Birefringence in the Boundary Layer of a Rarefied Heat Conducting Gas

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In a heat conducting gas of diatomic molecules enclosed between parallel plates, the second rank tensor polarization of the rotational angular momenta can be shown to be nonzero in a boundary layer near the plates, a few mean free paths thick. This phenomenon can be experimentally studied by measuring the birefringence of the boundary layer. Although the predicted effects are very small (Δn of the order of 10^{-14}), the existence of this effect is clearly demonstrated and qualitative results are obtained for N_2, CO and HD.

1. Introduction

In 1959 [1–3] Waldmann introduced the concept of nonequilibrium polarization in kinetic theory. At the 1964 semiar at Brown University [4] he discussed the consequences for the behavior of transport properties of polyatomic gases. He pointed out that as opposed to the situation in noble gases there exists here a class of tensorial transport phenomena that originates in a non-random distribution of molecular orientation. As an example he mentioned that as a consequence under certain conditions one might except birefringence. These concepts do not only play a dominant role in the kinetic theory of polyatomic gases in the dilute and rarefied gas regime (cf. the paper by Schmidt, Köhler, and Hess in this issue), they also give rise to an interesting class of boundary layer phenomena (see the paper by Vestner). Experiments in this field so far [5–8] had a serious drawback in as far as the boundary layer effects were not observed directly, but as changes in the overall behavior of the gas.

In 1977 Vestner and Beenakker suggested an experiment which would give direct insight in processes taking place in the boundary layer of a heat conducting gas of polyatomic molecules [9]. Starting with Waldmann's equations for the production and decay of polarizations at a surface they predicted that such a gas would be birefringent in a boundary layer a few mean free paths thick. It was suggested that by scanning the birefringence near the surface with a narrow laser beam, a direct indication of the extent to which tensor $\langle \overline{JJ} \rangle$ polarization is produced near wall might be obtained. This would yield an independent test on the validity of the kinetic theory for boundary layer phenomena as well as a determination of the generalized slip coefficients introduced by Waldmann [10].

This paper will present the first results of an experiment designed to measure the boundary layer birefringence. Before describing the procedure, an outline of the relevant theory will be given.

2. Theory of Boundary Layer Birefringence

Consider a rarefied gas of polyatomic molecules confined between parallel plates at temperatures T_+ and T_- (see Figure 1). The nonzero temperature difference will give rise to a heat flow $\boldsymbol{q}$ in the gas. Using the moment method, Vestner and Beenakker [9] derived two equations for the production

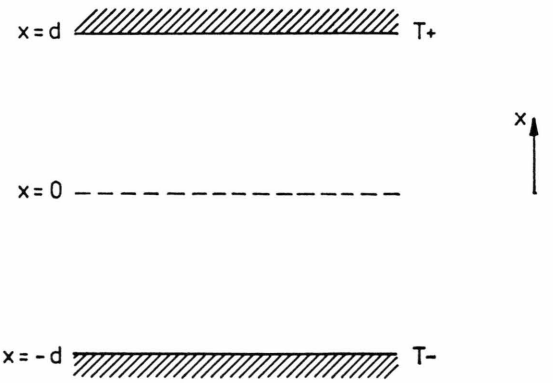

Fig. 1. Parallel plate arrangement with coordinate system.

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and decay of angular momentum polarization in the gas near the plates:

$$\underline{\mathbf{A}}^{(1200)} = -L\nabla\mathbf{A}^{(0200)} + \boldsymbol{q}\cdot\boldsymbol{\Delta} \tag{1}$$

and

$$\left(1 - L^2\frac{\mathrm{d}^2}{\mathrm{d}x^2}\right)\mathbf{A}^{(0200)} = 0\,. \tag{2}$$

