Birefringence in a nonequilibrium collisionless gas

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The birefringence in a nonequilibrium collisionless molecular gas in the presence of an external magnetic field is investigated. The nonequilibrium states produced by the temperature difference between the surfaces bounding the gas, the flow of the gas along the gap, and the motion of the surfaces relative to each other are considered. The effect is due to the tensor polarization of the moments of the gas molecules in the gap, and can be used to study nonspherical scattering of molecules by the surface of a solid. It is shown that the birefringence in the cases of the various nonequilibrium states is due to scattering processes with different spatial symmetries. An interesting characteristic that distinguishes the effect in question from the analogous phenomenon that occurs in the gas of a continuous medium is the nonmonotonic dependence of the phase difference on the coordinate of the axis of the ray in the gap in the presence of an external magnetic field.

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1. INTRODUCTION

In the general case, a monochromatic light ray passing through an anisotropic medium splits up into two linearly polarized waves, which propagate with different velocities. The polarization planes of these two waves are mutually perpendicular. This phenomenon is called birefringence of light.[1] The birefringence effect is due to the anisotropy of the permittivity of the medium: this anisotropy either can be an intrinsic property of the material, or it can be produced under the action of mechanical deformations, an electric field (the Kerr effect), or a magnetic field (the Cotton-Mouton effect). The permittivity anisotropy may also be due to the nonequilibrium state of the medium, especially when the medium is a gas. Thus, birefringence has been observed in an inhomogeneous CO_2-gas stream.[2] Theoretically, this effect was first predicted and described by Hess *et al.*[3,4] It is well known that the distribution function for an inhomogeneous gas consisting of molecules with rotational degrees of freedom is anisotropic both with respect to the directions of the velocities v of the molecules and with respect to the directions of the angular momentum vector **M** (Ref. 5). An important consequence of the anisotropy of the distribution function with respect to the directions of the vector M is the possibility of a tensor polarization of the angular-momentum vector, a polarization which in the case of optically anisotropic gas molecules makes the permittivity of the medium anisotropic. The tensor polarization of the vector M occurs as a result of the collisions of the nonspherical molecules with each other.

Birefringence in a highly rarefied gas in which the mean free path $\bar{l}$ of the molecules is much greater than the characteristic dimensions L of the device has been predicted by the present authors.[6] The situation considered is one in which the light propagates between two plane-parallel plates through a collisionless gas. The state of nonequilibrium in the system is assumed to be due to the existence of a temperature difference between the surfaces, or else of a pressure difference between the gap ends, which leads to the flow of the gas along the gap. The investigation of birefringence in a collisionless gas is of interest for a number of reasons. The birefringence effect manifests itself in first-order approximation in the small parameter μ (the nonsphericity parameter of the molecule), and, as the corresponding estimates show, can be investigated by modern methods.[7] In contrast to a continuous-medium gas, in which both the kinetic and the optical properties are determined by the local values of the macroscopic quantities and their derivatives, in a collisionless gas the analogous effects depend also on the geometry of the system. An essentially new property typical of a collisionless gas is the appearance of a macroscopic spatial inhomogeneity of the system upon application of an external magnetic field H. This property is connected with the role played by a magnetic field in the birefringence effect under discussion. Whereas in the known magneto-optical effects the magnetic field either modifies the structure of the levels of the molecules, or gives rise to the polarization of the moments of the molecules, orienting the moments parallel to the field vector H, in this case a weak magnetic field only changes the orientation of the moments of the molecules as a result of their precession about the direction of the H vector. Thus, the moment of a molecule changes its orientation during the free wall-to-wall flight of the molecule in the gap. Because of this, the birefringence effect depends on the coordinate of the ray in the gap. In its turn, the spatial inhomogeneity that is realized depends on the strength and direction of the magnetic field. Another interesting distinctive feature of birefringence in a collisionless gas is that the characteristics of the effect depend significantly not only on the type of nonequilibrium state, but also on whether the vector of the nonequilibrium state is a normal vector or a pseudovector. The latter, together with the behavior of the nonspherical-scattering probability under inversion, determines the character of the macroscopic inhomogeneity that arises in the gas.

In the present paper we establish and investigate the above-noted characteristics of birefringence in a collisionless nonequilibrium molecular gas. The analysis of the birefringence is based on symmetry considerations for the system. In particular, we investigate the birefringence in the case of the nonequilibrium state produced by a temperature difference between the sur-

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faces in the presence of an external magnetic field **H**, as well as the birefringence in the case of a gas flowing along the gap, or in the case in which the surfaces move in opposite directions (the Couette problem).

2. PERMITTIVITY TENSOR OF A NONEQUILIBRIUM COLLISIONLESS MOLECULAR GAS

In an optically anisotropic medium two waves with mutually perpendicular polarization planes, propagating in the same direction **q**, accumulate over a distance l a phase difference

$$\delta=2\pi l(n_1-n_2)/\lambda, \tag{1}$$

where λ is the wavelength of the light, while n_1 and n_2 are the refractive indices for the waves in the medium. Below we shall be interested in precisely the magnitude of the phase difference δ. As is well known,[10] the birefringence effect is determined by the symmetric part ε_{ik} of the permittivity tensor, i.e., $\varepsilon_{ik}=\varepsilon_{ki}$. We shall consider those wavelengths for which the medium is nonabsorbing, i.e., for which the symmetric tensor ε_{ik} is real. With allowance for this, let us write the equation for the refractive indices for waves propagating in the direction q in the form[1]

$$\begin{vmatrix} \varepsilon_{xx}-n^2 & \varepsilon_{xy} & \varepsilon_{xz} & q_x \\ \varepsilon_{xy} & \varepsilon_{yy}-n^2 & \varepsilon_{yz} & q_y \\ \varepsilon_{xz} & \varepsilon_{yz} & \varepsilon_{zz}-n^2 & q_z \\ q_x & q_y & q_z & 0 \end{vmatrix}=0. \tag{2}$$

In the case of a molecular gas we have for ε_{ik} the expression

$$\varepsilon_{ik}=\delta_{ik}+4\pi N\langle\alpha_{ik}\rangle, \tag{3}$$

where N is the number of molecules in 1 cm^3, $\langle\ldots\rangle$ denotes averaging with the nonequilibrium distribution function $f(\mathbf{v},\mathbf{M})$, and α_{ik} is the polarizability tensor for a linear molecule, averaged over the possible orientations of the axis of the molecule for a given direction of the angular momentum **M**:

$$\alpha_{ik}=\bar{\alpha}\delta_{ik}-{}^1/_2\Delta\alpha a\sum_m b_{ik}^m Y_{2m}(\mathbf{M}). \tag{4}$$

