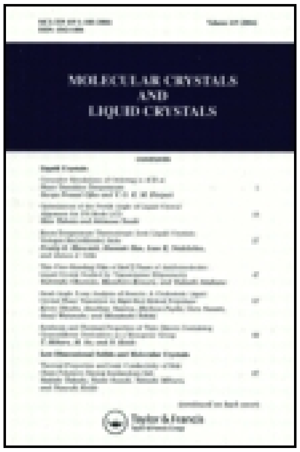

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Transport Processes in Anisotropic Fluids II. Coupling of Momentum and Energy Transport in a Nematic Mesophase

Jay Fisher [a] [b] & A. G. Fredrickson [a]
[a] Chemical Engineering Department, University of Minnesota, Minneapolis, 55455
[b] Amoco Chemicals Corp., Whiting, Indiana

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Transport Processes in Anisotropic Fluids

II.‡ Coupling of Momentum and Energy Transport in a Nematic Mesophase.§

JAY FISHER|| and A. G. FREDRICKSON

Chemical Engineering Department
University of Minnesota
Minneapolis 55455

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Introduction

It is a well established fact that transport processes in fluid substances of low molecular weight are adequately described by linear constitutive relations of the form

$$\mathbf{J} = -k\mathbf{\Phi}. \tag{1}$$

Here, $\mathbf{J}$ is the *flux* of the entity to be transported and $-\mathbf{\Phi}$ is the *driving force*—always the gradient of some potential function—for transport of that entity. The parameter k is the *conductivity*; it depends on the thermodynamic state of the phase in which transport is occurring, but not on the flux or the driving force. Classical examples of Eq. (1) are Fick's law of diffusion, Fourier's law of heat conduction, Ohm's law of electrical conduction, and Newton's law of momentum transport (viscous friction). In the first three cases cited above, a scalar quantity is transported so that $\mathbf{J}$ and $\mathbf{\Phi}$ are *vectors*; in the last case cited, a *vector* quantity is transported so that $\mathbf{J}$ and $\mathbf{\Phi}$ are *tensors* of second order.

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‡ The first paper in this series, "Interfacial Effects on the Viscosity of a Nematic Mesophase", by Jay Fisher and A. G. Fredrickson, was published in *Molecular Crystals and Liquid Crystals*.

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|| Present address: Amoco Chemicals Corp., Whiting, Indiana.

If Eq. (1) is strictly true, there can be no *coupling* between different transport processes. That is to say, the presence of a driving force for heat conduction (a temperature gradient) would not affect the flux of mass, nor would the presence of a driving force for momentum transport (a velocity gradient or shear field) affect the flux of heat, and so on. It is well-known[3] that certain kinds of couplings do in fact exist; these always involve fluxes which have the same tensorial character. In other words, couplings are found between heat and mass transport, but not between heat or mass transport and momentum transport.

It is a theorem of irreversible thermodynamics that coupling cannot exist between a vector transport process (heat, mass, electricity) and a tensor transport process (momentum transport). This result, known as *Curie's theorem*[4], is a consequence of two basic assumptions concerning transport processes; namely, that fluxes are *linear* in the driving forces, and that the material considered is *isotropic*.

Now liquid crystalline phases are surely not isotropic—indeed, perhaps the best name for them might be *anisotropic fluids*—and there exists evidence which could be interpreted in favor of a nonlinear constitutive equation for them. This last refers to the non-Newtonian viscosity found with mesophases.[18] We believe that the constitutive equations for mesophases are in fact quasi-linear in the driving forces and it is well-known that the non-Newtonian viscosity of mesophases can be explained without invoking nonlinear constitutive relations. Nevertheless, the anisotropy of liquid crystals remains, and this suggests that the laws governing transport in mesophases must be more complicated than the laws governing transport in ordinary fluids.

It is the purpose of this series of papers to elucidate these laws by experimental and theoretical studies of transport in mesophases. In the present paper, attention is focused on the process of heat conduction. We show experimentally that heat conduction is affected by momentum transport in a nematic mesophase, so that Curie's theorem is invalid for such materials. We also show experimentally that heat transfer is affected by an electric field, although the effect is probably due to convection caused by the field. We compare the results with a theory of transport based on the so-called " swarm model " of nematic mesophases. The theory will not fit the observations made with electric fields present, but we believe this to be due to artifacts caused by the field rather than to any basic failure of the theory.