The relevant polarizations are characterized by the quantities $\underline{\mathbf{A}}^{(1200)}$ (Kagan polarization) and $\mathbf{A}^{(0200)}$ (angular momentum polarization) which are proportional to the more familiar quantities $\langle\underline{\boldsymbol{\Phi}}^{(1200)}\rangle$ and $\langle\boldsymbol{\Phi}^{(0200)}\rangle$. These are defined so as to give (1) and (2) a simple form:

$$\mathbf{A}^{(0200)} = p\left(\frac{kT}{m}\right)^{1/2}\psi^{-1}\left\langle\sqrt{\frac{15}{2}}\,\frac{\overline{\boldsymbol{J}\boldsymbol{J}}}{J^2}\right\rangle, \tag{3}$$

$$\underline{\mathbf{A}}^{(1200)} = \left[\frac{\mathscr{S}(1200)}{\mathscr{S}(0200)}\right]^{1/2} p\left(\frac{kT}{m}\right)^{1/2} \cdot\psi^{-1}\left\langle\sqrt{15}\,\boldsymbol{W}\,\frac{\overline{\boldsymbol{J}\boldsymbol{J}}}{J^2}\right\rangle, \tag{4}$$

with

$$\psi = \frac{2}{5}(1+r^2)^{1/2}\,\frac{\mathscr{S}\binom{101+1}{1200}}{[\mathscr{S}(1200)\,\mathscr{S}(0200)]^{1/2}} \tag{5}$$

characterizing the strength of the polarizations in the notation as introduced by Thijsse et al. [11] and with $r = (2C_{\mathrm{rot}}/5k)^{1/2}$.

The quantity L in (1) defines a mean free path length

$$L = \frac{\sqrt{\pi}}{4}\,\frac{1}{n}\,\frac{1}{[\mathscr{S}(1200)\,\mathscr{S}(0200)]^{1/2}} \tag{6}$$

and the fourth rank isotropic tensor symbolized by $\boldsymbol{\Delta}$ in the same equation is given by

$$\Delta_{ijkl} = \tfrac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) - \tfrac{1}{3}\delta_{ij}\delta_{kl}\,. \tag{7}$$

From Eq. (2) $\mathbf{A}^{(0200)}$ may be determined apart from a constant. This constant depends on the applied boundary condition which may be derived from Waldmann's equation for the interfacial entropy production at the plates [10]:

$$\mathbf{A}^{(0200)} = \tilde{C}_{\mathrm{at}}\,\boldsymbol{q}\cdot\boldsymbol{n}\,\boldsymbol{n}\boldsymbol{n} + \tilde{C}_{\mathrm{a}}\,\boldsymbol{n}\cdot\underline{\mathbf{A}}^{(1200)}\,. \tag{8}$$

The vector $\boldsymbol{n}$ is normal to the plates. This condition states that tensor polarization at the wall is produced from the heat flow $\boldsymbol{q}$, with a strength characterized by $\tilde{C}_{\mathrm{at}}$, and from a Kagan polarization $\underline{\mathbf{A}}^{(1200)}$, characterized by $\tilde{C}_{\mathrm{a}}$. Solving (1) and (2) with this boundary condition, one finds for the tensor polarization

$$\mathbf{A}^{(0200)} = -\frac{\tilde{C}_{\mathrm{a}} - \tilde{C}_{\mathrm{at}}}{\tilde{C}_{\mathrm{a}} + 1}\,\frac{\sinh(x/L)}{\sinh(d/L)}\,\boldsymbol{n}\boldsymbol{q}\,. \tag{9}$$

With the general relations between the tensor polarization, the anisotropic part of the dielectric tensor and the birefringence [12], the following expression for the difference in index of refraction $\Delta\nu$ parallel and perpendicular to $\boldsymbol{n}$ is easily obtained:

$$\Delta\nu = \frac{1}{10\sqrt{3}}\,\frac{(\alpha_{\parallel} - \alpha_{\perp})}{\varepsilon_0}\,\frac{1}{kT}\left(\frac{m}{kT}\right)^{1/2} \cdot(1+r^2)^{-1/2}\,\frac{\mathscr{S}\binom{101+1}{1200}}{[\mathscr{S}(1200)\,\mathscr{S}(0200)]^{1/2}} \cdot q\,\frac{\tilde{C}_{\mathrm{a}} - \tilde{C}_{\mathrm{at}}}{\tilde{C}_{\mathrm{a}} + 1}\,\frac{\sinh(x/L)}{\sinh(d/L)}\,. \tag{10}$$

In this expression all effective cross sections are known from dilute gas bulk measurements. The only unknowns are $\tilde{C}_{\mathrm{a}}$ and $\tilde{C}_{\mathrm{at}}$. From (10) some characteristic features of boundary layer birefringence may be deduced immediately. The effect vanishes halfway between the plates ($x = 0$). Near the plates, where $|x| \approx d \ll L$, the birefringence decreases exponentially with the distance from the wall, i.e. as $\exp[-(d - |x|)/L]$.

