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PREDICTION OF LESLIE COEFFICIENTS FOR RODLIKE POLYMER NEMATICS

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ABSTRACT. Doi's molecular theory for the dynamics of rod-like polymers in concentrated solutions is here extended to include the effect of a molecular field. The resulting expression for the stress tensor is then linearized so as to obtain the Leslie coefficients in the nematic phase. Each of these coefficients takes the form of a basic viscosity - dependent on rod concentration and rotational diffusion constant - times a different function of the equilibrium order parameter. The predicted ordering of the three Miesowicz viscosities is in agreement with the experimental evidence.

INTRODUCTION. A molecular theory for the dynamics of rod-like polymers in concentrated solutions has been recently advanced by Doi[1]. The theory applies to both the isotropic and the nematic phases. It predicts the phase transition and shows the correct behavior of the shear viscosity in either phase.

The equations emerging from the theory are finally written by Doi in terms of the orientational order parameter tensor $\underset{\approx}{S}$ which is defined through the average

$$\underset{\approx}{S} = \left\langle \underset{\sim}{u}\underset{\sim}{u} - \frac{1}{3}\underset{\approx}{1} \right\rangle \tag{1}$$

where $\underset{\sim}{u}$ is a unit vector describing the rod orientation and $\underset{\approx}{1}$ is the unit tensor. Doi's equations give the rate of change of $\underset{\approx}{S}$ as

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$$d\underset{\approx}{S}/dt \;=\; \underset{\approx}{F}(\underset{\approx}{S}) \;+\; \underset{\approx}{G}(\underset{\approx}{S}) \qquad (2)$$

$$\underset{\approx}{F}(\underset{\approx}{S}) = -6\overline{D}_r \;\{(1-\frac{U}{3})\underset{\approx}{S}-U\left[\underset{\approx}{S}\cdot\underset{\approx}{S} - \frac{1}{3}(\underset{\approx}{S}:\underset{\approx}{S})\underset{\approx}{1}\right]+U(\underset{\approx}{S}:\underset{\approx}{S})\underset{\approx}{S}\;\} \qquad (3)$$

$$\underset{\approx}{G}(\underset{\approx}{S}) = \frac{1}{3}(\underset{\approx}{\kappa}+\underset{\approx}{\kappa}^{\dagger})+\left[\underset{\approx}{\kappa}\cdot\underset{\approx}{S}+\underset{\approx}{S}\cdot\underset{\approx}{\kappa}^{\dagger} - \frac{2}{3}(\underset{\approx}{\kappa}:\underset{\approx}{S})\underset{\approx}{1}\right] -2(\underset{\approx}{\kappa}:\underset{\approx}{S})\underset{\approx}{S} \qquad (4)$$

and the stress tensor $\underset{\approx}{\sigma}$ as

$$\underset{\approx}{\sigma} \;=\; -\,\frac{c\,k_B T}{2\overline{D}_r}\;\underset{\approx}{F}(\underset{\approx}{S}) \qquad (5)$$

In Eqs (2÷5), $\underset{\approx}{\kappa}$ is the velocity gradient and U is the parameter of the Maier-Saupe potential which depends on concentration c and temperature T; k_B is Boltzmann constant and $\overline{D}_r$ is a rotational diffusion constant which depends also on $\underset{\approx}{S}:\underset{\approx}{S}$.

The assumptions and the approximations used to derive Eqs (2÷5) do not seem very demanding. As explained by Doi[1], they are: i) the assumption of a Maier-Saupe type potential, ii) decoupling approximations of the form $\langle \underset{\sim}{u}\underset{\sim}{u}\underset{\sim}{u}\underset{\sim}{u}\rangle \simeq \langle \underset{\sim}{u}\underset{\sim}{u}\rangle\langle \underset{\sim}{u}\underset{\sim}{u}\rangle$, iii) an approximation concerning the orientational dependence of the diffusion constant. However, the specific form of the dependence of $\overline{D}_r$ on the orientational order parameter is of no consequence in what follows.

