Spectrum of Rayleigh scattering of light by a Boltzmann gas in a strong magnetic field

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The spectrum of Rayleigh scattering of light by a diatomic gas in a strong magnetic field is calculated from the solution of the Boltzmann transport equation. It is shown that the structure of the spectrum changes significantly when the magnetic field is applied. The depolarized constituent splits into five independent components. The widths of all the polarized components of the spectrum become functions of the angle between the field and the scattering vector. Detailed examination of the parameters of the spectrum in a magnetic field can be used as a basis for determining the magnetic-field dependence of all the transport coefficients.

1. INTRODUCTION

This paper presents a calculation of the spectrum of Rayleigh scattering of light in a gas consisting of anisotropic diatomic molecules placed in a sufficiently strong external magnetic field. It is found that the structure of both the polarized and, especially, the depolarized spectrum is very sensitive to the presence of the magnetic field. This is due to the partial "freezing" of the rotational degrees of freedom of the molecules by the magnetic field.[1–4] In their turn, the rotational degrees of freedom actively manifest themselves in the structure of the scattered spectrum.[5] This occurs both in its polarized component, through the dependence of the transport coefficients on the rotational degrees of freedom, and in the depolarized component of the spectrum, whose width is determined by collisions that alter the rotational angular momentum of the molecule. Moreover, the depolarized constituent of the scattered light has its own fine structure in the form of three narrow valleys (at the unshifted frequency and at the Mandel'shtam-Brillouin frequencies), whose intensities are determined by the coupling between the translational and rotational degrees of freedom during collisions. On the other hand, the width of the valley at the unshifted frequency is determined exclusively by the shear viscosity. Hence, the dependence of the width of the valley at the unshifted frequency on the direction of the magnetic field provides us with a direct way of studying shear viscosity as a function of the magnetic field.

The first theoretical paper by Kagan and Maksimov,[1] who showed that magnetic and electric fields influenced the transport coefficients of a gas through the vibrational degrees of freedom, was followed by an extensive series of papers devoted to this interesting phenomenon.[2–4,6–12] The dependence of transport coefficients on external electric and magnetic fields was investigated for nonequilibrium gases, and most experiments were concerned with the thermal conductivity at different gas pressures.[6–12] Precision studies of the structure of the spectrum of Rayleigh scattering of light by a gas in a magnetic field, which have recently become possible through the use of lasers and high-resolution techniques,[13] have yielded information on the magnetic-field dependence of all the transport coefficients, including shear and second viscosities, which are difficult to investigate under nonequilibrium gas conditions.

All the spectral parameters of light scattered by a gas in a magnetic field, including the transport coefficients, can be expressed in terms of the eigenfunctions and eigenvalues of the linearized Boltzmann collision integral. In the approximation in which the interaction potential of the gas molecules is only slightly anistropic, the angular dependence of the spectral parameters of the scattered light can be used to extract, in an explicit form, the contribution of translational and rotational degrees of freedom to all the transport coefficients.

2. STRUCTURE OF DEPOLARIZED CONSTITUENT OF THE SPECTRUM OF RAYLEIGH SCATTERING OF LIGHT BY A GAS IN A STRONG MAGNETIC FIELD

The Rayleigh spectrum of a gas is determined by the fluctuation correlation function[14]

$$\Phi(0, 0, \Gamma_2;\, t, \mathbf{r}, \Gamma_1) = \langle \delta f(\mathbf{r}, t, \Gamma_1)\, \delta f(0, 0, \Gamma_2) \rangle,$$

where t is the time, r is the coordinate, Γ represents the internal degrees of freedom of the molecule, δf is the departure of the distribution function f of the gas molecules from the equilibrium Boltzmann function f_0, and the angle brackets represent the usual averaging over the statistical ensemble. For simplicity, we confine our attention to the diatomic (or linear) molecule for which the internal degrees of freedom are the three components of velocity $\mathbf{v}$ and the three components of the angular momentum $\mathbf{M}$ of the molecule. When the magnetic field $\mathbf{B}$ is applied, these molecules begin to interact with the field because a rotating molecule has a magnetic moment $\boldsymbol{\mu} = \gamma \mathbf{M}$, where the gyromagnetic ratio γ depends on the internal structure of the diatomic molecule.[3] For paramagnetic molecules, $\gamma \simeq \mu_B/h$, where μ_B is the Bohr magneton, numerically equal to 0.93×10^{-20} erg/Oe. For most molecules, the ground state is nondegenerate with zero spin (nonparamagnetic molecules). The magnetic moment of the molecules is then due to the rotation of the nucleus, and the gyromagnetic ratio is determined by the nuclear magneton (for example, for nitrogen, $\gamma \simeq 0.25\mu_n/h$, where μ_n is the nuclear magneton, numerically equal to 5.05×10^{-24} erg/Oe).

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When the magnetic field $\mathbf{B}$ is present, the transport equation for the function $(r, t, \Gamma_1, \Gamma_2)$ for $t > 0$ is[1,15]

$$\frac{\partial\Phi}{\partial t}+\mathbf{v}_1\frac{\partial\Phi}{\partial\mathbf{r}}+\gamma[\mathbf{M}_1\times\mathbf{B}]\frac{\partial\Phi}{\partial\mathbf{M}_1}=\hat{I}_1\Phi \tag{1}$$

subject to the initial condition

$$\Phi(\mathbf{r}, 0, \Gamma_1, \Gamma_2)=f_0(\Gamma_1)\ \delta(\mathbf{r})\delta(\Gamma_1-\Gamma_2),$$

where $\hat{I}_1$ is the linearized integral Boltzmann operator[15] acting on the function δf. This is founded in the assumption that $\mu B \ll T$, where T is the gas temperature.