Theory

Any theory of transport in nematic mesophases must start with a description of the local structure of the mesophase and some assumptions as to the effect of externally applied influences on that structure. If the swarm model of the nematic mesophase is adopted, then the structure at a point is described by the distribution of swarm orientations in the neighborhood of the point. A distribution arises because swarms are subjected to random thermal torques as well as to non-random torques produced by the orienting influences of magnetic or electric fields, shear, or proximity to an interface. Equations of change describing the distribution of orientations have been developed by Fisher *et al.*,[8] and these should be applicable to the experiments described below.

In the continuum theory of Ericksen[5] and Leslie,[13] a different viewpoint is taken. The existence of swarms is not assumed, and local structure is described by a vector (ordinarily a *unit* vector, though this is not necessary) pointing in the direction of mean orientation at the point. Thus, the theory does not allow for a distribution of orientations, unless the mean orientation vector is permitted to have variable length. Equations of change for this vector are given in the references cited.

Once the description of local structure and the factors influencing it have been decided upon, the next step is to make some hypothesis concerning the effect of local structure upon the rates of transport processes. Obviously, this is most easily done by assuming that the conductivity depends on local structure. Eq. (1) will not serve here, however, since the existence of local structure must make the mesophase anisotropic. Hence, in place of Eq. (1), we write

$$\mathbf{J} = -\mathbf{K} \cdot \boldsymbol{\Phi} \tag{2}$$

where **K** is the conductivity tensor and a function of local structure.

If the swarm model is adopted, and if in addition one views the swarms as transversely isotropic solid bodies through which transport occurs by conduction alone, then a simple dependence of **K** on the distribution of swarm orientations results (Fisher *et al.*[8]). As applied to the problem of the effect of shear on thermal conductivity, this theory leads to the prediction that the conductivity should depend on the ratio of shear rate (σ) to the rotational diffusion coefficient (D_r) of swarms; the latter is of course a material constant.

Figure 1 shows the dependence of thermal conductivity on shear rate predicted by the theory of Fisher *et al.*[8] for the case where (a) the shear rate is uniform in the sample, (b) the surface-to-volume ratio of the sample is so small that the orienting influence of interfaces can be ignored, and (c) the imposed driving force (temperature gradient) is perpendicular to the flow but parallel to the speed gradient. In this figure, k is the ratio of the component of the heat flux vector in the direction of the driving force to the magnitude of the driving force. We shall call this the thermal conductivity, but it should be remembered that this is no longer a material property but is also dependent on shear rate as well as

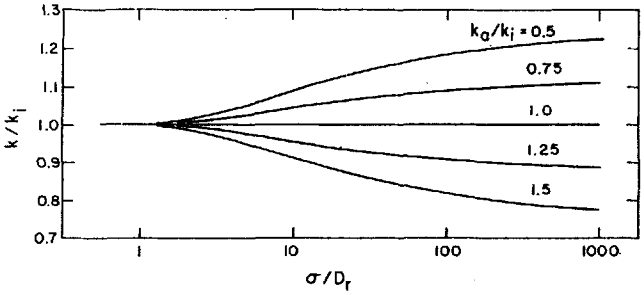

Figure 1. Variation of apparent conductivity with shear rate as predicted by the swarm theory. The transport flux is parallel to the velocity gradient but perpendicular to the velocity.

on the relative orientation of flow and temperature gradient. According to the swarm model, the sample should become isotropic at zero shear rate, and the corresponding isotropic conductivity—a material constant—is denoted by k_i. The curves shown in Fig. 1 have the ratio k_a/k_i as a parameter; the material constant k_a is the conductivity of a transversely isotropic swarm in its axial direction. Figure 1 predicts that shear will decrease the conductivity (for the case where flow and temperature gradient are perpendicular) if the axial conductivity is greater than the isotropic conductivity. Such a decrease is of course required under the assumption that shear tends to orient swarms with their axes parallel to the direction of flow.

