Anisotropic collisional relaxation of polarization moments and self-alignment of ions in a plasma

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(Submitted 9 March 1994)
Zh. Eksp. Teor. Fiz. **106**, 698–724 (September 1994)

A theory is developed for the relaxation of polarization moments of excited ions in a low-temperature plasma due to collisions with neutral atoms, taking into account the anisotropy of the distribution function with respect to the velocities of the colliding particles, which results from ion drift. It is found that in contrast to the isotropic relaxation produced by particles colliding in random directions, anisotropic relaxation can cause the collisional production and interconversion of polarization moments of the ions, including collisional self-alignment. The dynamical multipole moments of the velocity distribution function are introduced in order to describe the deviation from isotropy and to account for the forces between the colliding particles. A relation is established between the dynamical multipole moments and the ion drift velocity in the plasma, and selection rules are found for their contributions to the production and interconversion of polarization moments of different orders. The anisotropic collisional self-alignment of the angular momenta of drifting excited ions and the collisional intramultiplet mixing of the fine-structure levels are analyzed. The rate constants can be expressed in terms of the rate constants for the analogous processes in the most anisotropic possible case of collisions with oppositely directed monokinetic particle beams and in terms of the dynamical multipole moments of the velocity distribution function. A technique is proposed for studying these processes experimentally in terms of measurements of the intensities of the polarization components of ion spectral lines in a plasma. It is applied to measurements of the polarizations of the spectral lines emitted by Ar II ions from the $4p^2P^0$ and $4d'^2P$ excited doublet states in a low-temperature plasma to determine the ion drift velocity and find the rate constants for the collisional production and destruction of alignment in the $J=3/2$ doublet sublevels, and to determine the collisional transport of populations between the $J=1/2$ and 3/2 sublevels. The populations of the doublet sublevels, which are far from equilibrium in the plasma, are determined.

1. INTRODUCTION

The present work is devoted to the derivation of the theory of anisotropic collisional relaxation of polarization moments in atomic particles, and to applying it to excited ions drifting in a low-temperature plasma. Our purpose is to show that this theory permits a quantitative description of a variety of collisional processes in plasmas which lead to self-alignment of electron orbital angular momenta in ions drifting among neutral atoms. Self-alignment of ion angular momenta is demonstrated experimentally, and an experimental/theoretical technique is proposed which allows measurements of the intensities of the polarization components of ion spectral lines to provide a nondestructive contactless method for determining the ion drift velocity in the plasma, the degree of anisotropy of the velocity distributions of the ions and neutral atoms, and the rate constants for collisional processes that result in production and destruction of angular momentum alignment of the electron orbitals of the ions and transport a population between fine-structure multiplets of their energy levels.

These phenomena result from the drift self-alignment of the angular momenta of excited ions,[1] the mechanism for which depends essentially on intramultiplet mixing of their states in collisions with neutral atoms under conditions such that the velocity distribution functions of the colliding particles are anisotropic due to the ion drift. This is reflected in the structure of the present paper. We start by presenting a theory for the collisional relaxation of polarization moments and self-alignment of excited ions drifting in a low-temperature plasma. This theory is then used to analyze our experimental data on the polarization of the spectral lines of argon ions in a gas-discharge plasma.

2. ISOTROPIC AND ANISOTROPIC COLLISIONAL RELAXATION OF ATOMIC POLARIZATION MOMENTS

The polarization of light emitted by excited atoms and ions is determined by the ordering of the angular momenta J of their electron levels. This ordering is characterized qualitatively and quantitatively by the atomic polarization moments ρ_Q^K, which are irreducible spherical tensor components of the density matrix σ_{MM_1} of the ensemble of excited atoms. Their rank K assumes integer values from 0 to $2J$, where J is the total angular momentum quantum number of the electron shell of the excited atomic state. The projections Q of the polarization moments on the quantization axis run from $-K$ to $+K$. In Refs. 2–4 it was shown that the description of all possible types of ordering of the atomic angular momenta reduces to specifying the polarization moments ρ_Q^K,

 1063-7761/94/090384-15$10.00

and that they completely determine the polarization of the light emitted by the atoms. From the selection rules for dipole radiation it follows that the linear polarization of the light is determined by the values of five independent components of the rank-two polarization moment ρ_Q^2 (the so-called alignment tensor), while the circular polarization is determined by the three independent components of the rank-one polarization moment ρ_Q^1 (the so-called orientation vector).

Atomic collisions change the ordering of the atomic angular momenta. This gives rise to collisional relaxation of the polarization moments and hence to changes in the polarization of the light observed experimentally. This is the basis for polarization-spectroscopic techniques for studying the effect of collisions on the internal states of atomic particles; together with the quantum-mechanical collision theory, this is one of the fastest-growing areas in present-day optics. Reviews of research in this area can be found in Refs. 5–9.

Most work on the effect of collisions on the ordering of atomic angular momenta and light polarization has been carried out under the assumption of random collisions, so that all directions are equally probable for particle encounters (and the relative particle velocities are distributed randomly, i.e., isotropically). In this isotropic case the overall effect of collisions on the density matrix of the ensemble of excited atoms becomes spherically symmetric. According to the fundamental theorems of group theory there is no "mixing" of physical quantities belonging to different irreducible representations of the three-dimensional rotation group (determined by the rank K of the polarization moment ρ_Q^K) or to different rows of a single irreducible representation (determined by their projections Q). Consequently, isotropic collisions cause all polarization moments ρ_Q^K to decay independently of one another with rate constants γ^K that depend on the rank K, but not on the projection Q on the quantization axis. Among other things, this is responsible for the different rates of collisional destruction of alignment ($K=2$) and orientation ($K=1$), i.e., linear and circular polarization of light,[3,4] when these rates are independent of the polarization vector and the direction of observation.

The picture of collisional relaxation of the ordering of angular momenta of excited atoms and the polarization properties of light emitted by them becomes significantly more complicated in the anisotropic case when the velocity distribution of the colliding particles is characterized by some preferred spatial direction. In this case, anisotropic collisional relaxation of the atomic polarization moments occurs,[10–13] giving rise to a number of interesting properties which differ significantly from those in the isotropic case. Whereas isotropic collisional relaxation has only a destructive effect, so that it only causes collisional destruction of all kinds of order of atomic angular momenta and reduces the associated light polarization, anisotropic relaxation also has constructive effects. Those give rise to interactions and cause various kinds of ordering of the atomic angular momenta. The most illustrative example of this effect is the transformation of alignment into orientation, and vice versa, accompanied by the transformation of linear light polarization into circular.[11,12,14–24] Another is the creation of alignment of atomic angular momenta and linear light polarization[1,25–36] under anisotropic collisional conditions. Figuratively speaking, anisotropic relaxation transports anisotropy from the atomic particle velocity distribution to the angular momentum distribution of the electron shells.

The basic equations for collisional relaxation follow from simple but rigorous symmetry concepts. As already noted, the angular momentum ordering of the ensemble of atomic particles is determined by the polarization moments ρ_Q^K found by expanding the density matrix σ_{MM_1} in irreducible representations of the rotation group:

$$\rho_Q^K(J)=\sum_{M,M_1}(-1)^{J-M_1}\begin{bmatrix} J & J & K \\ M & -M_1 & Q \end{bmatrix}\sigma_{MM_1}. \tag{1}$$

In accordance with our previous remarks, isotropic collisions give rise to simple truly exponential decay laws for the polarization moments, described by the differential equations

$$\dot{\rho}_Q^K=-\gamma^K\rho_Q^K. \tag{2}$$

The isotropic collisional relaxation constants γ^K which enter into these equations are positive and depend only on the rank K of the polarization moment, not on its projection Q.

When the velocity distributions of the colliding particles are anisotropic the overall effect of the collisions on the state of the atomic ensemble does not have spherical symmetry, but only axial symmetry relative to the preferred direction in space in which encounters between the particles occur most frequently (or least frequently). Accordingly, the collisional relaxation of polarization moments is now classified by the irreducible representations of the group $C_{\infty v}$, consisting of rotations through an arbitrary angle about the anisotropy axis and mirror reflections in the plane perpendicular to this axis. As a result, the anisotropic relaxation of the polarization moments of an ensemble of excited atoms A in a gaseous medium of atoms B can be described by the equations

$$\dot{\rho}_Q^K=-n_B\sum_{K_1}\langle v\sigma_Q^{KK_1}\rangle\rho_Q^{K_1}. \tag{3}$$

Here n_B denotes the number density of the atoms of gas B. The matrix of rate constants $\langle v\sigma_Q^{KK_1}\rangle$ for the anisotropic collisional relaxation which appears on the right-hand side is diagonal in the projections Q of the polarization moments, but not in the rank K and K_1. Consequently, anisotropic collisions mix the polarization moments of different orders but conserve their projections Q on the anisotropy axis. A more detailed analysis of the symmetry properties reveals that when the projection Q is nonvanishing, polarization moments of arbitrary rank K and K_1 can undergo collisional interconversion, whereas for $Q=0$ this can occur only for those whose ranks K and K_1 are both even or both odd.