Here

$$\bar{\alpha}=(\alpha_\parallel+2\alpha_\perp)/3, \qquad \Delta\alpha=\alpha_\parallel-\alpha_\perp,$$

where $\alpha_\parallel$ and $\alpha_\perp$ are the principal values of the polarizability tensor of the molecule, and $a=2(6\pi/5)^{1/2}/3$. The spherical functions are defined in accordance with Ref. 8. The numbers b_{ik}^m determine the connection between the elements of the symmetric traceless second-rank tensor and the elements of the equivalent spherical tensor Y_{2m} (Ref. 9). According to (3) and (4), ε_{ik} has the form

$$\varepsilon_{ik}=\varepsilon_0\delta_{ik}-{}^1/_2N\Delta\alpha\bar{a}\sum_m b_{ik}^m\varphi_{2m}, \tag{5}$$

$$\varepsilon_0=1+4\pi N\bar{\alpha}, \qquad \bar{a}=4\pi a, \tag{6}$$

where ε_0 is the isotropic part of ε_{ik} and φ_{2m} is the second-rank spherical tensor defined by the expression

$$\varphi_{2m}=\int Y_{2m}(\mathbf{M})f(\mathbf{v},\mathbf{M})\,d\mathbf{v}\,d\mathbf{M}. \tag{7}$$

Thus, the main problem in the computation of the tensor ε_{ik} is the determination of the nonequilibrium distribution function $f(\mathbf{v},\mathbf{M})$ for the gas.

Let us consider a collisionless molecular gas located between two plane-parallel surfaces. We choose the system of coordinates such that the direction of the z axis coincides with the normal **k** to the lower surface. The planes $z=-L/2$ and $z=L/2$ correspond to the lower and upper surfaces. The distribution function $f(\mathbf{v},\mathbf{M})$ is determined by solving the kinetic equation for the collisionless gas

$$\mathbf{v}\frac{\partial f}{\partial \mathbf{r}}+\gamma[\mathbf{M}\times\mathbf{H}]\frac{\partial f}{\partial \mathbf{M}}=0 \tag{8}$$

with boundary conditions at each of the surfaces, which, in the general case, move with velocity $\mathbf{u}\perp\mathbf{k}$ in opposite directions:

$$\begin{gathered} |\mathbf{vk}|f^+(\mathbf{v},\mathbf{M})=\int\limits_{\mathbf{v}'\mathbf{k}<0}|\mathbf{v}'\mathbf{k}|W(\mathbf{v}'-\mathbf{u},\mathbf{M}'\to\mathbf{v}-\mathbf{u},\mathbf{M};\mathbf{k}) \\ \times f^-(\mathbf{v}',\mathbf{M}')d\mathbf{v}'\,d\mathbf{M}' \qquad (z=-L/2), \\ |\mathbf{vk}|f^-(\mathbf{v},\mathbf{M})=\int\limits_{\mathbf{v}'\mathbf{k}>0}|\mathbf{v}'\mathbf{k}|W(\mathbf{v}'+\mathbf{u},\mathbf{M}'\to\mathbf{v}+\mathbf{u},\mathbf{M};-\mathbf{k}) \\ \times f^+(\mathbf{v}',\mathbf{M}')d\mathbf{v}'\,d\mathbf{M}' \qquad (z=L/2), \end{gathered} \tag{9}$$

f^+ and f^- are the distribution functions for the molecules flying respectively upwards and downwards in the gap. The kernel of the integral equations (9) is the probability for scattering of nonspherical molecules with rotational degrees of freedom by the surface of a solid:

$$W(\mathbf{v}',\mathbf{M}'\to\mathbf{v},\mathbf{M};\mathbf{k})=W_0+\mu W_1(\mathbf{v}',\mathbf{M}'\to\mathbf{v},\mathbf{M};\mathbf{k}). \tag{10}$$

Here W_0 is the diffuse part of W, and does not depend on the orientation of the molecule, while μW_1 is that part of W which depends both on the orientations of the angular momenta M′ and M and on the velocities **v**′ and v before and after the collision. In the computations below we shall need the coefficients of the expansion of W_1 in terms of spherical functions of **M**′ and M:

$$W_1=\sum_{lml'm'} w_{l'm'lm}(\mathbf{v}'M'^2\to\mathbf{v},M^2;\mathbf{k})Y_{l'm'}(\mathbf{M}')Y_{lm}(\mathbf{M}). \tag{11}$$

The solution to Eq. (8) can be written in the form

$$f(\mathbf{v},\mathbf{M})=\sum_{lpk}\chi_{lp}(\mathbf{v},M^2)D_{pk}^{l*}(\Omega_H)Y_{lk}(\mathbf{M})\exp(ip\omega z/v_z), \tag{12}$$

where the $\chi_{lp}(\mathbf{v}M^2)$ are unknown functions, which can be determined in our problem in the first approximation in the small parameter μ ($\mu^2\sim10^{-3}$) from the solution to the system of equations (9), the $D_{pk}^l(\Omega_H)$ are the Wigner rotational functions[9], and the Ω_H are the Euler angles, which determine the orientation of the coordinate system $(x'y'z')$ connected with the field H ($z'\parallel$ H). Thus, the formulas (6)–(12) enable us to determine the permittivity tensor ε_{ik} for a given character of the nonequilibrium state. We shall determine the magnitude of the phase difference δ between the waves in accordance with the described solution scheme.

3. THE SYMMETRY OF THE SYSTEM

The properties of the phase difference δ are determined by the properties of the anisotropic part of the permittivity tensor of the medium. This part in turn on the symmetry of the system under consideration.