The shape of the Rayleigh spectrum $I(\omega)$ for the above gas is expressed in terms of the function Φ as follows:

$$I(\omega,\mathbf{q})=\mathrm{Re}\int d\Gamma_1\, d\Gamma_2\, c^*(\Gamma_1)\,\Phi(\omega,\mathbf{q},\Gamma_1,\Gamma_2)\,c(\Gamma_2),$$
$$\Phi(\omega,\mathbf{q},\Gamma_1,\Gamma_2)=\int_0^\infty dt\, d\mathbf{r}\,\Phi(\mathbf{r},t,\Gamma_1,\Gamma_2)\,e^{i(\mathbf{qr}-\omega t)}, \tag{2}$$

where $c(\Gamma) = e_{1s}\alpha_{st}(\Gamma)e_{2t}$, e_{1s} and e_{2t} are the polarization unit vectors of the incident and scattered waves, $\omega = \omega_2 - \omega_1$, $\mathbf{q} = \mathbf{k}_2 - \mathbf{k}_1$ are, respectively, the frequency and wave-vector differences for scattered (2) and incident (1) waves, and

$$\alpha_{st}(\Gamma)=\alpha\delta_{st}+\beta\left(\frac{M_s M_t}{M^2}-\frac{1}{3}\delta_{st}\right)$$

is the polarizability tensor of the gas molecules. The asterisk indicates complex conjugates, and repeated subscripts imply summation. When the scattered spectrum is analyzed with the aid of the transport equation (1), it is expedient[5] to transform from the description in terms of the correlation function $\Phi(\omega,\mathbf{q},\Gamma_1,\Gamma_2)$ to the new function χ:

$$\chi(\omega,\mathbf{q},\Gamma)=f_0^{-1}(\Gamma)\int\Phi(\omega,\mathbf{q},\Gamma,\Gamma')\,c(\Gamma')\,d\Gamma', \tag{3}$$

which satisfies the equation

$$\left\{i(\mathbf{qv})-i\omega+\gamma[\mathbf{M}\times\mathbf{B}]\frac{\partial}{\partial\mathbf{M}}-\hat{K}\right\}\chi=c(\Gamma), \tag{4}$$

where $\hat{K}$ is the linearized integral Boltzmann operator acting on the function χ. The shape of the spectrum is expressed in terms of χ, as follows:

$$I(\omega,\mathbf{q})=\mathrm{Re}\int d\Gamma c^*(\Gamma)f_0(\Gamma)\chi(\omega,\mathbf{q},\Gamma)=\mathrm{Re}\langle c|\chi\rangle, \tag{5}$$

where $\langle c|\chi\rangle$ is the scalar product of $c(\Gamma)$ and $\chi(\Gamma)$.

Before we proceed to the solution of (4), let us analyze the order of magnitude of all the terms in the equation. It is readily verified that this equation contains three independent frequency parameters, namely, the Doppler frequency shift in free motion qv, the precession frequency γB of the magnetic moment of the molecule in the magnetic field, and the gas-kinetic collision frequency ν, which is a characteristic eigenvalue of the collision operator $\hat{K}$. In this paper, we shall largely confine our attention to the following relationship between the parameters:

$$T\gg\gamma B\gg\nu\gg qv. \tag{6}$$

The numerical values of the pressures and magnetic fields that are necessary to satisfy (6) will be listed below. When (6) is satisfied, the term involving the magnetic field is the leading term in (4). The eigenfunctions of the operator $\gamma[\mathbf{M}\times\mathbf{B}]\partial/\partial\mathbf{M}$ are the spherical harmonics $Y_{LN}^{B}(\mathbf{M}/M)$, where the direction of $\mathbf{B}$ acts as the quantization axis. The corresponding purely imaginary eigenvalues are $i\gamma BN$. When only this term is retained in (1), the depolarized spectrum ($\mathbf{e}\cdot\mathbf{e}_2 = 0$) takes the form of five infinitely narrow components, located at the frequencies $\omega = 0$ $(N = 0)$, $\omega = \pm\gamma B$ $(N = \pm 1)$, $\omega = \pm 2\gamma B$ $(N = \pm 2)$. The integrated intensity of each component depends on the direction of the magnetic field as follows:

$$I(N=0)\propto 4(\mathbf{be}_1)^2(\mathbf{be}_2)^2,$$
$$I(N=\pm1)\propto\frac{2}{3}\left[(\mathbf{be}_1)^2+(\mathbf{be}_2)^2-4(\mathbf{be}_1)^2(\mathbf{be}_2)^2\right], \tag{7}$$
$$I(N=\pm2)\propto\frac{2}{3}\left[1-(\mathbf{be}_1)^2\right]\left[1-(\mathbf{be}_2)^2\right],\quad \mathbf{b}=\mathbf{B}/B.$$

We note that the total integrated intensity of all the five components is independent of the magnetic field $\mathbf{B}$ [this is readily verified by adding together all the components of the spectrum I_N in (7)], and is determined by the square of the symmetric component, β^2, of the polarizability tensor α_{ik}. Figure 1 illustrates the shape of the depolarized constituent of the spectrum for certain relationships between b and $\mathbf{e}_1,\mathbf{e}_2$.

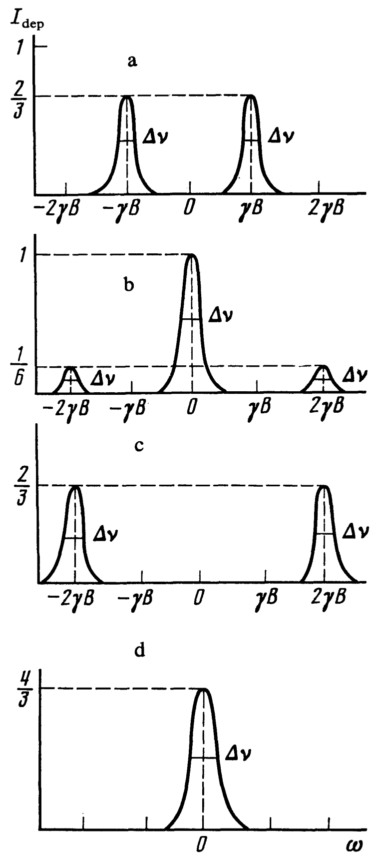

FIG. 1. Spectrum of depolarized ($\mathbf{e}_1\cdot\mathbf{e}_2 = 0$) Rayleigh scattering of light by a diatomic gas in a strong magnetic field $\gamma B > \Delta\nu$ (γB is the frequency of precession of the angular momentum around the magnetic field, and $\Delta\nu$ is the width of the depolarized component of the scattered spectrum. (a—vector $\mathbf{b}$ parallel to $\mathbf{e}_1$ or $\mathbf{e}_2$; b—vector $\mathbf{b}$ lies in the plane of the vectors $\mathbf{e}_1$ and $\mathbf{e}_2$ at the angle of 45°; c—vector $\mathbf{b}$ perpendicular to $\mathbf{e}_1$ and $\mathbf{e}_2$; d—shape of the spectrum for $\mathbf{B} = 0$.

