Convenient expressions of diffusion coefficients for free electrons in gases in the presence of Ramsauer effects

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The lateral- and longitudinal-diffusion coefficients for free electrons in weakly ionized gases (or in intrinsic semiconductors) under a steady state and uniform electric field **E** and with elastic collision frequency much larger than the inelastic one are obtained by the correlation function of the electron velocities. The mean-free-path method is used, since it allows expansions without divergences in the small number $W^2/\langle v^2\rangle$ where W is the electron drift velocity and $\langle v^2\rangle$ the mean-square value of the electron speed $\mathbf{v}$. The obtained explicit expressions are particularly convenient when there are Ramsauer effects which can produce up to 6% errors in the results obtained by the usual Boltzmann-Legendre expansion. Moreover, the obtained expressions are immediately generalized so as to give noise spectral densities including the "convective noise" found by Gurevich, which corresponds to the difference between the generalized longitudinal- and lateral-diffusion coefficients.

I. INTRODUCTION

In the last thirty years the differences between the lateral- and longitudinal-diffusion coefficients for free carriers in a scattering medium under the accelerating influence of an external electric field have been discovered.

The authors[1-7] who have developed the theory of the diffusion coefficients consider a stationary case and expand the relevant time-independent distribution function $f(\mathbf{r},\mathbf{v})$ for electrons in both spatial gradients and Legendre polynomials and substitute the expansion in the Boltzmann equation. The corresponding solutions are used to obtain transport quantities. When the spatial gradients are not parallel to the external electric field one must use expansions in spherical harmonics instead of in Legendre polynomials, as exhaustively shown in the work of Robson and Ness[8] or in that of Kumar, Skullerud, and Robson,[9] which can be considered as the most recent treatise on this subject.

The present approach is completely different. It applies the mean-free-path method to the velocity correlation functions by which explicit expressions of the diffusion coefficients can be obtained. This procedure, although mentioned as an introduction by Skullerud,[2] has not been used by the authors who work in the field of ionized gases, with the exception of a previous work by one of us[10] which is here synthesized and corrected for a second-order error in a normalization. On the contrary, the correlation functions are always used by the authors who work in the field of electric noise.

Besides giving a connection between the two fields of research, the method of the mean free paths applied to the correlation functions has the following other advantages.

(i) The position $\mathbf{r}$ of the electron does not appear in the correlation function of the electron velocity $\langle v(0)v(\tau)\rangle$ so that the integration over $\mathbf{r}$ (in order to find average transport quantities) reduces the distribution function $f(\mathbf{r},\mathbf{v})$ in the phase space to its marginal distribution function $f(\mathbf{v})$. Consequently, the density gradient expansion is no longer necessary.

(ii) It gives an independent and radically different expression for the difference between the longitudinal-diffusion coefficient D_x and the lateral-diffusion coefficient D_y (or D_z). For the values of the ratio D_x/D_y there are discrepancies among different authors, although Penetrante and Bardsley[11] have shown that Lucas[4] performed a nonconsistent approximation in the first equation of the two hierarchical equations of the P_1 approximation to the Boltzmann equation, so that Lucas's results are not reliable. Moreover the Monte Carlo simulations of Lucas and Saele[12] are affected by statistical fluctuations and the results of Francey and Jones[13] are unreliable because it is not correct to apply the steady-state theory of the Boltzmann equation to time-of-flight experiments. However, when the collision frequency ν is of the kind $\nu=\alpha v^n$, for $n=\frac{1}{2}$ (or $-\frac{1}{4}$ if we write $\nu=\beta v^{2n}$), there is a discrepancy of 3.5% even between reliable authors.[2,3,5,11]

(iii) D_x and $D_y=D_z$ are easily generalized by the correlation function so as to become $D_x(\omega)$ and $D_y(\omega)$, where ω is the angular frequency, which are proportional to the transversal and longitudinal spectral noises, respectively. The most important result, shown in a future paper, is that $D_x(\omega)$ contains the "convective noise" found by Gurevich,[14] and, we suppose, even the famous "excess" noise with frequency dependence $1/\omega$, whose origin is so far unknown. This noise, to which international conferences have been devoted, is probably due to runaway electrons.[15]

(iv) The mean-free-path method, with the expansion of the exponentials in which the auxiliary term b is introduced,[16,17] is particularly convenient when there are Ramsauer effects. In these cases the usual method can give results different by up to 40% from those obtained by the Monte Carlo method, as shown by Milloy and Watts,[18] although the cross sections considered by them were rather unphysical. However, also in real cases the results given by different reliable methods and the Monte Carlo one can differ by 6% for the transversal-diffusion coefficient $D_T = D_x = D_y$, as shown in Table I of Pitchford, Oneil, and Rumble[19] who used a Legendre expansion up to and including eight terms. On the contrary, the method here presented guarantees errors not exceeding a factor of order unity times m/M, where m and M are the electron and molecule mass, respectively. These small errors are guaranteed even in the unphysical case of a Ramsauer cross section reaching a zero value for $v = v_0'$. Notice that in this case the usual method gives an unphysical divergence in the calculation of the main term $\langle v^2/\nu_0(v)\rangle$, where $\nu_0(v)$ is the collision frequency which vanishes for $v = v_0'$. By contrast, our main term turns out to be $\langle v^2\nu_0/(\nu_0+b)^2\rangle$ which does not diverge when $\nu_0 \to 0$ because the auxiliary term b takes into account that the electron velocity changes during a free flight so that the collision frequency is no longer zero. In practical cases $\nu_0(v)$ never vanishes but it can reach small values comparable with b so that the errors in the usual method can be of some percents.[19]

(v) Explicit expressions are given for the diffusion coefficients, expressed as integrals over a single variable so that an experimentalist no longer has to solve systems of coupled differential equations. The explicit expressions are separately given for the lateral- and longitudinal-diffusion coefficients, with and without a Ramsauer effect. The inclusion of Ramsauer effects represents the main improvement with respect to the preceding quoted work of Ref. 10.

II. DIFFUSION COEFFICIENTS EXPRESSED BY VELOCITY CORRELATION FUNCTIONS

The generalized, lateral-diffusion coefficient $D_y(\omega)$ (where ω is the angular frequency) is proportional to the spectral density $J_y(\omega)=J_z(\omega)$ of the lateral electric diffusion noise when the particles have a charge e. They can be expressed (see Appendix A) by the correlation function

$$C_y(\tau)=C_z(\tau)=\langle v_y(0)v_y(\tau)\rangle=\langle v_z(0)v_z(\tau)\rangle$$

of the diffusing-particle velocity $v(t)$ taken at two sampling times $t=0$ and $t=\tau$, respectively. In the case of a wire section of length L containing N charged particles, we have

$$D_y(\omega)=\frac{L^2\pi}{2e^2N}J_y(\omega)=\int_0^\infty d\tau\langle v_y(0)v_y(\tau)\rangle\cos(\omega\tau) . \tag{1}$$

Similarly, the generalized longitudinal-diffusion coefficient $D_x(\omega)$ and the spectral density $J_x(\omega)$ of the longitudinal electric noise can be expressed by (see Appendix A)

$$D_x(\omega)=\frac{L^2\pi}{2e^2N}J_x(\omega) = \int_0^\infty d\tau[\langle v_x(0)v_x(\tau)\rangle-\langle v_x^2\rangle]\cos(\omega\tau) . \tag{2}$$

The velocity correlation function implies two averages, the first one is over the initial velocity $\mathbf{v}(0)=\mathbf{v}_i$ and has, as a weight, the stationary probability density $f(\mathbf{v}(0),0)$. The second average is over the final velocity $\mathbf{v}(\tau)=\mathbf{v}_f$ and has, as a weight, the transition probability density $f(\mathbf{v}_f,\tau\,|\,\mathbf{v}_i)$ which gives the transition from $\mathbf{v}_i$ at time $t=0$ to $\mathbf{v}_f$ at time $t=\tau$. Using polar coordinates so that $v_x = v\mu$, where $\mu=\mathbf{v}\cdot\mathbf{E}/vE$, we have

$$\langle v_x(0)v_x(\tau)\rangle=\int_0^\infty dv_i\, v_i^2\int_0^{2\pi}d\psi_i\int_{-1}^{1}d\mu_i\, f(v_i,\mu_i)v_i\mu_i \times\int_0^\infty dv_f\, v_f^2\int_0^{2\pi}d\psi_f\int_{-1}^{1}d\mu_f\,\tilde f(v_f,\mu_f,\psi_f,\tau\,|\,v_i,\mu_i,\psi_i)v_f\mu_f , \tag{3}$$

where, because of axial symmetry around v_i, the stationary probability density $f(v_i,\mu_i)$ does not depend on ψ_i. The correlation function for transverse velocities is obtained by replacing $v(1-\mu^2)$ for $v\mu$ in (3).

For $\tau\to\infty$ the transition probability density (or propagator) $\tilde f$ tends to the stationary f. The difficulty for the practical evaluation of D_x consists of obtaining $\tilde f$. Indeed, $\tilde f$ cannot be obtained by a Fokker-Planck method, because, owing to collisions, μ changes very rapidly. For the same reason the Boltzmann equation, in which the expansion of $\tilde f$ in Legendre polynomials has been substituted, would require a truncation of the series after a larger number of terms. Fortunately, this problem can be solved by the mean-free-path method.

