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Heat-Flow Birefringence in Gases

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Dedicated to Prof. Dr. L. Waldmann on the occasion of his 60th birthday

Heat-flow birefringence is described by a constitutive law which links the anisotropic part of the dielectric tensor with the gradient of the heat flux or the 2nd spatial derivative of the temperature field. For a gas of linear molecules, this relation is derived from transport-relaxation equations which, in turn, were obtained from the Waldmann-Snider equation. The magnitude of the heat-flow birefringence can be inferred from the Senftleben-Beenakker effect of the viscosity and of the heat conductivity. It is found to be of measurable size.

Transport processes in a fluid of optically anisotropic particles may lead to a birefringence, i. e. its dielectric tensor may possess a nonvanishing anisotropic part. Flow birefringence[1, 2] and acoustic birefringence[2, 3] are examples for such nonequilibrium birefringence phenomena. Both effects are described by a constitutive law which links the anisotropic part of the dielectric tensor with the gradient of the velocity. In a heat-conducting fluid the anisotropic part of the dielectric tensor can be proportional to the gradient of the heat flux or equivalently to the 2-nd spatial derivative of the temperature. A kinetic theory of this "heat-flow birefringence" is presented for gases of rotating molecules.

In contradistinction to liquids and colloidal solutions where birefringence is due to an alignment of the figure axis of the particles, in gases of rotating molecules it is connected with an alignment of the molecular rotational angular momentum[4]. This is reflected by the fact that the anisotropic part of the dielectric tensor is proportional to the tensor polarization of the rotational angular momentum. For the theoretical treatment of nonequilibrium birefringence phenomena the collision-induced tensor polarization has to be studied. Point of departure for such a theory is the Waldmann-Snider equation[5, 6], a generalized Boltzmann equation for particles with internal rotational degrees of freedom. Waldmann[5] mentioned flow birefringence as a possible application of his kinetic equation as early as 1957. The kinetic theory of flow birefringence, however, has been developed much later[7, 8]. This paper deals with the theory of the heat-flow birefringence. Its magnitude can be inferred from measurements of the influence of magnetic fields on the heat conductivity and the viscosity[9, 10] (Senftleben-Beenakker effect). It turns out to be of similar size as the flow birefringence in gases which has recently been observed experimentally for the first time[11]. Most of the results presented in this paper have been reported at the 7-th Symposium on Rarefied Gas Dynamics[12] (Pisa 1970).

This paper is divided into five major sections. Section 1 deals with the phenomenological description of heat-flow birefringence. A constitutive relation is introduced which links the anisotropic part of the dielectric tensor with the gradient of the heat flux or with the 2-nd spatial derivative of the temperature. Then it is indicated how the heat-flow birefringence can be measured in principle. For gases of linear molecules, the relation between the anisotropic part of the dielectric tensor and the tensor polarization of the molecular rotational angular momentum is stated in Section 2. The transport-relaxation equations for the macroscopic variables relevant for the present problem are presented in Section 3. These equations have been derived from the Waldmann-Snider equation by application of the moment method. They involve 13 relaxation coefficients, 7 of which are linearly independent. These coefficients can be expressed in terms of collision integrals obtained from the linearized Waldmann-Snider collision operator. In Section 4 it is indicated how the magnitudes of these relaxation coefficients can be inferred from measurements of the heat conductivity, of the shear and the bulk viscosities, and of the Senftleben-Beenakker effect of the heat conductivity and of the viscosity. Finally, in Section 5, the constitutive relation for heat-flow birefringence is derived from the transport-relaxation equations. The heat-flow birefringence coefficient is expressed in terms of relaxation coefficients or collision integrals which involve the scattering amplitude operator characterizing the binary collision processes. The order of magnitude of the heat-flow birefringence is estimated. In the Appendix a qualitative theory of the density dependence of the flow and the heat-flow birefringence is presented for low densities where wall collisions have to be taken into consideration.

§ 1. Phenomenological Description

The dielectric tensor $\boldsymbol{\epsilon}$ which determines the optical properties of a medium can be split into its isotropic and anisotropic parts:

$$\boldsymbol{\epsilon} = \varepsilon\,\boldsymbol{\delta} + \overline{\boldsymbol{\epsilon}}\,. \tag{1.1}$$

Here $\boldsymbol{\delta}$ is the (2-nd rank) unit tensor and ε is the isotropic dielectric coefficient. The symbol $\overline{\ldots}$ refers to the symmetric traceless part of a tensor, e. g. for two vectors $\boldsymbol{a}, \boldsymbol{b}$ one has

$$\overline{\boldsymbol{a}\,\boldsymbol{b}} = \tfrac{1}{2}\,(\boldsymbol{a}\,\boldsymbol{b} + \boldsymbol{b}\,\boldsymbol{a}) - \tfrac{1}{3}\,\boldsymbol{a}\cdot\boldsymbol{b}\,\boldsymbol{\delta}\,. \tag{1.2}$$

To study the effect of $\overline{\boldsymbol{\epsilon}} \neq 0$ on the index of refraction it is sufficient to consider the special case of light propagating parallel to one of the principal axes of the dielectric tensor. Let $\boldsymbol{e}_i$, $i = 1, 2, 3$ be unit vectors parallel to the principal axes. The index of refraction for linearly polarized light with the electric field vector parallel to $\boldsymbol{e}_i$ is given by

$$\nu_i = (\boldsymbol{e}_i\cdot\boldsymbol{\epsilon}\cdot\boldsymbol{e}_i)^{1/2}\,.$$

If the anisotropy of $\boldsymbol{\epsilon}$ is small, ν_i is approximately given by

$$\nu_i = \nu\,(1 + (1/2\,\varepsilon)\;\boldsymbol{e}_i\cdot\overline{\boldsymbol{\epsilon}}\cdot\boldsymbol{e}_i) \tag{1.3}$$

with $\nu = \sqrt{\varepsilon}$. The difference between the principal indices of refraction is a measure for the birefringence. Clearly for $\overline{\boldsymbol{\epsilon}} = 0$ one has $\nu_i = \nu$ for $i = 1, 2, 3$ and there is no birefringence.