If the continuum model of the mesophase is adopted, and if it is assumed that the conductivity tensor of Eq. (2) is transversely isotropic about the direction of mean orientation, then

$$\mathbf{K} = k_t \mathbf{I} + (k_a - k_t)\, \mathbf{dd} \tag{3}$$

where $\mathbf{I}$ is the unit tensor, k_t is the conductivity in any direction normal to $\mathbf{d}$, and $\mathbf{d}$ is the mean orientation vector of the continuum theory. In order to predict the effect of shear on thermal conductivity from this, one must know how $\mathbf{d}$ varies with shear rate. This problem has recently been considered by Atkin and Leslie[1] for the case of Couette flow between concentric cylinders. Unfortunately, it appears that numerical computation must be resorted to in order to evaluate $\mathbf{d}$ as a function of shear rate (and position!) so that we have prepared no graph, equivalent to Fig. 1, based on the continuum theory. Hence, it is possible at present to compare only the predictions of the swarm theory to the experimental data.

Experiments

EXPERIMENTAL APPARATUS

Thermal conductivity measurements were made in a concentric cylinder thermal conductivity cell. The cell was designed so that the outer cylinder could be rotated, thus shearing the fluid, and so that an electric field could be applied across the test fluid. A schematic illustration of the apparatus is shown in Figure 2, and an expanded diagram of the central portion with critical dimensions is shown in Figure 3.

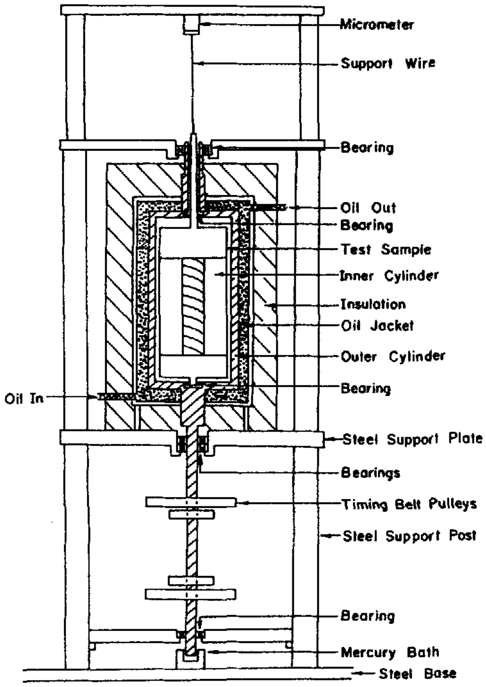

Figure 2. Schematic diagram of the experimental apparatus.

The central portion consists of two cylinders, vertically mounted with a narrow gap, 0.0508 cm or 0.02 inches, between them in which the test fluid is contained. The outer cylinder is supported from above and below by precision bearings as shown in Fig. 3. The inner cylinder is suspended from above by a steel wire and is centered within the outer cylinder by precision bearings as shown in Figure 2.

The inner cylinder was constructed in three sections. The

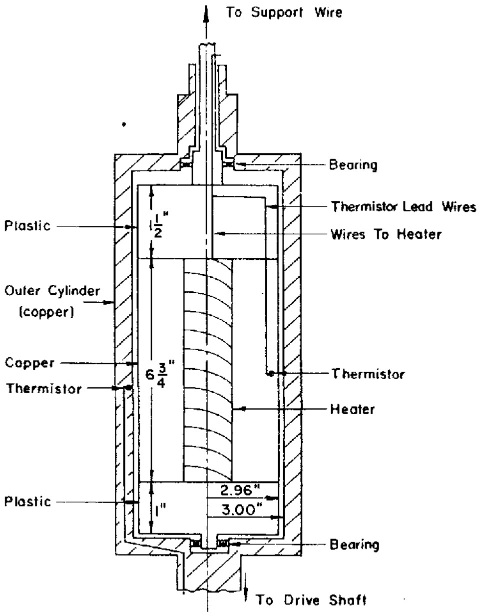

Figure 3. Detail diagram of the concentric cylinder thermal conductivity apparatus.

center section, which was the heating section, consisted of a copper cylinder with an electrical resistance heater running through its center. A thermistor to measure the temperature of the fluid at the inner cylinder was embedded in the copper halfway along its length and about $\frac{1}{8}$ inch from its outside edge. Above and below the heating section were sections of a G-7 silicone glass laminate. These served to thermally insulate the heating section from the end supports of the inner cylinder. The heating section was also electrically insulated from the outer cylinder and a wire was attached to the copper so that an electric field could be applied to the fluid.

