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Collision anisotropy and impact contour of spectral lines

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An impact theory of spectral-line broadening is constructed for transitions between degenerate states and anisotropic collisions. It is shown that the line contour for the subensemble over atoms with a given velocity consists of a set of Lorentz components that differ in width and in position. The number of components increases with increasing angular momentum of the combining states. The factors that mask the line splitting because of the anisotropy of the collisions, and the results of numerical calculations for Van der Waals and dipole-dipole interactions, are discussed.

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1. INTRODUCTION

It is customarily assumed that the impact contour of the spectral line corresponding to a transition between a pair of isolated levels has a Lorentz shape (see, e.g., Ref. 1). Actually, however, this conclusion of the theory presupposes a spherical symmetry of the average perturbation of the atomic oscillator by the buffer particles, i.e., the latter should be unpolarized (as is practically always the case), have an isotropic velocity distribution, and in addition, their average velocities $\bar{v}_b$ should greatly exceed the average velocities $\bar{v}$ of the radiating particles:

$$\rho_b(\mathbf{v}_b)=\rho_b(|\mathbf{v}_b|);\ \bar{v}_b\gg\bar{v}. \tag{1.1}$$

In other words, when the broadening cross section is averaged the radiating particles should be assumed immobile, while the buffer particles should be assumed to move isotropically. The conditions (1.1) serve as a formulation of the so called isotropic-collision (or isotropic-perturbation) model,[2-6] and if they are violated, the spectral lines have a more complicated structure, which in fact will be dealt with in the present article.

To explain the gist of the matter, we change over to a coordinate system connected with the radiating atom, which moves with velocity $\mathbf{v}$. In this frame a "wind" of buffer particles moving with group velocity $-\mathbf{v}$ blows, as it were, around the radiating atom. It is clear that the perturbation of the wave-emission process will have axial rather than spherical symmetry, and the symmetry axis is collinear with $\mathbf{v}$. For the electronic broadening, the conditions (1.1) are obviously satisfied. In broadening due to collisions with atoms, molecules, and ions, the model of isotropic collisions is adequate for the relatively lighter buffer gas, and its applicability is doubtful in the case of heavy perturbing particles.

The simplest manifestation of the "wind effect" consists in the fact that the impact width and the line shift turn out to depend on the velocity of the radiating atom. This dependence, discussed in the paper of Sobel'man and one of us,[7] and in many succeeding papers,[8-13] is the only consequence of the wind effect in the absence of collisional disorientation. In the opposite case, the line for atoms with a given velocity does not have a Lorentz shape.

The change of the line shape as a result of the anisotropy of the collisions was first established by Kazantsev[14] and was investigated in greater detail by Vdovin and Galitskiĭ.[15] In both papers they considered a transition between levels with total angular momentum 1 and 0, and the perturbation was due to the resonant dipole–dipole interaction, so an appreciable change in velocity was also taken into account. For the simpler model of relaxation constants, but also for the transition 1–0, it was shown in Refs. 16 and 17 that the line contour consists of two Lorentz components. In a recent paper, Baranova, Zel'dovich, and Yakovleva[18] predicted for Raman scattering of light polarization singularities also due to the anisotropy of the collisions. We shall note some related phenomena. The wind effect manifests itself in the production of the "hidden alignment" observed by Chaĭka.[19] The general theory of this phenomenon is discussed in article by D'yakonov and Perel'.[6] The alignment due to collisions with an atomic beam and leading to unique singularities of the fluorescence polarization was analyzed by Rebane.[20]

According to the theory developed below, the anisotropy of the collisions leads to a splitting of the spectral lines, and the number of the components is proportional to the angular momenta of the combining states. This raises naturally the question of why such a universal phenomenon was not observed (and still remains unobserved) during the many decades of experimental research, which led to accumulation of a tremendous empirical material on the contours of spectral lines and which serve as the basis of many conclusions on the physics of collisions. This question, which is fundamental for spectroscopy and atomic physics, is discussed in Sec. 4 after the exposition of the theory of the relaxation matrix (Sec. 2) and of the linear broadening theory (Sec. 3).

2. RELAXATION MATRIX

In the absence of a change of velocity in the collisions, the relaxation is described by a matrix whose elements can be written in the following form ($\varkappa q$ representation or representation of irreducible tensor operators):

$$\Gamma(JJ'\varkappa q|J_1J_1'\varkappa_1 q_1;\mathbf{v})=N_b\sum_{n_{b1}\mu q'}\int\rho_b(n_{b1}|\mathbf{v}-\mathbf{u}|)\,u\,d\mathbf{u}\,d\rho$$
$$\times D^{\varkappa}_{qq'}(\hat{\mathbf{v}},0)D^{\varkappa_1 *}_{q_1q'}(\hat{\mathbf{v}},0)D^{\varkappa}_{q'\mu}(0\beta0)D^{\varkappa_1 *}_{q'\mu}(0\beta0)\,\tilde{\sigma}(JJ'\varkappa\mu|J_1J_1'\varkappa_1\mu;n_{b1},\rho,u). \tag{2.1}$$

Here N_b, $\rho_b(n_b, |\mathbf{v}-\mathbf{u}|)$ are the concentration and distribution of the perturbing particles over the quantum numbers n_b and the velocities $\mathbf{v}_b=\mathbf{v}-\mathbf{u}$, and $\mathbf{u}$ is the relative velocity. The integral of $\tilde{\sigma}(\ldots)$ with respect to the impact parameter ρ determines the cross section, which we shall assume for the sake of simplicity to be calculated in the approximation of straight-line trajectories (although the latter is not obligatory):

$$\tilde{\sigma}(JJ'\varkappa\mu|J_1J_1'\varkappa_1\mu_1;n_{b1},\rho,u)=\sum_{\substack{MM_1M'M_1'\\ b,\nu_{b1}}}\{\delta_{JJ_1}\delta_{MM_1}\delta_{J'J_1'}\delta_{M'M_1'}\delta_{bb_1}$$
$$-\tilde{S}(JMb|J_1M_1b_1;\rho,u)\tilde{S}^*(J'M'b|J_1'M_1'b_1;\rho,u)\}$$
$$\times(-1)^{J'-M'+J_1'-M_1'}\langle JMJ'-M'|\varkappa\mu\rangle\langle J_1M_1J_1'-M_1'|\varkappa_1\mu_1\rangle. \tag{2.2}$$

The perturbing particles are assumed to be unpolarized (ρ_b does not depend on the projection ν_b of the spins, $b\equiv n_b, \nu_b$; n_b are scalar quantum numbers).

