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Energy loss of correlated charges in an electron gas

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The energy loss of a pair of charges in correlated motion through an electron gas is calculated using Lindhard's dielectric function. The results of numerical integrations are presented and in particular the cases of low and high velocities are described. Analytical expressions for the energy loss are given for the case of high velocities, which are in excellent agreement with the numerical results. A clear relationship between the energy loss of fast correlated charges and the partition rule for the stopping does not follow from this study. The results are in agreement with experimental data for the energy-loss ratio between molecular and atomic ions in thin carbon foils.

I. INTRODUCTION

The energy loss of charged particles in a degenerate electron gas has been a topic of great interest since the work of Lindhard and Winther,[1] because it became of considerable importance for the study of the energy loss of charged particles in real media.[2]

Whereas the energy loss of single atomic particles in matter has been widely studied for many years, the energy loss of swift ion clusters has been the subject of much more recent work, concerned with the incidence of swift molecular ions on thin solid films.[3-6] Molecular effects on the energy loss have been theoretically described[3,4] as a result of interference effects in the energy dissipated in the material, when its electrons are perturbed by the fields of external charges in correlated motion. Both of these treatments dealt with valence band electron excitations and made use of high-velocity approximations in calculating the energy loss, wherein the random motion of the electrons is neglected. Brandt, Ratkowski and Ritchie[3,6] have proposed a relation between the energy loss of ion clusters and the partition rule for the contribution of individual and collective electronic excitations to the stopping, which will be further analyzed here. On the other hand, Arista and Ponce[4] performed an analytical calculation of the energy loss, using simplified models to describe long-wavelength collective excitations and short-range individual excitations (for $k \ll k_c$ and $k \gg k_c$, respectively, where k_c is an appropriate cutoff wave number for the electron gas), and interpolating through the more complicated intermediate region ($k \sim k_c$).

We consider in this paper a more appropriate treatment for the energy loss of charges in correlated motion through an electron gas, using Lindhard's expression[7] for the dielectric constant of the medium. This provides a good description of collective and single-particle excitations with a self-consistent treatment of screening effects. Using the dielectric formalism we calculate the energy loss of two correlated charges; we present the results of numerical integrations, and also approximations valid at low and high velocities. These results are in addition compared with previous high-velocity approximations and several conclusions are drawn.

In Sec. II we express the energy loss of a cluster of nonrelativistic charges moving in a material medium, in terms of the longitudinal dielectric constant $\epsilon(\vec{k}, \omega)$. In Sec. III we treat the special case of two correlated charges moving in a degenerate electron gas, and present the results of full numerical integrations of the energy-loss expressions. In Sec. IV we consider in particular the case of low velocities and we show that interference effects in the energy loss are important when the internuclear separation is not large compared with the wavelength of the electrons at the Fermi surface. Using an appropriate approximation for the dielectric constant we obtain in Sec. V an analytical expression for the energy loss at high velocities, which is compared with the numerical results of Sec. III. The proposed relation[6] between the energy loss of swift ion clusters and the partition rules for the energy loss of single charges is considered in Sec. VI, and is found not to be in general agreement with the results of this work. In Sec. VII we summarize our conclusions and finally make some comments in relation with current studies of swift molecular ions traversing solid films.

II. DIELECTRIC FORMALISM FOR THE ENERGY LOSS

Let us consider a cluster of N charges $Z_i e$, moving with nonrelativistic velocity $\vec{v}$ in a material medium of longitudinal dielectric constant $\epsilon(\vec{k}, \omega)$. Neglecting small deviations of the individual velocities with respect to the average velocity $\vec{v}$, we

can write the corresponding charge density as

$$\rho(\vec{r},t)=\sum_i Z_i e\delta(\vec{r}-\vec{r}_i-\vec{v}t), \tag{1}$$

where $\vec{r}_i$ are the positions of the corresponding charges at time $t=0$.