The outer cylinder was made of copper and plated with 0.001 inch of nickel. The nickel plate was applied to reduce oxidation and the possibility of scratching. A thermistor was embedded midway along the length of the outer cylinder about $\frac{1}{8}$ inch from its inner surface.

Rotational speeds from 0.025 to 1000 revolutions per minute could be obtained. This gave shear rates from 0.2 to 8000 $\sec^{-1}$, although the highest used was 1000 $\sec^{-1}$. Because of the narrow gap width the shear rate would be uniform within about 3% throughout the fluid.

The outer cylinder was almost completely surrounded by an oil jacket which in turn was surrounded by a 3 inch thick layer of glass wool insulation. Mineral oil was pumped thru the oil jacket from a large constant temperature bath which was capable of maintaining the oil temperature constant within ± .01 °C.

The lead wires of the thermistor in the outer cylinder went down through the drive shaft and made contact with a stationary mercury bath. This procedure for getting the electrical signal off of the rotating shaft gave very little noise, and the mercury bath offered essentially no electrical resistance.

The resistance of the thermistors was measured using a Wheatstone bridge. It is believed that temperatures could be measured with a precision of ± 0.007 °C.

The power to the heater was provided by a D.C. power supply. The power input was measured by measuring the voltage across and the current thru the heater. The accuracy of the power measurement was ± 1% but in any series of experimental runs it was possible to maintain the power input to the heater constant within ± 0.3%.

A more complete description of the apparatus is given elsewhere.[6]

Materials

The only nematic mesophase studied was that of *p*-azoxyanisole. Preparation and criteria of purity were the same as those described in a previous paper.[7] Two calibrating fluids were also used. These were *n*-butanol, AR grade, obtained from the Mallinckrodt Chemical Co· and ethylene glycol obtained from Eastman Organic Chemicals.

Calibration of the Instrument

If it is assumed that all heat generated in the heater is conducted radially across the fluid and that no heat is conducted through the insulating sections of the inner cylinder, the thermal conductivity of the test liquid would be proportional to Q_h, the heat input to the inner cylinder, and inversely proportional to ΔT, the temperature difference between the cylinders. This simple situation does not obtain, however. In addition to the radial heat flux through the fluid there is also a heat flux through the insulation above and below the heating section of the inner cylinder and a non-radial heat flux through the fluid near the ends of the heated section. Rather than try to analyze this complex situation the thermal conductivity cell was calibrated with two fluids of known thermal conductivity.

Assume that the heat flux can be divided into two parts: that passing directly through the fluid and that which passes through the insulation at the ends of the heated section; *i.e.*,

$$Q_h = Q_f + Q_i . \tag{4}$$

The heat flux through the fluid, Q_f, is proportional to the thermal conductivity of the fluid and the temperature difference between the two cylinders. The heat flux through the insulation is assumed to be proportional to the temperature difference between the two cylinders. Thus

$$Q_h = kA\Delta T + B\Delta T . \tag{5}$$

where A and B are properties of the apparatus but not of the fluid under test.

Two compounds were used for calibration; these were ethylene glycol and *n*-butanol. The calibration runs were made at 70 °C at which temperature the thermal conductivities of the calibrating fluids are[23] 3.44×10^{-4} cal/cm sec °K for *n*-butanol and 6.21×10^{-4} cal/cm sec °K for ethylene glycol. Runs were made for each compound at 3 heating rates with ΔT varying from 0.6 to 1.8 °C. For all runs the variation of $Q_h/\Delta T$ for each compound was less than 0.7%. The data taken show that $A = 5.08 \times 10^3$ cm and $B = 0.168$ cal/sec °C for the apparatus.