Doi's theory is a molecular theory of nemato-dynamics. It is therefore interesting to perform a comparison with the classical phenomenological ELP theory by Ericksen[2], Leslie[3] and Parodi[4] (as shown by de Gennes[5] the more recent approach by the Harvard group is also equivalent to the ELP theory). In particular, it seems attractive to find predictive expressions of the Leslie coefficients in terms of molecular quantities.

However, Doi's and ELP theories cannot be compared immediately. It may be sufficient to note that in ELP theory the viscous stress is not symmetric in general, whereas Eq.(5) always gives a symmetric stress. On the other hand, Eqs (2÷5) predict in general a non-linear dependence of $\underset{\approx}{\sigma}$ on $\underset{\approx}{\kappa}$, whereas

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the ELP theory is limited to the linear range.

The stress symmetry, as given by Eq.(5), is due to the fact that in Doi's theory the effects of a "molecular field" have been left out. As shown e.g. by de Gennes[5], a molecular field arises from the distortion energy whenever the director is not uniform in the nematic or else from an external field, such as a magnetic field. In what follows, we shall extend Doi's theory to include such effects. As expected, the stress tensor becomes non-symmetric in the general case.

The resulting constitutive equation is then linearized to provide a comparison with the ELP theory. Definite predictions for the Leslie coefficients are thereby obtained.

THE CONTRIBUTION OF A MAGNETIC FIELD. For the sake of simplicity, we shall assume that the molecular field is made up entirely of an uniform magnetic field $\underset{\sim}{H}$. If $\chi = \chi_{\parallel} - \chi_{\perp}$ is the difference in magnetic susceptibility , parallel and perpendicular to the rod axis, the potential associated to the rod direction $\underset{\sim}{u}$ is written as

$$V(\underset{\sim}{u}) = -\tfrac{1}{2}\,\chi\,(\underset{\sim}{H}\cdot\underset{\sim}{u})^2 \qquad (6)$$

As shown by Doi[1], the kinetic equation for the rod distribution function $f(\underset{\sim}{u})$ is

$$\partial f/\partial t = \bar{D}_r\ \underset{\sim}{\nabla}\cdot\left[\underset{\sim}{\nabla} f + \frac{f}{k_B T}\ \underset{\sim}{\nabla} V_{tot}\right] - \underset{\sim}{\nabla}\cdot\ \dot{\underset{\sim}{u}} f \qquad (7)$$

where $V_{tot}(\underset{\sim}{u})$ is the total potential of a rod and $\dot{\underset{\sim}{u}}$ is the rate of change due to the velocity gradient

$$\dot{\underset{\sim}{u}} = \underset{\approx}{\kappa}\cdot\underset{\sim}{u} - (\underset{\sim}{u}\cdot\underset{\approx}{\kappa}\cdot\underset{\sim}{u})\underset{\sim}{u} \qquad (8)$$

By assuming that there is no external field, Doi substitutes for $V_{tot}(\underset{\sim}{u})$ a Maier-Saupe type potential. Multiplying $\underset{\sim}{u}\underset{\sim}{u} - 1/3\ \underset{\approx}{1}$ to both sides of Eq.(7) and integrating over $\underset{\sim}{u}$ gives Eqs (2÷4).

Here, we include in $\underset{\sim}{\nabla} V_{tot}$ the contribution from the magnetic field, which is

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$$\underset{\sim}{\nabla}V = -\chi\left[(\underset{\sim}{H}\cdot\underset{\sim}{u})\underset{\sim}{H} - (\underset{\sim}{H}\cdot\underset{\sim}{u})^2\underset{\sim}{u}\right] \tag{9}$$

and since

$$\iint(\underset{\sim}{\nabla}\cdot f\underset{\sim}{\nabla}V)(\underset{\sim}{u}\underset{\sim}{u} - \frac{1}{3}\underset{\approx}{1})d^2u = -\iint f(\underset{\sim}{u}\underset{\sim}{\nabla}V + \underset{\sim}{\nabla}V\underset{\sim}{u})d^2u =$$

$$= -\left\langle \underset{\sim}{u}\underset{\sim}{\nabla}V + \underset{\sim}{\nabla}V\underset{\sim}{u}\right\rangle \tag{10}$$

