Anisotropic collisional relaxation of atomic states with different electron angular momenta

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A range of characteristics of anisotropic collisional relaxation of atomic polarization moments is considered: collisional interconvertibility of the polarization moments of various ranks, the dependence of the relaxation rate on the projections of the polarization moments on the anisotropy axis, and multiexponential nature of transient relaxation processes involving atomic states with $j = 1, 3/2$, and 2. The symmetry properties of the matrix describing anisotropic collisional relaxation are identified and the numerical values of its elements are calculated by numerical integration of the impact-parameter-method equations for the investigated atom with neutral particles and ions. The results can be used in a quantitative description of collisional relaxation of various atomic states and of fine and hyperfine multiplets under conditions of extreme and partial anisotropy of the distribution of the relative velocities of the colliding particles.

INTRODUCTION

Relaxation of atomic states in gases under the influence of atomic collisions characterized by some preferred direction of the particle collisions exhibits a number of characteristics which distinguish it from the case of isotropic (random) collisions. The physical pattern of anisotropic collisional relaxation is much more elaborate than that of isotropic relaxation. For example, in the case of isotropic collisions the polarization moments of different ranks decay independently of one another (without mutual conversion) and all the components of the polarization moment of a given rank decay at the same rate.[1,2] However, in the case of anisotropic collisions the polarization moments of different ranks are interconvertible and the relaxation rate depends strongly on the projection of the component of the polarization moment on the anisotropy axis. The most striking example of interconvertibility of the polarization moments is the transfer from alignment to orientation,[3–10] which corresponds to a transition from the linear polarization of light to the circular polarization. Another example is the establishment of alignment of nonuniform populations of narrow-multiplet sublevels as a result of intramultiplet mixing under the influence of anisotropic collisions.[11–13] Interconvertibility of alignment and orientation is also responsible for a transient process characteristic of anisotropic relaxation: quantum polarization beats.[14] The dependence of the process of relaxation on the projection of the polarization moment along the anisotropy axis is manifested in particular by the appearance of the linear polarization of light as a result of different rates of decay of the longitudinal and transverse alignments.[15]

Anisotropic collisional relaxation was first calculated in Ref. 3 for an atomic state with angular momentum $j = 1$ by numerical integration of equations describing the impact parameter in the case of the $1/R^6$ (collisions of the investigated atom with neutral atoms of different gas) and $1/R^3$ (collisions of the investigated atom with ions) interaction laws. Similar calculations had been carried out[5,16] for atomic states with $j = 1$ and 2. The results of the latter calculations were reported in relative units, which made it difficult to use them in practice. More refined calculations of anisotropic relaxations in the $j = 1$ case were reported in Refs. 6 and 17.

An atomic state with angular momentum $j = 1$ is as yet the only case for which detailed numerical characteristics of anisotropic collisional relaxation are known. For this value of j the ordering of an electronic state of an atom is characterized solely by the polarization moments of the first and second ranks (orientation and alignment). In the case of higher values of the angular momentum the polarization moments have higher ranks ($\varkappa = 0, 1, \dots, 2j$) and the anisotropic collisional relaxation pattern exhibits an even greater variety and the number of its quantitative characteristics (independent elements of the relaxation matrix) increases.

We shall report the first calculations of the elements of the matrix describing anisotropic collisional relaxation of atoms in the case when $j = 3/2$ and 2. This makes it possible to extend the range of validity of the theory of anisotropic collisional relaxation by applying it to various spectral lines of atoms and atomic ions.

MATRIX OF ANISOTROPIC COLLISIONAL RELAXATION OF THE POLARIZATION MOMENTS

We shall consider an ensemble A of atoms which are in an electronic state characterized by an angular momentum j. Individual atoms in this ensemble are described by the wave functions

$$\Psi = \sum_{m=-j}^{j} c_m(t)\,\psi_{jm}, \tag{1}$$

where ψ_{jm} are the eigenfunctions of the operator of the square of the angular momentum $\hat{J}^2$ and its projection $\hat{J}_z$ along a fixed axis z of a laboratory coordinate system, and the whole ensemble is described by a density matrix

$$\sigma_{mm_1}(t) = \overline{c_m(t)\; c_{m_1}{}^*(t)}, \tag{2}$$

where the bar denotes averaging over the ensemble.

The polarization moments describing the ordering of the angular momenta of the electron shells of atoms in the ensemble A correspond to an expansion of the density matrix (2) in terms of irreducible representations of a group of three-dimensional rotations and are described by the expressions

$$\rho_q^{\varkappa}(t)=\sum_{mm_1}(-1)^{j-m_1}\begin{bmatrix} j & j & \varkappa \\ m & -m_1 & q \end{bmatrix}\sigma_{mm_1}(t). \qquad (3)$$

The ranks of the polarization moments have the values $\varkappa = 0, 1, \ldots, 2j$ and their z projections have the values $q = -\varkappa, -\varkappa+1, \ldots, \varkappa-1, \varkappa$.

We shall assume that the atoms in the ensemble A collide with perturbing particles B (rare-gas atoms or ions) and that these collisions are extremely anisotropic (so that the relative velocity $\mathbf{v}$ of each collision is exactly parallel to the z axis). Then, in view of the axial symmetry, the collisions retain the z projections of the polarization moment, but they result in mixing of the polarization moments of different ranks:

$$\dot{\rho}_q^{\varkappa}(t)=-\sum_{\varkappa_1}R_q^{\varkappa\varkappa_1}\rho_q^{\varkappa_1}(t). \qquad (4)$$

The dot denotes the time derivative, due to collisional processes.

