PHYSICAL REVIEW A VOLUME 5, NUMBER 6 JUNE 1972

Electron Correlations in the Unified Model for Stark Broadening

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(Received 2 December 1971)

An improved treatment of electron correlations is presented within the framework of a previously developed unified model for Stark broadening by plasma electrons. This treatment employs the method of the Bogoliubov-Born-Green-Kirkwood-Yvon hierarchy in connection with a closure relation corresponding essentially to the exclusion of simultaneous strong collisions. A line-shape expression is derived for the case that the perturbing electron gas is in thermal equilibrium. This expression depends on the dynamic dielectric constant and on the two-electron correlation function. If the latter is approximated by the linearized Debye-Hückel expression (low-density case), and if the result is simplified for the vicinity of the line center by using second-order perturbation theory for the atom-perturber interaction, the results of earlier treatments of electron correlations are recovered.

I. INTRODUCTION

Two important problems involved in the theory of Stark broadening of spectral lines have been treated separately in all previous investigations: the influence of the interaction between the plasma perturbers on the line profile, and the problem of "unification" of the "impact" and "quasistatic" theories of electron broadening.

The interaction between the charged particles of the plasma entails a shielding of their Coulomb fields. In connection with line broadening, this effect has generally been taken into account in calculations of distribution functions for the "low-frequency" component of the electric microfield,[1-6] and of autocorrelation functions for the "high-frequency" part of the microfield. The latter were needed in the impact theories of electron broadening.[7,8] Early treatments were based on cutoff procedures, truncating the collision integrals over the impact parameter at a shielding distance equal to the Debye length,[9,10] or to 1.123 times the Debye length.[11] In other investigations, static shielding effects have been introduced with the help of the linearized Debye-Hückel two-electron correlation function.[12,13] More recently, these approximations have been refined by expressing the autocorrelation function of the microfield in terms of the dynamic dielectric constant.[14-16] However, all these investigations used second-order perturbation theory for the plasma-atom interaction. This necessitated strong collision cutoffs for small impact parameters[17] and also prevented the resulting line-shape formulas from describing the transition to the quasistatic line wing.

Recently, theories have been developed which unify the impact and quasistatic aspects of the electron broadening in a coherent formalism, being valid from the collision-dominated line body up to the quasistatic wing. However, perturber correlations, which were basically contained in some of these approaches, were neglected at an early stage of the formalism. Shielding effects thus were treated by cutoff procedures (or Debye potentials), just as in the early impact theories.

The first unified treatment of electron broadening was based on a semiclassical many-body formalism.[18] The starting point was a generalized Liouville equation which was solved with Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) techniques using one essential assumption, namely that simultaneous strong electron collisions can be neglected. The method of using semiclassical statistics in connection with the exclusion of simultaneous strong collisions was pursued in later publications,[19-21] where more elegant expressions for the line profile have been given. Also, a quantum-mechanical formal line-shape expression has been proposed for the line wing.[22] Note that in all these treatments weak electron collisions are allowed to occur simultaneously, but, as already mentioned, without statistical interdependence. In Refs. 18 and 19 this restriction was introduced for the sake of mathematical simplicity, but was not necessary in principle, because the Liouville equation used there naturally contains the plasma interaction potential.

The purpose of this paper is to generalize the BBGKY method of Ref. 19 (hereafter referred to as I) to take full account of the electron part of the plasma interaction potential. This will lead us to a line-shape formula which contains both static and dynamic shielding of the electron gas. Therefore, our result will not only be unified with respect to electron broadening but also contain

properly the effects of electron correlations.

II. BASIC ASSUMPTIONS

Our formalism will be based on the classical-path approximation which includes the neglect of the back reaction of neutral atoms on the plasma perturbers. The validity of such an approach has been discussed in Paper I. A further assumption, also introduced in I, is the possibility of excluding simultaneous strong collisions. This has to be understood here in a generalized sense: not only strong electron-atom collisions, but also strong electron-electron collisions must be well separated in time. The strong-collision impact parameter for electron-atom collisions $b_{\min}$ is generally on the order of the Weisskopf radius b_W, but can be smaller in some cases of isolated lines,

$$b_{\min} \lesssim b_W = 3\hbar n^2/2m_e V_{\rm th} \ .$$

Here n is the principal quantum number of the excited state, m_e the electron mass, and $V_{\rm th}$ the thermal velocity of the electrons. The corresponding parameter for electron-electron collisions is the Landau length

$$b_L = e^2/m_e V_{\rm th}^2 \ ,$$

where e is the electron charge.

Hence, the validity conditions for the exclusion of simultaneous strong collisions are (a) $b_{\min} \ll \nu^{-1/3}$, (b) $b_L \ll \nu^{-1/3}$, where ν is the electron density.

A further condition needed for our formalism is imposed by the necessity that the cumulative effect of simultaneous weak collisions is sufficiently small as to cause only small changes of the atomic state during the plasma period ω_p^{-1}. As indicated in Appendix A, this leads to the inequality (c) $(b_W/\nu^{-1/3})^{3/2}(b_W/b_L)^{1/2}\ln(\lambda_D/b_{\min}) \ll 1$, where $\lambda_D = (kT_e/4\pi\nu e^2)^{1/2}$ is the Debye length. Note that this is one of the conditions for the impact theory to be valid around the line center. Hence, our unified formalism for electron broadening will be valid over the whole line profile if the impact theory is valid in the line core.

