Rotation of polarization plane of light in highly rarefied nonequilibrium gas

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The rotation of the polarization plane of light in a nonequilibrium, highly rarefied molecular gas between two plane-parallel solid surfaces is analyzed. The effect is attributed to Coriolis forces which act on the electrons in a rotating molecule and to the onset of a vector polarization along the directions of the angular momenta of the rotation of the molecules as they are scattered by a surface. The role of gyration vector is played by the pseudovector representing the rotational angular momentum of the gas molecules. The deviation of the gas from equilibrium is assumed to be caused by one of three factors: a difference between the temperatures of the surfaces, a difference between the gas pressures at the ends of the gap, and a translational motion of the surfaces with respect to each other, while the distance between them remains constant. For these different types of deviation from equilibrium, the rotation of the polarization plane of the light is caused by scattering processes which differ in spatial symmetry. The polarization-plane rotation can be utilized to study a nonspherical scattering of molecules by a solid surface. Imposing a weak magnetic field causes the angle through which the polarization plane is rotated to become a function of the coordinate of the beam axis in the gap, by virtue of a precession mechanism by which the field acts on the moments of the molecule.

I. INTRODUCTION

The distribution function of a nonequilibrium gas consisting of molecules having rotational degrees of freedom is anisotropic in terms of the directions of both the velocity **v** and the rotational moment **M** of the molecules (Ref. 1). The anisotropy of the distribution of the molecular gas with respect to the orientations of the moment of the molecules causes the dielectric constant of the gas to become a tensor, and the nonequilibrium gas becomes an optically anisotropic medium. One manifestation of optical anisotropy of a nonequilibrium gas is birefringence. This effect has been observed in flowing gaseous CO_2 (Ref. 2) and is described theoretically in Ref. 3. The birefringence in a nonequilibrium Knudsen gas was studied in Ref. 4. In contrast with a dense gas, in which an anisotropic distribution of the orientations of molecules is established as a result of nonspherical intermolecular collisions, a polarization of the rotational moments in a Knudsen gas arises as a result of nonspherical collisions of the molecules with a surface. Since the birefringence is determined by the symmetric part of the dielectric tensor, it is sensitive to the existence of a polarization of the rotational moments, described by a symmetric second-rank tensor, in the gas.

If a vector polarization of the rotational moments of the molecules arises in a nonequilibrium gas, the gas becomes a gyrotropic medium, capable of rotating the plane of polarization of light. We know (Ref. 5, for example) that for most gases intermolecular collisions lead to a tensor polarization of the rotational moments. Exceptional cases are highly polar molecules, for which there is also a vector polarization. Collisions of molecules with a surface lead to both a tensor polarization and a vector polarization of the rotational moments.[6] In the present paper we analyze the rotation of the polarization plane of light in a nonequilibrium Knudsen molecular gas between two plane-parallel surfaces.

The polarization-plane rotation is described by the antisymmetric imaginary part of the dielectric tensor, which is the dual of the pseudovector **g**, which is called "gyration vector."[7] In the Faraday effect the role of the gyration vector is played by the external magnetic field **B**, and the effect itself is a consequence of a dispersion of the polarizability of the molecules and of Lorentz forces which act on the electrons moving in the molecules in the field **B**. In the case $\mathbf{B} = 0$ the role of the gyration vector in a nonequilibrium molecular gas can be played by the vector average rotational moment of the molecules, $\langle \mathbf{M} \rangle$, which, like **B**, is a pseudovector, while the role of the Lorentz forces is played by Coriolis forces. Incorporating the Coriolis forces which exist during the rotation of molecules leads to an antisymmetric part of the tensor describing the scattering of light by nondegenerate molecules.[8] In an equilibrium gas, the distribution of the molecules with respect to rotational moment **M** is isotropic, and there is no polarization-plane rotation. In a nonequilibrium gas, as we will show below, it is possible to arrange conditions corresponding to $\langle \mathbf{M} \rangle \neq 0$. The appearance of a nonvanishing average moment with $\mathbf{B} = 0$ in a Knudsen molecular gas between two plane-parallel surfaces at different temperatures ("thermopolarization") has been studied and described previously.[9] In contrast with the known polarization-plane rotation in a rotating medium,[7] we will be discussing here types of deviations from equilibrium which do not lead to a rotation of the gas as a whole.

There are several reasons for the interest in research on polarization-plane rotation in a Knudsen gas. This effect arises in the first approximation in the small parameter μ, which represents the deviation of the molecules from a spherical shape, and it can be studied by advanced methods, as estimates show. In contrast with the gas of a continuous medium, in which both the kinetic and optical properties are determined by local values of macroscopic quantities and their derivatives, the corresponding effects in a Knudsen gas also depend on the geometry of the system. The polarization-plane rotation exhibits its most interesting features in a weak magnetic field, in which case the magnetic polarization of the molecules can be ignored ($B = 10^2$–10^3 G, $T \approx 300$ K) in comparison with the polarization which arises upon deviation from equilibrium. Such a field causes only a change in

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the orientation of the moments of the molecules by virtue of their precession around the direction of the vector **B**, and the moment of the molecules changes orientation as they move freely in the gap from wall to wall. Because of this effect, the polarization-plane rotation depends on the coordinate of the beam in the gap, as does the birefringence.[4] The precession gives rise to a vector polarization in the plane perpendicular to the field direction. This polarization depends on the strength of the field. The characteristics of the polarization-plane rotation depend strongly not only on the nature of the deviation from equilibrium but also on whether the deviation from equilibrium constitutes an ordinary vector or a pseudovector. The latter, along with the spatial-inversion symmetry properties of the nonspherical part of the probability for the scattering of molecules by the surface, determines the nature of the inhomogeneity which arises in the optical properties of the gas.

