Velocity distribution relaxation due to inelastic collisions

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A new mechanism is proposed which leads to efficient change in the velocity of an excited atom, namely, inelastic collisions. Along with the familiar mechanisms (resonance exchange by excitation, radiation capture, and collisions with a small impact parameter), the present mechanism also leads in a number of cases to a strong collision model. The effect of inelastic collisions on the velocity distribution of excited atoms is analyzed within the framework of the strong collision model. Stationary conditions, relaxation processes, and the spatially inhomogeneous problem are considered. It is shown that in a number of cases the mechanism can yield an appreciable contribution to the relaxation of the velocity distribution.

1. As is well known, the change in the velocity of atoms in collisions can exert an appreciable effect on the interaction of atoms with an electromagnetic field.[1] Numerous papers are now being devoted to this phenomenon, since it affects the spectral characteristics of lasers, the competition of waves in ring-shaped and linear gas lasers, the relation of the single-frequency and multiple-frequency powers, the width of the spectrum generated and so on. For description of the velocity change, a number of semiphenomenological models were proposed in[1-3], among which the strong-collision model has received special attention. According to this model, the probability of observing an atom at a velocity $\mathbf{v}$ after collision has the form of an equilibrium (Maxwellian) distribution $W(\mathbf{v})$, independently of the value of the velocity of the atom $\mathbf{v}'$ before the collision. In other words, the collision integral S_m in the kinetic equation for the distribution of atoms $\rho_m(\mathbf{v})$ on the level m is taken in the form

$$S_m=-\nu_m\rho_m(\mathbf{v})+\tilde{\nu}_m W(\mathbf{v})\langle\rho_m(\mathbf{v}')\rangle, \tag{1}$$

where ν_m and $\tilde{\nu}_m$ are known as the departure and arrival frequencies and $\langle\ldots\rangle$ denotes integration over $\mathbf{v}'$. The existence of strong collisions and their effect on the spectroscopic phenomena has been established reliably by experiment in many cases (see, for example,[4-7]). It has been shown that the model (1) satisfactorily describes resonant exchange of excitation,[8,9] dragging of resonance radiation,[4,10] and elastic collisions for the case of small impact parameters.[11] At the same time, the interpretation offered in[12] of the experimental data obtained in[6] for the $3s_2-2p_4$ transition of Ne gives, as it were, evidence of the existence of reasons other than those given above for the significant change in the velocity of the excited atoms.

In this paper, we wish to draw attention to a series of other processes which, in their effect on $\rho_m(\mathbf{v})$, reduce to the model (1). We are dealing with inelastic processes, which are usually described by only the first terms in (1). However, for each inelastic process that "withdraws" an atom from the level m, there exists an inverse process that "brings" an atom to the level m once again. If in this case the atom "arrives" with the Maxwellian distribution of $\mathbf{v}$, then S_m will also have the form (1). The problem is only whether the "arrival" frequency $\tilde{\nu}_m$ is large in comparison with the "departure" frequency ν_m, and under what conditions the "arrival" will have a Maxwellian distribution. It is obvious that $\tilde{\nu}_m\cong\nu_m$ if the state j to which the atom is transferred from the level m decays to the same level m chiefly because of collisions. The second part of the problem—the $\mathbf{v}$ distribution of the "arrival"—will be discussed below.

We note that the effect described of inelastic processes on the relaxation of the velocities is entirely analogous to the rotational-translational relaxation in molecular gases. However, in the case of molecular systems, the general considerations that have been set forth lose their cogency, inasmuch as translational relaxation takes place, as a rule, somewhat more rapidly than rotational relaxation, i.e., for relaxation of the velocity distribution, it is sufficient to take into account the elastic collisions and there is no necessity of bringing in the inelastic channels of velocity change. In the case of atomic systems, as will be shown below, the relaxation of the velocities through inelastic channels can play the decisive role in many cases of practical interest.