The object of the present section is to analyze the birefringence effect on the basis of an analysis of the symmetry of the system. For this purpose we construct a second-rank spherical tensor φ_{2m} from the vectors of the problem [see (7)]. The elements of the anisotropic part of the tensor ε_{ik} are linearly connected with the elements of the tensor φ_{2m} [see (5)]. The following vectors are involved in the problem: the vector **H**, the normal **k** to the lower surface (the surfaces can be made of different materials), the vector **X** characterizing the state of nonequilibrium in the system, and the vector $\mathbf{z} \parallel z$ characterizing the position of the ray in the gap. The point in introducing **z** lies in the fact that for $\mathbf{H} \neq 0$ the birefringence effect depends on the coordinate of the axis of the ray in the gap. Let us locate the origin of the vector **z** at the center of the gap and the terminus of the vector at the point through which the axis of the ray passes; **k** and **z** are vectors, **H** is a pseudovector, and **X** can be a vector or a pseudovector. Thus, if the nonequilibrium state is created by the motion of the surfaces in opposite directions with velocity $\mathbf{u} \perp \mathbf{k}$, then **X** is a pseudovector perpendicular to the vectors **u** and **k**. In the case of a gas flowing along the gap, **X** is a vector whose direction coincides with the direction of the flow. If, on the other hand, the nonequilibrium state is created by a temperature difference between the surfaces, then **X** is likewise a vector directed, say, from the surface with the lower temperature toward the surface with the higher temperature along the z axis. The general expression for the spherical tensor φ_{2m}, as expanded in terms of the irreducible tensors constructed from the components of the vectors of the problem, can be written in the form[9]

$$\varphi_{2m}=\sum_{s} \beta_s(H,X,z) \sum_{m_iM_j} Y_{l_1m_1}(\mathbf{H})$$
$$\times Y_{l_2m_2}(\mathbf{k})\langle l_1m_1l_2m_2|L_1M_1\rangle Y_{l_3m_3}(\mathbf{X})$$
$$\times Y_{l_4m_4}(\mathbf{z})\langle l_3m_3l_4m_4|L_2M_2\rangle\langle L_1M_1L_2M_2|2m\rangle, \qquad (13)$$
$$i=1,2,3,4; \quad j=1,2.$$

Here $\langle l_1m_1l_2m_2|l_3m_3\rangle$ is a Clebsch-Gordon coefficient and s denotes the set of indices over which the summation is performed, i.e., $s=(l_1, l_2, l_3, l_4, L_1, L_2)$. The coefficient β_s depends on the moduli of the vectors **H**, **X**, and **z**, and is determined by the probability W for scattering of the molecules by the surfaces. If W is invariant under inversion, β_s is a scalar; if not, the right-hand side of (13) contains terms in which β_s is a pseudoscalar; $m_2=m_4=0$, since $\mathbf{z} \parallel \mathbf{k} \parallel OZ$.

In the case of identical surfaces l_2 should be assumed to be even; if on the other hand the surfaces are different (i.e., if they scatter the molecules with different probabilities), then the right-hand side of (13) should contain terms with odd l_2. The birefringence effect in the case of a collisionless gas occurs in the first approximation in the parameter characterizing the nonequilibrium state[6]; therefore, we shall hereinafter set $l_3=1$. It is evident that the spatial inhomogeneity vanishes when the field **H** is switched off. We shall accordingly assume that the equality $l_4=0$ follows from the equality $l_1=0$. Then φ_{2m} can be rewritten in the form

$$\varphi_{2m}=\sum_{s} \beta_s(H,X,z) \sum_{m_1m_3} Y_{l_1m_1}(\mathbf{H}) Y_{l_20}(\mathbf{k})\langle l_1m_1l_20|L_1m_1\rangle$$
$$\times Y_{1m_3}(\mathbf{X}) Y_{l_40}(\mathbf{z})\langle 1m_3l_40|L_2m_3\rangle\langle L_1m_1L_2m_3|2m\rangle. \qquad (14)$$

TABLE I.

Row No.	$\mathbf{H} \neq 0$	X vector	W, invar. under inversion	Surfaces are identical	$\mathbf{X} \parallel z$	φ_{2m}, even rel. to middle of gap
1	−	−	−	−	+	+
2	−	+	+	−	+	+
3	−	−	−	−	−	+
4	−	+	+	−	−	+
5	−	+	−	+	−	+
6	−	−	+	+	−	+
7	+	−	−	−	−	+
8	+	+	+	−	−	+
9	+	+	−	+	−	+
10	+	−	+	+	−	+
11	+	+	−	−	−	−
12	+	−	+	−	−	−
13	+	−	−	+	−	−
14	+	+	+	+	−	−

If $H=0$, then, apart from a redefinition of β_s, we can write

$$\varphi_{2m}=\sum_{s} \beta_s(X) Y_{l_20}(\mathbf{k}) Y_{1m}(\mathbf{X})\langle l_201m|2m\rangle. \qquad (15)$$

In Table I we present the results of the investigation of the conditions in accordance with the formulas (14) and (15) when $\varphi_{2m} \neq 0$, as well as of the symmetry properties of the birefringence (φ_{2m}) with respect to reflection in the plane passing through the middle of the gap.

A plus sign in Table I denotes the assertion written in the top cell of the corresponding column. The minus signs in the first five columns denote respectively the assertions: $\mathbf{H}=0$, **X** is a pseudovector, W is not invariant under inversion, the surfaces are dissimilar, and **X** is arbitrarily oriented. Thus, the first five signs in a row formulate the physical conditions necessary for the existence of birefringence. The sixth sign in a row indicates the evenness (+) or oddness (−) of the birefringence (φ_{2m}) with respect to the middle of the gap under the prescribed conditions.

It can be seen from Table I that, when $\mathbf{X} \parallel z$ and $\mathbf{H}=0$, birefringence is possible only in the case of dissimilar surfaces (the rows 1 and 2). If, on the other hand, **X** is not parallel to z, then for $\mathbf{H}=0$ birefringence is also possible in the case of identical surfaces (the rows 5 and 6). For this to be so, it is necessary that the vector **X** characterizing the nonequilibrium state have the same symmetry with respect to the operation of inversion as the scattering probability W. As will be shown below, this situation in fact obtains. For example, if the state of nonequilibrium is created by the motion of the surfaces relative to each other (i.e., if **X** is a pseudovector and $\mathbf{X} \perp z$), then in the case of identical surfaces only that part of the probability W which is invariant under inversion makes a nonzero contribution to the effect. If, on the other hand, **X** is not parallel to z, and the surfaces are dissimilar (the rows 3 and 4), birefringence is also possible in the case in which the vector **X** and the scattering probability W transform differently under inversion. For $\mathbf{H} \neq 0$, the gas is inhomogeneous for the reasons indicated above.