Note that optical phenomena in a nonequilibrium gas can serve as a unique tool for identifying the contributions of the vector polarization (polarization-plane rotation) or tensor polarization (birefringence) of the rotational moments of molecules, while transport in a field[10] provides information which is summed over all types of polarizations.

In this paper we describe research into these features of polarization-plane rotation in a collisionless nonequilibrium molecular gas. The effect is analyzed on the basis of symmetry considerations regarding the system. In particular, the polarization-plane rotation is studied in the cases in which the deviation from equilibrium is caused by different factors: a difference between the temperatures of the surfaces, a flow of the gas along the gap, and a motion of the surfaces with respect to each other.

2. KINETIC DESCRIPTION OF POLARIZATION-PLANE ROTATION

The polarizability tensor α_{ik} of a rotating molecule, averaged over possible orientations of its axis with a given direction of its rotational moment, is[11]

$$\alpha_{ik}=\alpha_0\delta_{ik}+i\alpha_1 e_{ikl}m_l+\alpha_2(m_i m_k-{}^1/_3\delta_{ik}), \tag{1}$$

where $\mathbf{m}=\mathbf{M}/M$. The scalar ($\sim\alpha_0$) and symmetric ($\sim\alpha_2$) parts of the polarizability tensor are related to the geometric structure of the molecule, while the antisymmetric part ($\sim\alpha_1$) is caused by Coriolis forces and the dispersion of the polarizability tensor.[8] For most molecules we would have[11] $\alpha_1/\alpha_0\sim10^{-3}$–$10^{-4}$.

The dielectric tensor of a nonequilibrium rarefied gas can be written in the form $\varepsilon_{ik}=\varepsilon_{ik}^s+i\varepsilon_{ik}^a$, where the antisymmetric part is $\varepsilon_{ik}^a=e_{ikl}g_l$, and the gyration vector **g** is given by

$$\mathbf{g}=4\pi N\langle\alpha_1\mathbf{m}\rangle. \tag{2}$$

Here N is the density of molecules, and the angle brackets $\langle\ldots\rangle$ mean an average with the nonequilibrium distribution function $f(\mathbf{v},\mathbf{M})$ of the gas. The symmetry of the tensor ε_{ik} under the condition of a simultaneous change in the sign of magnetic field **B** in accordance with the Onsager relation $\varepsilon_{ik}(\mathbf{B})=\varepsilon_{ik}(-\mathbf{B})$ holds for only a medium at thermodynamic equilibrium.[12] With $\mathbf{B}=0$, the appearance of an antisymmetric part of ε_{ik} would thus be possible in only nonequilibrium systems. The quantity α_1 is a function of the rotational moment **M** and of the light frequency ω. The expression for g_l is conveniently written in the form

$$g_l=4\pi Nt_l{}^m\varphi_{1m},\qquad \varphi_{1m}=\int\alpha_1(M^2,\omega)Y_{1m}(\mathbf{m})f(\mathbf{v},\mathbf{M})\,d\mathbf{v}\,d\mathbf{M}, \tag{3}$$

where t_l^m are coefficients which determine the relationship between the components of the vector **M** and the equivalent spherical harmonics $Y_{1m}(\mathbf{m})$ (Ref. 13). The behavior of the polarization-plane rotation as a function of the frequency, ω, of the light wave, is determined (as in the Faraday effect) by the frequency dispersion of the polarizability $\alpha(\omega)$ of the molecules. In expression (1) we have $\alpha_1\sim\partial\alpha/\partial\omega$.

The angle (φ) through which the polarization plane of the light is rotated over a path length l in the gas is

$$\varphi=\frac{1}{2}\int\mathbf{gk}\,dl, \tag{4}$$

where **k** is the wave vector.

Our basic task in calculating the gyration vector **g** is thus to determine the nonequilibrium distribution function $f(\mathbf{v},\mathbf{M})$. In the present paper we examine the polarization-plane rotation in a collisionless molecular gas between two-plane parallel surfaces at $z=-L/2$ and $z=L/2$, where L is the distance between the surfaces. The distribution function $f(\mathbf{v},\mathbf{M})$ is found from the solution of the Boltzmann equation for a collisionless gas and the system of integral boundary conditions at each surface. The kernel of the integral boundary conditions is the probability for the scattering of molecules having rotational degrees of freedom by the surface: $W(\mathbf{v}',\mathbf{M}'\rightarrow\mathbf{v},\mathbf{M})$. To distinguish among the scattering processes which differ in spatial symmetry with respect to $\mathbf{M}'$ and **M** and to determine specifically which of these processes make nonzero contributions to the polarization-plane rotation, we expand W as an expansion in spherical harmonics of $\mathbf{M}'$ and **M**:

$$W=\sum_{lml'm'}W_{l'm'lm}(\mathbf{v}',\mathbf{M}'^2\rightarrow\mathbf{v},\mathbf{M}^2)Y_{l'm'}(\mathbf{M}')Y_{lm}(\mathbf{M}), \tag{5}$$

where if at least one of the indices l', l is nonzero we have $W_{l'm'lm}\sim\mu W_{0000}$, where $\mu^2\sim10^{-3}$ (Ref. 14).

