The diffusion approximation in the problem of migration of particles in a gas

S. G. Rautian

Institute of Automation and Electrometry, Siberian Branch of the Academy of Sciences of the USSR, Novosibirsk

(Submitted 21 May 1991)
Usp. Fiz. Nauk **161**, 151–170 (November 1991)

The problem of migration of particles in phase space constitutes a classical problem. Its correct, nonphenomenological formulation for gases that are not too dense is contained essentially in the work of Boltzmann,[1] where he formulated his famous kinetic equation

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla\right)\rho(\mathbf{r}\mathbf{v}t)=S \tag{1}$$

for the distribution function $\rho(\mathbf{r}\mathbf{v}t)$ ($\mathbf{r}$, $\mathbf{v}$, and t are the coordinate, the velocity, and the time). Under certain conditions the collision integral S can be expressed in terms of derivatives of ρ; this approximation, which is called the diffusion approximation, is widely applied in the most varied fields of physics. It has been discussed in detail in the literature, and has been included in textbooks.[2–8] In the recent past, however, the situation has changed so that the given, seemingly settled problem has required a certain modernization and rethinking.

In the second half of the sixties a new field of application of physical kinetics arose–laser physics, nonlinear laser spectroscopy, which is characterized by conditions quite unusual from the standpoint of traditional kinetics. The point is that the fundamental agent of laser spectroscopy–a plane monochromatic wave reacting in resonance with a gas, creates a sharply defined structure in the velocity distribution for the atoms existing in optically combining energy levels. The width of this structure can be considerably smaller than the general (Maxwellian) width of distribution (what we have said pertains to one Cartesian variable—the projection of $\mathbf{v}$ on the wave vector $\mathbf{k}$ of the wave). In contrast to this, the traditional physical concepts associated with the diffusion approximation started explicitly or implicitly with a pattern of relatively smooth nonequilibrium components of the velocity distribution.

A second new and important circumstance involves the large spectral power densities of laser radiation and the creation of considerable concentrations of excited states. In particular, very essential factors in the migration of atoms in excited states are their short duration, the features of the differential cross section in the region of small scattering angles, and other circumstances. As a result, in describing the migration of excited atoms, models prove to be of interest that strongly differ from those appropriate in the ground state.

Special approaches in the theory and in the overall physical picture of diffusion processes have been created by the particularly optical problem of the random walk of the dipole moment (or the polarization, or the coherence), which organically enter into the problem of Doppler broadening of spectral lines.

The study of the mentioned problems and a number of others in nonlinear spectroscopy has led to a revision of the conditions of applicability of the diffusion approximation for describing migration in $\mathbf{v}$-space. It has undergone some development, and the accent has been shifted in the common views. Besides, this is what usually happens in attempts to apply old, developed methods to new problems. Also the general tendency of recent decades toward weakening the role of phenomenology and increasing the relative weight of microscopical theories has had a substantial significance.

First of all, let us examine the traditional problem of migration of a small admixture of heavy particles (mass m) in a buffer gas of light particles (mass m_b, $m\gg m_b$). Bearing in mind the very simple case of structureless particles, we shall write the collision integral in the form

$$S=-\nu\rho(\mathbf{r}\mathbf{v}t)+\int A(\mathbf{v}|\mathbf{v}_1)\rho(\mathbf{r}\mathbf{v}_1t)d\mathbf{v}_1, \tag{2}$$

$$\nu=\int A(\mathbf{v}_1|\mathbf{v})d\mathbf{v}_1. \tag{3}$$

Here the kernel $A(\mathbf{v}|\mathbf{v}_1)$ of the collision integral is the number of $\mathbf{v}_1\to\mathbf{v}$ transitions per second owing to collisions with the buffer gas. The exit rate ν evidently gives the number of transitions per second from the point $\mathbf{v}$ into the entire velocity space.[1)]

Usually one argues as follows (see, e.g., Ref. 6, Sec. 21). It is actually evident that, when $m\gg m_b$, the variation of the velocity of the heavy particle owing to collisions is relatively small. In other words, the kernel $A(\mathbf{v}|\mathbf{v}_1)$ will be a sharply varying function of the velocity difference $\mathbf{v}_1-\mathbf{v}=\vec{\zeta}$, and a far smoother function of $\mathbf{v}_1$:

$$A(\mathbf{v}|\mathbf{v}_1)=a(\mathbf{v}_1,\vec{\zeta}),\quad \vec{\zeta}=\mathbf{v}_1-\mathbf{v}. \tag{4}$$

Therefore it is natural to use the expansion

$$A(\mathbf{v}|\mathbf{v}_1)\rho(\mathbf{r}\mathbf{v}_1t)=a(\mathbf{v}+\vec{\zeta},\vec{\zeta})\rho(\mathbf{r}\mathbf{v}+\vec{\zeta}t)=a(\mathbf{v},\vec{\zeta})\rho(\mathbf{r}\mathbf{v}t)$$
$$+\zeta_\alpha\frac{\partial}{\partial v_\alpha}\left(a(\mathbf{v},\vec{\zeta})\rho(\mathbf{r}\mathbf{v}t)\right)+\frac{1}{2}\zeta_\alpha\zeta_\beta\frac{\partial^2}{\partial v_\alpha\partial v_\beta}\left(a(\mathbf{v},\vec{\zeta})\rho(\mathbf{r}\mathbf{v}t)\right) \tag{5}$$

and write the collision integral in the form

$$S=\frac{\partial}{\partial v_\alpha}\left(A_\alpha\rho+\frac{\partial}{\partial v_\beta}(B_{\alpha\beta}\rho)\right). \tag{6}$$

Here the components of the vector A_α and the tensor $B_{\alpha\beta}$ are

$$A_\alpha=\int\zeta_\alpha a(\mathbf{v},\vec{\zeta})d\vec{\zeta},\quad B_{\alpha\beta}=\frac{1}{2}\int\zeta_\alpha\zeta_\beta a(\mathbf{v},\vec{\zeta})d\vec{\zeta}. \tag{7}$$

For an equilibrium distribution we have

$$\rho(\mathbf{r}\mathbf{v}t)=\rho_0\exp(-\mathbf{v}^2/\bar{v}^2),\quad \bar{v}^2=2T_b/m, \tag{8}$$

where T_b is the temperature of the buffer gas,[2)] and the collision integral must be zero. Consequently a relation must exist that connects the quantities A_α and $B_{\alpha\beta}$:

$$A_\alpha+\frac{\partial B_{\alpha\beta}}{\partial v_\beta}=\frac{2v_\beta}{\bar{v}^2}B_{\alpha\beta}. \tag{9}$$

Ultimately the kinetic equation acquires the form of the dif-

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fusion equation in v-space, which is called the Fokker–Planck equation:[6]

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla\right)\rho=\frac{\partial}{\partial v_\alpha}\left[B_{\alpha\beta}\left(\frac{2v_\beta}{\bar{v}^2}\rho+\frac{\partial\rho}{\partial v_\beta}\right)\right]. \tag{10}$$

The collision integral in Eq. (10) amounts to the divergence in v-space of a certain vector, which is naturally called the flux density of particles in velocity space due to collisions:

$$S=-\mathrm{div}_{\mathbf{v}}\mathbf{s},\quad s_\alpha=-\left(A_\alpha+\frac{\partial B_{\alpha\beta}}{\partial v_\beta}\right)\rho-B_{\alpha\beta}\frac{\partial\rho}{\partial v_\beta}. \tag{11}$$

Therefore the kinetic equation (10) has the form of the continuity equation in v-space:

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla\right)\rho+\mathrm{div}_{\mathbf{v}}\mathbf{s}=0, \tag{12}$$

"thus the conservation of the number of particles is automatically obeyed" (Ref. 6, Sec. 21).

Both the derivation and Eq. (10) itself leave the feeling of a certain dissatisfaction. First of all, by definition the set of quantities A_α and $B_{\alpha\beta}$ are the first and second moments of the kernel. Generally the moments of any function, in this case $a(\mathbf{v},\vec{\xi})$ serve as independent characteristics of it. Therefore the existence of a certain connection between A_α and $B_{\alpha\beta}$ seems unobvious and requires clarification. Clarification is all the more necessary in that under equilibrium conditions the collision integral in (2) is identically equal to zero for an arbitrary interaction of the colliding particles, for arbitrary values of any of their characteristics. In view of what we have said, the flux density in v-space due to collisions must automatically equal zero, and the additional condition (9), albeit ensuring this equality, is superfluous. We note, moreover, that it is not clear from the arguments that led to Eq. (10) to what is the small parameter equal that justifies the expansion (5). Finally, the expansion (5) itself, where the functions $a(\mathbf{v}_1,\vec{\xi})$ and $\rho(\mathbf{r}\mathbf{v}_1 t)$ figure "with equal rights" in the product $a(\mathbf{v}_1,\vec{\xi})\rho(\mathbf{r}\mathbf{v}_1 t)$, will be natural if the widths of the cofactors $a(\mathbf{v}_1,\vec{\xi})$ and $\rho(\mathbf{r}\mathbf{v}_1 t)$ are of the same order of magnitude as functions of the velocity. Perhaps the situation is just so in the traditional problems. However, for problems of nonlinear spectroscopy the stated conditions are not at all typical.