As can be seen from Table I, the evenness or oddness of the birefringence (φ_{2m}) under the operation $\mathbf{z} \to -\mathbf{z}$ is connected with the symmetry properties of the probability with respect to inversion and with the type of nonequilibrium state that obtains. Thus, in the case of identical surfaces, for φ_{2m} to be invariant under the substitution $\mathbf{z} \to -\mathbf{z}$, it is necessary that X and W have the same symmetry with respect to inversion (the rows 9 and 10). When X and W transform differently, the tensor φ_{2m} is odd with respect to the substitution $\mathbf{z} \to -\mathbf{z}$ (the rows 13 and 14). If the surfaces are dissimilar, then the tensor φ_{2m} is even (the rows 7 and 8) or odd (the rows 11 and 12) with respect to the substitution $\mathbf{z} \to -\mathbf{z}$, depending on whether X and W possess different symmetries or the same symmetry.

It should be borne in mind that the results given in Table I for the case of dissimilar surfaces (the rows 7, 8, 11, and 12) were obtained under the assumption that l_2 is odd [see the formula (14)]. Thus, for example, if for identical surfaces φ_{2m} is even with respect to the middle of the gap (i.e., with respect to the substitution $\mathbf{z} \to -\mathbf{z}$), then in the case of differing surfaces φ_{2m} is, when the other conditions are the same, a sum of even and odd components. By assuming l_2 to be odd, we consider only the odd component.

Let us point out that the Onsager symmetry relations are, generally speaking, not satisfied under nonequilibrium conditions, since they are derived under the assumption that only one energy dissipation mechanism operates—in the alternating electromagnetic field of the wave.[10] This situation is, in particular, realized in the well-known birefringence effect occurring in an inhomogeneous stream of a viscous fluid (the Maxwell effect), in which the internal friction constitutes an additional mechanism for energy dissipation, and leads to the violation of the Onsager symmetry relations.[10]

Birefringence occurs in a collisionless molecular gas only when the gas is in a nonequilibrium state. A weak magnetic field only modifies the then existing tensor polarization of the angular momenta as a result of their precession in the magnetic field. The distribution function of the nonequilibrium collisionless gas, which is necessary for the computation of the tensor φ_{2m} [see the formula (7)]. is determined by solving the Boltzmann kinetic equation (8) with the boundary conditions (9). The Boltzmann equation (8) is not invariant under H-field reversal. Therefore, the nonequilibrium distribution function contains terms both even and odd in the field. In consequence, the tensor φ_{2m} and the linearly related tensor ε_{ik} also contain terms that are odd in H. The presence of terms that are odd in H in the symmetric part of the permittivity tensor is due to the existence of the state of nonequilibrium in the system, and can be understood if account is taken of the precession mechanism of interaction between the angular momenta **M** of the molecules and the field H. Indeed, the angular precession velocity vector reverses sign when H is replaced by −H. Therefore, if in the field H the angular momentum **M** of a molecule turns as a result of the precession through an angle ψ in a time t, in the field −H the angular momentum of the molecule will turn during the same period of time through the angle $-\psi$. Since the birefringence is determined by the orientation of the angular momenta of the molecules, the permittivity tensor $\varepsilon_{ik}(\mathrm{H})$ does not, generally speaking, coincide with the tensor $\varepsilon_{ik}(-\mathrm{H})$.

Below, as a result of a kinetic computation in each specific state-of-nonequilibrium case, we find the tensor φ_{2m}, which, as can be verified, contains terms both even and odd in H. For simplicity, we compute the expressions for the phase difference δ only under conditions of even parity in H.

4. BIREFRINGENCE IN THE CASE OF DIFFERENT TEMPERATURES AT THE SURFACES

Let us set $\mathbf{u}=0$ in the equations (9). Substituting the expression (12) into the system of equations (9), and solving the system in the first approximation in the nonsphericity parameter μ, we obtain the anisotropic in the angular momentum M—part of the nonequilibrium distribution function of the gas.[11] The formula (7) has, in the linear approximation in ΔT ($\Delta T/T_0 \ll 1$), the form

$$\varphi_{2m}=\frac{\mu}{2\sqrt{\pi}}\frac{\Delta T}{T_0^2}\sum_p(-1)^m D_{p-m}^{2*}(\Omega_H)$$

$$\times D_{p0}^2(\Omega_H)\iint\limits_{v_z,v_z'>0} d^2\Gamma\, f_0(E)E'\Big[w_{2000}^{+}(-\Gamma,-\Gamma_3')$$

$$\times\exp\left(ip\frac{\omega(L/2+z)}{v_z}\right)-w_{2000}^{-}(-\Gamma_3,-\Gamma')\exp\left(ip\frac{\omega(L/2-z)}{v_z}\right)\Big]. \quad (16)$$

Here $f_0(E)$ is the Maxwellian energy (E) distribution with temperature T_0,

$$d^2\Gamma=d\Gamma d\Gamma',\quad \Gamma=(\mathbf{v},M^2),\quad \Gamma_3=(\mathbf{v}_3,M^2),$$
$$\mathbf{v}_3=\mathbf{v}-2\mathbf{k}(\mathbf{vk}),\quad d\Gamma=d\mathbf{v}MdM,\quad \omega=\gamma H,$$

γ is the gyromagnetic ratio for the molecule, z is the coordinate of the axis of the ray in the gap, w^+ pertains to the scattering of the molecules by the lower surface, while w^- pertains to the scattering by the upper surface. The expression (16) was derived with allowance for the reciprocal relation for, and the preservation of the Maxwellian character[11] of, the scattering probability.

The expression for $w_{ln00}(\Gamma,\Gamma';\mathbf{k})$ can be written as follows:

$$w_{ln00}(\Gamma,\Gamma';\mathbf{k})=|v_z'|f_0(E')\sum_{L_1l_3l_1l_1'}A_{l_1'l_1l0}^{L_1ll_3}(v'^2,v^2,M'^2,M^2)$$

$$\times\left(\frac{M'}{M_0}\right)^l\begin{pmatrix}L_1 & l & l_3\\ -n & n & 0\end{pmatrix}Y_{l_30}(\mathbf{k})\sum_{m_1m_1'}\langle l_1'm_1'l_1m_1|L_1-n\rangle\, Y_{l_1'm_1'}(\mathbf{v})\,Y_{l_1m_1}(\mathbf{v}'). \quad (17)$$

Below we shall assume in the computations that the coefficients A in (17) are constants.