Since the action of anisotropic collisions on the ensemble A not only has its own symmetry axis (z axis), but its own symmetry planes passing through this axis, it follows that the elements of the relaxation matrix $R_q^{\varkappa\varkappa_1}$ may couple only those polarization moments $\rho_q^{\varkappa}$ and $\rho_q^{\varkappa_1}$ which belong to the same irreducible representation of the symmetry group $C_{\infty v}$. If $q = 0$, this leads to the selection rule

$$R_0^{\varkappa\varkappa_1}=(-1)^{\varkappa+\varkappa_1}R_0^{\varkappa\varkappa_1}, \qquad (5)$$

which shows that $R_0^{\varkappa\varkappa_1} = 0$, where $\varkappa$ and $\varkappa_1$ are numbers of different parity: anisotropic collisions mix only those of the longitudinal components of the polarization moments $\rho_0^{\varkappa}$ and $\rho_0^{\varkappa_1}$ which have ranks of the same parity. (If $q \neq 0$, there is no such selection rule and the values of $R_q^{\varkappa\varkappa_1}$ are generally speaking different from zero for any $\varkappa$ and $\varkappa_1$.)

We can find elements of the matrix $R_q^{\varkappa\varkappa_1}$ by calculating the change in the state of an atom A when it collides with an incoming particle B. We shall introduce two Cartesian systems of coordinates with the origin at the nucleus of the atom A: a system x, y, z with the axes parallel to the corresponding axes in the laboratory coordinate system and a system $\tilde{x}, \tilde{y}, \tilde{z}$, in which the $\tilde{z}$ axis coincides with the z axis, whereas the $\tilde{x}$ and $\tilde{y}$ axes are rotated about the z axis by an angle φ, so that the trajectory of a particle B, regarded as rectilinear, lies in the $\tilde{x}\tilde{z}$ plane and passes by the nucleus of the atom A at a distance equal to the impact parameter B (Fig. 1).

We shall use $\tilde{\psi}_{jm}$ to denote the eigenfunctions of the operators $\hat{J}^2$ and $\hat{J}_z$ written in the coordinate system $\tilde{x}, \tilde{y}, \tilde{z}$, and we shall employ S for the scattering matrix in the same coordinate system. We shall assume that the collision begins at a time $t = -\infty$ and it ends at a time $t = +\infty$. If we describe the wave function of the atom A [Eq. (1)] in terms of the function $\tilde{\psi}_{jm}$, we find that

$$\Psi=\sum_{m=-j}^{j}\tilde{c}_m(t)\,\tilde{\psi}_{jm}. \qquad (6)$$

In view of the well-known property of spherical functions in the case of rotation about the z axis, we can show that the coefficients in Eqs. (1) and (6) are related by

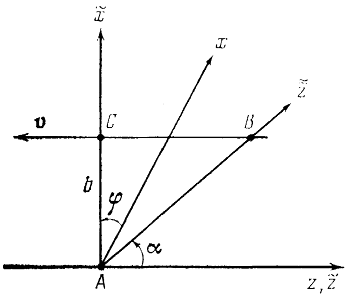

FIG. 1. Coordinate axes x, y, and z are parallel to the axes in the laboratory system; axes $\tilde{x}$ and $\tilde{y}$ are rotated about the z axis by an angle φ; axes $\tilde{z}$ and z coincide. An atom A being investigated is located at the origin of the coordinate system and a perturbing particle B flies past it along a strong line BC, parallel to the z axis and lying in the $\tilde{x}\tilde{z}$ plane. The angle α represents the instantaneous position of the particle B on its trajectory; the section AC of length b is the impact parameter. An axis z belongs to a "slave" coordinate system. It rotates by an angle π during the collision time. Axes y, $\tilde{y}$, $\tilde{\tilde{x}}$, and $\tilde{\tilde{y}}$ are not shown.

$$\tilde{c}_m(t)=e^{im\varphi}c_m(t). \qquad (7)$$

The coefficients of the expansion of the wave function (6) change as a result of collisions in accordance with the law

$$\tilde{c}_m(\infty)=\sum_{m'}\tilde{S}_{mm'}\tilde{c}_{m'}(-\infty), \qquad (8)$$

where elements of the $\tilde{S}$ matrix depend on the impact parameter b and on the collision velocity v, but are independent of the angle φ. Using Eqs. (7) and (2), we find that the changes in these coefficients correspond to the contribution to the change in the density matrix of the A atoms in a given j state, which is

$$\Delta\sigma_{mm_1}(\varphi)=\sum_{m'm_1'}e^{i(m_1+m'-m-m_1')\varphi}\left(\tilde{S}_{mm'}\tilde{S}^{*}_{m_1m_1'}-\delta_{mm'}\delta_{m_1m_1'}\right)\sigma_{m'm_1'}. \qquad (9)$$

Since atoms in the ensemble A undergo collisions with the B particles for all possible values of the angle φ, the contribution described by Eq. (9) should be averaged over this angle. The average contribution of one collision to the change in the density matrix of the ensemble A is

$$\Delta\sigma_{mm_1}=\sum_{m'm_1'}\left(\tilde{S}_{mm'}\tilde{S}^{*}_{m_1m_1'}-\delta_{mm'}\delta_{m_1m_1'}\right)\delta_{m_1+m'-m-m_1',0}\,\sigma_{m'm_1'}. \qquad (10)$$

We shall use N to denote the density of the perturbing B particles. The number of collisions per unit time which occur within a given interval b, $b + db$ of the impact parameter is $2\pi Nvbdb$. The contribution of all the collisions to the change in the density matrix of the A atoms is given by the equation

$$\dot{\sigma}_{mm_1}=2\pi Nv\int_0^{\infty}\Delta\sigma_{mm_1}b\,db. \qquad (11)$$

Using the transformation (3) to go over from the elements of the density matrix to the polarization moments, we obtain from Eq. (11), subject to Eq. (10), the following equation for the changes in the polarization moments of the ensemble of atoms A under the influence of anisotropic collisions:

$$\dot{\rho}_q^{\varkappa}=2\pi Nv\sum_{\substack{mm_1m'\\ m_1'\varkappa_1}}(-1)^{m_1'-m_1}\begin{bmatrix} j & j & \varkappa \\ m & -m_1 & q\end{bmatrix}\begin{bmatrix} j & j & \varkappa_1 \\ m' & -m_1' & q\end{bmatrix}$$

$$\times\int\limits_0^{\infty}(\mathfrak{S}_{mm'}\mathfrak{S}^{*}_{m_1m_1'}-\delta_{mm'}\delta_{m_1m_1'})\,b\,db\,\rho_q^{\varkappa_1}. \qquad (12)$$