An additional run at 120 °C was made with ethylene glycol to test the effect of absolute temperature on the calibration. The thermal conductivity as calculated from Eq. (5) was found to be 2% higher than that at 70 °C. Although there are no data for the thermal conductivity of ethylene glycol at this temperature, its thermal conductivity is known[23] to rise by about 0.7% per 10 °C between 15 and 90 °C. Thus the 2% rise in 50 °C was considered to be good indication that there were no major changes in the calibration at 120 °C

The thermal conductivity of ethylene glycol at 70 °C was also measured while shearing the fluid at rates of 100 and 200 sec^{-1}. There was no significant change in the thermal conductivity from that in the unsheared fluid.

POSSIBLE SOURCES OF ERROR

Possible sources of error in the measurement of thermal conductivity include (i) radiative transport, (ii) eccentricity of alignment of the cylinders, (iii) free convection, (iv) turbulence, (v) secondary flows due to eccentricity of alignment of the cylinders, (vi) viscous heating, (viii) ohmic heating (in those cases where an electric field was applied), and (vii) effects which disturb the measurement of cylinder temperatures. Calculations presented elsewhere[6] show that errors due to effects (i–ii and vi–viii) are essentially negligible or can be corrected for, and that effects (iii–iv) do not occur. Secondary flow due to misalignment of the cylinders is a theoretical possibility. However, the failure to find any effect of rotational speed of the outer cylinder on the thermal conductivity of ethylene glycol shows that such flows were not occurring in those experiments, and so we infer that they were not important in the experiment with *p*-azoxyanisole, either.

Results

The thermal conductivity of nematic *p*-azoxyanisole at 123 °C in the absence of externally applied fields was found to be 3.22×10^{-4} cal/sec cm °C. This agrees well with values of 3.31×10^{-4} cal/sec cm °C found by McCoy and Kowalczyk[14] and 3.2×10^{-4}

cal/sec cm °C found by Picot and Frederickson.[17] In addition, the measured thermal conductivity was independent of heat flux for temperature differences between the cylinders of 0.6 to 1.8 °C

The results for the apparent thermal conductivity of nematic *p*-azoxyanisole in the presence of a shear field are shown in Fig. 4.

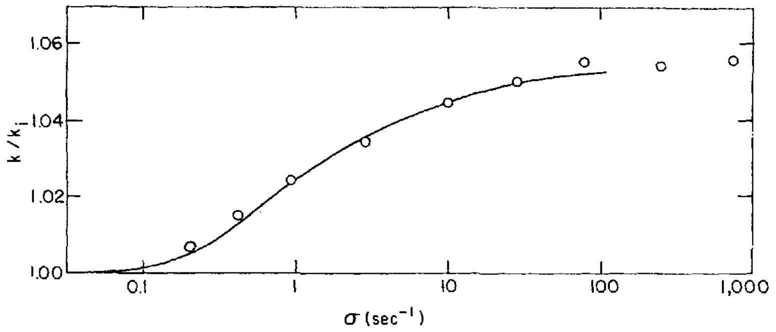

Figure 4. Effect of shear rate on the thermal conductivity of nematic *p*-azoxyanisole at 123 °C. Heat flux perpendicular to flow but parallel to velocity gradient. Solid line is the theoretical curve.

The thermal conductivity increases with increasing shear rate and appears to level off at about 3.41×10^{-4} cal/sec cm °C at shear rates above about 200 sec^{-1}. If the material were isotropic, the measured thermal conductivity should not change with shear.

When DC electric fields were applied across the fluid the apparent thermal conductivity rose with increasing field strength. At a field of 5000 volts/cm, the highest field used, the measured thermal conductivity was three times that with no electric field. However, several peculiar effects were noted during these experiments. When successively higher electric fields were applied the current through the fluid at first increased because of the higher fields but then dropped off to about $\frac{1}{4}$ of its initial value after about an hour. Also the electrical resistance of the fluid dropped from 10,000 ohms before the electric field experiments to about 750 ohms after the experiments. The thermal conductivity of the fluid with no applied field increased to about 1.5 times its initial value after the electric field experiments. It was noted upon taking the apparatus apart that the *p*-azoxyanisole had apparently reacted with the copper of the inner cylinder and in fact metallic copper was deposited on the nickel surface of the outer cylinder. Hence, experiments with DC fields were discontinued.