Assuming that the two surfaces are identical, i.e., that when their temperatures are equal they scatter the molecules with the same probability, and using (17) and the properties of the spherical functions under inversion and reflection in the xy plane, we find that

$$w_{2000}(-\Gamma,-\Gamma_3';\mathbf{k})=w_{2000}(-\Gamma_3,-\Gamma';-\mathbf{k})=w_{2000}(\Gamma_3,\Gamma';\mathbf{k}). \quad (18)$$

Then we have for φ_{2m} the expression:

$$\varphi_{2m}=\frac{\mu}{2\sqrt{\pi}}\frac{\Delta T}{T_0^2}\sum_p(-1)^m D^{2*}_{p-m}(\Omega_H)D_{p0}^{2}(\Omega_H)$$

$$\times\iint\limits_{v_z,v_z'\geqslant 0} d^2\Gamma f_0(E)E'w_{2000}(\Gamma_3,\Gamma';\mathbf{k})$$

$$\times\left[\exp\left(ip\frac{\omega(L/2+z)}{v_z}\right)-\exp\left(ip\frac{\omega(L/2-z)}{v_z}\right)\right]. \qquad (19)$$

Before computing the final expression for δ at prescribed directions of the vectors **H** and **q**, let us analyze the expression obtained for φ_{2m}, and thereby elucidate the properties of the function δ.

As can be seen from (16) and (18), for $\mathbf{H}=0$ birefringence will occur only in the case of dissimilar surfaces. This can be explained as follows. The **M**-vector polarization occurring as a result of the scattering of the gas by one of the surfaces gives rise to an anisotropy in the tensor ε_{ik}, and, consequently, makes it possible for birefringence to occur in the reflected gas stream. But because of the invariance of the scattering probability under inversion and reflection in the xy plane, the scattering by the other surface ($\mathbf{k}\rightarrow-\mathbf{k}$) will lead to exactly the same **M**-vector polarization (the surfaces are identical) with respect to the chosen coordinate system of the opposite sign, i.e., the inverted system. Thus, if the surfaces are identical, and if $\mathbf{H}=0$, then the **M**-vector polarizations that arise as a result of the scattering by the two surfaces cancel each other out, which leads to the disappearance of the birefringence effect. This result follows from the analysis of the symmetry of the system (the row 2 in Table I).

Let $\mathbf{H}\parallel\mathbf{k}$, then in (19) we have $d^2_{p0}(\theta_H=0)=\delta_{p,0}$ and, consequently, $\varphi_{2m}=0$. The nonoccurrence of birefringence at $\mathbf{H}\parallel\mathbf{k}$ can be understood on the basis of the following. The role of a magnetic field in the effect under discussion consists in the modification of the polarization of the gas as a result of the precession of the angular momenta of the molecules during the flight in the gap. But there is no preferred direction perpendicular to the z axis in the absence of a field in the system, and a magnetic field $\mathbf{H}\parallel z$ does not change this symmetry because the angular momenta of the molecules are not polarized by the field **H**. It is therefore not accidental that the application of a $\mathbf{H}\parallel\mathbf{k}$ field led in the expression for φ_{2m} to the equality $p=0$, which in fact implies that the field has been switched off, since the exponents p and $\omega=\gamma H$ in the expression for φ_{2m} are cofactors. Thus, $\varphi_{2m}(\mathbf{H}\parallel\mathbf{k})=\varphi_{2m}(\mathbf{H}=0)$.

It should be noted that the last result cannot be obtained from an analysis of the symmetry of the system in question [see the formulas (14) and (15)], since the mechanism underlying the effect of a magnetic field on the system is not concretized in the process. At the same time, if we recognize that a magnetic field H polarizes the angular momenta in its direction, and that this polarization is not connected with the nonspherical scattering, we find that in principle $\varphi_{2m}\neq 0$, and birefringence occurs. But this effect is vanishingly small, since the effective magnetic moment of a molecule is small: $\mu_{eff}H/k_BT\sim(10^{-7}-10^{-6})$.

The tensor φ_{2m}, as can be seen from (16) and (19), is a function not only of the field, but also of the coordinate z of the ray in the gap. This is due to the fact that the angle through which the magnetic moment of a molecule turns as a result of the precession depends not only on the magnetic field **H**, but also on the time during which this rotation occurs, and, hence, on the distance traveled by the molecule during this time (this is on the average the distance from a surface to the axis of the ray, i.e., $L/2+z$ or $L/2-z$). The tensor φ_{2m}, as can be verified from (19), reverses sign when the substitution $z\rightarrow-z$ is made in it, i.e., it is odd with respect to the middle of the gap $[\varphi_{2m}(z)=-\varphi_{2m}(-z)]$, and, as a result, $\varphi_{2m}(z=0)=0$. This follows from the fact that the scattering probability determined by the coefficient w_{2000} is invariant under inversion, while the vector **X** characterizing the state of nonequilibrium (in our case this is ΔT) changes its sign under inversion. In the case of identical surfaces this, as we saw from the symmetry analysis, makes the birefringence (φ_{2m}) odd with respect to the middle of the gap (see the row 14 in Table I). If, on the other hand, the surfaces are dissimilar (i.e., if $w^+\neq w^-$), then we can separate out in the expression (16) a term that is invaraint under the substitution $\mathbf{z}\rightarrow-\mathbf{z}$ (the row 8 in Table I).

Let us orient $\mathbf{H}\parallel y$ and $\mathbf{q}\parallel x$. Then from Eq. (2) we can easily determine the refractive indices n_1 and n_2 for the waves. Their difference is given by the expression

$$\Delta n=\varepsilon_0^{1/2}-\left(\varepsilon_0+\frac{N}{4}\left(\frac{3}{2}\right)^{1/2}\Delta\alpha\bar{\alpha}\varphi_{20}\right)^{1/2}\approx-\frac{N}{8}\left(\frac{3}{2}\right)^{1/2}\Delta\alpha\bar{\alpha}\varphi_{20},\quad \varepsilon_0\approx 1. \qquad (20)$$

As follows from (19) and (20), Δn is a function of w_{2000}. We retain in the series (17) only one term with the indices $l_1=0$, $l_1'=0$, $L_1=0$, and $l_3=2$, which were determined from a comparison of the theoretical and experimental data obtained in investigations of the thermomagnetic effect in N_2 or CO gases. After the performance of the integration in (19) over M, $\mathbf{v}$, M', v_x, and v_y, the expression for the phase difference δ will have the form

$$\delta=\frac{lN}{\lambda}\frac{\Delta T}{T_0}\mu(\alpha_\parallel-\alpha_\perp)K[I_2(\omega(\tau/2+t))-I_2(\omega(\tau/2-t))]. \qquad (21)$$

Here K is a dimensionless coefficient ($|K|\sim 1$),

$$\omega\tau=\gamma HL\left(\frac{m}{2T_0}\right)^{1/2},\quad \omega t=\gamma Hz\left(\frac{m}{2T_0}\right)^{1/2},\quad x=v_z\left(\frac{m}{2T_0}\right)^{1/2},$$
$$I_n(\omega t)=\int_0^\infty\exp(-x^2)\cos n\frac{\omega t}{x}dx, \qquad (22)$$

and m is the mass of the molecule.