3. SYMMETRY PROPERTIES

Whether a polarization-plane rotation can occur and the general properties of this effect (e.g., whether it is of even or odd parity with respect to the field) can be resolved on the basis of the spatial symmetry of the system. For this purpose, we construct a phenomenological expression for a gyration pseudovector from all the vectors of the problem. First, we have the macroscopic inhomogeneity **X**, which is either a vector (in the case of a thermal or density deviation from equilibrium) or pseudovector (if the deviation from equilibrium is caused by a motion of the surfaces). If, for example, the temperatures of the surfaces differ by ΔT, then the vector **X** is $\Delta T\mathbf{e}_z$ and is directed along the z axis, away from the colder surface toward the warmer ($\mathbf{e}_z$ is a unit vector along the z axis). If there is a gradient in the density (N) of the molecules along the gap, we have the vector $\mathbf{X}=\nabla N$. If the surfaces are moving in different directions at a velocity **u** which is perpendicular to the vector **n** (the normal to the surface), we have a pseudovector $\mathbf{X}=[\mathbf{u}\times\mathbf{n}]$ (Ref. 4). We will be assuming that the direction of the density gradient and the direction of the velocity of one of the surfaces are along the y axis.

In a Knudsen gas in a plane gap there is a physically distinct direction: that normal to the surfaces, along, say, the vector **n** for one of the surfaces. If the surfaces are identical, the gyration vector is not affected by an interchange of the surfaces (by which we mean a replacement of **n** by $-\mathbf{n}$ without changes in the directions of any other vectors of the problem). In the opposite case, combinations of vectors which are of odd parity with respect to **n** can also contribute to the expression for the gyration vector. When a magnetic field is turned on, the direction of the pseudovector **B** becomes physically preferred. In this case we should take account of the spatial inhomogeneity of the optical properties of the collisionless gas, which results from the change (due to precession in the field) in the orientations of the molecules during their free motion between the surfaces. When a field is applied to a plane gap there is a spatial inhomogeneity of the optical properties of the gas along the z axis perpendicular to the surfaces; i.e., the gyration vector depends on the vector $\mathbf{Z} = z\mathbf{e}_z$ which characteristizes the position of the axis of the light beam in the gap. The origin for the vector **Z** is conveniently put in the middle of the gap.

According to (3), the components of the gyration vector can be expressed in terms of the components of a spherical first-rank tensor φ_{1m}. A general expression for φ_{1m}, expanded in irreducible tensors constructed from the components of the vectors of the problem, can be written in the form

$$\varphi_{1m} = \sum_{p_t P_t} \beta_r \sum_{q_t Q_t} Y_{p_1 q_1}(\mathbf{X})\, Y_{p_2 q_2}(\mathbf{n}) \langle p_1 q_1 p_2 q_2 | P_1 Q_1 \rangle \\ \times Y_{p_3 q_3}(\mathbf{B})\, Y_{p_4 q_4}(\mathbf{Z}) \\ \times \langle p_3 q_3 p_4 q_4 | P_2 Q_2 \rangle \langle P_1 Q_1 P_2 Q_2 | 1m \rangle. \qquad (6)$$

Here the $\langle ... \rangle$ are Clebsch-Gordan coefficients, and $r = (p_1, p_2, p_3, p_4, P_1, P_2)$ is the set of indices over which the summation is carried out. The expansion coefficients β_r depend on the absolute values of the vectors **X**, **B**, and **Z** and are determined by the nonspherical part of the probability for the scattering of molecules by the surface. If the probability W is invariant under spatial inversion, then the β_r are scalars; in the opposite case, they are pseudoscalars. The reason is that the polarization of the gas along **M** arises upon a single scattering of gas molecules by the surface, so that the tensor φ_{1m} is nonvanishing even in the first approximation in the small parameter μ (which represents the deviation of the molecules from a spherical shape) and is linear in W_1.

In the coordinate system which we have selected, the vectors **n** and **Z** are collinear with the z axis, so we have $q_2 = q_4 = 0$ in expansion (6). We restrict the analysis to the first approximation in the deviation from equilibrium, **X**, and we set $p_1 = 1$. Expression (6) can then be rewritten as

$$\varphi_{1m} = \sum_{p_t P_t} \beta_r \sum_{q_t Q_t} Y_{1 q_1}(\mathbf{X})\, Y_{p_2 0}(\mathbf{n}) \langle 1 q_1 p_2 0 | P_1 q_1 \rangle Y_{p_3 q_3}(\mathbf{B})\, Y_{p_4 0}(\mathbf{Z}) \\ \times \langle p_3 q_3 p_4 0 | P_2 q_2 \rangle \langle P_1 q_1 P_2 q_2 | 1m \rangle. \qquad (7)$$

If the surfaces are identical, p_2 can be only even; if they are different (i.e., if the probabilities for the scattering of molecules by the surfaces are different), it can also be odd.

TABLE I. $\mathbf{B}=0$

	g_x	g_y	g_z
$[\mathbf{un}] \parallel \mathbf{e}_x$	W_+S_+	W_-S_-	0
$\nabla N \parallel \mathbf{e}_y$	W_+S_-	W_-S_+	0
$\Delta T \mathbf{e}_z$	0	0	W_-S_+

In the absence of a magnetic field we should set $p_3 = p_4 = 0$ in (7), since the spatial inhomogeneity of the effect along z also disappears when the field is turned off. With $\mathbf{B} = 0$, expression (7) thus takes the form

$$\varphi_{1m} = \sum_{p_2} \beta_r Y_{1m}(\mathbf{X})\, Y_{p_2 0}(\mathbf{n}) \langle 1 m p_2 0 | 1m \rangle. \qquad (8)$$

The possible appearance of $\langle \mathbf{M} \rangle \neq 0$ ($\varphi_{1m} \propto \langle \mathbf{M} \rangle$) in the absence of an external magnetic field was first pointed out in Ref. 9.

