Birefringence in the Boundary Layer of a Rarefied Heat Conducting Gas

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In a heat conducting diatomic gas enclosed between parallel plates, the second rank tensor polarization of the rotational angular momenta is non-zero only near the plates. Therefore the gas is birefringent only in a boundary layer which is a few mean free paths thick. The birefringence is calculated with the help of a differential equation and a boundary condition for the tensor polarization. The difference in the index of refraction for light polarized parallel, respectively perpendicular to the temperature gradient is evaluated for the gases N_2 and CO with the information obtained from the field effects on viscosity and on thermal conductivity, and from thermomagnetic pressure difference measurements. The effect is estimated to be of measurable size.

1. Introduction

In an optically anisotropic medium, the dielectric tensor $\boldsymbol{\epsilon}$ has a non-zero symmetric traceless part $\overline{\boldsymbol{\epsilon}}$:

$$\boldsymbol{\epsilon} = \bar{\varepsilon}\,\boldsymbol{\delta} + \overline{\boldsymbol{\epsilon}}\,. \qquad (1)$$

The index of refraction ν_i for light polarized parallel to the unit vector $\boldsymbol{e}_i$ (an allowed direction of the electric field for a given ray vector) is [1]

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

For small anisotropy, the difference in the index of refraction for light polarized parallel to $\boldsymbol{e}_1$, respectively $\boldsymbol{e}_2$ simply is obtained from

$$\delta\nu_{12} \equiv \nu_1 - \nu_2 \approx \frac{1}{2\,\bar{\nu}}\,(\boldsymbol{e}_1\,\boldsymbol{e}_1 - \boldsymbol{e}_2\,\boldsymbol{e}_2) : \overline{\boldsymbol{\epsilon}}\,, \qquad (3)$$

with $\bar{\nu} = \sqrt{\bar{\varepsilon}}$.

A fluid is anisotropic only in a non-equilibrium situation, e. g. flow birefringence and heat flow firefringence occur in the presence of a flow, respectively a heat flow. Phenomenologically, these two effects can be described by an ansatz relating the symmetric traceless part of the dielectric tensor to the gradients of the flow velocity $\boldsymbol{v}$ and the heat flux $\boldsymbol{q}$ [2]:

$$\overline{\boldsymbol{\epsilon}} = -2\,\beta_v\,\overline{\boldsymbol{\nabla}\boldsymbol{v}} - 2\,\beta_q\,\overline{\boldsymbol{\nabla}\boldsymbol{q}}\,. \qquad (4)$$

For dilute polyatomic gases, the kinetic theory of flow birefringence and of heat flow birefringence has been developed by Hess [2–7] starting from the quantum mechanical kinetic equation due to Waldmann [8] and Snider [9]. Recently, experiments have been performed by Baas et al.: flow birefringence has been measured for a large number of pure polyatomic gases [10, 11] and for binary mixtures of N_2, respectively HD, with the noble gases He, Ne, Ar [12]; heat flow birefringence has been studied for O_2 [13].

In this paper we are interested in birefringence occurring in a rarefied polyatomic gas at rest ($\boldsymbol{v} = 0$) with a constant heat flux ($\boldsymbol{\nabla}\,\boldsymbol{q} = 0$), an effect which is not contained in the ansatz (4). First, some general remarks are made about the kinetic theory of birefringence phenomena in gases, then a heat conducting gas between parallel plates is considered as a special case.

In a diatomic gas with polarizabilities $\alpha_{\|}, \alpha_{\perp}$ parallel and perpendicular to the molecular axis $\boldsymbol{u}$, the symmetric traceless part of the dielectric tensor is proportional to the nonequilibrium average of the tensor $\overline{\boldsymbol{u}\boldsymbol{u}}$, respectively the tensor $\overline{\boldsymbol{J}\boldsymbol{J}}$ of the rotational angular momenta $\boldsymbol{J}$ [4, 5]:

$$\overline{\boldsymbol{\epsilon}} = 4\,\pi\,n_0\,(\alpha_{\|} - \alpha_{\perp})\,\langle\overline{\boldsymbol{u}\boldsymbol{u}}\rangle = -2\,\pi\,n_0\,(\alpha_{\|} - \alpha_{\perp})\,\langle\overline{\boldsymbol{J}\boldsymbol{J}}(J^2 - \tfrac{3}{4})^{-1}\rangle\,; \qquad (5)$$