2. We consider the simplest case—arrival along a single inelastic channel under stationary conditions. The set of equations needed by us has the form

$$\Gamma_m\rho_m(\mathbf{v})=-\nu_{mj}\rho_m(\mathbf{v})+q_m(\mathbf{v})+\nu_{jm}W(\mathbf{v})\langle\rho_j\rangle, \tag{2}$$

$$\Gamma_j\rho_j(\mathbf{v})=-\nu_{jm}\rho_j(\mathbf{v})+q_j(\mathbf{v})+\nu_{mj}W(\mathbf{v})\langle\rho_m\rangle. \tag{3}$$

Here $q_m(\mathbf{v})$, $q_j(\mathbf{v})$ and Γ_m, Γ_j are the excitation rates and relaxation constants of the levels m, j, which do not include the frequencies ν_{mj} and ν_{jm} of the transitions $m\to j$ and $j\to m$. The quantities q_m, q_j and Γ_m, Γ_j can include, for example, spontaneous relaxation, elastic scattering, transitions along other inelastic channels and so on. The direct and inverse frequencies ν_{mj} and ν_{jm} are connected by the usual relations:

$$g_j\nu_{jm}=g_m\nu_{mj}\exp[-(E_m-E_j)/kT]. \tag{4}$$

We express $\langle\rho_j\rangle$ from (3) and substitute in (2); then

$$\begin{gathered}\Gamma_m\rho_m(\mathbf{v})=-\nu_{mj}\rho_m(\mathbf{v})+\tilde{\nu}_{mj}W(\mathbf{v})\langle\rho_m\rangle+\tilde{q}_m(\mathbf{v}),\\ \tilde{\nu}_{mj}=\nu_{mj}\frac{\nu_{jm}}{\Gamma_j+\nu_{jm}};\quad \tilde{q}_m(\mathbf{v})=q_m(\mathbf{v})+W(\mathbf{v})\frac{\nu_{jm}}{\Gamma_j+\nu_{jm}}\langle q_j\rangle.\end{gathered} \tag{5}$$

The first two terms in the right side of Eq. (5) correspond entirely to the strong-collision model (1). The arrival frequency $\tilde{\nu}_{mj}$ is always less than the departure frequency ν_{mj} because of the damping Γ_j of the state j, $\mathbf{v}$ along channels different from $j\to m$, but in the case $\nu_{jm}\gg\Gamma_j$ we have $\tilde{\nu}_{mj}\cong\nu_{mj}$. The physical content of the formula for $\tilde{\nu}_{mj}$ is obvious; after the inelastic pro-

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cess m → j, the atom returns in the subsequent collision acts with probability $\nu_{jm}/(\Gamma_j + \nu_{jm})$, and if the transition j → m serves as the dominant process of decay of the state j (i.e., if $\nu_{jm} \gg \Gamma_j$), then $\tilde{\nu}_{mj} \cong \nu_{mj}$. By itself, this exchange is well known, but one usually considers the total number of atoms on the level $\langle \rho_m \rangle$; here the inverse transitions are simply equivalent to a reduction in the rate of relaxation for $\langle \rho_m \rangle$:

$$(\Gamma_m + \nu_{mj} - \tilde{\nu}_{mj}) \langle \rho_m \rangle = \langle \tilde{q}_m \rangle. \quad (6)$$

It is important for us that the relaxation of the nonequilibrium velocity distribution to the equilibrium distribution W(**v**) takes place at a rate $\Gamma_m + \nu_{mj}$.

If there exist several inelastic channels with the subsequent return of the atoms (to the initial state m) with a Maxwellian distribution, then the collision integral keeps the structure (1), but ν_m, $\tilde{\nu}_m$ are given by the formulas

$$\nu_m = \sum_j \nu_{mj}, \qquad \tilde{\nu}_m = \sum_j \nu_{mj} \frac{\nu_{jm}}{\Gamma_j + \nu_{jm}}, \quad (7)$$

the physical meaning of which is clear from the discussion of Eqs. (5).

We now discuss the assumption of Maxwellian velocity distribution for the arrival case. For inelastic collisions of atoms with hot electrons of a gas-discharge plasma, a change in the velocity takes place, but it is so small that the strong-collision model could be assumed unconditionally ($\Delta v \sim \bar{v}_e m_e/m \sim 0.1 v$[13]). In the case of collisions with "heavy" particles (atoms of the gas itself or of a foreign gas; ions) and for the case $|E_m - E_j| \lesssim kT$, this assumption is evidently valid. In any case, if kT is several times $\Delta E = |E_m - E_j|$ and the inelastic transition m → j is also accompanied by significant angular scattering ($\theta \gtrsim 1$), then the strong-collision model turns out to be quite adequate for the real situation. This qualitative conclusion follows from the general properties of the kernel of the collision integral.[11]