In the absence of external fields and if no transport processes are going on, $\overline{\boldsymbol{\epsilon}} = 0$ holds true for fluids. In the presence of a flow velocity field $\boldsymbol{v}(\boldsymbol{x})$ flow birefringence may occur which is described by the constitutive law

$$\overline{\boldsymbol{\epsilon}} = -2\,\beta\,\overline{\nabla\boldsymbol{v}}\,, \tag{1.4}$$

where β is a phenomenological coefficient. Likewise, in the presence of a heat flux $\boldsymbol{q} = -\lambda\,\nabla T$ where λ is the heat conductivity and T is the temperature of the fluid, the ansatz

$$\boldsymbol{\epsilon} = -2\,\beta_\lambda\,\overline{\nabla\boldsymbol{q}} = 2\,\lambda\,\beta_\lambda\,\overline{\nabla\nabla}\,T \tag{1.5}$$

can be made for heat-flow birefringence. Here β_λ is another phenomenological coefficient characteristic for flow-birefringence. In Ref. [12] the coefficient $\gamma = -\lambda\,\beta_\lambda$ was used instead of β_λ. The coefficients β and β_λ may be positive or negative.

It is the task of the kinetic theory to justify the constitutive laws and to relate the coefficients occurring in them to molecular properties and, if possible, to other nonequilibrium phenomena. Before these points are treated for heat-flow birefringence in gases of rotating molecules it seems appropriate to discuss briefly how β_λ can be measured.

Consider stationary heat conduction through a fluid between two concentric cylinders with temperatures T_1, T_2 and radii R_1, R_2; $R_1 < R_2$. The mean free path l in the fluid is assumed to be very small compared with R_1. The temperature of the fluid at a point $\boldsymbol{x}$ is

$$T(\boldsymbol{x}) = T_1 + \frac{T_2 - T_1}{\ln(R_2/R_1)} \ln(r/R_1), \; R_1 \leqq r \leqq R_2, \tag{1.6}$$

with $\boldsymbol{r} = \boldsymbol{x} - \boldsymbol{e}_3 \, \boldsymbol{e}_3 \cdot \boldsymbol{x}$, $r = |\boldsymbol{r}|$. Here $\boldsymbol{x} = 0$ is a point on the axis of the cylinders and $\boldsymbol{e}_3$ is a unit vector parallel to it. From (1.6)

$$\overline{\nabla\nabla} T = (T_1 - T_2) \cdot (\ln(R_2/R_1))^{-1} \, (2 \, r^{-4} \, \overline{\boldsymbol{r}\boldsymbol{r}} - r^{-2} \, \overline{\boldsymbol{e}_3 \boldsymbol{e}_3}) \tag{1.7}$$

is obtained. Notice that $\boldsymbol{e}_3 \cdot \overline{\nabla\nabla} \cdot \boldsymbol{e}_3 \, T = 0$.

According to (1.5) and (1.7) the principal axes of the dielectric tensor are parallel to $\boldsymbol{e}_3$, $\boldsymbol{r}$, and $\boldsymbol{e}_3 \times \boldsymbol{r}$. Consider a thin beam of linearly polarized light which propagates parallel to the axis of the cylinders and intersects a plane perpendicular to it at the point $\boldsymbol{r}$. The difference between the indices of refraction $\nu_1 = \nu_{\parallel}$ and $\nu_2 = \nu_{\perp}$ for electric field vectors parallel and perpendicular to $\boldsymbol{r}$ is, due to (1.3), (1.5), (1.7), given by

$$\nu \, \delta\nu = \nu(\nu_{\parallel} - \nu_{\perp}) = 2 \, \beta_\lambda \, r^{-2} \, \lambda (T_1 - T_2) \, (\ln(R_2/R_1))^{-1}. \tag{1.8}$$

For gases $\nu = \sqrt{\varepsilon}$ can be approximated by 1. Clearly, the measurement of $\delta\nu$ allows the determination of β_λ.

§ 2. Dielectric Tensor and Tensor Polarization

Before the kinetic theory for heat-flow birefringence can be developed the anisotropic part $\overline{\boldsymbol{\epsilon}}$ of the dielectric tensor has to be expressed by an average of a molecular quantity. For gases, the dielectric tensor $\boldsymbol{\epsilon}$ is related to the molecular polarizability tensor $\boldsymbol{\alpha}$ by

$$\boldsymbol{\epsilon} = \boldsymbol{\delta} + 4 \pi n \langle \boldsymbol{\alpha} \rangle \tag{2.1}$$

where n is the number density of the gas and the bracket $\langle \ldots \rangle$ refers to an average evaluated with a nonequilibrium distribution operator. For gases of linear molecules the part of $\boldsymbol{\epsilon}$ which is relevant for birefringence can be written as [4, 7]

$$\overline{\boldsymbol{\epsilon}} = \varepsilon' \, \mathbf{a} \tag{2.2}$$

with

$$\mathbf{a} = \langle \boldsymbol{\Phi}_T \rangle , \tag{2.3}$$

$$\boldsymbol{\Phi}_T = \sqrt{15/2} \, (\langle J^2/(J^2 - \tfrac{3}{4}) \rangle_0)^{-1/2} \, (J^2 - \tfrac{3}{4})^{-1} \, \overline{\boldsymbol{J}\boldsymbol{J}} \approx \sqrt{15/2} \, J^{-2} \, \overline{\boldsymbol{J}\boldsymbol{J}} , \tag{2.4}$$

$$\varepsilon' = -2 \pi n (\alpha_{\parallel} - \alpha_{\perp}) \sqrt{2/15} \, (\langle J^2/(J^2 - \tfrac{3}{4}) \rangle_0)^{1/2}. \tag{2.5}$$