For nonrelativistic velocities, the electric field $\vec{E}(\vec{r},t)$ generated by the cluster is determined from Poisson's equation, which yields the simple algebraic relation

$$\vec{E}(\vec{k},\omega)=-(4\pi i\vec{k}/k^2)\rho(\vec{k},\omega)/\epsilon(\vec{k},\omega) \tag{2}$$

between the space-time Fourier transforms of $\vec{E}(\vec{r},t)$ and $\rho(\vec{r},t)$. Thus we obtain the following expression for the electric field:

$$\vec{E}(\vec{r},t)=\frac{-1}{(2\pi)^2}\sum_i Z_i e \times\int d^3k\,\frac{2i\vec{k}}{k^2}\,\frac{\exp[i\vec{k}\cdot(\vec{r}-\vec{r}_i-\vec{v}t)]}{\epsilon(\vec{k},\vec{k}\cdot\vec{v})}. \tag{3}$$

In these expressions the fields due to the external charges and the fields due to the polarization induced in the medium are summed. In particular, the force acting on the jth particle is

$$\vec{F}_j=Z_j e\vec{E}(\vec{r}_j+\vec{v}t,t) =\frac{Z_j e}{(2\pi)^2}\sum_i Z_i e\int d^3k\,\frac{2\vec{k}}{k^2} \times\left[\mathrm{Im}\left(\frac{1}{\epsilon(\vec{k},\vec{k}\cdot\vec{v})}\right)\cos(\vec{k}\cdot\vec{r}_{ji}) +\mathrm{Re}\left(\frac{1}{\epsilon(\vec{k},\vec{k}\cdot\vec{v})}\right)\sin(\vec{k}\cdot\vec{r}_{ji})\right], \tag{4}$$

where $\vec{r}_{ji}=\vec{r}_j-\vec{r}_i$. In Eq. (4) the force has been written explicitly in real form by using the physical requirement that the fields must be of real magnitudes, which imposes the following condition on the dielectric constant: $\epsilon(-\vec{k},-\omega)=\epsilon^*(\vec{k},\omega)$.

It is interesting to compare the behavior of the two terms in the integral with respect to a change in the sign of $\vec{r}_{ji}$, or what is equivalent, to compare the force that the i charge exerts on the j charge, with the force that the latter exerts on the first. We see that mutual forces acting through the term in $\mathrm{Re}(1/\epsilon)$ are opposed, and cancel out if we sum up the forces acting on the whole cluster of charges. The forces acting through the term in $\mathrm{Im}(1/\epsilon)$ are, on the contrary, dissipative (we are always considering the energy of the cluster, and not of the individual particles). Thus, the energy loss (per unit time) of the cluster of charges is given by

$$\frac{dW}{dt}=-\sum_j\vec{v}\cdot\vec{F}_j =\frac{e^2}{2\pi^2}\int d^3k\,\frac{\vec{k}\cdot\vec{v}}{k^2}\,\mathrm{Im}\left(\frac{-1}{\epsilon(\vec{k},\vec{k}\cdot\vec{v})}\right) \times\left(\sum_i Z_i^2+\sum_{i\neq j}Z_i Z_j\cos(\vec{k}\cdot\vec{r}_{ij})\right), \tag{5}$$

where we have separated the terms with $i=j$, which give the energy loss of totally independent charges, and the terms with $i\neq j$, which represent interference effects on the energy loss due to the simultaneous perturbation of the medium by the charges in correlated motion.

Since Eq. (5) consists of a sum of similar terms, the simplest case to deal with—without losing details of the general problem—is that of two charges $Z_1 e$ and $Z_2 e$ in correlated motion with velocity $\vec{v}$ and internuclear separation $\vec{r}_0$ ($\vec{r}_0=\vec{r}_1-\vec{r}_2$). We will restrict ourselves here to this case, for which the energy loss of both charges is given by

$$\frac{dW}{dt}=\frac{e^2}{2\pi^2}\int d^3k\,\frac{\vec{k}\cdot\vec{v}}{k^2}\,\mathrm{Im}\left(\frac{-1}{\epsilon(\vec{k},\vec{k}\cdot\vec{v})}\right) \times[(Z_1^2+Z_2^2)+2Z_1Z_2\cos(\vec{k}\cdot\vec{r}_0)]. \tag{6}$$

III. ELECTRON-GAS MODEL

In the following we will treat the energy loss of two correlated charges in a degenerate electron gas described by Lindhard's dielectric constant.[7]