The practically important process of quasiresonance excitation exchange between atoms of different types is described in the given scheme. In this case, by j we mean the state of an atom of a different type to which the excitation is transferred from the state m, and the collision frequencies ν_{jm}, ν_{mj} are determined by the concentrations of the "own" (N) and "foreign" (n) gases:

$$\nu_{mj} = \langle \sigma u \rangle n, \quad \nu_{jm} = \langle \sigma' u \rangle N. \quad (8)$$

In the given process, a completely different atom arrives from the ground state to replace an $\rho_m(\mathbf{v})$ atom and the assumption of the Maxwellian character of the distribution for arrival in the case $|E_m - E_j| < kT$ is completely reliable.

For direct transitions of the type A(m) + B → A(j) + B ± ΔE, the condition $\theta \gtrsim 1$ is satisfied if $\rho_0 \lesssim a$. Here ρ_0 is the "point" of intersection of the terms of the quasimolecule A + B corresponding to the initial and final states of the colliding particles,[14] and a is the radius of "nonpenetration." For example, for the reaction $Ne(3s_2) + He \rightarrow Ne(3s_3) + He + \Delta E$, we have $\sigma_0 \equiv \sigma(3s_2 \rightarrow 3s_3) \lesssim 1.3 \times 10^{-15}$ cm^2,[15] whence $\rho_0 \approx \sqrt{\sigma_0/\pi} \lesssim 2.0$ Å; from the data on diffusion of Ne in He ($\sigma_d \approx \pi a^2 \cong 2.5 \times 10^{-15}$ cm^2[16]) we obtain $a \approx (\sigma_d/\pi)^{1/2} \cong 2.8$ Å, so that $\rho_0 \lesssim a$. In this case, the angular scattering will be significant ($\theta \sim \pi$), i.e., the strong-collision model will describe satisfactorily the change in the velocity in this process.

3. The case considered above illustrates graphically the essence of the situation and the conditions under which it is necessary to take into account the relaxation of the velocities in inelastic channels. We now turn to the more general problem: the conditions are not stationary and are not spatially homogeneous, and the changes in the velocity are not necessarily described by the strong-collision model. In the case of a single inelastic channel we have, in place of (2), (3):

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\nabla + \Gamma_m\right) \rho_m(\mathbf{r}, \mathbf{v}, t) - S_m^{(e)} = q_m(\mathbf{r}, \mathbf{v}, t) + \int B_{mj}(\mathbf{v}, \mathbf{u}) \rho_j(\mathbf{r}, \mathbf{u}, t)\, d\mathbf{u}, \quad (9)$$

$$\left(\frac{\partial}{\partial t} + \mathbf{u}\nabla + \Gamma_j\right) \rho_j(\mathbf{r}, \mathbf{u}, t) - S_j^{(e)} = q_j(\mathbf{r}, \mathbf{u}, t) + \int B_{jm}(\mathbf{u}, \mathbf{v}) \rho_m(\mathbf{r}, \mathbf{v}, t)\, d\mathbf{v},$$

where $S_m^{(e)}$, $S_j^{(e)}$ are the elastic-collision integrals. Let $F_j(\mathbf{r}, \mathbf{u}, t; \mathbf{r}', \mathbf{u}', t')$ be the Green's function of the left side of the second of Eqs. (9); with the help of F_j, the first equation can be written down formally in the form

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\nabla + \Gamma_m\right) \rho_m(\mathbf{r}, \mathbf{v}, t) = \tilde{q}_m(\mathbf{r}, \mathbf{v}, t) + S_m^{(e)} + S_{jm}^{(i)}, \quad (10)$$

$$S_{jm}^{(i)} = -\nu_{mj} \rho_m(\mathbf{r}, \mathbf{v}, t) + \int A_{jm}^{(i)}(\mathbf{r}, \mathbf{v}, t | \mathbf{r}', \mathbf{v}', t') \rho_m(\mathbf{r}', \mathbf{v}', t')\, d\mathbf{r}'\, d\mathbf{v}'\, dt',$$