Inclusion of the collision term in (4) leads to the appearance of a finite width of each component of the depolarized scattered spectrum, which is equal to the frequency $\Delta\nu_N$ of collisions with a change in the rotational angular momentum of the molecule[5]:

$$\Delta\nu_N=\int Y_{2N}^*\left(\frac{\mathbf{M}}{M}\right)f_0(\Gamma)\hat{K}Y_{2N}\left(\frac{\mathbf{M}}{M}\right)d\Gamma$$
$$=\left\langle Y_{2N}\left(\frac{\mathbf{M}}{M}\right)|\hat{K}|Y_{2N}\left(\frac{\mathbf{M}}{M}\right)\right\rangle. \tag{8}$$

We recall that the quantity $\Delta\nu\lesssim\nu$ is connected only with the anisotropic part of the molecular potential [for a purely isotropic potential, $\hat{K}Y_{2N}(\mathbf{M}/M)=0$], so that $\Delta\nu$ is an independent collision parameter that, in general, does not coincide with ν.

We now proceed to the fine structure of the scattered spectrum due to the inclusion of the terms $i\mathbf{q}\cdot\mathbf{v}$ in (4). The operator $\gamma[\mathbf{M}\times\mathbf{B}]\partial/\partial\mathbf{M}$ divides the space of functions in which (4) operates into five classes of eigenfunctions corresponding to five eigenvalues. These classes are independent because they are not coupled by collisions by virtue of the condition $\nu\ll\gamma B$. The presence of the magnetic field ensures that, when the fine structure of the spectrum is calculated, the space of functions for which (4) must be solved is the class of functions with zero eigenvalues of the above operator: $\gamma[\mathbf{M}\times\mathbf{B}]\partial\chi/\partial M=0$.

The zeroth class of functions has the form

$$\chi(\mathbf{v},\mathbf{M})=\sum_l f_l(\mathbf{v},M)Y_{l0}^B\left(\frac{\mathbf{M}}{M}\right). \tag{9}$$

We note that the description of the spectrum within the framework of the space of the functions (9) is physically equivalent to a transformation from the variable $\mathbf{M}$ precessing in the magnetic field to the average between collisions $\langle\mathbf{M}\rangle=(\mathbf{M}\cdot\mathbf{b})\mathbf{b}$.[1)]

To solve (4) in the space of the functions (9), it is convenient to replace the collision operator $\hat{K}$ with $\tilde{K}=\hat{P}\hat{K}$, where $\hat{P}$ is the operator representing projection onto the space of the functions (9). The eigenfunctions χ_α of the operator $\tilde{K}$ are conveniently chosen in the form

$$\chi_{\alpha m}=\sum_{ls}\varphi_{\alpha m}^{sl}(v,M)Y_{sm}^B\left(\frac{\mathbf{v}}{v}\right)Y_{l0}^B\left(\frac{\mathbf{M}}{M}\right), \tag{10}$$

where $\varphi_{\alpha m}^{sl}(v,M)$ are real functions. The operator $\tilde{K}$ is real, so that the functions $\chi_{\alpha m}$ and $\chi_{\alpha-m}$ are related by $\chi_{\alpha m}=(-1)^m\chi_{\alpha-m}$, and the corresponding eigenvalues are $\nu_{\alpha m}=\nu_{\alpha-m}$. The subsequent calculations in the chosen space (9) are practically completely analogous to those in the absence of the magnetic field.[5] Equation (4) is expanded over the basis of the eigenfunctions of the collision operator $\tilde{K}$ (10). In this representation, (4) reduces to a coupled set of equations for the hydrodynamic and nonhydrodynamic modes. The hydrodynamic modes are the following five eigenfunctions of the collision operator $\tilde{K}$ with zero eigenvalues (these functions correspond to the five collision invariants, namely, the number of particles, the momentum $m\mathbf{v}$, and the energy ε): acoustic modes

$$\chi_1=\frac{1}{\sqrt{2}}\left(\frac{c_v}{c_p}\right)^{1/2}\left[1-\left(\frac{c_p}{c_v}\right)^{1/2}\frac{v_x}{v_0}+\frac{1}{c_v^{1/2}}\frac{\varepsilon-\bar{\varepsilon}}{\Delta\varepsilon}\right],$$
$$\chi_2=\frac{1}{\sqrt{2}}\left(\frac{c_v}{c_p}\right)^{1/2}\left[1+\left(\frac{c_p}{c_v}\right)^{1/2}\frac{v_x}{v_0}+\frac{1}{c_v^{1/2}}\frac{\varepsilon-\bar{\varepsilon}}{\Delta\varepsilon}\right], \tag{11a}$$

shear modes

$$\chi_3=\frac{v_y}{v_0},\quad \chi_4=\frac{v_z}{v_0}, \tag{11b}$$

and thermal-conduction modes

$$\chi_5=\left(\frac{c_v}{c_p}\right)^{1/2}\left(\frac{1}{c_v^{1/2}}-\frac{\varepsilon-\bar{\varepsilon}}{\Delta\varepsilon}\right). \tag{11c}$$

where c_p, c_v are the specific heats of the molecule with allowance for translational and rotational degrees of freedom at constant pressure and constant volume, respectively, $v_0=(T/m)^{1/2}$, T is the gas temperature, m is the mass of a molecule, $\bar{\varepsilon}=c_vT$ is the average energy of a molecule, and $\Delta\varepsilon=c_v^{1/2}T$ is the energy variance. When the magnetic field is present, the most convenient coordinate frame is that in which the x axis lies along the vector $\mathbf{q}$, the y axis lies in the plane of the vectors $\mathbf{q}$ and $\mathbf{B}$, and the z axis is perpendicular to this plane. The corresponding unit vectors are given by

$$\mathbf{e}_x=\frac{\mathbf{q}}{q},\quad \mathbf{e}_y=\frac{[[\mathbf{q}\times\mathbf{B}]\mathbf{q}]}{|[[\mathbf{q}\times\mathbf{B}]\mathbf{q}]|},\quad \mathbf{e}_z=\frac{[\mathbf{q}\times\mathbf{B}]}{|[\mathbf{q}\times\mathbf{B}]|}.$$

The functions (11) are written in this particular coordinate frame.