The ideas and doubts that we have presented become quite graphic if one uses the vanishing of the collision integral for equilibrium conditions, not at the end, but at the very start of the discussions. Actually, let us substitute the expression (8) into the formulas (2) and (3) for the collision integral. We find from the condition $S=0$ the connection between $A(\mathbf{v}|\mathbf{v}_1)$ and $A(\mathbf{v}_1|\mathbf{v})$ (the principle of detailed balance):

$$A(\mathbf{v}|\mathbf{v}_1)\exp(-\mathbf{v}_1^2/\bar{v}^2)=A(\mathbf{v}_1|\mathbf{v})\exp(-\mathbf{v}^2/\bar{v}^2), \tag{13}$$

Thereupon we can write the collision integral thus:

$$S=\int a(\mathbf{v},\mathbf{v}-\mathbf{v}_1)\left\{\exp\left[(\mathbf{v}_1^2-\mathbf{v}^2)/\bar{v}^2\right]\rho(\mathbf{r}\mathbf{v}_1 t)-\rho(\mathbf{r}\mathbf{v}t)\right\}d\mathbf{v}_1. \tag{14}$$

Now we note that the "smooth" variation of the kernel $a(\mathbf{v},\mathbf{v}-\mathbf{v}_1)$ contains $\mathbf{v}$ rather than $\mathbf{v}_1$, and in the integrand of the entry term actually not the product $a(\mathbf{v}_1,\mathbf{v}_1-\mathbf{v})\rho(\mathbf{r}\mathbf{v}_1 t)$ will be expanded, as was the case before, but the combination $\exp(\mathbf{v}_1^2/\bar{v}^2)\rho(\mathbf{r}\mathbf{v}_1 t)$ of the distribution function and the standard coefficient, which does not depend on the features of interaction of the particles in the collisions. In other words, Eq. (14) in explicit form gives the "slow" coefficient of $\rho(\mathbf{r}\mathbf{v}_1 t)$, and its width proves to be equal to $\bar{v}$. The expansion of the product $\exp(\mathbf{v}_1^2/\bar{v}^2)\rho(\mathbf{r}\mathbf{v}_1 t)$ in powers of $(\mathbf{v}_1-\mathbf{v})_\alpha$ leads to the relationship

$$Se^{\mathbf{v}^2/\bar{v}^2}=-A_\alpha\frac{\partial}{\partial v_\alpha}\left(e^{\mathbf{v}^2/\bar{v}^2}\rho(\mathbf{r}\mathbf{v}t)\right)+B_{\alpha\beta}\frac{\partial^2}{\partial v_\alpha\partial v_\beta}\left(e^{\mathbf{v}^2/\bar{v}^2}(\mathbf{r}\mathbf{v}t)\right). \tag{15}$$

As before, A_α and $B_{\alpha\beta}$ are defined by the formulas of (7).

Under equilibrium conditions we have $\exp(\mathbf{v}^2/\bar{v}^2)\rho(\mathbf{r}\mathbf{v}t)=\mathrm{const}$, and according to Eq. (15) we have $S=0$ without additional conditions of the type of (9). The conclusion that there are no connections among the coefficients remains in force also when we retain the derivatives of $\rho(\mathbf{r}\mathbf{v}t)$ of all orders. We should have expected this result, since the coefficients of the expansion are the moments of different orders of the kernel, and they are independent characteristics of it. On the other hand, as we can easily convince ourselves, the condition for going from Eq. (15) to Eq. (6) is given by the same equality (9). In other words, within the framework of the sequence of arguments (13)–(15), the condition (9) ensures the representation of the collision integral in the form of the divergence in v-space of a certain vector, which is interpreted as the flux density of particles in v-space, rather than the equality $S=0$ under equilibrium conditions.

The conclusions of the theory should not depend on the sequence of using the arguments, and the contradiction that arises can be only of methodological character. The point is that the relationship (9) between A_α and $B_{\alpha\beta}$ is, of course, not valid in the general case, yet it proves to be approximately valid, within the accuracy of calculating S, which is given by the terms dropped in the expansion of $\rho(\mathbf{r}\mathbf{v}t)$.

Indeed, let us use the explicit expression for the kernel:

$$A(\mathbf{v}|\mathbf{v}_1)=2N_b\int\sigma(\mathbf{u}|\mathbf{u}_1)\delta(\mathbf{u}^2-\mathbf{u}_1^2)\delta\left[\mathbf{v}-\mathbf{v}_1-\frac{\mu}{m}(\mathbf{u}-\mathbf{u}_1)\right]\times W_b(\mathbf{v}_1-\mathbf{u}_1)d\mathbf{u}d\mathbf{u}_1; \tag{16}$$

Here $\sigma(\mathbf{u}|\mathbf{u}_1)$ is the differential scattering cross section, $\mathbf{u}$ and $\mathbf{u}_1$ are the relative velocities after and before collision, μ is the reduced mass, N_b is the concentration of buffer particles, and $W_b(\mathbf{v}_1-\mathbf{u}_1)$ is their velocity distribution, which we should assume to be the equilibrium distribution:

$$W_b(\mathbf{v}_{b1})=(\sqrt{\pi}\bar{v}_b)^{-3}\exp(-\mathbf{v}_{b1}^2/\bar{v}_b^2), \tag{17}$$

$$\mathbf{v}_{b1}=\mathbf{v}_1-\mathbf{u}_1,\quad \bar{v}_b^2=2T_b/m_b,\quad \mu=mm_b/(m+m_b).$$

The δ-function in Eq. (16) reflects the laws of conservation of energy and momentum. Thus the kernel $A(\mathbf{v}|\mathbf{v}_1)$ is proportional to the differential cross section (or the partial rate of collisions N_b and $\sigma(\mathbf{u}|\mathbf{u}_1)$) averaged over the velocities of the buffer particles (or over the relative velocities) with account taken of the conservation laws.

By using the reciprocity theorem and Eq. (17), one can show[6,9] that Eq. (16) implies the relationship (13) between the kernels $A(\mathbf{v}|\mathbf{v}_1)$ and $A(\mathbf{v}_1|\mathbf{v})$ that arose above in a purely phenomenological way.

If we assume that $\sigma(\mathbf{u}|\mathbf{u}_1)$ depends only on u and the angle θ between $\mathbf{u}$ and $\mathbf{u}_1$, we can arrive at the formulas

$$\mathbf{A} = \nu_1 \mathbf{v}, \tag{18}$$

$$\nu_1 = \frac{\mu}{mv} N_b \int \sigma^{(1)}(u) \cos\vartheta W_b(\mathbf{v}-\mathbf{u}) u^2 d\mathbf{u}, \tag{19}$$

$$SpB_{\alpha\beta} = B_\parallel + 2B_\perp = \left(\frac{\mu}{m}\right)^2 N_b \int \sigma^{(1)}(u) W_b(\mathbf{v}-\mathbf{u}) u^3 d\mathbf{u}, \tag{20}$$

$$B_\parallel - B_\perp = \left(\frac{\mu}{m}\right)^2 \frac{N_b}{2} \int \left(\sigma^{(1)}(u) - \frac{3}{4}\sigma^{(2)}(u)\right) \times (3\cos^2\vartheta - 1) W_b(\mathbf{v}-\mathbf{u}) u^3 d\mathbf{u}, \tag{21}$$

$$\sigma^{(l)}(u) = 2\pi \int_0^\pi (1 - \cos^l\theta)\sigma(\mathbf{u}|\mathbf{u}_1)\sin\theta d\theta, \quad \cos\theta = \mathbf{u}\mathbf{u}_1/u^2, \quad \cos\vartheta = \mathbf{v}\mathbf{u}/vu. \tag{22}$$

The proportionality coefficient ν_1 between the vector $\mathbf{A}$ and $\mathbf{v}$ is called the transport frequency. The diffusion tensor $B_{\alpha\beta}$ is axially symmetric (the symmetry axis is directed along $\mathbf{v}$); $B_\parallel$ and $B_\perp$ are the components of the tensor in the $\mathbf{v}$-system parallel and perpendicular to $\mathbf{v}$.