The functions $\tilde{S}$ and $\tilde{S}^*$ in (2.2) are the elements of the scattering matrix referred to the $\mathbf{u}$-system (the z axis in it is directed along $\mathbf{u}$). The Wigner D matrices in (2.1) describe the successive transitions from the laboratory frame into the $\mathbf{v}$ and $\mathbf{u}$ systems, so that β denotes the angle between $\mathbf{u}$ and $\mathbf{v}$. The quantity $\tilde{\sigma}(\ldots)$ does not depend on the orientation of the $\mathbf{u}$-system in space; in addition, $\rho_b(n_b, |\mathbf{v}-\mathbf{u}|)$ depends only on β, but not on the azimuthal angle of $\mathbf{u}$. These circumstances are the causes of the equality of some of the indices of the D matrices and of the zero values of some of their arguments. For simplicity we shall leave out the indices J from $\tilde{\sigma}(\ldots)$ and $\Gamma(\ldots)$.

It is easy to illustrate the role of the wind effect by considering (2.1) for two limiting relations between the average velocities $\bar{v}$ and $\bar{v}_b$ of the radiating and buffer particles. If $\bar{v}\ll\bar{v}_b$ (the isotropic-perturbation model), then the relative velocity practically coincides with v_b, and we can discard $\mathbf{v}$ from $|\mathbf{v}-\mathbf{u}|$. Integration of the D matrices with respect to β leads then to $\delta_{\varkappa\varkappa_1}$, and expression (2.1) reduces to

$$\Gamma(\varkappa q|\varkappa_1q_1;\mathbf{v})=\delta_{\varkappa\varkappa_1}\delta_{qq_1}\Gamma_\varkappa;$$
$$\Gamma_\varkappa=N_b\sum_{n_b}\int\rho_b(n_b,u)\,u\,d\mathbf{u}\frac{1}{2\varkappa+1}\sum_\mu\int\tilde{\sigma}(\varkappa\mu|\varkappa_1\mu;n_b,\rho,u)\,d\boldsymbol{\rho}, \tag{2.3}$$

i.e., the relaxation matrix is diagonal in $\varkappa q$ and does not depend on q or $\mathbf{v}$.

In the opposite limit $\bar{v}\gg\bar{v}_b$, the relative velocity $\mathbf{u}$ practically coincides with $\mathbf{v}$, i.e.,

$$\rho_b(n_b,|\mathbf{v}-\mathbf{u}|)=\rho_b(n_b)\delta(\mathbf{v}-\mathbf{u}).$$

Under these conditions $\beta=0$, $D^{\varkappa}_{q\mu}(0)=\delta_{q\mu}$ and therefore

$$\Gamma(\varkappa q|\varkappa_1q_1;\mathbf{v})=N_bv\sum_{n_b}\rho_b(n_b)$$
$$\times\sum_\mu D^{\varkappa}_{q\mu}(\hat{v},0)D^{\varkappa_1 *}_{q_1\mu}(\hat{v},0)\int\tilde{\sigma}(\varkappa\mu|\varkappa_1\mu;n_b,\rho,v)\,d\boldsymbol{\rho}. \tag{2.4}$$

Expression (2.4) corresponds to almost immobile perturbing particles, and therefore the argument of the cross section is the velocity v. In contrast to (2.3), the quantities $\Gamma(\varkappa q|\varkappa_1q_1;\mathbf{v})$ in (2.4) are not diagonal in $\varkappa q$, and depend on q and $\mathbf{v}$; it is furthermore quite clear that the wind effect is the common cause of the aforementioned singularities: the dependence on both q and q_1 as well as on $\hat{v}$ is concentrated in the D matrices.

It must be emphasized that the anisotropic properties of the relaxation matrix manifest themselves only to the extent to which the collisional disorientation is significant. Actually, if the latter is absent, then it follows directly from the expression of the relaxation matrix in the JM representation

$$\Gamma(JMJ'M'|J_1M_1J_1'M_1';\mathbf{v})=N_b\sum_{n_b}\int\rho_b(n_b,|\mathbf{v}-\mathbf{u}|)$$
$$\times\sigma(JMJ'M'|J_1M_1J_1'M_1';n_b,\rho,u)\,u\,d\mathbf{u}\,d\rho, \tag{2.5}$$

that $\Gamma(\ldots)\propto\delta_{MM_1}\delta_{M'M_1'}$ if $\sigma\propto\delta_{MM_1}\delta_{M'M_1'}$. In this case the only manifestation of the wind effect is the dependence of $\Gamma(\ldots)$ on v.

For the discussions that follow it is convenient to rewrite (2.1) in a different form, wherein the tensor properties of the relaxation matrix are explicitly separated:

$$\Gamma(\varkappa q|\varkappa_1 q_1;\,\mathbf{v})=\sum_{LN}D_{N0}{}^{L}(\hat{v},0)\,(-1)^{\varkappa_1-q_1}\langle\varkappa q\varkappa_1-q_1|LN\rangle$$
$$\times[(2\varkappa+1)(2\varkappa_1+1)]^{1/2}\Gamma(JJ'\varkappa,J_1J_1'\varkappa_1,L;\,v). \tag{2.6}$$

We introduce here the notation

$$\Gamma(JJ'\varkappa,J_1J_1'\varkappa_1,L;\,v)\equiv\Gamma(\varkappa\varkappa_1L;\,v)$$
$$=N_b\sum_{n_b}\int_0^\infty \rho_b(n_b,L,v,u)\,u^3du\int\sigma(\varkappa\varkappa_1L;\,n_b,\rho,u)\,d\rho, \tag{2.7}$$

$$\rho_b(n_b,L,v,u)=2\pi\int_0^\pi \rho_b(n_b,|\mathbf{v}-\mathbf{u}|)D_{00}^{L}(0\beta0)\sin\beta d\beta, \tag{2.8}$$

$$\sigma(\varkappa\varkappa_1L;\,n_b,\rho,u)=[(2\varkappa+1)(2\varkappa_1+1)]^{-1/2}$$
$$\times\sum_{\mu}(-1)^{\varkappa_1-\mu}\langle\varkappa\mu\varkappa_1-\mu|L0\rangle\sigma(\varkappa\mu|\varkappa_1\mu;\,n_b,\rho,u). \tag{2.9}$$