In Eq. (6) the energy loss is given as a function of the relative orientations of $\vec{r}_0$ and $\vec{v}$. We will consider here the case in which the orientations of $\vec{r}_0$ are randomly distributed (in a statistical sense); this is, in fact, the case of greatest interest in connection with recent experimental work.[3,5] The mean energy loss $\langle dW/dt\rangle$, corresponding to random orientations of $\vec{r}_0$, may be obtained from Eq. (6) by simply replacing the factor $\cos(\vec{k}\cdot\vec{r}_0)$ within the integral by its angular average $\sin(kr_0)/(kr_0)$. Using the variable $\omega=\vec{k}\cdot\vec{v}$ we can write $d^3k=(k/v)\,dk\,d\omega\,d\phi$, and we get after integrating over the azimuthal angle ϕ

$$\left\langle\frac{dW}{dt}\right\rangle=\frac{2e^2}{\pi v}\int_0^\infty\frac{dk}{k}\int_0^{kv}\omega\,d\omega\,\mathrm{Im}\left(\frac{-1}{\epsilon(k,\omega)}\right) \times\left((Z_1^2+Z_2^2)+2Z_1Z_2\frac{\sin kr_0}{kr_0}\right). \tag{7}$$

In order to introduce Lindhard's expression for the dielectric constant of a degenerate electron gas, we write the integrals in terms of the reduced variables $u=\omega/kv_F$ and $z=k/2k_F$, where v_F is the Fermi velocity of the gas and $k_F=mv_F/\hbar$; we get, in this way,

$$\left\langle \frac{dW}{dt} \right\rangle = \frac{4\pi n e^4}{mv}\left[(Z_1^2+Z_2^2)L + 2Z_1Z_2 I \right], \tag{8}$$

with

$$L = \frac{6}{\pi\chi^2}\int_0^\infty z\,dz \int_0^w u\,du\, \mathrm{Im}\left(\frac{-1}{\epsilon(u,z)}\right), \tag{9}$$

$$I = \frac{6}{\pi\chi^2}\int_0^\infty z\,dz \int_0^w u\,du\, \mathrm{Im}\left(\frac{-1}{\epsilon(u,z)}\right)\frac{\sin(2k_F r_0 z)}{(2k_F r_0 z)}, \tag{10}$$

where $\chi^2 = e^2/\pi\hbar v_F$, n is the density of electrons, and the upper limit of integration over u is the reduced velocity $w = v/v_F$. The stopping number L is the same that appears when the energy loss of a single external charge is calculated, as done by Lindhard and Winther[1]; whereas the interference term I accounts for the additional contributions to the energy loss of correlated charges.

The dielectric constant is usually written

$$\epsilon(u,z) = 1 + (\chi^2/z^2)[\, f_1(u,z) + i f_2(u,z)\,]; \tag{11}$$

explicit expressions for the adimensional functions $f_1(u,z)$ and $f_2(u,z)$ have been given by Lindhard.[7] The integrals in Eqs. (9) and (10) extend over the regions in which $\mathrm{Im}[-1/\epsilon(u,z)] \neq 0$; thus we are led to consider the characteristic regions in the u-z plane illustrated in Fig. 1(a). The shaded region denoted by sp (because there single-particle excitations may occur) is where $f_2(u,z) \neq 0$, and corresponds to real transitions of individual electrons from the Fermi sphere to unoccupied states out of it, by absorbing the energy $\hbar\omega$ and the momentum $\hbar k$. Outside the sp region the conservation of energy and momentum forbids any single-particle excitation of free electrons. The resonance curve indicated with r in Fig. 1(a) is the region in which $\epsilon(u,z) = 0$ or $f_1(u,z) = -z^2/\chi^2$ [since $f_2(u,z) \equiv 0$ outside the sp region]. This defines a dispersion relation $u = u(z)$, corresponding to the existence of an undamped mode of collective oscillation of the electron gas (plasmon).