n_0 is the equilibrium value of the particle density. With the help of the kinetic theory based on the Waldmann-Snider equation, the average $\overline{\boldsymbol{J}\boldsymbol{J}}$ polarization can be expressed by the inhomogeneities of the flow velocity and the temperature. In the most simple approach, it is assumed that this type of $\overline{\boldsymbol{J}\boldsymbol{J}}$ polarization which is responsible for birefringence also is set up in a streaming gas. Furthermore, it is assumed here that its flux, $\langle \boldsymbol{c}\,\overline{\boldsymbol{J}\boldsymbol{J}}(J^2 - 3/4)^{-1}\rangle$, is the dominant polarization in a heat conducting gas ($\boldsymbol{c}$ is the molecular velocity). If these assump-

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tions are valid, the birefringence phenomena are intimately related with the field effects on viscosity and on thermal conductivity [14]. For the present problem it is convenient to work with the "tensor polarization" $\mathbf{A}^{(02)}$ and the "Kagan polarization" $\underline{\mathbf{A}}^{(12)}$ defined by:

$$\mathbf{A}^{(02)} = p_0 (k_B T_0/m)^{1/2} \langle \overline{\boldsymbol{J}\boldsymbol{J}} (J^2 - \tfrac{3}{4})^{-1} \rangle \cdot \psi^{-1} (\tfrac{2}{15} \langle J^2 (J^2 - \tfrac{3}{4})^{-1} \rangle_0)^{-1/2}, \quad (6)$$

$$\underline{\mathbf{A}}^{(12)} = p_0 \langle \boldsymbol{c} \overline{\boldsymbol{J}\boldsymbol{J}} (J^2 - \tfrac{3}{4})^{-1} \rangle \quad (7)$$
$$\cdot \psi^{-1} [\sigma(1200)/\sigma(0200)]^{1/2} [\tfrac{2}{15} \langle J^2 (J^2 - \tfrac{3}{4})^{-1} \rangle_0]^{-1/2}.$$

Here, T_0 and $p_0 = n_0 k_B T_0$ are the equilibrium values of temperature and pressure, k_B is Boltzmann's constant, m is the molecular mass, and $\langle \ldots \rangle_0$ denotes an equilibrium average. The constant ψ which characterizes the magnitude of the polarizations can be expressed by effective cross sections (for the notation see e. g. Ref. [15]) according to

$$\psi = \frac{\sqrt{2}}{5} \frac{\lambda_{\text{transl}}}{\lambda} \frac{\sigma\binom{0200}{2000}}{[\sigma(0200)\,\sigma(1200)]^{1/2}} \cdot \left[1 - \frac{\sigma\binom{1001}{1200}}{\sigma\binom{1010}{1200}} \frac{\sigma\binom{1010}{1001} + r\,\sigma(1010)}{\sigma(1001) + r\,\sigma\binom{1010}{1001}}\right], \quad (8)$$

where $r = (2\,c_{\text{rot}}/5\,k_B)^{1/2}$, and c_{rot} is the rotational heat capacity per molecule. The ratio of the translational (λ_{transl}) and total (λ) thermal conductivities is found from

$$\frac{\lambda_{\text{transl}}}{\lambda} = \frac{\sigma(1001) + r\,\sigma\binom{1010}{1001}}{\sigma(1001) + 2\,r\,\sigma\binom{1010}{1001} + r^2\,\sigma(1010)}. \quad (9)$$

The numerical values for ψ listed in Table I are calculated with the effective cross sections derived from the field effects on viscosity and thermal conductivity, see Reference [16].

Table I. Parameters for the birefringence at $T_0 = 300$ K.