III. CONVENIENT EXPRESSION FOR THE TRANSITION PROBABILITY DENSITY

We introduce the velocity $\mathbf{v}_0$ immediately after collisions, related to $\mathbf{v}$ by $\mathbf{v}=\mathbf{v}_0+\mathbf{a}t$, where $\mathbf{a}=e\mathbf{E}/m$ is the acceleration and t the time of flight. In scalar form, $v\mu=v_0\mu_0+at$ and

$$v(1-\mu^2)^{1/2}=v_0(1-\mu_0^2)^{1/2} ,$$

and the inverse functions are

$$v_0=(v^2+a^2t^2-2v\mu a t)^{1/2}$$

and

$$\mu_0=(v\mu-at)(v^2+a^2t^2-2v\mu at)^{-1/2} ,$$

so that

$$dv_0\,d\mu_0=dv\,d\mu\begin{vmatrix}\frac{\partial v_0}{\partial v} & \frac{\partial v_0}{\partial \mu}\\ \frac{\partial \mu_0}{\partial v} & \frac{\partial \mu_0}{\partial \mu}\end{vmatrix}=dv\,d\mu\left[\frac{v}{v_0}\right]^2 . \tag{4}$$

The probability $d^2p(v,\mu)$ of finding the electrons in the phase cell $v^2\,dv\,(-d\mu)\,d\psi$ is

$$d^3p=-v^2\,dv\,d\mu\,f(v,\mu)\,d\psi . \tag{5}$$

If we introduce the collision source $\mathcal{X}(v_0,\mu_0)$ defined as the time derivative, calculated at $t=0$, of the probability density $d^3p/(-v_0^2\,dv_0\,d\mu_0\,d\psi_0)$ in the "initial" phase space cell, then we can write the probability $d^4p_0(v_0,\mu_0,0)$ of finding electrons in the generalized phase cell (which includes dt) as

$$d^4p_0=-v_0^2\,dv_0\,d\mu_0\,d\psi_0\,dt\,\mathcal{X}(v_0,\mu_0) . \tag{6}$$

These electrons maintain the same initial velocity $\mathbf{v}_0=(v_0,\mu_0)$ until they collide. The variation dn of the number n of electrons surviving to collisions in the time interval dt is given by $dn=-\nu_0(v)ndt$ where $\nu_0=\nu_0(v)$ is the collision frequency so that, upon integration, we get

$$n=n_0\exp\left[-\int_0^t d\xi\,\nu_0[v(\xi)]\right] . \tag{6a}$$

Consequently, after a time of flight t we have

$$\begin{aligned}d^4p&=d^4p_0\exp\int_0^t -d\xi\,\nu_0(\xi)\\ &=-v_0^2\,dv_0\,d\mu_0\,d\psi_0\,dt\mathcal{X}(v_0,\mu_0)\\ &\quad\times\exp\left[-\int_0^t d\xi\,\nu_0[v(v_0,\mu_0\xi)]\right] .\end{aligned} \tag{7}$$

Integrating over all the times of flight, we obtain

$$\begin{aligned}d^3p&=-v_0^2\,dv_0\,d\mu_0\,d\psi_0\mathcal{X}(v_0,\mu_0)\\ &\quad\times\int_0^\infty dt\exp\left[-\int_0^t \nu_0[v(\xi)]d\xi\right] .\end{aligned} \tag{8}$$

Equating (5) with (8) and using (4) gives

$$f(v,\mu)=\mathcal{X}(v_0,\mu_0)\int_0^\infty dt\exp\left[-\int_0^t d\xi\,\nu_0[v(\xi)]\right] , \tag{9}$$

which is the desired relationship between f and $\mathcal{X}$ to be used in (3).

The advantage of the use of the "initial" quantities (immediately after collisions) is mainly felt in the transition probability density. Indeed during any flight the quantities immediately after the last collision do not change so that $\mathcal{X}(\mathbf{v}_{0f},\tau\,|\,\mathbf{v}_{0i})$ can be expressed by Dirac δ functions for each flight (recall that t_s is the sth time of flight)

$$\begin{aligned}\mathcal{X}(\mathbf{v}_{0f},\tau\,|\,\mathbf{v}_{0i})&=\mathcal{X}(v_{0f},\mu_{0f},\psi_{0f},\tau\,|\,v_{0i},\mu_{0i},\psi_{0i})\\ &=\sum_{s=1}^{\infty}A_s\delta(v_{0f}-v_{0s})\delta(\mu_{0f}-\mu_{0s})\delta(\psi_{0f}-\psi_{0s})\left[\mathcal{H}\left[\tau-\sum_{j=1}^{s-1}t_j\right]-\mathcal{H}\left[\tau-\sum_{j=1}^{s}t_j\right]\right] ,\end{aligned} \tag{10}$$

where A_s are normalization constants and $\mathcal{H}(\tau)$ is the Heaviside unit step function defined as $\mathcal{H}(\tau)=0$ for $\tau<0$ and $\mathcal{H}(\tau)=1$ for $\tau\geq 0$. If τ falls in the nth time of flight, (10) says that the final velocity $\mathbf{v}_{0f}$ is equal to $\mathbf{v}_{0n}$. Since, with the use of (9),

$$\begin{aligned}1&=\int_0^\infty dv\,v^2\int_{-1}^{1}d\mu\int_0^{2\pi}d\psi\,f(v,\mu)\\ &=\int_0^\infty dv_0\,v_0^2\int_{-1}^{1}d\mu_0\int_0^{2\pi}d\psi_0\mathcal{X}(v_0,\mu_0)\int_0^\infty dt\exp\left[-\int_0^t d\xi\,\nu_0[v(\xi)]\right] ,\end{aligned} \tag{11}$$

the normalization constants A_s appearing in (10) turn out to be given by

$$A_s^{-1}=v_{0s}^2\int_0^\infty dt\exp\left[-\int_0^t d\xi\,\nu_0[v(\xi)]\right] . \tag{12}$$

The $\mathbf{v}_{0s}$ appearing in (10) depends on $\mathbf{v}_{0(s-1)}$ so that the explicit expression of (10) expressed by the initial velocity $\mathbf{v}_{0i}$ at $\tau=0$ becomes extremely complicated after few steps. Fortunately the explicit calculation of the sth contribution to the diffusion calculation turns out to be of the order of $(W^2/\langle v\rangle^2)^{s/2}$ (where W is the electron drift velocity) with respect to the contribution due to the first term of Eq. (10). Detailed calculations relevant to the second flight ($s=2$) are reported in Appendix B of Ref. 10 (Cavalleri) (although with relative second-order errors due to wrong normalizations).

For free electrons in scattering media $W^2/\langle v\rangle^2 \simeq m/M$, where m and M are the masses of an electron and of a scattering center, respectively. Since the maximum value of m/M occurs for hydrogen and it is $\sim 3\times 10^{-4}$, the contribution of each flight with $s>1$ is less than 3×10^{-4} compared to that of the first flight. However the series due to the successive flights gives a contribution which can be of the same order as that of $s=1$. (*A posteriori* one ascertains that this is the case for D_x, while for $D_y=D_z$ the contribution due to the series remains small). Moreover, after a flight, the memory of the direction of the initial velocity is lost to within the small recoil of the collided molecule (or center of scattering). We can therefore consider in detail the first flight only, i.e., we leave unaltered only the first term of (10). For the rest of the series we do not consider the details of the single flights but we summarize their effect by a slow diffusion. In other words we take into account the rapid variations of $\mu=\cos\theta$ in the first flight and then use a Fokker-Planck approximation. Consequently, by (9), (10), and (12), we can write for the transition probability density $\tilde{f}$ [it does not make any difference to use either i (for initial) or 1 (for first flight)]:

$$\begin{aligned}\tilde{f}(v_f,\mu_f,\psi_f,\tau\,|\,v_i,\mu_i,\psi_i)\\ &\simeq v_{0f}^{-2}\delta(v_{0f}(\mathbf{v}_f)-v_{0i}(\mathbf{v}_i))\delta(\mu_{0f}(\mathbf{v}_f)-\mu_{0i}(\mathbf{v}_i))\\ &\quad\times\delta(\psi_{0f}(\mathbf{v}_f)-\psi_{0i}(\mathbf{v}_i))[\mathcal{H}(\tau)-\mathcal{H}(\tau-t_i)]\\ &\quad+\bar{f}(v_f,\mu_f,\tau\,|\,v_i,t_i)\mathcal{H}(\tau-t_i)\ ,\end{aligned} \tag{13}$$

where $\mathbf{v}_i$ and $\mathbf{v}_f$ are the initial and final velocities, respectively, while v_{0i} and v_{0f} are the corresponding velocities immediately after the preceding collisions. Both $\mathbf{v}_{0i}$ (the initial velocity corresponds to the first flight) and $\mathbf{v}_{0f}$ can be expressed as functions of $\mathbf{v}_i$ and $\mathbf{v}_f$ so as to have the same variables as in the left-hand side.

Notice that the "smoothed" transition probability $\bar{f}$ depends neither on ψ_f, because of axial symmetry around $\mathbf{E}$, nor on μ_i and ψ_i because the initial directions are no longer remembered after one collision. On the contrary $\tilde{f}$ depends on ψ_f, ψ_i, and μ_i. In particular $\tilde{f}$ changes abruptly after any collision, while $\bar{f}$ changes appreciably only after many collisions.