L is the angular momentum of the individual term in the expansion of $\rho_b(n_b,|\mathbf{v}-\mathbf{u}|)$ in spherical functions. $L=0$ corresponds to the isotropic part of the distribution, and $L>0$ to the anisotropic part. The elements that are not diagonal in $\varkappa$ contain only anisotropic terms while the diagonal elements contain both isotropic and anisotropic terms. The normalization in (2.6) is chosen such that the isotropic part ($L=0$), which is diagonal in $\varkappa q$ and does not depend on q, coincides with $\Gamma(\varkappa\varkappa 0;v)$ and with the value of $\Gamma(\varkappa q|\varkappa q;\mathbf{v})$ averaged over q or over $\hat{v}$:

$$\Gamma(\varkappa\varkappa0;\,v)=\frac{1}{2\varkappa+1}\sum_q\Gamma(\varkappa q|\varkappa q;\,\mathbf{v})=\frac{1}{4\pi}\int\Gamma(\varkappa q|\varkappa q;\,\mathbf{v})\,d\hat{v}. \tag{2.10}$$

Let us list some general consequences of (2.6). In the v-system the relaxation matrix is diagonal in q:

$$\Gamma(JJ'\varkappa q|J_1J_1'\varkappa_1q_1;\,\mathbf{v})=\delta_{qq_1}\Gamma_{\varkappa\varkappa_1}{}^{q}(v),$$
$$\Gamma_{\varkappa\varkappa_1}^{q}(v)=\sum_L(-1)^{\varkappa_1-q}\langle\varkappa q\varkappa_1-q|L0\rangle[(2\varkappa+1)(2\varkappa_1+1)]^{1/2}\Gamma(\varkappa\varkappa_1L;\,v). \tag{2.11}$$

The change of the sign of q and q_1 leads to the equation

$$\Gamma(\varkappa-q|\varkappa_1-q_1;\,\mathbf{v})=(-1)^{\varkappa-\varkappa_1+q-q_1}e^{2i(q_1-q)\varphi}\Gamma(\varkappa q|\varkappa_1q_1;\,-\mathbf{v}), \tag{2.12}$$

where φ is the azimuthal angle of $\mathbf{v}$ in the laboratory frame.

In the approximation (2.2) the differences of the energies of the states J and J_1, J' and J_1', as well as b and b_1 is much smaller than kT; under these conditions it follows from the reciprocity theorem

$$\Gamma(JJ'\varkappa q|J_1J_1'\varkappa_1q_1;\,\mathbf{v})=(-1)^{\varkappa-\varkappa_1+q-q_1}\Gamma(J_1J_1'\varkappa_1-q_1|JJ'\varkappa-q;\,-\mathbf{v}). \tag{2.13}$$

Combining the properties (2.12) and (2.13) we arrive at the relation

$$\Gamma(JJ'\varkappa q|J_1J_1'\varkappa_1q_1;\,\mathbf{v})=e^{2i(q_1-q)\varphi}\Gamma(J_1J_1'\varkappa_1q_1|JJ'\varkappa q;\,\mathbf{v}), \tag{2.14}$$

which simplifies in the v system:

$$\Gamma(JJ'\varkappa q|J_1J_1'\varkappa_1q;\,\mathbf{v})\equiv\Gamma_{\varkappa\varkappa_1}{}^{q}(v)=\Gamma_{\varkappa_1\varkappa}^{q}(v)\equiv\Gamma(J_1J_1'\varkappa_1q|JJ'\varkappa q;\,\mathbf{v}). \tag{2.15}$$

The scalar coefficients of the expansion (2.6) satisfy the following symmetry relations:

$$\Gamma(JJ'\varkappa,\,J_1J_1'\varkappa_1,\,L;\,v)=(-1)^{\varkappa-\varkappa_1}\Gamma(J_1J_1'\varkappa_1,\,JJ'\varkappa,\,L;\,v). \tag{2.16}$$

We note finally that with respect to the reversal of the sign of q, the quantities $\Gamma_{\varkappa\varkappa_1}^{q}(v)$ can be resolved into symmetrical and antisymmetrical parts corresponding to even and odd values of L in (2.11):

$$\Gamma_{\varkappa\varkappa_1}^{q}(v)=\Gamma_{\varkappa\varkappa_1}^{qs}(v)+\Gamma_{\varkappa\varkappa_1}^{qa}(v),\quad \Gamma_{\varkappa\varkappa_1}^{-q}(v)=(-1)^{\varkappa-\varkappa_1}[\Gamma_{\varkappa\varkappa_1}^{qs}(v)-\Gamma_{\varkappa\varkappa_1}^{qa}(v)]. \tag{2.17}$$

If parity is conserved in the interaction of the colliding particles, then the only nonzero terms in (2.6) and (2.11) are those with even values of L, $\Gamma_{\varkappa\varkappa_1}^{qa}(v)=0$, and the following additional symmetry relation holds:

$$\Gamma(\varkappa q|\varkappa_1q_1;\,\mathbf{v})=\Gamma(\varkappa q|\varkappa_1q_1;\,-\mathbf{v}),\quad \Gamma_{\varkappa\varkappa_1}^{q}(v)=(-1)^{\varkappa-\varkappa_1}\Gamma_{\varkappa\varkappa_1}^{-q}(v). \tag{2.18}$$

The considered matrix $\Gamma(JJ'\varkappa q|J_1J_1'\varkappa_1q_1;\mathbf{v})$ can be used to describe several different phenomena. In the present article we are interested only in the contour of the spectral line corresponding to a one-photon transition between a pair of isolated levels. In this problem we must put $J=J_1$, $J'=J_1'$. The same relaxation matrix determines also the line contour in multiphoton processes.

The relaxation matrix simplifies substantially if one of the combining levels is not subject to collisional disorientation,[1)] i.e.,

$$S(J'M'b|J'M_1'b_1;\,\rho,u)=\delta_{M'M_1'}S(b|b_1;\,\rho,u). \tag{2.19}$$

Under these conditions the JM representation is more convenient, and we can derive from (2.5) an equation similar to (2.6):

$$\Gamma(JMJ'M'|JM_1J'M_1';\,\mathbf{v})\equiv\Gamma(MM'|M_1M_1';\,\mathbf{v})$$
$$=\delta_{M'M_1'}(2J+1)^{1/2}\sum_{LN}D_{N0}{}^{L}(\hat{v},0)\,(-1)^{J-M_1}\langle JMJ-M_1|LN\rangle\Gamma(JJL;\,v), \tag{2.20}$$

where

$$\Gamma(JJL;\,v)\equiv\Gamma(L,v)=\frac{N_b}{(2J+1)^{1/2}}\sum_{n_{b_1}\mu}\int_0^\infty u^3\rho_b(n_{b_1},L,v,u)\,du$$
$$\times(-1)^{J-\mu}\langle J\mu J-\mu|L0\rangle\int\sigma(J\mu,J';\,n_{b_1},\rho,u)\,d\rho,$$
$$\sigma(J\mu,J';\,n_{b_1},\rho,u)=\sum_{\nu_{b_1},b}\{1-S(J\mu b|J\mu b_1;\,\rho,u)S^*(b|b_1,J',\rho,u)\}. \tag{2.21}$$

