Viscosity of a Polar Gas of Symmetric Top Molecules in Perpendicular Electric and Magnetic Fields

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A kinetic theory treatment of the influence of perpendicular magnetic and electric fields on the viscosity is given for a polar gas of symmetric top molecules. Expressions for the 9 independent viscosity coefficients are derived. In particular, the electric field influence on the transverse viscomagnetic pressure difference is studied.

In the presence of an external magnetic or electric field, the viscosity coefficient of a polyatomic gas becomes a field dependent 4-th rank tensor (Senftleben-Beenakker effect [1]). In the magnetic case, this tensor is determined by 5 independent viscosity coefficients [2], three being even and two being odd in the field. The latter give rise to transverse pressure differences [3]. In the electric case, the odd-in-field coefficients are absent because of parity reasons and only three independent even-in-field coefficients remain. All these coefficients have been related [4, 5] to collision integrals of the linearized Waldmann-Snider collision operator [6, 7].

If a polar gas is simultaneously influenced by a magnetic and an electric field, the symmetry of the system is lowered and the number of independent viscosity coefficients increases. The following treatment is confined to perpendicular electric and magnetic fields $\boldsymbol{E}$ and $\boldsymbol{H}$; unless $\boldsymbol{E} \parallel \boldsymbol{H}$, the general case can always be reduced to this case. From inspection of the viscosity tensors written down in a coordinate frame given by $\boldsymbol{E}$, $\boldsymbol{H}$ and $\boldsymbol{E} \times \boldsymbol{H}$, one infers the existence of 9 independent viscosity coefficients. In the present letter the magnetic- and electric field dependence of these coefficients is explained for a polar gas of symmetric top molecules, and the change of the transverse viscomagnetic pressure difference in an electric field is discussed.

Starting point of our kinetic treatment is the linearized Waldmann-Snider equation

$$\frac{\partial \Phi}{\partial t} + \boldsymbol{c} \cdot \frac{\partial \Phi}{\partial \boldsymbol{x}} + i\,\mathscr{L}(\boldsymbol{E}, \boldsymbol{H})\,\Phi + \omega(\Phi) = 0\,, \qquad (1)$$

for the relative deviation Φ of the distribution operator from equilibrium, $\omega(\Phi)$ is the linearized Waldmann-Snider collision operator and

$$i\,\mathscr{L}(\boldsymbol{E}, \boldsymbol{H}) \cdots = -\,[(\boldsymbol{\mu}_e \cdot \boldsymbol{E} + \boldsymbol{\mu}_m \cdot \boldsymbol{H}), \ldots]_- \qquad (2)$$

is the Liouville operator governing the free precessional motion of the electric and magnetic moments $\boldsymbol{\mu}_e$ and $\boldsymbol{\mu}_m$, respectively.

The nonequilibrium distribution Φ which depends on time t, position $\boldsymbol{x}$, molecular velocity $\boldsymbol{c}$, molecular rotational angular momentum $\boldsymbol{J}$ and its component along the body fixed symmetry axis, $J_\parallel$, is expanded into a series of moments (moment method, cf. Refs. [8, 9]). For our purpose, it is sufficient to take into account the following moments:

$$\boldsymbol{a} = (m/k_B T_0)^{1/2} \langle \boldsymbol{c} \rangle \quad (\propto \text{mean velocity}),$$

$$\mathbf{P} = (m/\sqrt{2}\, k_B T_0) \langle \overline{\boldsymbol{c}\,\boldsymbol{c}} \rangle \quad (\propto \text{friction pressure tensor}),$$

$$\mathbf{A} = (15/2 \langle J^2 (J^2 - 3/4) \rangle_0)^{1/2} \langle \overline{\boldsymbol{J}\,\boldsymbol{J}} \rangle \quad \text{(tensor polarization)}$$

and

$$\mathbf{B} = (15/2 \langle J_\parallel^2 J^2 (J^2 - 3/4) \rangle_0)^{1/2} \langle J_\parallel \overline{\boldsymbol{J}\,\boldsymbol{J}} \rangle .$$

The bracket $\langle \cdots \rangle$ denotes a nonequilibrium average and $\langle \cdots \rangle_0$ an equilibrium average; the bar $\overline{}$ means the symmetric irreducible part of a tensor. The following moment equations are obtained for the stationary case:

$$\omega^P \mathbf{P} + \omega^{PA} \mathbf{A} = -\sqrt{2}\, \overline{\nabla \boldsymbol{v}}\,, \qquad (3)$$

$$\omega^{AP} \mathbf{P} + \omega^A \mathbf{A} + \omega_H \mathscr{H} : \mathbf{A} + \omega_E \mathscr{E} : \mathbf{B} = 0, \qquad (4)$$