	$\alpha_{\parallel} - \alpha_{\perp}$ Å^3	ψ	$10^{14}\,\lambda\,\beta_T$ cm/K	L/l	C_t
N_2	0.69	0.049	8.13	1.03	3.7
CO	0.53	0.043	5.27	0.88	4.2
HD	0.30	0.044	6.19	3.30	10.2

With the help of the definition (6), the relation (5) can be written in the form

$$\bar{\boldsymbol{\varepsilon}} = -2\,\beta_T\,\mathbf{A}^{(02)}, \quad (10)$$

where β_T is given by

$$\beta_T = \pi (\alpha_{\parallel} - \alpha_{\perp})\,\psi (\tfrac{2}{15} \langle J^2 (J^2 - \tfrac{3}{4})^{-1} \rangle_0)^{1/2} \cdot (k_B T_0/m)^{-1/2} (k_B T_0)^{-1}. \quad (11)$$

In the bulk of the gas, the tensor polarization is proportional to $\overline{\nabla \boldsymbol{v}}$ and $\overline{\nabla \boldsymbol{q}}$, so that Eq. (10) leads to the constitutive law (4). For the coefficient β_v, Hess gives the expression [4, 5]

$$\beta_v = \frac{\pi}{2} (\alpha_{\parallel} - \alpha_{\perp}) \frac{\sigma\binom{0200}{2000}}{\sigma(0200)\,\sigma(2000)} \cdot \left(\frac{\pi}{15} \left\langle J^2 \left(J^2 - \frac{3}{4}\right)^{-1} \right\rangle_0 \right)^{1/2} (k_B T_0/m)^{-1/2}, \quad (12)$$

and β_q is related to β_v, β_T through [2]

$$\beta_q = \tfrac{2}{5}\,p_0^{-1} (\lambda_{\text{transl}}/\lambda)\,\beta_v + L\,\beta_T. \quad (13)$$

The length L is a characteristic free path determined by the relaxation constants of the polarizations:

$$L = \tfrac{1}{4} \sqrt{\pi}\, n_0^{-1} [\sigma(0200)\,\sigma(1200)]^{-1/2}. \quad (14)$$

According to Eqs. (12), (13) the coefficient β_v is independent of the gas density, whereas β_q is proportional to n_0^{-1}. Hence, flow birefringence is typical for the dilute gas regime, and heat flow birefringence becomes important in the bulk of a rarefied gas. In rarefied gases, however, the boundary layer effects have to be taken into account besides the bulk effects. In the following section, a situation is encountered where the tensor polarization is zero in the bulk of the gas, but is non-zero in the boundary layer. The resulting birefringence will be discussed in detail.

2. Tensor Polarization in a Heat Conducting Gas

Now, we consider a gas of diatomic molecules between infinite parallel plates located at $x = +d$ and $x = -d$ with the temperatures T_+ and T_-, respectively. In the most simple approximation, we assume that the translational temperature is equal to the rotational temperature everywhere. Then the temperature gradient is constant in space:

$$\nabla T = \frac{T_+ - T_-}{2\,d} (1 + C_t\,l/d)^{-1}\,\boldsymbol{e}. \quad (15)$$

Here, $\boldsymbol{e}$ is the unit vector in x-direction and l is a viscosity free path,

$$l = (k_B T_0/m)^{1/2}\,\eta/p_0 = \tfrac{1}{4} \sqrt{\pi} [n_0\,\sigma(2000)]^{-1}.$$

Since the temperature jump coefficient C_t is positive, the temperature gradient in the gas decreases with decreasing pressure.

In the bulk of the heat conducting gas, the angular momenta of the molecules are partially aligned. The experimental results for the field effects of thermal conductivity indicate that the Kagan polarization $\underline{\mathbf{A}}^{(12)}$ is the dominant type of alignment in most gases [14]. With the help of the moment method [17], a constitutive law for $\underline{\mathbf{A}}^{(12)}$ is derived [18] from the Waldmann-Snider equation:

$$\underline{\mathbf{A}}^{(12)} = -L\,\boldsymbol{\nabla}\,\mathbf{A}^{(02)} + \lambda\,\boldsymbol{\nabla} T\cdot\boldsymbol{\Delta}\,; \tag{16}$$

the symbol $\boldsymbol{\Delta}$ stands for the isotropic fourth rank tensor

$$\Delta_{\mu\nu,\mu'\nu'} = \tfrac{1}{2}(\delta_{\mu\mu'}\,\delta_{\nu\nu'} + \delta_{\mu\nu'}\,\delta_{\nu\mu'}) - \tfrac{1}{3}\,\delta_{\mu\nu}\,\delta_{\mu'\nu'}\,.$$