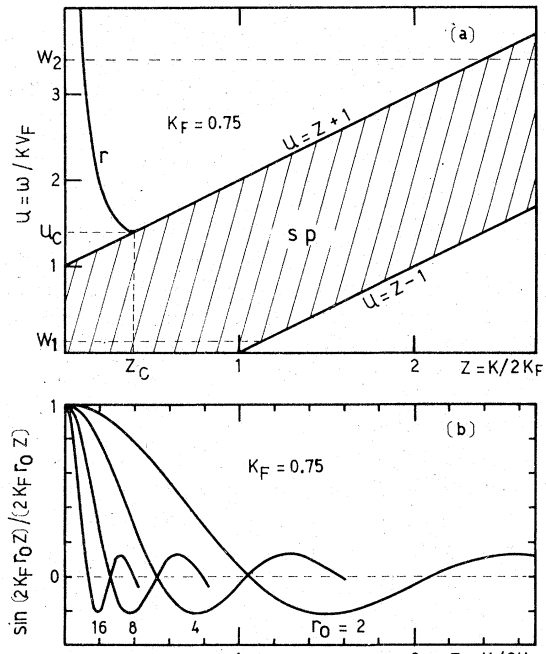

FIG. 1. (a) Regions of integration in the u, z variables. The dashed region, denoted by sp, corresponds to single-particle excitations; the curve r corresponds to resonant excitations of the electron gas (plasmons) for the case $k_F = 0.75$ a.u. (b) Interference factor $g(z) = \sin(2k_F r_0 z)/2k_F r_0 z$ as a function of z, for $k_F = 0.75$ a.u. and for internuclear distances $r_0 = 2$, 4, 8, and 16 a.u.

We can separate the contributions to the values of L and I in the following way:

$$L = L_s + L_r, \quad I = I_s + I_r, \tag{12}$$

where L_s and I_s are the contributions due to single-particle excitations. These are to be calculated by integrating Eqs. (9) and (10) over the sp region, in which $\epsilon_2(u,z) \neq 0$, and below the line $u = w$.

L_r and I_r are, on the other hand, the contributions due to collective excitations, obtained by integrating along the resonance curve r, where the double integrals reduce to line integrals. There is a minimum velocity v_c for plasmon excitation, corresponding to the point z_c, u_c (with $z_c = u_c - 1$), where the resonance curve merges into the sp region [Fig. 1(a)], that is, $v_c = u_c v_F$.[8]

In order to write the integrals for L_r and I_r it is convenient to express the energy-loss function as

$$\mathrm{Im}\left(\frac{-1}{\epsilon(u,z)}\right) = \frac{\pi\delta(u-u(z))}{\partial\epsilon_1(u,z)/\partial u}; \tag{13}$$

using this expression in Eqs. (9) and (10) we get

$$L_r = \frac{6}{\chi^2}\int_{z(w)}^{z_c} dz \left(\frac{zu}{\partial\epsilon_1(u,z)/\partial u}\right)_{u=u(z)} \tag{14}$$

and

$$I_r = \frac{6}{\chi^2}\int_{z(w)}^{z_c} dz\, \frac{\sin(2k_F r_0 z)}{2k_F r_0 z}\left(\frac{zu}{\partial\epsilon_1(u,z)/\partial u}\right)_{u=u(z)}, \tag{15}$$

where $z(w)$ is the minimum value of z that enters in the integration and corresponds to the maximum value of u: $u = w$. The integrals of Eqs. (14) and (15) were calculated starting from the point (u_c, z_c) and running over the resonance curve r, with small negative increments of z; for each new value of z the corresponding value of u was found from the condition $\epsilon_1(u,z) = 0$.

Figure 2 shows the results for I_s—the contribution of single-particle excitations to the interference term—as a function of the reduced velocity

w, for $v_F = 0.75$ a.u. and for several values of the internuclear separation r_0. We observe an approximate w^3 dependence for low velocities, whereas for large w, I_s tends to a constant value. This behavior is due to the strong variation of the interference factor $g(z) = \sin(2k_F r_0 z)/2k_F r_0 z$ shown in Fig. 1(b), which tends to cancel out the integration over large values of z (when I_s is calculated by integration over the sp region); it corresponds to the physical circumstance that "close collisions" (high-k excitations) do not contribute much to the interference effects.

The interference term I_r, corresponding to resonant excitations, is shown in Fig. 3 as a function of w. To analyze the effect of the interference factor $g(z)$ on the value of the integral we note that, for large velocities, the most important contribution to resonant excitations is given by the low-k region. With increasing w the integration of Eq. (15) covers smaller values of z, for which $g(z) \sim 1$; it may be seen that this gives place to a logarithmic behavior of I_r for sufficiently large values of w, as observed in Fig. 3. The value of w for which this dependence is reached increases with r_0. This behavior may be physically understood by observing that fast charges excite plasmons with a maximum wavelength proportional to v/ω_P (where $\omega_P^2 = 4\pi ne^2/m$ is the plasma frequency of the electron gas), so that if $v/\omega_P \gg r_0$ the excitation of the longest wavelength plasmons occurs as if the particles were united.

