

Influence of an Orienting Field on the Viscosity of a Molecular Liquid

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Z. Naturforsch. **39a**, 22–26 (1984); received October 14, 1983

From phenomenological consideration follows that both the field-induced anisotropy of the shear viscosity and the effect reciprocal to the flow birefringence contribute to the anisotropy of the effective viscosity in a Couette or a Poiseuille geometry, as well as to transverse effects and normal pressure differences. A Fokker-Planck equation approach links these mechanisms and yields a relation between two viscosity coefficients associated with the molecular orientation.

Application of an orienting magnetic or electric field renders the shear viscosity of a molecular liquid anisotropic; the effect reciprocal to the flow alignment gives rise to an additional anisotropy of the effective viscosity as it can, in principle, be observed in a Couette or a Poiseuille flow experiment. In lowest order in the applied field, both processes are governed by the 2nd rank alignment tensor. There is a strong similarity to the flow behavior of a nematic liquid crystal [1–3] where the magnitude of the alignment tensor, however, is determined by the (spontaneous) Maier-Saupe order parameter rather than by the magnitude of the applied field.

It is the purpose of this article, firstly to state the relevant constitutive relations as they follow from irreversible thermodynamics [4], secondly to indicate how the various phenomenological coefficients can be measured in a (plane) Couette or a (plane) Poiseuille flow arrangement and thirdly, to present results obtained from a mesoscopic theory based on a Fokker-Planck equation [5] for the orientational distribution function. Before these points are discussed, some remarks on the alignment and its optical detection via birefringence are in order.

Alignment, Birefringence

A liquid composed of (effectively) axisymmetric particles is considered. The unit vector parallel to the figure axis of a molecule is denoted by \mathbf{u} ; $f = f(t, \mathbf{r}, \mathbf{u})$ is the orientational distribution function with the normalization $\int f d^2u = 1$. The second

rank alignment tensor \mathbf{a} needed here is defined by

$$\mathbf{a} = \sqrt{\frac{15}{2}} \langle \overline{\mathbf{u}\mathbf{u}} \rangle, \quad (1)$$

where the numerical factor has been inserted for convenience; $\langle \dots \rangle = \int \dots f d^2u$ stands for an orientational average. The symbol $\overline{}$ indicates the symmetric traceless part of a tensor, e.g. one has

$$\overline{\mathbf{a}\mathbf{b}} = \frac{1}{2} (\mathbf{a}\mathbf{b} + \mathbf{b}\mathbf{a}) - \frac{1}{3} \mathbf{a} \cdot \mathbf{b} \delta$$

(where δ is the unit tensor) for the dyadic constructed from the components of the vectors \mathbf{a} and \mathbf{b} .

In the absence of any orienting torques, \mathbf{a} vanishes for an isotropic liquid. An applied magnetic field $\mathbf{H} = H \mathbf{h}$ where \mathbf{h} is the unit vector parallel to \mathbf{H} causes a non-zero equilibrium alignment

$$\mathbf{a} = \mathbf{a}^{\text{eq}} = \sqrt{\frac{3}{2}} a_{\text{eq}} \overline{\mathbf{h}\mathbf{h}}. \quad (2)$$

The scalar quantity a_{eq} with the properties $a_{\text{eq}} = \sqrt{5} \langle P_2(\mathbf{u} \cdot \mathbf{h}) \rangle$ and $\mathbf{a} : \mathbf{a} = a_{\text{eq}}^2$ is a measure for the degree of orientation. Here $P_2(x) = \frac{3}{2}(x^2 - \frac{1}{3})$ is the 2nd Legendre polynomial. In lowest order in the applied field one has

$$3\sqrt{5} a_{\text{eq}} = \left[\frac{\mu^2}{(k_B T)^2} P_2(\mathbf{u} \cdot \hat{\mu}) + \frac{\chi_a}{k_B T} \right] H^2 \quad (3)$$

