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Thermal Transport Phenomena in Nematic Liquid Crystals: A Review†

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Abstract—A brief review of the relevant literature available to date on heat transport in nematic liquid crystals is given.

1. Introduction

Within the past decade, research interest in liquid crystals has increased very rapidly due to the increased recognition of the potential applications of liquid crystals, especially the nematic ones. Though one has a better understanding of the structure and properties of these substances, it is essential to recognize the existing conflicts between the various schools of thought and the necessity for further explorations in this field. An excellent bibliography in liquid crystals research is available in Refs 2, 3, 6, 17, 23. The object of this article is to review the important literature available on heat conduction and associated phenomena in nematic liquid crystals. Earlier works related to this discussion are available as cross references in the literature cited.

2. Thermal Conductivity of Nematics

The earliest work on the thermal conductivity of nematic mesophases is by McCoy and Kowalczyk.(26) They have reported data on thermal conductivity at different temperatures over the nematic and isotropic range for three types of liquid crystals as listed in Table 1, which gives the details of measurement technique and some important data. Thermal conductivity decreases with increase in temperature in both nematic and isotropic ranges with a jump at the

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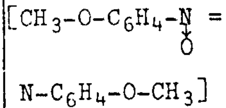
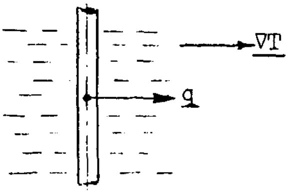
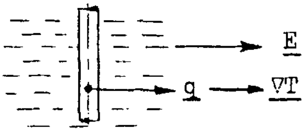
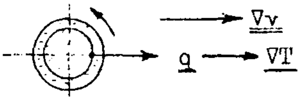
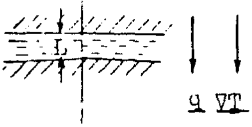
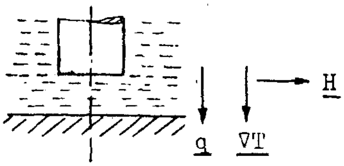
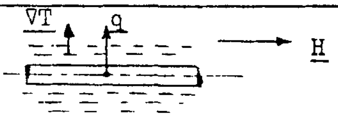
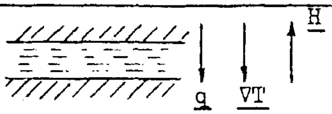
Liquid Crystal (Nematic Range °C)	Temperature (°C)	k x 10^4 cal/cm sec °C	
		k_o	$k_\perp$ or $k_\parallel$
* p-n-hexyloxy-benzoic acid (105-150.5)	124	3.19	-
* p-n-heptyloxy-benzoic acid (91-145)	130	3.50	-
PAA (118-135)  $[CH_3-O-C_6H_4-\underset{O}{\underset{\downarrow}{N}}=N-C_6H_4-O-CH_3]$	125	3.28	-
	125	3.20	5.0 †
	123	3.22	3.41 ($k_\perp$)
	125-6	3.24	3.60 †
	-	-	-
* DBA (124-142) $[n-C_{10}H_{21}-O-C_6H_4-COOH]$	134.9	1.633	1.348 ($k_\perp$)
MBBA (21-42) $[CH_3-O-C_6H_4-CH=N-C_6H_4-(CH_2)_3-CH_3]$	25-35	-	3.04 & 5.0 ($k_\perp$) ($k_\parallel$
APAPA (79.5-102) $[CH_3-O-C_6H_4-CH=N-C_6H_4-O-CO-CH_3]$	80-1	3.33	3.75

* p-n-alkoxybenzoic acids $[R-O-C_6H_4-COOH]$; $R=C_nH_{2n}$

† direction not known

Table 1

Remarks	Measurement Technique	Schematic Diagram	Ref
data over a wide temperature range	steady, non-flow, hot-filament cell with commercial sample		26
orientation unknown. $k=k_o + f(\underline{E})$	transient hot-wire, non-flow cell with & w/o electric field & comm. sample		32
possible convection error. $k_{\perp}>k_{\parallel}$ $k=k_o + f(\underline{\nabla v})$	steady Couette-flow cell (concentric cylinders) purified sample		13
surface orientation unknown $k = f(\frac{1}{L}, \frac{1}{\underline{\nabla T}})$	steady, non-flow, cone & plate cell with purified samples; boundary effects.		29,33
parallel surface orientation assumed $k=f(\underline{H})$ $k_{\perp}>k_{\parallel}$	steady, non-flow cell; comm. sample with magnetic field		23
$k_{\perp}<k_{\parallel}$ $k=k_o-f(\underline{H})$	transient hot-wire, non-flow cell; purified sample with magnetic field		42
$k_{\perp}<k_{\parallel}$ $k=f(\underline{H})$	steady, non-flow parallel plate cell; comm. sample with magnetic field		16 & priv comm
surface conditions unknown	same as in reference (29)	same as in reference (29)	34,35