$$A_{jm}^{(i)}(\mathbf{r}, \mathbf{v}, t; \mathbf{r}', \mathbf{v}', t') = \int B_{mj}(\mathbf{v}, \mathbf{u}) F_j(\mathbf{r}, \mathbf{u}, t; \mathbf{r}', \mathbf{u}', t') B_{jm}(\mathbf{u}', \mathbf{v}')\, d\mathbf{u}\, d\mathbf{u}',$$

$$\tilde{q}_m(\mathbf{r}, \mathbf{v}, t) = q_m(\mathbf{r}, \mathbf{v}, t) + \int B_{mj}(\mathbf{v}, \mathbf{u}) F_j(\mathbf{r}, \mathbf{u}, t; \mathbf{r}', \mathbf{u}', t') q_j(\mathbf{r}', \mathbf{u}', t')\, d\mathbf{u}\, d\mathbf{r}'\, d\mathbf{u}'\, dt'.$$

Thus, in this general case too, successive acts of inelastic transitions m → j, j → m lead to an "arrival term" in the collision integral $S_{jm}^{(i)}$; its kernel $A_{jm}^{(i)}$ is determined by the changes in the velocity in the transitions m → j, j → m (the factors $B_{mj}(\mathbf{v}, \mathbf{u})$ and $B_{jm}(\mathbf{u}, \mathbf{v}')$) and, in addition, by the elastic collisions experienced by the atom in state j (the factor $F_j(\mathbf{r}, \mathbf{u}, t; \mathbf{r}', \mathbf{u}', t')$). A singular feature of the kernel $A_{jm}^{(i)}$ that distinguishes its structure from the kernel of the elastic-collision integral is its dependence on **r**, t; **r**′, t′, i.e., the space-time nonlocalness of the "equivalent elastic collision" (in spite of the impact approximation). Physically, this difference is quite natural. In the case of elastic processes, the change in the velocity takes place within the time of the collision act $\tau_c \sim 10^{-12}$–10^{-13} sec, and it is assumed in the impact approximation that the conditions are constant in the time τ_c and over the interaction radius $\rho \sim \bar{v}\tau_c$; as a consequence of this, we have the space-time localness of the kernel of the elastic-collision integral $S_m^{(e)}$. For relaxation of the velocity over the inelastic channel, at least two successive collision acts separated in time and space, respectively, by the free path time and length: $\nu_m^{-1} \gg \tau_c$, $l \sim \bar{v}/\nu_m \gg \rho$ must occur. The conditions can change over these intervals, whence the above singularity of the kernels $A_{jm}^{(i)}$.

We recall that in the case of dragging of resonance radiation, two successive inelastic acts also take place—the radiation of a photon and its absorption by another atom. However, thanks to the large velocity of light, these acts (under laboratory conditions) are separated by a very small time interval and the collision integral is usually taken to be local in time. As a rule, it is necessary to take the spatial inhomogeneity into account.

4. We now analyze in more detail the features of the evolution of the velocity distribution under spatially

homogeneous conditions and for the case of instant "turning on" of the excitation:

$$q_m(\mathbf{r},\mathbf{v},t)=\begin{cases} q_m(\mathbf{v}), & t>0,\\ 0, & t<0.\end{cases} \tag{11}$$

The function $q_m(\mathbf{v})$ is not necessarily Maxwellian; it can be arbitrarily nonequilibrium. To simplify the calculations, we assume the strong-collision model. Under the conditions specified, it is more convenient to start not from Eq. (10), but directly from the set of equations (9), which we write down in the following form:

$$\left(\frac{\partial}{\partial t}+\Gamma_m+\nu_{mj}\right)\rho_m(\mathbf{v},t)=\nu_{jm}W(\mathbf{v})N_j(t)+q_m(\mathbf{v},t), \tag{12}$$

$$\left(\frac{d}{dt}+\Gamma_j+\nu_{jm}\right)N_j(t)=\nu_{mj}N_m(t),\quad \left(\frac{d}{dt}+\Gamma_m+\nu_{mj}\right)N_m=\nu_{jm}N_j+Q_m,$$
$$N_m(t)=\langle\rho_m(\mathbf{v},t)\rangle,\ N_j(t)=\langle\rho_j(\mathbf{v},t)\rangle,\ Q_m(t)=\langle q_m(\mathbf{v},t)\rangle. \tag{13}$$