(where T is the temperature of the liquid, k_B is the Boltzmann constant), for a Hamiltonian $\mathcal{H} = -\mu \cdot \mathbf{H} - \frac{1}{2} \chi_a \overline{\mathbf{u}\mathbf{u}} : \overline{\mathbf{H}\mathbf{H}}$ associated with the orientational energy; the effect of molecular correlations as discussed in [6] for the case of an orienting electric field has been disregarded. Here, $\mu = \mu \hat{\mu}$ is the

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magnetic moment; the Legendre polynomial occurring in (3) depends on the angle between $\boldsymbol{\mu}$ and the figure axis \mathbf{u} ; $\chi_a = \chi_{\parallel} - \chi_{\perp}$ is the difference between the magnetic susceptibilities for \mathbf{H} parallel and perpendicular to \mathbf{u} , respectively.

A nonzero 2nd rank alignment tensor renders a fluid birefringent. Let $\varepsilon_a = \varepsilon_{\parallel} - \varepsilon_{\perp}$ be the difference between the dielectric coefficient of a completely aligned fluid with the electric field parallel and perpendicular to \mathbf{u} . Then one has, for the anisotropic (symmetric traceless) part $\boldsymbol{\varepsilon}$ of the dielectric tensor $\boldsymbol{\varepsilon}$ which is responsible for the birefringence:

$$\overline{\boldsymbol{\varepsilon}} = \varepsilon_a \langle \overline{\mathbf{u} \mathbf{u}} \rangle = \sqrt{\frac{2}{15}} \varepsilon_a \mathbf{a}. \quad (4)$$

As a consequence, the difference $\delta v = v_{\parallel} - v_{\perp}$ between the indices of refraction v_{\parallel} and v_{\perp} for linearly polarized light with the \mathbf{E} -field vectors parallel and perpendicular to $\mathbf{H} \sim \mathbf{h}$ is given by

$$(v_{\parallel} + v_{\perp})(v_{\parallel} - v_{\perp}) = 2 v \delta v = \frac{1}{\sqrt{5}} \varepsilon_a a_{\text{eq}}. \quad (5)$$

For a small alignment, the quantity $v = \frac{1}{2}(v_{\parallel} + v_{\perp})$ is practically equal to the “isotropic” index of refraction $\frac{1}{3}v_{\parallel} + \frac{2}{3}v_{\perp}$; then one has $\delta v \sim H^2$, cf. (3).

Due to (5), the magnitude a_{eq} of the alignment tensor can be determined from Cotton-Mouton effect [7] measurement when the quantity ε_a is known.

An expression analogous to (2) holds true for an applied electric field; the relation analogous to (3) has to be modified by internal field corrections [6]. Furthermore, it should be mentioned that there exists, in principle, a nonequilibrium contribution to the Kerr effect in liquid containing mobile ions [8].

Constitutive Relations

To study the influence of a molecular orientation on the flow properties of a fluid, the alignment tensor has to be taken into account as an additional macroscopic variable. In [4, 9], this idea has been used to study nonequilibrium phenomena both in the isotropic and nematic phases of liquid crystals by an extension of the standard methods of irreversible thermodynamics. With higher rank alignment tensors or terms nonlinear in the 2nd rank alignment tensor \mathbf{a} disregarded, the constitutive relation for the friction pressure tensor (symmetric

traceless) derived in [4] is written as

$$\overline{\mathbf{p}} = -2 P_k \tau_p (\boldsymbol{\gamma} + \sqrt{6} \sigma \overline{\mathbf{a} \cdot \boldsymbol{\gamma}}) - \sqrt{2} P_k \tau_{pa} \left(\frac{\partial}{\partial t} \mathbf{a} - 2 \overline{\boldsymbol{\omega} \times \mathbf{a}} \right) \quad (6)$$

where

$$\boldsymbol{\gamma} \equiv \overline{\nabla \mathbf{v}}, \quad \boldsymbol{\omega} = \frac{1}{2} \nabla \times \mathbf{v} \quad (7)$$

are the symmetric traceless part of the gradient of the flow velocity \mathbf{v} and the vorticity associated with the antisymmetric part of the velocity gradient tensor. In (6), the reference pressure $P_k = n k_B T$ is the kinetic pressure of a fluid with the number density n and the temperature T . The quantities τ_p and τ_{pa} are phenomenological relaxation time coefficients (the subscripts “p” and “a” refer to “pressure” and “alignment”), σ is an additional coefficient specifying the anisotropy of the shear viscosity tensor.