Eq.(2) becomes

$$d\underset{\approx}{S}/dt = \underset{\approx}{F}(\underset{\approx}{S}) + \underset{\approx}{M}(\underset{\approx}{S}) + \underset{\approx}{G}(\underset{\approx}{S}) \tag{11}$$

where

$$\underset{\approx}{M}(\underset{\approx}{S}) = \frac{\bar{D}_r\chi}{k_BT}\left[\frac{2}{3}(\underset{\sim}{H}\underset{\sim}{H} - \frac{H^2}{3}\underset{\approx}{1}) - \frac{2}{3}H^2\underset{\approx}{S} + \underset{\sim}{H}\underset{\sim}{H}\cdot\underset{\approx}{S} + \underset{\approx}{S}\cdot\underset{\sim}{H}\underset{\sim}{H} - \right.$$

$$\left. - \frac{2}{3}(\underset{\sim}{H}\underset{\sim}{H}:\underset{\approx}{S})\underset{\approx}{1} - 2(\underset{\sim}{H}\underset{\sim}{H}:\underset{\approx}{S})\underset{\approx}{S}\right] \tag{12}$$

Following Doi and Edwards^6, the stress tensor is obtained by calculating the free energy change due to a hypothetical infinitesimal deformation. However, we must account of the fact that the stress is not symmetric in this case. If $\underset{\approx}{Q}$ describes the hypothetical deformation, in the sense that a vector $\underset{\sim}{r}$ embedded in the continuum changes to $(\underset{\approx}{1} + \underset{\approx}{Q})\cdot\underset{\sim}{r}$ after the deformation, the change in free energy density ΔA and the stress $\underset{\approx}{\sigma}$ are related by

$$\Delta A = \underset{\approx}{Q}:\underset{\approx}{\sigma} \tag{13}$$

When $\underset{\approx}{\sigma}$ is symmetric, Eq.(13) and Eq.(A-12) of Doi and Edwards^6 coincide.

The free energy density contributed by the magnetic field is $c\langle V\rangle$, where V is given by Eq.(6). The corresponding change associated to $\underset{\approx}{Q}$ is

$$\Delta A = c\left\langle \underset{\sim}{\nabla}V\cdot\underset{\approx}{Q}\cdot\underset{\sim}{u}\right\rangle = c\ \underset{\approx}{Q}:\left\langle \underset{\sim}{u}\underset{\sim}{\nabla}V\right\rangle \tag{14}$$

To the stress given by Eq.(5), the magnetic field then adds the contribution

$$\underset{\approx}{\sigma}_M = c\left\langle \underset{\sim}{u}\underset{\sim}{\nabla}V\right\rangle = -c\chi\left[\left\langle \underset{\sim}{u}\underset{\sim}{u}\right\rangle\cdot\underset{\sim}{H}\underset{\sim}{H} - (\left\langle \underset{\sim}{u}\underset{\sim}{u}\right\rangle:\underset{\sim}{H}\underset{\sim}{H})\left\langle \underset{\sim}{u}\underset{\sim}{u}\right\rangle\right] \tag{15}$$

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It may be noted that the first term on the right-hand side of Eq.(15) is generally not symmetric.

By combining Eqs (5) and (15), the total stress may be written as

$$\underset{\approx}{\sigma} = - \frac{ck_BT}{2\bar{D}_r}\left[\underset{\approx}{F}(\underset{\approx}{S})+\underset{\approx}{M}(\underset{\approx}{S}) - \frac{\bar{D}_r\chi}{k_BT}(\underset{\sim}{H}\underset{\sim}{H}\cdot\underset{\approx}{S} - \underset{\approx}{S}\cdot\underset{\sim}{H}\underset{\sim}{H})\right] \tag{16}$$

Eqs (11) and (16) provide the rheological constitutive equation when a magnetic field is present. For a given velocity gradient $\underset{\approx}{\kappa}(t)$ (as observed when moving with the fluid velocity), Eq.(11) gives $\underset{\approx}{S}(t)$, and $\underset{\approx}{\sigma}(t)$ is then calculated by means of Eq.(16). In the general case, the response is non-linear and viscoelastic.