As a result of the mixing of polarization moments of different ranks K and K_1, and because the relaxation rate constants depend on the projections Q, anisotropic collisions exhibit polarization phenomena which are impossible in principle for random isotropic collisions. These include the examples mentioned above, collisional conversion of alignment into orientation accompanied by the transition from linear to circular light polarization, collisional production of the longitudinal alignment component ρ_0^2 of atomic angular momenta parallel to the anisotropy axis in conjunction with the appearance of linear light polarization, and other interesting effects as well: dependence of the rate of collisional depolarization of light on the direction of polarization with respect to the anisotropy axis,[37–39] dependence of the efficiency of collisionally induced population transport between atomic states on the polarization of the light,[40–49] and transient beat polarization processes due to the simultaneous collisional relaxation of polarization moments of different orders.[24,50–53]

3. NECESSARY CONDITION FOR COLLISIONAL PRODUCTION OF ALIGNMENT

Among the varied anisotropic collisional relaxation processes for the polarization moments we will be interested here primarily in collisional production of alignment of the angular momentum of electron shells in connection with the excitation of ions drifting in a plasma. Just as with other typical anisotropic collisional relaxation processes, this proceeds more effectively as the degree of anisotropy of the velocity distribution function of the colliding particles increases; it disappears entirely for an isotropic spherically symmetric velocity distribution. Along with anisotropic collisions, collisional production of alignment also requires collisional transport of populations between different atomic energy levels (collisional "mixing" just of degenerate magnetic M_J for a single value of J does not suffice). This implies that the kinetic energy of the colliding particles must be sufficiently large in order to mix the different atomic levels, primarily the J components of the atomic multiplets. This important condition for the collisional production of alignment[28] is related to the conservation of particle number and the symmetry of the matrix of rate constants $\langle v\sigma_Q^{KK_1}\rangle$ in Eq. (3), which describe the anisotropic collisional rates of the atomic polarization moments. If the particle trajectories are rectilinear, the matrix of rate constants has a symmetry property associated with time reversal:[54]

$$\langle v\sigma_Q^{KK_1}\rangle=\langle v\sigma_Q^{K_1K}\rangle. \tag{4}$$

Let us now consider the situation in which slow collisions do not mix atomic states with different energies $\mathscr{E}_J$, but can mix degenerate M_J levels. Then the total population n_J of the level $\mathscr{E}_J$ is related to its zeroth-order polarization moment by

$$n_J=(2J+1)^{1/2}\rho_0^0(J), \tag{5}$$

and does not change as a result of collisions. The contribution of the collisions to the time derivative of this polarization moment therefore vanishes. Using the corresponding equation from Eqs. (4) to express this contribution we have

$$\dot{n}_J=-n_B(2J+1)^{1/2}\sum_K \langle v\sigma_0^{0K}\rangle\rho_0^K(J). \tag{6}$$

By virtue of the conservation of the occupation number n_J of the level $\mathscr{E}_J$, the right-hand side of this relation is equal to zero for arbitrary instantaneous values of the polarization moments $\rho_0^K(J)$. Consequently, all rate constants $v\sigma_0^{0K}$ corresponding to the collisional transformation of polarization moments of nonzero order K of level J to this particular occupation number n_J are equal to zero. For the symmetry property (4) it now follows that all the rate constants $\langle v\sigma_0^{K0}\rangle$ corresponding to production of the longitudinal components ρ_0^K of polarization moments of nonzero rank K for any given level J from this same occupation number simultaneously vanish. This also includes the rate constant for the production of the longitudinal alignment $\rho_0^2(J)$. Thus, in the absence of collisional transitions between different energy levels $\mathscr{E}_J$ no alignment of angular momenta occurs, despite the anisotropic nature of the collisions.

4. RATE CONSTANTS FOR COLLISIONAL RELAXATION OF THE POLARIZATION MOMENTS OF IONS FOR TOTALLY AND PARTLY ANISOTROPIC VELOCITY DISTRIBUTION FUNCTIONS OF THE COLLIDING PARTICLES

We first consider the most anisotropic possible case of colliding excited ions A and atoms of an admixture gas B, with oppositely directed monokinetic beams such that the relative velocity is parallel to the z axis and has a fixed value v. In this case the effective cross sections for the collisional relaxation of the polarization moments of the ions can be calculated directly from the solution of the system of differential equations for the impact parameter[54] describing the change in the degenerate excited ion state during the collision. We have performed such calculations numerically for rectilinear particle trajectories, assuming that the splitting of the degenerate A ion energy level with orbital electron angular momentum L in the field of the B atom at a distance R is given by

$$E_{LM}(R)=\frac{3M^2-L(L+1)}{3L(L+1/2)}\frac{\Delta C}{R^6}. \tag{7}$$

Here M is the projection of the orbital angular momentum L of the electron shell of the A ion on the A→B direction. The quantity ΔC is determined by the A–B London interaction along with the orientation energy of the A ion quadrupole moment in the field of the B atom dipole induced by the charge of the A ion.

We have numerically integrated the system of $2L+1$ differential equations using the impact parameter method for different values of the orbital angular momentum of L of the excited A ion for a large number (500) of particle trajectories having different values of the impact parameter. This has enabled us to find the effective cross sections $\sigma_q^{\kappa\kappa_1}(v)$ as a function of the relative particle velocity v for the most anisotropic possible collisional relaxation of orbital polarization moments $\rho_q^\kappa(L)$ of the excited ions. The result is given in the form

$$\sigma_q^{\kappa\kappa_1}(v)=a_q^{\kappa\kappa_1}[\Delta C/\hbar v]^{2/5}, \tag{8}$$

where $a_q^{\kappa\kappa_1}$ are dimensionless numbers tabulated in Ref. 54. From Eq. (8) the rate constants for collisional relaxation of the orbital polarization moments of ions in the most anisotropic possible case of monokinetic collisions are equal to

$$v\sigma_q^{\kappa\kappa_1}(v)=a_q^{\kappa\kappa_1}[\Delta C/\hbar]^{2/5}v^{3/5}. \tag{9}$$

In real situations the relative velocity distribution $F(\mathbf{v})$ of the colliding particles is only partly anisotropic, so that there can be a spread in the vectors $\mathbf{v}$ in both their absolute value and the angle they make with the z axis. In each microscopically small volume we will regard the distribution function to be axisymmetric with respect to the local ion drift direction, which we take to be parallel to the z axis. The collisional relaxation of the polarization moments can now be represented as a superposition of an infinite number of totally anisotropic relaxation processes corresponding to collisions in monokinetic beams with all possible velocity vectors $\mathbf{v}$, taken with weights $F(\mathbf{v})$. As a result we find the following formula for the rate constants for collisional relaxation of the polarization moments of the excited ions:

$$\langle v\sigma_q^{\kappa\kappa_1}\rangle=\sum_{q_1q_2}\int D^{\kappa}_{qq_1}(\phi,\theta,\psi)D^{\kappa_1^*}_{q_2q_1}\times(\phi,\theta,\psi)F(\mathbf{v})\sigma_{q_1}^{\kappa\kappa_1}(v)v^3\sin\theta d\theta d\psi d\phi. \tag{10}$$

The polar angle θ is measured from the z axis. Substituting (9) in (10) we obtain

$$\langle v\sigma_q^{\kappa\kappa_1}\rangle=[\Delta C/\hbar]^{2/5}\sum_{l,q_1}(-1)^{q+q_1}\begin{bmatrix}\kappa & \kappa_1 & l\\ q & -q & 0\end{bmatrix}\times\begin{bmatrix}\kappa & \kappa_1 & l\\ q_1 & -q_1 & 0\end{bmatrix}a_{q_1}^{\kappa\kappa_1}\int F(\mathbf{v})\times P(\cos\theta)v^{13/5}dv\sin\theta d\theta. \tag{11}$$

The coefficient matrix $a_q^{\kappa\kappa_1}$ has the symmetry property[54]

$$a_q^{\kappa\kappa_1}=(-1)^{\kappa-\kappa_1}a_{-q}^{\kappa_1\kappa}. \tag{12}$$

Using the symmetry property of the Clebsch–Gordan coefficients we deduce[55] that only terms with even l remain in the sum (11).

We expand the velocity distribution function in Legendre polynomials:

$$F(\mathbf{v})=\sum_{l=0}^{\infty}f_l(v)P_l(\cos\theta), \tag{13}$$

$$f_l(v)=[(2l+1)/4\pi]\int F(\mathbf{v})P_l(\cos\theta)\sin\theta d\theta d\phi. \tag{14}$$

Substituting (14) into (11) yields, after integration over angles, the following expression for the rate constants for collisional relaxation of the orbital polarization moments of the ions:

$$\langle v\sigma_q^{\kappa\kappa_1}\rangle=\left|\frac{\Delta C}{\hbar}\right|^{2/5}\sum_{l,q_1}(-1)^{q+q_1}\begin{bmatrix}\kappa & \kappa_1 & 2l\\ q & -q & 0\end{bmatrix}\times\begin{bmatrix}\kappa & \kappa_1 & 2l\\ q_1 & -q_1 & 0\end{bmatrix}a_{q_1}^{\kappa\kappa_1}\frac{4\pi}{4l+1}\times\int_0^{\infty}f_{2l}(v)v^{13/5}dv. \tag{15}$$

5. DYNAMIC MULTIPOLE MOMENTS OF THE VELOCITY DISTRIBUTION FUNCTION

We introduce dimensionless quantities which we will call the dynamic multipole moments for the distribution function of the relative velocity of the colliding particles:

$$S_{2l}=[4\pi/(4l+1)][M_AM_B/(M_A+M_B)kT]^{3/10}\int_0^{\infty}f_{2l}(v)v^{13/5}dv. \tag{16}$$

To within a constant factor, these are integrals of the even coefficients (14) in the expansion of the velocity distribution function, evaluated with weight $v^{13/5}$, obtained using the power ($n=6$) in the dynamical law (7) for the splitting of the degenerate energy level of the excited A ion in the field of the B atom.