3. Experimental Method and Results

Experimentally a measurement of the birefringence as a function of wall distance is inconvenient, because it requires moving either the plates or the probe beam. As the dependence on position is contained in the factor $\sinh(x/L)$ and L is inversely proportional to p, measuring how the birefringence varies as a function of p is experimentally more attractive. Experimental curves of $\Delta\nu$ versus p may then be compared with the theoretical prediction using (10), providing a direct test of the validity of the kinetic theory for boundary phenomena.

The measuring cell in which boundary layer birefringence was detected consists essentially of the hot and cold plate arrangement shown schematically in Figure 1. Several considerations determine the choice of materials and dimensions of such an apparatus. As the laser beam used to measure the birefringence has a diameter of about 0.5 mm, measurements in the boundary cannot be performed closer than 2 mm to the wall, in order to avoid problems with scattering. Thus measure-

ments have to take place at pressures so low that L is of the order of 1—10 mm, i.e., $p = 5—0.5$ Pa. Consequently light metals and polymers are not suited for an apparatus because they can degas substantially at low pressures. In our cell we used only glass and stainless steel, materials which allow baking up to 300 °C. Mass spectrometer analysis revealed that only minute amounts ($10^{-4} — 10^{-3}$ Pa) of hydrogen and carbon monoxide were left after baking.

Furthermore the equations presented in Sect. 2 apply only to gases where the influence of the walls becomes negligible in the bulk. Therefore the distance between the walls has to be so large that $d/L \gg 1$. To satisfy this a d value of 5 cm was chosen. The level of birefringence which may be detected is proportional to the path length of the laser beam in the gas. The cell discussed here has a path length of 50 cm.

Both plates were kept at 298 K by circulating water from a thermostate controlled bath through the tubing (see Figure 2). Rapid modulation of the temperature gradient and thus $\boldsymbol{q}$ is achieved by passing current through a wire grid of parallel platinum wires of 50 μm diameter with 5 mm spacings, placed at a distance of 1 mm from one of the plates. Close to the grid the temperature distribution will be nonplanar. At distances larger than the spacing of the wires, the inhomogeneity disappears rapidly, as in the well-known electrostatic case (cf. [13]). Hence the equitemperature surface near the furthest cold plate will be flat just as in the parallel plate case. Passing current at intervals of 10 seconds allowed phase-sensitive detection of the boundary layer birefringence, and reduced the noise to such a low level that a resolution of $\Delta\nu \approx 2 \times 10^{-15}$ was achieved. This proved to be sufficient to perform measurements in N_2, CO and HD at room temperature (298 K) and with temperature differences up to 40 K. For the optical arrangement see [14].

Measurements were performed as a function of the temperature difference ΔT between the wires and the cold plates because it is difficult to measure the driving heat flux, $\boldsymbol{q}$, directly. This complicates the interpretation of the data by the fact that the relation between ΔT and $\boldsymbol{q}$ is influenced by Knudsen effects. In a simple flat plate arrangement one would have