The coefficient in front of the quantity $\rho_q^{\varkappa_1}$ on the right-hand side of Eq. (12) represents an element of the relaxation matrix $R_q^{\varkappa\varkappa_1}$ defined in accordance with Eq. (4). Therefore, we obtain the following explicit expression for the elements of the matrix of the anisotropic collisional relaxation of the polarization moments, expressed in terms of the elements of the scattering matrix:

$$R_q^{\varkappa\varkappa_1}=2\pi Nv\sum_{\substack{mm_1\\ m'm_1'}}(-1)^{m_1'-m_1+1}\begin{bmatrix} j & j & \varkappa \\ m & -m_1 & q\end{bmatrix}\begin{bmatrix} j & j & \varkappa_1 \\ m' & -m_1' & q\end{bmatrix}$$

$$\times\int\limits_0^{\infty}(\mathfrak{S}_{mm'}\mathfrak{S}^{*}_{m_1m_1'}-\delta_{mm'}\delta_{m_1m_1'})\,b\,db. \qquad (13)$$

When we allow for the behavior of the Clebsch-Gordan coefficients when the signs of all the lower indices are reversed, the above expression yields the following asymmetry property of the relaxation matrix:

$$R_{-q}^{\varkappa\varkappa_1}=(-1)^{\varkappa+\varkappa_1}R_q^{\varkappa\varkappa_1}. \qquad (14)$$

In the next section we shall show that in the case of rectilinear trajectories the scattering matrix has the symmetry property

$$\mathfrak{S}_{m'm}=(-1)^{m-m'}\mathfrak{S}_{mm'}. \qquad (15)$$

Using Eq. (13), we find that the elements of the matrix of the anisotropic collisional relaxation process do not change as a result of transposition of the upper indices:

$$R_q^{\varkappa\varkappa_1}=R_q^{\varkappa_1\varkappa}. \qquad (16)$$

We note that the matrix R describes relaxation of an isolated electronic state with a given value of j, without collision-induced transitions to other electronic states. This corresponds to the situation when the number of atoms in a given j state is not affected by collisions: $\dot{n}_j=0$. The number of atoms is related to the polarization moment of zeroth rank by the expression $n_j=(2j+1)^{1/2}\rho_0^0$. Using the relaxation equation (4), we find that the condition for conservation of the number n_j of particles in a given electronic state can be written in the form

$$\dot{\rho}_0^{0}=-\sum_{\varkappa_1}R_0^{0\varkappa_1}\rho_0^{\varkappa_1}=0. \qquad (17)$$

This condition is satisfied for all values of the electronic polarization moments $\rho_0^{\varkappa}$. Hence, it follows that the coefficients in front of all the polarization moments on the right-hand side vanish so that the elements of the relaxation matrix become $R_0^{0\varkappa}=0$ for all values of $\varkappa$. Allowing for the symmetry properties of Eq. (16), we can write this result in the form

$$R_0^{0\varkappa}=R_0^{\varkappa 0}=0. \qquad (18)$$

The symmetry properties of the R matrix of Eqs. (14) and (16), and also the selection rule of Eq. (5) and the property of Eq. (18) mean that the number of independent elements of the matrix is considerably lower than the total number of its elements.[16,18]

DESCRIPTION OF COLLISIONS

We shall introduce a "slave" coordinate system $\tilde{\tilde{x}}, \tilde{\tilde{y}}, \tilde{\tilde{z}}$, obtained by rotation of the coordinate system $\tilde{x}, \tilde{y}, \tilde{z}$ by an angle α about the y axis. The $\tilde{\tilde{z}}$ axis of the slave system is directed along a line joining instantaneous positions of the colliding particles A and B. During the collision this axis rotates by an angle π. The eigenfunctions of the atom A, corresponding to quantization of the angular momentum along the axis of the slave coordinate system, will be denoted by $\tilde{\tilde{\psi}}_{jm}$.

Writing down the wave function of the atom A in the form

$$\Psi=\sum_{m=-j}^{j}\tilde{\tilde{c}}_m(t)\,\tilde{\tilde{\psi}}_{jm}, \qquad (19)$$

we find that changes in the coefficients $\tilde{\tilde{c}}_m$ in the course of a collision are described by the following system of equations derived within the framework of the impact parameter method[19]:

$$i\,\frac{d\tilde{c}_m}{dt}=\frac{W_m(t)}{\hbar}\,\tilde{\tilde{c}}_m(t)-i\sum_{m_1}\left\langle\tilde{\tilde{\psi}}_{jm}\middle|\frac{d}{dt}\,\tilde{\tilde{\psi}}_{jm_1}\right\rangle\tilde{\tilde{c}}_{m_1}(t). \qquad (20)$$

Here, W_m is the energy of the interaction between the atoms A and B, calculated for the case when the projection of the angular momentum of an electron shell of the A atom along the line AB is m. We shall assume that this energy is described by an expression corresponding to the first nonvanishing term of the multipole expansion:

$$W_m(t)=C_m/R^n(t), \qquad (21)$$

where $R(t)$ is the time-dependent distance between the particles A and B. The case $n=6$ describes the van der Waals interaction of neutral atoms A and B and the case $n=3$ describes the interaction of an ion B with the quadrupole moment of an electron shell of an atom A.