Table I shows the results of an analysis of the polarization-plane rotation carried out through the use of expansion (8) for three types of deviation from equilibrium. For $\mathbf{X} = \Delta T \mathbf{e}_z$, for example, we have a gyration vector $\mathbf{g} \neq 0$ only if the scattering probability is invariant under inversion (W_-) and the surfaces are identical (S_+); in this case we have $\mathbf{g} \| \mathbf{X}$. The existence of terms in expansion (5) of the probability W which are not invariant under spatial inversion was discussed in Ref. 6. For an $\mathbf{X} = \nabla N$ parallel to $\mathbf{e}_y$ we have a vector $\mathbf{g} \neq 0$ in two cases: if the scattering probability is invariant under inversion (W_+) and the surfaces are different (S_-), or if the probability is not invariant under inversion and the surfaces are identical. In the first of these cases we have $\mathbf{g} \parallel \mathbf{e}_x$, i.e., $\mathbf{g} \perp \mathbf{X}$, and in the second $\mathbf{g} \parallel \mathbf{e}_y$, i.e., $\mathbf{g} \parallel \mathbf{X}$. For $\mathbf{X} = [\mathbf{u} \times \mathbf{n}]$ parallel to $\mathbf{e}_x$, we again have $\mathbf{g} \neq 0$ in two cases: if the scattering probability is invariant under inversion and the surfaces are identical ($\mathbf{g} \parallel \mathbf{e}_x$, i.e., $\mathbf{g} \parallel \mathbf{X}$); if the probability is not invariant under inversion and the surfaces are different ($\mathbf{g} \parallel \mathbf{e}_y$, i.e., $\mathbf{g} \perp \mathbf{X}$).

In a magnetic field, the possibilities for the occurrence of a polarization-plane rotation are considerably richer. Table II shows the results of an analysis of these possibilities found through the use of expansion (7) for $\mathbf{X} = [\mathbf{u} \times \mathbf{n}]$; Table III shows the corresponding results for $\mathbf{X} = \nabla N$; and Table IV shows the corresponding results for $\mathbf{X} = \Delta T \mathbf{e}_z$. A note is in order here. Generally speaking, expansion (7) takes no account of the mechanism by which the magnetic field influences the polarization-plane rotation. A precession mechanism could give rise to a gyration vector $\mathbf{g}_B \perp \mathbf{B}$ which, along with $\mathbf{g}_0$ (in the case $\mathbf{B} = 0$), would make up the

TABLE II. $[\mathbf{un}] \parallel \mathbf{e}_x$

	g_x	g_y	g_z
B_x	W_+S_+	$W_-S_+B_+Z_-$ $W_-S_-B_+Z_+$	$W_-S_+B_-Z_-$ $W_-S_-B_-Z_+$
B_y	$W_+S_+B_+Z_+$ $W_+S_-B_+Z_-$	W_-S_-	$W_+S_+B_-Z_+$ $W_+S_-B_-Z_-$
B_z	$W_+S_+B_+Z_+$ $W_+S_-B_+Z_-$ $W_-S_+B_-Z_-$ $W_-S_-B_-Z_+$	$W_+S_+B_-Z_+$ $W_+S_-B_-Z_-$ $W_-S_+B_+Z_-$ $W_-S_-B_+Z_+$	0

TABLE III. $\nabla N \parallel \mathbf{e}_y$

	g_x	g_y	g_z
B_x	W_+S_-	$W_-S_+B_+Z_+$ $W_-S_-B_+Z_-$	$W_-S_-B_-Z_+$ $W_-S_-B_-Z_-$
B_y	$W_+S_+B_+Z_-$ $W_+S_-B_+Z_+$	W_-S_+	$W_+S_+B_-Z_-$ $W_+S_-B_-Z_+$
B_z	$W_+S_+B_+Z_-$ $W_+S_-B_+Z_+$ $W_-S_+B_-Z_+$ $W_-S_-B_-Z_-$	$W_+S_+B_-Z_-$ $W_+S_-B_-Z_+$ $W_-S_+B_+Z_+$ $W_-S_-B_+Z_-$	0

total vector **g**. The terms in (7) which lead to a vector $\mathbf{g}_B \| B$ should thus be eliminated. The reason is that in the kinetic calculation below we will take account of only the precession mechanism for the effect of the field on the polarization-plane rotation, which operates under the condition $\mu B \ll k_B T$ ($T \approx 300$ K, $B \leqslant 10^3$ G). Magnetic polarization of the molecules—which we are not discussing here—leads to a $\mathbf{g}_B \| \mathbf{B}$.