$$q = \lambda \left(1 + \frac{K}{p}\right)^{-1} \frac{\Delta T}{2d} . \tag{11}$$

In our case one has complications because the heat

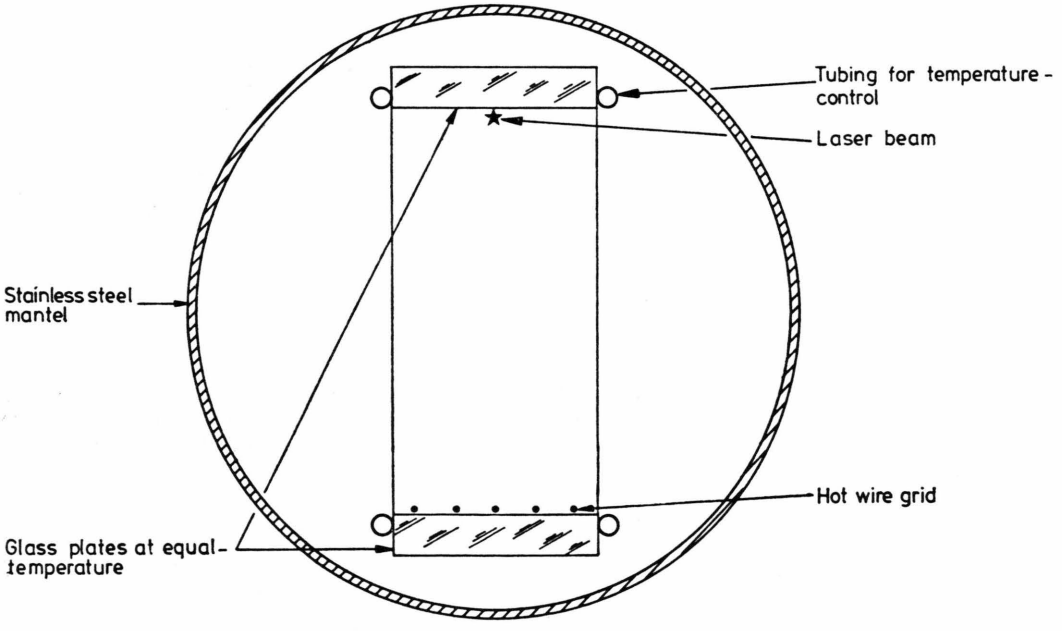

Fig. 2. Cross section of the cell in which boundary layer birefringence is measured. The laser beam passes perpendicularly to the plane of drawing.

source is a grid. For small mean free path one expects an expression of the form

$$q = A\,\lambda\left(1 + \frac{K'}{p}\right)^{-1} \frac{\Delta T}{2d} \tag{12}$$

with a different value of K' and a geometrical factor A. Both A and K' are not well-known. This gives rise to systematic uncertainties in the quantitative interpretation of the results. At lower pressures, when the mean free path becomes of the order of the grid spacing; large deviations will occur. At all pressures q will remain proportional to ΔT and consequently the same holds for the resulting birefringence. This was checked by measurements in N_2 with the laser beam at 2 mm from the cold plate at three different pressures. (The temperature of the wires was monitored through their resistance, pressures were measured with a differential capacitance type manometer, Barocel 10 T). The result is shown in Figure 3. The effect is seen to vary linearly with ΔT as is expected. Thus the quantity $\Delta v/\Delta T$ may be used to compare measurements at various pressures. Such measurements are shown in Figs. 4, 5 and 6 for N_2, CO and HD. The experimental points are seen to pass through a maximum with decreasing pressure. This behavior may be shown to be in qualitative agreement with Equation (10). For x and $d \gg L$ the factor $\sinh(x/L)/\sinh(d/L)$ may be approximated by

$$\exp\left[-\frac{(d - |x|)}{L}\right].$$

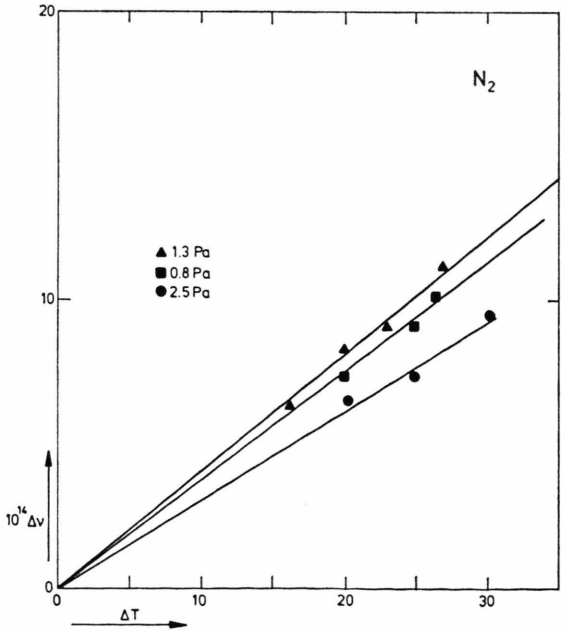

Fig. 3. Birefringence for N_2 as a function of applied temperature difference at three pressures.