The angle α is related to the time t measured from the beginning of the collision:

$$\operatorname{ctg}\alpha=-vt/b. \qquad (22)$$

If instead of the time t we use the angle α, we can reduce the system of equations (20) to

$$i\,\frac{d\tilde{\tilde{c}}_m}{d\alpha}=\frac{C_m\sin^{n-2}\alpha}{b^{n-1}\hbar v}\,\tilde{\tilde{c}}_m+\frac{i}{2}\,[(j+m+1)^{1/2}(j-m)^{1/2}\tilde{\tilde{c}}_{m+1}$$

$$-(j-m+1)^{1/2}(j+m)^{1/2}\tilde{\tilde{c}}_{m-1}]. \qquad (23)$$

SYMMETRY OF THE SCATTERING MATRIX UNDER TIME REVERSAL

The system of equations (23), valid in the model of rectilinear trajectories, shows that the scattering matrix exhibits time-reversal symmetry [Eq. (15)], which leads to the symmetry property described by Eq. (16) for the anisotropic collisional relaxation matrix. We shall now prove this.

The system of differential equations (23) behaves as a matrix which transforms the values of the quantities $\tilde{\tilde{c}}_m(0)$ given for $\alpha = 0$ into their running values $\tilde{\tilde{c}}_m(\alpha)$. Denoting the matrix of this transformation by U, we find that the general solution of the system for arbitrary initial values of $\tilde{\tilde{c}}_m(0)$ can be described by the expression

$$\tilde{\tilde{c}}_m(\alpha) = \sum_{m_1} U_{mm_1}(\alpha)\, \tilde{\tilde{c}}_{m_1}(0). \qquad (24)$$

The elements of the matrix $U_{mm_1}(\alpha)$ with a fixed value of the index m_1 form, in particular, the solution of the system $\tilde{\tilde{c}}_m(\alpha) = U_{mm_1}(\alpha)$, which corresponds to the initial condition $\tilde{\tilde{c}}_m(0) = U_{mm_1}(0) = \delta_{mm_1}$.

In the slave coordinate system the scattering matrix is related to the U matrix by

$$\tilde{\tilde{S}}_{mm_1} = U_{mm_1}(\pi). \qquad (25)$$

If the system of equations (23) is subjected to complex conjugation, transposition of the indices in accordance with the scheme $m \to -m$, and transformation $\alpha \to \pi - \alpha$, the result is a system of equations

$$i\frac{d\tilde{\tilde{c}}^{\,*}_{-m}(\pi-\alpha)}{d\alpha} = \frac{C_m \sin^{n-2}\alpha}{b^{n-1}\hbar v}\tilde{\tilde{c}}^{\,*}_{-m}(\pi-\alpha) + \frac{i}{2}\left[(j+m+1)^{1/2}(j-m)^{1/2}\tilde{\tilde{c}}^{\,*}_{-m-1}(\pi-\alpha) - (j-m+1)^{1/2}(j+m)^{1/2}\tilde{\tilde{c}}^{\,*}_{-m+1}(\pi-\alpha)\right]. \qquad (26)$$

Hence, it follows that in the case when a certain set of energies $\tilde{\tilde{c}}^{(1)}_m(\alpha)$ satisfies the system of equations (23), the same system is satisfied also by a second set which is $\tilde{\tilde{c}}^{(2)}_m(\alpha) = \tilde{\tilde{c}}^{(1)}_{-m}(\pi - \alpha)$. If the first set is assumed to represent a particular solution $\tilde{\tilde{c}}^{(1)}_m(\alpha) = U_{mm_1}(\alpha)$, we find that the second solution is described by

$$\tilde{\tilde{c}}^{(2)}_m(\alpha) = U^*_{-m,+m_1}(\pi-\alpha). \qquad (27)$$

At the point $\alpha = \pi$ this solution satisfies the condition $\tilde{\tilde{c}}^{(2)}_m(\pi) = U^*_{-m,m_1}(\pi) = \delta_{-m,m_1}$. It therefore follows from Eq. (27) that in the general solution of the system (24) the quantities $\tilde{\tilde{c}}_m(\alpha)$ are expressed in terms of their values specified at the point $\alpha = \pi$:

$$\tilde{\tilde{c}}_m(\alpha) = \sum_{m_1} U^*_{-m,-m_1}(\pi-\alpha)\, \tilde{\tilde{c}}_{m_1}(\pi). \qquad (28)$$

Assuming that $\alpha = 0$, we obtain

$$\tilde{\tilde{c}}_m(0) = \sum_{m_1} U^*_{-m,-m_1}(\pi)\, \tilde{\tilde{c}}_{m_1}(\pi). \qquad (29)$$

Next, the relationship between the matrix U and the inverse scattering matrix is

$$U^*_{-m,-m_1}(\pi) = \tilde{\tilde{S}}^{-1}_{mm_1}. \qquad (30)$$

Using Eq. (25), we obtain

$$\tilde{\tilde{S}}^*_{-m,-m_1} = \tilde{\tilde{S}}^{-1}_{mm_1}. \qquad (31)$$

Applying now the unitary property of the $\tilde{\tilde{S}}$ matrix, we find that it has the following symmetry property

$$\tilde{\tilde{S}}_{mm_1} = \tilde{\tilde{S}}_{-m_1,-m}. \qquad (32)$$

Since the $\tilde{\tilde{z}}$ axis of the slave coordinate system for $\alpha = \pi$ is rotated by an angle π relative to the $\tilde{z}$ axis, the scattering matrix expressed in the coordinate system $\tilde{x}, \tilde{y}, \tilde{z}$ is

$$\tilde{S}_{mm_1} = (-1)^{j+m}\tilde{\tilde{S}}_{-m,m_1}. \qquad (33)$$

It follows from the last two expressions that in the case of rectilinear trajectories the $\tilde{S}$ matrix does indeed have the symmetry property described by Eq. (15) and the R matrix has the symmetry property described by Eq. (16).