Using (16) we can write a formula for the relaxation rate constants of the orbital polarization moments (15) as follows:

$$\langle v\sigma_q^{\kappa\kappa_1}\rangle=g_n\sum_{l,q_1}(-1)^{q+q_1}\begin{bmatrix}\kappa & \kappa_1 & 2l\\ q & -q & 0\end{bmatrix}\times\begin{bmatrix}\kappa & \kappa_1 & 2l\\ q_1 & -q_1 & 0\end{bmatrix}a_{q_1}^{\kappa\kappa_1}S_{2l}. \tag{17}$$

The coefficient outside the summation sign is equal to

$$q=|\Delta C/\hbar|^{2/5}|2(M_A+M_B)kT/M_AM_B|^{3/10}. \tag{18}$$

From the triangle condition on the Clebsch–Gordan coefficients it follows that summation over l in Eq. (17) is restricted by the requirement

$$|\kappa-\kappa_1|\leqslant 2l\leqslant\kappa+\kappa_1. \tag{19}$$

For an electron state of the A ion with angular momentum L the ranks κ and κ_1 of the polarization moments can assume values from 0 to $2L$. From (19) it follows that collisional relaxation is determined by the dynamic multipole moments S_{2l} of even order $2l=0,2,4,\dots,2L$. If all even dynamic multipole moments S_{2l} (except the zeroth) in the relative velocity distribution function vanish, while the odd moments have arbitrary values, then the collisional relaxation will end up being completely isotropic.

Collisional transformation of the polarization moments of order κ and κ_1 is possible only when the rate constant $\langle v\sigma^{\kappa\kappa_1}\rangle$ in (17) is nonzero. The ranks of the dynamic multipole moments S_{2l} of the velocity distribution function that take part in collisional relaxation of the polarization moments of rank κ and κ_1 are determined by (19). This is illus-

TABLE I. Involvement of the dynamic multipole moments of the distribution function of the relative velocities of the colliding particles in relaxation processes and in interconversion of the polarization moments for excited atomic particles.

κ	κ_1				
	0	1	2	3	4
0	S_0	–	S_2	–	S_4
1	–	S_0, S_2	S_2	S_2, S_4	S_4
2	S_2	S_2	S_0, S_2, S_4	S_2, S_4	S_2, S_4, S_6
3	–	S_2, S_4	S_2, S_4	S_0, S_2, S_4, S_6	S_2, S_4, S_6
4	S_4	S_4	S_2, S_4, S_6	S_2, S_4, S_6	S_0, S_2, S_4, S_6, S_8

Note. In the diagonal cells ($\kappa=\kappa_1$) the dynamical multipole moments which determine the collisional damping of the polarization moments of order κ are given, while in the off-diagonal cells ($\kappa\neq\kappa_1$) the multipole moments which determine the collisional interconversion of polarization moments of order κ and κ_1 are given.

trated in Table I, which extends over polarization moments of rank κ from 0 to 8. The polarization moment with $\kappa=0$ is proportional to the population of the ion state in question (and to the total intensity of the light emitted from it in transitions); the polarization moments with $\kappa=1$ and 2 (orientation and alignment) determine the circular and linear polarization respectively of light emitted in dipole transitions, while polarization moments of higher rank ($\kappa=3,4,...$) are not involved in light emission, but can affect its polarization indirectly because its relaxation is combined with that of orientation and alignment. From Table I we see that the dynamic quadrupole moment S_2 occupies a special place among the dynamic multipole moments of the velocity distribution function. It controls the most important anisotropic relaxation processes, collisional production of alignment from populations ($\kappa=2$, $\kappa_1=0$), and conversion of alignment into orientation ($\kappa=1$, $\kappa_1=0$).

6. THE RELATIVE-VELOCITY DISTRIBUTION FUNCTION FOR COLLISIONS BETWEEN DRIFTING IONS AND NEUTRAL IONS IN A PLASMA

In a low-pressure discharge plasma, the velocities of the neutral B atoms are isotropic to a good approximation and are described by a Maxwellian

$$f_B(\mathbf{v}_B)=(\beta/\pi)^{3/2}e^{-\beta v_B^2}, \quad (20)$$

where $\beta=M_B/2kT$. The ion distribution function in the plasma is less accurately known. Here we assume that in each macroscopically small volume of the plasma it takes the form of a Maxwellian with an imposed drift:[56]

$$f_A(\mathbf{v}_A)=(\alpha/\pi)^{3/2}\exp\{-\alpha|\mathbf{v}-v_0\mathbf{e}_z|^2\}, \quad (21)$$

where $\alpha=M_A/2kT$. Here $v_0\mathbf{e}_z$ denotes the ion drift velocity vector in the small volume, M_A and M_B are the ion and atom masses respectively, and T is the temperature. The distribution of the relative velocities $\mathbf{v}=\mathbf{v}_B-\mathbf{v}_A$ is found by integrating the product of Eqs. (20) and (21) over velocities $\mathbf{v}_A$ and $\mathbf{v}_B$ with a three-dimensional delta function $\delta(\mathbf{v}-\mathbf{v}_A+\mathbf{v}_B)$. As a result we find

$$f(\mathbf{v})=(\gamma/\pi)^{3/2}\exp\{-\gamma(v^2+2vv_0\cos\theta+v_0^2)\}, \quad (22)$$

where

$$\gamma=M_AM_B/2kT(M_A+M_B) \quad (23)$$

and θ is the angle between the relative velocity and the local ion drift direction, i.e., the z axis. The distribution function of the relative velocities of the B particles and A ions, Eq. (22), is a Maxwellian displaced in the z direction. The surfaces on which the probability of A–B collisions is constant are spheres in the space of the relative velocities with a common center at the point $v_x=v_y=0$, $v_z=v_0$. When the drift velocity v_0 vanishes, the relative velocity distribution (22) is isotropic, and it becomes more anisotropic as v_0 increases. The anisotropy can be quantified by the ratio of the ion drift velocity to the purely Maxwellian value of the most probable relative velocity between the ions and neutral particles, i.e., the dimensionless parameter

$$\lambda=v_0\sqrt{\gamma}. \quad (24)$$

As λ increases, the spheres on which the distribution function (22) is constant move farther and farther along the z axis from the origin of coordinates, and in the limit $\lambda\to\infty$ this distribution approaches the most anisotropic one possible. This is illustrated in Fig. 1.

Substituting (24) into (14) and performing the integration, we find the coefficients of the multipole expansion of the velocity distribution function:

$$f_l(v;\lambda)=\frac{(2l+1)\lambda^2}{2\pi^3 v_0}\exp\{-\lambda[1+(v/v_0)]^2\}$$
$$\times(v/v_0)^{1/2}I_{l+1/2}(2\lambda v/v_0), \quad (25)$$

where $I_{l+1/2}$ is a Bessel function of imaginary argument. Substituting this result into (16) yields the following explicit expression for the even dynamic multipole moments of the velocity distribution function:

$$S_{2l}=\lambda^{2l}e^{-\lambda^2}\left[\Gamma\left(\frac{2l+18/5}{2}\right)/\Gamma\left(2l+\frac{3}{2}\right)\right]$$
$$\times\Phi\left(\frac{2l+18/5}{2};2l+\frac{3}{2};\lambda^2\right). \quad (26)$$

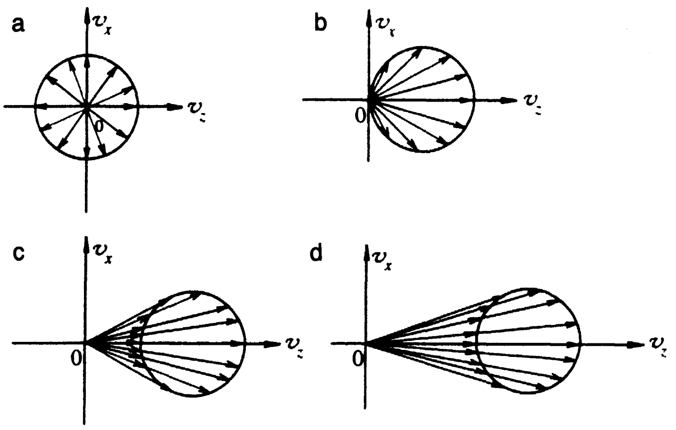

FIG. 1. Shape of the surfaces on which the relative velocity distribution of the drifting excited A ions and neutral B atoms is constant in a low-temperature plasma for different values of the anisotropy parameter $\lambda = v_0[M_A M_B/2kT(M_A+M_B)]^{1/2}$. All the relative velocity vectors $\mathbf{v}$ whose points lie on the indicated surfaces are equally probable. The z axis is parallel to the ion drift velocity $\mathbf{v}_0$. a) $\lambda=0$, isotropic relative velocity distribution; b) $\lambda=1$; c) $\lambda=2$; d) $\lambda=3$.

This contains the confluent hypergeometric function Φ and the gamma function Γ. For small values of the anisotropy parameter λ the dynamic multipole moments S_{2l} are proportional to λ^{2l}, while for large values of λ they behave as $\lambda^{3/5}$. The dynamic multipole moments S_0, S_2, S_4, S_6, and S_8 calculated from (26) are plotted versus λ in Fig. 2.

7. ANISOTROPIC COLLISIONAL RELAXATION OF THE POLARIZATION MOMENTS FOR INTRAMULTIPLET MIXING

In Sec. 3 we saw that collisional alignment of the atomic angular momenta is impossible when collisions do not induce transitions between different energy levels $\mathscr{E}_J$. The situation changes when the anisotropic collisions mix different J states. The derivatives of the populations n_J of the J levels in Eqs. (6) then fail to vanish, and anisotropic collisional alignment can be produced from the occupation numbers n_J. To calculate this process we must find the rate constants that appear in Eq. (1) for collisional relaxation of the polarization moments, which describe the ordering of the angular momenta $\mathbf{J}=\mathbf{L}+\mathbf{S}$ of the fine structure of the level $\mathscr{E}_J$.