IV. GENERALIZED DIFFUSION COEFFICIENTS EXPRESSED BY THE ADDITIONAL TERM b

Before using the transition probability density given by (13) it is convenient to expand its slowly varying part $\bar{f}$ around $t_i=0$, i.e.,

$$\begin{aligned}\bar{f}(v_f,\mu_f,\tau\,|\,v_i,t_i)&=\bar{f}(v_f,\mu_f,\tau-t_i\,|\,v_i,0)\\ &\simeq\bar{f}(v_f,\mu_f,\tau\,|\,v_i)-t_i\left[\frac{\partial\bar{f}}{\partial\tau}\right]_{t_i=0}\ .\end{aligned} \tag{14}$$

Since $\bar{f}$ is a slowly varying function of τ of the kind $\bar{f}\sim\exp(-m\nu t/M)$ the contribution to D due to the second term of (14) turns out to be of the order m/M compared to that due to the first term, so that it can be neglected. For the same reason [to obtain D in (1) and (2) we integrate over τ from 0 to ∞] we can neglect t_i compared to τ in the second term of (13). By these approximations, putting

$$v_i\mu_i=v_{0i}\mu_{0i}+at_i\ , \tag{15a}$$

$$v_f\mu_f=v_{0i}\mu_{0i}+a(t_i+\tau)\ , \tag{15b}$$

substituting (14), (4), (9), and (13) in equation (3), and integrating over ψ_i and ψ_f yields

$$\begin{aligned}\langle v_x(0)v_x(\tau)\rangle&=2\pi\int_0^\infty dv_{0i}\,v_{0i}^2\int_{-1}^1 d\mu_{0i}\,\chi(v_{0i},\mu_{0i})\int_0^\infty dt_i\left[\exp\left[-\int_0^{t_i}d\xi\,\nu_0(\xi)\right]\right](v_{0i}\mu_{0i}+at_i)\\ &\qquad\times(v_{0i}\mu_{0i}+at_i+a\tau)[\mathcal{H}(\tau)-\mathcal{H}(\tau-t_i)]\\ &\quad+4\pi^2\int_0^\infty dv_i\,v_i^2\int_{-1}^1 d\mu_i f(v_i,\mu_i)v_i\mu_i\int_0^\infty dv_f\,v_f^2\int_{-1}^1 d\mu_f\,\bar{f}(v_f,\mu_f,\tau\,|\,v_i)v_f\mu_f\mathcal{H}(\tau)\ .\end{aligned} \tag{16}$$

Let us expand f and $\bar{f}$ in Legendre polynomials

$$f(v_i,\mu_i)=\sum_0^\infty P_s(\mu_i)f_s(v_i)\ , \tag{17a}$$

$$\bar{f}(v_f,\mu_f,\tau\,|\,v_i)=\sum_0^\infty P_s(\mu_f)\bar{f}_s(v_f,\tau\,|\,v_i)\ . \tag{17b}$$

Substituting (17a) and (17b) in (16) and the result in (2), after integration over τ in the first term, and over μ_i and μ_f in the second term, recalling the orthogonality of the Legendre polynomials and the fact that only $P_1(\mu)=\mu$ appears in (16), we obtain

$$\begin{aligned}D_x(\omega)&=2\pi\int_0^\infty dv_{0i}\,v_{0i}^2\int_{-1}^1 d\mu_{0i}\,\chi(v_{0i},\mu_{0i})\int_0^\infty dt_i\left[\exp\left[-\int_0^{t_i}d\xi\,\nu_0(\xi)\right]\right]\\ &\qquad\times\frac{1}{\omega}\left[(v_{0i}^2\mu_{0i}^2+3v_{0i}\mu_{0i}at_i+2a^2t_i^2)\sin(\omega t_i)\right.\\ &\qquad\qquad\left.+\frac{1}{\omega}(v_{0i}\mu_{0i}a+a^2t_i)[\cos(\omega t_i)-1]\right]\\ &\quad+\int d\tau\cos(\omega\tau)\left[\tfrac{16}{9}\pi^2\int_0^\infty dv_i\,v_i^3f_1(v_i)\int_0^\infty dv_f\,v_f^3\bar{f}_1(v_f,\tau\,|\,v_i)-\langle v_x\rangle^2\right]\ .\end{aligned} \tag{18}$$

Let us expand the exponential appearing in (18) with the introduction of an auxiliary term bt and the notation[16,17]

$$\dot{\nu}_0=\frac{d\nu_0}{dv},\quad \ddot{\nu}_0=\frac{d^2\nu_0}{dv^2}\ .$$

We get

$$\exp\left[-\int_0^t d\xi\,\nu_0(\xi)\right]=[\exp(-\nu_0 t-bt)]\exp(bt+R)$$
$$\simeq\left[1+bt-\tfrac{1}{2}at^2\dot{\nu}_0\mu+\tfrac{1}{2}b^2t^2-\tfrac{1}{2}abt^3\dot{\nu}_0\mu+\tfrac{1}{8}a^2t^4\dot{\nu}_0^2\mu^2\right.$$
$$\left.-\tfrac{1}{6}a^2t^3\ddot{\nu}_0\mu^2-\tfrac{1}{6}a^2t^3(1-\mu^2)\frac{\dot{\nu}_0}{v}\right]\exp(-\nu_0 t-bt)\ . \quad (19)$$

We have truncated after the second order in the acceleration a since we want the leading term and no more than the relative second order.[16]

It is also convenient to express the collision source χ as a function of the isotropic component $f_0(v)$ of the usual distribution function $f(v,\mu)$. Still with second-order accuracy we have[10,17]

$$\chi(v,\mu)=\nu_0 f_0-\frac{a\nu_1}{\nu_0-\nu_1}\frac{df_0}{dv}\mu+a^2\left[-\frac{1}{3v^2}\frac{d}{dv}\left[\frac{v^2}{\nu_0-\nu_1}\frac{df_0}{dv}\right]+\frac{\nu_2}{\nu_0-\nu_2}\frac{2v}{3}\frac{d}{dv}\left[\frac{1}{\nu_0-\nu_1}\frac{1}{v}\frac{df_0}{dv}\right]P_2(\mu)\right]\ , \quad (20)$$

where:

$$\nu_0(v)=\frac{1}{2}\int_{-1}^{1}d\mu\,\nu(v,\mu),\quad \nu_1(v)=\frac{1}{2}\int_{-1}^{1}d\mu\,\nu(v,\mu)\mu,\quad \nu_2(v)=\frac{1}{2}\int_{-1}^{1}d\mu\,\nu(v,\mu)P_2(\mu)\ , \quad (21)$$

$\nu(v,\mu)$ being the differential collision frequency and $P_2(\mu)=(3\mu^2-1)/2$ the Legendre second polynomial. Substituting (19) and (20) in (18) gives, still truncating the terms in a^s with $s>2$,

$$D_x(\omega)=D_{x1}(\omega)+D_{x2}(\omega)+D_{x3}(\omega)\ , \quad (22)$$

where D_{x1} is the contribution due to the main term $\nu_0 f_0$ of (20) substituted in the first term of (18), D_{x2} comes from the other terms of (20) and the first term of (18), and D_{x3} is the second term of (18). We get, writing v instead of v_0 for simplicity,

$$D_{x1}(\omega)=\frac{4\pi}{3}\int_0^\infty dv\,v^4\nu_0 f_0\left[\frac{1}{H^2}+2b\frac{k}{H^4}+\left[b^2+\frac{12a^2}{v^2}\right]\frac{3k^2-\omega^2}{H^6}-12\left[\frac{47}{15}\frac{a^2}{v}\dot{\nu}_0+\frac{1}{5}a^2\ddot{\nu}_0\right]k\frac{k^2-\omega^2}{H^8}\right.$$
$$\left.+\tfrac{9}{5}a^2\dot{\nu}_0^2\frac{5k^4-10\omega^2k^2+\omega^4}{H^{10}}+\frac{3a^2}{\omega^2v^2}\frac{k^2-\omega^2}{H^4}-\frac{a^2}{\omega^2}\dot{\nu}_0 k\frac{k^2-3\omega^2}{vH^6}-\frac{3a^2}{v^2\omega^2k^2}+\frac{a^2}{v\omega^2k^3}\dot{\nu}_0\right]\ , \quad (23)$$

where use is made of the integrals (23)–(28) of Ref. 10 (Cavalleri) in which

$$k=\nu_0+b \quad (24)$$

and

$$H^2=k^2+\omega^2\ . \quad (25)$$

Moreover,

$$D_{x2}(\omega)=\tfrac{4}{3}\pi a^2\int_0^\infty dv\,v^4\left\{\frac{\nu_1}{\nu_0-\nu_1}\frac{df_0}{dv}\left[\frac{3}{5H^6}(3k^2-\omega^2)\dot{\nu}_0-\frac{6k}{vH^4}-\frac{1}{v\omega^2}\left[\frac{k}{H^2}-\frac{1}{k}\right]\right]\right.$$
$$\left.+\left[\frac{4}{15}\frac{v\nu_2}{\nu_0-\nu_2}\frac{d}{dv}\left[\frac{1}{\nu_0-\nu_1}\frac{1}{v}\frac{df_0}{dv}\right]-\frac{1}{3v^2}\frac{d}{dv}\left[\frac{v^2}{\nu_0-\nu_1}\frac{df_0}{dv}\right]\right]\frac{1}{H^2}\right\}\ . \quad (26)$$

$D_{x3}(\omega)$ is given by the second term of (18).