All the other eigenfunctions of the operator $\tilde{K}$ with nonzero eigenvalues $\nu_{\alpha m}$ are called nonhydrodynamic. The connection between the above modes is implemented by the operator $i\mathbf{qv}$. The set of equations written in the basis of the eigenfunctions of the operator K [equivalent to the intergral equation (4)] assumes the form[5]

$$\begin{aligned}
-i(\omega+qu_{3B})a_1+\Gamma_1a_1&=c_1-i\Delta c_1,\\
-i(\omega-qu_{3B})a_2+\Gamma_2a_2&=c_2-i\Delta c_2,\\
-i\omega a_3+\Gamma_3a_3+\Gamma_{34}a_4&=-i\Delta c_3,\\
-i\omega a_4+\Gamma_4a_4+\Gamma_{43}a_3&=-i\Delta c_4,\\
-i\omega a_5+\Gamma_5a_5&=c_5,\\
-i\omega a_{\alpha m}+\nu_{\alpha m}a_{\alpha m}&=c_{\alpha m}-i\sum_{k\leq5}(\mathbf{qv})_{\alpha m,k}a_k,\quad \alpha>5,
\end{aligned} \tag{12}$$

where a_i are the coefficients in the expansion of the solution $\chi(\omega,\mathbf{q},\Gamma)$ of (4), with the operator $\tilde{K}$ replacing $\hat{K}$, in terms of the eigenfunctions χ_i (10) of the operator $\tilde{K}$: $a_i=\langle\chi_i|\chi\rangle$. The widths $\Gamma_1,\ldots,\Gamma_5$ of the hydrodynamic modes in second order of perturbation theory in the parameter qv/ν are given by

$$\Gamma_i=\sum_{\substack{\alpha>5\\m}}(\mathbf{qv})_{i,\alpha m}(\mathbf{qv})_{\alpha m,i}(\nu_{\alpha m})^{-1}. \tag{13}$$

The remaining quantities are defined as follows:

$$\Gamma_{34}=\sum_{\substack{\alpha>5\\m}}(\mathbf{qv})_{3,\alpha m}(\mathbf{qv})_{\alpha m,4}(\nu_{\alpha m})^{-1},\quad \Gamma_{43}=\Gamma_{34}^*,$$
$$c_i=\langle\chi_i|c\rangle,\quad \Delta c_i=\sum_{\substack{\alpha>5\\m}}(\mathbf{qv})_{i,\alpha m}c_{\alpha m}(\nu_{\alpha m})^{-1}, \tag{14}$$
$$(\mathbf{qv})_{\alpha m,k}=\langle\chi_{\alpha m}|\mathbf{qv}|\chi_k\rangle=\int\chi_{\alpha m}\mathbf{qv}f_0\chi_k\,d\Gamma,\quad k=1,\ldots5.$$

Since the operator $\widetilde{K}$ is T-even, we have $\Delta c_1 = \Delta c_2$ and $\Gamma_1 = \Gamma_2$. The coefficient

$$c_1=c_2=\frac{1}{\sqrt{2}}\left(\frac{c_v}{c_p}\right)^{1/2}\alpha(\mathbf{e}_1\mathbf{e}_2),\quad c_3=c_4=0,\quad c_5=c_p^{-1/2}\alpha(\mathbf{e}_1\mathbf{e}_2)$$

satisfies, as before, the Landau-Placzek relation

$$\frac{(c_1)^2+(c_2)^2}{(c_1^2)+(c_2)^2+(c_5)^2}=\frac{c_v}{c_p}.$$

For frequencies $\omega \lesssim \Delta\nu$, the scattered spectrum intensity is given by

$$I(\omega,\mathbf{q})=\operatorname{Re}\sum_{k\leq 5} c_k^* a_k+\operatorname{Re}\sum_{\substack{\alpha>5\\ m}} c_{\alpha m}^* a_{\alpha m}. \qquad (15)$$

We note that the first term in (15) describes a narrow triplet in the polarized spectrum: the central component is determined by the intensity $(c_5)^2$ and the width Γ_5 of the thermal-conduction mode, whilst the Mandel'shtam-Brillouin acoustic components that are shifted by the amount $\pm\Omega_{ac} = qu_{ac}$ have the intensities $(c_1)^2 = (c_2)^2$ and widths $\Gamma_1 = \Gamma_2$.

The second term in (15) describes the depolarized part of the spectrum (we consider only the portion of the depolarized spectrum that is unshifted by the magnetic field) with intensity of the order of $(c_{\alpha m})^2$ and width of the order of $\Delta\nu_{\alpha m}$ (the quantities $\Delta\nu_{\alpha m}$ are interpreted as representing the eigenvalues in the set $\nu_{\alpha m}$ that determine the frequency of collisions with a change in the rotational angular momentum **M**). Moreover, this term also incorporates the fine structure of the depolarized scattered radiation, i.e., the narrow valleys at the line center and at the acoustic frequencies, where the valley depths are determined by the quantities $(\Delta c_3)^2$, $(\Delta c_4)^2$, and $(\Delta c_1)^2$, and the widths by Γ_3, Γ_4, and Γ_1, respectively. We must now proceed to the dependence of these parameters of the spectrum on the magnetic field.

3. MAGNETIC-FIELD DEPENDENCE OF THE SPECTRAL PARAMETERS OF POLARIZED SCATTERED RADIATION

It is well-known[14] that the widths of the narrow components in the scattered spectrum are determined by the damping of the hydrodynamic modes, and can be expressed in terms of the transport coefficients, namely, the shear and second viscosities and the thermal conductivity. When the magnetic field is applied, these coefficients become anisotropic and are tensors.[1,15]

We begin by examining Γ_5, which is determined exclusively by the thermal-conductivity tensor $\varkappa_{ij}$:

$$\Gamma_5=\sum_{ij} q_i q_j \varkappa_{ij}\frac{m}{\rho c_p},$$

where ρ is the gas density (in g $\cdot$ cm^{-3}):

$$\varkappa_{ij}=\frac{\rho}{m}\sum_{\substack{\alpha>5\\ k}}\frac{\langle c_p^{1/2}\chi_5|v_i|\chi_{\alpha k}\rangle\langle\chi_{\alpha k}|v_j|c_p^{1/2}\chi_5\rangle}{\nu_{\alpha k}}. \qquad (16)$$

Substituting the explicit expressions (10) for the eigenfunctions $\chi_{\alpha k}$ in (16), and separating the angular dependence on the velocity **v**, we obtain

$$\varkappa_{ij}=\sum_{k=0,\pm1}\langle n_i|Y_{1k}^B(\mathbf{n})\rangle\langle Y_{1k}^B(\mathbf{n})|n_j\rangle c_k,$$
$$c_k=\frac{1}{3}\sum_{\alpha>5}\frac{\rho}{m}\frac{|\langle c_p^{1/2}\chi_5|v|\varphi_{\alpha k}^{10}\rangle|^2}{\nu_{\alpha k}}\geqslant 0,\quad n_i=\frac{v_i}{v}. \qquad (17)$$