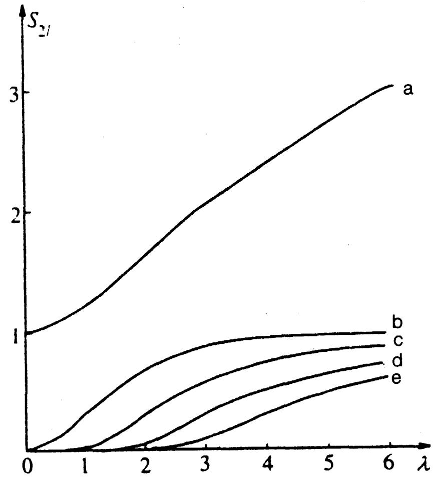

FIG. 2. Dynamic multipole moments S_{2l} of different orders for the relative velocity distribution function of the drifting excited ions and neutral atoms with a degenerate ion energy level split according to $\Delta E = \Delta C/R^6$ as a function of the anisotropy parameter λ: a) $S_0(\lambda)$; b) $S_2(\lambda)$; c) $S_4(\lambda)$; d) $S_6(\lambda)$; e) $S_8(\lambda)$.

For narrow fine-structure multiplets, these rate constants can be expressed in terms of the relaxation rate constants $\langle v\sigma_q^{\kappa\kappa_1}\rangle(L)$ of the orbital polarization moments $\rho_q^\kappa(L)$ (Refs. 57–59). Assume that the splitting $\Delta\mathscr{E}$ of the multiplet is considerably less than the kinetic energy of the colliding particles, and that the condition

$$|\Delta\mathscr{E}|/\hbar \ll v/R, \tag{27}$$

holds. The latter implies that the frequency of the multiplet splitting is small compared with the angular frequency associated with the passage of the B particle past the excited A particle at a typical collisional separation R. Then to a good approximation we can assume that during the collision the coupling between the $\mathbf{L}$ and $\mathbf{S}$ vectors is disrupted, and the orbital angular momentum vector $\mathbf{L}$ is reoriented. The direction of the latter determines the A–B interaction energy in Eq. (7). But the spin $\mathbf{S}$ of the electron shell, whose direction does not depend on this interaction, does not affect the short-lived collision process, and it maintains its orientation.[57] Consequently, in collisions only the orbital polarization moments $\rho_q^\kappa(L)$ relax directly. Their relaxation laws are given by Eq. (17). After the collision the disrupted (L,S) coupling is re-established, and the polarization moments of the fine structure of the $\mathscr{E}_J$ level form again.

These disruption and re-establishment processes for the (L,S) coupling are completely described by the algebra of angular momenta. As a result we find the following expression for the rate constants for anisotropic relaxation of the polarization moments of the J-component multiplet's fine structure:

$$\begin{aligned}\langle v\sigma_Q^{KK_1}\rangle(J,J_1) = \sum_{\kappa,\kappa_1,l,p,q} & (-1)^{K+K_1+Q+p+q-l}(2J+1) \\ & \times(2J_1+1)(2l+1)[(2K+1)(2K_1 \\ & +1)(2\kappa+1)(2\kappa_1 \\ & +1)]^{1/2}\begin{Bmatrix} S & S & l \\ L & L & \kappa \\ J & J & K\end{Bmatrix} \\ & \times\begin{Bmatrix} S & S & l \\ L & L & \kappa_1 \\ J_1 & J_1 & K_1\end{Bmatrix}\begin{Bmatrix}\kappa_1 & \kappa & p \\ K & K_1 & l\end{Bmatrix} \\ & \times\begin{bmatrix}\kappa_1 & \kappa & p \\ q & -q & 0\end{bmatrix}\begin{bmatrix}K_1 & K & p \\ Q & -Q & 0\end{bmatrix} \\ & \times\langle v\sigma_q^{\kappa\kappa_1}\rangle(L).\end{aligned} \tag{28}$$

The ranks of the polarization moments of the J levels are denoted by K and K_1, and their projections by Q, while κ, κ_1, and q denote the ranks and projection of the purely orbital polarization moments $\rho_q^\kappa(L)$.

The equations for anisotropic collisional relaxation of the polarization moments of the fine-structure multiplets can be written in the form

$$\dot{\rho}_Q^K(J) = -n_{\rm B}\sum_{KK_1} \langle v\sigma_Q^{KK_1}\rangle(J,J_1)\rho_Q^{K_1}. \tag{29}$$

8. ANISOTROPIC COLLISIONAL ALIGNMENT OF A NARROW ^2P DOUBLET

Now we consider the collisional alignment of the $J=3/2$ level of a narrow ^2P doublet of an excited ion resulting from its own drift through the neutral atoms in the positive column of a gas discharge.

In accordance with (5), the populations of the $J=1/2$ and 3/2 levels of the doublet can be expressed in terms of the corresponding zeroth-order polarization moments: $n_{1/2}=\sqrt{2}\rho_0^0(1/2)$, $n_{3/2}=2\rho_0^0(3/2)$. We will describe the longitudinal alignment of the $J=3/2$ level by the quantity $a_{3/2}=2\rho_0^2(3/2)$. Focusing on the block with $Q=0$ in the system of equations (29), which describes collisional relaxation of the populations of the doublet levels together with longitudinal alignment of the $J=3/2$ level, we obtain the system of equations

$$\begin{aligned}\dot{n}_{1/2}&=n_{\rm B}[-\langle v\sigma_p\rangle n_{1/2}+(1/2)\langle v\sigma_p\rangle n_{3/2}+(1/2)\\&\quad\times\langle v\sigma_a\rangle a_{3/2}]-\Gamma_0 n_{1/2}+F_{1/2},\\ \dot{n}_{3/2}&=n_{\rm B}[\langle v\sigma_p\rangle n_{1/2}-(1/2)\langle v\sigma_p\rangle n_{3/2}-(1/2)\\&\quad\times\langle v\sigma_a\rangle a_{3/2}]-\Gamma_0 n_{3/2}+F_{3/2},\\ \dot{a}_{3/2}&=n_{\rm B}[\langle v\sigma_a\rangle n_{1/2}-(1/2)\langle v\sigma_a\rangle n_{3/2}-\langle v\sigma_d\rangle a_{3/2}]\\&\quad-\Gamma_0 a_{3/2}.\end{aligned} \tag{30}$$

Here we have introduced the abbreviated notation $\langle v\sigma_p\rangle$ for the rate constant for for transport of population between the $J=1/2$ and $J=3/2$ levels, $\langle v\sigma_a\rangle$ for collisional production of alignment, and $\langle v\sigma_d\rangle$ for collisional destruction (depolarization) of alignment. These are positive quantities and can be expressed in terms of the rate constants $\langle v\sigma_Q^{KK_1}\rangle(J,J_1)$ which appear in Eqs. (28):

$$\begin{aligned}\langle v\sigma_p\rangle&=-\sqrt{2}\langle v\sigma_0^{00}\rangle(1/2,3/2)=-\sqrt{2}\langle v\sigma_0^{00}\rangle\\&\quad\times(3/2,1/2),\\ \langle v\sigma_a\rangle&=-\sqrt{2}\langle v\sigma_0^{20}\rangle(3/2,1/2)=-\sqrt{2}\langle v\sigma_0^{02}\rangle\\&\quad\times(1/2,3/2),\\ \langle v\sigma_d\rangle&=\langle v\sigma_0^{22}\rangle(3/2,3/2).\end{aligned} \tag{31}$$

The radiative damping constant is denoted by Γ_0 in Eqs. (30); $F_{1/2}$ and $F_{3/2}$ are the joint intensities for population of the $J=1/2$ and 3/2 levels of the doublet by processes in the plasma.

In Eqs. (30) the rate of collisional transport of population from the $J=1/2$ level to the $J=3/2$ level is twice the rate of transport in the opposite direction; these rates are inversely proportional to the statistical weights $2J+1$ of the levels. The fact that the ratio of the direct and inverse population transport rates does not depend on the multiplet splitting $\Delta\mathscr{E}$ is a shortcoming of the extreme simplification in the (L,S)-coupling disruption model during collisions. Specifically, it follows from the principle of detailed balance[61] that collision-induced transitions between levels in which the energy increases by $|\Delta\mathscr{E}|$ are "inhibited" in comparison with the inverse transitions, where the magnitude of this "inhibition" is given by the Boltzmann factor

$$y=\exp(-|\Delta\mathscr{E}|/kT_{\rm eff}), \tag{32}$$

where $T_{\rm eff}$ is the effective temperature of the ensemble of colliding particles. It is determined by the mean squared relative velocity, $T_{\rm eff}=M_{\rm A}M_{\rm B}\langle v^2\rangle/3k(M_{\rm A}+M_{\rm B})$. For the velocity distribution (22) this mean value exceeds the pure Maxwellian value by the square of the drift velocity v_0^2. Hence the effective temperature of the collisions exceeds the average temperature T of the plasma particles and is given by

$$T_{\rm eff}=[1+(3/2)\lambda^2]T \tag{33}$$

In normal fine-structure multiplets the $J=3/2$ level lies above the $J=1/2$ level. For these, the y factors from (32) should be included in the terms of Eqs. (30) describing collisional transitions $J=1/2\rightarrow J=3/2$ with an increase in energy. Thus we obtain equations for the combined anisotropic relaxation of the populations and longitudinal alignment of the levels of a narrow ^2P-doublet, we find by means of the principle of detailed balance:

$$\begin{aligned}\dot{n}_{1/2}&=n_{\rm B}[-y\langle v\sigma_p\rangle n_{1/2}+(1/2)\langle v\sigma_p\rangle n_{3/2}+(1/2)\\&\quad\times\langle v\sigma_a\rangle a_{3/2}]-\Gamma_0 n_{1/2}+F_{1/2},\\ \dot{n}_{3/2}&=n_{\rm B}[y\langle v\sigma_p\rangle n_{1/2}-(1/2)\langle v\sigma_p\rangle n_{3/2}-(1/2)\\&\quad\times\langle v\sigma_a\rangle a_{3/2}]-\Gamma_0 n_{3/2}+F_{3/2},\\ \dot{a}_{3/2}&=n_{\rm B}[y\langle v\sigma_a\rangle n_{1/2}-(1/2)\langle v\sigma_a\rangle n_{3/2}-\langle v\sigma_d\rangle a_{3/2}]\\&\quad-\Gamma_0 a_{3/2}.\end{aligned} \tag{34}$$

From these equations we deduce several important consequences for combined anisotropic collisional relaxation of populations and longitudinal alignment of the levels of a narrow fine-structure multiplet.