Direct calculation shows that the values of A_α and $B_{\alpha\beta}$ from Eqs. (18)–(21) do not satisfy the equality (9). However, if we adopt the inequalities

$$m_b/m \ll 1, \quad v^2/\bar{v}_b^2 = m_b v^2/m\bar{v}^2 \ll 1, \tag{23}$$

then the corresponding approximate values are given by the formulas

$$B_{\alpha\beta} = B\delta_{\alpha\beta}, \quad B = (1/2)\nu_1 \bar{v}^2, \quad \mathbf{A} = \nu_1 \mathbf{v},$$

$$\nu_1 = \frac{8}{3\sqrt{\pi}} \frac{m_b}{m} N_b \bar{v}_b \int_0^\infty \sigma^{(1)}(u) \exp\left(-\frac{u^2}{\bar{v}_b^2}\right) \frac{u^5 du}{\bar{v}_b^6} \tag{24}$$

and the condition (9) is satisfied. The relative corrections to the first approximation of (24) are equal in order of magnitude to m_b/m and $m_b v^2/m\bar{v}^2$, and one must not retain them, since the dropped terms containing the third and fourth derivatives of ρ are of the same order of smallness. Actually, simple estimates of the terms containing the nth-order derivatives of ρ are:

$$n=1: \ \frac{v}{\delta v}\nu_1\rho, \qquad n=2: \ \left(\frac{\bar{v}}{\delta v}\right)^2 \nu_1\rho,$$
$$n=3: \ \frac{v}{\partial v}\nu_1\rho\frac{m_b}{m}\left(\frac{\bar{v}}{\partial v}\right)^2, \qquad n=4: \ \left(\frac{\bar{v}}{\partial v}\right)^2 \nu_1\rho\frac{m_b}{m}\left(\frac{\bar{v}}{\delta v}\right)^2. \tag{25}$$

Here δv is the characteristic width of the nonequilibrium component of the function $\rho(\mathbf{r}vt)$. Evidently the expansion parameter of the collision integral is the quantity

$$\frac{m_b}{m}\left(\frac{\bar{v}}{\partial v}\right)^2. \tag{26}$$

This coincides with m_b/m if $\delta v \sim \bar{v}$. We see also from the estimates of (25) that the first term of the expansion can be either smaller ($v < \bar{v}^2/\delta v$) or larger ($v > \bar{v}^2/\delta v$) than the second term. Hence we must retain the terms with first and second derivatives of ρ.[3)] Yet the third- and fourth-order terms are small with respect to the parameter m_b/m, as are the corrections to the values of A_α and $B_{\alpha\beta}$ calculated by the approximate formulas of (24).

Thus the relationship (9) is only approximately valid when the transport rate and $B_\parallel \approx B_\perp \approx B$ are taken to be independent of the velocity v.

Sometimes the independence of B on v is advanced as a hypothesis not associated with the condition (9) that supplements it. Here we see an error, at least, when we are dealing with the Brownian motion of a particle in a gas. Of course, without concretizing the physical meaning of the arguments and the functions, in a purely phenomenological approach, a dependence of ν_1 and B on the variables of the problem is quite possible (see, e.g., Refs. 4 and 8).

Problems close in spirit arise in the opposite limiting case $m \ll m_b$, when the velocities of the buffer particles on the average are considerably smaller than those of the impurity particles ($\bar{v} \gg \bar{v}_b$). Upon assuming in Eq. (14) that $\mu/m = m_b(m+m_b) \approx 1$ and substituting $\delta(\mathbf{v}_1 - \mathbf{u}_1)$ in place of $W_b(\mathbf{v}_1 - \mathbf{u}_1)$, we obtain

$$A(\mathbf{v}|\mathbf{v}_1) = 2N_b\sigma(\mathbf{v}|\mathbf{v}_1)\delta(\mathbf{v}^2 - \mathbf{v}_1^2). \tag{27}$$

In other words, in this approximation the buffer particles are assumed to be immobile, while the relative velocities $\mathbf{u}$ and $\mathbf{u}_1$ are assumed to coincide respectively with $\mathbf{v}$ and $\mathbf{v}_1$. The limiting relationship (27) is known as the Lorentz model, according to which the modulus of the velocity does not vary in collision, while the angular dependence of the kernel and of the differential cross section on the angle between $\mathbf{v}$ and $\mathbf{v}_1$ coincide.

The distribution $W_b(\mathbf{v}_1 - \mathbf{u}_1)$, which was replaced by $\delta(\mathbf{v}_1 - \mathbf{u}_1)$, actually has the width $\bar{v}_b$ (see Eq. (17)). Therefore, as simple estimates show, the mean-square variation of the modulus of the velocity is

$$\langle (v - v_1)^2 \rangle \equiv \frac{1}{\nu}\int (v - v_1)^2 A(\mathbf{v}|\mathbf{v}_1) d\mathbf{v}_1 \approx \frac{\nu_1}{\nu}\bar{v}_b^2, \quad v \lesssim \bar{v}. \tag{28}$$

The variation of the modulus of the velocity on the average is relatively small, and we can proceed using the diffusion approximation with respect to the distribution of the particles over the modulus of the velocity v.

Let us examine the spatially homogeneous problem ($\nabla\rho = 0$); we shall integrate the kinetic equation over the directions $\hat{\mathbf{v}}$ of the vector $\mathbf{v}$ and take account of the fact that the kernel depends only on v, v_1, and the angle between $\mathbf{v}$ and $\mathbf{v}_1$:

$$\frac{\partial}{\partial t}\bar{\rho}(vt) = \int_0^\infty \bar{A}(v_1|v)\left[e^{(v_1^2 - v^2)/\bar{v}^2}\bar{\rho}(v_1 t) - \bar{\rho}(vt)\right] v_1^2 dv_1, \tag{29}$$

$$\bar{\rho}(vt) = \int \rho(\mathbf{v}) d\hat{\mathbf{v}}, \quad \bar{A}(v_1|v) = \int A(\mathbf{v}_1|\mathbf{v}) d\hat{\mathbf{v}}_1. \tag{30}$$

Now let us expand $\exp(v_1^2/\bar{v}^2)\bar{\rho}(v_1 t)$ in Eq. (29) in powers of $v_1 - v$ and restrict the expression to second-order terms:

$$e^{v^2/\bar{v}^2}\frac{\partial\bar{\rho}}{\partial t} = -a\frac{\partial}{\partial v}\left(e^{v^2/\bar{v}^2}\rho\right) + b\frac{\partial^2}{\partial v^2}\left(e^{v^2/\bar{v}^2}\bar{\rho}\right). \tag{31}$$

Here we have introduced the following notation for the first and second moments:

$$a = -\int_0^\infty (v_1 - v)\bar{A}(v_1|v)v_1^2 dv_1 = -\int (v_1 - v)A(\mathbf{v}_1|\mathbf{v})d\mathbf{v}_1, \tag{32}$$

$$b = \frac{1}{2}\int_0^\infty (v_1 - v)^2\bar{A}(v_1|v)v_1^2 dv_1 = \frac{1}{2}\int (v_1 - v)^2 A(\mathbf{v}_1|\mathbf{v})d\mathbf{v}_1. \tag{33}$$

As in the previous case ($m \gg m_b$), under equilibrium conditions the collision integral equals zero for arbitrary moments a and b. However, if one tries to reduce Eq. (31) to the continuity equation for the function $v^2\bar{\rho}(vt)$,[6]

$$v^2\frac{\partial}{\partial t}\bar{\rho} = \frac{\partial}{\partial v}\left[v^2 b\left(\frac{2v}{\bar{v}^2}\bar{\rho} + \frac{\partial\bar{\rho}}{\partial v}\right)\right], \tag{34}$$

then one must impose the condition

$$c \equiv a + \frac{\partial b}{\partial v} - \frac{2}{v}\left(\frac{v^2}{\bar{v}^2} - 1\right)b = 0. \tag{35}$$

For exact values of a and b, the relationship (35) is not fulfilled, but proves valid in the case

$$m \ll m_b, \quad v \gg \bar{v}_b. \tag{36}$$

We have already discussed the role of the inequality $m \ll m_b$, yet the condition $v \gg \bar{v}_b$ perhaps is not fully obvious. It involves the fundamental condition of applicability of the diffusion approximation: in the course of any time interval Δt of interest, a large number n of collisions must occur, i.e., $n = \nu\Delta t \gg 1$ (see, e.g., Ref. 8). In view of what we have said, in the diffusion approximation it is physically valid to study the intervals $v^2 \sim (\Delta v)^2 \sim \bar{v}_b^2 n \sim \bar{v}_b^2$.

Thus, also in the case of heavy buffer particles the condition (35) is satisfied only at the level of accuracy that is inherent in the diffusion approximation. In this connection it is of interest to estimate the order of magnitude of the terms dropped in Eq. (35). Here the values of the coefficients are important, and hence we must use some type of models that allow calculation of the moments in full. For the calculations the model is simplest in which the combinations $u\sigma^{(1)}(u)$, $u\sigma^{(2)}(u)$, and $u\sigma(u)$ do not depend on u. For this model we have

$$c = 15(m/m_b - \bar{v}_b^2/6v^2)a.$$

Consequently, actually the condition of applicability of the diffusion approximation is stricter by an order of magnitude:

$$15\frac{m}{m_b}\left(1 - \frac{\bar{v}^2}{6v^2}\right) \ll 1. \tag{37}$$

Similar results are obtained for a model that assumes $\sigma^{(l)}(u)$ and $\sigma(u)$ to be independent of the velocity u.