The quantity $\mathbf{a}$ is referred to as tensor polarization. The rotational angular momentum operator of a molecule is denoted by $\hbar \boldsymbol{J}$. The coefficients $\alpha_{\parallel}$ and $\alpha_{\perp}$ occurring in Eq. (2.5) are the polarizabilities of a molecule for the electric field vector parallel and perpendicular to the molecular axis. The average of a molecular quantity $\Psi = \Psi(\boldsymbol{V}, \boldsymbol{J})$ is given by

$$n \langle \Psi \rangle = \int \mathrm{d}^3 V \, \mathrm{Tr} \, \Psi f \tag{2.6}$$

with $n = \int \mathrm{d}^3 V \, \mathrm{Tr} \, f$. Here $f = f(t, \boldsymbol{x}, \boldsymbol{V}, \boldsymbol{J})$ is the nonequilibrium distribution operator of the gas, $\boldsymbol{V}$ is the velocity of a particle with mass m in units of $(2 k T/m)^{1/2}$, and "Tr" denotes the trace over the magnetic substates and a summation over the rotational states. Equilibrium averages evaluated with the equilibrium distribution f_0 instead of f are denoted by $\langle \ldots \rangle_0$.

According to Eq. (2.2), the (collision-induced) tensor polarization $\mathbf{a}$ has to be calculated for the determination of the birefringence caused by a transport process.

§ 3. Transport-Relaxation-Equations

The nonequilibrium distribution operator which is eventually needed for the evaluation of nonequilibrium averages obeys the Waldmann-Snider equation [5, 6]. From this generalized Boltzmann equation an infinite set of transport-relaxation equations for the moments of the distribution operator can be derived by use of the moment method [13]. For most physical applications it is sufficient to consider a finite subset of these moment equations. The moments which are taken into consideration are the macroscopic variables needed to specify the nonequilibrium state of the gas. In addition to the tensor polarization $\mathbf{a}$, the following macroscopic variables are taken into account for the treatment of the heat-flow birefringence: the translational and internal temperatures T_{t} and T_{i}, the corresponding heat fluxes $\boldsymbol{q}_{\mathrm{t}}$ and $\boldsymbol{q}_{\mathrm{i}}$, the friction pressure tensor $\mathbf{p} = n m \langle \overline{\boldsymbol{c}\boldsymbol{c}} \rangle$ where $\boldsymbol{c} = (2 k T/m)^{1/2} \boldsymbol{V}$ is the particle velocity, and the "Kagan vector"

$$\mathbf{b} = \langle \boldsymbol{c} \cdot \boldsymbol{\Phi}_T \rangle , \tag{3.1}$$

which is the vector part of the tensor polarization flux. The other parts of this tensor can be ignored for the present problem. Furthermore, a term involving the flow velocity $\boldsymbol{v}$ is retained in the equation for $\mathbf{p}$. Though this term is not needed for heat-flow birefringence it facilitates the comparison with flow birefringence.

The relevant transport-relaxation equations are

$$\boldsymbol{q}_{\mathrm{t}} + (5\,k/2\,m)\,p_0\,(\nabla T_{\mathrm{t}} + (2\,T_0/5\,p_0)\,\nabla\cdot\mathbf{p}) + \omega_{\mathrm{t}}\,\boldsymbol{q}_{\mathrm{t}} + r\,\omega_{\mathrm{ti}}\,\boldsymbol{q}_{\mathrm{i}} + \sqrt{\tfrac{3}{2}}\,p_0\,\omega_{\mathrm{tK}}\,\mathbf{b} = 0\,, \tag{3.2}$$

$$\dot{\boldsymbol{q}}_{\mathrm{i}} + (c_{\mathrm{i}}/m)\,p_0\,\nabla T_{\mathrm{i}} + r^{-1}\,\omega_{\mathrm{it}}\,\boldsymbol{q}_{\mathrm{t}} + \omega_{\mathrm{i}}\,\boldsymbol{q}_{\mathrm{i}} + \sqrt{\tfrac{3}{2}}\,r^{-1}\,p_0\,\omega_{\mathrm{iK}}\,\mathbf{b} = 0\,, \tag{3.3}$$

$$\dot{\mathbf{b}} + (k\,T_0/m)\,\nabla\cdot\mathbf{a} + \sqrt{\tfrac{2}{3}}\,p_0{}^{-1}\,\omega_{\mathrm{Kt}}\,\boldsymbol{q}_{\mathrm{t}} + \sqrt{\tfrac{2}{3}}\,r\,p_0{}^{-1}\,\omega_{\mathrm{Ki}}\,\boldsymbol{q}_{\mathrm{i}} + \omega_{\mathrm{K}}\,\mathbf{b} = 0\,, \tag{3.4}$$

$$\dot{\mathbf{p}} + 2\,p_0\,(\overline{\nabla\boldsymbol{v}} + (2/5)\,p_0{}^{-1}\overline{\nabla\boldsymbol{q}_{\mathrm{t}}}) + \omega_\eta\,\mathbf{p} + \sqrt{2}\,p_0\,\omega_{\eta\mathrm{T}}\,\mathbf{a} = 0\,, \tag{3.5}$$

$$\dot{\mathbf{a}} + (3/5)\,\overline{\nabla\mathbf{b}} + (1/\sqrt{2})\,p_0{}^{-1}\,\omega_{\mathrm{T}\eta}\,\mathbf{p} + \omega_{\mathrm{T}}\,\mathbf{a} = 0\,. \tag{3.6}$$