Taking into account the statistical weights of the doublet levels and the Boltzmann factor under thermal equilibrium conditions, we find that the populations of the doublet levels are related by $yn_{1/2}-(1/2)n_{3/2})=0$. Here the terms describing collisional transport of the populations n_J between levels $J=1/2\leftrightarrow 3/2$ vanish. This can be seen from the first two equations of Eqs. (34), and agrees with the principle of detailed balance.

The first two terms on the right-hand side of the third of Eqs. (34) describe the anisotropic collisional production of $a_{3/2}$ alignment from the populations. Since they both contain the same rate constant $\langle v\sigma_a\rangle$, the efficiency with which alignment is produced is proportional to the quantity $\Delta n=yn_{1/2}=(1/2)n_{3/2}$, i.e., to the amount by which the populations of the doublet levels deviated from their equilibrium values. In other words, anisotropic collisions can give rise to alignment only from nonequilibrium populations of fine-structure multiplet levels, and the efficiency of this process increases when the populations deviate from their equilibrium values.

Finally, the third terms in square brackets on the right-hand sides of the first two of Eqs. (34) describe the effect of alignment of the angular momenta of the ensemble of ions in the $J=3/2$ state on the rates of transitions induced by collisions between the fine-structure multiplet levels. This phenomenon was predicted theoretically in Ref. 41 for the general case of anisotropic collisions. Like other characteristic features of anisotropic collisional relaxation, the effect of alignment on transport of populations is more pronounced as the distribution function of the velocities of the colliding particles becomes more anisotropic. Calculations[41,47] show that in the most anisotropic possible collisions with oppositely directed beams, the efficiency for collisional transport of population between levels of a fine-structure multiplet varies as a function of the magnitude and sign of the alignment of the $J=3/2$ level by a factor of 5–10. For quantitative calculations of the combined anisotropic relaxation of populations and alignment of doublet levels according to Eqs. (34), we need to know the rate constants $\langle v\sigma_p\rangle$, $\langle v\sigma_d\rangle$, and $\langle v\sigma_a\rangle$ that appear in them. We have used Eq. (17) to calculate these constants using the expression (26) for the dynamic multipole moments S_{2l} of the distribution function of the relative velocities of the excited ions in the ^2P state drifting in a medium of neutral atoms. The values of the quantities $a_q^{\kappa\kappa_1}$ were used in Ref. 54. Table II displays the rate constants calculated in this manner which determine the combined isotropic collisional relaxation of the populations and alignment of the levels of a narrow ^2P doublet of drifting ions in a plasma with different values of the anisotropy parameter λ given by (24).

TABLE II. Rate constants for the processes for collisional production of alignment, $\langle v\sigma_a\rangle$; destruction of alignment, $\langle v\sigma_d\rangle$; and transport of populations, $\langle v\sigma_p\rangle$, for the components of the narrow ^2P doublet of excited ions drifting in a plasma as a function of the anisotropy parameter λ.

λ	$\langle v\sigma_a\rangle$	$\langle v\sigma_d\rangle$	$\langle v\sigma_p\rangle$
0	0	2.87	2.00
0.2	0.023	2.90	2.01
0.4	0.091	3.00	2.06
0.6	0.20	3.15	2.13
0.8	0.34	3.34	2.23
1.0	0.49	3.57	2.35
1.2	0.66	3.82	2.48
1.4	0.83	4.07	2.62
1.6	1.00	4.33	2.76
1.8	1.16	4.58	2.91
2.0	1.31	4.83	3.06
2.2	1.46	5.07	3.20
2.4	1.60	5.30	3.35
2.6	1.73	5.53	3.49
2.8	1.85	5.75	3.64
3.0	1.97	5.97	3.77
3.2	2.08	6.18	3.91
3.4	2.19	6.39	4.04
3.6	2.29	6.60	4.17
3.8	2.39	6.80	4.31
4.0	2.49	6.99	4.43
4.2	2.58	7.19	4.56
4.4	2.67	7.38	4.68
4.6	2.76	7.57	4.80
4.8	2.84	7.75	4.92
5.0	2.92	7.93	5.04

Note. The rate constants are expressed in units of $|\Delta C\hbar|^{2/5}\gamma^{3/10}$.

9. RELATION BETWEEN THE RATE CONSTANTS OF THE COLLISIONAL PROCESSES AND THE POLARIZATION PROPERTIES OF LIGHT

In a plasma, a wide variety of physical processes take place simultaneously. This greatly complicates the analysis because we do not know the values of the corresponding (sometimes numerous) parameters which determine the plasma state in each specific experimental situation. Hence the polarization-spectroscopic technique is worthy of attention. This technique, proposed in Ref. 30, allows one to determine the ion drift velocity, the rate processes for processes by which alignment is produced and destroyed collisionally, and the transport of population between fine-structure sublevels of the ion multiplets to be determined just from measuring the relative intensities of the polarization components of the light emitted by excited ions. We briefly describe the theoretical basis for this technique.

The intensity of light emitted in the transition $(J,L)\rightarrow(J_0,L_0)$ and polarized in the direction of the unit vector $\mathbf{e}$ is equal to

$$I_e=C(L,L_0)(-1)^{J+J_0}(2J+1)(2J_0+1)\times\begin{Bmatrix} L_0 & J_0 & S \\ J & L & 1 \end{Bmatrix}\sum_{K,Q,q_1,q_2}(-1)^{K+Q}(2K+1)^{1/2}\begin{Bmatrix} J & J & K \\ 1 & 1 & J \end{Bmatrix}\begin{bmatrix} K & 1 & 1 \\ -Q & q_1 & q_2 \end{bmatrix}e^*_{q_1}e_{q_2}\rho^K_Q(J). \tag{35}$$

The coefficient $C(L,L_0)$ depends on the oscillator strength of the transition; under the condition that the splitting is small in comparison with the frequency of the optical transition, it is independent of J and J_0; here e_{q1} and e_{q2} denote the components of the polarization unit vector e projected on the circular unit vectors. For the case of interest to us, involving optical transitions from the levels of the ^2P doublet ($L=1, J=1/2$ or $3/2$), Eq. (35) yields the following relations which connect the populations of the doublet levels and the alignment of the $J=3/2$ level to the experimentally observed light intensities I_x, I_y, and I_z polarized parallel to the Cartesian axes:

$$(I_y-I_z)_{J=3/2}=C(1,L_0)\sqrt{2}(-1)^{J_0+3/2}(2J_0+1)$$
$$\times\begin{Bmatrix} 3/2 & 3/2 & 2 \\ 1 & 1 & J_0 \end{Bmatrix}$$
$$\times\begin{Bmatrix} L_0 & J_0 & 2 \\ 3/2 & 1 & 1 \end{Bmatrix}^2 a_{3/2}, \tag{36a}$$

$$(I_y+I_z/2)_{J=3/2}=C(1,L_0)(-1)^{J_0+3/2}(2J_0+1)$$
$$\times\begin{Bmatrix} 3/2 & 3/2 & 0 \\ 1 & 1 & J_0 \end{Bmatrix}$$
$$\times\begin{Bmatrix} L_0 & J_0 & 1/2 \\ 3/2 & 1 & 1 \end{Bmatrix}^2 n_{3/2}, \tag{36b}$$

$$(I_y+I_z/2)_{J=1/2}=C(1,L_0)(-1)^{J_0+1/2}(2J_0+1)$$
$$\times\begin{Bmatrix} 1/2 & 1/2 & 0 \\ 1 & 1 & J_0 \end{Bmatrix}$$
$$\times\begin{Bmatrix} L_0 & J_0 & 1/2 \\ 1/2 & 1 & 1 \end{Bmatrix}^2 n_{1/2}. \tag{36c}$$

Now we consider a plasma in steady state, where the time derivatives on the left-hand side of Eqs. (34) vanish. From the third equation of this system it follows that in steady state we have

$$a_{3/2}/[yn_{1/2}-(1/2)n_{3/2}]=\langle v\sigma_a\rangle p/[\langle v\sigma_d\rangle p+kT\Gamma_0]. \tag{37}$$

Here we have expressed the density of the neutral atoms in terms of the pressure p and temperature T in the plasma. It is essential that this ratio be independent of the rates $F_{1/2}$ and $F_{3/2}$ of the plasma processes populating the levels of the doublet, whose magnitudes are generally unknown. Now we express the quantities $a_{3/2}$, $n_{1/2}$, and $n_{3/2}$ in terms of the intensities of the polarized light using the relations (36). As a result we obtain a formula which directly relates the rate constants for collisional production of alignment $\langle v\sigma_a\rangle$ and its destruction $\langle v\sigma_d\rangle$ with the experimentally observed intensities of the polarized light components.