Similarly, for the tensor polarization $\mathbf{A}^{(02)}$ a second order differential equation is obtained,

$$\left(1 - L^2\frac{d^2}{dx^2}\right)\mathbf{A}^{(02)} = 0\,, \tag{17}$$

which is homogeneous for a heat conducting gas between parallel plates since $\boldsymbol{v} = 0$, and $\boldsymbol{\nabla\nabla}\,T = 0$ in the present approximation. This differential equation can be solved if a boundary condition for the tensor polarization is available. With Waldmann's thermodynamic method [19] such a boundary condition can be derived [18] from the interfacial entropy production with the result:

$$\mathbf{A}^{(02)} = \tilde{C}_{at}\,\boldsymbol{q}\cdot\boldsymbol{n}\,\overline{\boldsymbol{n}\boldsymbol{n}} + \tilde{C}_a\,\boldsymbol{n}\cdot\underline{\mathbf{A}}^{(12)}\,, \tag{18}$$

where $\boldsymbol{n} = \pm\boldsymbol{e}$ at $x = \pm d$. The phenomenological coefficients $\tilde{C}_{at}$ and $\tilde{C}_a > 0$ characterize the production of tensor polarization by molecule-wall collisions from a normal heat flux and a Kagan polarization. From an analysis of thermomagnetic pressure difference data [20], values for $\tilde{C}_a$ and $\tilde{C}_{at}$ have been found with rather large uncertainties; typical values are listed in Table II.

Table II. Birefringence for $(T_- - T_+)/2\,d = 1$ K/cm, $L/d = 0.2$, calculated from Eqs. (21), (24) with the parameters from Table I and $\tilde{C}_a$, $\tilde{C}_{at}$ from Reference [20]. The pressure p_0 refers to a free path of $L = 1$ cm.

	$\tilde{C}_a$	$\tilde{C}_{at}$	$10^{14}\,\Gamma$	$10^{14}\,\delta\nu_{12}$ ($x = 0.9\,d$)	p_0 torr
N_2	1.0	−0.74	7.07	2.49	0.0041
CO	1.0	−0.53	4.03	1.25	0.0035

The solution of Eqs. (16) – (18) yields the result already known from the theory of the thermomagnetic pressure difference (for $L/d \ll 1$) [21],

$$\mathbf{A}^{(02)}(x) = -\frac{\tilde{C}_a - \tilde{C}_{at}}{\tilde{C}_a + 1}\,\frac{\sinh x/L}{\sinh d/L}\,\overline{\boldsymbol{e}\boldsymbol{q}}\,, \tag{19}$$

with $\boldsymbol{q} = -\lambda\,\boldsymbol{\nabla}\,T$. At the walls $x = \pm d$, the tensor polarization has the non-zero value

$$\mathbf{A}^{(02)}(\pm d) = \mp\frac{\tilde{C}_a - \tilde{C}_{at}}{\tilde{C}_a + 1}\,\overline{\boldsymbol{e}\boldsymbol{q}}\,.$$

With increasing distance from the plates, $\mathbf{A}^{(02)}$ decreases (essentially exponentially) over a length of the order of the free path L to its bulk value of zero. According to Eq. (10), the dielectric tensor has the same spatial dependence:

$$\overline{\boldsymbol{\epsilon}}(x) = 2\,\beta_{\mathrm{T}}\,\frac{\tilde{C}_a - \tilde{C}_{at}}{\tilde{C}_a + 1}\,\frac{\sinh x/L}{\sinh d/L}\,\overline{\boldsymbol{e}\boldsymbol{q}}\,. \tag{20}$$

Hence, the gas is birefringent only in a boundary layer of thickness L. With $\bar{\nu} \approx 1$, Eqs. (3), (15), (20) lead to the relations

$$\delta\nu_{12}(x) = \delta\nu_{12}(d)\,\frac{\sinh x/L}{\sinh d/L}\,, \tag{21}$$

$$\delta\nu_{12}(d) = [(\boldsymbol{e}_1\cdot\boldsymbol{e})^2 - (\boldsymbol{e}_2\cdot\boldsymbol{e})^2]\,\Gamma\,(1 + C_t\,l/d)^{-1}\,, \tag{22}$$

where Γ is an abbreviation for

$$\Gamma = \lambda\,\beta_{\mathrm{T}}\,\frac{T_- - T_+}{2\,d}\,\frac{\tilde{C}_a - \tilde{C}_{at}}{\tilde{C}_a + 1}\,. \tag{23}$$