Here $p_0 = n\,k\,T_0$ is the equilibrium pressure of a gas with the equilibrium temperature T_0. The quantity r is an abbreviation for $(5\,k/2\,c_{\mathrm{i}})^{1/2}$ where c_{i} is the heat capacity per particle associated with its rotational motion. The relaxation coefficients $\omega\,..$ can be expressed in terms of collision integrals obtained from the Waldmann-Snider equation. More precisely, with

$$\boldsymbol{\Phi}_{\mathrm{t}} = (2/\sqrt{5})\,(V^2 - 5/2)\,\mathbf{V}, \qquad \boldsymbol{\Phi}_{\mathrm{i}} = \sqrt{2\,k/c_{\mathrm{i}}}\,(\varepsilon_{\mathrm{i}} - \langle\varepsilon_{\mathrm{i}}\rangle_0)\,\mathbf{V}, \tag{3.7}$$

$$\boldsymbol{\Phi}_{\mathrm{K}} = \sqrt{6/5}\,\mathbf{V}\cdot\mathbf{\Phi}_{\mathrm{T}}\,, \ \mathbf{\Phi}_\eta = \sqrt{2}\,\overline{\mathbf{V}\,\mathbf{V}} \tag{3.8}$$

where ε_{i} is the internal rotational energy of a molecule in units of $k\,T_0$ and with $\mathbf{\Phi}_{\mathrm{T}}$ given by (2.4), the relaxation coefficients are defined by

$$\omega_{nn'} = \tfrac{1}{3}\,\langle\boldsymbol{\Phi}_n\cdot\omega(\boldsymbol{\Phi}_{n'})\rangle_0 \quad \text{for} \quad n, n' = \mathrm{t}, \mathrm{i}, \mathrm{K}\,, \tag{3.9}$$

$$\omega_{nn'} = \tfrac{1}{5}\,\langle\mathbf{\Phi}_n : \omega(\mathbf{\Phi}_{n'})\rangle_0 \quad \text{for} \quad n, n' = \eta, \mathrm{T}\,, \tag{3.10}$$

and $\omega_{nn} = \omega_n$. Here $\omega(\ldots)$ is the linearized Waldmann-Snider collision operator. Notice that the normalization factors occurring in the definition for the $\boldsymbol{\Phi}_n$, $\mathbf{\Phi}_n$ have been chosen such that

$$\langle\boldsymbol{\Phi}_n\cdot\boldsymbol{\Phi}_n\rangle_0 = 3 \quad \text{for} \quad n = \mathrm{t}, \mathrm{i}, \mathrm{K} \qquad \text{and} \qquad \langle\mathbf{\Phi}_n : \mathbf{\Phi}_n\rangle_0 = 5 \quad \text{for} \quad n = \eta, \mathrm{T}\,.$$

From time reversal invariance of the intermolecular interaction potential follow the symmetry relations

$$\omega_{\mathrm{ti}} = \omega_{\mathrm{it}}\,, \qquad \omega_{\mathrm{tK}} = \omega_{\mathrm{Kt}}\,, \qquad \omega_{\mathrm{iK}} = \omega_{\mathrm{Ki}}\,, \qquad \omega_{\eta\mathrm{T}} = \omega_{\mathrm{T}\eta}\,, \tag{3.11}$$

From the explicit form of the collision integrals the useful relations

$$\omega_{\mathrm{t}} = (2/3)\,\omega_\eta - (5/3)\,r^{-1}\,\omega_{\mathrm{ti}}\,, \qquad \omega_{\eta\mathrm{T}} = \sqrt{3}\,\omega_{\mathrm{tK}} \tag{3.12}$$

can be inferred. Thus of the 13 relaxation coefficients occurring in Eqs. (3.2) – (3.6) 7 only are linearly independent. Effective collision cross sections $\sigma\,..$ can be defined by

$$\omega\,.. = n\,v_{\mathrm{th}}\,\sigma\,.. \tag{3.13}$$

where n is the number density and $v_{\mathrm{th}} = (8\,kT_0/\pi m_{\mathrm{red}})^{1/2}$ is a thermal velocity, $m_{\mathrm{red}} = \frac{1}{2}\,m$ is the reduced mass of a pair of molecules. The diagonal relaxation coefficients and effective cross sections are positive, the nondiagonal ones may have both signs. To compare with the effective cross sections $\sigma(\ldots)$ used by McCourt and Moraal [14] and by the Leiden group [15, 16], it is noticed that

$$\sigma_{\mathrm{t}} = \sigma(1010)\,, \quad \sigma_{\mathrm{i}} = \sigma(1001)\,, \tag{3.14}$$

$$\sigma_{\mathrm{it}} = -\sigma\binom{1010}{1001}\,, \quad \sigma_\eta = \sigma(2000)\,. \tag{3.15}$$

If $\mathbf{\Phi}_{\mathrm{T}}$ [cf. (2.4)] is replaced by

$$\mathbf{\Phi}_{02} = \sqrt{15/2}\,\langle J^2(J^2 - 3/4)\rangle_0{}^{-1/2}\,\overline{\boldsymbol{J}\,\boldsymbol{J}} \quad \text{one has}$$

$$\sigma_{\mathrm{T}} = \sigma(0200)\,, \qquad \sigma_{\eta\mathrm{T}} = \sigma\binom{2000}{0200}\,, \tag{3.16}$$

$$\sigma_{\mathrm{K}} = \sigma(1200)\,, \quad \sigma_{\mathrm{tK}} = -\sqrt{\tfrac{5}{3}}\,\sigma\binom{1010}{1200}\,, \quad \sigma_{\mathrm{iK}} = \sqrt{\tfrac{5}{3}}\,\sigma\binom{1001}{1200}\,. \tag{3.17}$$

For further details on relaxation coefficients and effective cross sections see References [13–17].

Before the heat-flow birefringence is treated it seems appropriate to consider the expressions for the viscosity and the heat conductivity which can be derived from Eqs. (3.2) – (3.6) and to discuss how the relaxation coefficients or the effective cross sections involved can be determined from experimental data.