If the light particles are electrons, while the heavy particles are atoms or molecules, then the condition (37) is fulfilled with much room to spare; however, for a mixture of atomic or molecular gases, the coefficient 15 is important in practice, since the fulfillment of the inequality (37) requires $m/m_b \sim 10^{-2}$. Even for $m = 2$ (molecular hydrogen) and $m = 4$ (helium), we need $m_b = 200$ and 400, respectively.

The estimates of (25) and the parameter of (26) characterize the cases in which the entire collision integral is written in the diffusion approximation. However, this approach is not at all obligatory. The point is that the kernel $A(\mathbf{v}|\mathbf{v}_1)$ consists of components that differ strongly in their angular properties, and one can apply the diffusion approximation to describe the contribution of only small-angle scattering. Let us study this problem in greater detail.

For collisions of heavy particles (atoms, molecules) under gas-kinetic conditions, the differential cross section as a rule has three components–isotropic or almost isotropic (i), small-angle classical (c), and diffractive (d). If we use the concepts of the quasiclassical theory of scattering, we can say that isotropic scattering corresponds to values of the impact parameters smaller than the radius of the electron shells (or the radius of repulsion) of the particles. Small-angle classical scattering arises from long-range forces, e.g., van der Waals attraction. Finally, the diffractive component of scattering arises from the region of values of impact parameters exceeding the Weisskopf radius, where the interaction changes the phase of the wave function by less than $\pi/2$, and it must be treated quantum-mechanically in the Born approximation.

Estimates of the effective scattering angles for the stated three components are:

$$\theta_i \sim 1, \quad \theta_c \sim U/T \sim 0{,}1, \quad \theta_d \sim \frac{\lambda}{\rho_W} = \frac{\hbar}{\mu\bar{u}\rho_W} \sim 10^{-2}. \tag{38}$$

Here U, λ, and ρ_W are the interaction potential, the de Broglie wavelength, and the Weisskopf radius of the colliding particles. Since the change in velocity is associated with the scattering angle θ by the simple relationship (see Eq. (16))

$$\Delta v = |\mathbf{v} - \mathbf{v}_1| = \frac{\mu}{m}|\mathbf{u} - \mathbf{u}_1| = 2\frac{\mu}{m}u\sin\frac{\theta}{2}, \tag{39}$$

the kernel $A(\mathbf{v}|\mathbf{v}_1)$ consists of three components having sharply differing characteristic widths:

$$\Delta v_i \sim (\mu/m)^{1/2}\bar{v}, \quad \Delta v_c \sim 0{,}1(\mu/m)^{1/2}\bar{v},$$
$$\Delta v_d \sim 10^{-2}(\mu/m)^{1/2}\bar{v} \tag{40}$$

(Here u is replaced by the mean thermal relative velocity $\bar{u} = (m/\mu)^{1/2}\bar{v}$). It is pertinent to recall that the effective cross section of the diffractive component is approximately equal to $\pi\rho_W^2$ and constitutes about half of the total effective cross section ($\approx 2\pi\rho_W^2$).

The smallness parameter of the diffusion approximation, which equals the ratio of the effective widths of the kernel (Δv) and the nonequilibrium component of the distribution δv, is given, of course, by the widest part of the kernel,

$$\Delta v/\delta v \sim \Delta v_i/\partial v \sim (\mu/m)^{1/2}\bar{v}/\delta v,$$

in agreement with Eq. (26).

As is known, in ordinary transport phenomena—diffusion, viscosity, and thermal conduction—the main role is played by the isotropic and the small-angle classical scattering (see, e.g., Refs. 10 and 11). In these processes one usually need not take the diffractive component into account[4)] against the background of the others, since its contribution to the transport cross sections $\sigma^{(l)}(u)$ is infinitesimally small ($1 - \cos^l\theta \approx l\theta^2/2$ for small θ; see Eq. (22)). How-

ever, in problems of nonlinear spectroscopy of gases, the situation substantially differs, and the diffractive component of the scattering under some conditions can prove decisive.

As we have already mentioned, owing to absorption and stimulated emission of a plane monochromatic wave, the velocity distributions of the atoms in the combining levels acquire a sharply marked structure having the characteristic width $\delta v \sim \Gamma k$, where Γ is the radiative and impact width of the spectral line for the given transition (the so-called Bennett structure; see, e.g., Refs. 9, 12, and 13). For relatively low pressures (less than 1 Torr), the standard estimate is $\Gamma \sim 10^8\ \mathrm{s}^{-1}$ and

$$\delta v/\bar{v} \sim \Gamma/k\bar{v} \sim 10^{-2}.$$

Thus the Bennett structure can be extremely sharp on the scale of the overall width of the distribution of $\bar{v}$.

It is natural to use the diffusion approximation only in the part of the collision integral arising from small-angle (or only from diffractive) scattering, while keeping the integral form for the isotropic component:

$$S = S^{\mathrm{i}} + S^{\mathrm{d}}, \tag{41}$$

$$S^{\mathrm{d}} = \frac{\partial}{\partial v_\alpha}\left[A_\alpha^{\mathrm{d}}\rho + \frac{\partial}{\partial v_\beta}(B_{\alpha\beta}^{\mathrm{d}}\rho)\right], \tag{42}$$

$$A_\alpha^{\mathrm{d}} = \int (\mathbf{v}_1 - \mathbf{v})_\alpha A^{\mathrm{d}}(\mathbf{v}_1|\mathbf{v})d\mathbf{v}_1,$$

$$B_{\alpha\beta}^{\mathrm{d}} = \frac{1}{2}\int (\mathbf{v}_1 - \mathbf{v})_\alpha (\mathbf{v}_1 - \mathbf{v})_\beta A^{\mathrm{d}}(\mathbf{v}_1|\mathbf{v})d\mathbf{v}_1,$$

$$A^{\mathrm{d}}(\mathbf{v}|\mathbf{v}_1) = 2N_{\mathrm{b}}\int \sigma^{\mathrm{d}}(\mathbf{u}|\mathbf{u}_1)\delta(u^2 - u_1^2)\delta\left[\mathbf{v} - \mathbf{v}_1 - \frac{\mu}{m}(\mathbf{u} - \mathbf{u}_1)\right] \times W_{\mathrm{b}}(\mathbf{v}_1 - \mathbf{u}_1)d\mathbf{u}d\mathbf{u}_1. \tag{43}$$

Here $A^{\mathrm{d}}(\mathbf{v}|\mathbf{v}_1)$ and $\sigma^{\mathrm{d}}(\mathbf{u}|\mathbf{u}_1)$ are the diffractive (or small-angle) components of the kernel and of the differential cross section, while the term S^{i} involves the isotropic scattering and is described by formulas like (2) and (3). The small parameter in the expansion in (42) is the combination

$$\Delta v_{\mathrm{d}}/\delta v \sim (\mu/m)^{1/2}\theta_{\mathrm{d}}\bar{v}/\delta v. \tag{44}$$

We note the "practical disparity" of the effective angle θ_{d} and the mass ratio μ/m in the formation of the small value of the ratios Δv_{d} and δv: we can consider the values $\theta_{\mathrm{d}} \sim 10^{-1}$–$10^{-2}$ to be typical, whereas we should count the values $\mu/m = 10^{-2}$–10^{-4} "equivalent" to them as unique.