The results for the apparent thermal conductivity of nematic *p*-azoxyanisole in the presence of a 60 Hz. AC electric field are shown in Fig. 5. The thermal conductivities were again calculated from Eq. (5), but with the temperature drop corrected for ohmic heating. Although there was no apparent reaction of the fluid in these experiments some peculiar effects were observed again. There was no detectable change in apparent thermal conductivity for fields below 160 volts/cm. For fields from 400 to 1100 volts/cm there was a significant rise in the thermal conductivity. When the field was raised from 0 to 200 volts/cm the apparent thermal conductivity with 200 volts/cm was the same as with no field; however, when a 400 volt/cm field was applied for a minute and then reduced to 200 volts/cm a 5% to 7% rise in the apparent thermal conductivity was observed. This procedure was repeated several times yielding the same results.

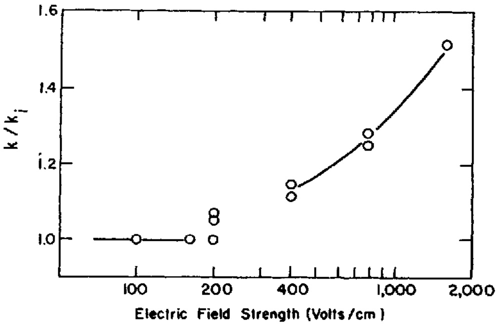

Figure 5. Effect of a 60 Hz. AC electric field on the apparent thermal conductivity of nematic *p*-azoxyanisole at 123 °C. Heat flux and electric field parallel.

Discussion of Results

The experiments reported above prove that heat conduction in *p*-azoxyanisole is influenced by momentum transport, in violation of Curie's theorem. The results of the experiments were reproducible within ±0.5%. The results obtained with isotropic ethylene glycol show that convection or secondary flows induced by the shear are not important factors in measuring the thermal conductivity.

There is however some question as to the significance of the results obtained with electric fields across the fluid. The effects noted in the 200 volt/cm range lead one to suspect that there are influences other than molecular orientation affecting the apparent thermal conductivity. Relaxation times for molecular orientation are short compared to the time scale of the experiments and one would not expect the orientation to depend strongly on the history of the electric field. It seems probable that the effects noted with 200 volt/cm fields, and at least most of the rise in apparent thermal conductivity were due to convection currents set up within the fluid by the electric field. Such effects have been found in nematic mesophases by Naggiar[15] and in isotropic dielectric fluids by Schmidt and Leidenfrost,[20] Gross and Porter,[9] and others. Heilmeier, Zanoni, and Barton[11] have demonstrated that electric fields do cause convection in nematic liquid crystals. It is possible that the large " domains " noted by Williams and Heilmeier[24] and by Heilmeier[10] are due to convection; indeed, they may be convection cells of the type familiar in hydrodynamics (see, *e.g.*, Chandrasekhar).[3] The effects we observed with fields of the order of 200 volts/cm could be explained by assuming that a field of that strength is not sufficient to initiate convection but could sustain it once it had been initiated by a stronger field of (say) 400 volts/cm.

In this paper, no further attempt will be made to analyze the results obtained with electric fields. Instead, we focus attention on the results obtained with a shear field present. We assume that the effects noted are due to shear induced orientation of domains. In Fig. 4, the experimental results for thermal conductivity as a function of shear are compared with the prediction of the swarm theory. Three material constants from the theory were determined for *p*-azoxyanisole at 123 °C from the experimental results. These were: (1) the thermal conductivity of the disordered fluid, $k_i = 3.22 \times 10^{-4}$ cal/cm sec °C; (2) the thermal conductivity along the long axis of the swarms, $k_a = 2.82 \times 10^{-4}$ cal/cm sec °C; and (3) the rotational diffusion coefficient, $D_r = 0.075\ \text{sec}^{-1}$. The theoretical curve in Fig. 4 has been plotted assuming the foregoing values for k_a and D_r.