§ 4. Viscosity, Heat Conductivity

For a discussion of stationary transport phenomena in the hydrodynamical regime the "normal solutions" of the transport-relaxation Eqs. (3.2) to (3.6) have to be studied. The normal solutions are obtained if all time derivatives and the spatial derivatives of nonconserved quantities are put equal to zero. Furthermore, just one nonequilibrium temperature $T_\mathrm{t} = T_\mathrm{i} = T$ is retained. Then Eqs. (3.5), (3.6) yield

$$\mathbf{p} = -2\,\eta\,\overline{\nabla \boldsymbol{v}}\,. \tag{4.1}$$

The viscosity η is given by

$$\eta = \eta_\mathrm{iso}\,(1 + A_{\eta\mathrm{T}})\,, \tag{4.2}$$

with

$$A_{nn'} = \frac{\omega_{nn'}\,\omega_{n'n}}{\omega_n\,\omega_{n'}} = \frac{\sigma_{nn'}\,\sigma_{n'n}}{\sigma_n\,\sigma_{n'}}\,. \tag{4.3}$$

If no alignment would be set up in a viscous flow $(\omega_{\eta\mathrm{T}} = \omega_{\mathrm{T}\eta} = 0)$ η would be equal to the isotropic viscosity

$$\eta_\mathrm{iso} = p_0/\omega_\eta\,. \tag{4.4}$$

The quantity $A_{\eta\mathrm{T}}$ is typically of the order 10^{-2} or even smaller. Terms nonlinear in $A_{\eta\mathrm{T}}$ have been disregarded in Equation (4.2). Likewise, Eqs. (3.2), (3.3), and (3.4) yield

$$\boldsymbol{q}_\mathrm{t} = -\lambda_\mathrm{t}\,\nabla T, \qquad \boldsymbol{q}_\mathrm{i} = -\lambda_\mathrm{i}\,\nabla T, \tag{4.5}$$

with the translational and internal heat conductivities

$$\lambda_\mathrm{t} = \lambda_\mathrm{t}^\mathrm{iso}\,(1 + B_\mathrm{tK})\,, \qquad \lambda_\mathrm{i} = \lambda_\mathrm{i}^\mathrm{iso}\,(1 + B_\mathrm{iK})\,. \tag{4.6}$$

The total heat conductivity is

$$\lambda = \lambda_\mathrm{t} + \lambda_\mathrm{i} = \lambda_\mathrm{iso}\,(1 + B_{\lambda\mathrm{K}})\,. \tag{4.7}$$

The isotropic values of these transport coefficients are

$$\lambda_\mathrm{t}^\mathrm{iso} = \frac{5\,k}{2\,m}\,\frac{p_0}{\omega_\mathrm{t}}\left(1 - r^{-1}\,\frac{\omega_\mathrm{ti}}{\omega_\mathrm{i}}\right)(1 - A_\mathrm{it})^{-1}, \tag{4.8}$$

$$\lambda_\mathrm{i}^\mathrm{iso} = \frac{c_\mathrm{i}}{m}\,\frac{p_0}{\omega_\mathrm{i}}\left(1 - r\,\frac{\omega_\mathrm{ti}}{\omega_\mathrm{t}}\right)(1 - A_\mathrm{it})^{-1}, \tag{4.9}$$

$$\lambda_\mathrm{iso} = \frac{5\,k}{2\,m}\,\frac{p_0}{\omega_\mathrm{t}}\left(1 - 2\,r^{-1}\,\frac{\omega_\mathrm{ti}}{\omega_\mathrm{i}} + r^{-2}\,\frac{\omega_\mathrm{t}}{\omega_\mathrm{i}}\right)(1 - A_\mathrm{it})^{-1}. \tag{4.10}$$

If terms of higher than 2-nd power in ω_tK, ω_iK are disregarded, the quantities B_tK, B_iK and $B_{\lambda\mathrm{K}}$ are given by

$$B_\mathrm{tK} = \frac{\tilde{\omega}_\mathrm{tK}\,\omega_{\lambda\mathrm{K}}}{\omega_\mathrm{t}\,\omega_\mathrm{K}}\,(1 - A_\mathrm{it})^{-1}, \qquad B_\mathrm{iK} = \frac{\tilde{\omega}_\mathrm{iK}\,\omega_{\lambda\mathrm{K}}}{\omega_\mathrm{i}\,\omega_\mathrm{K}}\,r^{-1}\,\frac{\lambda_\mathrm{t}^\mathrm{iso}}{\lambda_\mathrm{i}^\mathrm{iso}}\,(1 - A_\mathrm{it})^{-1}, \tag{4.11, 12}$$

$$B_{\lambda\mathrm{K}} = \frac{\lambda_\mathrm{t}^\mathrm{iso}}{\lambda_\mathrm{iso}}\,\frac{\omega_{\lambda k}^2}{\omega_\mathrm{t}\,\omega_\mathrm{K}}\left(1 - r^{-1}\,\frac{\omega_\mathrm{ti}}{\omega_\mathrm{i}}\right)(1 - A_\mathrm{it})^{-1} = \frac{5\,k}{2\,m}\,\frac{p_0}{\lambda_\mathrm{iso}}\,\frac{1}{\omega_\mathrm{K}}\left[\frac{\omega_{\lambda\mathrm{K}}}{\omega_\mathrm{t}}\left(1 - r^{-1}\,\frac{\omega_\mathrm{it}}{\omega_\mathrm{i}}\right)(1 - A_\mathrm{it})^{-1}\right]^2, \tag{4.13}$$

with

$$\tilde{\omega}_\mathrm{tK} = \omega_\mathrm{tK} - (\omega_\mathrm{it}/\omega_\mathrm{i})\,\omega_\mathrm{iK}\,, \qquad \tilde{\omega}_\mathrm{iK} = \omega_\mathrm{iK} - (\omega_\mathrm{it}/\omega_\mathrm{t})\,\omega_\mathrm{tK}\,, \tag{4.14}$$

$$\omega_{\lambda\mathrm{K}} = \omega_\mathrm{tK} + r\,(\lambda_\mathrm{i}^\mathrm{iso}/\lambda_\mathrm{t}^\mathrm{iso})\,\omega_\mathrm{iK} = \omega_\mathrm{tK} + r^{-1}\,\omega_\mathrm{iK}\cdot(\omega_\mathrm{t} - r\,\omega_\mathrm{it})/(\omega_\mathrm{i} - r^{-1}\,\omega_\mathrm{it})\,. \tag{4.15}$$