Condition (b) is equivalent with the requirement that the Debye sphere contains a great number of charged particles (kinetic plasma). Up to now, no theory of line broadening or plasma kinetics exists for cases where this condition is not fulfilled. For not-too-high principal quantum number n, condition (c) is included in (a). We have written conditions (a) and (c) for the electrons, for which they are usually well fulfilled. This would not be the case for the ions which, therefore, must be treated by a separate formalism. Following Ref. 3, we assume that the total plasma microfield can be split up into two stochastically independent parts: a "low-frequency" component $\vec{F}$ (being due to "dressed" ions and including ion-ion and ion-electron correlations) and a "high-frequency" component arising from a homogeneous gas of electrons moving in a uniform neutralizing positive background. The validity of this decomposition has been discussed in Ref. 23. According to this description we incorporate the low-frequency component $\vec{F}$ in the unperturbed Hamiltonian of the radiating atom, $H^0 = H^0(\vec{F})$. Neglecting the time dependence of F, H^0 then contains a static Stark effect. The total atomic Hamiltonian is

$$H = H^0(\vec{F}) + \sum_{j=1}^{N} V(\vec{r}_j) \ , \tag{1}$$

where $V(\vec{r}_j)$ is the potential due to the presence of one plasma electron at position $\vec{r}_j$. The sum of all electron potentials in the Hamiltonian (1) represents the contribution of the high-frequency component of the plasma microfield.

III. FORMALISM

As in I, we start from the general semiclassical line-shape formula[7,8] which describes the dipole radiation corresponding to transitions between two well-separated level groups, a and b, of the atom:

$$L(\omega) = \frac{1}{\pi}\,\mathrm{Re} \sum_{\alpha\alpha'\beta\beta'} \rho_{\alpha\alpha}\, \vec{D}_{\alpha\beta} \cdot \vec{D}^*_{\alpha'\beta'} \times \int_0^\infty e^{-i(\omega_0+\omega)t} \langle\langle \alpha\beta | \{T_b(t) T_a^*(t)\}_{\rm av} | \alpha'\beta' \rangle\rangle\, dt \ . \tag{2}$$

In this formula, the following notations are used: ω is the angular frequency measured from the line center ω_0. The summation subscripts α, α' and β, β' label the substates of the upper and lower atomic level groups a and b, respectively. ρ is the atomic density operator and $\vec{D}$ the dipole moment operator, with $\rho_{\alpha\alpha}$ and $\vec{D}_{\alpha\beta}$ denoting corresponding matrix elements. In the direct product $T_b T_a^*$, T_a and T_b denote the atomic time-evolution operator at time t, acting only on subspaces a and b, respectively. $\langle\langle \alpha\beta | T_b T_a^* | \alpha'\beta' \rangle\rangle$ ("doubled-atom" notation[7]) stands for $\langle \alpha | T^* | \alpha' \rangle \langle \beta | T | \beta' \rangle$. The symbol $\{\cdots\}_{\rm av}$ means ensemble average with respect to the plasma perturbers.

Equation (2) is based on the neglect of transitions induced by the plasma perturbers between levels of different groups (no-quenching assumption). The same approximation leads to a "projected Schrödinger equation" for T_a and T_b in the subspaces a and b of the atomic Hilbert space

$$\frac{\partial T_{a,b}}{\partial t} = -\frac{i}{\hbar}\left(H^0_{a,b}(\vec{F}) + \sum_{j=1}^{N} V_{a,b}(\vec{r}_j(t)) \right) T_{a,b} \ , \qquad T_{a,b}(0) = 1 \tag{3}$$

where $\vec{r}_j(t)$ describes the trajectory of electron

j, and

$$H^0_{a,b} \equiv P_{a,b} H^0 P_{a,b}\,, \quad V_{a,b} \equiv P_{a,b}\, V P_{a,b}\,,$$

P_a and P_b being the projection operators on subspaces a and b, respectively.

According to our decomposition of the plasma microfield, the ensemble average in (2) is carried out in two steps,

$$\{T_b\, T_a^*\}_{\rm av} = \{\{T_b\, T_a^*\}_{\rm hf}\}_{\rm lf} = \int_0^\infty W_{\rm lf}(F)\{T_b\, T_a^*\}_{\rm hf}\, dF\,, \tag{4}$$

where hf and lf refer to the high-frequency and low-frequency components of the microfield, and $W_{\rm lf}(F)$ is the low-frequency microfield distribution. The most elaborate evaluations of this function have been presented in Refs. 4–6.

Our aim is the evaluation of the high-frequency average $\{T_b\, T_a^*\}_{\rm hf}$. To this end, we consider the atomic-state vector $|z(t)\rangle$ (with components z^γ on some basis) and project it into the Hilbert subspaces a and b:

$$|x\rangle \equiv P_a\,|z\rangle\,, \quad |y\rangle \equiv P_b\,|z\rangle\,. \tag{5}$$

Since we have

$$|x(t)\rangle = T_a(t)|x(0)\rangle\,, \quad |y(t)\rangle = T_b(t)|y(0)\rangle$$

for any initial values $|x(0)\rangle$ and $|y(0)\rangle$, we may, in particular, choose the initial vector independent of the initial configuration of the plasma electrons. The desired operator average can then be related to the average of the dyadic product $|yx^*\rangle\rangle \equiv |y\rangle\,|x^*\rangle$ by the equation

$$\{|y(t)x^*(t)\rangle\rangle\}_{\rm hf} = \{T_b(t)T_a^*(t)\}_{\rm hf}\,|y(0)x^*(0)\rangle\rangle\,. \tag{6}$$

It is shown in I that the quantity on the left-hand side of Eq. (6) is related to a tensor $\Phi_N(\vec{r}_1, \vec{v}_1, \ldots, \vec{r}_N, \vec{v}_N; t)$ depending on the positions $\vec{r}_i$ and velocities $\vec{v}_i$ of the N plasma electrons, and on time t. This tensor has been defined in I by means of a distribution function for the variables of the atom-plasma system. An alternate definition of Φ_N is mathematically equivalent[24] but may be more transparent for physical interpretation. Let $f_N(\vec{r}_1, \vec{v}_1, \ldots, \vec{r}_N, \vec{v}_N, t)$ be the distribution function of the electron gas in the $6N$-dimensional phase space and

$$|z(t)\rangle = |z(\vec{r}_1, \vec{v}_1, \ldots, \vec{r}_N, \vec{v}_N; t)\rangle$$

be the atomic state corresponding to a specified configuration $(\vec{r}_1, \vec{v}_1, \ldots, \vec{r}_N, \vec{v}_N)$ at time t. Then [using definitions (5)], one has

$$\Phi_N(\vec{r}_1, \vec{v}_1, \ldots, \vec{r}_N, \vec{v}_N; t) \equiv |y(t)x^*(t)\rangle\rangle\, f_N(\vec{r}_1, \vec{v}_1, \ldots, \vec{r}_N, \vec{v}_N; t)\,.$$