EQUILIBRIUM CONDITION. In the absence of a velocity gradient, $\underset{\approx}{S}$ will be axially symmetric around the direction of the magnetic field (we are here assuming that χ is positive). Indicating such direction by a unit vector $\underset{\sim}{n}$, we write the equilibrium value of $\underset{\approx}{S}$ as

$$\underset{\approx}{S}_{eq} = S(\underset{\sim}{n}\underset{\sim}{n} - \frac{1}{3}\underset{\approx}{1}) \tag{17}$$

where S is determined by the condition

$$\underset{\approx}{F}(\underset{\approx}{S}_{eq}) + \underset{\approx}{M}(\underset{\approx}{S}_{eq}) = \underset{\approx}{0} \tag{18}$$

i.e.

$$(1 - \frac{U}{3})S - \frac{1}{3}US^2 + \frac{2}{3}US^3 = \frac{1}{9}\frac{\chi H^2}{k_BT}(1+S-2S^2) \tag{19}$$

and the stress $\underset{\approx}{\sigma}$ is obviously isotropic.

With respect to the result by Doi[1], the equilibrium value of the order parameter given by Eq.(19) is somewhat increased by the magnetic contribution. However, for ordinary values of χ and H, this effect is expected to be minor. For $U < 3$, S is essentially zero and the system is isotropic. For $U > 3$, it becomes a nematic with an S-value of 0.5 or larger.

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SLOW FLOWS. If the velocity gradient $\underset{\approx}{\kappa}(t)$ is sufficiently small, one expects that the nematic axis remains aligned with the magnetic field in spite of the flow. In other words, we expect that the order parameter tensor may be written as

$$\underset{\approx}{S} = \underset{\approx}{S}_{eq} + \underset{\approx}{S}' \tag{20}$$

with $\underset{\approx}{S}'$ a perturbation of order $\underset{\approx}{\kappa}$.

Furthermore, if $\underset{\approx}{\kappa}(t)$ varies slowly along the trajectories we may neglect memory effects and write Eq.(11) as

$$\underset{\approx}{F}(\underset{\approx}{S}) + \underset{\approx}{M}(\underset{\approx}{S}) + \underset{\approx}{G}(\underset{\approx}{S}) = \underset{\approx}{0} \tag{21}$$

which can be expanded to provide a linear algebraic relationship between $\underset{\approx}{S}'$ and $\underset{\approx}{\kappa}$.

By using Eqs (20) and (21), the viscous stress is written in a more convenient form as

$$\underset{\approx}{\sigma} = \frac{ck_BT}{2\bar{D}_r}\left[\underset{\approx}{G}(\underset{\approx}{S}_{eq}) + \frac{\bar{D}_r\chi H^2}{k_BT}(\underset{\sim}{n}\underset{\sim}{n}\cdot\underset{\approx}{S}'-\underset{\approx}{S}'\cdot\underset{\sim}{n}\underset{\sim}{n})\right] \tag{22}$$

Here, $\underset{\approx}{G}(\underset{\approx}{S}_{eq})$ is already explicit in $\underset{\approx}{\kappa}$ and $\underset{\sim}{n}$, whereas the second term is to be obtained by solving Eq.(21).