The matrix (2.20) has a remarkable property that is very important for practical problems, namely, it is diagonal in the $\mathbf{v}$-system

$$\Gamma(MM'|M_1M_1';\,\mathbf{v})=\delta_{MM_1}\delta_{M'M_1'}\Gamma_M(v),$$
$$\Gamma_M(v)=(2J+1)^{1/2}\sum_L(-1)^{J-M}\langle JMJ-M|L0\rangle\Gamma(L,v). \tag{2.22}$$

It is easy to prove properties similar to (2.10), (2.12)–(2.14), and (2.17):

$$\Gamma(0,v)=\frac{1}{2J+1}\sum_M\Gamma_M(v)=\frac{1}{4\pi}\int\Gamma(MM|MM;\,\mathbf{v})\,d\hat{v}, \tag{2.23}$$

$$\Gamma(-M-M'|-M_1-M_1';\,\mathbf{v})=(-1)^{M-M_1}e^{2i(M_1-M)\varphi}\Gamma(MM'|M_1M_1';\,-\mathbf{v}), \tag{2.24}$$

$$\Gamma(MM'|M_1M_1';\,\mathbf{v})=(-1)^{M-M_1}\Gamma(-M_1-M_1'|-M-M';\,-\mathbf{v})$$
$$=e^{2i(M_1-M)\varphi}\Gamma(M_1M_1'|MM';\,\mathbf{v}), \tag{2.25}$$

$$\Gamma_M(v)=\Gamma_M{}^{s}(v)+\Gamma_M{}^{a}(v),\quad \Gamma_{-M}(v)=\Gamma_M{}^{s}(v)-\Gamma_M{}^{a}(v). \tag{2.26}$$

The symmetrical and antisymmetrical parts are given by the even and odd values of L in (2.22); for interactions that conserve parity, we have $\Gamma_M^a(v)=0$.

The relaxation-matrix properties proved above follow from general symmetry considerations and are in no way connected with the concrete form of the scattering amplitudes. We assume now validity of the "power-law model" frequently used in the impact theory of broadening: the angular variables and the distance between the colliding particles in the interaction potential are separable, and the dependence on the distance follows the power law r^{-s}. In this case the argument of the scattering matrix in (2.2) and (2.21) is the combination $\eta=\rho u^{1/(s-1)}$, which enables us to determine the dependence of $\Gamma(\varkappa\varkappa_1 L; v)$ on v.

Let $\rho_b(n_b, v_b)$ be of the form

$$\rho_b(n_b,\mathbf{v}_b)=\rho_b(n_b)W(\mathbf{v}_b),\quad W(\mathbf{v}_b)=(\sqrt{\pi}\bar{v}_b)^{-3}\exp[-\mathbf{v}_b^2/\bar{v}_b^2]. \tag{2.27}$$

Then

$$\rho_b(n_b,L,v,u)=\rho_b(n_b)\left(\frac{\sqrt{2}}{\bar{v}_b}\right)^3\exp\left\{-\frac{(v^2+u^2)}{\bar{v}_b^2}\right\}\left(\frac{\bar{v}_b^2}{2uv}\right)^{1/2}I_{L+1/2}\left(\frac{2uv}{\bar{v}_b^2}\right), \tag{2.28}$$

where $I_{L+1/2}(z)$ are Bessel functions of the first kind and of imaginary argument, while for the coefficient $\Gamma(\varkappa\varkappa_1 L; v)$, defined by (2.7), we obtain the expressions

$$\Gamma(\varkappa\varkappa_1 L; v)=N_b\bar{v}_b^\beta K(s,L,v)\,2\pi\int_0^\infty\sigma(\varkappa\varkappa_1 L;\eta)\,\eta\,d\eta, \tag{2.29}$$

$$\sigma(\varkappa\varkappa_1 L; v)=\sum_{n_b}\rho_b(n_b)\sigma(\varkappa\varkappa_1 L,n_b,\eta), \tag{2.30}$$

$$K(s,L,v)=\zeta^L\Phi\left(\frac{L-\beta}{2},L+\frac{3}{2};-\zeta^2\right)\Gamma\left(\frac{L+\beta}{2}+\frac{3}{2}\right)\Big/\Gamma\left(L+\frac{3}{2}\right), \tag{2.31}$$

where

$$\beta=\frac{s-3}{s-1},\quad \zeta=\frac{v}{\bar{v}_b},$$

$\Phi(\alpha,\gamma;x)$ is a confluent hypergeometric function.[21] According to (2.31), the isotropic part of the relaxation matrix ($L=0$) retains a finite value at $v=0$. This part reflects the role of the wind effect in the absence of disorientation, while expression (2.29) at $L=0$ agrees with the previously obtained one.[11] For the anisotropic part, on the other hand, the lowest are the expansion terms with $L=1$ or 2, and at small $v/\bar{v}_b$ they behave like $v/\bar{v}_b$ or $(v/\bar{v}_b)^2$. At the same time we have at $v\gg\bar{v}_b$

$$K(s,L,v)\approx(v/\bar{v}_b)^\beta, \tag{2.32}$$

i.e., the asymptotic dependence on v is the same for all L.

From (2.32), just as from the general limiting expression (2.4), it follows that

$$\Gamma^q_{\varkappa\varkappa_1}(v)=N_b v^\beta\cdot 2\pi\int_0^\infty\sigma(\varkappa q|\varkappa_1 q;\eta)\,\eta\,d\eta. \tag{2.33}$$

In view of the monotonic increase (at a rate v) of the anisotropic part of the relaxation matrix, as well as its ratio to the isotropic part, Eq. (2.33) corresponds to the strongest manifestation of the wind effect.

The noted regularities are clearly seen in Fig. 1, which shows plots of the functions $K(s,L,v)/K(s,0,0)$ for $s=3,6$ and $L=0,2,4$. The figure shows also the Maxwellian distributions in $|v|$ for the radiating particles at mass ratios $m_b/m=1/4$, 1 and 4. Comparing the Maxwellian curves 7–9 and the plots 1–6, we note that the wind effect is insignificant at $m_b\lesssim m$.