Summarizing the tabulated results, we note that under conditions such that we have a gyration vector $\mathbf{g}_0 \neq 0$ in the absence of a field the application of a field **B** which is not parallel to $\mathbf{g}_0$ results in the appearance of (in addition to $\mathbf{g}_0$) a vector $\mathbf{g}'_B$, which has two components which are perpendicular to each other and to the vector **B**. One of these components, which lies in the plane of $\mathbf{g}_0$ and **B**, is even in the field (B_+), while the other, which is perpendicular to this plane, is odd in the field (B_-). Both are even with respect to z (Z_+). According to the Onsager relation, the imaginary antisymmetric part of the tensor ε_{ik} should be odd in **B** (Ref. 12). This property, however, holds only for a medium in thermodynamic equilibrium. The tensor ε_{ik} has terms which are both odd and even with respect to **B** because of the deviation of the system from equilibrium. Furthermore, a magnetic field lifts the restrictions on the symmetry properties of the system with respect to interchange of the surfaces. The resultant additional gyration vector $\mathbf{g}''_B$ has for an interchange of the surfaces symmetry properties which differ from those of $\mathbf{g}_0$ and $\mathbf{g}'_B$. Like $\mathbf{g}'_B$ it has two components in the plane perpendicular to the vector **B**. Like $\mathbf{g}'_B$, the component of the vector $\mathbf{g}''_B$ which is even in the field lies in the plane of $\mathbf{g}_0$ and **B**, while the odd component is perpendicular to this plane. In contrast with $\mathbf{g}_B$ however, both components of $\mathbf{g}''_B$ are odd along $z(Z_-)$. It should be kept in mind here that as light propagates along the z axis the contribution of the components of the gyration vector **g** which are odd in z, and also the polarization-plane rotation, turn out to be zero because of the integration over z in (4). If, on the other hand, the beam is directed along the gap (perpendicular to $\mathbf{e}_z$), then the contribution of $\mathbf{g}''_B$ to the effect may also vanish, because of the finite thickness of the beam and the ensuing need for averaging over z in this case. Generally speaking, a spatial inhomogeneity of the polarization-plane rotation in a beam of finite thickness results in a depolarization of the light.

TABLE IV. $\Delta T \mathbf{e}_z$

	g_x	g_y	g_z
B_x	0	$W_-S_+B_-Z_-$ $W_-S_-B_-Z_+$	$W_-S_-B_+Z_+$ $W_-S_-B_+Z_-$
B_y	$W_-S_+B_-Z_+$ $W_-S_-B_-Z_-$	0	$W_-S_-B_+Z_+$ $W_-S_-B_+Z_-$
B_z	0	0	W_-S_+

4. POLARIZATION-PLANE ROTATION FOR THE CASE OF SURFACES AT DIFFERENT TEMPERATURES

The problem of determining nonequilibrium function f for the case in which the surfaces have different temperatures (T and $T + \Delta T$) was solved in Ref. 10. The dependence of f on **M** arises in this case even in the first approximation in the parameter μ (a measure of the deviation from a spherical shape). Also restricting the present discussion to the linear approximation in $\Delta T / T$, we write an expression for φ_{1m}:

$$\varphi_{1m} = \frac{1}{2\pi^{1/2}} \frac{\Delta T}{k_B T^2} \sum_p (-1)^m D^{1*}_{p-m}(\Omega_B) D_{p0}^{1}(\Omega_B)$$
$$\times \int\limits_{v_z\, v_z' \gtrless 0} \int d\Gamma\, d\Gamma'\, f_M(E) E' \alpha_1(\omega, M^2) R_{p0}(\Gamma, \Gamma', B, z), \tag{9}$$
$$R_{p0} = W^{+}_{1000}(-\Gamma, -\Gamma_3', \mathbf{n}) \exp[ip\gamma B(L/2+z)/v_z]$$
$$- W^{-}_{1000}(-\Gamma_3, -\Gamma', -\mathbf{n}) \exp[ip\gamma B(L/2-z)/v_z].$$

Here D^{1}_{p-m} are the Wigner rotation functions,[13] Ω_B are the Euler angles which specify the orientation of the coordinate system (x',y',z') associated with the field **B** ($z' \parallel \mathbf{B}$), $f_M(E)$ is a Maxwellian distribution, E is the energy of the molecule, $\Gamma = (\mathbf{v}, M)$, $\Gamma_3 = (\mathbf{v}_3 M)$, $\mathbf{v}_3 = \mathbf{v} - 2\mathbf{n}(\mathbf{vn})$, $d\Gamma = d\mathbf{v} M dM$, γ is the gyromagnetic ratio of the molecule, and W^+ and W^- refer to scattering by the lower and upper surfaces, respectively.

As in Ref. 10, we will be using the following expansion for W_{lk00}:

$$W^{\pm}_{lk00}(\Gamma, \Gamma', \mathbf{n}) = |v_z'| f_M(E) \sum_s A_{ls}^{\pm} \left(\frac{M'}{\langle M \rangle}\right)^l \begin{pmatrix} L_1 & l & l_3 \\ -k & k & 0 \end{pmatrix}$$
$$\times Y_{l_3 0}(\mathbf{n}) \sum_{m_1' m_1} \langle l_1' m_1' l_1 m_1 | L_1 - k \rangle Y_{l_1' m_1'}(\mathbf{v}') Y_{l_1 m_1}(\mathbf{v}), \tag{10}$$

where $s = (L_1, l_3, l_1', l_1)$, $\langle M \rangle$ is the average value of the rotational moment, (:::) are the Wigner $3j$ symbols, and we are assuming that the coefficients A_{ls} are constants.