The analogous set of equations in the case of the "elastic variant" of the strong-collision model has the form

$$\left(\frac{\partial}{\partial t}+\Gamma_m+\nu_m\right)\rho_m(\mathbf{v},t)=\bar{\nu}_mW(\mathbf{v})N_m(t)+q_m(\mathbf{v},t), \tag{14}$$
$$\left(\frac{d}{dt}+\tilde{\Gamma}_m\right)N_m(t)=Q_m(t),\quad \tilde{\Gamma}_m=\Gamma_m+\nu_m-\bar{\nu}_m\equiv\tau^{-1},$$

whence it is easy to obtain the result

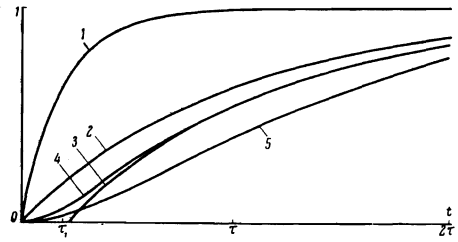
$$N_m(t)=Q_m\tau[1-\exp(-t/\tau)],$$
$$\rho_m(\mathbf{v},t)=q_m(\mathbf{v})\tau_1[1-e^{-t/\tau_1}]+Q_mW(\mathbf{v})\tau_2\left[1-\frac{\tau}{\tau_2}e^{-t/\tau}+\frac{\tau_1}{\tau_2}e^{-t/\tau_1}\right], \tag{15}$$
$$\tau_1=(\Gamma_m+\nu_m)^{-1},\quad \tau_2=\bar{\nu}_m/\tilde{\Gamma}_m(\Gamma_m+\nu_m)=\bar{\nu}_m\tau\tau_1.$$

It is seen from (15) that the stationary distribution over $\mathbf{v}$ contains the terms proportional to $q_m(\mathbf{v})$ and $W(\mathbf{v})$. The first part is established (curve 1 in the figure) in the time τ_1, which serves as the "accumulation time" for this part (the factor τ_1 for $q_m(\mathbf{v})$). The equilibrium part is "accumulated" in a time $\tau_2=\tau-\tau_1$, and is established, roughly speaking, in a time $\tau+\tau_1$: for small $t\ll\tau_1$, the equilibrium part in $\rho_m(\mathbf{v},t)$ increases $\sim t^2$, and then (in the case of practical interest, $\tau\gg\tau_1$) approaches a stationary value according to an exponential law, shifted by the interval τ_1 (curves 3, 4 in the figure). The above features of the evolution of $\rho_m(\mathbf{v},t)$ are quite understandable: the equilibrium part arises from the nonequilibrium part ($\sim q_m(\mathbf{v})$, for example, the Bennett "peak") over the entire lifetime τ of the atom on the level, but is shifted in time by an amount τ_1 relative to the growth curve of the total number $N_m(t)$ of particles (curve 2). This evolution pattern, which is characteristic for $\tau_1\ll\tau$, is preserved qualitatively until $\tau_1=\tau/2$ (to which corresponds $\nu_m=\bar{\nu}_m=\Gamma_m$), when the equilibrium part increases according to the law $[1-\exp(-t/\tau)]^2$ (curve 5 in the figure).

We now turn our attention to the strong-collision model in the "inelastic" variant (Eqs. (12), (13)). As in (14), $\rho_m(\mathbf{v},t)$ is found by a quadrature:

$$\langle\rho_m(\mathbf{v},t)=q_m(\mathbf{v})\tau_1[1-e^{-t/\tau_1}]+\nu_{jm}W(\mathbf{v})\int_0^t N_j(t')e^{-(t-t')/\tau_1}dt',$$
$$\tau_1=(\Gamma_m+\nu_{mj})^{-1}, \tag{16}$$

while the nonequilibrium part, as was to be expected, is identical with that in (15). The equilibrium part is different as a consequence of the fact that is arises from the transition $j\to m$, and the integral in (16) contains $N_j(t')$, which does not change according to the simple exponential law (like $N_m(t)$ in (14), (15)): the "double-act" nature of the strong collisions leads to a set of equations



Curves illustrating the evolution of the velocity distribution in the strong collision model after instantaneous starting (at t = 0) of the excitation: curve 1–function $1-e^{-t/\tau_1}$; 2–function $1-e^{-t/\tau}$; 3–function $1-\tau e^{-t/\tau_1}/\tau_2$; 5–function $(1-e^{-t/\tau})^2$; $\tau=5\tau_1$ for all curves.