The expression for the transversal diffusion coefficient $D_y=D_z$ is obtained by substituting $v\cos\psi(1-\mu^2)^{1/2}$ instead of $v\mu$ in (3). The integration over ψ cancels the contribution coming from the second term of (13). By the same procedure leading to (18) we get

$$D_y=\pi\int_0^\infty dv_{0i}\,v_{0i}^4\int_{-1}^{1}d\mu_{0i}\chi(v_{0i},\mu_{0i})(1-\mu_{0i}^2)\int_0^\infty dt\frac{1}{\omega}\sin(\omega t)\exp\left[-\int_0^t d\xi\,\nu_0(\xi)\right]\ . \quad (27)$$

If we substitute (19) and (20) in (27) we get

$$D_y(\omega)=D_z(\omega)=D_{y1}(\omega)+D_{y2}(\omega)\ , \quad (28)$$

where, still writing v instead of v_0,

$$D_{y1}(\omega)=\frac{4\pi}{3}\int_0^\infty dv\, v^4\nu_0 f_0\left[\frac{1}{H^2}+\frac{2kb}{H^4}+b^2\frac{3k^2-\omega^2}{H^6}+\frac{3}{5}a^2\dot{\nu}_0^2\frac{5k^4-10\omega^2k^2+\omega^4}{H^{10}}-\frac{4}{5}a^2\left[\ddot{\nu}_0+\frac{4}{v}\dot{\nu}_0\right]k\frac{k^2-\omega^2}{H^8}\right], \tag{29}$$

$$D_{y2}^{(\omega)}=\frac{4}{3}\pi a^2\int_0^\infty dv\, v^4\left\{\frac{\nu_1\dot{\nu}_0}{\nu_0-\nu_1}\frac{df_0}{dv}\frac{3k^2-\omega^2}{5H^6}-\left[\frac{2}{15}\frac{v\nu_2}{\nu_0-\nu_2}\frac{d}{dv}\left[\frac{1}{\nu_0-\nu_1}\frac{1}{v}\frac{df_0}{dv}\right]+\frac{1}{3v^2}\frac{d}{dv}\left[\frac{v^2}{\nu_0-\nu_1}\frac{df}{dv}\right]\right]\frac{1}{H^2}\right\}. \tag{30}$$

V. ORDINARY DIFFUSION COEFFICIENTS EXPRESSED BY THE ADDITIONAL TERM b

For $\omega\to 0$ the generalized diffusion coefficients become the ordinary diffusion coefficients which we write here

$$D_x=D_{x1}+D_{x2}+D_{x3}\ , \tag{31}$$

where

$$D_{x1}=\frac{4}{3}\pi\int_0^\infty dv\, v^4\nu_0 f_0\left[\frac{1}{(\nu_0+b)^2}+\frac{2b}{(\nu_0+b)^3}+\frac{3}{(\nu_0+b)^4}\left[b^2+\frac{9}{v^2}a^2\right]-\frac{2}{(\nu_0+b)^5}\left[\frac{79}{5}\frac{a^2}{v}\dot{\nu}_0+\frac{6}{5}a^2\ddot{\nu}_0\right]+\frac{9a^2\dot{\nu}_0^2}{(\nu_0+b)^6}\right], \tag{32}$$

$$D_{x2}=\frac{4}{3}\pi a^2\int_0^\infty dv\, v^4\left\{\frac{\nu_1}{\nu_0-\nu_1}\left[\frac{9\dot{\nu}_0}{5(\nu_0+b)^4}-\frac{5}{v(\nu_0+b)^3}\right]\frac{df_0}{dv}+\frac{1}{(\nu_0+b)^2}\left[\frac{4}{15}\frac{v\nu_2}{\nu_0-\nu_2}\frac{d}{dv}\left[\frac{1}{\nu_0-\nu_1}\frac{1}{v}\frac{df_0}{dv}\right]-\frac{1}{3v^2}\frac{d}{dv}\left[\frac{v^2}{\nu_0-\nu_1}\frac{df_0}{dv}\right]\right]\right\}, \tag{33}$$

$$D_{x3}=\int_0^\infty d\tau\left[\frac{16\pi^2}{9}\int_0^\infty dv_i\, v_i^3 f_1(v_i)\int_0^\infty dv_f\, v_f^3\bar{f}_1(v_f,\tau\,|\,v_i)-\langle v_x\rangle^2\right]. \tag{34}$$

Similarly

$$D_y=D_{y1}+D_{y2}\ , \tag{35}$$

where

$$D_{y1}=\frac{4}{3}\pi\int_0^\infty dv\, v^4\nu_0 f_0\left[\frac{1}{(\nu_0+b)^2}+\frac{2b}{(\nu_0+b)^3}+\frac{3b^2}{(\nu_0+b)^4}+\frac{3a^2\dot{\nu}_0^2}{(\nu_0+b)^6}-\frac{4a^2}{5(\nu_0+b)^5}\left[\ddot{\nu}_0+\frac{4}{v}\dot{\nu}_0\right]\right], \tag{36}$$

$$D_{y2}=\frac{4}{3}\pi a^2\int_0^\infty dv\, v^4\left\{\frac{\nu_1\dot{\nu}_0}{\nu_0-\nu_1}\frac{df_0}{dv}\frac{3}{5(\nu_0+b)^4}-\frac{1}{(\nu_0+b)^2}\left[\frac{2}{15}\frac{v\nu_2}{\nu_0-\nu_2}\frac{d}{dv}\left[\frac{1}{\nu_0-\nu_1}\frac{1}{v}\frac{df_0}{dv}\right]+\frac{1}{3v^2}\frac{d}{dv}\left[\frac{v^2}{\nu_0-\nu_1}\frac{df_0}{dv}\right]\right]\right\}. \tag{37}$$

We have thus the desired expressions (31)–(37) to within the auxiliary term b which is explicitly obtained in the next section.

As shown in Ref. 16, the terms multiplied by a^2 are of the second order in $(m/M)^{1/2}$ with respect to the leading terms, given by (32) for the longitudinal diffusion coefficient, and by (36) for the transversal diffusion coefficient. Since m is the electron mass and M the molecule mass, the maximum ratio m/M occurs for hydrogen and it is equal to 2.72×10^{-4}, while for other gases (or atoms in a lattice of a solid) the ratio is still smaller. It seems that also D_{x3} is of second order because f_1 and $\bar{f}_1$ are each proportional to a. But the decay time of $\bar{f}_1$ is of the order $(M/m)\nu_0^{-1}$ and the integration over time in (35) introduces a factor M/m which is just the inverse of what a^2 introduces. Consequently, D_{x3} is of the order of the leading term.

By our procedure and approximations, we estimate the errors introduced in the second-order terms to 30%, so that those ones relative to the leading terms are less than one part in 10^4. Moreover, in all the practical cases, $\nu_2<\nu_1\ll\nu_0$ so that the first two terms of (33) and (37) can be neglected without modifying the relative error of 10^{-4}.

The second-order terms are mainly due, in the integration over v, to the contribution of the integrals from zero to a small value compared to $\langle v\rangle$. This is due to the fact that, for $v \to 0$, the collision frequency $\nu_0 = N\sigma v$ is proportional to v (because σ tends to a constant value). Now, if we neglected b, these second-order terms would diverge for $v \to 0$ and, with $b \neq 0$, they give the largest part of their contribution during the integration from 0 to a small value of the order $b/N\sigma$.

We could even neglect completely the terms in a^2, but we preserve them only to calculate b so as to develop a correct expression (to within 10^{-4}) for the leading terms.

A fourth-order expansion in the acceleration would give a better approximation even for the second-order term because there is a kind of back flow which slightly modifies the coefficients of the second-order terms. Fortunately, without carrying out the extremely long calculation, the comparison with the rigorous expansion of (18) in the case $\nu_0 \propto v$ suggests such small modifications because logarithmic terms have to vanish. This will be done in the next section so that in the conclusions the convenient expressions to be used by experimentalists will be given.

VI. CALCULATION OF THE ADDITIONAL PARAMETER *b* AND MODIFICATION OF THE SECOND-ORDER COEFFICIENTS

The generalized diffusion coefficients obtained in the preceding section present no divergences for $\omega \neq 0$ even if we put $b = 0$. However, errors are present which tend to infinity for $\omega \to 0$ if we leave $b = 0$. For nonzero molecule temperature T the collision frequency never vanishes and therefore no divergences arise in the diffusion coefficients. However, for small T values big errors are present which remain large even for $T \simeq 300$ K. The divergences (or at least the big errors in D_x and $D_y = D_z$) arise when ν_0 vanishes. This occurs for $v \to 0$ because the collision cross section σ approaches a finite value so that

$$\nu_0 = N\sigma v = \alpha v \ , \tag{38}$$

where N is the molecule concentration and $\alpha = N\sigma$ a constant.

We will therefore leave b in our expressions and evaluate an explicit expression for it. In practice we can assume (38) as the expression of ν for $v \to 0$, and calculate the corresponding b value. Indeed, as shown in Ref. 16, b can be written in the form

$$b = za/v \ , \tag{39}$$

where z is a dimensionless factor we evaluate by the following procedure.