We break W_{1000} up into two parts: C_{10} and H_{10}. The first is invariant under spatial inversion, while the second changes sign:

$$\begin{aligned} W^{+}_{1000}(-\Gamma, -\Gamma_3', \mathbf{n}) &= C_{10}^{+}(\Gamma_3, \Gamma', \mathbf{n}) + H_{10}^{+}(\Gamma_3, \Gamma', \mathbf{n}), \\ W^{-}_{1000}(-\Gamma_3, -\Gamma', -\mathbf{n}) &= C_{10}^{-}(\Gamma_3, \Gamma', \mathbf{n}) + H_{10}^{-}(\Gamma_3, \Gamma', \mathbf{n}). \end{aligned} \tag{11}$$

Using expansion (10) and the symmetry properties of the spherical harmonics, we see that the indices $l_1' + l_1 + l_3$ and $m_1' + m_1$ for C_{10} are even, while those for H_{10} are respectively odd and even. It is not difficult to verify that the terms C_{10} do not contribute to expression (9), by virtue of the integration over the angles specifying the orientations of the vectors **v** and **v**′ in the xy plane and the symmetry properties of the $3j$ symbols. A polarization-plane rotation is therefore possible in the case of different surface temperatures only if the probability for the scattering of the molecules is not invariant under spatial inversion.

In the absence of a field ($B = 0$) the tensor φ_{1m} is not zero if the surfaces are identical (in the sense $H_{10}{}^{+} = H_{10}{}^{-}$); on the contrary, we have $\varphi_{1m} = 0$ if $H_{10}{}^{+} = -H_{10}{}^{-}$, i.e., if the surfaces are different. Furthermore, in the absence of a field the tensor φ_{1m} degenerates to φ_{10}, so the direction of the gyration vector $\mathbf{g}_0$ is the z axis. When a field is applied, the tensor φ_{1m} is nonzero not only in the case $H_{10}{}^{+} = H_{10}{}^{-}$ but also in the case $H_{10}{}^{+} = -H_{10}{}^{-}$. In the former case φ_{1m} and thus the gyration vector $\mathbf{g}$ are even functions of the coordinate z, while in the latter case they are odd.

In the case of identical surfaces the gyration vector can be written as follows according to (3) and (9):

$$\mathbf{g}=\mathbf{g}_0+\mathbf{g}^{-}+\mathbf{g}^{+}=g_{\Delta T}(\mathbf{e}_z+[\mathbf{e}_z\mathbf{b}]I_1+[[\mathbf{e}_z\mathbf{b}]\mathbf{b}]I^2),$$

$$I_1=\frac{1}{I_0}\int\limits_0^\infty P_{l_1}(x)\exp(-x^2)\sin\frac{\Omega\tau}{2x}\cos\frac{\Omega t}{2x}\,dx, \tag{12}$$

$$I_2=\frac{1}{I_0}\int\limits_0^\infty P_{l_1}(x)\exp(-x^2)\left(1-\cos\frac{\Omega\tau}{2x}\cos\frac{\Omega t}{2x}\right)dx,$$

$$I_0=\int\limits_0^\infty P_{l_1}(x)\exp(-x^2)\,dx,$$

where $\mathbf{b} = \mathbf{B}/B$, $x = v_z(m/2k_BT)^{1/2}$, $\Omega = \gamma B$, $\tau = L(m/2k_BT)^{1/2}$, $t = z(m/2k_BT)^{1/2}$ and $P_{l_1}(x)$ is a polynomial of degree l_1. For the very simple model W_{1000}, for which only the terms with $l_1 = 0$ are nonzero in expansion (10), the polynomial $P_0(x)$ is unity.

It follows from (12) that the gyration vector $\mathbf{g}$ has three components, one of which ($\mathbf{g}_0$) does not depend on the field and is directed along the z axis, while the two others do depend on the field. One of the latter two ($\mathbf{g}^{-}$) is perpendicular to the plane formed by the vectors $\mathbf{e}_z$ and $\mathbf{b}$ and is odd in the field; the other ($\mathbf{g}^{+}$), which lies in the plane of $\mathbf{e}_z$ and $\mathbf{b}$ and is perpendicular to $\mathbf{b}$, is even in the field. The results of the kinetic theory thus agree completely with conclusions which follow from asymmetry analysis of the system.

The dependence of the vectors $\mathbf{g}^{+}$ and $\mathbf{g}^{-}$ on the absolute value B of the field, for a given orientation of the field, is a damped oscillation with a characteristic period $\Omega\tau \approx 1$, where Ω is the precession frequency of the rotational moment of the molecule in the field, and τ is the average time taken by a molecule to pass from one surface to the other. A value $\Omega\tau \approx 1$ corresponds to a single precession of $\mathbf{M}$ around the direction of $\mathbf{B}$ as the molecule moves from one surface to the other; $\Omega\tau \approx 2$ corresponds to a twofold precession, etc. The damping of the oscillations is a consequence of the velocity distribution of the molecules. A similar behavior as a function of the absolute value of the field, B, is found in the thermomagnetic effect.[10]

The absolute value of the gyration vector satisfies the proportionality $g_{\Delta T} \sim \mu(\Delta T/T)\alpha_1 N$, and for $\Delta T/T \approx 0.1$ and $N = 10^{19}\ \mathrm{m}^{-3}$ it is 10^{-16} in order of magnitude. Consequently, the angle through which the polarization plane of visible light would rotate over a one-meter path length under these conditions would be $\approx 10^{-9}$ rad. Since the polarization-plane rotation is odd in the macroscopic-inhomogeneity vector of the gas, it can be multiplied by forcing the beam to traverse the same path repeatedly by means of reflections. The polarization-plane rotation would thus be amenable to experimental study by advanced methods, which are capable of measuring polarization-plane rotations within an error of 10^{-9} rad (Ref. 15).