Integration over $\mathbf{u}$ and $\mathbf{u}_1$ reduces the relationship (43) for A_α^{d} and $B_{\alpha\beta}^{\mathrm{d}}$ to formulas analogous to (18)–(22). However, the assumption of sharply directed forward scattering (θ_{d} is the small parameter) leads to a simplification. Evidently we have

$$1 - \cos^2\theta = 2(1 - \cos\theta) - (1 - \cos\theta)^2,$$

and for small angles we can drop the term $(1 - \cos\theta)^2$ in the definition (22) of the transport cross section $\sigma^{(2)}(u)$. Consequently, for small-angle scattering we have $\sigma_{\mathrm{d}}^{(2)}(u) = 2\sigma_{\mathrm{d}}^{(1)}(u)$, and Eqs. (18)–(22) acquire the form

$$\mathbf{A}^{\mathrm{d}} = \nu_1^{\mathrm{d}}\mathbf{v}, \tag{18a}$$

$$\nu_1^{\mathrm{d}} = \frac{\mu}{mv}N_{\mathrm{b}}\int \sigma_{\mathrm{d}}^{(1)}(u)\cos\vartheta W_{\mathrm{b}}(\mathbf{v} - \mathbf{u})u^2 d\mathbf{u}, \tag{19a}$$

$$B_\parallel^{\mathrm{d}} + 2B_\perp^{\mathrm{d}} = \left(\frac{\mu}{m}\right)^2 N_{\mathrm{b}}\int \sigma_{\mathrm{d}}^{(1)}(u)W_{\mathrm{b}}(\mathbf{v} - \mathbf{u})u^3 d\mathbf{u}, \tag{20a}$$

$$B_\perp^{\mathrm{d}} - B_\parallel^{\mathrm{d}} = \frac{1}{4}\left(\frac{\mu}{m}\right)^2 N_{\mathrm{b}}\int \sigma_{\mathrm{d}}^{(1)}(u)(3\cos^2\vartheta - 1)W_{\mathrm{b}}(\mathbf{v} - \mathbf{u})u^3 d\mathbf{u}. \tag{21a}$$

Thus, in the case under discussion A_α^{d} and $B_{\alpha\beta}^{\mathrm{d}}$ are determined by the single transport cross section $\sigma_{\mathrm{d}}^{(1)}(u)$. Explicit integration over the angle ϑ between $\mathbf{v}$ and $\mathbf{u}$ ($\cos\vartheta$ also enters into $W_{\mathrm{b}}(\mathbf{v} - \mathbf{u})$) enables us to convince ourselves of the correctness of the identity

$$A_\alpha^{\mathrm{d}} + \frac{\partial B_{\alpha\beta}^{\mathrm{d}}}{\partial v_\beta} = \frac{2v_\beta}{\bar{v}^2}B_{\alpha\beta}^{\mathrm{d}}, \tag{9a}$$

analogous to (9), whereby the component S^{d} of the collision integral can be written in canonical form:

$$S^{\mathrm{d}} = \frac{\partial}{\partial v_\alpha}\left[B_{\alpha\beta}^{\mathrm{d}}\left(\frac{2v_\beta}{\bar{v}^2}\rho + \frac{\partial\rho}{\partial v_\beta}\right)\right]. \tag{10a}$$

We emphasize that the relationships (9a), (10a), and (19a)–(21a) are valid for an arbitrary mass ratio m/m_{b}, as long as the value of the parameter $(\mu/m)^{1/2}\theta_{\mathrm{d}}$ remains small. When $m_{\mathrm{b}}/m \sim 1$ or > 1, the quantities ν_1 and $B_\parallel^{\mathrm{d}}$, $B_\perp^{\mathrm{d}}$ substantially depend on the velocity v, as we can derive directly from Eqs. (19a)–(21a). Moreover, also in contrast to the case $m \gg m_{\mathrm{b}}$, the anisotropy of the tensor $B_{\alpha\beta}^{\mathrm{d}}$ proves to be appreciable. Besides, this can be considered to be one of the manifestations of the dependence of $B_\parallel$ and $B_\perp$ on v. We note also that the inequality $B_\perp^{\mathrm{d}} > B_\parallel^{\mathrm{d}}$ holds for small-angle scattering, whereas in the model of isotropic scattering[5] we have $B_\perp < B_\parallel$.

We should emphasize that we have assumed implicitly above a rather fast decline of the differential cross section with increasing scattering angle, which brings about a real decline in the means of $1 - \cos\theta$, $(1 - \cos\theta)^2$, etc. Of course, this is the situation in models with an exponential law of decline. However, with power-function laws complications can occur. For example, for Rutherford scattering we have $\sigma(\mathbf{u}|\mathbf{u}) \propto 1/\sin^4\theta/2$; in this case the means of $1 - \cos\theta$ and of $(1 - \cos\theta)^l$, $l \geqslant 2$, differ only in the value of the Coulomb logarithm.

Thus it is precisely in singling out the small-angle component of the scattering that the diffusion approximation leads to the general form of the Fokker–Planck equation with coefficients dependent on the velocity, with anisotropy of the diffusion tensor, and with the nontrivial relationship (9a) between A_α^{d} and $B_{\alpha\beta}^{\mathrm{d}}$. Yet the traditional case of migration of a heavy particle in a lighter buffer gas is highly simplified, since the transport rate and the diffusion coefficient do not depend on the velocity.

We recall that applicability of the diffusion approximation requires a large number of collisions. Therefore, when we single out the small-angle scattering and describe it with a differential operator we assume that the small-angle scattering cross section is far larger than that of isotropic scattering.

Let us turn now to analyzing the features inherent in the migration of excited particles. Here the new and very important factor is the finite lifetime caused by spontaneous radiative decay and inelastic processes in collisions. For the

ground state the lifetime is not bounded, and steady-state conditions correspond to the equilibrium distribution, i.e., statistical equilibrium between the migrating particles and the buffer gas.[6)] The duration of an excited state is finite, and it can prove to be too short for attainment of an equilibrium distribution. Therefore the steady-state velocity distribution in the excited state often proves to be nonequilibrium. Such conditions arise in regard to many degrees of freedom, when "there isn't time" for the collisions to establish an equilibrium distribution.

Let us illustrate the role of the short lifetime of an excited state with a very simple diffusion model. Let the index j denote belonging to a level having the energy E_j. The kinetic equation for the element of the density matrix $\rho_{jj}(\mathbf{r}\mathbf{v}t)$ has the form (see, e.g., Ref. 9)

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla+\Gamma_j\right)\rho_{jj}=S_j+q_j. \tag{45}$$

Here Γ_j is the decay constant, S_j is the elastic component of the collision integral, q_j describes the entry into the level j owing to collisions and interaction with radiation. The quantity q_j does not contain ρ_{jj} in explicit form and is usually treated as the right-hand side of Eq. (45). Therefore we shall study below the equation

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla+\Gamma_j\right)\rho_{jj}=S_j. \tag{46}$$

In the diffusion approximation and for a model in which the transport frequency ν_1 and the diffusion coefficient depend on v, the latter equation has the form

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla+\Gamma_j\right)\rho_{jj}=\nu_1\frac{\partial}{\partial\mathbf{v}}\left(\mathbf{v}\rho_{jj}+\frac{\bar{v}^2}{2}\frac{\partial\rho_{jj}}{\partial\mathbf{v}}\right). \tag{47}$$

The Green's function of this equation is well known (see, e.g., Refs. 3, 14, and 15):

$$f(\mathbf{r}\mathbf{v}t|\mathbf{r}'\mathbf{v}')=\left[2\pi(GP-H^2)^{1/2}\right]^{-3}\exp\left\{-\Gamma_j t-\frac{G\vec{\xi}^2-2H\vec{\xi}\vec{\eta}+P\vec{\eta}^2}{2(GP-H^2)}\right\}, \tag{48}$$

$$G=\frac{\bar{v}^2}{2}\left(1-e^{-2\nu_1 t}\right),\quad H=\frac{\bar{v}^2}{2\nu_1}\left(1-e^{-\nu_1 t}\right)^2,$$

$$P=\frac{\bar{v}^2}{\nu_1^2}\left[\nu_1 t-1+e^{-\nu_1 t}-\frac{1}{2}\left(1-e^{-\nu_1 t}\right)^2\right], \tag{49}$$

$$\vec{\xi}=\mathbf{r}-\mathbf{r}'-\mathbf{v}'\left(1-e^{-\nu_1 t}\right)/\nu_1,\quad \vec{\eta}=\mathbf{v}-e^{-\nu_1 t}\mathbf{v}'. \tag{50}$$

The Fourier transform of the Green's function of (48) with respect to the variables $\mathbf{r}$ and $\mathbf{v}$, which is sometimes more convenient, is given by the formula

$$f(\mathbf{k}\vec{\varkappa}t|\mathbf{r}'\mathbf{v}')=\exp\left[-\Gamma_j t-i\mathbf{p}\mathbf{k}-i\mathbf{g}\vec{\varkappa}-\frac{1}{2}\left(P\mathbf{k}^2+2H\mathbf{k}\vec{\varkappa}+G\vec{\varkappa}^2\right)\right],$$

$$\mathbf{p}=\mathbf{r}'+\mathbf{v}'\left(1-e^{-\nu_1 t}\right)/\nu_1,\quad \mathbf{g}=e^{-\nu_1 t}\mathbf{v}'. \tag{48a}$$

Here $\mathbf{k}$ and $\vec{\varkappa}$ are the Fourier variables conjugate to $\mathbf{r}$ and $\mathbf{v}$, respectively.

