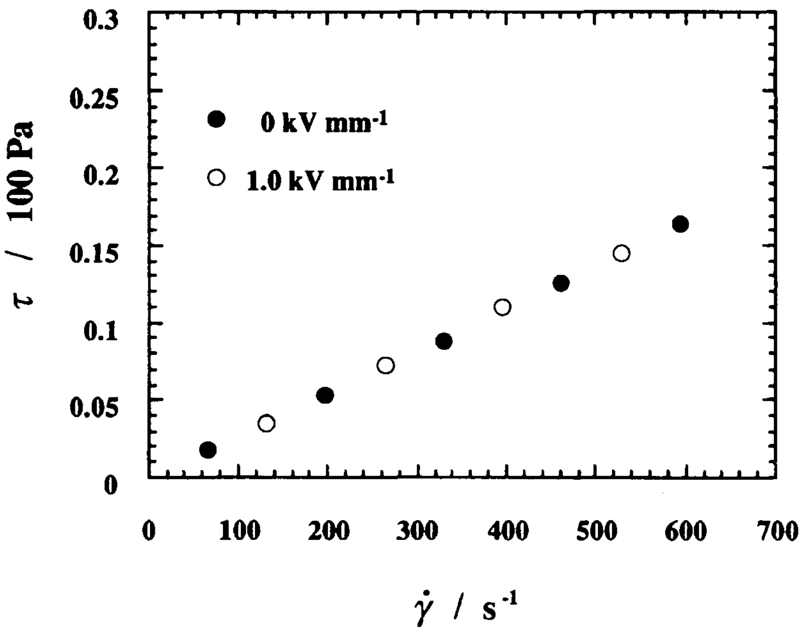

FIGURE 11 Rheology, shear stress vs shear rate, with and without the electric field (1 kV mm^{-1}, 200 Hz) at 313.2 K in the Is phase. No distinct rheological change occurs by the application of the electric field.

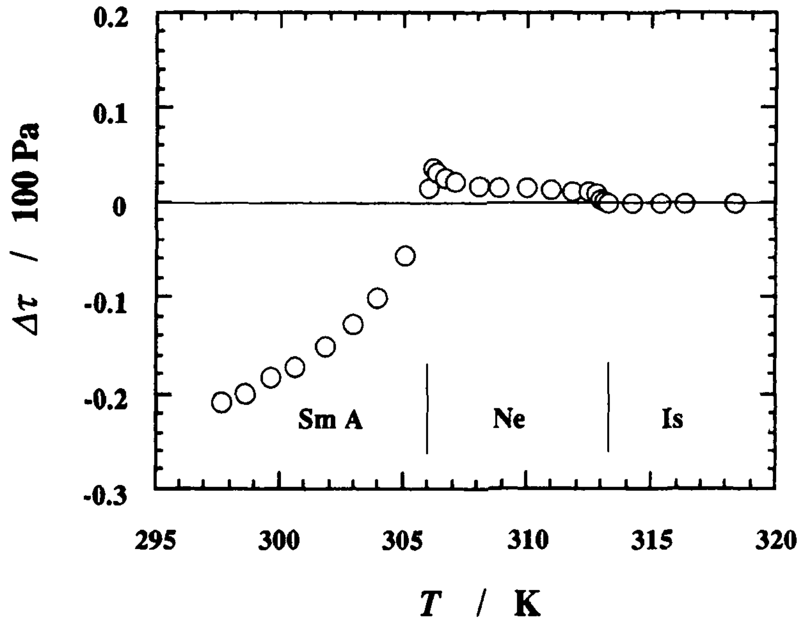

FIGURE 12 Temperature dependence of the induced shear stress Δτ measured at a shear rate of 32.95 s^{-1} in SmA, Ne and Is Phases. The amplitude and the frequency of the applied electric field are 1 kV mm^{-1} and 200 Hz, respectively.

increases in the Ne phase and does not change in the Is phase. The ER effect in the SmA phase is suggested to occur as a result of the fractional change of the two coexisting geometries of the smectic layers. In the Ne phase, the ER effect strongly depends on temperature; near T_{AN} there appears a peak in the viscosity followed by a gradual decrease to a constant value when increasing the amplitude of the electric field, while far from T_{AN} the viscosity increases monotonically and saturates at high fields. On the basis of the Leslie-Ericksen theory, the viscosity peak near T_{AN} and the saturation of the ER effect at high fields are successfully interpreted in terms of the orientational change of the director upon application of the electric field. The viscosity peak is understood to appear as a result of the large α_1 near T_{AN}, which might be due to the Ne to SmA phase transition. In the low field region where the flow alignment of the director is not possible, the observed ER effect is interpreted as caused by the orientational change of the director from the direction perpendicular to both the velocity and the velocity gradient directions to the velocity gradient plane.

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