From this it follows immediately that the quantity we wish to calculate is given by

$$\{|y(t)x^*(t)\rangle\rangle\}_{\rm hf} = \int \Phi_N(\vec{r}_1, \vec{v}_1, \ldots, \vec{r}_N, \vec{v}_N; t) \times d^3r_1 d^3v_1 \cdots d^3r_N d^3v_N\,. \tag{7}$$

The tensor Φ_N has been shown to satisfy a differential equation of the Liouville type [Eq. (6) of I],

$$\frac{\partial}{\partial t}\Phi_N + \frac{i}{\hbar}\bar{H}^0\Phi_N + \sum_{j=1}^N \left(\bar{V}(\vec{r}_j)\Phi_N + \vec{v}_j \cdot \frac{\partial}{\partial \vec{r}_j}\Phi_N - \frac{1}{m_e}\frac{\partial W_N}{\partial \vec{r}_j}\cdot\frac{\partial}{\partial \vec{v}_j}\Phi_N \right) = 0\,, \tag{8}$$

with the initial condition [chosen in accordance with (6)]

$$\Phi_N(\vec{r}_1, \vec{v}_1, \ldots, \vec{r}_N, \vec{v}_N; 0) = |y(0)x^*(0)\rangle\rangle\, f_N(\vec{r}_1, \vec{v}_1, \ldots, \vec{r}_N, \vec{v}_N; 0)\,.$$

Here, the following abreviations have been used:

$$\bar{H}^0 \equiv H_b^0 - H_a^{0*}\,, \quad \bar{V}(\vec{r}_j) \equiv V_b(\vec{r}_j) - V_a^*(\vec{r}_j)\,;$$

further,

$$W_N \equiv \sum_{j=1}^N \sum_{l=j+1}^N W(\vec{r}_{jl})\,,$$

with

$$W(\vec{r}) \equiv e^2/|\vec{r}|\,, \quad \vec{r}_{jl} \equiv \vec{r}_j - \vec{r}_l\,,$$

denotes the interaction potential of the electron gas. The action of an operator O_a or O_b on the tensor Φ_N has to be understood as follows: If $\Phi_N^{\beta\alpha}$ are the components of Φ_N, and $O^\alpha_{a/\alpha'}$ or $O^\beta_{b/\beta'}$ the matrix elements of O_a or O_b, the components of the tensor $O_a\Phi_N$ are

$$(O_a\Phi_N)^{\beta\alpha} = \sum_{\alpha'} O^\alpha_{a/\alpha'}\Phi_N^{\beta\alpha'}\,,$$

and those of $O_b\Phi_N$ are

$$(O_b\Phi_N)^{\beta\alpha} = \sum_{\beta'} O^\beta_{b/\beta'}\Phi_N^{\beta'\alpha}\,.$$

The next step of our treatment will be similar to the familiar BBGKY method known in statistical mechanics.[25] For the sake of brevity we shall use the short-hand notations $(1, 2\ldots)$ instead of $(\vec{r}_1, \vec{v}_1, \vec{r}_2, \vec{v}_2, \ldots)$ and $d(j)$ instead of $d^3r_j d^3v_j$, and also omit the time argument t when it is not needed explicitly.

Let us define the tensors Φ_s (with $s = 0, 1, \ldots, N$) by

$$\Phi_s(1, 2, \ldots, s; t) \equiv \int \Phi_N(1, 2, \ldots, N; t)\, d(s+1)\ldots d(N)\,.$$

In particular, for $s = 0$, this expression is identical with the average dyadic product (7):

$$\Phi_0(t) = \{|y(t)x^*(t)\rangle\rangle\}_{\rm hf}\,. \tag{9}$$

Let us also introduce the s-electron distribution

functions ($s=1,2,\ldots,N$):

$$f_s(1,2,\ldots,s;t)\equiv\int f_N(1,2,\ldots,N;t)\,d(s+1)\cdots d(N).$$

These functions satisfy the well-known BBGKY hierarchy of equations.[25] A similar chain of equations can be derived for the Φ_s. By integrating Eq. (8) over the variables of all but s electrons, we obtain an equation which couples Φ_s to Φ_{s+1}:

$$\frac{\partial}{\partial t}\Phi_s+\frac{i}{\hbar}\overline{H}^0\Phi_s+\sum_{j=1}^{s}\left(\frac{i}{\hbar}\overline{V}(j)\Phi_s+\vec{v}_j\cdot\frac{\partial}{\partial\vec{r}_j}\Phi_s-\frac{1}{m_e}\sum_{\substack{l=1\\ l\neq j}}^{s}\frac{\partial W(\vec{r}_{jl})}{\partial\vec{r}_j}\cdot\frac{\partial}{\partial\vec{v}_j}\Phi_s\right)$$

$$=-\frac{i}{\hbar}(N-s)\int\overline{V}(s+1)\Phi_{s+1}\,d(s+1)+(N-s)\sum_{j=1}^{s}\int\frac{\partial W(\vec{r}_{j,s+1})}{\partial\vec{r}_j}\cdot\frac{\partial}{\partial\vec{v}_j}\Phi_{s+1}\,d(s+1)\ . \qquad (10)$$

Writing this equation for $s=0,1,2,\ldots,N$ yields a hierarchy of $N+1$ coupled equations for the functions Φ_0, Φ_1, ..., Φ_N, subject to the initial conditions

$$\Phi_s(1,\ldots,s;0)=\Phi_0(0)f_s(1,\ldots,s;0)\ . \qquad (11)$$

As usual in kinetic theory, we truncate this hierarchy after the second ($s=1$) equation by introducing a closure hypothesis, expressing the three-body function $\Phi_2(1,2)$ (which couples two electrons to the atom) in terms of two-body and one-body functions. In our case of electron broadening, the following closure relation turns out to be particularly convenient[26]:

$$\Phi_2(1,2)=\Phi_1(1)f_1(2)+\Phi_1(2)f_1(1)+\Phi_0 f_2(1,2)-2\Phi_0 f_1(1)f_1(2)\ . \qquad (12)$$

We show in Appendix A that the approximation connected with this relation corresponds to the exclusion of simultaneous strong electron-atom and electron-electron collisions, and that its validity domain is delimited by the conditions (a)–(c) introduced in Sec. II.