The relaxation coefficients occurring in these expressions are also of importance for heat-flow birefringence. Therefore, some remarks on their possible experimental determination are in order. The quantities $A_{..}$ and $B_{..}$ are rather small. Thus to a good approximation η_iso and λ_iso as given by Eqs. (4.4), (4.10) are equal to the measured values of the viscosity and the heat conductivity, e. g. one has

$$\omega_\eta \approx p_0/\eta\,, \qquad \sigma_\eta \approx k\,T_0/v_\mathrm{th}\,\eta\,. \tag{4.16}$$

With the help of the relation (3.12) a rough estimate for ω_t and ω_i can be obtained if ω_it is put equal to zero. In this approximation which is accurate to within 20% one has $\omega_\mathrm{t}/\omega_\eta \approx 0.67$ and

$$\omega_\mathrm{i}/\omega_\eta \approx (c_\mathrm{i}/\mathrm{k})\,(m\,\lambda/k\,\eta - 15/4)^{-1}\,. \tag{4.17}$$

If data are available for the bulk viscosity η_v, the relaxation coefficient ω_it can be determined by the relation

$$-\frac{6}{5}\,r\,\frac{\omega_{\text{it}}}{\omega_\eta}=\frac{k\,c_{\text{i}}}{[(3/2)\,k+c_{\text{i}}]^2}\,\frac{\eta}{\eta_{\text{v}}}\,. \tag{4.18}$$

Then better values for ω_{t} and ω_{i} can be found [20].

The magnitude of the nondiagonal relaxation coefficients $\omega_{\eta\text{T}}$, ω_{tK}, ω_{iK} and the diagonal coefficients ω_{T}, ω_{K} which are of crucial importance for heat-flow birefringence can be inferred from the Senftleben-Beenakker effect of the viscosity and of the heat conductivity. This can be understood as follows. In the presence of a magnetic field with magnitude H the rotational angular momentum $\hbar\boldsymbol{J}$ of a molecule with the gyromagnetic ratio γ undergoes a precessional motion about the field direction with the precession frequency $\omega_{\text{H}}=\gamma H$. This precessional motion partially destroys the alignment set up in a transport situation. For $\omega_{\text{H}}\gg\omega_{\text{T}}$, ω_{K}, i. e. if a molecule makes many precessions between two collisions, η and λ approach their isotropic values η_{iso} and λ_{iso}. Hence the coefficients $A_{\eta\text{T}}$ and $B_{\lambda\text{K}}$ are associated with the magnetic-field induced change of the viscosity and the heat conductivity. More specifically, if the tensor polarization $\mathbf{a}$ and the Kagan vector $\boldsymbol{b}$ are the only significant types of alignment set up by a viscous flow and in the presence of a heat flux, respectively, one has [10, 16, 21, 22]

$$(\Delta\eta/\eta)_{\text{sat}}=-A_{\eta\text{T}}\,, \tag{4.19}$$

$$(\Delta\lambda_\perp/\lambda)_{\text{sat}}=-(9/10)\,B_{\lambda\text{K}}\,. \tag{4.20}$$

Here $\Delta\eta=\eta(H)-\eta$ is the difference between the viscosity $\eta(H)$ in the presence of a magnetic field with magnitude H and the field-free viscosity η. Likewise $\Delta\lambda_\perp=\lambda_\perp(H)-\lambda$ is the difference between the heat conductivity $\lambda_\perp(H)$ for a magnetic field perpendicular to the temperature gradient and the field-free value λ. The subscript "sat" refers to the saturation value which is approached for $\omega_{\text{H}}\gg\omega_{\text{T}}$ and $\omega_{\text{H}}\gg\omega_{\text{K}}$, respectively. The relaxation coefficients ω_{T} and ω_{K} can be inferred from the value of the magnetic field for which $\Delta\eta/\eta$ and $\Delta\lambda_\perp/\lambda$ attain half of their saturation values. They occur also in connection with the collisional and diffusional broadening of the depolarized Rayleigh line [4, 7, 23].

§ 5. Heat-flow Birefringence

For a discussion of birefringence the tensor polarization $\mathbf{a}$ has to be calculated from the transport-relaxation equations. In a steady state situation Eq. (3.6) yields

$$\mathbf{a}=-\frac{1}{\sqrt{2}\,p_0}\,\frac{\omega_{\eta\text{T}}}{\omega_{\text{T}}}\,\mathbf{p}-\frac{3}{5\,\omega_{\text{T}}}\,\overline{\nabla\boldsymbol{b}}\,. \tag{5.1}$$