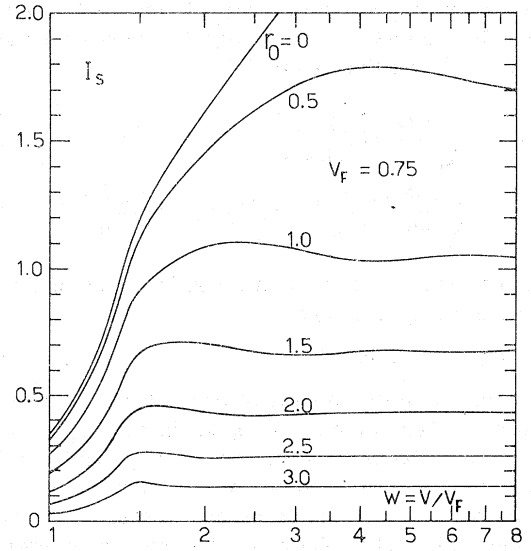

FIG. 2. Results of numerical integration of the interference term I_s (corresponding to single-particle excitations) as a function of the reduced velocity $w = v/v_F$ for $v_F = 0.75$ a.u. and for several values of the internuclear distance r_0.

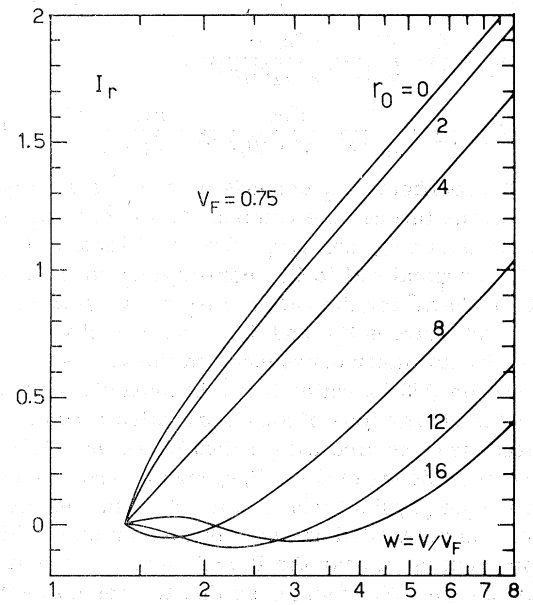

FIG. 3. Results of numerical integration of the interference term I_r (corresponding to resonant excitations) as a function of the reduced velocity $w = v/v_F$ for $v_F = 0.75$ a.u. and for several values of the internuclear distance r_0.

The results for L_s and L_r correspond to the curves for $r_0 = 0$, shown in Figs. 2 and 3; the calculation of these terms was previously described by Lindhard and Winther,[1] and so does not deserve special comments here.

IV. LOW VELOCITIES

We consider in this section the case $v \ll v_F$; that is, low reduced velocities as indicated by w_1 in Fig. 1(a). It is clear that in this case there is no excitation of plasmons, the only excitations that take place being those in a thin horizontal stripe given by $0 \leq u \leq w$ and $0 \leq z \lesssim 1$. In this region we can use the following expression for f_2 (Ref. 7):

$$f_2(u,z) = \tfrac{1}{2}\pi u\,, \tag{16}$$

which is applicable over almost the whole region of integration (except for a small triangle just above the point $z = 1, u = 0$). We then get, for $u \ll 1$, the energy-loss function as

$$\mathrm{Im}\left(\frac{-1}{\epsilon(u,z)}\right) \cong \frac{\pi}{2}\chi^2 \frac{uz^2}{[z^2 + \chi^2 f_1(0,z)]^2}\,. \tag{17}$$