After writing Eq. (10) for $s=0$ and 1, we insert the expression (12) for Φ_2 in the integral terms on the right-hand side of the equation for $s=1$. Assuming the electron gas to be homogeneous and in equilibrium over the characteristic lengths of interest, we may use the first BBGKY equation for $f_1(1,t)$:

$$\frac{\partial f_1(1,t)}{\partial t}=\frac{N-1}{m_e}\int\frac{\partial W(\vec{r}_{13})}{\partial\vec{r}_1}\cdot\frac{\partial f_2(1,3)}{\partial\vec{v}_1}\,d(3)=0\ ,$$

and thus obtain

$$\left(\frac{\partial}{\partial t}+\frac{i}{\hbar}\overline{H}^0\right)\Phi_0(t)=-\frac{i}{\hbar}N\int\overline{V}(3)\Phi_1(3;t)d(3)\ , \qquad (13)$$

$$\left(\frac{\partial}{\partial t}+\frac{i}{\hbar}\overline{H}_0+\frac{i}{\hbar}\overline{V}(1)+\vec{v}_1\frac{\partial}{\partial\vec{r}_1}\right)\Phi_1(1;t)=-\frac{i(N-1)}{\hbar}f_1(1)\int\overline{V}(3)\Phi_1(3;t)d(3)$$

$$+\frac{N-1}{m_e}\frac{\partial f_1(1)}{\partial\vec{v}_1}\cdot\int\frac{\partial W(\vec{r}_{13})}{\partial\vec{r}_1}\Phi_1(3;t)\,d(3)-\frac{i(N-1)}{\hbar}\int\overline{V}(3)g(1,3)d(3)\Phi_0(t)\ , \qquad (14)$$

where

$$g(1,2)\equiv f_2(1,2)-f_1(1)f_1(2)$$

is the two-electron equilibrium correlation function.

Equations (13) and (14) are closed in Φ_0 and Φ_1 and will therefore serve to calculate Φ_0 whose Fourier transform is needed in the line-shape formula [see Eqs. (2), (6), and (9)]. Since in our formalism Fourier and Laplace transforms of most of the quantities involved are needed, we introduce a general definition, denoting the Fourier and Laplace transform of an arbitrary function $h(\vec{r}_1,\vec{r}_2,\ldots;t)$ of position vectors $\vec{r}_j$ and of time by

$$\tilde{h}(\vec{k}_1,\vec{k}_2,\ldots;p)\equiv\int_{-\infty}^{+\infty}\cdots\int_{-\infty}^{+\infty}e^{-i[\vec{k}_1\cdot\vec{r}_1+\vec{k}_2\cdot\vec{r}_2\cdots]}d^3r_1\,d^3r_2\cdots\int_0^{+\infty}e^{-pt}\,dt\,h(\vec{r}_1,\vec{r}_2,\ldots;t)\ . \qquad (15)$$

If h does not depend on time, $\tilde{h}$ is defined as to be just the Fourier transform, if it depends only on time, it is just the Laplace transform. For brevity we shall write $\tilde{\vec{V}}(\vec{k})=\tilde{V}(\vec{k})$. We further define

$$\bar{g}(\vec{k},\vec{v}_2)\equiv\int\int_{-\infty}^{+\infty} e^{-i\vec{k}\cdot(\vec{r}_1-\vec{r}_2)}\, g(1,2)\,d^3r_1\,d^3v_1 \quad . \tag{16}$$

To solve Eqs. (13) and (14), we make extensive use of Green's functions. Let us first introduce an operator $R(1,2;t)$ defined by the equation

$$\left(\frac{\partial}{\partial t}+\frac{i}{\hbar}\bar{H}^0+\frac{i}{\hbar}\bar{V}(1)+\vec{v}_1\cdot\frac{\partial}{\partial\vec{r}_1}\right)R(1,2;t) = \frac{N-1}{m_e}\frac{\partial f_1(1)}{\partial\vec{v}_1}\cdot\int\frac{\partial W(\vec{r}_{13})}{\partial\vec{r}_1}R(3,2;t)\,d(3) \quad , \tag{17}$$

with initial condition

$$R(1,2;0)=\delta(\vec{r}_1-\vec{r}_2)\delta(\vec{v}_1-\vec{v}_2) \quad . \tag{18}$$

If we use Eq. (13) to eliminate the first integral term on the right-hand side of Eq. (14) (using $N\approx N-1$), the operator R may serve us as a Green's function to express Φ_1 explicitly in terms of Φ_0. Taking the Fourier and Laplace transform of Φ_1, we obtain

$$\tilde{\Phi}_1(\vec{k},\vec{v}_1;p)=S(\vec{k},\vec{v}_1;p)\tilde{\Phi}_0(p) \quad , \tag{19}$$

with

$$S(\vec{k},\vec{v}_1;p)=\int\int\tilde{R}(\vec{k},\vec{v}_1,\vec{k}',\vec{v}_2;p)\left[f_1(2)\left(p+\frac{i}{\hbar}\bar{H}^0\right)\delta(\vec{k}')-\frac{iN}{\hbar(2\pi)^3}\tilde{V}(\vec{k}')\bar{g}(\vec{k}',\vec{v}_2)\right]d^3v_2\,d^3k' \quad . \tag{20}$$

$\tilde{\Phi}_0(p)$ can now be expressed explicitly in terms of $\Phi_0(0)$ by combining (19) with the first equation of the hierarchy [Eq. (13)]. We obtain

$$\tilde{\Phi}_0(p)=[p+(i/\hbar)\bar{H}^0+K(p)]^{-1}\Phi_0(0) \tag{21}$$

with

$$K(p)=-\frac{iN}{\hbar(2\pi)^3}\int\int\tilde{V}(\vec{k})S(\vec{k},\vec{v};p)\,d^3k\,d^3v \quad .$$

From Eqs. (2), (4), (6), (9), and (21), it can easily be seen that $K(p)$ becomes the electronic collision operator ("width and shift operator") of the line profile if we put $p=i\omega+0$.