According to Eqs. (3.2) – (3.10) the friction pressure tensor and the Kagan vector are given by

$$\mathbf{p}=-2\,\eta\,(\overline{\nabla\boldsymbol{v}}+(2/5\,p_0)\,\overline{\nabla\boldsymbol{q}_{\text{t}}})\,, \tag{5.2}$$

$$\boldsymbol{b}=\sqrt{\frac{2}{3}}\,\frac{1}{p_0\,\omega_{\text{K}}}\,(\omega_{\text{Kt}}\,\boldsymbol{q}_{\text{t}}+r^{-1}\,\omega_{\text{Ki}}\,\boldsymbol{q}_{\text{i}})-\frac{k\,T_0}{m\,\omega_{\text{K}}}\,\nabla\cdot\mathbf{a}\,, \tag{5.3}$$

for η see Equation (4.2). If the expressions (5.2), (5.3) are inserted into Eq. (5.1) and spatial derivatives of higher than 2-nd order are neglected,

$$\mathbf{a}=\;2\,\eta\,\frac{\omega_{\eta\text{T}}}{\sqrt{2}\,p_0\,\omega_{\text{T}}}\,\overline{\nabla\boldsymbol{v}}-2\,\eta\,\lambda_{\text{t}}\,\frac{2}{5\,\sqrt{2}\,p_0{}^2\,\omega_{\text{T}}}\left(\omega_{\eta\text{T}}+\frac{\sqrt{3}}{2}\,\frac{\omega_\eta}{\omega_{\text{K}}}\,\omega_{\lambda\text{K}}\right)\overline{\nabla\nabla}T \tag{5.4}$$

is found. Here η and λ_{t} can be approximated by their isotropic expressions (4.4), (4.8), for $\omega_{\lambda\text{K}}$ see Equation (4.15). The anisotropic part $\overline{\boldsymbol{\epsilon}}$ of the dielectric tensor is proportional to $\mathbf{a}$, cf. Equation (2.2). Thus Eq. (5.4) provides a justification for the constitutive laws (1.4), (1.5). Furthermore, the flow and heat-flow birefringence coefficients β and β_λ can be inferred from Equation (5.4). With

$$\beta_0=-\varepsilon'\,\frac{\eta}{\sqrt{2}\,p_0}=\frac{2\,\pi\,(\alpha_\parallel-\alpha_\perp)}{\sqrt{15}}\,\frac{\eta}{k\,T_0} \tag{5.5}$$

these coefficients are given by

$$\beta=\beta_0\,\omega_{\eta\text{T}}/\omega_{\text{T}}\,, \tag{5.6}$$

$$\beta_\lambda = \frac{2}{5\,p_0}\,\frac{\lambda_\mathrm{t}}{\lambda}\left(\frac{\omega_{\eta\mathrm{T}}}{\omega_\mathrm{T}} + \frac{\sqrt{3}}{2}\,\frac{\omega_\eta\,\omega_{\lambda\mathrm{K}}}{\omega_\mathrm{T}\,\omega_\mathrm{K}}\right)\beta_0\,. \tag{5.7}$$

Due to Eqs. (3.12), (4.15), β_λ can also be written as

$$\begin{aligned}\beta_\lambda &= \frac{2}{5\,p_0}\,\frac{\lambda_\mathrm{t}}{\lambda}\left(1+\frac{1}{2}\,\frac{\omega_\eta}{\omega_\mathrm{K}}\right)\beta + \frac{\sqrt{3}}{5\,p_0}\,r\,\frac{\lambda_\mathrm{i}}{\lambda}\,\frac{\omega_\eta\,\omega_{\mathrm{iK}}}{\omega_\mathrm{T}\,\omega_\mathrm{K}}\,\beta_0\\ &= \frac{2}{5\,p_0}\,\frac{\lambda_\mathrm{t}}{\lambda}\left[1+\frac{1}{2}\,\frac{\omega_\eta}{\omega_\mathrm{K}}\left(1+r\,\frac{\lambda_\mathrm{i}}{\lambda_\mathrm{t}}\,\frac{\omega_{\mathrm{iK}}}{\omega_{\mathrm{tK}}}\right)\right]\beta\,,\end{aligned} \tag{5.8}$$

with $r=(5\,k/2\,c_\mathrm{i})^{1/2}$. Clearly, β and β_λ depend linearly on $\omega_{\eta\mathrm{T}}$ and $\omega_{\lambda\mathrm{K}}$, the coefficients which characterize the strength of the collision-induced coupling between the tensor polarization and the friction pressure and between the Kagan vector and the heat flux. This is in contradistinction to the Senftleben-Beenakker effect of the viscosity and of the heat conductivity, cf. Eqs. (4.19), (4.20) and Equations (4.3) and (4.13).

Notice that the dependence of β and β_λ on the density n or on the pressure $p_0 = n\,k\,T_0$ is different. The flow birefringence coefficient β is independent of the number density. This is typical for transport coefficients governing the hydrodynamic regime of a dilute gas. According to Eqs. (5.7), (5.8), the heat-flow birefringence coefficient β_λ is inversely proportional to the number density, i. e. it is a Burnett-type transport coefficient. This result holds true as long as a typical mean free path of a molecule, say $l=(n\,\sigma_\eta)^{-1}$, is very small compared with a characteristic macroscopic length L. For heat conduction through a gas between two concentric cylinders with radii R_1 and R_2 as it has been considered in § 1, l has to be small compared both with R_1 and $R_2 - R_1$. This condition has to be kept in mind if one wants to estimate whether the heat-flow birefringence is of measurable size. In the Appendix a qualitative discussion of flow and heat-flow birefringence is presented for densities where the condition $l \ll L$ breaks down.