Since the coefficients $\varphi_{\alpha k}^{10}$ and the eigenvalues $\nu_{\alpha k}$ depend only on the modulus of k, the constants c_k in (17) also have the property $c_k = c_{-k}$. Using this relation, we obtain the following expressions for the $\varkappa_{ij}$ (see Refs. 1–4):

$$\varkappa_{ij}=\varkappa\delta_{ij}+\varkappa_1 b_i b_j,\quad \varkappa=c_1,\quad \varkappa_1=c_0-c_1. \qquad (18)$$

The antisymmetric part of the thermal-conductivity tensor appears when $\gamma B\sim\nu$, but does not provide a contribution to Γ_5 [according to (16)]. Using the explicit form of the magnetic-field dependence of the thermal-conductivity tensor,[1–4] we can now determine the analogous relation for the width of the thermal-conduction component of the polarized spectrum:

$$\Gamma_5=\frac{m}{\rho c_p}q^2\{\varkappa+\varkappa_1\cos^2\theta\},\quad \cos\theta=(\mathbf{b}\mathbf{e}_x). \qquad (19)$$

It is clear from this expression that the width Γ_5 is a function of the angle θ between the magnetic field and the scattering vector $\mathbf{q}(\mathbf{e}_h = \mathbf{q}/q)$. By varying the angle θ, it is possible to separate experimentally the isotropic from the anisotropic components of the width Γ_5 and thus separate the coefficients $\varkappa$ and $\varkappa_1$ in the thermal-conductivity tensor. Kagan and Maksimov[3] have shown that, in the case of a weakly anisotropic potential for which $\Delta\nu/\nu \ll 1$, it turns out that $\varkappa_1/\varkappa\sim(\Delta\nu/\nu)^2$, i.e., the ratio of the anisotropic part of the linewidth Γ_5 to its isotropic part is of the order of $(\Delta\nu/\nu)^2$.

We now turn to the determination of the quantities Γ_3, Γ_4, and Γ_{34}, Γ_{43} that determine the widths of the shear modes. These coefficients are related to the shear viscosity tensor η_{ijkl} as follows:

$$\Gamma_3=\frac{1}{\rho}\sum_{ijkl}q_j q_k\eta_{ijkl}e_{yi}e_{yl},\quad \Gamma_{34}=\frac{1}{\rho}\sum_{ijkl}q_j q_k\eta_{ijkl}e_{yi}e_{zl},$$
$$\Gamma_4=\frac{1}{\rho}\sum_{ijkl}q_j q_k\eta_{ijkl}e_{zi}e_{zl},\quad \Gamma_{43}=\Gamma_{34}^*, \qquad (20)$$

where

$$\eta_{ijkl}=\rho\sum_{\substack{\alpha>5\\ m}}\frac{\langle v_i v_j-{}^1/_3\delta_{ij}v^2|\chi_{\alpha m}\rangle\langle\chi_{\alpha m}|v_k v_l-{}^1/_3\delta_{kl}v^2\rangle}{\nu_{\alpha m}}\frac{1}{v_0^2}.$$

Isolating in (20) the angular dependence on the velocity **v** in an explicit form, we obtain

$$\eta_{ijkl}=\sum_{m=0,\pm1,\pm2}\left\langle n_i n_j-\frac{1}{3}\delta_{ij}\Big|Y_{2m}^B(\mathbf{n})\right\rangle\times\left\langle Y_{2m}^B(\mathbf{n})\Big|n_k n_l-\frac{1}{3}\delta_{kl}\right\rangle d_m,$$
$$d_m=\sum_{\alpha>5}\frac{\rho}{v_0^2}\frac{|\langle v^2|\varphi_{\alpha m}^{20}\rangle|^2}{\nu_{\alpha m}}\frac{4}{3}\left(\frac{1}{15}\right)^2. \qquad (21)$$

Since the coefficients satisfy the relation $d_m = d_{-m}$, it is convenient to write the tensor η_{ijkl} in the form (see Refs. 1–3)

$$\eta_{ijkl}=\eta\,(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}-{}^2/{}_3\delta_{ij}\delta_{kl})+\eta_1\,(b_ib_k\delta_{jl}+b_ib_l\delta_{jk}+b_jb_l\delta_{ik}+b_jb_k\delta_{il}-4b_ib_jb_kb_l)$$
$$+\eta_2\,(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}-\delta_{ij}\delta_{kl}+b_ib_j\delta_{lk}+b_lb_k\delta_{ij}+b_ib_jb_kb_l-b_ib_k\delta_{jl}-b_ib_l\delta_{jk}-b_jb_k\delta_{il}-b_jb_l\delta_{ik}), \qquad (22)$$
$$\eta=[d^{(0)}+d^{(1)}+d^{(2)}+d^{(-1)}+d^{(-2)}],\quad \eta_1=d^{(1)}-d^{(0)},\quad \eta_2=d^{(2)}-d^{(0)}.$$

In the zero-order approximation, the coefficient η in (22) is identical with the scalar viscosity coefficient in the absence of the magnetic field, and the quantities η_1 and η_2 are zero. (The coefficients d_m are then equal to one another.) The ansiotropic part of the viscosity tensor appears because the zeroth eigenfunctions of the form $\chi_{\alpha 0}\propto Y_{20}(\mathbf{M}/M)$ become mixed with functions of the form $\chi_{\alpha 0}\propto Y_{20}(\mathbf{v}/v)$. This mixing leads to a correction of the first order in $\Delta\nu/\nu$ to the coefficient $d^{(0)}$. The corrections to the coefficients $d_{m\neq 0}$ appear only in the second order of perturbation theory in the parameter $\Delta\nu/\nu$. As a result, in first-order perturbation theory, the coefficients η_1 and η_2 are equal and of the order of $\eta\Delta\nu/\nu$. In the absence of the magnetic field, the viscosity tensor contains additional terms due to the admixture of the functions $Y_{2m}(\mathbf{v}/v)$ to the zeroth eigenfunctions of the form $Y_{2m}(\mathbf{M}/M)$, where $m\neq 0$. This contribution is also of the order of $\eta\Delta\nu/\nu$.

Since the above contribution is positive for the elements of the tensor η_{ijij}, we may conclude that the application of the magnetic field produces a reduction in the tensor components η_{ijij} by an amount of the order of $\eta\Delta\nu/\nu$, and thus a reduction in the width Γ_3 and Γ_4 by the amount $\Gamma_3\Delta\nu/\nu$.