As can be seen from (21) and (22), the phase difference δ changes its sign when ΔT is replaced by $-\Delta T$, as well as when **z** is replaced by $-\mathbf{z}$. The latter also follows from the analysis of the symmetry of the system (the row 14 in Table I), the fact that δ is an odd function of z being unrelated with the parallelism of the vector characterizing the state of nonequilibrium and the z axis, as obtains here. We can, in principle, choose those orientations of **H** and **q** for which the answer for δ will also contain terms that are odd in **H**, but qualitatively the picture will not change.

The dependence of the phase difference δ on the field is shown in Fig. 1 (the curve 1). The oscillations in δ

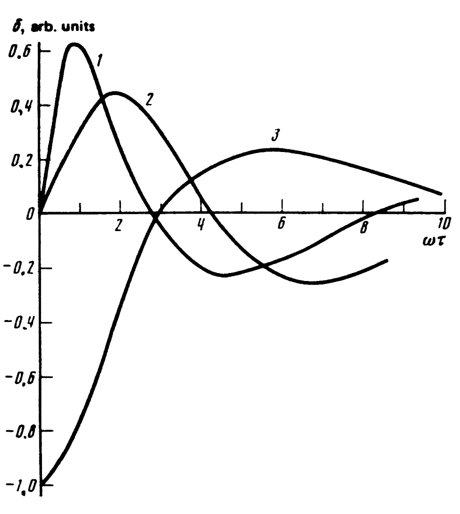

FIG. 1. Dependence of the phase difference δ on $\omega\tau$ for $z = L/4$ and $\mathbf{q} \parallel x$: 1) $\Delta T \neq 0$, $\mathbf{H} \parallel y$; 2) $\Delta N \neq 0$, $\mathbf{H} \parallel z$ ($\delta \times 10^2$); 3) $\mathbf{u} \neq 0$, $\mathbf{H} \parallel z$.

as the field intensity is increased is due to the single, double, etc., precession of the angular momentum of the molecules during the flight between the walls. The damping of the oscillations is due to the presence of a velocity distribution for the molecules.

Let us make order-of-magnitude estimates for the phase difference δ for $T_0 = 300$ K, $H = 0.5 \times 10^3$ Oe, $\Delta T = 100$ K, $l = 50$ cm, $L = 0.2$ cm, $N = 10^{13}$ cm^{-3}, $\lambda = 5 \times 10^3$ Å, $\alpha_{\parallel} - \alpha_{\perp} = 0.7$ Å^3 (for N_2), and $\mu = (0.3-1) \times 10^{-1}$. The substitution of these parameter values into (21) leads to the value $\delta \sim 10^{-7}$ rad. Modern methods allow us to measure the phase difference to within 10^{-9} rad.[7] Let us note that the above-presented estimate for δ pertains to the case of scattering of N_2 or CO gas by a gold or platinum surface, for which the magnitude of the nonsphericity parameter for a molecule is known from thermomagnetic effect investigations ($\mu^2 \sim 10^{-3}$) and the occurrence of ($w_{2000} \neq 0$) scattering processes leading to the tensor polarization of the gas molecules has been established. Physically, the model used for w_{2000} implies that the scattering process is isotropic with respect to the directions of the velocities $\mathbf{v}$ and $\mathbf{v}'$ and anisotropic like Y_{20} with respect to the directions of the angular momentum of a molecule before or after a collision.

5. THE BIREFRINGENCE EFFECT IN A GAS STREAM

Let us now consider birefringence in the case of the flow of a collisionless gas along the gap ($\nabla N \parallel y$) when $\Delta T = 0$. The problem of determining the nonequilibrium distribution function $f(\mathbf{v}, \mathbf{M})$ of the gas under these conditions is solved in Ref. 12. The tensor φ_{2m} can thus be computed:

$$\varphi_{2m} = -\frac{\mu}{2\sqrt{\pi}} L \frac{\nabla N}{N} \sum_{pk} (-1)^m D^{2*}_{p-m}(\Omega_H) \times D_{p0}{}^2(\Omega_H) \iint\limits_{v_z, v_z' > 0} \frac{v_y'}{v_z'} f_0(E) R_{pk}(\Gamma, \Gamma', \omega, z)\, d^2\Gamma,$$

$$R_{pk} = w_{2k00}(-\Gamma, -\Gamma_3'; \mathbf{k}) \exp\left(ip \frac{\omega(L/2+z)}{v_z}\right) + w_{2k00}(-\Gamma_3, -\Gamma'; -\mathbf{k}) \exp\left(ip \frac{\omega(L/2-z)}{v_z}\right). \tag{23}$$

The index k in the right member of (23) assumes only the values ± 1. This can be obtained through integration over $\mathbf{v}'$ with allowance for the expression (17) for w_{ln00}.