In the experimental part of this work we studied the polarization of the spectral lines emitted in transitions from the J levels of the excited $4d'\ {}^2P$ doublet of the argon ion to the $4p'\ {}^2D^0_{3/2}$ level or from the J levels of the $4p\ {}^2P^0$ doublet to the $4s\ {}^2P_{3/2}$ level. In the former case we have

$$\frac{\langle v\sigma_a\rangle p}{\langle v\sigma_d\rangle p+kT\Gamma_0}=\frac{12.5(I_y-I_z)_{3/2}}{5(I_y+0.5I_z)_{3/2}-y(I_y+0.5I_z)_{1/2}}, \tag{38a}$$

and in the latter we have

$$\frac{\langle v\sigma_a\rangle p}{\langle v\sigma_d\rangle p+kT\Gamma_0}=\frac{(I_y-I_z)_{3/2}}{(I_y+0.5I_z)_{3/2}-2y(I_y+0.5I_z)_{1/2}}. \tag{38b}$$

The subscripts 1/2 and 3/2 indicate the value of J associated with the level from which the light is emitted.

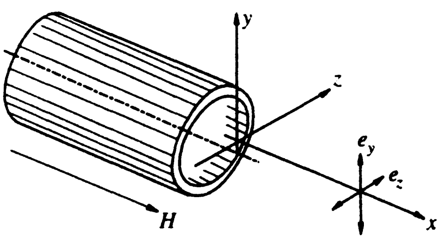

FIG. 3. Cylindrical hollow cathode and its associated coordinate system for polarization radiation observed from a macroscopically small negative glow region of the discharge plasma.

Equations (38a, b) are the key to the theoretical interpretation of measurements of the polarization of ion lines in a plasma. The results of these measurements and their analysis are given in the next section.

In conclusion we note that this approach is also applicable for describing polarization phenomena in the case of more complicated excited multiplets (not necessarily ${}^2P$ doublets) of the drifting ions.

10. EXPERIMENTAL OBSERVATION OF THE DRIFT SELF-ALIGNMENT OF IONS IN LOW-TEMPERATURE PLASMAS

For polarization effects associated with ion drift to be observable, the drift velocity must be large in the radiating part of the discharge. This condition is not satisfied in normal gas-discharge systems (the positive column of a constant-current discharge or a capacitive high-frequency discharge), where the ion drift velocities are typically low in this region. For this reason we chose to study the gas discharge inside a hollow cathode.

The electric field profile in such a discharge is very nonuniform: its strength falls off sharply as a function of distance from the cathode surface toward the center of the discharge.[62] Mass-spectrometric studies have shown[63] that essentially all positive ions are formed in the negative glow region and diffuse (when the mean free path is of order 10^{-1} for pressures of order 0.1 torr) toward the cathode, acquiring kinetic energy at its surface, which determines the potential drop in the discharge gap. At the periphery of the negative glow region near the cathode dark space there is a transition region of finite extent, in which there is a sizable potential gradient and high ion drift velocities.

We have studied a gas discharge in argon with a cylindrical hollow aluminum cathode having a diameter of 1 cm and a length of 5 cm at a discharge potential of 80–100 V, discharge current of 10–60 mA, and pressure 0.1–0.4 torr. We measured the intensities I_z and I_y of the linear polarization of the spectral lines emitted by the discharge along sight lines parallel to the x axis of the cathode (Fig. 3) and by using a diaphragm to isolate narrow regions (with cross section less than 1 mm^2) of the discharge emission at various distances from the cathode surface. The intensities of the individual linearly polarized light components were measured by means of a photomultiplier tube connected to an electronic integrator, and the small difference I_z-I_y in the

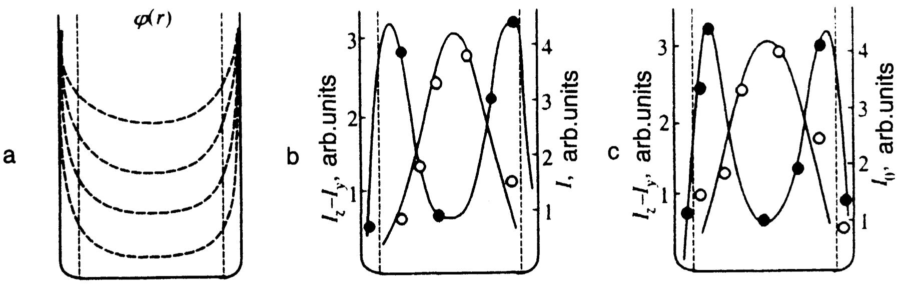

FIG. 4. a) Qualitative sketch of the equipotential surfaces in a discharge with a cylindrical hollow cathode (following Ref. 62). b) Radial dependence of the total light intensity $I = I_y + I_z$ and the polarization signal $I_z - I_y$ for the Ar II spectral line $\lambda = 3819$ Å ($4d'\ ^2P_{3/2} - 4p'\ ^2D_{3/2}$) in the discharge of a hollow cathode. c) Radial dependence of the total light intensity $I_0 = I + I_z$ and the polarization signal $I_z - I_y$ of the Ar II ion spectral line $\lambda = 4764$ Å ($4p\ ^2P^0_{3/2} - 4s\ ^2P_{1/2}$) in the discharge of a hollow cathode.

intensity of light linearly polarized along the radius of the cylindrical cathode and parallel to its surface respectively was measured using a magnetic polarization spectrometer.[64]

In accordance with the comments made in Sec. 3, collisional alignment can be expected in the case of narrow spectral multiplets effectively mixed by collisions. We therefore studied narrow doublets of excited Ar^+ ions. For calibration purposes, in addition to the ion spectral lines we also studied the lines of neutral Ar atoms. The aggregate of experimental data which we obtained for different Ar^+ and Ar ions and in different geometrical arrangements of the observation region enabled us to draw two general conclusions.

1. The peak of the total radiation intensity $I_z + I_y$ both from ions and from neutral atoms under our experimental conditions occurs in the central region of the hollow cathode, i.e., in the region with the highest density of the fast electrons that excite them (see Fig. 4).

2. For the ion lines the polarization signal $I_z - I_y$ increases rapidly as we scan the observation region from the center of the discharge toward the cathode surface, and it attains its largest value at the edge of the negative glow region adjacent to the cathode dark space at a distance of about 1.5 mm from the cathode surface (Fig. 4). But the intensity of these same ion lines as a function of the location of the observation region is qualitatively quite different and is described by a bell-shaped curve. It is noteworthy that the polarization signal $I_z - I_y$ for the ion lines in the boundary region of the negative glow is more than a factor of ten stronger than the corresponding signal for the neutral atoms, despite the enormous difference in the densities of the neutral atoms and the ions (see Fig. 5a, b).

The first of these facts points at collisions with fast electrons as the general reason for excitation of ions and neutral atoms. The second fact implies that there exists a very different explanation for the alignment of angular momenta of the neutral atoms and the ions at the edge of the glow region. Whereas for the neutral atoms alignment is produced by anisotropic electron collisions, the self-alignment of ion angular momenta is due to collisions with the neutral atoms, which are anisotropic in nature because of the ion drift, whose velocity reaches its peak value in this region.

We have studied two groups of spectral lines of the Ar^+ ions in greatest detail, corresponding to optical transitions from the $4d'^2P_{1/2,3/2}$ and $4p^2P^0_{1/2,3/2}$ levels of the excited Ar II doublets respectively to the $4p'^2D^0_{3/2}$ and $4s^2P_{3/2}$ levels. The splitting of the levels of the first of these excited doublets is equal to $\Delta\mathscr{E} = 0.01$ eV$=110$ K, while that of the levels of the second doublet is equal to $\Delta\mathscr{E} = 0.07$ eV$=770$ K.

From the experimentally measured total intensities $I_y + I_z$ and the polarization signal $I_y - I_z$ we found the values of I_y and I_z appearing respectively on the right-hand side of Eq. (38a) (for the two $4d'^2P_{1/2,3/2} - 4p'^2D^0_{3/2}$ spectral emission lines) or on the right-hand side of Eq. (38b) (for the two $4p^2P^0_{1/2,3/2} - 4s^2P_{3/2}$ spectral emission lines). Representing this combination as $A(I_y, I_z)$ for brevity we have from Eqs. (38a) and (38b)

$$\langle v\sigma_a \rangle / [\langle v\sigma_d \rangle + kT\Gamma_0/p] = A(I_y, I_z). \tag{39}$$

For specified temperature T and pressure p and constant radiative damping Γ_0, the left-hand side of this relation depends on the anisotropy parameter λ through the rate constants $\langle v\sigma_a \rangle$ and $\langle v\sigma_d \rangle$, whose evaluation was described above (see Table II). For an arbitrary gas-discharge regime this enables us to use measurements of the linearly polarized spectral line intensities to determine the anisotropy parameter λ, and along with it [using Eq. (24)] the ion drift velocity v_0. The value of λ thus obtained was used to calculate the dynamic multipole moments S_{2l} of the velocity distribution function from Eq. (26), and then using Eq. (17) to calculate the rate constants for collisional production of alignment $\langle v\sigma_a \rangle$ and for its collisional destruction (depolarization) $\langle v\sigma_d \rangle$, and the collisional transport of populations $\langle v\sigma_p \rangle$ between levels of the excited fine-structure multiplet of the drifting ions being studied.