Subtracting Eq.(18) from Eq.(21) and expanding to first order in $\underset{\approx}{S}'$ and $\underset{\approx}{\kappa}$ gives

$$\left[1 - \frac{U}{3} + \frac{2}{3}US + \frac{2}{3}US^2 + \frac{2}{3}\bar{\chi}(1+2S)\right]\underset{\approx}{S}'-(US+\bar{\chi})(\underset{\sim}{n}\underset{\sim}{n}\cdot\underset{\approx}{S}'+\underset{\approx}{S}'\cdot\underset{\sim}{n}\underset{\sim}{n}) +$$

$$+ 2S(US+\bar{\chi})(\underset{\sim}{n}\underset{\sim}{n}:\underset{\approx}{S}')\underset{\sim}{n}\underset{\sim}{n} + \frac{2}{3}(1-S)(US+\bar{\chi})(\underset{\sim}{n}\underset{\sim}{n}:\underset{\approx}{S}')\ \underset{\approx}{1} =$$

$$= \frac{1}{6\bar{D}_r}\{\frac{1}{3}(1-S)(\underset{\approx}{\kappa}+\underset{\approx}{\kappa}^{\dagger})+S(\underset{\approx}{\kappa}\cdot\underset{\sim}{n}\underset{\sim}{n}+\underset{\sim}{n}\underset{\sim}{n}\cdot\underset{\approx}{\kappa}^{\dagger})-2S^2(\underset{\approx}{\kappa}:\underset{\sim}{n}\underset{\sim}{n})\underset{\sim}{n}\underset{\sim}{n} -$$

$$- \frac{2}{3}S(1-S)(\underset{\approx}{\kappa}:\underset{\sim}{n}\underset{\sim}{n})\ \underset{\approx}{1}\ \} \quad ; \qquad \bar{\chi} = \frac{1}{6}\ \frac{\chi H^2}{k_BT} \tag{23}$$

Pre - and postmultiplying Eq.(23) by $\underset{\sim}{n}\underset{\sim}{n}$ and taking the difference gives

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$$\frac{\bar{D}_r\chi H^2}{k_B T}(\underset{\sim}{n}\underset{\sim}{n}\cdot\underset{\approx}{S}'-\underset{\approx}{S}'\cdot\underset{\sim}{n}\underset{\sim}{n})=\frac{S(1-S)}{2+S}\left[\underset{\sim}{n}\underset{\sim}{n}\cdot(\underset{\approx}{\kappa}+\underset{\approx}{\kappa}^{\dagger})-(\underset{\approx}{\kappa}+\underset{\approx}{\kappa}^{\dagger})\cdot\underset{\sim}{n}\underset{\sim}{n}\right]+$$

$$+\ \frac{3S^2}{2+S}\,(\underset{\sim}{n}\underset{\sim}{n}\cdot\underset{\approx}{\kappa}^{\dagger}-\underset{\approx}{\kappa}\cdot\underset{\sim}{n}\underset{\sim}{n}) \tag{24}$$

where use has been made of the relationship which links S, U and χ, Eq.(19). Finally, by letting

$$\underset{\approx}{A}=\tfrac{1}{2}\,(\underset{\approx}{\kappa}+\underset{\approx}{\kappa}^{\dagger}) \tag{25}$$

$$\underset{\approx}{\Omega}=\tfrac{1}{2}\,(\underset{\approx}{\kappa}-\underset{\approx}{\kappa}^{\dagger}) \tag{26}$$

the viscous stress takes the form

$$\underset{\approx}{\sigma}=\frac{ck_BT}{2\bar{D}_r}\left[\frac{2}{3}\,(1-S)\underset{\approx}{A}+2S\,\underset{\sim}{n}\underset{\sim}{n}\cdot\underset{\approx}{A}-2S^2(\underset{\approx}{A}:\underset{\sim}{n}\underset{\sim}{n})\underset{\sim}{n}\underset{\sim}{n}\right.$$

$$\left.-\ S(1+\frac{3S}{2+S})\ \underset{\sim}{n}\underset{\sim}{n}\cdot\underset{\approx}{\Omega}\ +\ S(1-\frac{3S}{2+S})\underset{\approx}{\Omega}\cdot\underset{\sim}{n}\underset{\sim}{n}\right] \tag{27}$$

where we have dropped an irrelevant isotropic term.