As can be seen from (23), here the birefringence is due to another process involving the scattering ($w_{2\pm100} \neq 0$) of the molecules by the surface. It is possible to split the coefficient $w_{2\pm100}$ into two parts $K_{2\pm1}$ and $D_{2\pm1}$, the first of which is invariant under inversion, while the second changes its sign in such a transformation:

$$\begin{aligned} w_{2k00}(-\Gamma, -\Gamma_3'; \mathbf{k}) &= -D_{2k}{}^+(\Gamma_3, \Gamma'; \mathbf{k}) - K_{2k}{}^+(\Gamma_3, \Gamma'; \mathbf{k}), \\ w_{2k00}(-\Gamma_3, -\Gamma'; -\mathbf{k}) &= -D_{2k}{}^-(\Gamma_3, \Gamma'; \mathbf{k}) + K_{2k}{}^-(\Gamma_3, \Gamma'; \mathbf{k}). \end{aligned} \tag{24}$$

The plus superscript in (24) pertains to scattering by the lower surface; the minus superscript, to scattering by the upper surface. Substituting (24) into (23), and setting $\mathbf{H} = 0$, we obtain the following results. In the presence of a gas flow along the gap the birefringence can also be determined by that part ($D_{2\pm1}$) of the probability which is invariant under inversion. If the surfaces are identical, i.e., if $D^+_{2\pm1} = D^-_{2\pm1}$ and $K^+_{2\pm1} = K^-_{2\pm1}$, then φ_{2m} is determined only by that part of the probability which changes sign under inversion, i.e., by $D_{2\pm1}$ (the row 5 in Table I). If, on the other hand, the surfaces are dissimilar, φ_{2m} is determined by both the $D_{2\pm1}$ and $K_{2\pm1}$ terms (the row 4). That part of φ_{2m} which is determined by the $K_{2\pm1}$ terms then changes its sign upon the interchange of the surfaces.

Below we shall assume that the surfaces are identical, and that $\mathbf{H} \neq 0$. Then, using (24), we can rewrite the expression for R_{pk} entering into (23) in the form

$$\begin{aligned} R_{pk} = &-D_{2k}(\Gamma_3, \Gamma'; \mathbf{k}) \left[\exp\left(ip \frac{\omega(L/2+z)}{v_z}\right) + \exp\left(ip \frac{\omega(L/2-z)}{v_z}\right)\right] \\ &-K_{2k}(\Gamma_3, \Gamma'; \mathbf{k}) \left[\exp\left(ip \frac{\omega(L/2+z)}{v_z}\right) - \exp\left(ip \frac{\omega(L/2-z)}{v_z}\right)\right]. \end{aligned} \tag{25}$$

As can be seen from (23) and (25), that part (K_{2k}) of the probability which is invariant under inversion gives rise to a φ_{2m} (birefringence) that is odd with respect to the middle of the gap (i.e., with respect to the operation $\mathbf{z} \to -\mathbf{z}$), while the noninvariant part (D_{2k}) gives rise to an even φ_{2m}. Notice that the results agree with the results obtained from the analysis of the symmetry of the system (the rows 14 and 9 in Table I). From the foregoing it follows, in particular, that the measurement of the phase difference δ as a function of z in $\mathbf{H} \neq 0$ can provide information about the presence of both the invariant and the noninvariant—under inversion—parts of the probability for scattering of molecules by the surface of a solid.

Let us assume that $\mathbf{H} \parallel \mathbf{k}$. Then, as can be seen from (23) and (25), $\varphi_{2m} \neq 0$ only when $m = \pm 1$. In fact,

$$d_{-pm}^{2}(\theta_H = 0) = \delta_{-p,m}, \quad d_{pk}{}^2(\theta_H = 0) = \delta_{p,k},$$

but $k = \pm 1$, and therefore $m = \pm 1$. The elements of the permittivity tensor then have the form

$$\begin{aligned} &\varepsilon_{xx} = \varepsilon_{yy} = \varepsilon_{zz} = \varepsilon_0, \quad \varepsilon_{xy} = 0, \\ &\varepsilon_{yz} = -\frac{N}{4i} \Delta\alpha\bar{a}(\varphi_{21} + \varphi_{2-1}), \quad \varepsilon_{xz} = \frac{N}{4} \Delta\alpha\bar{a}(\varphi_{21} - \varphi_{2-1}). \end{aligned} \tag{26}$$

Using (2) and (26), we can find the refractive indices for the two waves:

$$n_1=(\varepsilon_0+\varepsilon_{yz})^{1/2}, \quad n_2=(\varepsilon_0-\varepsilon_{yz})^{1/2} \quad \text{for} \quad \mathbf{q}\parallel x; \tag{27}$$

$$n_1=(\varepsilon_0+\varepsilon_{xz})^{1/2}, \quad n_2=(\varepsilon_0-\varepsilon_{xz})^{1/2} \quad \text{for} \quad \mathbf{q}\parallel y; \tag{28}$$

$$n_1=n_2=\varepsilon_0^{1/2} \quad \text{for} \quad \mathbf{q}\parallel z. \tag{29}$$

The results of the investigation of the thermomagnetic effect allowed us to establish the presence of only that part of the probability which is invariant under inversion (i.e., $K_{2\pm1}$). Therefore, we shall, in computing δ, proceed from the condition that $D_{2\pm1}=0$. A comparison of the theoretical and experimental results obtained in the thermomagnetic-effect investigations on N_2 and CO gases interacting with Au or Pt surfaces showed that the summation indices in the expression (17) for $K_{2\pm1}$ assume only the values $l_1=1$, $l_1'=1$, $L_1=1$, $l_3=2$. Using these values, we can obtain an explicit expression for the phase difference δ. Let $\mathbf{q}\parallel x$. After the integration in the formula (23) with allowance for (25), we obtain

$$\delta=\mu\frac{LN}{\lambda}\frac{\Delta N}{N}(\alpha_\parallel-\alpha_\perp)K(U_1(\omega(\tau/2+t))-U_1(\omega(\tau/2-t))]. \tag{30}$$

Here K is a dimensionless coefficient ($|K|\sim1$). The integral $U_1(\omega t)$ has the form

$$U_1(\omega t)=\int_0^\infty x\exp(-x^2)\cos\frac{\omega t}{x}dx. \tag{31}$$

The estimation of the phase difference δ with $\Delta N/N\sim1/3$ and the same values of the remaining parameter for the N_2 gas yields $\delta\sim10^{-9}$ rad. Figure 1 (the curve 2) shows a plot of the dependence.

It can be seen from (30) that the phase difference δ changes sign when ΔN is replaced by $-\Delta N$, as well as when $\mathbf{z}$ is replaced by $-\mathbf{z}$. The latter result follows from the analysis of the symmetry of the system (the row 14 in Table I). It should be borne in mind here that, in computing (30), we restricted ourselves to only that part of the scattering probability which is invariant under inversion. In the opposite case the expression for δ would have also contained terms that are odd in z (the row 9). As in the preceding problem, the expression for δ contains only terms that are even in H. At the same time, as can be seen from (23), the expression for φ_{2m} in the general case also contains terms that are odd in H. The damped oscillation of δ as the field intensity is increased is also due to the precession of the angular momenta of the molecules and the existence of a velocity distribution for these particles.