The measured dependence of the functions $A(I_y, I_z)$ on the argon pressure at $T = 330$ K is shown in Figs. 6 and 7 respectively for the doublet spectral lines $4d'^2P_{1/2,3/2} - 4p'^2D^0_{3/2}$ ($\lambda = 383.0$ and 381.9 nm) and $4p^2P^0_{1/2,3/2} - 4s^2P_{3/2}$ ($\lambda = 466.8$ and 454.5 nm). We carried out a theoretical analysis of the experimental results using values of the radiative damping constants for the $4d'^2P_{1/2,3/2}$ and $4p^2P^0_{1/2,3/2}$ excited doublet states, equal respectively to $\Gamma_0 = 1.73 \cdot 10^8\ s^{-1}$ (from Ref. 65) and $1.11 \cdot 10^8\ s^{-1}$ (from Ref. 66), and the rate constants for the combined London and charge-quadrupole interaction, which deter-

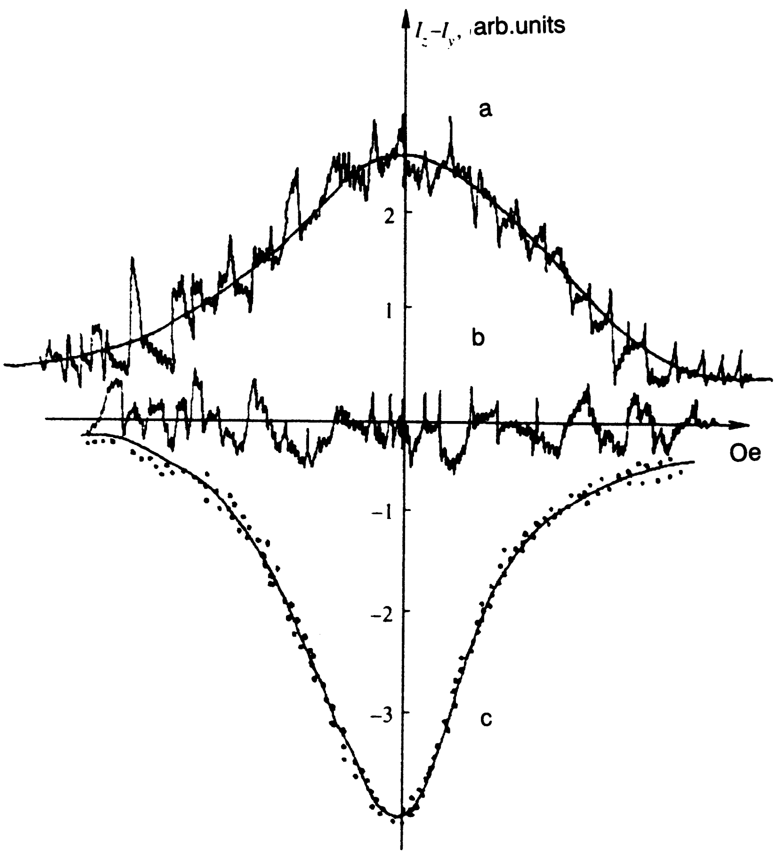

FIG. 5. Comparison of the polarization-magnetic signals $I_z(H)-I_y(H)$ of the ion and atomic lines at the edge of the negative glow region of the hollow cathode and in the positive column of the discharge. a) Ar II ion line $\lambda=4545$ Å ($4p\ ^2P^0_{3/2}-4s\ ^2P_{3/2}$) at the edge of the negative glow of the hollow cathode. b) Ar I atomic line $\lambda=4300$ Å ($5p[2_{1/2}]_2-4s[1_{1/2}]_1$) at the edge of the negative glow of a hollow cathode. c) Ar I atomic line $\lambda=4300$ Å ($5p[2_{1/2}]_2-4s[1_{1/2}]_1$) in the positive column of a constant-current discharge.

mined the splitting of the degenerate excited ion level in the course of a collision with a neutral atom, amounting under these conditions respectively to $\Delta C=2150\cdot 10^{-60}$ and $365\cdot 10^{-60}$ erg·cm^6. We calculated the quantities ΔC using the model potential method of Kratzer and Simons,[67] which is similar to the quantum defect technique.

With this approach only one parameter remained free in the theoretical values of the rate constants $\langle v\sigma_a\rangle$ and $\langle v\sigma_d\rangle$ determined in Table II: the anisotropy parameter λ of the velocity distribution function. The magnitude of λ was determined from the requirement that the theoretically calculated left-hand side of relation (39) agree with its experimentally measured right-hand side. The results of the theoretical calculations of $A(I_y,I_z)$ for both of the Ar II ion doublets considered as a function of pressure are shown in Figs. 6 and 7 by the solid traces.

Knowledge of the anisotropy parameter λ of Eq. (18), together with the rate constants of the anisotropic collisional processes, enables us also to determine the ion drift velocity v_0. The theoretical analysis of the quantity $A(I_y,I_z)$ measured experimentally thus yields detailed information about the ion drift and the rate constants for a variety of different anisotropic collisional processes involving excited drifting ions in a plasma. Tables III and IV display the results which we obtained in this way for Ar^+ ions in the excited states $4d'\,^2P_{1/2,3/2}$ and $4p\,^2P^0_{1/2,3/2}$ in the region at the edge of the negative glow, where the alignment signal of the ion lines is largest.

Comparison of the data of these tables obtained from independent experimental measurements with two different

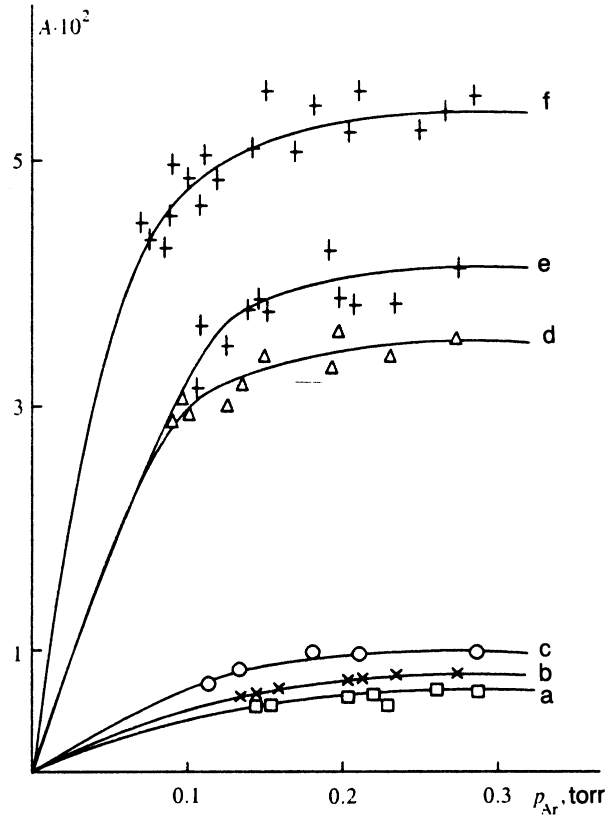

FIG. 6. The quantity $A(I_y,I_z)$ determined from Eq. (38a) as a function of the argon pressure p_{Ar} for the Ar II ion doublet spectral lines $4d'\ ^2P_{1/2,3/2}-4p'\ ^2D^0_{3/2}$ in the discharge of a hollow cathode of diameter 10 mm at various distances z from the cathode axis (in mm): a) $z=0$; b) 0.5; c) 1; d) 2.25; e) 2.6; f) 3.

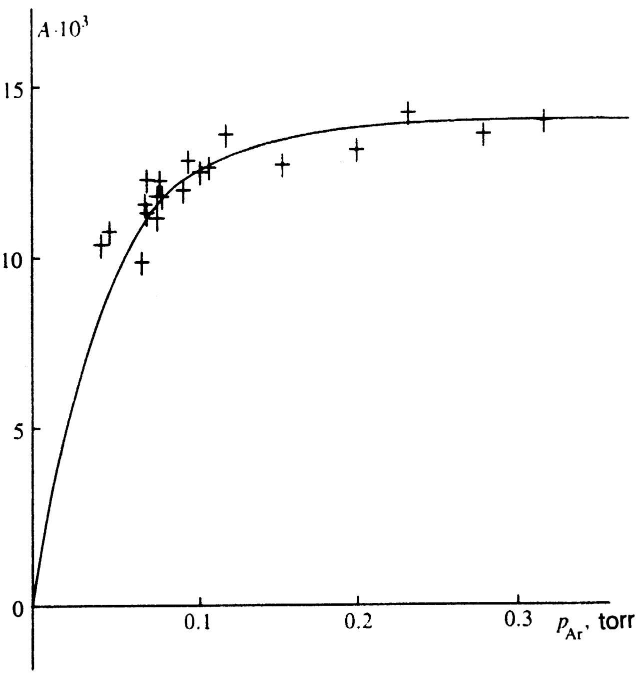

FIG. 7. Experimentally measured and theoretically calculated quantity $A(I_y, I_z)$ determined by Eq. (38b) as a function of the argon pressure in the discharge of a hollow cathode for the Ar II ion spectral doublet $4p\ ^2P^0_{1/2,3/2}-4s\ ^2P_{3/2}$.

excited argon ion doublets reveals good agreement between the ion drift velocities determined by the method presented here (to within 20%).

In analyzing the data on the rate constants of collisional processes it should be kept in mind that when the walls of the discharge tube are held at a constant temperature T, the ion mean free path falls off as the pressure p increases. This is accompanied by a reduction in the average ion drift velocity v_0 and the anisotropy parameter λ. Along with λ, the effective temperature T_{eff} given by Eq. (35) for the distribution of the relative velocities of the ions and neutral atoms also decreases (by almost a factor of six as we go from a pressure of 0.02 torr to a pressure of 0.30 torr; see Table IV), which corresponds to a decrease in the average relative velocity of the colliding ions and neutral atoms by almost a factor of 2.5. Consequently, as the pressure p increases, the rate constants of the collisional processes decrease. Among them the rate constant $\langle v\sigma_a\rangle$ for collisional production of alignment decreases the fastest, since this process is entirely due to the anisotropy of the relaxation and vanishes at $\lambda=0$. For the rate constants of depolarization $\langle v\sigma_d\rangle$ and population transport $\langle v\sigma_p\rangle$, the pressure dependence is weaker: they remain nonzero even in the isotropic case, i.e., when there is no ion drift (at $\lambda=0$).