of second order and the development follows the "double exponential" law:

$$N_j(t)=Q_m\tau'\frac{\nu_{mj}}{\Gamma_j+\nu_{jm}}\left[1-\frac{\lambda_1}{\lambda_1-\lambda_2}e^{-\lambda_2 t}+\frac{\lambda_2}{\lambda_1-\lambda_2}e^{-\lambda_1 t}\right],$$
$$N_m(t)=Q_m\tau'\left[1-\frac{\lambda_1-\tilde{\Gamma}_m'}{\lambda_1-\lambda_2}e^{-\lambda_2 t}+\frac{\lambda_2-\tilde{\Gamma}_m'}{\lambda_1-\lambda_2}e^{-\lambda_1 t}\right],$$
$$\tilde{\Gamma}_m'\equiv\tau'^{-1}=\Gamma_m+\nu_m-\bar{\nu}_m'=\Gamma_m+\frac{\nu_{mj}\Gamma_j}{\Gamma_j+\nu_{jm}}=\frac{\lambda_1\lambda_2}{\lambda_1\lambda_2-\Gamma_m-\nu_{mj}}, \tag{17}$$
$$\lambda_{1,2}=\frac{1}{2}\{(\Gamma_m+\nu_{jm}+\Gamma_j+\nu_{mj})\pm$$
$$\pm[(\Gamma_m+\nu_{jm}+\Gamma_j+\nu_{mj})^2-4(\Gamma_j\nu_{mj}+\Gamma_m\nu_{jm}+\Gamma_j\Gamma_m)]^{1/2}\}.$$

Accordingly, $\rho_m(\mathbf{v},t)$ evolves according to the more complicated law

$$\rho_m(\mathbf{v},t)=q_m(\mathbf{v})\tau_1[1-e^{-t/\tau_1}]$$
$$+Q_m\tau_2'W(\mathbf{v})\left\{1-\frac{1}{\tau_2'}\left[\frac{\lambda_1-\Gamma_m-\nu_{mj}}{\lambda_2(\lambda_1-\lambda_2)}e^{-\lambda_2 t}+\frac{\Gamma_m+\nu_{mj}-\lambda_2}{\lambda_1(\lambda_1-\lambda_2)}e^{-\lambda_1 t}\right]\right.$$
$$\left.+\frac{\tau_1}{\tau_2'}e^{-t/\tau_1}\right\},\quad \tau_2'=\tau'-\tau_1,\quad \tau_1=(\Gamma_m+\nu_{mj})^{-1}. \tag{18}$$

The term $(\tau_1/\tau_2')e^{-t/\tau_1}$ in the curly brackets has a complete analog in (15). But the expression in the square brackets in (18) replaces the simple term $(\tau/\tau_2)e^{-t/\tau}$ from (15). The physical meaning of the damping constants λ_1, λ_2 will be different depending on the relation between Γ_m, Γ_j, ν_{jm} and ν_{mj}. If $\Gamma_m=\Gamma_j=\Gamma$, then

$$\lambda_1=\Gamma+\nu_{mj}+\nu_{jm},\quad \lambda_2=\Gamma, \tag{19}$$

i.e., the smaller of the roots $\lambda_{1,2}$ characterizes the decay time of the levels, and the larger the transitions between then. Here the evolution of the equilibrium part in $\rho_m(\mathbf{v},t)$ is qualitatively the same as in the "elastic" case (see the figure); the only difference is that for small t, the increase takes place according to the t^3 (and not t^2) law and the exponential part of the development is shifted not by τ_1 but approximately by τ_1+1/Γ.

Of practical interest is the case $\Gamma_m+\nu_{mj}\gg\Gamma_j+\nu_{jm}$ (it is realized, for example, in a helium-neon mixture in the case of the exchange $He(2^1S_0)-Ne(3s_2)$). Here we have

$$\lambda_1=\Gamma_m+\nu_{mj}+\frac{\nu_{jm}\nu_{mj}}{\Gamma_m+\nu_{mj}},\quad \lambda_2=\Gamma_j+\nu_{jm}\frac{\Gamma_m}{\Gamma_m+\nu_{mj}}\equiv\tilde{\Gamma}_j, \tag{20}$$

i.e., λ_2 characterizes the rate of decay of the longer-lived state j with account of the decay from the state m (cf. with $\tilde{\Gamma}_m'$ in (17)), and λ_1 is practically identical with τ_1^{-1}. Even in this case, consequently, the general character of the evolution of $\rho_m(\mathbf{v},t)$ is conveyed by the curves shown in the figure, but λ_2 characterizes the decay of the state j and not m.