From the measured value of the rotational diffusion coefficient and the calculations of Fisher *et al.*[8]—see especially Table 2 of that paper—it is possible to make a qualitative prediction concerning the effect of shear on the viscosity of *p*-azoxyanisole. From the table in the cited paper, swarm orientation should be virtually complete (degree of orientation = 0.8) at a shear rate of about 100 sec^{-1} if D_r is 0.075 sec^{-1}. Thus, the viscosity should become constant at shear rates of this order of magnitude and larger. The viscosity data of Fisher and Frederickson[7] taken at low shear rates are in agreement with this expectation.

Concluding Remarks

The effects of a shear field on thermal conductivity that we have found are consistent with the theory of Fisher *et al.*[8] The data also allow estimates to be made of several material constants appearing in that theory. This, however, is not sufficient to establish the validity of the theory; for that purpose we require that material constants obtained in a first set of experiments shall describe quantitatively the results of a second set of experiments, without the necessity of estimating the values of new constants from the results of the second set of experiments.

The experiments with electric fields were an attempt to partially satisfy the criterion laid down above. The results of these experiments were inconsistent with the theory of orientation by an electric field and the constants derived from the shear experiments. We believe, however, that this was due to the complicating effects of convection. In retrospect, it seems that experiments on the effect of *magnetic* fields on thermal conductivity would have been more appropriate, as such fields apparently produce no convection and there is little doubt about the direction of orientation that they produce. It is hoped that the results of such experiments will soon be forthcoming.

Finally, we comment on two conceptual possibilities concerning transport in liquid crystals. Stewart, Holland, and Reynolds[22] report data which they cite as evidence for an orienting effect of a heat flux. In a later note,[21] Stewart states that the effects noted were not due to a number of causes, among which he lists convection. Picot[16] rejected this conclusion; he felt that the effects noted were due to convection. Some of the data reported above reinforce Picot's conclusion: the measured thermal conductivity was independent of heat flux in the range of temperature differences used (0.6–1.8 °C). Hence, Fisher *et al.*[8] adhered to Picot's rejection of heat flux as a possible orienting influence, and added no term involving the heat flux or the temperature gradient to the factors influencing the orientation of swarms.

The other possibility concerns the nature of the conductivity tensor, **K**. According to the hypotheses advanced in this paper, **K** depends only on the local and instantaneous structure of the mesophase. However, it is conceivable that the conductivity tensor could also depend on the *spin* of domains at the point in question. In the Ericksen–Leslie theory[5,13] of liquid crystals, the momentum flux tensor is indeed assumed to depend on spin as well as on static orientation. A simple physical mechanism of energy transport involving spin is easily imagined: A spinning domain in a temperature gradient will pick up thermal energy along the portion of its periphery on the " hot side " of the domain. Rotation of the domain will move the heated portion of the periphery to the " cold side " of the domain, where the excess thermal energy will be lost.

It is easy to show that this mechanism will not be important in *p*-azoxyanisole at the shear rates used in these experiments. The spin rate of a domain in a shear field will be of the same order of magnitude as the magnitude of the shear rate. Thus, the time required to transport a temperature difference the distance of one domain diameter will be of the order of $1/2\sigma$; with the largest shear rate used here, the foregoing time is about 2×10^{-3} sec. In *p*-azoxyanisole, swarms are thought to contain about 10^5 molecules; their diameter would thus be about 4×10^{-6} cm. By Einstein's relation, the time required for conduction through a domain to transport a temperature difference is of order of magnitude $d^2/2\alpha$, where d is the domain diameter and α is its thermal diffusivity. The axial thermal conductivity of *p*-azoxyanisole is 2.82×10^{-4} cal/sec cm °C as found above; its density is 1.15 gm/cm[19] and its specific heat is 0.53 cal/gm °C.[12] Thus, $\alpha = 4.63 \times 10^{-4}$ cm^2/sec, and the time of transport is about 2×10^{-8} sec. We see that the spin mechanism of transport is too slow to contribute significantly to the transport rate.

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