Now, insertion of the field-induced equilibrium alignment (2) into (6) leads to

$$\overline{\mathbf{p}} = -2 \eta \boldsymbol{\gamma} - 2 \tilde{\eta}_1 \overline{\mathbf{h} \mathbf{h} \cdot \boldsymbol{\gamma}} - 2 \tilde{\eta}_2 \left(\overline{\mathbf{h} \frac{\partial}{\partial t} \mathbf{h}} - \overline{\boldsymbol{\omega} \times \mathbf{h} \mathbf{h}} \right) \quad (8)$$

with the viscosity coefficients

$$\eta = P_k \tau_p, \quad \tilde{\eta}_1 = 3 \eta \sigma a_{\text{eq}}, \quad \tilde{\eta}_2 = \sqrt{3} P_k a_{\text{eq}} \tau_{pa}. \quad (9)$$

Notice that both $\tilde{\eta}_1$ and $\tilde{\eta}_2$ are proportional to a_{eq} and consequently to H^2 in lowest order. In (8), it has been assumed that the magnitude H of the field \mathbf{H} does not depend on the time t .

Before it is discussed how the various viscosity coefficients (9) can be measured, it is worth mentioning that τ_{pa} and consequently $\tilde{\eta}_2$ are related to the flow birefringence. In fact, phenomena associated with τ_{pa} are a manifestation of the effect reciprocal [10] to the flow alignment [4] $\mathbf{a} = -\sqrt{2} \tau_{ap} \boldsymbol{\gamma}$. The relaxation time coefficient τ_{ap} is equal to τ_{pa} occurring in (7, 9) by a Onsager symmetry relation. The flow-birefringence is determined by [4], cf. (4),

$$\overline{\boldsymbol{\varepsilon}} = -2 \beta \boldsymbol{\gamma}, \quad \beta = (\varepsilon_a / \sqrt{15}) \tau_{ap}. \quad (10)$$

Thus due to $\tau_{ap} = \tau_{pa}$, the viscosity $\tilde{\eta}_2$ is related to the flow birefringence coefficient β by

$$\varepsilon_a \tilde{\eta}_2 = 3 \sqrt{5} P_k a_{\text{eq}} \beta. \quad (11)$$

Furthermore, it should be mentioned that the relaxation time coefficients have the properties [4]

$$\tau_p > 0, \quad \tau_a > 0, \quad \tau_p \tau_a > \tau_{ap}^2, \quad (12)$$

where τ_a is the (bare) relaxation time of the alignment. Notice that $\tau_{ap} = \tau_{pa}$ and consequently $\tilde{\eta}_2$ and β may have either sign.

No general relation analogous to (11) seems to exist for $\tilde{\eta}_1$. However, for the special model to be considered later, $\tilde{\eta}_1$ is found to be proportional to $\tilde{\eta}_2$.

For applications of (6) and other constitutive relations to the non-Newtonian flow behavior and the viscoelasticity which, in principle, allow the determination of the relaxation time coefficients, see [11, 12].

Effective Viscosities

Next, the consequences of (8) are studied for a constant magnetic field and a planar flow geometry as realised in a planar Couette or a planar Poiseuille flow. With the x - and y -axis of the coordinate system chosen to be parallel to the flow velocity v and its gradient, respectively, one has

$$\gamma = \gamma \mathbf{e}^x \mathbf{e}^y, \quad \omega = -\frac{1}{2} \gamma \mathbf{e}^z, \quad \gamma = \partial v_x / \partial y, \quad (12)$$

where the $\mathbf{e}^{x,y,z}$ are unit vectors parallel to the coordinate axes. In the Couette and Poiseuille arrangements one has $\gamma = \text{const.}$ and $\gamma' \equiv \partial \gamma / \partial y = \text{const.}$ respectively.