For the further evaluation of the expression $S(\vec{k},\vec{v};p)$, we next determine the operator R from Eqs. (17) and (18). To this end, we use the atomic time-evolution operator $Q(\vec{r},\vec{v};t)$ describing the time development of the atom in the presence of one perturber moving on the straight line $\vec{r}+\vec{v}t$. Q is defined by the Schrödinger equation

$$\frac{\partial}{\partial t}Q+\frac{i}{\hbar}[\bar{H}^0+\bar{V}(\vec{r}+\vec{v}t)]Q=0 \quad , \tag{22}$$

with the initial condition

$$Q(\vec{r},\vec{v};0)=1 \quad .$$

By an integration along the characteristics [for more details of the calculation see Eqs. (9)–(17) of Paper I], Eq. (17) may be given the following form:

$$R(1,2;t)=Q(\vec{r}_1-\vec{v}_1t,\vec{v}_1;t)\delta(\vec{r}_{12}-\vec{v}_1t)\delta(\vec{v}_1-\vec{v}_2) + \frac{N}{m_e}\frac{\partial f_1(1)}{\partial\vec{v}_1}\int_0^t Q(\vec{r}_1-\vec{v}_1\tau,\vec{v}_1;\tau)\,d\tau \times\int\frac{\partial}{\partial\vec{r}_1}W(\vec{r}_{13}-\vec{v}_1\tau)R(3,2;t-\tau)\,d(3) \quad .$$

Taking the Fourier and Laplace transform of this equation and integrating over the velocity vector $\vec{v}_1$ leads to

$$\int D(\vec{k},\vec{k}'';p)\left[\int\tilde{R}(\vec{k}'',\vec{v}_1,\vec{k}',\vec{v}_2;p)\,d^3v_1\right]d^3k'' = \tilde{Q}(\vec{k}+\vec{k}',\vec{v}_2;p+i\vec{k}\cdot\vec{v}) \quad , \tag{23}$$

where the kernel D is given by

$$D(\vec{k},\vec{k}';p)\equiv\delta(\vec{k}-\vec{k}') - \frac{iN}{m_e(2\pi)^3}\int\frac{\partial f_1(1)}{\partial\vec{v}_1}\cdot\vec{k}'\tilde{W}(\vec{k}') \times\tilde{Q}(\vec{k}-\vec{k}',\vec{v}_1;p+i\vec{k}\cdot\vec{v}_1)\,d^3v_1 \quad . \tag{24}$$

To solve Eq. (23), we again introduce a Green's function $G(\vec{k},\vec{k}';p)$ defined by

$$\int D(\vec{k},\vec{k}'';p)G(\vec{k}'',\vec{k}';p)\,d^3k''=\delta(\vec{k}-\vec{k}') \quad . \tag{25}$$

The unknown in Eq. (23) can then be expressed as

$$\int\tilde{R}(\vec{k},\vec{v}_1,\vec{k}',\vec{v}_2;p)\,d^3v_1 = \int G(\vec{k},\vec{k}'';p)\tilde{Q}(\vec{k}''+\vec{k}',\vec{v}_2;p+i\vec{k}''\cdot\vec{v}_2)\,d^3k'' \quad . \tag{26}$$

Before determining G from Eqs. (25) and (24), let us insert the formal expression (26) in Eq. (20) (after integration over $\vec{v}_1$). With the help of the relation (B1) of Appendix B, and using the fact that $G(\vec{k},0;p)=\delta(\vec{k})$, the operator $K(p)$ takes the following form:

$$K(p)=\frac{-N}{\hbar^2(2\pi)^6}\iiiint\tilde{V}(\vec{k})G(\vec{k},\vec{k}';p)\tilde{Q}(\vec{k}'-\vec{k}'',\vec{v};p+i\vec{k}'\cdot\vec{v})\tilde{V}(\vec{k}'')\left(\frac{\varphi(v)}{V}+N\bar{g}(\vec{k}'',\vec{v})\right)d^3k\,d^3k'\,d^3k''\,d^3v \quad . \tag{27}$$

Here,

$$\varphi(\vec{v}_1) = V f_1(1) = (m_e/2\pi k T_e)^{3/2} e^{-m_e v^2/2kT_e}$$

denotes the Maxwell distribution (V being the plasma volume).

In order to derive an explicit expression for the function $G(\vec{k}\vec{k}'; p)$, we slightly change the form of Eq. (24). Using the relation (B1) of Appendix B for $\vec{k} \to \vec{k} - \vec{k}'$, $p \to p + i\vec{k}' \cdot \vec{v}$, we obtain a new expression for the kernel D:

$$D(\vec{k},\vec{k}';p) = D^{(0)}(\vec{k},\vec{k}';p) + D^{(1)}(\vec{k},\vec{k}';p) \ ,$$

where

$$D^{(0)}(\vec{k},\vec{k}';p) = \epsilon\left(\vec{k}, p + \frac{i}{\hbar}\bar{H}^0\right)\delta(\vec{k}-\vec{k}') \ ,$$

$$D^{(1)}(\vec{k},\vec{k}';p) = -\frac{\nu}{(2\pi)^6 \hbar m_e} \iint \frac{\partial\varphi(\vec{v})}{\partial\vec{v}} \cdot \vec{k}'\tilde{W}(\vec{k}')\tilde{Q}(\vec{k}-\vec{k}'', \vec{v}_1, p + i\vec{k}\cdot\vec{v}_1)\tilde{V}(\vec{k}''-\vec{k}')\left(p + \frac{i}{\hbar}\bar{H}^0 + i\vec{k}'\cdot\vec{v}\right)^{-1} d^3v\, d^3k'' \ .$$

Here

$$\epsilon(\vec{k}, s) = 1 - \frac{i\nu}{m_e} \int \frac{\partial\varphi(\vec{v})}{\partial\vec{v}} \cdot \vec{k}\,\tilde{W}(\vec{k})(s + i\vec{k}\cdot\vec{v})^{-1} d^3v \tag{28}$$

is the dielectric constant of the electron gas.