We note that this change in the width Γ_3 and Γ_4 that occurs when the magnetic field is applied is greater than the change in Γ_5 because it arises in lower order of the perturbation theory in $\Delta\nu/\nu$. This is so because, by virtue of the invariance of the collision operator under space inversion, the operator $\hat{K}$ connects the functions $Y_{lm}(\mathbf{v}/v)$ and $Y_{lm}(\mathbf{M}/M)$ only when l is even. Hence, in the case of thermal conductivity ($l=1$), this matrix element is zero, whereas, for shear viscosity ($l=2$), it is not. The result is that the change in the shear viscosity in the magnetic field is proportional to $\Delta\nu/\nu$, and the change in the thermal conductivity to $(\Delta\nu/\nu)^2$. In our view, the authors of Ref. 3 did not adequately justify the assumption that any matrix element of the form $\langle Y_{lm}(\mathbf{v}/v)|\hat{K}\,|Y_{lm}(\mathbf{M}/M)\rangle$ is zero, so that the changes in shear viscosity and thermal conductivity in the magnetic field are proportional to $(\Delta\nu/\nu)^2$.

We shall now use the expressions for η_{ijkl} given by (22) to calculate Γ_3, Γ_4, Γ_{34}, Γ_{43}:

$$\Gamma_3=(q^2/\rho)\,[\eta+\eta_1+(\eta_2/4-\eta_1)\sin^2 2\theta],$$
$$\Gamma_4=(q^2/\rho)\,[\eta+\eta_1+(\eta_2-\eta_1)\sin^2\theta], \qquad (23)$$
$$\Gamma_{34}=\Gamma_{43}=0,\quad \cos\theta=\mathbf{be}_x,\quad \sin\theta=\mathbf{be}_y.$$

As can be seen from (12), the shear modes are independent of one another in the chosen coordinate frame, and manifest themselves in the depolarized scattered spectrum only in the form of narrow valleys with intensities proportional to $(\Delta c_i)^2$, $i=3,4$. Substituting the explicit form of the functions $\chi_{\alpha m}$ into the expression (14) for Δc_i, and recalling that only the functions $\chi_{\alpha m}$ with $m=0$ contribute to the sum over m, we obtain

$$(\Delta c_4)^2\propto(\mathbf{be}_z)^2(\mathbf{bq})^2(\mathbf{e}_1\mathbf{b})^2(\mathbf{e}_2\mathbf{b})^2=0,$$
$$(\Delta c_3)^2\propto(\mathbf{be}_y)^2(\mathbf{bq})^2(\mathbf{e}_1\mathbf{b})^2(\mathbf{e}_2\mathbf{b})^2\propto(\mathbf{e}_1\mathbf{b})^2(\mathbf{e}_2\mathbf{b})^2\sin^2 2\theta. \qquad (24)$$

It is clear from (24) that the valley in the depolarized spectrum appears only for the mode χ_3, and has the maximum depth when the angle between **b** and **q** is $\theta=45°$. The valley vanishes when **b** is parallel or perpendicular to **q**. The contrast of the valley is

$$\frac{(\Delta c_3)^2}{c_\alpha^2}\frac{\Delta\nu}{\Gamma_3}\sim\left(\frac{\Delta\nu}{\nu}\right)^3,$$

just as in the absence of the magnetic field,[5] and is independent of the gas density. The dependence of the valley intensity on the polarizations of the incident and scattered waves [Eq. (24)] is the same as for the intensity of the main component of depolarized scattering [see (7)]. The valley contrast is therefore the same for the depolarized I_{VH} and I_{HH} components of the scattered radiation (V and H denote, respectively, the vertical and horizontal directions of polarization of the incident and scattered radiation). We note that the valley does not appear at all in the absence of the I_{HH} component.[5]

By studying the dependence of the valley width on the direction of the magnetic field, it is possible to separate experimentally the isotropic from the anisotropic components in the expression for the valley width Γ_3. Within the framework of perturbation theory in the parameter $\Delta\nu/\nu$, this is insufficient for a complete description of all the components of the viscosity tensor. In fact, when the rotational degrees of freedom are taken into account in the first order in $\Delta\nu/\nu$, it is clear from (21) that the result is a change in the coefficient d_0 alone, and the sign of the change is positive. Hence, using (22), we obtain $\eta_1=\eta_2=-\Delta\eta$, where $\Delta\eta>0$ is the change in η when the rotational degrees of freedom are turned on, $\eta=\eta(v)+\Delta\eta$, and the width of the valley assumes the form

$$\Gamma_3=(q^2/\rho)\,[\eta(v)+{}^3/{}_4\Delta\eta\sin^2 2\theta]. \qquad (25)$$

Thus, the θ-independent part of the valley width governs the component of the viscosity that depends only on translational degrees of freedom. The positive increment $\Delta\eta$ can then be determined from the anisotropic part of the width Γ_3, and hence we can find all three viscosity coefficients ($\eta_1=\eta_2=-\Delta\eta$, $\eta=\eta(v)+\Delta\eta$), and thus completely characterize the magnetic-field dependence of the viscosity tensor.

We now proceed to the magnetic-field dependence of the width Γ_1 of the acoustic mode. Using (13), we write

$$\Gamma_1=\sum_{ij}\frac{1}{2\rho}q_iq_j\xi_{ij},$$
$$\xi_{ij}=\rho\sum_{\substack{\alpha>5\\ m}}\frac{\langle\sqrt{2}\chi_1|v_i|\chi_{\alpha m}\rangle\langle\chi_{\alpha m}|v_j|\sqrt{2}\chi_1\rangle}{\nu_{\alpha m}}, \qquad (26)$$
$$\chi_1=\frac{1}{\sqrt{2}}\left(\frac{c_v}{c_p}\right)^{1/2}\left[1-\left(\frac{c_p}{c_v}\right)^{1/2}\frac{v_x}{v_0}+\frac{1}{c_v^{1/2}}\frac{\varepsilon-\bar{\varepsilon}}{\Delta\varepsilon}\right].$$