In the absence of decay ($\Gamma_j=0$, ground state) one usually does not consider short times $\nu_1 t\ll 1$, since they constitute only the very onset of evolution, while we are interested in the duration of the transition phase of the process ($\sim 1/\nu_1$) and relatively long times $\nu_1 t\gg 1$,

$$G=\bar{v}^2/2,\quad P=\bar{v}^2 t/\nu_1=2Dt,\quad D=\bar{v}^2/2\nu_1, \tag{51}$$

when memory is lost of the initial velocity ($\vec{\eta}=\mathbf{v}$), the variance G of the distribution over $\mathbf{v}$ is close to $\bar{v}^2/2$, and the variance of the displacements P increases by the "diffusion" law:

$$f(\mathbf{r}\mathbf{v}t|\mathbf{r}'\mathbf{v}')=\left[\pi\bar{v}^2(2t\nu_1)^{1/2}\right]^{-3}\exp\left[-\frac{\mathbf{v}^2}{\bar{v}^2}-\frac{(\mathbf{r}-\mathbf{r}')^2}{2\bar{v}^2 t/\nu_1}\right]. \tag{52}$$

For excited states we are interested in the opposite limiting case[15] when everything is finished in relatively short times,

$$\nu_1 t\lesssim\nu_1/\Gamma_j\ll 1, \tag{53}$$

and the variances are small:

$$G=\bar{v}^2\nu_1 t\ll\bar{v}^2,\quad H=\bar{v}^2\nu_1 t^2/2,\quad P=\bar{v}^2\nu_1 t^3/3\ll\bar{v}^2 t/\nu_1, \tag{54}$$

that is, within the lifetime of the excited state the sharply marked structure "has no time" to expand and remains narrow on the scale of $\bar{v}$. It is precisely in this limiting case that the variance of the velocity distribution increases in proportion to t (according to a "diffusion" law), while the variance of the displacement is $P\propto t^3$. Moreover, the effect of braking plays a smaller role here than diffusion,

$$(1-e^{-\nu_1 t})|\mathbf{v}'|\approx\nu_1 t|\mathbf{v}'|\ll(\nu_1 t)^{1/2}\bar{v}, \tag{55}$$

provided, of course, that the initial velocity v' does not exceed the mean thermal v by a large factor. As a result the Green's function of (48) acquires the form

$$f(\mathbf{r}\mathbf{v}t|\mathbf{r}'\mathbf{v}')=3\sqrt{3}(\pi\nu_1|t^2\bar{v}^2)^{-3}\exp\left(-\Gamma_j t-\frac{\mathbf{v}^2-\mathbf{v}'^2}{2\bar{v}^2}\right)$$
$$\times\exp\left\{-\frac{(\mathbf{v}-\mathbf{v}')^2}{2\bar{v}^2\nu_1 t}-\frac{6}{\bar{v}^2\nu_1 t^3}\left[\mathbf{r}-\mathbf{r}'-(\mathbf{v}+\mathbf{v}')\frac{t}{2}\right]^2\right\}. \tag{56}$$

In the given approximation the "braking effect" was manifested in the universal asymmetric coefficient $\exp[-(\mathbf{v}^2-\mathbf{v}'^2)/2\bar{v}^2]$. We can easily convince ourselves (e.g., by using Eq. (13)) that in an arbitrary kernel of the collision integral this factor is the sole coefficient asymmetric with regard to permutation of $\mathbf{v}$ and $\mathbf{v}'$, namely

$$A(\mathbf{v}|\mathbf{v}_1)=e^{(v_1^2-v^2)/2\bar{v}^2}A_s(\mathbf{v}|\mathbf{v}_1)=e^{(v_1^2-v^2)/2\bar{v}^2}A_s(\mathbf{v}_1|\mathbf{v}).$$

Here the function $A_s(\mathbf{v}|\mathbf{v}_1)$ is symmetric with respect to permuting $\mathbf{v}$ and $\mathbf{v}_1$. Hence we can say that in the approximation of (53) the asymmetry of the kernel is simply "transferred" to the Green's function. The steady-state population of the excited level j is created by continuous excitation, which is described by the right-hand side of Eq. (45). Let us study the spatially homogeneous steady-state problem, upon assuming that

$$q_j=Q_j\delta(\mathbf{v}-\mathbf{v}').$$

Then we have

$$\rho_{jj} = Q_j F_j(\mathbf{v}|\mathbf{v}');$$

$$F_j(\mathbf{v}|\mathbf{v}') = \int_0^\infty \left[\pi\bar{v}^2(1-e^{-2\nu_1 t})\right]^{-3/2} \exp\left[-\Gamma_j t - \frac{(\mathbf{v}-e^{-\nu_1 t}\mathbf{v}')^2}{(1-e^{-2\nu_1 t})\bar{v}^2}\right] dt. \tag{57}$$

The function $F_j(\mathbf{v}|\mathbf{v}')$ amounts to the solution of the equation

$$\Gamma_j F_j = \nu_1 \frac{\partial}{\partial \mathbf{v}}\left(\mathbf{v}F_j + \frac{\bar{v}^2}{2}\frac{\partial F_j}{\partial \mathbf{v}}\right) + \delta(\mathbf{v}-\mathbf{v}'), \tag{58}$$

i.e., the Green's function of the steady-state, spatially homogeneous problem. For rapidly decaying excited states (condition (53)), we have[7]

$$F_j(\mathbf{v}|\mathbf{v}') = \int_0^\infty \frac{dt}{(2\pi\bar{v}^2\nu_1 t)^{3/2}} \exp\left(-\Gamma_j t - \frac{|\mathbf{v}-\mathbf{v}'|^2}{2\bar{v}^2\nu_1 t} - \frac{\mathbf{v}^2-\mathbf{v}'^2}{2\bar{v}^2}\right)$$

$$= \frac{1}{4\pi\alpha^2\bar{v}^2|\mathbf{v}-\mathbf{v}'|\Gamma_j} \exp\left\{-\frac{|\mathbf{v}-\mathbf{v}'|}{\alpha\bar{v}} - \frac{\mathbf{v}^2-\mathbf{v}'^2}{2\bar{v}^2}\right\},$$

$$\alpha = \left(\frac{\nu_1}{2\Gamma_j}\right)^{1/2} \ll 1. \tag{59}$$

A characteristic feature of the Green's function of (59) is the already mentioned small asymmetry with respect to the point $\mathbf{v} = \mathbf{v}'$ and the integrable singularity at this point. One can show[9] that the arbitrary kernel of the collision integral contains the factor $1/|\mathbf{v}-\mathbf{v}'|$. Consequently, similarly to the asymmetric coefficient, the singularity of the kernel in the approximation of (53) is "transferred" to the Green's function of (59). The scale of the structure in the velocity distribution is given by the parameter $\alpha\bar{v} = (\nu_1/2\Gamma_j)^{1/2}\bar{v} \ll \bar{v}$.

The function (57) is closely associated with the solution of the steady-state equation

$$\Gamma_j \rho_{jj}(\mathbf{v}) = -\nu\rho_{jj}(\mathbf{v}) + \int A(\mathbf{v}|\mathbf{v}_1)\rho_{jj}(\mathbf{v}_1)d\mathbf{v}_1 + Q_j\delta(\mathbf{v}-\mathbf{v}') \tag{60}$$

having the Keilson–Storer model kernel[9,18]

$$A(\mathbf{v}|\mathbf{v}_1) = A_{KS}(\mathbf{v}|\mathbf{v}_1) = \nu\left[\pi(1-\gamma^2)\bar{v}^2\right]^{-3/2} \exp\left[-\frac{(\mathbf{v}-\gamma\mathbf{v}_1)^2}{(1-\gamma^2)\bar{v}^2}\right]. \tag{61}$$

By using the method of successive approximations we can arrive at the formulas of (62):

$$\rho_{jj}(\mathbf{v}) = Q_j F_{KS}(\mathbf{v}|\mathbf{v}'),\ \Gamma_j F_{KS}(\mathbf{v}|\mathbf{v}')$$

$$= \frac{1}{1+n}\left\{\delta(\mathbf{v}-\mathbf{v}') + n\sum_{l=1}^{\infty} \frac{[n/(1+n)]^l}{[\pi\bar{v}^2(1-\gamma^{2l})]^{3/2}} \times \exp\left[-\frac{(\mathbf{v}-\gamma^l\mathbf{v}')^2}{(1-\gamma^{2l})\bar{v}^2}\right]\right\};$$

$$n = \nu/\Gamma_j \tag{62}$$

The Green's function (57) of the diffusion approximation and the regular component of the function $F_{KS}(\mathbf{v}|\mathbf{v}')$ are approximately equal if we can replace the series in Eq. (62) with an integral and assume that

$$\nu_1 t = l\ln\frac{1}{\gamma} \approx (1-\gamma)l, \quad l\ln\left(1+\frac{1}{n}\right)$$

$$= \Gamma_j t \frac{1-\gamma}{(-\ln\gamma)}\ln\left(1+\frac{1}{n}\right)^n \approx \Gamma_j t, \tag{63}$$

which corresponds to the conditions

$$\nu_1/\nu = 1-\gamma \ll 1, \quad n = \nu/\Gamma_j \gg 1, \tag{64}$$

which must be fulfilled for applicability of the diffusion approximation.