(n = 1,2....).

nematic–isotropic transition temperature. For *p*-azoxyanisole (PAA), their data indicate a 0.5% decrease in thermal conductivity per °C over the nematic range. In view of the later observations(29,32) on the influence of boundaries, the preceding data have to be considered with some reservation to be discussed in the next section. Picot and Fredrickson(32) reported the influence of a solid interface and electric field on thermal conduction in a nematic mesophase at a fixed temperature. For the first time, they established the effect of a solid–nematic interface on heat conduction by demonstrating the decreasing conductivity from the surface to the bulk fluid. The inhomogeneity in orientation caused by the interface extends to a distance of the order of 20 μ into the bulk. In the absence of any other orienting influence, this was attributed to the orienting influence of the interface.

Recent observations in the literature (4,27,31) seem to indicate the generation of convective flow currents in nematics in the presence of electric fields. Hence the apparent increase in thermal conductivity with the increase in electric field strength reported in Ref. 32 can either be attributed to the increased convective heat transfer or to the orienting effect of electric field. This work is inconclusive on this question. Also, the non-uniform electric field in the cylindrical system and the uncontrolled surface orientation in the experiments cited makes any conclusion on the orienting effect (in the case of externally applied electric field) very uncertain. The technique of transient hot-wire probe for thermal conductivity measurements in an inhomogeneous material needs careful consideration since the solution of the transient conduction equation is not as simple as in the case of homogeneous media. This becomes even more critical in view of the anisotropic thermal transport behaviour(40) of nematics.

Fisher and Fredrickson(13) studied the effect of hydrodynamic shear field on thermal conductivity of nematic PAA. Their conclusion on the nature of thermal conductivity anisotropy (see Table 1 and Eq. (2))—i.e. $k_\perp > k_\parallel$ is questionable because of a possible error in experimental data caused by possible convection effects due to hydrodynamic instability in the annulus of the Couette-flow cell.(33) The cell gap was large enough (0.5 mm) to make the surface influences negligible as compared to the hydrodynamic orienting influence. However there was some possibility of eccentricity between the two cylinders of the Couette-flow apparatus, possibly resulting in secondary convective flows.

Patharkar *et al.*(29) used a cone-and-plate cell of non-flow type which removed most of the major mechanical and other problems. Their results establish the effects of interface and temperature gradient on heat conduction and also affirm the results of Picot and Fredrickson.(32) Rajan *et al.*(34,35) have reported similar observations for APAPA (*p*-anisylidene *p*-aminophenyl-acetate. The influence of temperature gradient on thermal transport phenomena is new and will be discussed subsequently. Yun *et al.*(42) have reported thermal conductivity data on nematic *n*-decyloxy *p*-benzoic acid (DBA) in the presence of magnetic fields, obtained with the hot-wire method. The results are very conclusive—i.e., positive thermal conductivity anisotropy ($k_\parallel > k_\perp$) as compared to the negative anisotropy ($k_\perp > k_\parallel$ for PAA) of Fisher and Fredrickson,(13) as interpreted from their results with a knowledge of the well understood influence of magnetic field on nematic mesophases—i.e. orientation parallel to the static magnetic field. The conductivity was found to be higher near the interface than in the bulk as observed by Picot and Fredrickson.(32)

The most recent work in this area is by Guyon *et al.*(16) They studied the effect of magnetic field on thermal conductivity of nematic *p*-methoxy benzylidene *p*-*n*-butylaniline (MBBA). Their conclusions on the sign of the conductivity anisotropy is the same as that of Yun *et al.*(42) They did not observe any surface influences with controlled surface orientation when the gap in the parallel plate cell was varied up to about 700 μ.

3. Boundary Effects in Liquid Crystals Transport Phenomena

It has been well known that near solid boundaries, the interface dictates some orientation, which is indestructible by external fields and which depends on the nature and prior treatment of the solid surface.(1a,6a) This is not clearly understood from a theoretical viewpoint. It is reported in the liquid crystal literature that clean glass surfaces orient the nematic molecular axis normal to the surface whereas unclean or rubbed (induced surface charges) glass and clean metallic surfaces (silver, gold, platinum) create parallel orientation. The preferred orientation is attributed to the surface couple stress on the liquid crystals.(11) The orientation boundary condition as prescribed by the interface competes with the external field orienting influences to give a resultant orientation pattern.