We shall now take into account the following properties of the eigenfunctions of the operator $\tilde{K}$: (1) the functions $\chi_{\alpha m}$ must contain the **v** harmonics of the same parity because of the invariance of the operator $\tilde{K}$ under space inversion and (2) the functions $\chi_{\alpha m}$ are orthogonal to the five hydrodynamic functions. By using these properties, we can extract from the tensor ξ_{ij} the terms that are due to the conductivity and viscosity tensors $\varkappa_{ij}$ and η_{ijkl}:

$$\xi_{ij}=\rho\sum_{\substack{\alpha>5\\ m}}\frac{\langle v_x/v_0+c_v^{-1/2}\chi_5|v_i|\chi_{\alpha m}\rangle\langle\chi_{\alpha m}|v_j|v_x/v_0+c_v^{-1/2}\chi_5\rangle}{\nu_{\alpha m}}$$

$$=\varkappa_{ij}m\left(\frac{1}{c_v}-\frac{1}{c_p}\right)+\eta_{ixjx}+\delta_{ix}\delta_{jx}\zeta+\frac{\rho}{3}\delta_{ix}\times\sum_{\alpha>5}\frac{1}{v_0^2}\frac{\langle v^2|\chi_{\alpha 0}\rangle\langle\chi_{\alpha 0}|v_jv_x-{}^1/{}_3\delta_{jx}v^2\rangle}{\nu_{\alpha 0}}+\frac{\rho}{3}\delta_{jx}\sum_{\alpha>5}\frac{1}{v_0^2}\frac{\langle v_iv_x-{}^1/{}_3\delta_{ix}v^2|\chi_{\alpha 0}\rangle\langle\chi_{\alpha 0}|v^2\rangle}{\nu_{\alpha 0}},\qquad(27)$$

where

$$\zeta=\frac{\rho}{9}\sum_{\alpha>5}\frac{1}{v_0^2}\frac{\langle|v^2|\varphi_{\alpha 0}{}^{00}|^2}{\nu_{\alpha 0}}$$

is the usual second viscosity coefficient that is independent of the direction of the magnetic field.[2] The last two terms in (27) give the additional contribution to the second viscosity that depends on the direction of the magnetic field and vanishes when we take the average over the direction of **b**. Substituting the explicit expressions for the $\chi_{\alpha 0}$ from (10) into these terms, and extracting the angular dependence of **v**, we obtain

$$\xi_{ij}=\varkappa_{ij}m\left(\frac{1}{c_v}-\frac{1}{c_p}\right)+\eta_{ixjx}+\delta_{ix}\delta_{jx}\zeta+\zeta_1\{b_ib_j(\delta_{ix}+\delta_{jx})-{}^2/{}_3\delta_{ix}\delta_{jx}\},$$

$$\zeta_1=\frac{2}{3}\cdot\frac{1}{15}\rho\sum_{\alpha>5}\frac{1}{v_0^2}\frac{\langle v^2|\varphi_{\alpha 0}{}^{00}\rangle\langle\varphi_{\alpha 0}{}^{20}|v^2\rangle}{\nu_{\alpha 0}},\qquad(28)$$

$$\Gamma_1=\frac{1}{2c_v}\Gamma_5+\frac{q^2}{2\rho}\zeta+\frac{q^2}{2\rho}\zeta_1\left\{\cos^2\theta-\frac{1}{3}\right\}+\frac{q^2}{2\rho}\left\{\frac{4}{3}\eta+\eta_1\sin^2 2\theta+\frac{4}{3}\eta_2\sin^4\theta\right\}.$$

It is clear from (28) that the width Γ_1 contains contributions to the isotropic and anisotropic components of thermal conductivity and shear viscosity as well as terms due to second viscosity. Estimates of ζ_1,ζ yield $\zeta_1\sim\eta\Delta\nu/\nu\sim\zeta\,(\Delta\nu/\nu)^2$, i.e., the anisotropic part of Γ_1, due to second viscosity, is of the same order as the corresponding increment due to shear viscosity. However, these increments have different angular dependence. The anisotropic increment due to the thermal conductivity appears in the next order in the parameter $\Delta\nu/\nu$, so that its contribution to the linewidth Γ_1 need not be taken into account. The diagonal elements of the tensor ξ_{ij} are reduced when the magnetic field is applied, and this is responsible for the corresponding reduction in the linewidth Γ_1 by the amount $\Delta\Gamma\sim(\Delta\nu/\nu)\Gamma_1$. The dependence of $\Gamma_1,\Gamma_3,\Gamma_5$ on the direction of the magnetic field is shown in Fig. 2.

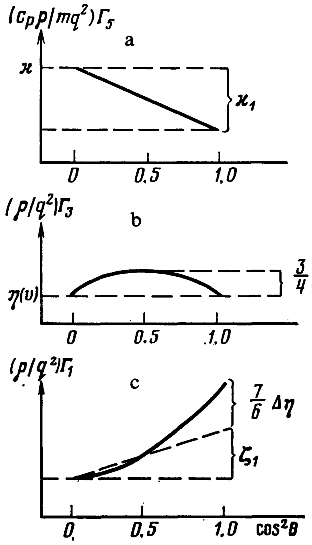

FIG. 2. The widths $\Gamma_5,\Gamma_3,\Gamma_1$ as functions of the angle θ between the direction of the magnetic field and the scattering vector **q**: a—$\varkappa=\varkappa_\perp$ is the value of the thermal conductivity when the magnetic field is perpendicular to the temperature gradient; $\varkappa+\varkappa_1=\varkappa_\parallel<\varkappa_\perp$, $\varkappa_1/\varkappa\sim(\Delta\nu/\nu)^2$ and $\varkappa_\parallel$ is the thermal conductivity when the magnetic field is parallel to the temperature gradient; b—$\eta(v)$ is the component of shear viscosity that is associated with only the translational degrees of freedom of the molecule; $\Delta\eta$ is the contribution to the shear viscosity due to the rotational degrees of freedom; c—$\xi=\varkappa_2m/c_pc_v+\frac{1}{2}\zeta-\frac{1}{3}\zeta_1+\frac{2}{3}\eta(v)-\frac{1}{2}\Delta\eta$, ζ_1 is the anisotropic part of the second viscosity.

Finally, we must consider the quantity Δc_1 that determines the intensity of the valleys in the depolarized spectrum at acoustic frequencies. Using (14) for Δc_1, we obtain

$$(\Delta c_1)^2\infty(e_1b)^2(e_2b)^2\left\{A_1-\frac{1}{3}A_2+A_2\cos^2\theta\right\}^2,$$

$$A_1=\sum_{\alpha>5}\frac{\langle v^2|\varphi_{\alpha 0}{}^{00}\rangle\langle\varphi_{\alpha 0}{}^{02}|1\rangle}{\nu_{\alpha 0}},$$

$$A_2=\frac{2}{15}\sum_{\alpha>5}\frac{\langle v^2|\varphi_{\alpha 0}{}^{20}\rangle\langle\varphi_{\alpha 0}{}^{02}|1\rangle}{\nu_{\alpha 0}}.\qquad(29)$$

It is clear from (29) that the dependence of the valley intensity $(\Delta c_1)^2$ on the angle θ between the vectors **b** and **q** differs from the corresponding angular dependence for the quantity $(\Delta c_3)^2$ in (24). In particular, Δc_1 vanishes when $\cos^2\theta=1/3-A_1/A_2$, i.e., the angle for which the valleys on the acoustic components disappear is determined by the ratio A_1/A_2, so that, in contrast to Δc_3, this angle depends on the particular properties of the gas. Using perturbation theory in the parameter $\Delta\nu/\nu$, it is readily verified that $A_1/A_2\sim\Delta\nu/\nu$.