The dependence on the velocity is described by the function $K(s,L,v)$ also in the model (2.19): the quantity $\Gamma(JJL;v)$ defined by (2.21) is proportional to $K(s,L,v)$, i.e.,

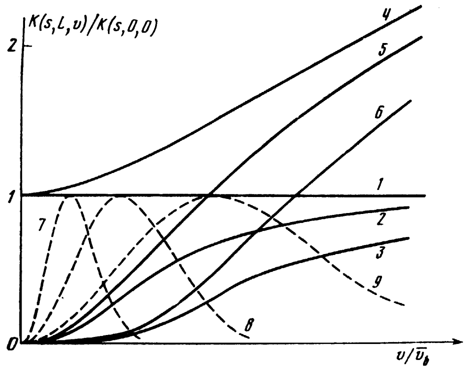

FIG. 1. Plots of the functions $K(s,L,v)/K(s,0,0)$: $s=3$ for curves 1, 2, 3 and $s=6$ for curves 4, 5, 6. Curves 1, 4; 2, 5; and 3, 6 correspont to $L=0$, 2, and 4. Dashed curves —Maxwellian distribution for $|v|$ at $m_b/m=1/4$ (curve 7), 1 (8), and 4 (9).

$$\Gamma(JJL,v)=N_b\bar{v}_b^\beta K(s,L,v)\cdot 2\pi\int_0^\infty\sigma(JJL,\eta)\,\eta\,d\eta,$$

$$\sigma(JJL,\eta)=\sum_{n_b\mu}\rho_b(n_b)(-1)^{J-\mu}\langle J\mu J-\mu|L0\rangle(2J+1)^{-1/2}\sigma(J\mu,J',n_b,\eta). \tag{2.34}$$

3. CONTOUR OF SPECTRAL LINE

The general expression for the specific absorbed (or emitted) power $P(\Omega)$ can be represented in the form

$$P(\Omega)=2\hbar\omega\,\mathrm{Re}\sum_q\langle iG_q^*\rho_{mn}(1q,\mathbf{v})\rangle, \tag{3.1}$$

where

$$\Omega=\omega-\omega_{mn},\quad G_q=dE_q/2\sqrt{3}\hbar,$$

$\rho_{mn}(\varkappa q,\mathbf{v})$ is an element of the density matrix, d is the reduced matrix element of the dipole moment, ω_{mn} is the Bohr frequency for the $m-n$ transition, and E_q is the spherical component of the electric field intensity of the monochromatic wave (frequency ω).

The kinetic equation for $\rho_{mn}(\varkappa q,\mathbf{v})$ (the relaxation-constant model)

$$(\gamma-i\Omega')\rho_{mn}(\varkappa q,\mathbf{v})+\sum_{\varkappa_1 q_1}\Gamma(\varkappa q|\varkappa_1 q_1;\mathbf{v})\rho_{mn}(\varkappa_1 q_1,\mathbf{v})=-i\sum_\sigma B(\varkappa q|1\sigma)G_\sigma;$$

$$\Omega'=\Omega-\mathbf{kv}=\omega-\omega_{mn}-\mathbf{kv} \tag{3.2}$$

contains in its right-hand side G_σ and a certain matrix $B(\varkappa q\,|\,1\sigma)$ that depends on the singularities of the processes of excitation and relaxation of the combining levels (γ is the spontaneous half-width). If it is assumed that the rate of excitation of the levels m and n is isotropic, then as a result of the hidden alignment the matrix $B(\varkappa q\,|\,1\sigma)$ turns out to be nondiagonal.

We consider below a simpler model, corresponding to an equilibrium distribution over the magnetic sublevels

$$B(\varkappa q|1\sigma)=\delta_{\varkappa 1}\delta_{q\sigma}NW(\mathbf{v}), \tag{3.3}$$

where N is the concentration of the absorbing particles and $W(\mathbf{v})$ is their distribution of the velocities, which will be assumed to be Maxwellian. This model is realized, for example, for transitions from the ground state.

It is convenient to solve the kinetic equation (3.2) in the $\mathbf{v}$ system, where the relaxation matrix is diagonal in

q. In this system of coordinates

$$\rho(1q,\mathbf{v})=-iG_qNW(\mathbf{v})R_q(\Omega'),\tag{3.4}$$

$$R_q(\Omega')=[\hat{\Gamma}^q+(\gamma-i\Omega')\hat{E}]^{-1}_{\varkappa=\varkappa_1=1}.\tag{3.5}$$

The subsequent transition into the laboratory system leads to the relations

$$P(\Omega)=2\pi\hbar\omega N\sum_q|G_q|^2I(\Omega),\tag{3.6}$$

$$I(\Omega)=\frac{1}{3\pi}\operatorname{Re}\left\langle\left\{\sum_q R_q(\Omega')+\frac{3}{4}\left(\cos^2\vartheta-\frac{1}{3}\right)\left[\sum_q R_q(\Omega')-3R_0(\Omega')\right]\right.\right.$$
$$\left.\left.+\frac{3}{2}\cos\vartheta[R_1(\Omega')-R_{-1}(\Omega')](\xi_+-\xi_-)\right\}W(\mathbf{v})\right\rangle,\tag{3.7}$$

where ϑ is the angle between $\mathbf{v}$ and the wave vector $\mathbf{k}$, $\xi_\pm = |G_{\pm1}|^2/\Sigma_q|G_q|^2$.

The first two terms in (3.7) do not depend on the polarization, while the third term reverses sign on going from the right to the left-hand polarization ($\xi_+=1,\xi_-=0$ and $\xi_+=0,\xi_-=1$; the quantization axis is along $\mathbf{k}$). The polarization dependence of $I(\Omega)$ vanishes when (2.18) is satisfied, since $R_1(\Omega')=R_{-1}(\Omega')$ in this case. In the absence of Doppler broadening, the only nonzero term is the isotropic term

$$I(\Omega)=\frac{1}{3\pi}\operatorname{Re}\left\langle\sum_q R_q(\Omega')W(\mathbf{v})\right\rangle,\tag{3.8}$$

provided, of course, that $W(\mathbf{v})$ depends only on $|\mathbf{v}|$.

To determine the spectral structure of the contour we note that the elements of the reciprocal matrix (3.5) can be represented in the form of a partial-fraction expansion or as a linear combination of Lorentzians:

$$R_q(\Omega')=\sum_{k=1}^{r_q}Z_k^q[\gamma+\Gamma_k^q-i(\Omega'-\Delta_k^q)]^{-1},\tag{3.9}$$

where $\Gamma_k^q+i\Delta_k^q$ are the eigenvalues of the matrix $\hat{\Gamma}^q$, and the coefficients Z_k^q do not depend on the frequency Ω'. Thus, the anisotropy of the collision causes splitting of the spectral line into several Lorentzian components with different widths and positions of the maxima. This splitting is universal for all processes between isolated levels with arbitrary values of the angular momenta J and J'. For the transition $J=1-J'=0$, considered in Refs. 14–17, the quantities $R_q(\Omega')$ contain one Lorentzian each, and the spectral lines consist of three or two components (the latter for even potentials). In the general case of arbitrary values of J and J' the number of components r can be easily obtained by determining the rank of the relaxation matrix. The results are summarized in the table. With increasing J and J' the number r increases rapidly, approximately like $6J$ (or $3J$).