QUANTITATIVE CHARACTERISTICS OF ANISOTROPIC COLLISIONAL RELAXATION OF ATOMIC STATES WITH $j=1$, 3/2, AND 2

The elements of the scattering matrix $\tilde{\tilde{S}}_{mm_1}$ in the slave coordinate system were calculated by numerical integration of the system of equations (23) subject to the initial conditions $\tilde{\tilde{c}}_m(0) = \delta_{mm_1}$. Then, using Eq. (33), we calculated from Eq. (13) the values of the elements of the relaxation matrix $R^{\varkappa\varkappa_1}_q$. All the calculations were made on a computer. In solving the system of equations (23) the precision was monitored using the unitary property of the $\tilde{\tilde{S}}$ matrix. Calculating the integrals in Eq. (13), we went over from integration with respect to the impact parameter b to integration with respect to the Massey parameter, defined by (for a $1/R^n$ interaction law)

$$B_n = \Delta C / b^{n-1}\hbar v, \qquad (34)$$

where ΔC is the absolute value of the difference of the quantities C_m occurring in Eq. (21) and corresponding to the maximum ($m = j$) and minimum ($m = 0$ or 1/2) values of the projection of the electron angular momentum of an atom A on a line joining the colliding particles A and B.

A detailed numerical solution of the system of differential equations (23) was obtained in the range $0 \leqslant B_n \leqslant 100$ of the Massey parameter (in steps of B_n, amounting to 0.02 for $B_n \leqslant 0.1$ and 0.2 for $B_n > 0.1$). In the case of large Massey parameters ($B_n > 100$) it was replaced by the adiabatic approximation obtained by crossing out the quantities $\tilde{\tilde{c}}_{m+1}$ and $\tilde{\tilde{c}}_{m-1}$ on the right-hand side in the system (23).

The elements of the anisotropic collisional relaxation matrix $R^{\varkappa\varkappa_1}_q$ calculated by this method for electronic states with angular momenta $j = 1$, 3/2, and 2 are listed in Table I. (The results for $j = 1$ were obtained earlier[15] and are included for the sake of comparison.) This table includes only the independent elements of the relaxation matrix. Their number for $j = 1$, 3/2, and 2 is 6, 14, and 26, respectively. The other elements of the R matrix can be determined from the results in the table using Eqs. (5), (14), (16), and (18). The elements of the relaxation matrix are expressed in the following units:

$$s_n \equiv N v^{(n-3)/(n-1)} \left|\frac{\Delta C}{\hbar}\right|^{2/(n-1)} \qquad (35)$$

We shall now turn to the characteristics of anisotropic collisional relaxation, which follow from numerical values of the elements of the R matrix.

The diagonal elements of the matrix $R^{\varkappa\varkappa}_q$ determine the rates of decay of the polarization moments under the influence of collisions. Collisional decay of the longitudinal com-

TABLE I. Elements of the anisotropic collisional relaxation matrix $R_q^{\varkappa\varkappa_1}$ for electron states $j = 1, 3/2$, and 2.

$j=1$

$\varkappa$	$\varkappa_1$	q	$n=6$	$n=3$	$\varkappa$	$\varkappa_1$	q	$n=6$	$n=3$
1	1	0	4.61	6.56	2	2	0	1.45	1,64
1	1	1	1,97	2,41	2	2	1	2,94	3.48
1	2	1	$0.96i$	$-0,39i$	2	2	2	2.55	3.56

$j = {}^3/_2$

$\varkappa$	$\varkappa_1$	q	$n=6$	$n=3$	$\varkappa$	$\varkappa_1$	q	$n=6$	$n=3$
1	1	0	4,37	5.67	2	2	2	2.57	3.14
1	1	1	1.62	1.89	2	3	1	$0,66i$	$0.25i$
1	2	1	$0.58i$	$-0.20i$	2	3	2	$0.87i$	$0.00i$
1	3	0	-0.69	-1.22	3	3	0	3,33	3.84
1	3	1	$-0,31$	-0.74	3	3	1	2.58	3.01
2	2	0	2,29	3.62	3	3	2	2,84	3.42
2	2	1	3.71	4.95	3	3	3	2,50	3.28

$j = 2$

$\varkappa$	$\varkappa_1$	q	$n=6$	$n=3$	$\varkappa$	$\varkappa_1$	q	$n=6$	$n=3$
1	1	0	3.47	3.54	2	4	2	0.03	-0.33
1	1	1	1.37	1.13	3	3	0	3.33	3,70
1	2	1	$0.60i$	$0.00i$	3	3	1	2,81	3.51
1	3	0	-0.53	-0.76	3	3	2	3.46	4.00
1	3	1	-0.18	-0.44	3	3	3	2.42	2,52
1	4	1	$0.10i$	$-0.10i$	3	4	1	$0,52i$	$0.22i$
2	2	0	2.12	3.12	3	4	2	$0.60i$	$0.17i$
2	2	1	3,39	3.98	3	4	3	$0,75i$	$0.12i$
2	2	2	2,22	2.23	4	4	0	1.48	2.37
2	3	1	$0.32i$	$0.10i$	4	4	1	2.22	2.48
2	3	2	$0.64i$	$-0.03i$	4	4	2	2.43	2.37
2	4	0	-0.26	-1.03	4	4	3	2.67	2.69
2	4	1	-0.26	-0.98	4	4	4	2.10	2.28

ponent of the orientation ρ_0^1 occurs in all the cases considered in our calculations ($j = 1$, 3/2, and 2; $n = 6$ and 3) approximately 2.5 times faster than the decay of the transverse component ρ_1^1. This means that the orientation vector induced by some external agency (for example, by a light pulse) not only does not decay in the course of anisotropic relaxation, but rotates across the anisotropy axis: in the course of decay the longitudinal component disappears first and then only the transverse component remains.

In the case of the polarization moment of the second rank (alignment) the component ρ_1^2 inclined at an angle of 45° to the anisotropy axis decays in all cases 1.5–2 times faster than the longitudinal component ρ_0^2. The transverse alignment component ρ_2^2 for $j = 1$ decays much faster than the longitudinal component ρ_0^2. This suppresses linear polarization of light along this anisotropy axis when an ensemble of relaxing atoms is illuminated with unpolarized light.[15] An increase in the angular momentum j equalizes the decay constants of the longitudinal and transverse alignment for an interaction law with $n = 6$, whereas for $n = 3$ the sign of the difference between these decay constants is reversed (the longitudinal alignment begins to decay faster). In the case of the polarization moments of the third and fourth rank it is difficult to find any definite relationship governing the values of the corresponding elements R_q^{33} and R_q^{44} of the relaxation matrix, but the anisotropic nature of the relaxation process manifested by the dependences of these elements on the projection q is manifested quite clearly.