We take the first term of (18) with $\omega = 0$ and with χ given by the first term of (20), i.e., $\chi = \nu_0 f_0$ in which we use $\nu_0 = \alpha v$. We obtain

$$D_{x1}^{\text{rig}} = 2\pi\alpha \int_0^\infty dv\, v^3 f_0(v) \int_{-1}^1 d\mu \int_0^\infty dt (v^2\mu^2 t + 5v\mu a t^2/2 + 3a^2 t^3/2) \exp\left[-\alpha \int_0^t d\xi\, v(\xi)\right] . \tag{40}$$

The corresponding approximate expression to (40) is Eq. (32) in which the truncated expansion (19) has been used. If we put $b = 0$ in (32), the resulting expression is the most affected by errors. That is why we chose (40), and the corresponding (32), to obtain z.

For convenience we take $f_0 = \exp(-h^2 v^2)$. If we put $a = 0$ in both (32) and (40) we obtain the same leading term. In order to calculate the second-order term we put

$$x = hv, \quad y = \alpha t/h, \quad c = ah^2/\alpha \ , \tag{41}$$

expand the exponent of the exponential appearing in (40), and subtract the leading term. We get

$$\begin{aligned} D_{x1}^{\text{rig}} - D_{x1}(a=0) = {} & \frac{2\pi}{\alpha h^4} \int_0^\infty dx\, x^3 \exp(-x^2) \\ & \times \int_{-1}^1 d\mu \int_0^\infty dy (x^2\mu^2 y \exp(-xy)\{-1 + \exp[-\mu c y^2/2 - (1-\mu^2)c^2 y^3/6x]\} \\ & + (5xy^2\mu c/2 + 3c^2 y^3/2)\exp[-xy - \mu c y^2/2 - (1-\mu^2)c^2 y^3/6x]) \\ = {} & \frac{2\pi}{\alpha h^4} 5.831 c^2 \ , \end{aligned} \tag{42}$$

where the result has been obtained by a computer.

The approximate expression D_{x1}^{ap} is obtained from (32) by putting

$$\begin{aligned} & b = za/v, \quad x = h^2 v^2, \quad f_0[v(x)] = \exp(-x) \ ; \\ & c = ah^2/\alpha, \quad S = x + zc \ . \end{aligned} \tag{43}$$

We get

$$D_{x1}^{\text{ap}} = \frac{2\pi}{3\alpha h^4} \int_0^\infty dx \exp(-x) \left[\frac{x^3}{S^2} + \frac{2zcx^3}{S^3} + \frac{3(z^2+9)c^2x^3}{S^4} - \frac{158c^2x^4}{5S^5} + \frac{9c^2x^5}{S^6} \right] . \tag{44}$$

The first two terms inside the large parentheses of (44) are

$$\frac{x^3}{S^2}+\frac{2zcx^3}{S^3}=x-\frac{3z^2c^2}{S}+\frac{5z^3c^3}{S^2}-\frac{2z^4c^4}{S^3}\ , \tag{45}$$

and the first term in the right-hand side of (45) gives the leading term.

The integrals appearing in (44) are calculated by the method shown in Appendix E of Ref. 16 and we get

$$D_{x1}^{\rm ap}=\frac{2\pi}{3\alpha h^4}\left[1-3z^2c^2(-\gamma-\ln cz)+5c^3z^3\frac{1}{cz}-2z^4c^4\frac{1}{2c^2z^2}+3c^2(z^2+9)(-\tfrac{11}{6}-\gamma-\ln cz)\right.$$
$$\left.-\tfrac{158}{5}c^2(-\tfrac{25}{12}-\gamma-\ln cz)+9c^2(-\tfrac{137}{60}-\gamma-\ln cz)\right]\ , \tag{46}$$

where γ is the Euler constant.

The logarithmic terms, absent in (42), do not vanish exactly in (46) (only those multiplied by z^2 cancel each other exactly). This is due to our truncation of the expansion and this fact is suggestive of what the successive terms would give. We therefore change some coefficients so as to obtain an exact cancellation of the logarithmic terms. The modified expression which indirectly takes into account a wider expansion and replaces (44), is

$$D_{x1}^{\rm ap}=\frac{2\pi}{3\alpha h^4}\int_0^\infty dx\,\exp(-x)\times\left[\frac{x^3}{S^2}\frac{2zcx^3}{S^3}+\frac{3(z^2+21)c^2x^3}{S^4}-\frac{50c^2x^4}{S^5}-\frac{13c^2x^5}{S^6}\right]. \tag{47}$$

An expansion like (46) gives, after simplification

$$D_{x1}^{\rm ap}=\frac{2\pi}{3\alpha h^4}[1+c^2(18.34-1.5z^2)]\ . \tag{48}$$

Equating $D_{x1}^{\rm ap}-D_{x1}(a=0)$ to (42) yields

$$z=z_{\parallel}=0.751\ . \tag{49}$$

The rigorous expression of the first term of the transversal coefficient is obtainable from (27) by putting $\omega=0$. Still using $\chi=\alpha v\exp(-h^2v^2)$ we get

$$D_{y1}^{\rm rig}=\pi\alpha\int_0^\infty dv\,v^5f_0(v)\times\int_{-1}^{1}d\mu(1-\mu^2)\times\int_0^\infty dt\,t\exp\left[-\alpha\int_0^t d\xi\,v(\xi)\right]. \tag{50}$$

Again using (41) we transform (50) in the following form convenient for the computer

$$D_{y1}^{\rm rig}-D_{y1}(a=0)=\frac{\pi}{\alpha h^4}\int_0^\infty dx\,x^5\exp(-x^2)\times\int_{-1}^{1}d\mu(1-\mu^2)\int_0^\infty dy\,y\exp(-xy)\times[-1+\exp(-\mu cy^2/2-(1-\mu^2)c^2y^3/6x)]$$
$$=(\pi/\alpha h^4)(-0.595c^2)\ . \tag{51}$$

The approximate expression $D_{y1}^{\rm ap}$ is obtained from (36) by the use of (43)

$$D_{y1}^{\rm ap}=\frac{2\pi}{3\alpha h^4}\int_0^\infty dx\,\exp(-x)\left[\frac{x^3}{S^2}+\frac{2zcx^3}{S^3}+\frac{3z^2c^2x^3}{S^4}-\frac{16c^2x^4}{5S^5}+\frac{3c^2x^5}{S^6}\right]. \tag{52}$$

With the use of (45) and the method exposed in Appendix E of Ref. 16 we obtain

$$D_{y1}^{\rm ap}=\frac{2\pi}{3\alpha h^4}\left[1-3z^2c^2(-\gamma-\ln cz)+5c^3z^3\frac{1}{cz}-2z^4c^4\frac{1}{2z^2c^2}+3c^2z^2(-\tfrac{11}{6}-\gamma-\ln cz)\right.$$
$$\left.-\tfrac{16}{5}c^2(-\tfrac{25}{12}-\gamma-\ln cz)+3c^2(-\tfrac{137}{60}-\gamma-\ln cz)\right]. \tag{53}$$

Here also the logarithmic terms do not vanish completely and a small modification of one coefficient is required. Precisely we change the coefficient $\frac{16}{5}$ of the third term inside the large parentheses of (53) by 3. However it is still simpler and more convenient for the case in which a Ramsauer effect occurs, to neglect the last two terms. We get

$$D_{y1}^{\rm ap}=\frac{2\pi}{3\alpha h^4}\int_0^\infty dx\,\exp(-x)\left[\frac{x^3}{S^2}+\frac{2zcx^3}{S^3}+\frac{3z^2c^2x^3}{S^4}-\frac{3c^2x^4}{S^5}+\frac{3c^2x^5}{S^6}\right]$$

$$=\frac{2\pi}{3\alpha h^4}[1-c^2(0.6+1.5z^2)]\ . \tag{54}$$

Equating $D_{y1}^{\rm ap}-D_{y1}(a=0)$ to (53) yields

$$z=z_\perp=0.442\ . \tag{55}$$

Still more compact and simple expressions can be obtained which approximate in an excellent way the rigorous expressions. For the longitudinal-diffusion coefficient it is

$$D_{x1}^{\rm ap}=\frac{2\pi}{3\alpha h^4}\int_0^\infty dx\,\exp(-x)\left[\frac{x^3}{S^2}+\frac{2zcx^3}{S^3}+\frac{3z^2c^2x^3}{S^4}+\frac{54c^2x^2}{S^3}\left[1-\frac{x}{S}\right]\right]\ . \tag{56}$$

Its expansion to second order yields

$$D_{x1}^{\rm ap}=\frac{2\pi}{3\alpha h^4}[1+c^2(18-1.5z^2)]\ . \tag{57}$$

Equating $D_{x1}^{\rm ap}-D_{x1}(a=0)$ to (42) gives

$$z_\parallel=0.581\ . \tag{58}$$

For $D_{y1}^{\rm ap}$ it is sufficient to retain only the first three terms of the integrand

$$D_{y1}^{\rm ap}=\frac{2\pi}{3\alpha h^4}\int_0^\infty dx\,\exp(-x)\left[\frac{x^3}{S^2}+\frac{2zcx^3}{S^3}+\frac{3z^2c^2x^3}{S^4}\right]$$

$$=\frac{2\pi}{3\alpha h^4}(1-1.5c^2z^2)\ . \tag{59}$$

Equating $D_{y1}^{\rm ap}-D_{y1}(a=0)$ to (51) yields

$$z=z_\perp=0.771\ . \tag{60}$$

VII. EVALUATION OF b WHEN THERE IS A RAMSAUER EFFECT

With the aim of calculating $b=za/v$, i.e., z, when there is a Ramsauer effect at $v=v'$, we schematize it as

$$\begin{aligned}&\nu_0(v)=A=\text{const}\quad\text{for } v\le 0.9v' \text{ and } v\ge 1.1v'\ ,\\ &\nu_0(v)=B\,|v-v'|=10A\,|v-v'|/v'\\ &\qquad\text{for } 0.9v'\le v\le 1.1v'\ .\end{aligned} \tag{61}$$