TABLE I. Number of Lorentz components in the line contour ($J\geqslant J'$).

	Parity conservation			Parity nonconservation		
	r_0	r_1	r	r_0	$r_{\pm1}$	r
Integer J and J' Half-integer J and J'	J $J'+{}^1/_2$	$J+J'$ $J+J'$	$2J+J'$ $J+2J'+{}^1/_2$	$2J'+1$	$J+J'$	$4J'+2J+1$
Disorientation on one of the levels (J)						
Integer J and J' Half-integer J and J'			$J+1$ $J+{}^1/_2$			$2J+1$

In the model (2.19), the relaxation matrix is diagonal in the JM representation, and the line contour consists of Lorentzians whose parameters are given directly by the elements $\Gamma_M(v)$[2)]:

$$I(\Omega)=\frac{1}{\pi}\operatorname{Re}\sum_M\left\langle\left\{\frac{1}{\Gamma_M(v)-i\Omega'}\left[\frac{1}{2J+1}+\frac{3}{4}\left(\cos^2\vartheta-\frac{1}{3}\right)\right.\right.\right.$$
$$\left.\times\left(\frac{1}{2J+1}-\langle JMJ'-M|10\rangle^2\right)\right]+\frac{1}{2}\cos\vartheta\,\langle JMJ'1-M|11\rangle^2$$
$$\left.\left.\times\left[\frac{1}{\Gamma_M(v)-i\Omega'}-\frac{1}{\Gamma_{-M}(v)-i\Omega'}\right](\xi_+-\xi_-)\right\}W(\mathbf{v})\right\rangle.\tag{3.10}$$

Here the number of the components is much less (see the table), and is independent of J'. The value of J' depends only on the weight with which the different Lorentzians enter in $I(\Omega)$.

At relatively small values of $J,J'(J,J'=3/2,2)$ the order of the characteristic levels is higher than the second, and it becomes difficult to determine the eigenvalues of the relaxation matrices in closed form. Interest attaches therefore to approximate calculations of the elements $R_q(\Omega')$. One can regard, for example, the perturbation to be the off-diagonal elements of the matrix $\hat{\Gamma}^q$, and then

$$R_q(\Omega')=\frac{1}{\Gamma_{11}^q-i\Omega'}+\sum_{\varkappa\neq1}\frac{[\Gamma_{1\varkappa}^q]^2}{[\Gamma_{11}^q-i\Omega']^2[\Gamma_{\varkappa\varkappa}^q-i\Omega']}.\tag{3.11}$$

Thus, the off-diagonal elements give a correction of the order of $[\Gamma_{1\varkappa}^q]^2/\Gamma_{11}^q\Gamma_{\varkappa\varkappa}^q$.

We can assume the perturbation to be the entire anisotropic part of $\hat{\Gamma}^q$. Taking into account the first equation of (2.10) and the fact that the isotropic part of $\hat{\Gamma}^q$ is independent of q, we obtain from (3.7) [with the aid of (3.11)]

$$I(\Omega)=\frac{1}{\pi}\operatorname{Re}\left\langle\frac{W(\mathbf{v})}{\Gamma_1-i\Omega'}\left\{1+\sum_{\varkappa,L\neq0}\left(\frac{2\varkappa+1}{3}\right)^{1/2}\frac{[\Gamma(1\varkappa L;v)]^2}{[\Gamma_1-i\Omega'][\Gamma_\varkappa-i\Omega']}\right.\right.$$
$$\left.\left.-\left(\frac{3}{2}\right)^{1/2}\cos\vartheta\frac{\Gamma(111;v)}{\Gamma_1-i\Omega'}(\xi_+-\xi_-)-\frac{3^{1/2}}{2}\left(\cos^2\vartheta-\frac{1}{3}\right)\frac{\Gamma(112;v)}{\Gamma_1-i\Omega'}\right\}\right\rangle;\tag{3.12}$$
$$\Gamma_\varkappa\equiv\Gamma(\varkappa\varkappa0;\ v).$$

In the absence of Doppler broadening the second line in (3.12) vanishes and the corrections to the Lorentz contour are given by the ratio $[\Gamma(1\varkappa L;v)]^2/\Gamma_1\Gamma_\varkappa$, i.e., it turns out to be of second order of smallness. This circumstance, noted already by Kazantsev,[14] is general for all arbitrary transitions. The anisotropic terms in (3.12) contain factors $\Gamma(11L;v)/\Gamma_1$ which are of first order of smallness. However, the average values of the factors $\cos\vartheta$ and $\cos^2\vartheta-1/3$ is zero, and this introduces an additional smallness in the case of weak Doppler broadening. In the opposite limiting case the impact broadening is of no significance at all. Therefore the anisotropic terms in (3.7) can play a certain role only in the intermediate situation, when the Doppler and impact broadenings are comparable. However, it is difficult to estimate this role from general considerations, and we shall return to this question in a discussion of the numerical calculations.

An expression analogous to (3.12) can be obtained also for the model of "disorientation on one level": it is necessary to expand the Lorentz terms in (3.10) in powers of the anisotropic part of the relaxation constants and carry out in explicit form summation over M:

$$I(\Omega)=\frac{1}{\pi}\operatorname{Re}\left\langle\frac{W(\mathbf{v})}{\Gamma-i\Omega'}\left\{1+\sum_{L\neq 0}\left[\frac{\Gamma(L,v)}{\Gamma-i\Omega'}\right]^2\right.\right.$$
$$+\left(\frac{3}{2}\right)^{1/2}\frac{(2J+1)^{1/2}}{\Gamma_1-i\Omega'}(-1)^{J+J'}\left[\frac{3}{2}\left(\cos^2\vartheta-\frac{1}{3}\right)\begin{Bmatrix}J' & J & 1\\ 2 & 1 & J\end{Bmatrix}\Gamma(2,v)\right.$$
$$\left.\left.\left.-3^{1/2}\cos\vartheta\begin{Bmatrix}J' & J & 1\\ 1 & 1 & J\end{Bmatrix}\Gamma(1,v)(\xi_+-\xi_-)\right]\right\}\right\rangle,\quad \Gamma=\Gamma(JJ0,v). \tag{3.13}$$

From (3.13) follow obviously the conclusions discussed above.