In the Couette flow case one infers from (8)

$$p_{xy} = -\eta_{\text{eff}} \gamma, \quad (13)$$

$$\begin{aligned} p_{xz} &= -\frac{1}{2} (\tilde{\eta}_1 - \tilde{\eta}_2) h_y h_z \gamma, \\ p_{yz} &= -\frac{1}{2} (\tilde{\eta}_1 + \tilde{\eta}_2) h_y h_z \gamma \end{aligned} \quad (14)$$

for the nondiagonal elements of the pressure tensor. Furthermore, the normal pressures differences

$$\begin{aligned} \frac{1}{2} (p_{xx} - p_{yy}) &= \tilde{\eta}_2 h_x h_y \gamma, \\ \frac{1}{2} (p_{xx} + p_{yy}) - p_{zz} &= -\tilde{\eta}_1 h_x h_y \gamma \end{aligned} \quad (15)$$

are found. Here the $h_{x,y,z}$ are the components of the unit vector $\mathbf{h} = H^{-1} \mathbf{H}$ with respect to the coordinate axes. Notice that the quantities (14, 15) vanish in the absence of an orienting field. The dependence of the effective viscosity η_{eff} of (13) on the coefficients η , $\tilde{\eta}_1$, $\tilde{\eta}_2$ and the direction of \mathbf{h} is discussed later.

In the case of a Poiseuille flow, the force density

$$\mathbf{k} = \nabla \cdot \overline{\mathbf{p}} \quad (16)$$

is the quantity of interest. From (8) with (12) one now infers

$$k_x = -\eta_{\text{eff}} \gamma', \quad (17)$$

$$k_y = -(\tilde{\eta}_1 + \tilde{\eta}_2) \gamma' h_x h_y, \quad (18)$$

$$k_z = -\frac{1}{2} (\tilde{\eta}_1 + \tilde{\eta}_2) \gamma' h_x h_z.$$

Notice that the “transverse” effects (18) vanish in the absence of a field. The “longitudinal” effect (17), i.e. the flow resistance is governed by the same effective viscosity η_{eff} as already occurred in (13). This quantity is given by

$$\eta_{\text{eff}} = \eta + \frac{1}{2} \tilde{\eta}_1 (h_x^2 + h_y^2 - \frac{2}{3}) + \frac{1}{2} \tilde{\eta}_2 (h_x^2 - h_y^2). \quad (20)$$

Thus one has for the Miesowicz [1–3] viscosities $\eta_{A,B,C}$ corresponding to \mathbf{h} parallel to $\mathbf{e}^{x,y,z}$, respectively,

$$\begin{aligned} \eta_A &= \eta + \frac{1}{6} \tilde{\eta}_1 + \frac{1}{2} \tilde{\eta}_2, \\ \eta_B &= \eta + \frac{1}{6} \tilde{\eta}_1 - \frac{1}{2} \tilde{\eta}_2, \\ \eta_C &= \eta - \frac{1}{3} \tilde{\eta}_1, \end{aligned} \quad (21)$$

and consequently

$$\begin{aligned} \eta &= \frac{1}{3} (\eta_A + \eta_B + \eta_C), \\ \tilde{\eta}_1 &= \frac{3}{2} [\frac{1}{2} (\eta_A + \eta_B) - \eta_C], \\ \tilde{\eta}_2 &= \eta_A - \eta_B. \end{aligned} \quad (22)$$

Notice, (20) implies $\eta(45^\circ) = \frac{1}{2} (\eta_A + \eta_B)$ where $\eta(45^\circ)$ in the effective viscosity for \mathbf{h} parallel to $\mathbf{e}^x \pm \mathbf{e}^y$. This special result is valid only up to order H^2 . In higher order, a contribution of the form $-2 \tilde{\eta}_3 \overline{\mathbf{h} \mathbf{h}} \cdot \gamma \cdot \mathbf{h}$ in (8) with $\tilde{\eta}_3 \sim H^4$ leads to $2 \eta(45^\circ) - (\eta_A + \eta_B) = \tilde{\eta}_3 \neq 0$.