We take again the first term of (18) with $\omega=0$ and $\chi=\nu_0 f_0=\nu_0\exp(-hv)$, thus obtaining, with the use of (61),

$$\begin{aligned}D_{x1}^{\rm rig}=&\,2\pi A\left[\int_0^{0.9v'}dv\,v^2e^{-hv}+\int_{1.1v'}^\infty dv\,v^2e^{-hv}\right]\\ &\times\int_{-1}^1 d\mu\int_0^\infty dt(v^2\mu^2t+2.5v\mu at^2+1.5a^2t^3)\exp(-At)+(20\pi A/v')\\ &\times\int_{0.9v'}^{1.1v'}dv\,v^2\,|v-v'|\,e^{-hv}\\ &\times\int_{-1}^1 d\mu\int_0^\infty dt(v^2\mu^2t+2.5v\mu at^2+1.5a^2t^3)\\ &\times\exp\left[-(10A/v')\int_0^t d\xi\,|(v^2+a^2\xi^2+2a\xi v\mu)^{1/2}-v'|\right]\ .\end{aligned} \tag{62}$$

The first two integrals are easily solved and we obtain, by the use of (41)

$$\int_0^{0.9v'}(\cdots)+\int_{1.1v'}^\infty(\cdots)=\frac{2\pi}{Ah^5}(15.9505+3467.7824c^2)\ . \tag{63}$$

To calculate the contribution around the Ramsauer effect (i.e., for $0.9v'<v<1.1v'$) we put, similarly to (41) and with the additional convenient assumption $hv'=1$,

$$x=hv,\quad hv'=1,\quad y=10At,\quad c=ah/10A\ . \tag{64}$$

We get

$$\int_{0.9v'}^{1.1v'}(\cdots)=\frac{2\pi}{10Ah^5}\int_{0.9}^{1.1}dx\,x^2\,|x-1|\,e^{-x}\int_{-1}^1 d\mu\int_0^\infty dy(x^2y\mu^2+2.5cx\mu y^2+1.5c^2y^3)\times\exp\left[-\int_0^y dy'\,|(x^2+c^2y'^2+2cxy'\mu)^{1/2}-1|\right]\ . \tag{65}$$

The approximate expression can be obtained by expanding the exponential in (62) and introducing the auxiliary term b, and also by the use of (61) and (64). Here also we find a convenient compact expression which reads

$$D_{x\|}^{\mathrm{ap}}=\frac{2\pi}{Ah^5}\left[\frac{2}{3}\left[\int_0^{0.9}dx\,x^6e^{-x}+\int_{1.1}^{\infty}dx\,x^6e^{-x}\right]\left[\frac{1}{H^2}+\frac{20zc}{H^3}+\frac{300c^2z^2}{H^4}-2.7\frac{(10c)^{1.962}}{H^{4.038}}\right]\right.$$
$$\left.+\frac{1}{15}\int_{0.9}^{1.1}dx\,x^6e^{-x}|x-1|\left[\frac{1}{F^2}+\frac{2zc}{F^3}+\frac{3c^2z^2}{F^4}-2.7\frac{c^{1.962}}{F^{4.038}}\right]\right], \quad (66)$$

where

$$H=x+10zc,\quad F=x\,|x-1|+zc\ . \quad (67)$$

The first two integrals of (66) give the same result as (63). The comparison of (65) with (66) regards only $\int_{0.9}^{1.1}$. The calculations, performed by a computer, lead to

$$z_{R\|}=0.5\ . \quad (68)$$

For the transversal diffusion coefficient we get in a similar way

$$D_{y\|}^{\mathrm{rig}}=\frac{\pi}{Ah^5}\left[\int_{-1}^{1}d\mu(1-\mu^2)\int_0^{\infty}dy\,0.01ye^{-y/10}\left[\int_0^{0.9}dx\,x^4e^{-x}+\int_{1.1}^{\infty}dx\,x^4e^{-x}\right]\right.$$
$$\left.+0.1\int_{0.9}^{1.1}dx\,x^4|x-1|e^{-x}\int_{-1}^{1}d\mu(1-\mu^2)\int_0^{\infty}dy\,y\exp\left[-\int_0^{y}dy'\,|(c^2y'^2+x^2+2\mu cxy')^{1/2}-1|\right]\right]. \quad (69)$$

The compact, convenient expression of $D_{y\|}^{\mathrm{ap}}$ is similar to (66) and reads

$$D_{y\|}^{\mathrm{ap}}=\frac{4\pi}{3Ah^5}\left[\int_0^{0.9}dx\,x^6e^{-x}+\int_{1.1}^{\infty}dx\,x^6e^{-x}\right]\left[\frac{1}{H^2}+\frac{20zc}{H^3}+\frac{300c^2z^2}{H^4}-2.3\frac{(10c)^{1.962}}{H^{4.038}}\right]$$
$$+\frac{2\pi}{15Ah^5}\int_{0.9}^{1.1}dx\,x^6e^{-x}|x-1|\left[\frac{1}{F^2}+\frac{2zc}{F^3}+\frac{3c^2z^2}{F^4}-2.3\frac{c^{1.962}}{F^{4.038}}\right]. \quad (70)$$

Here also the first two terms give the same result (up to c^2 terms) of the corresponding terms of the rigorous expression. The comparison of the contribution $\int_{0.9}^{1.1}$ leads to

$$z_{R\perp}=0.5\ . \quad (71)$$

VIII. GENERAL RESULTS FOR THE EXPERIMENTALISTS IN THE CASE OF RAMSAUER EFFECTS

We summarize our results to be used by experimentalists when $\nu_2<\nu_1\ll\nu_0$, where ν_0, ν_1, and ν_2 are given by (21), $\nu(v,\mu)$ being the differential collision frequency. They are convenient because they are expressed by integrals over a single variable which is the velocity v. When there is a Ramsauer effect at a speed v' the integral is conveniently split into three parts. The explicit expression for the transversal diffusion coefficient is

$$D_{y\|}^{\mathrm{ap}}=\frac{4\pi}{3}\int_0^{0.9v'}dv\,v^4\nu_0f_0\left[\frac{1}{(\nu_0+b_\perp)^2}+\frac{2b_\perp}{(\nu_0+b_\perp)^3}+\frac{3b_\perp^2}{(\nu_0+b_\perp)^4}\right]$$
$$+\frac{4\pi}{3}\int_{0.9v'}^{1.1v'}dv\,v^4\nu_0f_0\left[\frac{1}{(\nu_0+b_R)^2}+\frac{2b_R}{(\nu_0+b_R)^3}+\frac{3b_R^2}{(\nu_0+b_R)^4}-2.3\left[\frac{\nu_0v'^2}{|v-v'|}\right]^{0.076}\frac{a^{1.962}}{(\nu_0+b_R)^{4.038}v^{2.038}}\right]$$
$$+\frac{4\pi}{3}\int_{1.1v'}^{\infty}dv\,v^4\nu_0^{-1}f_0[1+3a^2\dot{\nu}_0^2\nu_0^{-4}-\tfrac{4}{5}a^2\nu_0^{-3}(\ddot{\nu}_0+4\dot{\nu}_0/v)]\ , \quad (72)$$

where

$$\dot{\nu}_0=d\nu_0/dv,\quad \ddot{\nu}_0=d^2\nu_0/dv^2;\quad b_\perp=0.771a/v\ \text{ with } a=eE/m;\quad b_R=0.5a/v\ . \quad (73)$$

In the last integral, where divergences never occur, we have put $b=0$ and kept the original coefficients appearing in (36). The explicit expression of the longitudinal-diffusion coefficient is

$$D_{x1}^{\rm ap} = \frac{4\pi}{3}\int_0^{0.9v'} dv\, v^4 \nu_0 f_0 \left[\frac{1}{(\nu_0+b_\parallel)^2}+\frac{2b_\parallel}{(\nu_0+b_\parallel)^3}+\frac{3b_\parallel^2-54a^2v^{-2}}{(\nu_0+b_\parallel)^4}+\frac{54a^2v^{-2}}{\nu_0(\nu_0+b_\parallel)^3}\right]$$
$$+\frac{4\pi}{3}\int_{0.9v'}^{1.1v'} dv\, v^4\nu_0 f_0\left[\frac{1}{(\nu_0+b_R)^2}+\frac{2b_R}{(\nu_0+b_R)^3}+\frac{3b_R^2}{(\nu_0+b_R)^4}-2.7\left[\frac{\nu_0 v'^2}{|v-v'|}\right]^{0.076}\frac{a^{1.962}}{(\nu_0+b_R)^{4.038}v^{2.038}}\right]$$
$$+\frac{4\pi}{3}\int_{1.1v'}^{\infty} dv\, v^4\nu_0^{-1} f_0\left[1+\frac{27a^2}{v^2\nu_0^2}-\frac{12a^2}{5\nu_0^3}\left[\frac{79}{6v}\dot\nu_0+\ddot\nu_0\right]+\frac{ga^2\dot\nu_0^2}{\nu_0^4}\right]$$
$$+\int_0^\infty d\tau\left[\frac{16\pi^2}{9}\int_0^\infty dv_i\, v_i^3 f_1(v_i)\int_0^\infty dv_f\, v_f^3\bar f_1(v_f,\tau\,|\,v_i)-\langle v_x\rangle^2\right], \quad (74)$$

where

$$b_\parallel=0.581a/v,\quad b_R=0.5a/v\ . \quad (75)$$

In both (72) and (74) the distribution function of the speed v is given by the Chapman-Cowling expression

$$f_0=A^*\exp\left[-\int_0^v dv\frac{v}{(kt/m)+\frac{1}{3}Mm^{-1}a^2(\nu_0-\nu_1)^{-2}}\right], \quad (76)$$

where A^* is a normalization constant, k the Boltzmann constant, T the absolute temperature, M and m the masses of the gas molecule and of the electron, respectively.