$$\omega_E \mathscr{E} : \mathbf{A} + \omega^B \mathbf{B} = 0\,. \qquad (5)$$

Here, ω^A, ω^P, ω^B are the relaxation frequencies of the corresponding tensors and $\omega^{AP} = \omega^{PA}$ is the coupling coefficient of friction pressure tensor and tensor polarization; $\omega_H = g_{eff}\, \mu_N H/\hbar$ is the magnetic Larmor frequency where μ_N is the nuclear magneton and $g_{eff} = g_\perp (1 + (g_\parallel - g_\perp)/g_\perp \langle J_\parallel^2/J^2 \rangle_0)$ is an effective rotational g-factor [10] and

$$\omega_E = \mu_e E \langle J^2 \rangle_0^{-1} (\langle J^2 J_\parallel^2 (J^2 - 3/4) \rangle_0 / \langle J^2 (J^2 - 3/4) \rangle_0)^{1/2} \hbar$$

is the effective electric Larmor precession frequency with μ_e being the electric dipole moment. The fourth rank tensors $\mathscr{H}$ and $\mathscr{E}$ describe the

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rotation of irreducible 2nd rank tensors around the respective magnetic and electric field directions $\boldsymbol{h}$ and $\boldsymbol{e}$ [11]. By use of the technique of projection operators developed by Hess and Waldmann [11], Eq. (5) is easily solved to give $\mathbf{B}$ in terms of $\mathbf{A}$. Insertion of the result in Eq. (4) yields

$$\sum_{m,n=-2}^{+2}(1+m^2\varphi_E{}^2+i\,n\,\varphi_H)\,\mathscr{P}^{(m)}(\boldsymbol{e})\colon\mathscr{P}^{(n)}(\boldsymbol{h})\colon\mathbf{A} = -(\omega^{AP}/\omega^A)\,\mathbf{P}\,. \tag{6}$$

In Eq. (6), $\mathscr{P}^{(m)}(\boldsymbol{e})$ and $\mathscr{P}^{(n)}(\boldsymbol{h})$ are the respective fourth rank projection operators for the electric and magnetic case [11], $\varphi_E=(\omega_E{}^2/\omega^A\omega^B)^{1/2}$ and $\varphi_H=\omega_H/\omega^A$ are the corresponding effective precession angles. The double sum of the l.h.s. of Eq. (6) can be regarded as a 6×6 matrix (acting on the 6-column $\mathbf{A}$) if a matrix label is identified with a pair of tensor indices. Since the tensors are traceless, the original system of 6 linearly dependent tensor equations can be reduced to a system of 5 linearly independent equations. Choosing $\boldsymbol{h}$ in z-direction, $\boldsymbol{e}$ in y-direction and $\boldsymbol{e}\times\boldsymbol{h}$ in x-direction and the $\overline{zz}$, $\overline{yy}$, $\overline{xy}$, $\overline{yz}$, $\overline{xz}$-components as the independent ones, one can solve Eq. (6) for $\mathbf{A}$ by matrix inversion:

$$\mathbf{A} = -(\omega^{AP}/\omega^A)\,\mathscr{A}(\boldsymbol{E},\boldsymbol{H})\colon\mathbf{P}\,. \tag{7}$$

Switching back to 6 components, we obtain the following expressions for the 9 non-vanishing elements of the $\mathscr{A}$-tensor:

$$\begin{aligned}
&\mathfrak{A}_{\overline{yy},\overline{yy}} = 2(f_1f_2+g_1)/3(f_1f_2+4f_3g_1)\,,\\
&\mathfrak{A}_{\overline{yy},\overline{xy}} = -\mathfrak{A}_{\overline{xy},\overline{yy}}\\
&\qquad = -g_2f_2/(f_1f_2+4f_3g_1),\\
&\mathfrak{A}_{\overline{yy},\overline{zz}} = \mathfrak{A}_{\overline{zz},\overline{yy}}\\
&\qquad = -(f_1f_2+4g_1)/3(f_1f_2+4f_3g_1)\,,\\
&\mathfrak{A}_{\overline{zz},\overline{zz}} = 2(f_1^2+4g_1)/3(f_1f_2+4f_3g_1)\,,\\
&\mathfrak{A}_{\overline{zz},\overline{xy}} = -\mathfrak{A}_{\overline{xy},\overline{zz}}\\
&\qquad = (f_2-1)\,g_2/2(f_1f_2+4f_3g_1)\,,\\
&\mathfrak{A}_{\overline{xy},\overline{xy}} = f_2/2(f_1f_2+4f_3g_1)\,,\\
&\mathfrak{A}_{\overline{xz},\overline{xz}} = f_1/2(f_1f_2+g_1)\,,\\
&\mathfrak{A}_{\overline{xz},\overline{yz}} = -\mathfrak{A}_{\overline{yz},\overline{xz}} = g_2/2(f_1f_2+g_1)\,,\\
&\mathfrak{A}_{\overline{yz},\overline{yz}} = f_2/2(f_1f_2+g_1)\,.
\end{aligned} \tag{8}$$