Equation (25) can now be written in the form

$$G(\vec{k},\vec{k}';p) = \epsilon^{-1}\left(\vec{k}, p + \frac{i}{\hbar}\bar{H}^0\right) \times \left[\delta(\vec{k}-\vec{k}') - \int D^{(1)}(\vec{k},\vec{k}'';p)G(\vec{k}'',\vec{k}';p)\,d^3k''\right] \ .$$

This equation is particularly suitable for an iterative solution with the lowest-order approximation

$$G(\vec{k},\vec{k}';p) = \epsilon^{-1}\left(\vec{k}, p + \frac{i}{\hbar}\bar{H}^0\right)\delta(\vec{k}-\vec{k}') \ . \tag{29}$$

By writing the corresponding iteration series for the quantity

$$\int \tilde{V}(\vec{k})G(\vec{k},\vec{k}';p)\,d^3k$$

[which is needed in (27)], it can be shown[27] that the relative correction to the lowest-order solution (29) is on the order of b_W/λ_D. Since for a kinetic plasma $\nu^{-1/3}$ is much smaller than λ_D, we have $b_W \ll \lambda_D$ whenever condition (a) is fulfilled. Therefore, the approximation (29) is less restrictive than the closure hypothesis (12).

After inserting the expression (29) in Eq. (27), the operator $K(p)$ still contains the two undetermined functions $\bar{g}(\vec{k},\vec{v})$ and $\tilde{Q}(\vec{k},\vec{v};p)$. The former is related to the two-electron equilibrium correlation function [see Eq. (16)], which is generally well approximated[25] by

$$g(1,2) \simeq f_1(1)f_1(2) \times \left\{\exp\left[-\frac{1}{kT_e}W(\vec{r}_{12})\exp\left(\frac{-|\vec{r}_{12}|}{\lambda_D}\right)\right] - 1\right\} \ . \tag{30}$$

In cases where condition (b) is well fulfilled one may replace (30) by the linearized Debye-Hückel expression[25] [corresponding to the first two terms of the series expansion of the outer exponential in (30)]. This leads to

$$N\bar{g}(\vec{k},\vec{v}) = -[\varphi(v)/V]\frac{1}{1+k^2\lambda_D^2} \ ,$$

and since

$$\epsilon(\vec{k},0) = 1 + 1/k^2\lambda_D^2 \ ,$$

we have the relation

$$N\bar{g}(\vec{k},\vec{v}) + \varphi(\vec{v})/V = \epsilon^{-1}(\vec{k},0)\varphi(\vec{v})/V \ . \tag{31}$$

Using (29) and (31), the operator $K(p)$ takes the form

$$K(p) = \frac{-\nu}{(2\pi)^6\hbar^2} \iiint \tilde{V}(\vec{k})\epsilon^{-1}\left(\vec{k}, p + \frac{i}{\hbar}\bar{H}^0\right)\tilde{Q}(\vec{k}-\vec{k}', \vec{v}; p + i\vec{k}\cdot\vec{v})\tilde{V}(\vec{k}')\,\epsilon^{-1}(k', 0)\varphi(\vec{v})\,d^3k\,d^3k'\,d^3v \ . \tag{32}$$

For numerical applications, it appears to be more convenient to invert the Fourier transforms in the integrand of the expression (32). To this end, we introduce the "dynamically shielded" potential[28]

$$V_S(\vec{r}, p) = [1/(2\pi)^3] \int \tilde{V}(\vec{k})\epsilon^{-1}(\vec{k}, p)\, e^{i\vec{k}\cdot\vec{r}}\, d^3k \ . \tag{33}$$

Using this definition, Eq. (32) becomes

$$K(p) = -\frac{\nu}{\hbar^2}\int_0^\infty e^{-pt}\,dt \iint V_S\left(\vec{r} + \vec{v}t, p + \frac{i}{\hbar}\bar{H}^0\right) \times Q(\vec{r},\vec{v},t)V_S(\vec{r},0)\varphi(\vec{v})\,d^3r\,d^3v \ . \tag{34}$$

We notice that in this expression the two potential factors V_S are shielded whereas the potential in the Schrödinger equation (22) for Q is not.

Therefore, Eq. (34) cannot be obtained from the former unified theory[19-21] by simply replacing V everywhere with V_S. On the other hand, it cannot be excluded that such a result would be obtained if the cluster expansion would be extended to higher order [instead of just using Eq. (12)] and if the exact solution for $G(\vec{k},\vec{k}';p)$ could be used instead of (29). We were able to show that using an exact expression for $G(\vec{k},\vec{k}';p)$ without extending the cluster expansion would not suffice to replace the Coulomb potential by a shielded potential in Eq. (22). Any difference between such a result and Eq. (34) would be small, however, because shielding only becomes effective at large distances where Q can be replaced by the zero-order term of its perturbation expansion.

For a final evaluation of Eq. (34), the "one-perturber" atomic time-evolution operator $Q(\vec{r},\vec{v},t)$ has to be determined from the Schrödinger equation (22). Various approximate solutions of this equation have been discussed in previous investigations[19,20,29-31]; in particular, an algorithm for a sequence of exponential approximate solutions of equations of the type (22) has been derived,[30] and the second member of this sequence has been used in Ref. 19. For the special case of complete degeneracy of the unperturbed Hamiltonian and for a pure dipole interaction V, the exact solution of Eq. (22) has recently been found.[32-34] Therefore, in cases when the level splitting due to the ionic microfield does not influence the collision operator significantly, calculations for hydrogen lines on the basis of the exact operator Q of Refs. 32-34 should be most promising.