To illustrate the evolution in the inelastic model, the case $\Gamma_j = \Gamma_m = \nu_{mj} = \nu_{jm} \equiv \Gamma$ is instructive; here the curly bracket in (18) goes over into $[1-\exp(-\Gamma t)]^3$, in contrast with $[1-\exp(-\Gamma t)]^2$ for (15).

5. It should be clear from the previous sections that a long lifetime Γ_j^{-1} of the state that is the partner of (j) adds to the effectiveness of the "strong collisions along the inelastic channel." For small Γ_j, the spatial inhomogeneity of the conditions (diffusion on the wall, for example) can turn out to be significant, especially if we are dealing with a metastable state. We now consider this problem in more detail.

From the analogy between nonstationarity and the spatial inhomogeneity, it is easy to perceive that the Knudsen case corresponds to the region of small t ($N_j \sim t^2$)—the length of the path is much greater than the minimal dimensions of the vessel. This case is comparatively complicated for analysis (see, for example, the solution of a similar problem in[17]) and does not appear frequently in spectroscopic practice at the present time. We shall therefore not consider it here. The diffusion approximation evidently corresponds to the region of exponential growth in the figure—the free path length is much less than the scale of the inhomogeneity. In this case, consequently, one can expect that account of the spatial inhomogeneity will reduce to redetermination of the relaxation constants.

We now consider the very simple but most important case in which it is necessary to consider the spatial diffusion only of the state j (the metastable He (2^1S_0) in the He—Ne mixture). With the help of the standard method we can obtain the following expression for $\rho_m(\mathbf{r}, \mathbf{v})$ (a cylindrical vessel of radius R, $\mathbf{r}$ is the transverse coordinate, zero boundary conditions, stationary problem):

$$\rho_m(\mathbf{r},\mathbf{v}) = q_m(\mathbf{r},\mathbf{v})\tau_1 + W(\mathbf{v})\sum_{l=1}^{\infty} J_0\left(\mu_l \frac{r}{R}\right)\left\{Q_m{}^l\tau_2{}^l + \frac{\nu_{jm}}{\Gamma_j{}^l+\nu_{jm}} Q_j{}^l\tau^l\right\}$$

$$\Gamma_j{}^l = \Gamma_j + D\frac{\mu_l{}^2}{R^2}, \quad \frac{1}{\tau^l} = \Gamma_m + \nu_{mj}\frac{\Gamma_j{}^l}{\Gamma_j{}^l+\nu_{jm}}, \tag{21}$$

$$\tau_2{}^l = \tau^l - \tau_1, \quad \tau_1 = (\Gamma_m + \nu_{mj})^{-1}.$$

Here J_0 is a Bessel function of the first kind of order zero, μ_l is its l-th root, Q_m^l, Q_j^l are coefficients of the expansion of $Q_m(\mathbf{r})$, $Q_j(\mathbf{r})$ in the Bessel functions $J_0(\mu_l r/R)$. It is seen from a comparison of (21) with (17), (18) that each Bessel component of $\rho_m(\mathbf{r}, \mathbf{v})$ has its own effective times τ_2^l, τ^l, the structure and meaning of which are entirely analogous to what we have had earlier.

Generalization to the case of many channels is obvious—the effective frequencies of arrival and departure are determined by the rule (7) with the replacement of Γ_j by Γ_j^l. In the case of a single inelastic channel and nonstationary conditions, the evolution of each Bessel component in $\rho_m(\mathbf{r}, \mathbf{v})$ will be determined by formulas of the type (18) and in the expressions for $\lambda_{1,2}^l$, which are similar to (17), it is necessary to substitute Γ_j^l in place of Γ_j.