As mentioned earlier, Picot and Fredrickson(32) experimentally established for the first time, the surface influence on heat conduction in liquid crystals, which has been confirmed by later workers.(14,29,34,42) A qualitative model describing the surface orienting effect is as follows(32):

At the solid surface, the molecules are strongly adsorbed, and oriented with their dipolar axis normal to the surface. This adsorbed layer interacts with the adjacent molecules to create further orientation in the same direction as the adsorbed layer. This effect decreases as the distance from the surface increases due to the competing effect of thermal or rotary Brownian motions that tend to destroy or randomize any ordered arrangement of the domains.(36) Thus, the material has isotropic properties (in the nematic range) because of random orientations at large distances from the surface—i.e., at distances much larger than the interfacial oriented layer thickness. This conclusion is based on Bose and Ornstein's " swarm " hypothesis. According to Zocher's " distortion and continuum " hypothesis, such random orientation in the bulk is not recognized. Neither of these two hypotheses completely explains all of the phenomena occurring in liquid crystals. The preceding qualitative model still does not explain the surface effects for non-polar molecules.

The molecular description of liquid crystals using statistical means as given by Maier and Saupe(2,24) introduces the concept of a distribution of orientations or an average " order parameter " to explain many of the properties of liquid crystals.(5,6,24,41) All these theories for liquid crystal structure are discussed in detail in the literature.(2,3,6) Rajan *et al.*(33,34) used this statistical description of orientation patterns and the idea of random bulk orientations to analyze the results of Patharkar *et al.*(29) and labelled the analysis as a " swarm–continuum " theory. Thermal conductivity data for certain nematic liquid crystals near an interface have been successfully analyzed by this concept using the " anisotropy or deviatoric orientation tensor " **A** (analogous to the order parameter) to describe the orientations.(40)

$$A_{ij}(\mathbf{r}) = \left\langle \cos\theta_i(\mathbf{r})\cos\theta_j(\mathbf{r}) - \frac{\delta_{ij}}{3} \right\rangle \tag{1}$$

where $\boldsymbol{\delta}_i \cos \theta_i = \mathbf{n} =$ " director " or unit vector along the molecular axis.

$\delta_{ij} = \boldsymbol{\delta}_i \cdot \boldsymbol{\delta}_j =$ Kronecker delta

$\boldsymbol{\delta}_i =$ unit vector along the ith axis

$\mathbf{r} =$ position vector

$\langle\ \rangle =$ ensemble average for all orientations as described by a suitable distribution function $f(\theta_i, \theta_j)$

Invariance to rotation by 180° or transverse isotropy of the swarm (molecular aggregate) is assumed in the above tensorial description of orientations. A linear constitutive relation between the heat flux vector and the temperature gradient is found in the literature(21):

$$\begin{aligned} q_i &= \alpha_1 T_{,i} + \alpha_2 n_j T_{,j} n_i \\ &\equiv -k_0(\delta_{ij} + \lambda_1 A_{ij}) T_{,j} \end{aligned} \tag{2}$$

where $k_0 = (k_\parallel + 2k_\perp)/3$

$\lambda_1 =$ conductivity anisotropy

$= (k_\parallel - k_\perp)/k_0$

$k_\parallel$ and $k_\perp$ are the principal conductivities parallel and perpendicular respectively to the major axis of the spheroidal swarm. For steady state, no external field situations, Sullivan(40) derived the following:

$$\tau v_h A_{ij,h} = \delta_c^2 A_{ij,hh} - A_{ij} \tag{3}$$

where $\tau =$ orientation relaxation time

$v_h =$ flow velocity

$A_{ij} = A^\circ_{ij}$ at the boundaries (3a)

Rajan *et al.*(33) define δ_c as an interfacial oriented layer thickness at which distance from the surface A_{ij} is 0.368 of the value of A°_{ij}. Their analysis of the thermal conductivity data using the preceding equations, gives δ_c to be of the order of 30 μ and 140 μ respectively for PAA and APAPA. They also give a critical discussion on the conductivity anisotropy. Yun and Fredrickson(42a) have reported a similar analysis. Thermal conductivity data in the past lack this information.

It is generally believed that long-range order is the main defining feature of a nematic mesophase. Uniform orientation throughout the bulk of the sample is supposed even in the absence of external forces or fields. This conclusion is perhaps drawn because of the fact that most of the optical and other experiments were conducted in samples of small sizes—i.e., sizes that are comparable to δ_c. The real answer to this can come only from experimentation with large size samples. One experiences a great difficulty in the optical experiments with large size samples because of the opacity of the nematics.