The results presented in this section show that the two viscosity coefficients $\tilde{\eta}_1$ and $\tilde{\eta}_2$ associated with the molecular alignment can be determined independently either by measuring the field-induced change of the effective viscosity (20) for various directions of the magnetic field or by investigating the transverse effects (14, 18) on the normal pressure differences (15). In nematic liquid crystals where the magnetic-field-induced anisotropy of the viscosity is rather large, differences between the viscosities η_A , η_B , η_C , cf. (21), have been discovered quite some time ago [1–3, 13] for a more recent careful experimental study, see [14]. In the case of

isotropic liquids, the effect of a magnetic field on the viscosity is expected to be rather small. Thus differential measuring techniques are required analogous to those developed for the study of the influence of a magnetic field on the transport properties of molecular gases [15–19] (Senftleben-Beenakker effect); also transverse effects [19–20] have been measured in gases and liquid crystals [21]. For a discussion of the experimental arrangements needed to determine all viscosity coefficients see [19, 22, 23].

Results of a Fokker-Planck Equation Approach

In a mesoscopic theory which is intermediate between the phenomenological irreversible thermodynamics and a true microscopic theory, the dynamics of the alignment can be treated on the basis of a (generalised) Fokker-Planck equation for the orientational distribution function. For colloidal solutions, this approach has been developed some time ago [7, 24]; for its extension to the flow alignment of liquid crystals and the non-Newtonian viscosity of molecular liquids see [5] and [12], respectively. It should be mentioned that the influence of an orienting electric field on the viscosity of a colloidal solution was also studied in [24]. The basic physics underlying the present theory is described as follows. The orientational dynamics of a molecule is governed by a friction torque (which implies that \mathbf{u} undergoes a rotational diffusion) and by orienting torques which may stem from an external field or from an internal molecular field. In the presence of a viscous flow, the molecules acquire an average angular velocity equal to the vorticity $\boldsymbol{\omega} = \frac{1}{2} \text{rot } \mathbf{v}$ of the flow field and an additional orienting torque $\sim \mathbf{u} \times (\boldsymbol{\gamma} \cdot \mathbf{u})$ is exerted. The size of the latter torque is characterized by a parameter R which depends on the “shape” of a particle, e.g. a hydrodynamic model yields $R = (p^2 - 1)(p^2 + 1)$ where $p = a/b$ is the ratio between the semiaxes a and $b = c$ of an ellipsoid of revolution. In general, R will not be given by this hydrodynamic expression, but one still has $R > 0$ for prolate and $R < 0$ for oblate molecules.

From the Fokker-Planck equation the following equation is derived for the alignment tensor [5, 12] \mathbf{a} :

$$\frac{\partial}{\partial t} \mathbf{a} - 2(\boldsymbol{\omega} \times \mathbf{a} + \frac{3}{7} R \overline{\boldsymbol{\gamma} \cdot \mathbf{a}}) + 6w(\mathbf{a} - \mathbf{a}^{\text{eq}}) = \sqrt{\frac{6}{5}} R \boldsymbol{\gamma}, \quad (23)$$

where \mathbf{a}^{eq} is the field-induced equilibrium alignment e.g. given by (2, 3). Comparison with the phenomenological theory shows that the relaxation time coefficients τ_a and τ_{ap} are related to the rotational diffusion coefficient w and to the molecular nonsphericity parameter R by [5, 12]

$$\tau_a = (6w)^{-1}, \quad \tau_{\text{ap}} \tau_a^{-1} = -\sqrt{\frac{3}{5}} R. \quad (24)$$

As a consequence, the flow birefringence coefficient β is now given by

$$\beta = -(\varepsilon_a/5) R \tau_a. \quad (25)$$

In (23), terms involving the 4-th rank alignment tensor have been disregarded.