The off-diagonal (in respect of $\varkappa$ and $\varkappa_1$) elements of the relaxation matrix $R_q^{\varkappa\varkappa_1}$ describe collisional interconvertibility of the polarization moments of different ranks: alignment and orientation (ρ_1^2 and ρ_1^1) orientation and hexadecapole alignment (ρ_1^1 and ρ_1^4), orientation and octupole orientation (ρ_0^1 and ρ_0^3, ρ_1^1 and ρ_1^3), alignment and octupole orientation (ρ_1^2 and ρ_1^3, ρ_2^2 and ρ_2^3), alignment and hexadecapole alignment (ρ_0^2 and ρ_0^4, ρ_1^2 and ρ_1^4, ρ_2^2 and ρ_2^4), and also interconvertible hexadecapole alignment and octupole orientation (ρ_q^4 and ρ_q^3, where $q = 0$, 1, 2, or 3).

Classification of the components of the polarization moments $\rho_q^\varkappa$ in accordance with irreducible representations of the symmetry group $C_{\infty v}$ is given in Table II and it demonstrates in the general case of an arbitrary value of j, the division of these components into a set of concurrently relaxing quantities. We must bear in mind that in the case of two-dimensional irreducible representations E the quantities $\rho_q^\varkappa$ and $\rho_{-q}^\varkappa$ form independently relaxing sets (the value of the projection of the polarization moment q is conserved during relaxation).

One further property of the relaxation matrix should be noted here. It follows from the definition of the polarization moments (3), after allowance for the Hermitian nature of the density matrix σ, that

TABLE II. Classification of the polarization moments of an ensemble of atoms in a state with an angular momentum j, described in terms of irreducible representations of the symmetry group $C_{\infty v}$.

Irreducible representation	Components of polarization moments belonging to given irreducible representation	Multiplicity of repetition of irreducible representation
A_1	$\rho_0^0, \rho_0^2, \rho_0^4, \ldots, \rho_0^{2[j]}$	$[j]+1$
A_2	$\rho_0^1, \rho_0^3, \rho_0^5, \ldots, \rho_0^{2[j+1/2]-1}$	$[j+1/2]$
E_1	$\rho_{\pm 1}^1, \rho_{\pm 1}^2, \rho_{\pm 1}^3, \ldots, \rho_{\pm 1}^{2j}$	$2j$
E_2	$\rho_{\pm 2}^2, \rho_{\pm 2}^3, \rho_{\pm 2}^4, \ldots, \rho_{\pm 2}^{2j}$	$2j-1$
....	....	....
E_{2j}	$\rho_{\pm 2j}^{2j}$	1

Note. Here, $[j]$ denotes the integer part of the number j.

$$\rho_{-q}^{\varkappa *}=(-1)^q\rho_q^{\varkappa}. \tag{36}$$

Hence, and from Eq. (4), it follows that the relaxation matrix elements satisfy the relationship

$$R_q^{\varkappa\varkappa_1}=R_{-q}^{\varkappa\varkappa_1 *}. \tag{37}$$

Combining this result with Eq. (14), we have

$$R_q^{\varkappa\varkappa_1}=(-1)^{\varkappa+\varkappa_1}R_q^{\varkappa\varkappa_1 *}. \tag{38}$$

It is therefore clear that the off-diagonal (in terms of $\varkappa$) matrix elements $R_q^{\varkappa\varkappa_1}$, responsible for the interconvertibility of the polarization moments of different ranks, are all either purely real or purely imaginary: the former applies in the case when the sum ($\varkappa+\varkappa_1$ is even) and the latter applies when this sum is odd.

Among the processes of interconvertibility of the polarization moments the most important is the transition from the alignment ρ_1^2, which is inclined at an angle of 45° with respect to the z axis and is induced by anisotropic collisions, to the transverse orientation ρ_1^1 (accompanied by the transition from the linear to the circular polarization of light). The effectiveness of this transition is described by the matrix element R_1^{12}. In the case of collisions with neutral particles (when $n=6$) if $j=3/2$ or 2, this matrix element represents about 60% of its value in the case when $j=1$, so that an increase in the angular momentum of the electron shell reduces slightly the collisional transformation of alignment into orientation. In the case of collisions with ions (in the $n=3$ case) the matrix element R_1^{12} falls rapidly in absolute value on increase of j and vanishes for $j=2$ (within the limits of the calculation error).

It is clear from Table I that the off-diagonal (in terms of $\varkappa$) matrix elements $R_q^{\varkappa\varkappa_1}$ are systematically smaller than the diagonal elements $R_q^{\varkappa\varkappa}$. Then, in special cases some of the off-diagonal elements are very close to zero (R_2^{23} for $j=3/2$ or 2 and $n=3$; R_1^{12} for $j=2$ and $n=3$; R_2^{24} for $j=2$ and $n=6$).

Collisional interconvertibility of the polarization moments is responsible for the complex multiexponential nature of relaxation of these moments under anisotropic collision conditions. The components of the polarization moments $\rho_q^{\varkappa}$ become grouped into concurrently relaxing sets and collisional relaxation of the values in each set considered as a function of time is described by

$$\rho_q^{\varkappa}(t)=\sum_{s=1}^{p}C_s e^{-\lambda_s t}.$$

where the number of the exponents λ_s is equal to the number of the quantities $\rho_q^{\varkappa}$ in a given set. If $j=2$, we have the following sets of concurrently relaxing components of the polarization moments: ρ_0^1 and ρ_0^3; ρ_0^2 and ρ_0^4; $\rho_1^1, \rho_1^2, \rho_1^3$, and ρ_1^4; ρ_2^2, ρ_2^3, and ρ_2^4; ρ_3^3 and ρ_3^4; ρ_4^4. Similar sets are obtained in the $j=3/2$ case by crossing out the components of the polarization moment of the fourth rank ρ_q^4, and the sets corresponding to $j=1$ are obtained by dropping the components of the polarization moments of the fourth and third ranks ρ_q^4 and ρ_q^3.