The knowledge of the expression (34) for $p=i\omega+0$ permits us to write the line-shape formula explicitly. Using Eqs. (2), (4), (6), (9), and (21), we find

$$L(\omega)=\frac{1}{\pi}\,\mathrm{Re}\sum_{\alpha\alpha'\,\beta\beta'}\rho_{\alpha\alpha}\,\vec{D}_{\alpha\beta}\,\vec{D}^{*}_{\alpha'\beta'}\int_0^\infty W_{1t}(F)\,dF\,\langle\langle\alpha\beta\,|\,[i\,(\omega+\overline{H}^0/\hbar)+K(i\omega+0)]^{-1}\,|\,\alpha'\beta'\rangle\rangle\quad .$$

We note in conclusion that from the "width and shift operator" $K(i\omega+0)$ as given by Eq. (34), we may reproduce the "unified" result of Refs. 19-21 and the "dynamically shielded" impact formula of Refs. 14-16 by using corresponding approximations. The former is obtained from (34) by putting $\epsilon(\vec{k},p)=1$ in Eq. (33) and introducing Debye cutoffs (or Debye potentials) instead. The latter can be derived by approximating $Q(\vec{r},\vec{v};t)$ by $e^{-it\overline{H}^0/\hbar}$, the zero-order term of its perturbation series, and introducing a strong-collision cutoff. In contrast to the results of Refs. 14-16, 19-21, the integrations in (34) do not need to be cutoff at either small or large impact parameters.

APPENDIX A

The closure relation (12) can easily be verified for the case that two of the three particles (atom, electron 1, and electron 2) undergo a strong collision (in the sense as defined in Sec. II) while the interaction of these particles with the third one is weak in comparison and thus negligible. Let us assume that the strong collision occurs between the atom and electron 1. Then $\Phi_2(1,2)$, $\Phi_1(2)$, and $f_2(1,2)$ factorize to become $\Phi_1(1)f(2)$, $\Phi_0 f_1(2)$, and $f_1(1)f_1(2)$, respectively, and Eq. (11) is identically fulfilled. The same argument holds when electron 2 is strongly coupled to the atom. When the strong collision occurs between the two electrons, we have $\Phi_2(1,2)=\Phi_0 f_2(1,2)$, $\Phi_1(1)=\Phi_0 f_1(1)$, and $\Phi_1(2)=\Phi_0 f_1(2)$, and Eq. (11) is satisfied again.

If the three particles interact simultaneously but weakly (in the sense as defined in Sec. II), Eq. (12) can be verified by a perturbation treatment of the hierarchy (10) up to first order.[27] We here choose a slightly different method which is less rigorous but somewhat simpler and physically more transparent. Let us introduce formal coupling parameters λ_1, λ_2, and λ_{12} (characterizing the strength of interaction between the atom and electrons 1 and 2, and between the two electrons, respectively,) and let us write Φ_1, Φ_2, and f_2 as

$$\Phi_1(1;\lambda_1),\ \Phi_1(2;\lambda_2),\ \Phi_2(1,2;\lambda_1,\lambda_2,\lambda_{12}),$$

$$f_2(1,2;\lambda_{12}).$$

In the case that some or all of the λ's vanish, the three particles are partially or completely decorrelated:

$$\Phi_1(1;0)=\Phi_0 f_1(1),\quad \Phi_2(1,2;0,0,0)=\Phi_0 f_1(1)f_1(2)\ , \tag{A1a}$$

$$f_2(1,2,0)=f_1(1)f_1(2), \tag{A1b}$$

$$\Phi_2(1,2;\lambda_1,0,0)=\Phi_1(1,\lambda_1)f_1(2)\ ,$$

$$\Phi_2(1,2;0,0,\lambda_{12})=\Phi_0 f_2(1,2;\lambda_{12}),\ \text{etc.} \tag{A1c}$$

To first order in the coupling parameters we have

$$\Phi_1(1;\lambda_1)=\Phi_0 f_1(1)+\lambda_1\frac{\partial}{\partial\lambda_1}\Phi_1(1;\lambda_1)\big|_{\lambda_1=0}+O(\lambda_1^2)\ , \tag{A2a}$$

$$\Phi_1(2;\lambda_2)=\Phi_0 f_1(2)+\lambda_2\frac{\partial}{\partial\lambda_2}\Phi_1(2;\lambda_2)\big|_{\lambda_2=0}+O(\lambda_2^2)\ , \tag{A2b}$$

$$f_2(1,2;\lambda_{12})=f_1(1)f_1(2)$$

$$+\lambda_{12}\frac{\partial}{\partial\lambda_{12}}f_2(1,2;\lambda_{12})\big|_{\lambda_{12}=0}+O(\lambda_{12}^2)\ , \tag{A2c}$$

$$\Phi_2(1,2;\lambda_1,\lambda_2,\lambda_{12})=\Phi_0 f_1(1)f_1(2)$$
$$+\lambda_1\frac{\partial}{\partial\lambda_1}\Phi_2(1,2;\lambda_1,0,0)\big|_{\lambda_1=0}$$
$$+\lambda_2\frac{\partial}{\partial\lambda_2}\Phi_2(1,2;0,\lambda_2,0)\big|_{\lambda_2=0}$$
$$+\lambda_{12}\frac{\partial}{\partial\lambda_{12}}\Phi_2(1,2;0,0,\lambda_{12})\big|_{\lambda_{12}=0}$$
$$+O(\lambda_1^2,\lambda_2^2,\lambda_{12}^2,\lambda_1\lambda_2,\lambda_1\lambda_{12},\lambda_2\lambda_{12})\ . \tag{A3}$$