As discussed earlier, Guyon *et al.*(16) and Yun *et al.*(42) using thermal conductivity data in magnetic fields, have shown that DBA and MBBA exhibit positive conductivity anisotropy compared to the negative anisotropy of PAA observed by Fisher.(13) Their observations also indicate that the interface orientation is perpendicular to the surface.

The influence of interfaces on the hydrodynamics of nematic liquid crystals has been clearly demonstrated by Fisher and Fredrickson.(14) Viscosity of PAA as observed in a Poiseuille flow capillary viscometer seems to be a strong function of the size of the capillary. Two major conclusions may be drawn from this work.

(a) nematic mesophases exhibit non-Newtonian shear thinning behaviour as expected for the hydrodynamics of rod like molecules in flow.

(b) the interface orientation is affected by the hydrodynamic shear and tends toward parallel orientation in the direction of flow at the surfaces.

They were able to correlate their results in an empirical manner using Ericksen's(12) continuum theory with some success. A theoretical analysis of the boundary effects on viscometric problems using the theory of elasticity for nematic mesophases has been recently given by Lee and Eringen.(18) A basic assumption underlying their analysis is that a nematic liquid crystal (without any external effects) has an axis of orientation with non-random degree of order which is different from the random or isotropic order suggested by Fisher.(14) The question of random order in the nematic state as distinguished from random isotropic melt state is yet unsolved though some experimental evidence of random nematic state can be concluded from recent literature.(28a)

4. Temperature Gradient Effects in Nematic Liquid Crystals

In the bulk of liquid crystals away from interfaces the orientation pattern is influenced by various fields,(11) shear, electromagnetic, temperature gradient and small temperature changes. The effect of temperature gradient was first observed by Lehmann(19) in 1900. He observed violent rotations in liquid crystal droplets in the presence of a temperature gradient. Anzelius(1) attempted to analyze this effect of temperature gradient competing with the boundary effects. He did not use the now known continuum principles. Oseen(28) criticized the static theory of Anzelius basically for this reason and put forward a dynamic explanation of this phenomenon predicting a non-steady solution for the orientation. Leslie(21) used the principles of continuum theory to explain the rotation phenomenon in cholesteric liquid crystals (special case of twisted Nematics). He called this the thermomechanical coupling effect and derived the conditions required for static and dynamic solutions for the orientation director **n**. If the boundary effects are dominant, **n** can have a static solution. Otherwise, **n** varies periodically with time with the bulk fluid translational velocity being zero. The possibility of convective cell generation due to Bénard instability(10) in a temperature gradient was not considered in his analysis. The corresponding continuum theory for nematic liquid crystals does not explicitly incorporate thermal gradients as an orienting agent, since in the first approximation mechanical and thermal effects are uncoupled. However the theory could be amended to incorporate thermal effects in nematics—i.e., by introducing into the constitutive equation for the intrinsic orientation, body force terms which are non-linear in the temperature gradient.(22)

The first systematic experimental observation of temperature gradient effect was made by Stewart(37) in his X-ray diffraction studies on nematic PAA. He concluded that a small temperature gradient of 1 or 2 °C/cm, produces a preferred orientation of the liquid crystal " swarms " or molecular aggregates ($10^5 - 10^6$ in number with axes parallel to one another) with their long axes perpendicular to the direction of maximum temperature gradient. The transverse orientation of the nematic mesophases in a heat flow(38,39) could not be explained by the differential thermal expansion, the momentum or radiation pressure of high frequency acoustic waves, the anisotropy of heat conduction and the reorientations caused by unavoidable convection flow currents generated due to instability in a temperature gradient field.(25) Such orientations in heat flow were not observed in solid–liquid suspensions. It is doubtful whether the velocity gradient caused by the radiation pressure of the acoustic waves (transverse and longitudinal) would be sufficient to produce enough torque for reorienting the nematic mesophase.

Picot and Fredrickson(32) and Yun *et al.*(42) did not observe any such effects of heat conduction for a two fold variation in the impressed heat flux. They rejected Stewart's observations as being due to natural convection flow effects. Fisher and Fredrickson(13) also did not observe any temperature gradient effects in this flow-cell for conductivity measurements in PAA up to 36 °C/cm.