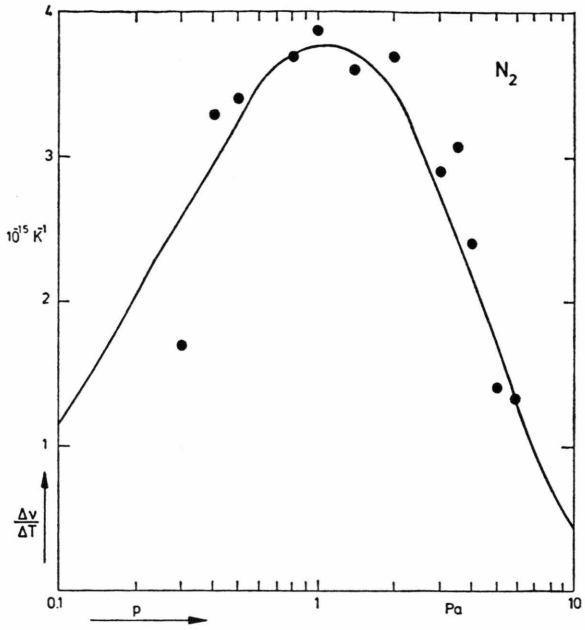

Fig. 4. The observed birefringence in N_2 divided by the temperature difference plotted versus the pressure. The solid line is calculated using Eqs. (10) and (12) with $K' = 0.40$ Pa.

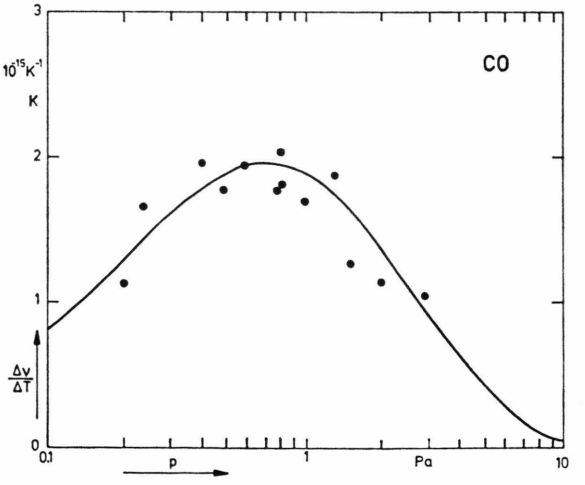

Fig. 5. The observed birefringence in CO divided by the temperature difference plotted versus the pressure. The solid line is calculated using Eqs. (10) and (12) with $K' = 0.45$ Pa.

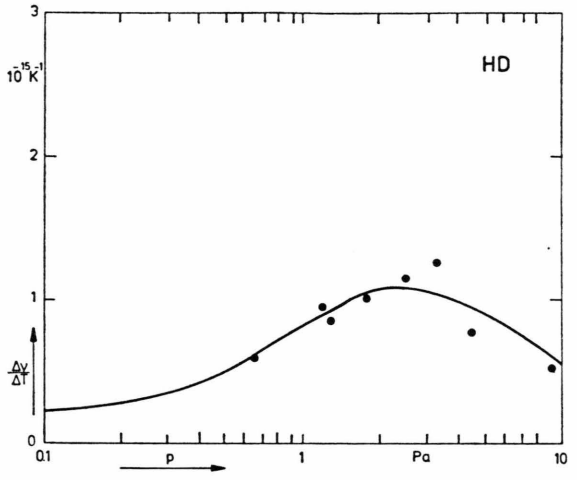

Fig. 6. The observed birefringence in HD divided by the temperature difference plotted versus the pressure. The solid line is calculated using Eqs. (10) and (12) with $K' = 2.0$ Pa.

Hence as L varies as $1/p$ the effect should increase when the pressure is lowered. On the other hand q decreases at lower pressure (cf. (12)) which causes the birefringence to eventually vanish. This explains the drop at the low pressure side. The curves drawn in Figs. 4, 5 and 6 are derived from (10) using q given by (12), taking for K' the value of a flat arrangement [9]. They are scaled to the experimental value of $\Delta\nu$ at the top. As the position of the peak (maximum value) occurs at pressures where the mean free path is no longer small compared to the grid structure one has to be careful to draw further quantitative conclusions from the observed agreement. Hence we will refrain from deriving values for

$$(\tilde{C}_a - \tilde{C}_{at})/(1 + \tilde{C}_a)$$

based on our results.

Summarizing, one may say that the results of this experiment firmly establish the existence of a birefringent boundary layer in a rarefied heat conducting gas of polyatomic molecules. A quantitative test of the theory requires further changes in the cell to enable an accurate determination of q; such improvements are in progress.

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