In particular, if the light is travelling parallel to the plates, the two possible polarizations are $\boldsymbol{e}_1 = \boldsymbol{e}$ and $\boldsymbol{e}_2 \perp \boldsymbol{e}$:

$$\delta\nu_{12}(d) = \Gamma\,(1 + C_t\,l/d)^{-1}\,. \tag{24}$$

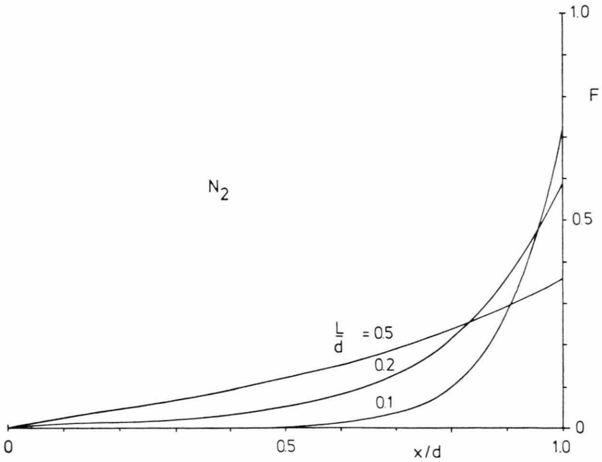

Fig. 1. Pressure dependence of birefringence for N_2 according to Eqs. (21), (24):
$F(x) = \delta\nu_{12}(x)/\Gamma = (1 + C_t\,l/d)^{-1}\,\sinh x/L/\sinh d/L$.
The parameters $C_t = 3.7$ and $L/l = 1.03$ from Table I have been used.

Hence, for vanishing free path, $\delta\nu_{12}(d)$ is independent of pressure. For low pressures, the decay length L of $\delta\nu_{12}(x)$ is large, but (due to the temperature jump) the boundary value $\delta\nu_{12}(d)$ is smaller than its high pressure limit Γ. This type of pressure dependence of the effect is displayed in Fig. 1 for the gas N_2.

3. Estimate for the Magnitude of the Birefringence

Birefringence in the boundary layer can be observed only if the free path L is large enough. On the other hand, the theory presented here is applicable only at relatively small Knudsen numbers L/d, e.g. $L/d = 0.2$. These two conditions can be met by an appropriate choice of the plate spacing and of the gas pressure.

In Table I, some quantities needed for the estimate of $\delta\nu_{12}$ at room temperature ($T_0 = 300$ K) are listed. With the values for the effective cross sections from Ref. [16], the coefficient ψ and the ratio L/l are calculated from Eqs. (8), (9), (14) for the gases N_2, CO and HD. Taking the known values for the polarizabilities [22, 11] and for the thermal conductivity [23], the quantity $\lambda\,\beta_T$ is then found from Eq. (11). The temperature jump coefficient C_t as obtained from thermal conductivity measurements at low pressures [24] is also given in Table I. In Table II, characteristic values for the parameters $\tilde{C}_a$ and $\tilde{C}_{at}$ as derived from thermomagnetic pressure difference data [20] are presented for N_2 and CO. For the gas HD, thermomagnetic pressure difference cannot be described adequately [20] by the simple theory of Ref. [21], so that it is not possible to determine values for $\tilde{C}_a$ and $\tilde{C}_{at}$. With a temperature gradient of $(T_- - T_+)/2\,d = 1$ K/cm and a Knudsen number of $L/d = 0.2$, the quantity Γ is now calculated from Equation (23). From Equation (21), $\delta\nu_{12}$ is found as a function of the position. According to the values for $\delta\nu_{12}(x = 0.9\,\mathrm{d})$ given in Table II, birefringence in the boundary layer should be measurable since it is of the same order of magnitude as flow birefringence [11] and heat flow birefringence [13]. Notice that $\delta\nu_{12}(x)$ should be positive for $x > 0$, i.e. near the cold plate.

In such an experiment, the decay length L and the ratio $(\tilde{C}_a - \tilde{C}_{at})/(\tilde{C}_a + 1)$ could be determined, thus providing a crucial test for the theoretical treatment of the polarizations which is e.g. applied in the theory of the thermomagnetic pressure difference. This information could prove very helpful for the gas HD.

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