In (74) $f_1(v)$ is the first anisotropic component in the two terms Legendre expansion of the distribution function and is related to $f_0(v)$ by

$$f_1(v)=-\frac{a}{\nu_0-\nu_1}\frac{df_0}{dv}\ . \quad (77)$$

What remains to be calculated is the transition probability $\bar f_1(v_f,\tau\,|\,v_i)$ which requires the solution of the Boltzmann equation in the Fokker-Planck approximation.

Consequently, the transversal-diffusion coefficient given by (72) is completely explicit and needs no solutions of other equations, while the longitudinal-diffusion coefficient requires the Green function $\bar f_1(v_f,\tau\,|\,v_i)$ to be obtained from the Boltzmann equation.

The last term of (74) gives the main difference between the transversal- and longitudinal-diffusion coefficients and, when generalized, represents an additional noise which we suppose can give the famous and still unexplained $1/f$ noise.

IX. SOME NUMERICAL EXAMPLES

We consider the case of gas molecules behaving as rigid spheres, i.e., a collision frequency of the kind $\nu_0=N\sigma v$. This behavior is common to all gases for $v\to 0$ as already discussed in Sec. 6, Eq. (38), and for neon in an important wide range of electron speeds where the cross section is $\sigma\simeq 2\times 10^{-16}$ cm^2. The relevant distribution function, Eq. (76), with the use of the dimensionless field parameters[2]

$$v_0^2=kT/m,\quad x=v/v_0,\quad \epsilon^2=m/M,\quad \mathcal{E}=eE/(N\epsilon\sigma kT)\ , \quad (78)$$

becomes

$$f_0(v,T)=v_0^{-3}f_0'(x,v_0,\mathcal{E})\ , \quad (79)$$

with

$$f_0'(x,v_0,\mathcal{E})=\tilde A v_0^{-3}\exp\left[-\int_0^x dx\frac{x^3}{x^2+\mathcal{E}^2/3}\right]$$
$$=A\exp(-x^2/2)(1+3x^2/\mathcal{E}^2)^{\mathcal{E}^2/6}\ , \quad (80)$$

where the normalization constant is obtained from

$$1=4\pi\int_0^\infty dx\, x^2 f_0'(x,v_0,\mathcal{E})\ . \quad (81)$$

We calculate the transversal-diffusion coefficient by Eq. (72) without Ramsauer effect, i.e., dropping the second term. The limits of the first and third integral are rather arbitrary and we use $0.1v_0$ since the use of the additional parameter b is important for low speeds only. We also introduce a dimensionless diffusion coefficient $d_\perp$ which, with the use of Eq. (78) and (79), reads

$$d_\perp=N\sigma(m/kT)^{1/2}D_\perp$$
$$=\frac{4\pi}{3}\left\{\int_0^{0.1}dx\, x^7 f_0'(x,v_0,\mathcal{E})\left[\frac{1}{(x^2+0.771\epsilon\mathcal{E})^2}+\frac{1.542\epsilon\mathcal{E}}{(x^2+0.771\epsilon\mathcal{E})^3}+\frac{1.7833(\epsilon\mathcal{E})^2}{(x^2+0.771\epsilon\mathcal{E})^4}\right]\right.$$
$$\left.+\int_{0.1}^\infty dx\, x^3 f_0'(x,v_0,\mathcal{E})\left[1-\frac{1}{5}\left[\frac{\epsilon\mathcal{E}}{x^2}\right]^2\right]\right\}. \quad (82)$$

If we neglect the second order corrections, Eq. (82) reduces to that used by Skullerud[2] and Parker and Lowke,[3] which is

$$d_1^0=\frac{4\pi}{3}\int_0^\infty dx\, x^3 f_0'(x,v_0,\mathcal{E})\ . \tag{83}$$

The numerical differences between the results obtained from Eq. (82) by a computer and those obtained from Eq. (83) are of order $\epsilon^2=m/M\simeq 10^{-4}$, almost two orders beyond the present experimental accuracy. Our simple formulas are in this case ready for future more accurate experiments.

X. CONCLUSIONS

By the method of the mean free path we have found explicit expressions for the generalized diffusion coefficients $D_x(\omega)$ and $D_y(\omega)$. The application of these expressions, given by Eqs. (18), (22), (23), (26), (28) and (30), to the electric noise is left to a future paper. It is interesting that the mean-free-path method, together with the use of the velocity correlations, does not need the density gradient expansion and can give directly $D_x(\omega)$, from which the ordinary longitudinal coefficient $D_x=D_x(\omega=0)$ is immediately obtained.

In our method an auxiliary term b is introduced which takes into account the variation of the collision frequency $\nu_0(v)$ during a free path because of the electron velocity variation due to the external electric field **E**. This term b removes therefore the unphysical divergence that arises in a hypothetical case of a Ramsauer minimum at a speed v_0' such that $\nu_0(v_0')=0$. Just in this extreme case we have performed numerical calculations in Sec. VII by using the rigorous expression (which never diverges) and the approximate expansion (in which b is introduced) which is the useful, final, explicit expression. By the chosen $b=0.5a/v$ (where $a=eE/m$), the differences between the two expressions are of m/M order, where m and M are the electron and molecule mass, respectively. Consequently, in the real cases where $\nu(v_0')\neq 0$ although small, we guarantee the same approximation of m/M order, while the best values in the literature present discrepancies with the Monte Carlo method up to 6%.

The explicit expressions for the diffusion coefficients are given in Sec. VIII by Eqs. (72)–(75).

When there is no Ramsauer effect, our results, although less important, have been numerically compared in Sec. IX with standard results as obtained by Skullerud.[2] The discrepancies are still of m/M order and it is with this accuracy that we guarantee our expressions that we still report here in the most compact form for the convenience of experimentalists.

By use of Eqs. (36), (37), and the more compact expression (59), we have for the transversal-diffusion coefficient

$$D_y=\frac{4}{3}\pi\int_0^\infty dv\, v^4\left\{\nu_0 f_0\left[\frac{1}{(\nu_0+b)^2}+\frac{2b}{(\nu_0+b)^3}+\frac{3b^2}{(\nu_0+b)^4}\right]+\frac{a^2}{5}\frac{df_0}{dv}\frac{3\nu_1\dot\nu_0}{(\nu_0-\nu_1)(\nu_0+b)^4}\right.$$
$$\left.-\frac{a^2}{(\nu_0+b)^2}\left[\frac{2v\nu_2}{15(\nu_0-\nu_2)}\frac{d}{dv}\left[\frac{1}{\nu_0-\nu_1}\frac{1}{v}\frac{df_0}{dv}\right]+\frac{1}{3v^2}\frac{d}{dv}\left[\frac{v^2}{\nu_0-\nu_1}\frac{df_0}{dv}\right]\right]\right\}, \tag{84}$$

where $a=eE/m$ is the acceleration due to the external field E, and

$$b=0.771a/v\ , \tag{85}$$

$$\nu_0=\nu_0(v)=\frac{1}{2}\int_{-1}^{1}d\mu\,\nu(v,\mu)\ , \tag{86}$$

where $\mu=\mathbf{a}\cdot\mathbf{v}/(av)$ and $\nu(v,\mu)$ is the differential collision frequency. Moreover

$$\nu_1=\nu_1(v)=\frac{1}{2}\int_{-1}^{1}d\mu\,\nu(v,\mu)\mu\ , \tag{87}$$

$$\nu_2=\nu_2(v)=\frac{1}{4}\int_{-1}^{1}d\mu\,\nu(v,\mu)(3\mu^2-1)\ , \tag{88}$$

$$\dot\nu_0=d\nu_0(v)/dv\ . \tag{89}$$

When the elastic collision frequency is much larger than the inelastic collision frequency, the isotropic component $f_0=f_0(v)$ of the velocity distribution function is given by the Chapman-Cowling-Davidov expression (76).