Using Eqs. (A1) and (A2), we find

$$\lambda_1\frac{\partial}{\partial\lambda_1}\Phi_2(1,2;\lambda_1,0,0)\big|_{\lambda_1=0}$$
$$=f_1(2)\lambda_1\frac{\partial}{\partial\lambda_1}\Phi_1(1;\lambda_1)\big|_{\lambda_1=0}$$
$$=f_1(2)[\Phi_1(1;\lambda_1)-\Phi_0 f_1(1)]+O(\lambda_1^2)$$

and correspondingly

$$\lambda_1\frac{\partial}{\partial\lambda_1}\Phi_2(1,2;0,\lambda_2,0)$$
$$=f_1(1)[\Phi_1(2;\lambda_2)-\Phi_0 f_1(2)]+O(\lambda_2^2)\ ,$$
$$\lambda_{12}\frac{\partial}{\partial\lambda_{12}}\Phi_2(1,2;0,0,\lambda_{12})\big|_{\lambda_{12}=0}$$
$$=\Phi_0[f_2(1,2,\lambda_{12})-f_1(1)f_1(2)]+O(\lambda_{12}^2)\ .$$

On inserting these equations in (A3) we obtain the closure relation (12) to first order in the coupling parameters λ_1, λ_2, and λ_{12}.

Note that Eq. (12) is not valid for the case that the three particles interact simultaneously and strongly. Hence, our formalism is valid only if strong collisions are well separated in time, i.e., if conditions (a) and (b) of Sec. II are fulfilled.

A problem still arises from the fact that even though the perturbation expansion in the coupling parameters λ makes sense for $\Phi_2(1,2)$ (corresponding to two simultaneous weak collisions on the atom), the contributions of the higher orders in the integral terms of the hierarchy do not necessarily decrease with increasing powers of λ. In fact, the integral terms contain the cumulative effects of many weak collisions, and we have to make sure whether our perturbation treatment accounts for these effects. Let us consider the most pessimistic case of long times ($t>\omega_p^{-1}$, ω_p being the plasma frequency). Then all collisions are completed and the perturbation has its largest effect on the time variation $\partial\Phi_0(t)/\partial t$. Our expansion would then coincide with that used in the derivation of the collision operator of the impact theory.[7-9] The condition for this expansion to be valid is

$$\left|\frac{1}{\Phi}\frac{\partial\Phi}{\partial t}\omega_p^{-1}\right|\ll 1\ ,$$

where Φ denotes some characteristic component of the tensor $\Phi_0(t)$. Explicitly, this condition is given by the inequality (c) of Sec. II.

APPENDIX B

Using the identity

$$Q^{-1}(\vec{r},\vec{v};-t)\equiv Q(\vec{r}-\vec{v}t,\vec{v};t)$$

which follows from Eq. (20), and the relation

$$\frac{d}{dt}Q^{-1}=-Q^{-1}\frac{dQ}{dt}Q^{-1}\ ,$$

we obtain

$$\frac{d}{dt}Q(\vec{r}-\vec{v}t,\vec{v};t)$$
$$=-\frac{i}{\hbar}Q(\vec{r}-\vec{v}t,\vec{v};t)[\overline{H}^0+\overline{V}(\vec{r}-\vec{v}t)]\ .$$

Taking the Fourier and Laplace transform, this equation is readily transformed to

$$\tilde{Q}(\vec{k},\vec{v};p+i\vec{k}\cdot\vec{v})\left(p+\frac{i}{\hbar}\overline{H}^0\right)=(2\pi)^3\delta(\vec{k})$$
$$-\frac{i}{\hbar(2\pi)^3}\int\tilde{Q}(\vec{k}-\vec{k}',\vec{v};p+i\vec{k}\cdot\vec{v})\tilde{V}(\vec{k}')d^3k'\ . \tag{B1}$$

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PHYSICAL REVIEW A VOLUME 5, NUMBER 6 JUNE 1972

Charge Dependence of Ionization Energy Loss for Relativistic Heavy Nuclei

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(Received 18 October 1971)

The ionization energy loss of relativistic heavy nuclei is calculated using the exact Mott cross section for close collisions. Deviations from the Bloch formula are computed in some typical cases and found to be significant for nuclei with $Z \gtrsim 20$.

I. INTRODUCTION

Methods for calculating ionization energy loss of charged particles traversing matter predate the beginning of quantum mechanics. The original classical calculation by Bohr[1] was followed by Bethe's quantum-mechanical treatment[2] and Williams' method of impact parameters.[3] Bloch[4] has indicated the connection between the classical and quantum-mechanical methods, and Mott[5] has demonstrated the equivalence of Bethe's method and the method of impact parameters. Elaborations and extensions of this early work to include such things as shell corrections and the density effect have been given, and there are review articles which give a complete summary of all this work.[6]

One result of these calculations is that the average energy loss of a particle of charge Ze and velocity β is proportional to Z^2 when $Z/137\beta \ll 1$. When this condition is not satisfied, Bloch's formula[7] gives a more general Z dependence which reduces to the classical result given by Bohr[1] when $Z/137\beta \gg 1$. Bloch's correction is present in the nonrelativistic case and can be thought of as the result of a modification of the minimum scattering angle below which no energy transfer takes place.[8]

If nuclear collisions are ignored, charged particles lose energy primarily by collisions with atomic electrons. It is convenient to divide these collisions into "distant collisions," in which electron-binding effects are included, and "close collisions," in which the binding energy of the electrons is ignored.[9] In the relativistic case, there are corrections to the Born approximation for close collisions of electrons with heavy nuclei ($20 < Z < 120$) which are not included in Bloch's original calculation. In this paper, these corrections are computed numerically using the exact Mott cross section[10] transformed to the frame in which the electron is initially at rest. The usual treatment of distant collisions is assumed to be valid here (see Sec. II B for a discussion of this assumption). The total energy loss is computed as a function of Z and compared with Bloch's formula in some typical cases.

II. ENERGY-LOSS CALCULATION

A. Close Collisions

In order to obtain the energy loss of a heavy par-