Replacing this expression in Eqs. (9) and (10), and using Eq. (8), we get for the energy loss per unit pathlength

$$\left\langle \frac{dE}{dx} \right\rangle = \frac{1}{v}\left\langle \frac{dW}{dt} \right\rangle \cong \frac{4\pi ne^4}{mv_F^2} w[(Z_1^2 + Z_2^2)C_L + 2Z_1 Z_2 C_I]\,, \tag{18}$$

with

$$C_L = \frac{L}{w^3} = \int_0^1 \frac{z^3 dz}{[z^2 + \chi^2 f_1(0,z)]^2}\,, \tag{19}$$

$$C_I = \frac{I}{w^3} = \int_0^1 \frac{z^3 dz}{[z^2 + \chi^2 f_1(0,z)]^2} \frac{\sin(2k_F r_0 z)}{2k_F r_0 z}\,. \tag{20}$$

The coefficient C_L depends only on the density of electrons through the value of χ^2, whereas C_I depends both on χ^2 and $k_F r_0$. The first term in Eq. (18), proportional to C_L, corresponds to the result of Lindhard[7] for the energy loss of two slow independent charges $Z_1 e$ and $Z_2 e$, and it is characterized by its linear dependence on the velocity. Equation (18) indicates that this linearity subsists for the energy loss of slow correlated charges (because C_I is also independent of the velocity). The dependence of C_I on $k_F r_0$ may be understood in simple physical terms; for $v \ll v_F$ the electrons that can be excited are only those located in a thin spherical shell near the Fermi surface (of width $\hbar\omega \sim 2\hbar k_F v \ll E_F = \frac{1}{2} m v_F^2$), so that the relative velocity between these electrons and the external charges is $\sim v_F$; the natural parameter to describe the interferences is then the ratio between r_0 and the wavelength $\lambda\!\!\!^{-}_F = k_F^{-1} = \hbar/mv_F$ of the electrons at the Fermi surface.

The values of C_L and C_I have been calculated for $v_F = 0.6$ and 0.9 a.u., by numerical integration of Eqs. (19) and (20). In Fig. 4 we show the results

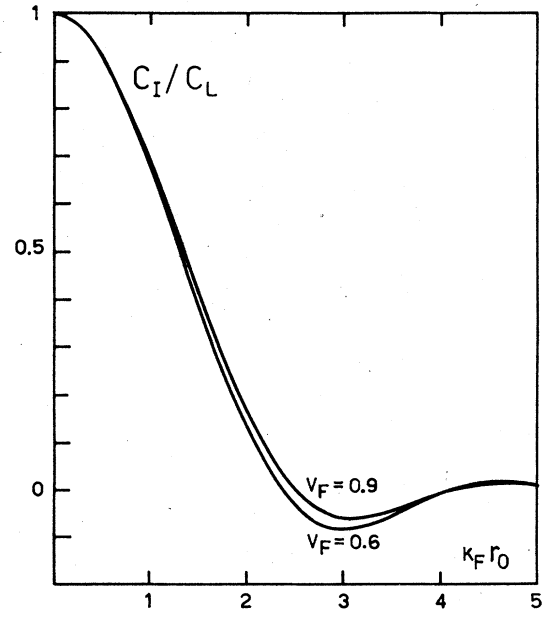

FIG. 4. Ratio between the interference coefficient C_I and Lindhard's coefficient C_L for the case of low velocities, as a function of $k_F r_0$. We show the results for $v_F = 0.6$ and 0.9 a.u.

for the relation C_I/C_L as a function of $k_F r_0$. We conclude from these results that, for low velocities, interference effects in the energy loss are important for small internuclear distances, such that $k_F r_0 \lesssim 2$.

V. HIGH VELOCITIES

In order to calculate the energy loss for velocities $v \gg v_F$ [large reduced velocities, as indicated by w_2 in Fig. 1(a)], we can use the following approximate expression for $\epsilon(k,\omega)$ (Ref. 7):

$$\epsilon(k,\omega) \cong 1 + \frac{\omega_P^2}{(\hbar k^2/2m)^2 - (\omega + i\delta)^2}\,, \tag{21}$$

where δ is an infinitesimally small positive quantity. This approximation corresponds to the case $\omega/k \gg v_F$, but otherwise arbitrary k, and describes in a very simple way the collective and individual electronic excitations. For $\delta \to 0$, we get