To obtain a feeling for the order of magnitude of the difference $\delta\nu$ of the indices of refraction caused by heat-flow birefringence, N_2 at room temperature is considered. In this case one has

$$\begin{aligned}\beta_0 &\approx 6\cdot 10^{-15}\ \mathrm{s}\,,\\ \lambda/k &\approx 2\cdot 10^{19}\ \mathrm{cm}^{-1}\,\mathrm{s}^{-1}\,,\\ 2\,\lambda_\mathrm{t}/5\,\lambda &\approx 0.3\,.\end{aligned}$$

With the experimental arrangement discussed in § 1 $r^{-2}(\ln R_2/R_1)^{-1} \approx 10\ \mathrm{cm}^{-2}$ can be realized by either $R_1 = 0.05$ cm, $R_2 = 1$ cm, $r \approx 0.2$ cm or $R_1 = 1.0$ cm, $R_2 = 1.1$ cm, $r \approx R_1$. For $(T_1 - T_2)/T_0 \approx 0.1$ and at a number density $n \approx 0.3\cdot 10^{17}\ \mathrm{cm}^{-3}$ which corresponds to a pressure of roughly 1 Torr and a mean free path of $5\cdot 10^{-3}$ cm,

$$\delta\nu \approx 20\left(\frac{\omega_{\eta\mathrm{T}}}{\omega_\mathrm{T}} + \frac{\sqrt{3}}{2}\,\frac{\omega_\eta}{\omega_\mathrm{T}}\,\frac{\omega_{\lambda\mathrm{K}}}{\omega_\mathrm{K}}\right)\cdot 10^{-13} \tag{5.9}$$

is obtained from Equations (1.8), (5.7). Ratios of nondiagonal and diagonal relaxation coefficients as they occur in Eq. (5.9) are typically of the order of 0.1. Thus $|\delta\nu| \approx 10^{-13}$ can be expected. It has been demonstrated by the flow birefringence experiments of Baas that $|\delta\nu|$ as small as 10^{-14} can be measured safely. Hence heat-flow birefringence should be measurable too.

It seems worth mentioning that the relaxation coefficients $\omega_{\eta\mathrm{T}}$, ω_T, ω_{tK}, ω_{iK} would vanish for molecules with angle independent interaction potential and consequently with a spherical scattering amplitude. Hence heat-flow birefringence, much as flow birefringence and the Senftleben-Beenakker effect, is a sensitive tool for obtaining information on the nonsphericity of the scattering amplitude and of the molecular interaction potential [16, 19, 24].

In summary, for a gas of linear molecules, the constitutive law for heat-flow birefringence has been derived from the Waldmann-Snider equation via the transport-relaxation equations. By Eq. (5.7), the heat-flow birefringence coefficient is expressed in terms of quantities characterizing the properties of single molecules and their binary interaction. Furthermore, its magnitude can be related to the Senftleben-Beenakker effect of the viscosity and of the heat conductivity. Measurements of the heat-flow birefringence in gases are possible and desirable. It might also be worthwhile to look for heat-flow birefringence in colloidal solutions though no kinetic theory and no estimate of the size of the effect exists for this case.

Appendix

Density Dependence of Flow and Heat-flow Birefringence

The formulae (5.6), (5.7) for the coefficients β and β_λ pertain to densities n which are high enough such that a typical mean free path l of a molecule is small compared with a characteristic macroscopic length L. If this condition is no longer fulfilled the collisions of the molecules with the wall have to be taken into consideration. Qualitatively, this can be achieved by replacing all diagonal relaxation coefficients (also those occurring in the expressions for η and λ) according to

$$\omega_{\mathrm{k}} \to \omega_{\mathrm{k}} + \omega_{\mathrm{wall}} = \omega_{\mathrm{k}} (1 + l_{\mathrm{k}} L^{-1}) \qquad \text{(A.1)}$$

where $\omega_{\mathrm{wall}} = v_{\mathrm{th}} L^{-1}$ is the wall collision frequency, v_{th} is a mean thermal velocity and the mean free path l_{k} is defined by $l_{\mathrm{k}} = v_{\mathrm{th}} \, \omega_{\mathrm{k}}{}^{-1} = (n \, \sigma_{\mathrm{k}})^{-1}$. Here "k" stands for η, T, t, ... Upon the assumption that wall collisions give no contribution to the non-diagonal relaxation coefficients and that all the l_{k} are approximately equal, the density dependence of $\delta \nu$ for flow birefringence is

$$\delta \nu = x^2 (1 + x)^{-2} (\delta \nu)_{\mathrm{d.g.}} \qquad \text{(A.2)}$$

with
$$x = L \, l^{-1} = n/n^*, \quad n^* = (\sigma L)^{-1}. \qquad \text{(A.3)}$$

Here $(\delta \nu)_{\mathrm{d.g.}}$ is the dilute gas value which is approached for high densities, viz. for $x \gg 1$ or $n \gg n^*$. Half of this value is reached for $x = 1 + \sqrt{2} \approx 2.4$, i. e. for $n \approx 2.4 \, n^*$. For small densities the flow birefringence increases quadraticly with the number density. To obtain a feeling for the order of magnitude of n^* it is noticed that $n^* \approx 0.3 \cdot 10^{16} \ \mathrm{cm}^{-3}$ for $\sigma \approx 30 \cdot 10^{-16} \ \mathrm{cm}^2$ and $L \approx 0.1$ cm. At room temperature this number density corresponds to a pressure of 0.1 Torr.

The difference between the indices of refraction due to heat-flow birefringence is now given by

$$\delta \nu = x^2 (1 + x)^{-3} (\delta \nu \, n/n^*)_{\mathrm{d.g.}} , \qquad \text{(A.4)}$$

where the expression $(\delta \nu \, n/n^*)_{\mathrm{d.g.}}$ is determined by Eq. (1.8) with β_λ given by Equation (5.7). Notice that $(\delta \nu \, n/n^*)_{\mathrm{d.g.}}$ is independent of the number density. According to (A.4), the heat-flow birefringence increases quadraticly with the number density at low densities. For $x = 2$ or $n = 2 \, n^*$ it reaches its maximum value $(\delta \nu)_{\max} = (4/27) \, (\delta \nu \, n/n^*)_{\mathrm{d.g.}}$ and for $n \gg n^*$ it decreases inversely proportional to the number density.

These qualitative considerations should yield the general trend of the low density behaviour of the flow and the heat-flow birefringence. A more detailed theory analogous to Vestner's [25] treatment of the Knudsen corrections of the Senftleben-Beenakker effect and of the thermomagnetic pressure difference [26] is desirable.

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