The multiexponential nature of relaxation of the polarization moments can be observed directly in transient processes (for example, in the case of free decay after pulsed excitation). The functions $\rho_q^{\varkappa}(t)$ may then vary nonmonotonically with time and under certain conditions, if among the exponents λ_s there are complex values, there may be even damped oscillations (polarization beats—see Ref. 14).

For example, in the specific case of interconvertibility of alignment and orientation, free decay of the polarization moments under the influence of anisotropic collisions is described (for $j=1$) by the equations

$$\dot{\rho}_1^1=-R_1^{11}\rho_1^1-R_1^{12}\rho_1^2, \qquad \dot{\rho}_1^2=-R_1^{21}\rho_1^1-R_1^{22}\rho_1^2, \tag{39}$$

where in view of Ref. 16 we have $R_1^{21}=R_1^{12}$. The solution of these equations is given by the expression with $p=2$. The exponential decay constants are then

$$\lambda_{1,2}={}^1/_2[R_1^{11}+R_1^{22}\pm(4R_1^{12}R_1^{21}+(R_1^{22}-R_1^{11})^2)^{1/2}]. \tag{40}$$

If

$$\omega^2\equiv|R_1^{12}|^2-{}^1/_4(R_1^{22}-R_1^{11})^2>0, \tag{41}$$

the radicand in Eq. (40) is negative and the exponential decay of the alignment and orientation is accompanied by their oscillations at a frequency ω. Turning back to the numerical values of the elements of the R matrix given in Table I, we find that this polarization beat regime is obtained in the $j=1$ case for the law of interaction with $n=6$, but not for the law of interaction with $n = 3$. We can give also other examples when concurrent relaxation of the polarization moments in time is in the form of damped oscillations (for example, relaxation of the transverse alignment ρ_2^2 together with the component of the octupole orientation ρ_2^3 when $j = 3/2$ and $n = 6$). It should also be pointed out that a strongly nonmonotonic time dependence of the polarization moment can occur also in the absence of beats (when all the λ_s are real).

In the case of rectilinear trajectories it follows from Eq. (18) that collisional creation of the polarization moments $\rho_0^\varkappa$ with $\varkappa \neq 0$ from a population ρ_0^0 of an isolated j level is impossible. (The role of bending and splitting of trajectories in such processes is considered in Ref. 20.) The situation changes drastically when the total angular momentum F of an atom consists of a relaxing electron angular momentum j and a nonrelaxing angular momentum (for example, nuclear spin momentum) i, and when the multiplet of levels originating from a given electronic state j is sufficiently narrow for effective collisional mixing. In this case the order of the total angular momentum $\mathbf{F} = \mathbf{j} + \mathbf{i}$ of an ensemble of atoms is described by "large" polarization moments:

$$\rho_Q{}^K(FF_1) = \sum_{\substack{MM_1 mm_1\\ \mu\mu_1\varkappa q}} (-1)^{F_1-M_1} \begin{bmatrix} F & F_1 & K \\ M & -M_1 & Q \end{bmatrix} \times \begin{bmatrix} j & i & F \\ m & \mu & M \end{bmatrix} \begin{bmatrix} j & i & F_1 \\ m_1 & \mu_1 & M_1 \end{bmatrix} \times \begin{bmatrix} j & j & \varkappa \\ m & -m_1 & q \end{bmatrix} \sigma_{\mu\mu_1} \rho_q^\varkappa, \qquad (42)$$

where $\sigma_{\mu\mu_1}$ is the density matrix for projections of the angular momentum i. Anisotropic collisional relaxation of large polarization moments occurs subject to selection rules for the projections Q and the ranks K, and subject to similar selection rules for $\varkappa$ and q in the case relaxation of an isolated j level. However, it is important that in the case of levels of a (j, i) multiplet there are no selection rules of the type (18), so that generally speaking the matrix elements coupling large polarization moments characterized by $K \neq 0$ and the populations of the multiplet levels $\rho_0^0(FF)$ differ from zero. This is explained by the fact that in the course of relaxation the populations of the individual components of a multiplet are not conserved: only the total population of all the levels of the multiplet is conserved. Therefore, in the case of the individual components of a multiplet we can no longer write down the conditions (17) for conservation of the population.

This collisional creation of order under the influence of anisotropic collisions in the case of intramultiplet mixing was considered in Refs. 11–13 and 17 in connection with the creation of alignment of narrow multiplets in hyperfine and fine structures. It is important to note that in this case the relaxation matrix of large polarization moments $\rho_Q^K(FF_1)$ (matrix $R_Q^{KK_1}$) is obtained from the anisotropic collisional relaxation matrix $R_q^{\varkappa\varkappa_1}$ of pure electron polarization moments by suitable addition of the angular momenta on the basis of kinematic considerations. The relationship between these matrices is given by[17]

$$R_Q^{KK_1}(FF_1; F_2F_3) = \sum_{\substack{\varkappa\varkappa_1 l\\ pq}} (-1)^{F^*}(2j+1) [(2\varkappa+1)(2\varkappa_1+1)(2K+1)(2K_1+1) \times (2F+1)(2F_1+1)(2F_2+1)(2F_3+1)]^{1/2} \times \begin{Bmatrix} i & i & l \\ j & j & \varkappa \\ F & F_1 & K \end{Bmatrix} \begin{Bmatrix} i & i & l \\ j & j & \varkappa_1 \\ F_2 & F_3 & K_1 \end{Bmatrix} \cdot \times \begin{Bmatrix} \varkappa_1 & \varkappa & p \\ K & K_1 & l \end{Bmatrix} \begin{bmatrix} \varkappa_1 & \varkappa & p \\ q & -q & 0 \end{bmatrix} \begin{bmatrix} K_1 & K & p \\ Q & -Q & 0 \end{bmatrix} R_q^{\varkappa\varkappa_1}, \qquad (43)$$

where $F^* \equiv F_1 + F_3 - F - F_2 + K + K_1 - l - p + Q + q$ and the quantities in the braces represent the $6j$ and $9j$ symbols.