Let us call attention to $\delta(\mathbf{v}-\mathbf{v}')$ in the expression (62) for the Green's function $F_{KS}(\mathbf{v}|\mathbf{v}')$. The given term, which is also characteristic of short-lived states, evidently describes the fraction of the atoms that have not undergone even a single elastic collision in their time of existence in the excited state. The fraction of such atoms (integrated over $\mathbf{v}$) is $\Gamma_j/(\Gamma_j+\nu) = 1/(1+n) \ll 1$. However, we must not drop the δ-function *a priori*,[8] since it can lead to a considerably sharper structure than the regular part, owing to which the amplitude of this sharp component can prove to be rather large. Consequently the Green's function of the diffusion equation for the steady-state, spatially homogeneous problem has the form

$$\frac{1}{1+n}\left[\frac{1}{\Gamma_j}\delta(\mathbf{v}-\mathbf{v}') + nF_j(\mathbf{v}|\mathbf{v}')\right], \quad n = \nu/\Gamma_j, \tag{65}$$

where $F_j(\mathbf{v}|\mathbf{v}')$ is given by Eq. (57) or (59).

Thus the diffusion approximation and the theory based on the Keilson–Storer model kernel Eq. (61) are equivalent from the standpoint of parameterizing the system: the first and second moments of the kernel of the collision integral contain the very same transport frequency of collisions ν_1, which is taken to be independent of the velocity; in the Keilson–Storer model ν_1 is expressed in terms of the parameter γ:

$$\mathbf{A} = \nu_1\mathbf{v} = (1-\gamma)\nu\mathbf{v},$$

$$\frac{1}{4}(1-\gamma^2)\nu\bar{v}^2 \approx \frac{1}{2}(1-\gamma)\nu\bar{v}^2 = \frac{1}{2}\nu_1\bar{v}^2 = B,$$

$$n = \frac{\nu}{\Gamma_j} = \frac{\nu_1}{(1-\gamma)\Gamma_j}. \tag{66}$$

If one must retain the term $\delta(\mathbf{v}-\mathbf{v}')$ in Eq. (65), then the problem requires three quantities–n, ν_1, and B–for parameterization.

The short duration of excited states, which restricts migration in velocity space, makes possible a highly distinctive variant of the so-called "cascade diffusion". The point is that a transition from high enough excited states to the ground state often occurs in cascade fashion and by different channels. Consequently, for any concrete level a complex structure can be formed in the velocity distribution, consisting of several components with varying degrees of diffusion broadening and displacement owing to the braking effect (we can point to Ref. 19 as an example of analysis of a structure of this type).

Up to now we have been discussing the migration of particles, excited or unexcited, i.e., on the formal level–the diagonal elements of the density matrix. In treating the migration of dipole moments (or coherence, or nondiagonal elements), new ideas are drawn in. Of interest are the coherences, both between the magnetic sublevels of a degenerate state, and between stationary states of differing energy. Analysis of the features of migration of coherence of the first type requires taking into account disorienting collisions, the anisotropy of interaction of the colliding particles, and the dependence of the cross sections on factors other than the scattering angle. Here (as was true everywhere above), we shall digress from level degeneracy, and focus attention on "optical" coherence, coherence of the second type.

According to correlation theory, the space-time Fourier transform of the coherence is closely connected with the form of the contour of spectral lines (see, e.g., Refs. 9, 14, and 20). Therefore the problem of Doppler broadening of spectral lines amounts to a transposition into spectral language of the problem of migration of coherence in velocity space.

The first fruitful attempts to apply the diffusion approximation to the problem of broadening of spectral lines go back as far as the fifties.[21,22] It was shown that elastic collisions, by retarding the displacement of dipole moments, decrease the role of Doppler broadening due to thermal motion. In the limit of high pressures they lead to a Lorentz contour having the so-called diffusion width:

$$I(\omega)=\frac{1}{\pi}\frac{\Gamma+\gamma_d}{(\Gamma+\gamma_d)^2+(\omega-\omega_0-\Delta)^2},\quad \nu_1\gg k\bar{v},\ |\omega-\omega_0|, \tag{67}$$

$$\gamma_d=(k\bar{v})^2/2\nu_1; \tag{68}$$

Here Γ and Δ are the half-width and the displacement of the line caused by spontaneous relaxation and by the interaction during collisions. The quantity γ_d, which is called the diffusion half-width, describes the residual contribution of the motion of the emitter.

At the beginning of the seventies a series of studies[23–27] appeared, in which the kernel of the collision integral in the kinetic equation for the nondiagonal element $\rho_{mn}(\mathbf{r}\mathbf{v}t)$ of the density matrix was expressed in terms of the scattering amplitude. Namely, the equation for $\rho_{mn}(\mathbf{r}\mathbf{v}t)$ has the form

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla+\gamma_{mn}+\nu_{mn}\right)\rho_{mn}(\mathbf{r}\mathbf{v}t)=\int A_{mn}(\mathbf{v}|\mathbf{v}_1)\rho_{mn}(\mathbf{r}\mathbf{v}_1t)d\mathbf{v}_1+q_{mn}(\mathbf{r}\mathbf{v}t), \tag{69}$$

Here γ_{mn} and $q_{mn}(\mathbf{r}\mathbf{v}t)$ describe the spontaneous decay and the excitation of coherence (e.g., by light), while we have

$$\nu_{mn}=\frac{2\pi\hbar}{i\mu}\int\left(f_{mn}(\mathbf{u}|\mathbf{u})-f^*_{nn}(\mathbf{u}|\mathbf{u})\right)W_b(\mathbf{v}-\mathbf{u})d\mathbf{u}, \tag{70}$$

$$A_{mn}(\mathbf{v}|\mathbf{v}_1)=2N_b\int\sigma_{mn}(\mathbf{u}|\mathbf{u}_1)\delta(\mathbf{u}^2-\mathbf{u}_1^2)\delta\left[\mathbf{v}-\mathbf{v}_1-\frac{\mu}{m}(\mathbf{u}-\mathbf{u}_1)\right]\times W_b(\mathbf{v}_1-\mathbf{u}_1)d\mathbf{u}d\mathbf{u}_1, \tag{71}$$

$$\sigma_{mn}(\mathbf{u}|\mathbf{u}_1)=f_{mm}(\mathbf{u}|\mathbf{u}_1)f^*_{nn}(\mathbf{u}|\mathbf{u}_1). \tag{72}$$

Here $f_{jj}(\mathbf{u}|\mathbf{u}_1)$ are the amplitudes of elastic scattering in the combining states $j=m,\ n$. Thus the nondiagonal element obeys a kinetic equation of the Boltzmann type. However, the exit frequency ν_{mn} and the kernel $A_{mn}(\mathbf{v}|\mathbf{v}_1)$ generally turn out to be complex. If the scattering in the states m and n is the same, $f_{mm}(\mathbf{u}|\mathbf{u}_1)=f_{nn}(\mathbf{u}|\mathbf{u}_1)$, then ν_{mn} and $A_{mn}(\mathbf{v}|\mathbf{v}_1)$ are real and coincide with the analogous quantities of the collision integrals in the equations for the number of particles $\rho_{jj}(\mathbf{r}\mathbf{v}t),\ j=m,\ n$.

A remarkable feature of the problem of broadening of spectral lines consists of the fact that one must deal in it with an "extremely inhomogeneous" problem. Actually, in the linear approximation, e.g., the excitation has the form

$$q_{mn}(\mathbf{r}\mathbf{v}t)=Q\exp[i(\mathbf{k}\mathbf{r}-\omega t)], \tag{73}$$

that is, it has a fine spatial inhomogeneity with the scale $\lambda=2\pi/k\sim10^{-4}$ cm. We can represent the solution of Eq. (69) as:

$$\rho_{mn}(\mathbf{r}\mathbf{v}t)=Q\int F_{mn}(\mathbf{r}\mathbf{v}t|\mathbf{r}'\mathbf{v}'t')\exp[i(\mathbf{k}\mathbf{r}'-\omega t')]d\mathbf{r}'d\mathbf{v}'dt'. \tag{74}$$

Here $F_{mn}(\mathbf{r}\mathbf{v}t|\mathbf{r}'\mathbf{v}'t)$ is the Green's function, which satisfies the equation

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla+\gamma_{mn}+\nu_{mn}\right)F_{mn}(\mathbf{r}\mathbf{v}t|\mathbf{r}'\mathbf{v}'t')=\int A_{mn}(\mathbf{v}|\mathbf{v}_1)F_{mn}(\mathbf{r}\mathbf{v}_1t|\mathbf{r}'\mathbf{v}'t')d\mathbf{v}_1+\delta(\mathbf{r}-\mathbf{r}')\delta(\mathbf{v}-\mathbf{v}')\delta(t-t'). \tag{75}$$

Thus, to calculate $\rho_{mn}(\mathbf{r}\mathbf{v}t)$ by Eq. (74) we must actually know the function, which in the initial stage of its evolution is δ-shaped in $\mathbf{r}\mathbf{v}$-space, and find its Fourier transform. Contrary to this, in problems of the populations, one often encounters spatially homogeneous conditions, and one deals only with migration in velocity space, and the calculations are substantially simplified.