To end the section, we note two general conclusions. It is easy to show that the integrated (with respect to frequency) line intensity

$$I_\infty=\int_{-\infty}^{\infty}I(\Omega)\,d\Omega=1$$

"is not sensitive" to the wind effect, as should be the case, since I_∞ does not depend at all on any of the singularities of the relaxation of the dipole moment. In particular, the anisotropic corrections to (3.14) and (3.15) alter the shape of the contour, but do not influence I_∞. From relations (3.7) and (3.11) it follows that the "wing" of the line has a universal form

$$I(\Omega)\approx\frac{1}{\pi\Omega^2}\operatorname{Re}\langle W(v)[\gamma+\Gamma(110;v)]\rangle;\qquad |\Omega|\gg k\bar{v},\ |\gamma+\Gamma(110;v)|. \tag{3.14}$$

Formula (3.14) refines the well known position of the impact theory of broadening (see, e.g., Ref. 1), namely, it indicates the exact meaning of the parameter that is important for the intensity of the "wing"—the isotropic part of the diagonal element $\varkappa=\varkappa_1=1$ of the relaxation matrix.

4. NUMERICAL CALCULATION. DISCUSSION

As already noted, the multicomponent character of the line as a result of the anisotropy of the collisions is a perfectly universal effect. At the same time the literature, to our knowledge, contains no experimental proof of its existence. It is therefore natural to analyze the factors that can mask the manifestation of the anisotropy. As one such factor we note, first of all, phase modulation of an atomic oscillator. It is clear that with increasing phase randomization the relative contribution of the disorientation to the line broadening will decrease, i.e., the role of an anisotropy effects should decrease. The foregoing pertains to relatively strong phase modulation: a small contribution of this modulation may turn out to be useful, since it contributes to an increase of the distance between the components of the structure. The latter considerations are in agreement with the results of numerical calculations of the relaxation matrix performed in Ref. 16 for the transition 1–0 and for van der Waals interaction. In the optical region of the spectrum, the strong phase modulation is more the rule rather than an exception, and here it can annihilate the influence of the wind effect, except for the dependence of the width and shift on the velocity of the radiating particle.

The broadening theory developed in Sec. 3 was constructed within the framework of the model of relaxation constants, i.e., no account is taken of the change of the velocity in the collisions. This change of velocity plays, as is well known, the role of spectral exchange between line components, and can result in collapse of the structure, analogous to the Dicke narrowing,[22,7] or to other types of collapse.[5] We shall demonstrate it using as an example the transition 1–0, describing the change of the velocity by the model of strong collisions and neglecting the Doppler broadening.

Under the foregoing conditions, in place of the system (3.2) we have [we assume $\varkappa=\varkappa_1=0$ and employ the model (3.3)]

$$(\gamma-i\Omega)\rho(1q,\mathbf{v})=-\sum_{q_1}\Gamma(1q|1q_1;v)\rho(1q_1,\mathbf{v})+\nu W(\mathbf{v})\langle\rho(1q,\mathbf{v})\rangle-iG_qNW(\mathbf{v}). \tag{4.1}$$

Equation (4.1) differs from the Kazantsev equation[14] obtained for resonant dipole-dipole interaction only in the structure of the integral arrival term: in (4.1) we have discarded its anisotropic part.

The solution of (4.1) is elementary, after which we obtain for $I(\Omega)$ in (3.6)

$$I(\Omega)=\frac{1}{\pi}\operatorname{Re}\frac{\langle J(\Omega)\rangle}{1-\langle\nu J(\Omega)\rangle}, \tag{4.2}$$

$$J(\Omega)=\frac{1}{3}\sum_q W(\mathbf{v})[\gamma+\Gamma_{11}{}^q(v)-i\Omega]^{-1}. \tag{4.3}$$

If we neglect terms of the form $(\Gamma_{11}^1-\Gamma_{11}^0)^4/(\Gamma_{11}^0\Gamma_{11}^1)^2$ (of fourth order of smallness) and assume that ν and $\Gamma\equiv\Sigma_q\Gamma_{11}^q/3$ are independent of v (the latter is realized in r^{-3} interaction), then it follows from (4.2) and (4.3) that

$$I(\Omega)=\frac{1}{\pi}\operatorname{Re}\{[\gamma+\Gamma-i\Omega]/(\gamma_1-i\Omega)(\gamma_2-i\Omega)\},$$
$$\gamma_{1,2}=\gamma+\Gamma-\nu/2\pm[(\nu/2)^2+{}^2/_9\langle W(\mathbf{v})[\Gamma_{11}{}^1-\Gamma_{11}{}^0]^2\rangle]^{1/2}. \tag{4.4}$$

Expression (4.4) is typical for a contour of a line consisting of two components and collapsing as a result of spectral exchange characterized by the frequency ν: depending on the relation between ν and the difference $|\Gamma_{11}^1-\Gamma_{11}^0|$ of the relaxation constants, the line contour either consists of two components having different widths ($\nu\ll\Gamma$), or takes the form of a single Lorentzian

$$I(\Omega)=\frac{1}{\pi}\operatorname{Re}\frac{1}{\Gamma+\gamma-\nu-i\Omega},\qquad \nu^2\gg\langle W(\mathbf{v})[\Gamma_{11}{}^1-\Gamma_{11}{}^0]^2\rangle. \tag{4.5}$$

The last relation demonstrates clearly that the change of the velocity in collisions can completely mask the line splitting due to anisotropy of the collisions.

Besides the factors noted above, to observe the spectral manifestations of the wind effect it is essential in principle that the anisotropy of the collisions manifest itself only in the second order of smallness [see (3.12), (3.13), and the discussion that follows].

Proceeding to discuss the numerical calculations, it should be noted that at the present time the data on the possible values of the constants $\Gamma(\varkappa\varkappa_1L;v)$ or $\Gamma(JJL;v)$ are quite limited. Namely, the relaxation matrix with allowance for the anistropy of the collisions was calculated only for the transition 1–0 in the case of dipole-dipole[14,15] and van der Waals[16] interactions. As shown above, the results of Refs. 14–16 can be used also for the transitions 1–1 and 1–2, if it is assumed that the collisions perturb only one of the levels with $J=1$ [see (2.20), (2.21), and (3.10)]. It is for these cases that numerical calculations were performed of the $I(\Omega)$ line contour.

The calculations have shown that individual Lorentzian components differ from one another quite significantly, up to 25% (depending on the mass ratio m_b/m, see Fig. 2). However, the entire line contour is accounted for with accuracy not worse than 95% by the term of the expansion (2.6) or (2.20) with $L=0$, i.e., by the isotropic part of the relaxation matrix, and the contribution of the anisotropic part is not more than 5%. The foregoing is illustrated in Fig. 3, which shows plots of the functions $I(\Omega)$ corresponding to the 1–0 transition and to van der Waals interaction. An increase of the ratio of $\bar{v}/\bar{v}_b$ influences the shape, width, and shift of the line as a whole, but likewise principally via the isotropic part.