5. POLARIZATION-PLANE ROTATION IN THE PRESENCE OF A DENSITY GRADIENT

Using the expression derived in Ref. 16 for the distribution function f of a gas in the case in which a deviation from equilibrium is caused by a density gradient along the gap ($\nabla N \parallel \mathbf{e}_y$), we can calculate the tensor φ_{1m} which determines the gyration vector $\mathbf{g}$ according to (3):

$$\begin{aligned}\varphi_{1m}&=-\frac{1}{2\pi^2}\frac{L\nabla N}{N}\sum_{p,k}(-1)^m D^{1*}_{p-m}(\Omega_B)D_{pk}{}^1(\Omega_B)\\&\times\int\limits_{v_z}\int\limits_{v_z'\geqslant 0} d\Gamma\,d\Gamma'\,f_M(E)\frac{v_y'}{v_z'}\alpha_1(\omega,M^2)R_{pk}(\Gamma,\Gamma',B,z),\\R_{pk}&=W^{+}_{1k00}(-\Gamma,-\Gamma_3',\mathbf{n})\exp[ipB\gamma(L/2+z)/v_z]\\&+W^{-}_{1k00}(-\Gamma_3,-\Gamma',-\mathbf{n})\exp[ip\gamma B(L/2-z)/v_z].\end{aligned} \tag{13}$$

The index k over which the summation is carried out in (13) takes on only the values ± 1 [as can be verified by integrating (13) over $\mathbf{v}'$ and using expression (10) for W_{1k00}].

It can be seen from (13) that the polarization-plane rotation is caused by a mechanism for the scattering of molecules by the surface ($W_{1k00} \neq 0$) which differs from that in the case of a thermal deviation from equilibrium. The quantities W_{1k00} can be broken up into two parts, K_{1k} and D_{1k}, the first invariant under inversion, while the second changes sign:

$$\begin{aligned}W^{+}_{1k00}(-\Gamma,-\Gamma_3',\mathbf{n})&=-D_{1k}{}^{+}(\Gamma_3,\Gamma',\mathbf{n})-K_{1k}{}^{+}(\Gamma_3,\Gamma',\mathbf{n}),\\W^{-}_{1k00}(-\Gamma_3,-\Gamma',-\mathbf{n})&=-D_{1k}{}^{-}(\Gamma_3,\Gamma',\mathbf{n})+K_{1k}{}^{-}(\Gamma_3,\Gamma',\mathbf{n}).\end{aligned} \tag{14}$$

It is not difficult to see that the indices $l_1' + l_1 + l_3$ and $m_1' + m_1$ in expansion (10) are respectively odd and even for k_{1k}, while for D_{1k} for both are odd.

In the case $B = 0$ and with identical surfaces ($D_{1k}{}^{+} = D_{1k}{}^{-}$, $K_{1k}{}^{+} = K_{1k}{}^{-}$), a rotation of the polarization plane is possible only with a part D_{1k} of the scattering probability which is invariant under inversion. It can be verified that the gyration vector $\mathbf{g}$ is parallel to $\mathbf{e}_y$ in this case. If, on the other hand, the surfaces are different, the part K_{1k} of the scattering probability which is invariant under inversion will contribute to the polarization-plane rotation. The component of $\mathbf{g}$ which arises in this case is directed along the x axis. Setting $m = 0$ in (13), and making use of the unitarity of the Wigner functions, we can verify that we have $\varphi_{10} = 0$, i.e., that $\mathbf{g}$ is perpendicular to $\mathbf{e}_z$.

We now consider the case $\mathbf{B} \neq 0$. It follows from (13) that when a field is applied the polarization-plane rotation becomes spatially nonuniform along z; i.e., the vector polarization of the gas along $\mathbf{M}$ depends on the coordinate z of the point at which the polarization is considered, so the polarization-plane rotation depends on the coordinate z of the beam in the gap. For the case of identical surfaces, the part of the scattering probability which is invariant under inversion leads to an odd dependence of φ_{1m} and thus of $\mathbf{g}$ on z, while the noninvariant part leads to an even dependence. If, on the other hand, the surfaces are different, then the difference $K_{1k}{}^{+} - K_{1k}{}^{-}$, which makes a contribution to φ_{1m} which is even in z, and the difference $D_{1k}{}^{+} - D_{1k}{}^{-}$, which makes an odd contribution, will figure in (13). Comparing these results with the results of the symmetry analysis of the polarization-plane rotation, we find total agreement.

In analyzing the dependence of the polarization-plane rotation on the absolute value and orientation of the field B we assume that the surfaces are identical and that the scattering probability is invariant under inversion ($W_{1k00} = K_{1k}$). The expression for the gyration vector in this case is

$$\mathbf{g}=\mathbf{g}^{-}+\mathbf{g}^{+}=g_{\nabla N}([\mathbf{b}\mathbf{e}_x]I_3+[\mathbf{b}[\mathbf{b}\mathbf{e}_x]]I_4),$$

$$I_3=\frac{1}{I_0}\int_0^\infty xP_{l_1}(x)\exp(-x^2)\cos\frac{\Omega\tau}{2x}\sin\frac{\Omega t}{x}\,dx,$$

$$I_4=\frac{1}{I_0}\int_0^\infty xP_{l_1}(x)\exp(-x^2)\sin\frac{\Omega\tau}{2x}\sin\frac{\Omega t}{x}\,dx, \qquad (15)$$