The space and time dependence of an excitation of the type of (73) is characteristic of single-photon processes of emission and absorption. In the case of multiphoton excitation of coherence, the combination $\Sigma_i(\pm\mathbf{k}_i)\mathbf{r}$ with the wave vectors $\mathbf{k}_i$ of the interacting waves appears in $q_{mn}(\mathbf{r}\mathbf{v}t)$ instead of $\mathbf{k}\mathbf{r}$. Here the sign is chosen according to whether the photon i is emitted or absorbed. In certain combinations of the vectors $\mathbf{k}_i$ the inhomogeneity can prove to be large-scale. In the other, spectral language this implies a substantial mutual compensation of the Doppler shifts of the frequencies of the different waves.

Let us return to the problem of the diffusion approximation with respect to $\rho_{mn}(\mathbf{r}\mathbf{v}t)$. We note in advance that the properties of the cross sections $\sigma_{mn}(\mathbf{u}|\mathbf{u}_1)$ and the kernels $A_{mn}(\mathbf{v}|\mathbf{v}_1)$ have been studied considerably less fully than the analogous quantities for the populations. Evidently $\sigma_{mn}(\mathbf{u}|\mathbf{u}_1)$ is a complex oscillating function of the scattering angle for the case $f_{mm}(\mathbf{u}|\mathbf{u}_1)\neq f_{nn}(\mathbf{u}|\mathbf{u}_1)$. If $f_{mm}(\mathbf{u}|\mathbf{u}_1)$ and $f_{nn}(\mathbf{u}|\mathbf{u}_1)$ differ strongly enough, then generally we can drop the integral term in Eqs. (69) and (75). Physically this is an obvious result: the large difference between $f_{mm}(\mathbf{u}|\mathbf{u}_1)$ and $f_{nn}(\mathbf{u}|\mathbf{u}_1)$ implies a large difference of the scattering phases in the states m and n, i.e., a large collisional difference of the phases of the atomic oscillator; in other words, the lifetime of coherence arises from phase memory, and the evolution of

$\rho_{mn}(\mathbf{r}\mathbf{v}t)$ in velocity space is no longer significant. However, a number of contrary cases exist in which the migration in v-space has time to be manifested within the phase-memory time, and we must keep the integral term in Eqs. (69) and (75). Of especial interest from this standpoint are the spectral characteristics of ions and their Coulomb scattering.[19,28,29] The point is that, in the case of Coulomb interaction, the scattering amplitudes are the same for all j, and hence there are no phase effects.

A small value of the parameter $(m_b/m)^{1/2}\theta_d$ (owing to θ_d, or m_b/m, or both) allows one to use the diffusion approximation for calculating the coherence in full analogy with the cases discussed above of the populations. In going over to the differential form of the collision operator, one in fact uses the dependence of $\sigma_{mn}(\mathbf{u}|\mathbf{u}_1)$ only on the scattering angle and u (but not on the azimuthal angle), and also the fact that $A_{mn}(\mathbf{v}|\mathbf{v}_1)$ depends on $\mathbf{v}$ and $\mathbf{v}_1$ via the factor

$$\delta\left[\mathbf{v}-\mathbf{v}_1-\frac{\mu}{m}(\mathbf{u}-\mathbf{u}_1)\right]W_b(\mathbf{v}_1-\mathbf{u}_1). \tag{76}$$

The same properties were important also in the case of the populations. Therefore the Fokker–Planck equation for $\rho_{mn}(\mathbf{r}\mathbf{v}t)$ has the standard form

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla+\gamma_{mn}+\nu_{mn}-\tilde{\nu}_{mn}\right)\rho_{mn}$$
$$=\frac{\partial}{\partial v_\alpha}\left[B^{mn}_{\alpha\beta}\left(\frac{2v_\beta}{\bar{v}^2}\rho_{mn}+\frac{\partial\rho_{mn}}{\partial v_\beta}\right)\right],$$
$$\tilde{\nu}_{mn}=\int A_{mn}(\mathbf{v}_1|\mathbf{v})d\mathbf{v}_1, \tag{77}$$

Here the complex tensor $B^{mn}_{\alpha\beta}$ is calculated by Eqs. (7) or (43), but with the use of the kernel $A_{mn}(\mathbf{v}|\mathbf{v}_1)$. What we have said implies, in particular, that the microscopic theory (but not phenomenological considerations) imply the possibility of introducing into Eq. (77) the concept of the density of collisional coherence flux in v-space:

$$s^{mn}_\alpha=-B^{mn}_{\alpha\beta}\left(\frac{2v_\beta}{\bar{v}^2}\rho_{mn}+\frac{\partial\rho_{mn}}{\partial v_\beta}\right),$$

and the vanishing of this flux density in the case of an equilibrium velocity distribution $\rho_{mn}(\mathbf{r}\mathbf{v}t)$. Caution in using the general phenomenological considerations developed in application to the properties of distributions of particles is obligatory, since the quantity $\rho_{mn}(\mathbf{r}\mathbf{v}t)$ itself serves as a characteristic of nonequilibrium: a system with nonzero coherence is statistically not at equilibrium. Nevertheless, if $\rho_{mn}(\mathbf{r}\mathbf{v}t)$ has a Maxwellian velocity distribution, then the elastic component of the collision integral in the kinetic equations (69) and (77) can give only the very simple relaxation term $(\nu_{mn}-\tilde{\nu}_{mn})\rho_{mn}$, which reflects the role of phase jumps of the atomic oscillator in collisions.

The general ideas and concrete situations discussed above have shown, I hope, rather graphically the existence of really distinctive, unusual conditions under which the migration of particles in resonance interaction with laser radiation occurs in a gas. In many of their properties the phenomena being discussed are close to those that occur in beams, or perhaps more precisely, in gas jets having a degree of collimation of the order of $\Gamma/k\bar{v}$. The Bennett structure can be treated as a sort of beam or jet in velocity space. In one Cartesian coordinate—the projection of the velocity on the wave vector of the active wave—the jet is sharply bounded by an interval of the order of $\Gamma/k \ll \bar{v}$, while in the two orthogonal components it possesses an almost Maxwellian distribution. The overall picture of the relaxation of the Bennett structure corresponds more to the beam ideology than to the standard pictures developed in application to transport phenomena.

I think that precisely by the unusualness of the situation and by its contrast with the prevailing canons can one explain the following curious historical fact. The existence of the Bennett structure was first established in 1962.[30] However, only after 17 years[31] were its mechanical consequences acknowledged; Bennett jets (or beams) of atoms on two optically combining levels are braked by the buffer gas in equal measure, whereby the gas as a whole acquires a macroscopic mechanical motion. This phenomenon, which has been called light-induced drift, proved to be the beginning of a new field–gas kinetics in the field of laser radiation, with interesting consequences with regard to atomic and molecular physics, technology, astrophysics, and other fields.[9)] I mention this problem, not at all in order to discuss its essence to any extent whatever, but exclusively to stress the importance of the methodological aspects in the systems of our knowledge and their dependence on "typical physical conditions".

I take pleasure in thanking M. I. D'yakonov, V. I. Perel', A. M. Shalagin, and D. A. Shapiro for interesting discussions, useful to me, of the questions touched upon here.

[1)] The velocity as a fundamental variable seems to us more convenient than the frequently used momentum.

[2)] The buffer gas, by assumption, exists in a state of statistical equilibrium. In the converse case, parallel with the problem of migration of the particles, one must study the evolution of the buffer toward an equilibrium state.

[3)] One can explain the need to keep the terms containing the first and second derivatives of ρ and v_α also in another way. The diffusion term leads to a broadening of the region of localization of the particles in v-space. Dynamic friction acts in the opposite direction. The mutual compensation of diffusion and friction ensures the stability of the equilibrium distribution, as stated by Eq. (9).

[4)] An exception are the light gases at low temperatures.[10]

[5)] For isotropic scattering ($\sigma(\mathbf{u}|\mathbf{u})=$ const) we have $\sigma^{(2)}=(2/3)\sigma^{(1)}$, and in Eq. (21) we obtain $\sigma^{(1)}-(3/4)\sigma^{(2)}=(1/2)\sigma^{(1)}$. As regards the integral over ϑ, it is always positive.

[6)] Of course, we have in mind spatially homogeneous conditions. Cold or hot walls or other forms of artificially maintained substantial spatial inhomogeneity can lead (and actually do so in many cases) to a sharp breakdown of equilibrium between the different components of the gas mixture and the various degrees of freedom.

[7)] Reference 17 gives a one-dimensional analog of the Green's function of (59).

[8)] This circumstance has been emphasized also in Ref. 18.

[9)] Material of review character on light-induced drift and associated problems can be found in Refs. 32–35.

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Translated by M. V. King