6. BIREFRINGENCE IN THE CASE OF THE NONEQUILIBRIUM STATE CREATED BY THE MOTION OF THE SURFACES (THE COUETTE PROBLEM)

Let the upper plate move along the y axis with velocity $\mathbf{u}$; the lower plate, along the same axis with velocity $\mathbf{u}$ in the opposite direction. Substituting the expression (12) for $f(\mathbf{v},\mathbf{M})$ in the system of integral equations (9) with $\mathbf{u}\neq0$ ($\mathbf{u}\perp\mathbf{k}$), we find the nonequilibrium distribution function $f(\mathbf{v},\mathbf{M})$ of the gas in the first approximation in the scattering-nonsphericity parameter μ. The integrals φ_{2m} have, in the first approximation in the parameter u/v_t, the form

$$\begin{gathered}\varphi_{2m}=-\frac{\mu}{2\sqrt{\pi}}\frac{4u}{v_T^2}\sum_{pk}(-1)^m D^{2*}_{-pm}(\Omega_H)D_{pk}^2(\Omega_H)\\ \times\int\int\limits_{v_z,\,v_z'>0} v_y'f_0(E)F_{pk}(\Gamma,\Gamma',\omega,z)\,d^2\Gamma,\\ F_{pk}=w_{2k00}(-\Gamma,-\Gamma_3';\mathbf{k})\exp\left(ip\frac{\omega(L/2+z)}{v_z}\right)\\ -w_{2k00}(-\Gamma_3,-\Gamma';-\mathbf{k})\exp\left(ip\frac{\omega(L/2-z)}{v_z}\right).\end{gathered} \tag{32}$$

The index k, as in (23), assumes only the values ±1. This can be verified by performing the integration over $\mathbf{v}'$ in (32) with allowance for the expression (17) for w_{2k00}. Thus, the birefringence is determined by the same molecule-scattering process on the surface, that determines the birefringence in a gas stream along the gap. But there is an important difference. In front of the second term in the expression for F_{pk} stands the minus sign, and not the plus sign, as is the case in (23). This is a consequence of the fact that the vector characterizing the state of nonequilibrium here is a pseudovector. Indeed, the invariant and noninvariant—under inversion—parts of the probability exchange roles when the expression (24) is substituted into (32). As follows from the analysis of the symmetry of the system, the symmetry of the tensor φ_{2m} with respect to the middle of the gap changes when the vector characterizing the state of nonequilibrium is replaced by a pseudovector. Moreover, in the case of identical surfaces, the symmetry of that part of the scattering probability which makes the nonzero contribution to the effect in $\mathbf{H}=0$ changes. This can be seen from a comparison of the rows 7–10 and 11–14 in Table I, as well as the rows 5 and 6. Thus, in the case of identical surfaces and $\mathbf{H}=0$, the birefringence is determined by that part of the probability which is invariant under inversion, i.e., by $K_{2\pm1}$ (in the problem with a gas stream the effect is determined by the noninvariant part $D_{2\pm1}$). If the surfaces are dissimilar, then that part of φ_{2m} which is determined by $D_{2\pm1}$ changes its sign upon the interchange of the surfaces.

Let us write down F_{pk} with allowance for (24), assuming that the surfaces are identical:

$$\begin{gathered}F_{pk}=-D_{2k}(\Gamma_3\Gamma';\mathbf{k})\left[\exp\left(ip\frac{\omega(L/2+z)}{v_z}\right)-\exp\left(ip\frac{\omega(L/2-z)}{v_z}\right)\right]\\ -K_{2k}(\Gamma_3,\Gamma';\mathbf{k})\left[\exp\left(ip\frac{\omega(L/2+z)}{v_z}\right)+\exp\left(ip\frac{\omega(L/2+z)}{v_z}\right)\right].\end{gathered} \tag{33}$$

As can be seen from (33) and (32), that part of the probability which is invariant under inversion (i.e., $K_{2\pm1}$) gives rise to φ_{2m} that is even with respect to the middle of the gap (i.e., with respect to the operation $\mathbf{z}\to-\mathbf{z}$), while the noninvariant part ($D_{2\pm1}$) gives rise to an odd φ_{2m}. These results also agree with the results obtained from the analysis of the symmetry of the system (the rows 10 and 13 in Table I).

Let us again assume that $\mathbf{H}\parallel\mathbf{k}$. Then $\varphi_{2m}\neq0$ when $m=\pm1$. The elements of the permittivity tensor ε_{ik} are given by the formulas (26). Limiting ourselves to that part of the probability which is invariant under inversion (i.e., to $K_{2\pm1}$), and taking into account the values of the summation indices in the expression for $K_{2\pm1}$, we obtain the following explicit expression for δ as a function of z and H for $\mathbf{H}\parallel\mathbf{k}$ and $\mathbf{q}\parallel x$:

$$\delta=\mu\frac{u}{v_T}\frac{l}{\lambda}N(\alpha_\parallel-\alpha_\perp)K[U_1(\omega(\tau/2+t))+U_1(\omega(\tau/2-t))]. \qquad (34)$$

Here K is a dimensionless coefficient ($|K|\sim 1$). The integral U_1 is given by the formula (31). The estimation of the phase difference δ with $u/v_t\sim 1$ and the same values of the remaining parameters for the N_2 gas yields the value 10^{-7} rad.

As can be seen from (34), the computed expression for the phase difference is an odd function of the velocity u and an even function of the field **H** and the coordinate z of the ray in the gap. The evenness of δ as a function of z is due to the invariance of that part of the scattering probability which is determined by the coefficient $K_{2\pm 1}$ and the invariance under inversion of the vector characterizing the state of nonequilibrium in this problem (the row 10 in Table I).

Figure 1 (curve 3) shows a plot of the phase difference δ as a function of the field. It can be seen that $\delta(\mathbf{H}=0)\neq 0$. This is due to the fact that in this problem the vector characterizing the state of nonequilibrium is a pseudovector, and in this case, as follows from the symmetry (the row 6), that part of the probability which is invariant under inversion when $\mathbf{H}=0$ makes a nonzero contribution to the permitivity tensor and, consequently, to δ.

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