It should be noted that the result comes out independent of the intensity of the molecular field, i.e. of χ, but for its possible influence on the equilibrium order parameter S. The effect of the molecular field is essentially that of determining the direction of the nematic axis, $\underset{\sim}{n}$. Here we have assumed that $\underset{\sim}{n}$ is constant along a trajectory, but the result is readily extended to a variable $\underset{\sim}{n}$ by a suitable change of reference frame. In fact, Eq.(27) becomes frame indifferent if $-\ \underset{\approx}{\Omega}\cdot\underset{\sim}{n}$ is substituted for by the vector

$$\underset{\sim}{N}=\dot{\underset{\sim}{n}}-\underset{\approx}{\Omega}\cdot\underset{\sim}{n} \tag{28}$$

THE LESLIE COEFFICIENTS. Within the common factor $ck_BT/2\bar{D}_r$, the Leslie coefficients predicted by Eq.(27) are

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$$\alpha_1 = -2S^2 \qquad \alpha_2 = -S(1+\frac{3S}{2+S}) \qquad \alpha_3 = -S(1-\frac{3S}{2+S})$$

$$\alpha_4 = \frac{2}{3}(1-S) \qquad \alpha_5 = 2S \qquad \alpha_6 = 0 \tag{29}$$

which duly satisfy the Parodi relation $\alpha_2+\alpha_3 = \alpha_6-\alpha_5$.

The Miesowicz viscosities come out as

$$\eta_a = \tfrac{1}{2}\,\alpha_4 = \frac{ck_BT}{6\bar{D}_r}(1-S)$$

$$\eta_b = \tfrac{1}{2}(\alpha_3+\alpha_4+\alpha_6) = \frac{ck_BT}{6\bar{D}_r}\,\frac{(1-S)^2}{1+S/2} \tag{30}$$

$$\eta_c = \tfrac{1}{2}(\alpha_4+\alpha_5-\alpha_2) = \frac{ck_BT}{6\bar{D}_r}\,\frac{(1+2S)^2}{1+S/2}$$

showing the correct ordering $\eta_c > \eta_a > \eta_b$ as experimentally observed. For example, for $S = 0.5$, we find $\eta_a/\eta_b = 2.5$, $\eta_c/\eta_b = 16$. These values are larger than experimentally found for thermotropic nematics. However, the model considered here strictly applies to very long thin rods, i.e. to lyotropic rigid polymers. It is perhaps to be expected that for shorter molecules such as those of thermotropic systems, the contribution of the rotational diffusion is but a part of the viscous stress. Also the prediction $\alpha_6 = 0$ does not compare favorably with some results on thermotropic systems[5]. The theoretical result $\alpha_6 = 0$ might be particularly sensitive to the decoupling approximation adopted here : $\langle \underset{\sim}{uuuu} \rangle \simeq \langle \underset{\sim}{uu} \rangle \langle \underset{\sim}{uu} \rangle$.

The characteristic ratio for flow orientation in simple shear is found to be

$$\lambda = -\frac{\gamma_2}{\gamma_1} = \frac{\alpha_5-\alpha_6}{\alpha_3-\alpha_2} = \frac{2}{3}\,\frac{1+S/2}{S} > 1 \tag{31}$$

which implies stability over the whole range of S-values, $\cos 2\theta = 1/\lambda$ determining the angle θ that the director

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chooses with respect to the flow lines when the molecular field is absent or negligible[5]. The shear viscosity η which is obtained in these conditions corresponds to a symmetric stress (zero torque) and it is derived either from Eq.(27) or directly from Eqs (2-5) in the limit of small shear rates. It was in fact calculated by Doi[1] as

$$\eta = \frac{ck_B T}{6\bar{D}_r} \; \frac{(1-S)^2(1+2S)(1+3S/2)}{(1+S/2)^2} \qquad (32)$$

It is then found $\eta_c > \eta > \eta_b$ for all values of S. Conversely, the ratio η/η_a, which decreases by increasing S, crosses unity when $S = (\sqrt{33} - 1)/8 \simeq 0.593$. This crossing of η and η_a is perhaps to be expected also for thermotropic systems and it might be amenable to experimental verification.

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