In view of the above conclusions, the elements of the anisotropic collisional relaxation matrix $R_q^{\varkappa\varkappa_1}$ listed in Table I are suitable not only for the calculation of the various characteristics of anisotropic relaxation of an isolated electronic state with a given value of j, but also for the description of similar characteristics of multiplets which appear on addition of a relaxing electron angular momentum to a nonrelaxing nuclear spin momentum $\mathbf{i}$ (or in the case of narrow fine-structure multiplets when a nonrelaxing electron orbital momentum l is added to a nonrelaxing electron spin momentum $\mathbf{s}$).

The quantities $R_q^{\varkappa\varkappa_1}$ listed in Table I correspond to the extreme anisotropic case of collisions of the particles at rest A with a monokinetic beam of particles B, parallel exactly to the z axis. It is easier to consider a partly anisotropic case in which these idealized conditions are not satisfied. Let us assume that the function describing the real distribution of the relative velocities $\mathbf{v} = \mathbf{v}_B - \mathbf{v}_A$ has an axial symmetry and is of the form $f(v,\theta)$, where θ is the angle of tilt of the vector $\mathbf{v}$ relative to the z axis and v is the absolute value of this vector. If we represent collisional relaxation of the distribution of the velocities $f(v,\theta)$ in the form of a sum of an infinite number of extremely anisotropic relaxation processes corre-

TABLE III. Constants of isotropic collisional relaxation of electronic states $j = 1$, $3/2$, and 2.

	$j=1$		$j={}^3/_2$		$j=2$	
	$n=6$	$n=3$	$n=6$	$n=3$	$n=6$	$n=3$
γ^1	2.85	3.79	2.54	3.15	2.07	1.93
γ^2	2.48	3.14	2.99	3.96	2.67	3.11
γ^3	–	–	2.74	3.32	2.96	3.39
γ^4	–	–	–	–	2.26	2.45
γ^2/γ^1	0.87	0.83	1.22	1.26	1.29	1.61
γ^3/γ^1	–	–	1.12	1.05	1.43	1.76
γ^4/γ^1	–	–	–	–	1.09	1.27

sponding to monokinetic distributions, we find that the matrix of partly anisotropic collisional relaxation is described by the following expression containing elements of the matrix $R_q^{\varkappa\varkappa_1}$ and of Wigner's D function:

$$\langle R_q^{\varkappa\varkappa_1}\rangle=\sum_{q_1}\int D_{qq_1}^{\varkappa}(\varphi,\theta,\psi)D_{qq_1}^{\varkappa_1 *}(\varphi,\theta,\psi)$$

$$\times\ f(v,\theta)R_{q_1}^{\varkappa\varkappa_1}(v)\sin\theta\, d\theta\, d\varphi\, d\psi. \qquad (44)$$

The isotropic relaxation matrix of the polarization moments subjected to collisions along random directions can also be expressed in terms of the extremely anisotropic relaxation matrix $R_q^{\varkappa\varkappa_1}$. In the isotropic case the polarization moments relax under the influence of collisions in accordance with the law

$$\dot{\rho}_q^{\varkappa}=-\gamma^{\varkappa}\rho_q^{\varkappa}. \qquad (45)$$

The isotropic collisional relaxation constants of the polarization moments occurring in the above expression are found by averaging the relevant elements of the R matrix over the index $\varkappa$:

$$\gamma^{\varkappa}=\frac{1}{2\varkappa+1}\sum_{q=-\varkappa}^{\varkappa}R_q^{\varkappa\varkappa}. \qquad (46)$$

Table III gives our calculated isotropic collisional relaxation constants for the electronic states with $j=1$, 3/2, and 2 corresponding to the anisotropic collisional relaxation matrices $R_q^{\varkappa\varkappa_1}$ listed in Table I. These constants $\gamma^{\varkappa}$ are somewhat larger than those calculated earlier (for $n=6$) when $j=1$ (Refs. 21–24), $j=3/2$ (Refs. 25–27), and $j=2$ (Refs. 24 and 28), differing from the latter by 5–10%. This clearly due to the fact that in the cited papers a detailed numerical integration of the system of equations (23) was carried out in a narrower range of values of the Massey parameter B_n and in the calculations made by us.

CONCLUSIONS

The results reported above can be used in quantitative calculations in a wide range of phenomena associated with anisotropic collisional relaxation of atomic states in the case of extreme anisotropy (as in beam experiments) and also in the case of partly anisotropic velocity distributions. This includes the problem of anisotropic relaxation of atomic states in the case of monochromatic excitation when the anisotropy is due to the Doppler effect,[6,7,29] and also an interesting group of problems of anisotropic relaxation in a plasma when the anisotropy is due to the drift of ions or electrons. In particular, in the case of a plasma one can expect collisional alignment of nonequilibrium populations of levels of spectral multiplets[17,30–32] (self-alignment of ionic and atomic states) and collisional orientation from alignment (self-orientation of the angular momenta).[33,34,35] Investigations of these processes can give valuable information on the ion drift velocity in a plasma and on the rate constants of various elementary processes.[36]

The knowledge of numerical values of elements of the anisotropic collisional relaxation matrix for various values of the electron angular momenta j makes it possible to extend greatly the range of quantitative applications of the theory of anisotropic collisional relaxation to various atomic states and fine or hyperfine multiplets under conditions of extreme or partial anisotropy of the distribution of the relative velocities of the colliding particles.

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