6. As an example, we consider the $3s_2$ state of neon, for which we can point out five independent channels, which lead to the strong collision model:

$$\nu_m = \nu_0 + A_{m0} + \nu_1 + \nu_2 + \nu_3,$$

$$\tilde{\nu}_m = \nu_0 + A_{m0} + \nu_1\frac{\nu_1'}{\Gamma_1+\nu_1'} + \nu_2\frac{\nu_2'}{\Gamma_2+\nu_2'} + \nu_3\frac{\nu_3'}{\Gamma_2+\nu_3'}. \tag{22}$$

The terms ν_0, A_{m0} describe the resonance exchange of excitation in the collisions of Ne($3s_2$) and Ne(1p) and dragging of resonance radiation. The frequency ν_1 corresponds to the exchange Ne($3s_2$) $\rightleftarrows$ He(2^1S_0), and the frequencies ν_2, ν_3 to inelastic transitions of neon atoms Ne($3s_2$) $\rightleftarrows$ Ne($3s_3$) for the case of collisions with helium and neon, respectively; ν_j' are the frequencies of the inverse transitions; Γ_1, Γ_2 are the probabilities of damping of the states He(2^1S_0) and Ne($3s_3$). According to literature data (here and elsewhere, p_{Ne} and p_{He} are in Torr), $A_{m0} = 2.5\times10^7\ \text{sec}^{-1}$[18]; $\nu_0 = 1.68\,\pi\lambda^3 A_{m0}N = 0.40\times10^7\, p_{Ne}\,\text{sec}^{-1}$[18,19].

At the present time, an estimate can be made very simply of the role of inelastic collisions with helium. In accord with[20], $\nu_1' = 0.2\times10^7\, p_{Ne}\,\text{sec}^{-1}$. Then, using (4), we get, at T = 300°K, $\nu_1 = 0.4\times10^7\, p_{He}\,\text{sec}^{-1}$. Since $\Gamma_1 \ll \nu_1'$ under typical laser conditions, we have $\tilde{\nu}_1 \approx \nu_1 \approx 0.4\times10^7\, p_{He}\,\text{sec}^{-1}$. Consequently, the contribution to "strong collisions" due to exchange of excitation with helium significantly exceeds ν_0 (since $p_{He} \gg p_{Ne}$) and at high helium pressures approaches the maximum contribution A_{m0} that can be contributed by dragging of resonance radiation (the case of complete dragging in a spatially homogeneous field).

Literature data[15] indicate that ν_2 is close to ν_1 in magnitude. Setting $\nu_2 \approx 0.4\times10^7\, p_{He}\,\text{sec}^{-1}$, we have $\nu_2' \approx 1.2\times10^7\, p_{He}\,\text{sec}^{-1}$; inasmuch as $\Gamma_2 \cong 6\times10^6\ \text{sec}^{-1}$,[21], at a pressure $p_{He} \sim 2$ Torr, we already get $\nu_2' > \Gamma_2$ and $\tilde{\nu}_2 \sim \nu_2$.

In addition to the three inelastic channels shown in (22), other such channels can be considered.

Thus, for high mixture pressures, processes in collisions with neon and helium can play a significant (and under certain conditions decisive) role in the relaxation of the velocity distribution according to the "strong collision" model, and should be taken into account along with the processes which were considered earlier. Here there is an opportunity to gain understanding of the reasons for certain phenomena that were not previously clear. As an example, we point to the work of Smith,[12] who reached the conclusion that the experimental data of[6] for the transition $3s_2 - 2p_4$ of neon in a mixture with helium cannot be explained solely by radiation dragging from the level $3s_2$, and that one must assume the existence of other "strong collision" mechanisms, the frequency of which increases with increasing p_{He}. In our opinion, such a mechanism may be the redistribution of the velocities as a result of inelastic collisions, as was pointed out above.

It is interesting to note that the process of transfer of the excitation from neon to helium and back does not fit completely into the framework of the usual "strong-collision" model. The fact is that the transfer of energy from the metastable 2^1S_0 atoms of helium to the neon atoms takes place with an energy deficit, as a consequence of which the excited neon atoms are "cooled." The indicated effect can explain the incomplete agreement of the experimental data for the mixture of neon with helium (pointed out in[6]) with the results of a calculation according to the strong collision model: the edge of the Doppler background turned out to be lower

than follows from calculation. At the same time, measurements with pure neon[6] gave significantly better agreement.

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