To make use of (23) for the viscosity problem, it is noticed that the friction pressure tensor \mathbf{p} can be written as [11, 12]

$$\mathbf{p} = -2\eta_{\text{iso}} \boldsymbol{\gamma} + \mathbf{p}_a, \quad (26)$$

where η_{iso} would be the viscosity if the coupling between the translational motion and orientation of the molecules could be switched off. The additive term

$$\mathbf{p}_a = \sqrt{2} P_k \tau_{\text{pa}} \tau_a^{-1} (\mathbf{a} - \mathbf{a}^{\text{eq}}) \quad (27)$$

is the friction pressure tensor associated with the alignment. Elimination of the difference $\mathbf{a} - \mathbf{a}^{\text{eq}}$ with the help of (23), subsequent use of $\mathbf{a} \approx \mathbf{a}^{\text{eq}} = \sqrt{\frac{3}{2}} a^{\text{eq}} \overline{\mathbf{h} \mathbf{h}}$ and of the relation (24), as well as $\tau_{\text{ap}} = \tau_{\text{pa}}$ leads to an expression of the form (8) where the viscosity coefficients are now given by

$$\eta = \eta_{\text{iso}} + \frac{3}{5} P_k R^2 \tau_a, \quad (28)$$

$$\tilde{\eta}_1 = -\frac{3}{7} R \tilde{\eta}_2, \quad (29)$$

$$\tilde{\eta}_2 = -\frac{3}{5} P_k R \sqrt{5} a_{\text{eq}} \tau_a. \quad (30)$$

Notice that $\tilde{\eta}_2 < 0$ and $\tilde{\eta}_2 > 0$ for prolate and oblate particles respectively, and $\tilde{\eta}_1 > 0$ in both cases. The relation (29) between $\tilde{\eta}_1$ and $\tilde{\eta}_2$ implies

$$\eta_B - \eta_A = \frac{7}{2} R \left[\frac{1}{2} (\eta_A + \eta_B) - \eta_C \right] \quad (31)$$

for the effective viscosity coefficients discussed in the previous section.

It should be stressed again that (11) is more general than the model dependent relations (28–31). The relaxation time τ_a occurring in (28, 30) can be determined experimentally by various methods, e.g. broadening of the depolarized Rayleigh line, study

of the viscoelasticity or the "inertia" of the Cotton-Mouton effect.

Some remarks on the pretransition region of the isotropic phase of a nematic liquid crystal are in order. There, in equilibrium alignment (3) underlying the Cotton-Mouton effect (5) and the flow birefringence (10) are increased by the factor $(1 - T^*/T)^{-1}$ where T^* is a temperature slightly below the temperature T_K where the isotropic-nematic transition takes place. This experimentally well established behavior [1–3] which has not been taken into account here can be understood within the phenomenological theory [4, 25] and it also follows from the Fokker-Planck equation approach [5].

The relations (29, 30) for the viscosity coefficients $\tilde{\eta}_1$, $\tilde{\eta}_2$ remain valid but now a_{eq} is enhanced over the expression (3) by the factor $(1 - T^*/T)^{-1}$, (28) is not affected. On the right hand side of (11) a factor $(1 - T^*/T)$ has to be inserted if it is understood that β and a_{eq} are inferred from birefringence measurements. In any case, the pretransitional increase of a_{eq} will help to detect the field-induced anisotropy of the viscosity in the isotropic liquid phase.

Concluding Remarks

In this article, the anisotropy of the effective viscosity, the normal pressure differences and the transverse viscosity effects have been considered in lowest order in the magnitude of an orienting field. In this case there are two additional viscosity coefficients associated with the molecular alignment. One of them is linked with the flow birefringence via an Onsager-symmetry relation; the Fokker-Planck equation yields a second inter-relation. In general, there exists one further viscosity coefficient (which is of higher order in the orienting field); it seems to be small even in nematic liquid crystals [14] where one has a rather strong alignment.

Experimental studies of the field-induced anisotropy of the viscosity are desirable for liquids where equilibrium and nonequilibrium birefringence studies have been made or can also be made; liquids composed of relatively large non-spherical molecules, as well as nematic liquid crystals in the isotropic phase are good candidates. The theory for the viscosity presented here could also be adopted to nonspherical magnetic colloids.

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