The intensities I_z and I_y of the polarization components of light emitted at various distances from the cathode were measured for the $4d'\,^2P_{1/2,3/2}$ excited doublet state, and we analyzed the dependence of $A(I_y, I_z)$ on the argon pressure as described above. As a result we determined the radial dependence of the anisotropy parameter λ and the ion drift velocity v_0 associated with it. This enabled us to construct radial profiles $v_0(z)$ of the ion drift velocity at different pressures in the plasma (Fig. 8).

In Sec. 3 it was noted that the necessary condition for collisional production of alignment, in addition to the anisotropy of the collisions, is that the ratio of the populations of the sublevels of the multiplet mixed by collisions differ from its equilibrium value. For ^2P doublets this equilibrium value is equal to $2y$, where y is given by Eq. (32). For measurements of light emitted in $^2P^0_J-{}^2P_{3/2}$ transitions the deviation in the level populations of the $^2P^0_{1/2,3/2}$ doublet from their equilibrium values is characterized by the quantity

$$(n_{3/2}/n_{1/2})-2y=0.4[(I_y+I_z/2)_{J=3/2}/(I_y+I_z/2)_{J=1/2}]-2y. \tag{40}$$

Figure 9 shows the ratio of the intensities which appears on the right-hand side of this relation, measured experimentally for the sublevels of the doublet $4p^2P^0$ of Ar II. From the figure it is clear that in the plasma the sublevels of this doublet are populated in a very nonequilibrium fashion: the $J=3/2$ level is depleted, while the $J=1/2$ level has an excess population, which creates favorable conditions for collisional self-alignment of the angular momentum of the electron shells of the drifting ions and for observation of the associated polarization phenomena.

TABLE III. The anisotropy of the collisional relaxation, drift velocity, and rate constants for anisotropic collisional processes of the $4d'\ ^2P_{1/2,3/2}$ doublet of Ar^+ ions in a low-pressure plasma at 330 K.

Pressure, torr	Boltzmann factor y	Anisotropy parameter λ	Drift velocity v_0, 10^5 cm/s	Rate constants for the collisional processes, 10^{-9} cm³/s		
				$\langle v\sigma_a\rangle$	$\langle v\sigma_d\rangle$	$\langle v\sigma_p\rangle$
0.02	0.98	4.65	2.43	2.49	6.84	4.34
0.04	0.98	4.50	2.35	2.44	6.71	4.26
0.06	0.98	4.40	2.30	2.40	6.63	4.21
0.08	0.98	4.35	2.27	2.32	6.48	4.13
0.10	0.97	4.00	2.09	2.23	6.28	3.98
0.12	0.97	3.62	1.89	2.07	5.98	3.77
0.14	0.96	3.22	1.68	1.88	5.57	3.53
0.16	0.96	2.92	1.52	1.73	5.29	3.34
0.20	0.94	2.42	1.26	1.45	4.78	3.02
0.30	0.91	1.79	0.93	1.04	4.11	2.61

TABLE IV. The anisotropy of the collisional relaxation, drift velocity, and rate constants for anisotropic collisional processes of the $4p'\ {}^2P^0_{1/2,3/2}$ doublet of Ar^+ ions in a low-pressure plasma at 330 K.

Pressure, torr	Boltzmann factor y	Anisotropy parameter λ	Drift velocity v_0, 10^5 cm/s	Rate constants for the collisional processes, 10^{-9} cm^3/s		
				$\langle v\sigma_a\rangle$	$\langle v\sigma_d\rangle$	$\langle v\sigma_p\rangle$
0.02	0.842	4.34	2.22	1.17	3.25	2.05
0.04	0.825	4.07	2.09	1.11	3.14	1.98
0.06	0.807	3.83	1.97	1.06	3.03	1.91
0.08	0.787	3.62	1.83	1.01	2.94	1.85
0.10	0.765	3.42	1.79	0.97	2.85	1.80
0.12	0.746	3.25	1.70	0.93	2.77	1.75

11. CONCLUSION

The results obtained here show that the theory of anisotropic collisional relaxation of the polarization moments of atomic particles can be applied to excited ions in a plasma, and provides a basis for quantitative spectral polarimetric investigations of the drift of excited ions and a variety of anisotropic collisional processes involving them. In the model we have adopted, the anisotropy of the relative velocity distribution function and its dynamic multipole moments depend on the single parameter λ, whose value can be determined by analyzing experimentally measured intensities of the polarization components of ion spectral lines. Knowing the value of λ in turn enables us to determine the ion drift velocity and the rate constants for the collisional processes of production and destruction of alignment and intramultiplet mixing of the excited ion states.

The internal consistency of this experimental–theoretical approach is supported by the good agreement (to within 20%) in the values of the ion drift velocity obtained in this way from independent experimental data for two different excited fine-structure multiplets of Ar II ions, despite the great (more than a factor of ten) difference in the size of their splitting and the dispersion-polarization constants of ion interactions in these excited doublet states with neutral atoms of the ambient gaseous medium. This circumstance, as well as the internal consistency of the theory of anisotropic collisional relaxation developed here, encourages us to believe that this experimental–theoretical spectropolarimetric technique is widely applicable to the study of anisotropic collisional relaxation processes and the drift self-alignment of ion angular momenta resulting from them in various plasmas.

We touch briefly on the extent to which these results depend on the choice of the drift ion distribution function $f_A(\mathbf{v}_A)$ being a Maxwellian with an imposed drift, Eq. (21). In Sec. 4 we have seen that collisional self-alignment of drifting ions is determined by only one dynamic multipole moment of the relative velocity distribution function (22), its quadrupole moment $S_2(\lambda)$, which depends on the anisotropy parameter λ of Eq. (24). Our methodology is based on measurements of the polarization signal from the self-alignment of the excited ions. In consequence of this, different distributions of the relative ion-atom velocity distribution having the same dynamic quadrupole moment $S_2(\lambda)$ are essentially equivalent. On the other hand, for a given ion drift velocity $\mathbf{v}_0$ (i.e., the average value of the ordered velocity vector) the anisotropy parameter λ and along with it the dynamical quadrupole moment $S_2(\lambda)$ of the distribution function of the relative velocities depends rather weakly on the specific choice of the ion velocity distribution function $f_A(\mathbf{v}_A)$. In fact, the "opposite" of a Maxwellian with an imposed drift

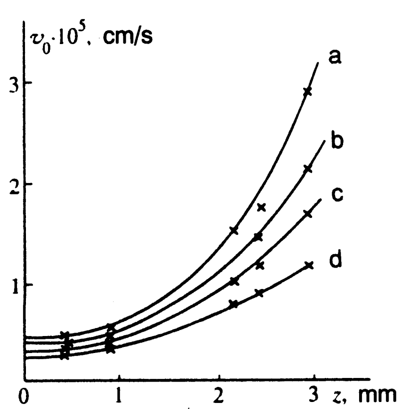

FIG. 8. Argon ion drift velocity as a function of the distance z to the axis of a cylindrical hollow cathode of diameter 10 mm, measured for the discharge current of 20 mA at various argon pressures (p_{Ar}, in torr): a) 0.12; b) 0.16; c) 0.20; d) 0.30.

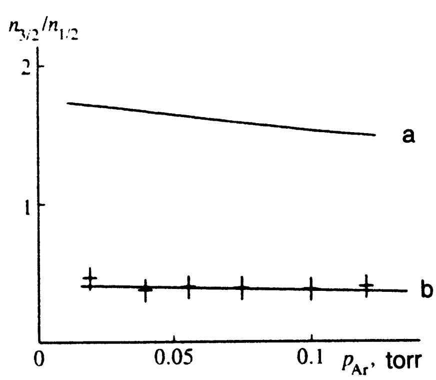

FIG. 9. Deviation of the population ratio $n_{3/2}/n_{1/2}$ of the $J=3/2$ and 1/2 levels of the Ar II $4p\ {}^2P^0_{1/2,3/2}$ ion doublet from its equilibrium value in the range of argon pressures studied: a) equilibrium value of the population ratio $(n_{3/2}/n_{1/2})=2y$, calculated from Eqs. (32) and (33) using the values of the anisotropy parameter λ measured experimentally; b) population ratio $n_{3/2}/n_{1/2}$ experimentally measured from the intensities of the Ar II ion doublet spectral components $4p\ {}^2P^0_{1/2,3/2}-4s\ {}^2P_{3/2}$.

as in Eq. (21) is the monokinetic ion drift velocity distribution

$$f_A(\mathbf{v}_A)=\delta(\mathbf{v}_A-\mathbf{v}_0). \tag{41}$$

Using the Maxwellian velocity distribution $f_B(\mathbf{v}_B)$ for the neutral atoms, Eq. (20), we find that this monokinetic ion velocity distribution (41) corresponds to the following relative velocity distribution:

$$f(\mathbf{v})=(\beta/\pi)^{3/2}\exp\{-\beta(v^2+2vv_0\cos\theta+v_0^2)\}. \tag{42}$$

It differs from the relative velocity distribution (22) by having the parameter β given by (20) instead of γ given by Eq. (23). The values of the anisotropy parameter λ (24) for the distributions (41) and (22) are related by

$$(\beta/\gamma)^{1/2}=[(M_A+M_B)/M_A]^{1/2}. \tag{43}$$

For atoms and ions with the same mass $M_A=M_B$, this ratio is equal to 1.41. Thus, even for the two different ion distribution functions (22) and (41), which are completely different, the anisotropy parameters differ by only 40%. Thus, there is reason to believe that the error in determining the parameter λ given by Eq. (24) and the ion drift velocity v_0 associated with it resulting from the imprecision in the model velocity distribution function (21) of the drifting ions is no more than 10–20%.

As more detailed experimental measurements are made, the theory can be refined by using more detailed drift ion velocity distribution functions[68–71] and by going from the dispersion-polarization model to a more accurate description of the interaction between the colliding particles.

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Translated by David L. Book