$$I_0=\int_0^\infty xP_{l_1}(x)\exp(-x^2)\,dx.$$

It follows from (15) that we have $\mathbf{g}^{+}\perp\mathbf{g}^{-}$, $\mathbf{g}^{+}\perp\mathbf{B}$, $\mathbf{g}^{-}\perp\mathbf{B}$, and $\mathbf{g}^{-}\perp\mathbf{e}_x$. Both $\mathbf{g}^{+}$ and $\mathbf{g}^{-}$ are odd with respect to z, while $\mathbf{g}^{+}$ is even and $\mathbf{g}^{-}$ odd with respect to $\mathbf{B}$. With $\mathbf{B}=0$ or $\mathbf{B}\parallel\mathbf{e}_x$, we have $\mathbf{g}=0$. These results agree with results which are shown in Table III and which correspond to the condition $W_{+}S_{+}$. The dependence of $\mathbf{g}$ on the absolute value of the field consists of damped oscillations which correspond to single, twofold, etc., precessions of the rotational moment of the molecules around the field direction over the time taken by the molecules to move between the surfaces. The absolute value of the gyration vector satisfies the proportionality $g_{\nabla N}\sim\mu L\nabla N\alpha_1$; with $N=10^{19}\ \mathrm{m}^{-3}$ and $L\nabla N=0.1N$, it is 10^{-16} in order of magnitude. This result leads to the same estimate of the rotation of the polarization plane as in the case of the thermal deviation from equilibrium.

6. ROTATION OF THE POLARIZATION PLANE IN THE CASE OF A DEVIATION FROM EQUILIBRIUM CAUSED BY A MOTION OF THE SURFACES

In accordance with the procedure of Ref. 4, we determine the distribution function f and, in accordance with (3), the tensor φ_{1m}, which is given in the first approximation in the parameter u/V_T (the measure of the deviation from equilibrium) and in the small parameter μ (the measure of the deviation from a spherical shape) by

$$\varphi_{1m}=-\frac{1}{2\pi^{1/2}}\frac{4u}{v_T{}^2}\sum_{pk}(-1)^m D^{1*}_{p-m}(\Omega_B)D_{pk}{}^1(\Omega_B)$$

$$\times\int\limits_{v_z}\int\limits_{v_z'\geqslant 0} d\Gamma\,d\Gamma'\,f_M(E)\,v_y'\alpha_1(\omega,M^2)F_{pk}(\Gamma,\Gamma',B,z), \qquad (16)$$

$$F_{pk}=\overset{+}{W}_{1k00}(-\Gamma,-\Gamma_3',\mathbf{n})\exp[ip\gamma B(L/2+z)/v_z]$$

$$-\overset{-}{W}_{1k00}(-\Gamma_3,-\Gamma',-\mathbf{n})\exp[ip\gamma B(L/2-z)/v_z].$$

Here we have $k=\pm 1$, as in (13). The same parts of the scattering probability thus make a nonzero contribution to the effect as in the problem involving a density gradient. Note, however, that the symmetry properties of the expression found for the tensor φ_{1m} here differ from those of φ_{1m} of the preceding section of the paper because the vector representing the deviation from equilibrium is a pseudovector when the surfaces are in relative motion. For example, when the vector representing the deviation from equilibrium is replaced by a pseudovector there is a change in the parity of the tensor φ_{1m} with respect to the coordinate z. Because of this point, we will not present the results of the kinetic calculation in the same degree of detail as we did in Sec. 5. We simply note that the results agree completely with the results found from the symmetry analysis. For example, in the case of identical surfaces, with a scattering probability invariant under inversion, the gyration vector is nonzero even with $\mathbf{B}=0$. When a field is imposed, we again find the components $\mathbf{g}^{+}$ and $\mathbf{g}^{-}$ which supplement $\mathbf{g}_0$. The first is even in the field, and the second odd:

$$\mathbf{g}=\mathbf{g}_0+\mathbf{g}^{-}+\mathbf{g}^{+}=g_u(\mathbf{e}_x+[\mathbf{b}\mathbf{e}_x]I_5+[\mathbf{b}[\mathbf{b}\mathbf{e}_x]]I_6),$$

$$I_5=\frac{1}{I_0}\int_0^\infty xP_{l_1}(x)\exp(-x^2)\sin\frac{\Omega\tau}{2x}\cos\frac{\Omega t}{x}\,dx, \qquad (17)$$

$$I_6=\frac{1}{I_0}\int_0^\infty xP_{l_1}(x)\exp(-x^2)\left(1-\cos\frac{\Omega\tau}{2x}\cos\frac{\Omega t}{2x}\right)dx,$$

$$I_0=\int_0^\infty xP_{l_1}(x)\exp(-x^2)\,dx.$$

Here we have $\mathbf{g}^{+}\perp\mathbf{g}^{-}$, $\mathbf{g}^{+}\perp\mathbf{B}$, $\mathbf{g}^{-}\perp\mathbf{B}$, and $\mathbf{g}^{-}\perp\mathbf{e}_x$. Both $\mathbf{g}^{+}$ and $\mathbf{g}^{-}$ are even in z. With $\mathbf{B}=0$ or $\mathbf{B}\parallel\mathbf{e}_x$, the components $\mathbf{g}^{+}$ and $\mathbf{g}^{-}$ are zero. These results correspond to those shown in Table II for the condition $W_{+}S_{+}$. The dependence of $\mathbf{g}^{+}$ and $\mathbf{g}^{-}$ on the absolute value of the field consists of damped oscillations with increasing B. The absolute value of the gyration vector satisfies the proportionality $g_u\sim\mu(u/v_T)\alpha_1 N$. With $u/v_T=0.1$ and $N=10^{19}\ \mathrm{m}^{-3}$ we find the same magnitude for the rotation of the polarization plane of the light as in the preceding cases.

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