The foregoing conclusions can be illustrated using as the example the limiting case $\bar{v}\gg\bar{v}_b$, when the anisotropy effects are maximal, and we have from (2.11) and (2.33)

$$\Gamma_{11}{}^{q}(v)=A_q(v/\bar{v})^{\beta},\quad A_q=a_q\Gamma_0(\bar{v}/\bar{v}_b)^{\beta},$$
$$\beta=(s-3)/(s-1),\quad \Gamma_0\equiv\Gamma(110;0). \tag{4.6}$$

Here Γ_0 corresponds to the value $\Gamma(110;v)$ at $v=0$, and the coefficients a_q, which can be expressed in terms of the cross section with the aid of relations (2.7), (2.11), (2.29), and (2.32), specify the ratios of the relaxation constants with different q at $\bar{v}\gg\bar{v}_b$. For simplicity we consider the line shift and the broadening due to the spontaneous decay; then

$$R_q(\Omega)=\frac{4}{\sqrt{\pi}A_q}\int_0^{\infty}\frac{z^{2+\beta}\,dz}{z^{2\beta}+(\Omega/A_q)^2}e^{-z^2},\quad \bar{v}\gg\bar{v}_b. \tag{4.7}$$

Thus, in the considered limiting case the contour of the component is a universal function whose scale on the frequency axis is given by the quantity A_q. We consider now the maximum value of the intensity in the summary contour, which is obtained at $\Omega=0$:

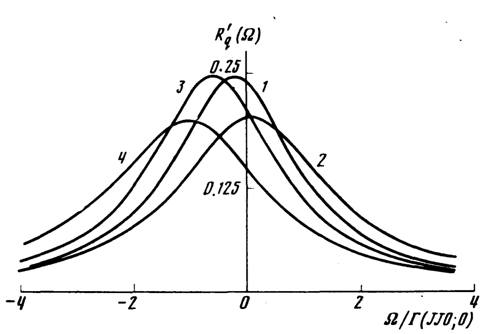

FIG. 2. Plots of the functions Re $R_q(\Omega)$ for the transition 1—0, $q=0$ (curves 1 and 3) and $q=1$ (curves 2 and 4), mass ratio $m_b/m=4$. The impact-broadengin parameters were chosen in the following manner: $a=b=1/7$, $\Gamma''(110;v)=0$ for curves 1,2; $a=-5/8$, $b=1/7$, $\Gamma''(110;v)$ for curves 3 and 4, where

$$a+bi=\lim\frac{\Gamma(112;v)}{\Gamma'(110;v)},\quad v\to\infty.$$

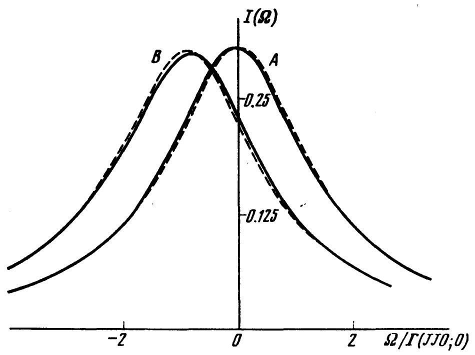

FIG. 3. Line contour $I(\Omega)$ in the absence of Doppler broadening. Solid curves—exact calculation, dashed curves—for the isotropic part of the relaxation matrix, Curves A and B have the same parameters as curves 1 and 2 (A) and 3 and 4 (B) in Fig. 2.

$$I(0)=\frac{1}{3\pi}\sum_q R_q(0)=\frac{C}{3}\sum_q A_q^{-1}=\frac{C}{A}\left[1+\frac{2}{9}\frac{(A_1-A_0)^2}{A_1A_0}\right],$$
$$A=\frac{1}{3}\sum_q A_q,\quad C=2\Gamma\left(\frac{3-\beta}{2}\right)\Big/\pi^{1/2}, \tag{4.8}$$

where A corresponds according to (2.10) to the isotropic part of the relaxation matrix. The influence of the anisotropic part on $I(0)$ is described in the second term in the square brackets of (4.8). In accordance with the results of Refs. 14–16 we have $|A_0-A_1|/A=0.3$, i.e., the individual components of the line differ in width and in maximum intensity by approximately 30%. For the contour as a whole, however, the correction for the anisotropy amounts to only 2%, in accord with the statements made above.

As shown by numerical calculations, a relatively small Doppler broadening ($k\bar{v}$ of the order of the impact width at $v=\bar{v}$) hardly changes the quantitative relations, which are illustrated in Figs. 2 and 3, although under the given conditions $I(\Omega)$ contains terms of first order in the anisotropy [see (3.12) and the discussion that follows].

Thus, the corrections to the contour of the spectral line on account of the anisotropy of the collisions turn out to be relatively small. The principal manifestation of the wind effect reduces to a dependence of the isotropic part of the relaxation matrix on the velocity. Of course, the foregoing is based on numerical values of $\Gamma(\varkappa\varkappa_1L;v)$, which so far are known only in two cases.[14-16] It is not excluded that for other types of interactions of colliding particles the ratios $|A_0-A_1|/A_q$ in (4.8) will be significantly larger.

The general conclusion formulated above is closely connected with the assumption that the ensemble of radiating atoms is isotropic. Otherwise an effective separation should take place of some of the $R_q(\Omega)$, and the corrections for the anisotropy of the collisions become of the first order of smallness. This is the situation, for example, in experiments with beams,[20] and in nonlinear saturation spectroscopy.[23]

In conclusion, we point out a number of problems for the analysis of which the relaxation-matrix theory developed above can be useful. Besides the already noted question of the contour of nonlinear resonance, this pertains to the problem of the shape of lines connected with nonresonant multiphoton processes. It is known, in particular, that Raman scattering of light by molecular hydrogen is accompanied by an anomalously small phase modulation.[24] This circumstance, as emphasized above, is favorable for manifestation of the collision anisotropy. The "hidden alignment" due to the wind effect is described by the relaxation matrix (2.1) with $J = J' = J_1 = J_1'$. A matrix of general form enters in the collapse problem in its usual formulation,[5] i.e., in the analysis of polarization exchange between transitions with close Bohr frequencies.

[1)] In the optical region of the spectrum one uses frequently also a stronger assumption, according to which one of the levels is not perturbed at all, see, e.g., Ref. 1.

[2)] To simplify the notation, we have left out from (3.10)–(3.13) the quantity γ, which can be assumed to be included in the isotropic part of the relaxation matrix.

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