The longitudinal-diffusion coefficient is given [as can be obtained by Eqs. (31)–(34) and the more compact expression (56)], by

$$D_x=\frac{4}{3}\pi\int_0^\infty dv\, v^4\left\{\nu_0 f_0\left[\frac{1}{(\nu_0+b)^2}+\frac{2b}{(\nu_0+b)^3}+\frac{3b^2}{(\nu_0+b)^4}+\frac{54a^2}{v^2(\nu_0+b)^3}\left[\frac{1}{\nu_0}-\frac{1}{\nu_0+b}\right]\right]\right.$$
$$+a^2\left[\frac{\nu_1}{\nu_0-\nu_1}\left[\frac{9\dot\nu_0}{5(\nu_0+b)^4}-\frac{5}{v(\nu_0+b)^3}\right]\frac{df_0}{dv}\right.$$
$$\left.\left.+\frac{4v\nu_2}{15(\nu_0+b)^2(\nu_0-\nu_2)}\frac{d}{dv}\left[\frac{1}{\nu_0-\nu_1}\frac{1}{v}\frac{df_0}{dv}\right]-\frac{1}{3v^2(\nu_0+b)^2}\frac{d}{dv}\left[\frac{v^2}{\nu_0-\nu_1}\frac{df_0}{dv}\right]\right]\right\}$$
$$+\int_0^\infty d\tau\left[-\langle v_x\rangle^2+\frac{16\pi^2}{9}\int_0^\infty dv_i\, v_i^3 f_1(v_i)\int_0^\infty dv_f\, v_f^3\bar f_1(v_f,\tau\,|\,v_i)\right], \tag{90}$$

where, in the last integral, f_1 is given by

$$f_1(v) = -a(\nu_0-\nu_1)^{-1}\frac{df_0}{dv} \quad (91)$$

and $\bar{f}_1(v_f,\tau\,|\,v_i)$ can be obtained from the P_1 approximation to the Boltzmann equation.

In the large majority of real gases, $\nu_2 < \nu_1 \ll \nu_0$ so that Eqs. (89) and (90) can be strongly simplified.

APPENDIX A: CONNECTION BETWEEN DIFFUSION COEFFICIENTS AND NOISE SPECTRAL DENSITIES

The diffusion coefficient expressed by the velocity correlation function has been given by Kubo,[20] in the case of zero drift velocity $\mathbf{W}=\langle \mathbf{v}\rangle$. For reader's convenience we will here report the complete treatment when $W\neq 0$, moreover showing the connection between the generalized diffusion coefficient and the noise spectral density.

An electric field $\mathbf{E}=E\mathbf{i}$ is parallel to the x axis so that $\mathbf{W}=W_x\mathbf{i}=\langle v_x\rangle\mathbf{i}$. The longitudinal-diffusion coefficient D_x is defined as the position's mean-square spreading of a charge-carrier ensemble initially prepared in the position x_0, divided by $2t$. The so-defined D_x is independent of the time t provided t is much larger than the mean free time of flight of the charge carriers. In particular we can take the limit for $t\to\infty$, i.e.,

$$D_x = \lim_{t\to\infty}\left[\frac{1}{2t}\langle [x(t)-(x_0+Wt)]^2\rangle\right] . \quad (A1)$$

Now it is

$$x-(x_0+Wt) = \int_0^t dt[v_x(t)-W] = \int_0^t dt\, v_{x\mathrm{rel}}(t) , \quad (A2)$$

where $v_{x\mathrm{rel}}$ is the velocity relative to the mass center of the ensemble, i.e., the velocity measured by an observer having the velocity W with respect to the laboratory.

Substituting (A2) in (A1) gives

$$D_x = \lim_{t\to\infty}\left[\frac{1}{2t}\int_0^t dt_1\int_0^t dt_2\langle v_{x\mathrm{rel}}(t_1)v_{x\mathrm{rel}}(t_2)\rangle\right] . \quad (A3)$$

Let us put $\tau=t_2-t_1$ and change the variables in the following way

$$dt_1\,dt_2 = d\tau\,dt_1\begin{vmatrix}\dfrac{\partial t_1}{\partial t_1} & \dfrac{\partial t_1}{\partial \tau}\\ \dfrac{\partial t_2}{\partial t_1} & \dfrac{\partial t_2}{\partial \tau}\end{vmatrix} = d\tau\,dt_1\begin{vmatrix}1 & 0\\ 1 & 1\end{vmatrix} = d\tau\,dt_1 . \quad (A4)$$

If E is constant or if $\Delta E/E \ll 1$ during a time of flight, the process can be considered as stationary and the correlation function C_x of $v_{x\mathrm{rel}}$ depends only on $\tau=t_2-t_1$, i.e.,

$$\begin{aligned}C_x(t_1,t_2) &= \langle v_{x\mathrm{rel}}(t_1)v_{x\mathrm{rel}}(t_2)\rangle\\ &= \langle v_{x\mathrm{rel}}(t_1)v_{x\mathrm{rel}}(t_1+\tau)\rangle\\ &= \langle v_{x\mathrm{rel}}(0)v_{x\mathrm{rel}}(\tau)\rangle\\ &= C_x(\tau) .\end{aligned} \quad (A5)$$

Substituting (A4) and (A5) in (A3) and therefore changing the limits of integration gives

$$D_x = \lim_{t\to\infty}\left[\frac{1}{2t}\int_0^t dt_1\int_{-t_1}^{t-t_1} d\tau\, C_x(\tau)\right] . \quad (A6)$$

Since (A6) is the limit of the ratio between two divergent expressions (for $t\to\infty$), we can apply Hospital's rule

$$\begin{aligned}D_x &= \lim_{t\to\infty}\left[\frac{d}{2\,dt}\int_0^t dt_1\int_{-t_1}^{t-t_1} d\tau\, C_x(\tau)\right]\\ &= \lim_{t\to\infty}\left[\frac{1}{2}\int_{-t}^{0} d\tau\, C_x(\tau)+\frac{1}{2}\int_0^t dt_1\, c_x(t-t_1)\right] .\end{aligned} \quad (A7)$$

Let us perform the limit in the first integral and let us put $\xi=t-t_1$ in the second. Since $dt_1=-d\xi$, by changing the limits of integration we get

$$\begin{aligned}D_x &= \frac{1}{2}\int_{-\infty}^{0} d\tau\, C_x(\tau)+\lim_{t\to\infty}\left[\frac{1}{2}\int_t^0 -d\xi\, C_x(\xi)\right]\\ &= \frac{1}{2}\int_{-\infty}^{0} d\tau\, C_x(\tau)+\frac{1}{2}\int_0^\infty d\xi\, C_x(\xi) = \int_0^\infty d\tau C_x(\tau) .\end{aligned} \quad (A8)$$

The two transversal diffusion coefficients D_y and D_z are equal because of axial symmetry around $\mathbf{E}$. By the same procedure—and the simplification $\langle v_y\rangle=\langle v_x\rangle=0$, which implies $v_{y\mathrm{rel}}=v_y$ and $v_{z\mathrm{rel}}=v_z$—we get

$$D_y = D_z = \int_0^\infty d\tau\, C_y(\tau) = \int_0^\infty d\tau\, C_z(\tau) . \quad (A9)$$

The generalized diffusion coefficient is defined as

$$D_x(\omega) = \int_0^\infty d\tau\, C_x(\tau)\cos(\omega\tau) \quad (A10)$$

and coincides with D_x for $\omega\to 0$, i.e., $D_x(0)=D_x$. Because of (A2) and (A5), (A10) is equal to (2) of the main text. In a similar way (1) is obtained.

Let us now consider a wire section of length L, having plane parallel ends, containing N carriers each of charge e and with a velocity v_{sx} along the x axis of the wire. Ramo's theorem[21] gives, for the current $i(t)$ induced on the end planes,

$$i(t) = \frac{e}{L}\sum_{s=1}^{N} v_{sx}(t) . \quad (A11)$$

The mean value of the current is

$$\langle i(t)\rangle = \frac{e}{L}\sum_{s=1}^{N}\langle v_{sx}(t)\rangle = \frac{e}{L}N\langle v_x\rangle , \quad (A12)$$

and the current fluctuation is

$$\Delta i = \frac{e}{L}\left[\sum_{s=1}^{N} v_{sx}(t) - N\langle v_x\rangle\right] . \quad (A13)$$

$\Delta i=\Delta i(t)$ is a stochastic variable $y(t)$ whose spectral density is defined by

$$G(\omega) = \frac{2}{\pi}\int_0^\infty dt\langle y(0)y(t)\rangle\cos(\omega t) . \quad (A14)$$

If $y(t)=\Delta i(t)$, (A13) in (A14) gives

$$J_{\parallel}(\omega)=\frac{2e^2}{\pi L^2}\int_0^\infty dt\sum_{s=1}^{N}\sum_{p=1}^{N}[\langle v_{sx}(0)v_{px}(t)\rangle-N\langle v_{sx}(0)\langle v_x\rangle\rangle-N\langle v_{px}(t)\langle v_x\rangle\rangle+N^2\langle v_x\rangle^2]\cos(\omega t)$$

$$=\frac{2e^2}{\pi L^2}\int_0^\infty dt\left[\sum_{s=1}^{N}\langle v_{sx}(0)v_{sx}(t)\rangle+\sum_{s=1}^{N}\sum_{p\neq s=1}^{N}\langle v_{sx}(0)v_{px}(t)\rangle-N^2\langle v_x\rangle^2-N^2\langle v_x\rangle^2+N^2\langle v_x\rangle^2\right]\cos(\omega t)$$

$$=\frac{2e^2}{\pi L^2}\int_0^\infty dt(N\langle v_x(0)v_x(t)\rangle+N(N-1)\langle v_x\rangle^2-N^2\langle v_x^2\rangle)\cos(\omega t)$$

$$=\frac{2e^2}{\pi L^2}N\int_0^\infty dt(\langle v_x(0)v_x(t)\rangle-\langle v_x\rangle^2)\cos(\omega t)\ . \qquad \text{(A15)}$$

By comparing (A10) with (A15) we obtain the second step of (2) of the main text.

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