A formal explanation of the observation of Stewart *et al.*(38) using the continuum theory is given by Davison.(7,8) He obtained a linear theory for heat conduction by assuming that the director **n** varies only infinitesimally from a fixed direction and by applying the theorems of linear elastostatics.

$$\mathbf{n} = \mathbf{n}_0 + (\boldsymbol{\Omega} \times \mathbf{n}_0) \tag{4}$$

and

$$\boldsymbol{\Omega} \times \mathbf{n}_0 = 0$$

where $\boldsymbol{\Omega}$ is a dimensionless pseudovector such that $\mathrm{d}\boldsymbol{\Omega}/\mathrm{d}t$ is the spin vector of the director $\mathbf{n}_0$. By considering the Helmholtz Free energy in the system and by suitable energy balances, he derived the relation for the dissipation or entropy production inequality different from the Clausius–Duhem inequality where the entropy flux is just the heat flux divided by temperature.

By a suitable choice of constitutive relation for the various fluxes and fields as in Leslie-Ericksen theory, he concludes the following:

If the orientation and temperature boundary conditions are imposed, static orientations do not effect the temperature distribution. But, the presence of a temperature field $T(\mathbf{r},t)$ affects the reorientation field by introducing an intrinsic body moment on the director. If boundaries prescribe this moment also, the coupling effect gets very complicated. Consider the planar orientation

$$\theta = \theta(r,z) \qquad \text{(orientation angle)}$$

and

$$\boldsymbol{\Omega} = (0, \boldsymbol{\Omega}(r,z), 0) \tag{5}$$

Davison's field equations for a symmetric, isotropic system give

$$\Omega_{,rr}+\Omega_{zz,}=MT_{,rz}$$

and

$$T_{,rr}+T_{,zz}=0 \tag{6}$$

where M = body moment elastic constant.

Equation (6) can be solved easily if it is assumed that the boundaries fix the reference orientation $\mathbf{n}_0$ and that the reorientation spin $\boldsymbol{\Omega}$ vanishes at the boundaries and where the temperature gradient is zero.

Davison's equations become highly complicated in other general type of orientations and also where the thermal conductivity anisotropy is non-zero. As mentioned earlier, Patharkar *et al.*⁽²⁹⁾ demonstrated the influence of thermal gradients on heat conduction in nematic liquid crystals. A strong coupling between the boundary and temperature gradient effect is also indicated by their results. Any error due to hydrodynamic instabilities caused by natural convection was prevented making the temperature gradient along the direction of gravity in the conductivity cell. All other possible sources of error such as radiation, end conduction etc. were corrected for in their results. Thermal conductivity decreases at all gaps with the increase in temperature gradient (up to 200 °C/cm). Recently Dubois-Violette⁽⁹⁾ used the continuum and elasticity theories⁽¹⁵⁾ to analyze the hydrodynamic instability in a thermal field for a nematic liquid crystal. She relates the hydrodynamic instability in a parallel plate cell to the direction of ∇T in relation to Earth's gravity and the anisotropy of thermal conductivity. She states the conditions for the onset of instability (analogous to Bénard type convection cells in isotropic systems). For ∇T against gravity, systems with negative conductivity anisotropy ($\lambda_1 < 0$) are stable and for ∇T along gravity, systems with positive anisotropy ($\lambda_1 > 0$) are stable. For PAA⁽²⁹⁾ ($\lambda_1 < 0$) with negative anisotropy one should expect instability if ∇T is in the direction of gravity which was not observed by Patharkar⁽³⁰⁾ in his optical studies in thermal fields. Though Dubois-Violette's analysis explains the rotations observed by Lehmann, the decrease in thermal conductivity with increase in ∇T cannot be explained as one should expect increased heat transfer between the plates when natural convection cells exist. Partharkar⁽³⁰⁾

points out the existence of a critical temperature gradient (Rayleigh number) in each gap at which natural convection starts thereby increasing the effective heat transfer between the cell plates. It is to be noted that any theoretical statement of critical temperature gradient must recognize the temperature limits for the mesophase as well as the effect of temperature on the order parameter.⁽⁵⁾

5. Concluding Remarks

In this review article, we have presented a detailed discussion on the interfacial and temperature gradient effects in the heat transport in liquid crystals. It is clear from this discussion that more experimental information is needed in various liquid crystals to get a clear understanding of the above mentioned phenomena. There seems to be a vast amount of theoretical analyses available, but it goes without saying that the fundamental questions on the bulk orientations in the natural state of the liquid crystals (random or partially ordered) and the mechanism of surface orientations must be answered beyond any doubt in order to apply any of the available theories. Again, this seems possible only if experiments are performed in liquid crystal samples free of any boundary effects—i.e., systems with large volume to surface ratio. Systematic optical and X-ray studies on these appear to be the only prospective resource to achieve this goal.

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