Note, that $\mathfrak{A}_{\overline{xx},\ldots}=-\mathfrak{A}_{\overline{yy},\ldots}-\mathfrak{A}_{\overline{zz},\ldots}$ etc. The field dependent functions $f_1\ldots g_2$ are given by

$$\begin{aligned}
&f_1=1+\varphi_E{}^2,\quad f_2=1+4\varphi_E{}^2,\\
&f_3=1+3\varphi_E{}^2;
\end{aligned} \tag{9}$$

$$g_1=\varphi_H{}^2,\quad g_2=\varphi_H\,. \tag{10}$$

From Eqs. (7), (3), the viscosity tensor $\boldsymbol{\eta}(\boldsymbol{E},\boldsymbol{H})$ in our coordinate frame is obtained as

$$\boldsymbol{\eta}(\boldsymbol{E},\boldsymbol{H}) = (p/\omega^P)\left(\boldsymbol{\Delta}+\frac{(\omega^{AP})^2}{\omega^P\omega^A}\,\mathscr{A}(\boldsymbol{E},\boldsymbol{H})\right), \tag{11}$$

where p is the pressure and $\boldsymbol{\Delta}$ is the fourth rank isotropic unit tensor

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

For $H=0$ or $E=0$ the well known formulae for the electric [5] and magnetic [4] Senftleben-Beenakker effect are recovered. From Eq. (11) the relative change of the viscosity in perpendicular electric and magnetic fields can be inferred as

$$\begin{aligned}
\Delta\boldsymbol{\eta}/\eta = &-((\omega^{AP})^2/\omega^P\omega^A)\\
&\cdot(\mathscr{A}(E=0,H=0)-\mathscr{A}(\boldsymbol{E},\boldsymbol{H}))\,.
\end{aligned} \tag{12}$$

Of particular experimental interest is the influence of the electric field on the transverse viscomagnetic pressure difference. For a rectangular channel with linear dimensions $L_x, L_y \gg L_z$, a gas flow $\boldsymbol{v}$ in x-direction ($\boldsymbol{H}$ in z-direction and $\boldsymbol{E}$ in y-direction) the velocity gradient tensor is $\boldsymbol{\nabla v}\approx\partial v/\partial z\,\boldsymbol{e}_x\boldsymbol{e}_z$. With the help of the Navier Stokes equation a transverse pressure difference $(\delta p)_y$ due to an applied pressure difference $(\delta p)_x$ in x-direction can be calculated from Eqs. (8), (11):

$$\begin{aligned}
(\delta p)_y = &-((\delta p)_x/L_x)\,L_y\,\frac{(\omega^{AP})^2}{\omega^P\omega^A}\,\varphi_H\\
&\cdot[(1+\varphi_E{}^2)(1+4\varphi_E{}^2)+\varphi_H{}^2]^{-1}\,.
\end{aligned} \tag{13}$$

For an experimental setup with $L_x=35$ mm, $L_y=15$ mm and a pressure $p=1$ torr, $(\delta p)_y/(\delta p)_x$ is calculated for the gas NF_3 as function of H with E as parameter. Values for φ_H/H and $(\omega^{AP})^2/\omega^P\omega^A$ are taken from van Ditzhuysen's measurements [10] on the magnetic Senftleben-Beenakker effect of polar gases while φ_E/E is inferred from the experiments [12] on the electric Senftleben-Beenakker effect of viscosity. The result is shown in Figure 1. With increasing E, the magnitude of the viscomagnetic transverse pressure difference decreases and at the same time the position of the maximum is shifted towards higher H-values. This decrease is obviously due to the additional precission of the molecules in the electric field which gives rise to a further destruction of tensor polarization.

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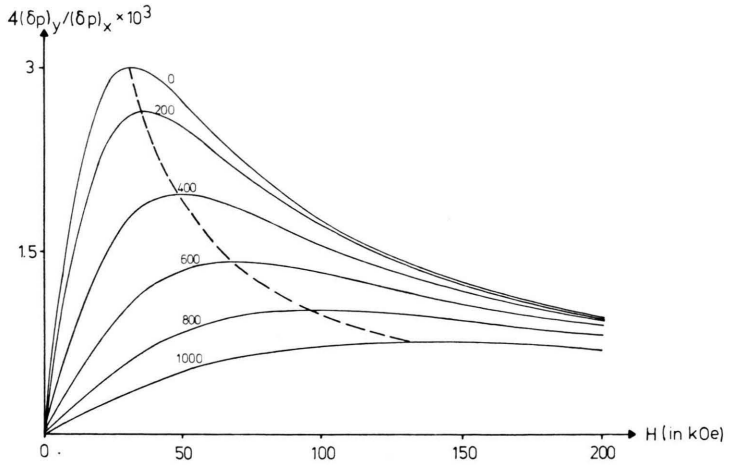

Fig. 1. The dimensionless transverse viscomagnetic pressure difference $4\,(\delta p)_y/(\delta p)_x$ for NF_3 at 1 torr and 300 K as function of magnetic field strength H_z (in kOe) for several values of the electric field strength E_y (in V/cm).

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