Detailed examination of the structure of the spectrum of Rayleigh scattering of light by a dense gas ($\nu\gg qv$, which corresponds to gas pressures $P\gtrsim 1$ atm), enables us to determine experimentally the magnetic-field dependence of all the transport coefficients (thermal conductivity, shear viscosity, and second viscosity). The magnetic field for which the above calculations are valid is defined by condition (6), and its order of magnitude for paramagnetic molecules (for example, oxygen) is $B>500$ Oe for $P\simeq 1$ atm and $\gamma B/T\sim 5\times10^{-4}$. For $T\simeq 10$ atm and $\gamma B/T\sim 5\times10^{-3}$, the field is $B>5000$ Oe; for nonparamagnetic molecules (such as nitrogen), $B>10^6$ Oe for $P\simeq 1$ atm and $\gamma B/T\sim 10^{-4}$.

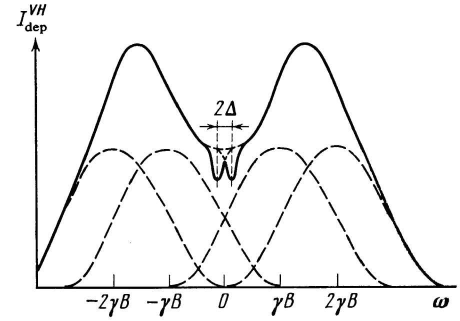

FIG. 3. Fine structure of the VH component of the depolarized scattered spectrum in a magnetic field $\gamma B \sim \Delta\nu$, $\mathbf{b}\|\mathbf{q}$. 2Δ is the splitting of the shear mode χ_3 and χ_4 due to effects that are odd in the magnetic field [see (30)].

4. EFFECT ON THE STRUCTURE OF THE SCATTERED SPECTRUM OF EFFECTS THAT ARE ODD IN THE MAGNETIC FIELD

It is well-known that, when the magnetic field is introduced, the transport-coefficient tensors acquire terms that are odd in the magnetic field.[1–4] These terms vanish in the limit of sufficiently strong fields ($\gamma B \gg \nu$). It is readily verified [cf. (16) and (26)] that the widths Γ_5 and Γ_1 of the Rayleigh and Mandel'shtam-Brillouin components of the polarized scattered radiation are insensitive to these effects. Odd effects can appear in the scattered spectrum only in the fine structure of the depolarized constituent at the unshifted frequency.

When the magnetic field is not too strong ($\gamma B \sim \Delta\nu$), the shear viscosity tensor η_{ijkl} is characterized by five independent quantities, two of which, Im d_1 and Im d_2 [cf. Eq. (7.2) in Ref. 3], characterize the effects that are odd in the field. These terms are most clearly defined in the spectrum when the magnetic field lies along the scattering vector ($\mathbf{b}\|\mathbf{q}$). The solution of the secular equation for the modes χ_3 and χ_4 then leads to two complex-conjugate eigenvalues:

$$\begin{aligned}\lambda_1&=(q^2/\rho)\{\mathrm{Re}d_1+i(\mathrm{Im}d_1+{}^1/_2\mathrm{Im}d_2)\}=\Gamma+i\Delta,\\ \lambda_2&=(q^2/\rho)\{\mathrm{Re}d_1-i(\mathrm{Im}d_1+{}^1/_2\mathrm{Im}d_2)\}=\Gamma-i\Delta.\end{aligned} \tag{30}$$

The appearance of these two complex-conjugate eigenvalues signifies that the fine structure of the depolarized constituent of the scattered spectrum contains two valleys with equal widths Γ that are shifted relative to the unshifted frequency by the amount $\pm\Delta$. The ratio of the parameters Δ and Γ is then given by the following order-of-magnitude relation:

$$\frac{\Delta}{\Gamma}\sim\left(\frac{\Delta\nu}{\nu}\right)^2\frac{\xi^2}{1+\xi^2},\quad \xi=\frac{\gamma B}{\Delta\nu}.$$

Thus, by determining Δ from the splitting in the spectrum, we can determine the quantity Im d_1 + 1/2 Im d_2 and thus estimate the contribution of odd effects to the shear viscosity tensor. When $\mathbf{b}\|\mathbf{q}$, the depolarized spectrum in the presence of the magnetic field consists of four components at the frequencies $\pm\gamma B$ and $\pm 2\gamma B$ for the VH polarization, and three components at the frequencies $\pm 2\gamma B$ and $\omega = 0$ for the HH polarization. The above fine-structure splitting effect should be looked for in the VH-polarized scattered radiation because the fine structure itself appears for $\mathbf{b}\|\mathbf{q}$ only because of the admixture of the shear modes χ_3,χ_4 to functions of the form $Y(\mathbf{M}/M)$, which contribute to depolarized scattering at frequencies $\pm\gamma B$. The intensity of the two split profiles is the same. Figure 3 illustrates this situation in a qualitative way. The fine structure does not appear at all in the case of HH polarization because the two components of the depolarized spectrum at the frequencies $\pm\gamma B$ are then absent. We note that the spectrum is symmetric relative to the frequency $\omega = 0$. Hence, a change in the sign of the magnetic field, equivalent to the substitution $\omega \to -\omega$, has no effect on the form of the spectrum.

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[1)]This situation is analogous to the splitting of purely rotational Raman scattering from the Rayleigh component. A transformation is then made from the description in terms of the Euler angles characterizing the orientation of the molecule in space to the description in terms of the angular momentum vector $\mathbf{M}$ averaged over the rotational period of the molecule.

[2)]It is readily shown that this definition of the second viscosity is the same as the generally accepted one[15]

$$\zeta=\frac{\rho}{mT}\left\langle\frac{mv^2}{3}-\frac{\varepsilon(\Gamma)}{c_v}\right|\hat{\hat{K}}^{-1}\left|\frac{mv^2}{3}-\frac{\varepsilon(\Gamma)}{c_v}\right\rangle$$

subject to the normalization adopted above, namely,

$$\int f_0 d\Gamma=1,\qquad \hat{K}^{-1}|\varepsilon(\Gamma)\rangle=0.$$

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