$$\mathrm{Im}\left(\frac{-1}{\epsilon(k,\omega)}\right) \cong \frac{\pi\omega_P^2}{2\omega_k}[\delta(\omega - \omega_k) - \delta(\omega + \omega_k)]\,, \tag{22}$$

with

$$\omega_k^2 = \omega_P^2 + (\hbar k^2/2m)^2\,. \tag{23}$$

Using Eq. (22) in Eq. (7) we get

$$\left\langle \frac{dW}{dt} \right\rangle = \frac{4\pi n e^4}{mv} \int_{k_1}^{k_2} \frac{dk}{k} \left((Z_1^2 + Z_2^2) + 2Z_1 Z_2 \frac{\sin(kr_0)}{kr_0} \right), \tag{24}$$

where the values of k_1 and k_2, for $(mv^2/\hbar\omega_P)^2 \gg 1$, are given by

$$k_1 \cong \omega_P/v\,, \quad k_2 \cong 2mv/\hbar\,, \tag{25}$$

which correspond to the extreme momentum transfers $\hbar k_1$ and $\hbar k_2$. Expressing the energy loss as in Eq. (8), we finally obtain

$$L = \int_{k_1}^{k_2} \frac{dk}{k} = \ln\left(\frac{2mv^2}{\hbar\omega_P}\right), \tag{26}$$

$$I = \int_{k_1}^{k_2} \frac{dk}{k} \frac{\sin(kr_0)}{kr_0} = F\left(\frac{r_0\omega_P}{v}\right) - F\left(\frac{2mvr_0}{\hbar}\right), \tag{27}$$

where the function $F(x)$ is given in terms of the cosine integral $\mathrm{Ci}(x)$ as

$$F(x) = x^{-1}\sin x - \mathrm{Ci}(x)\,. \tag{28}$$

In contrast with the case of low velocities, two characteristic distances appear here, $r_{\max} = v/\omega_P$ and $r_{\min} = \hbar/2mv$, which are the reciprocals of the wave numbers corresponding to minimum and maximum momentum transfer, respectively, as given in Eq. (25). They may be physically interpreted as the adiabatic distance for plasmon excitation and the minimum impact parameter for the collision between a classical particle and an electron. By comparison of r_0 with these char-

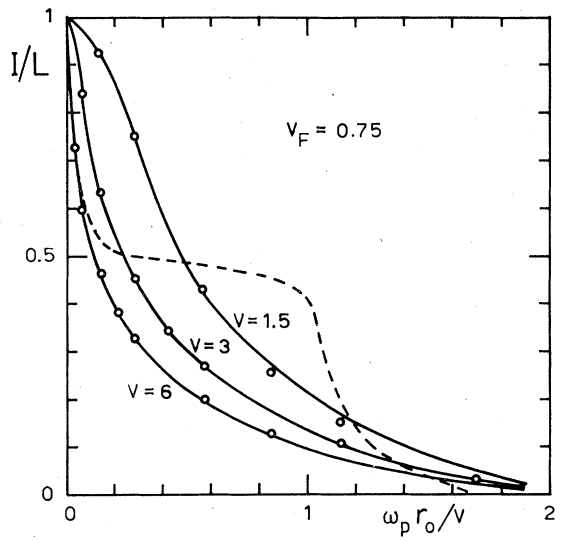

FIG. 5. Ratio between the interference term I and Lindhard's stopping number L as a function of $\omega_P r_0/v$ for $v_F = 0.75$ a.u. and for velocities $v = 1.5$, 3.0, and 6.0 a.u. The solid lines are the results of the analytical expressions [Eqs. (26) and (27)]; the dashed line corresponds to calculations by Brandt *et al.* (Ref. 3), the circles are the results of numerical integrations using Lindhard's dielectric constant.

acteristic distances we can distinguish the following limiting cases.

a. Separated charges. If r_0 is much larger than the adiabatic distance, $r_0 \gg v/\omega_P$ (and consequently $r_0 \gg \hbar/2mv$), we can use the limit $F(x) \to 0$ (for $x \to \infty$), and then $I = 0$. The energy loss is given by the first term in Eq. (8), with L given by Eq. (26). This corresponds to the case of separated charges dissipating energy independently.

b. United charges. This is the case for $r_0 \ll r_{\min} = \hbar/2mv$, so that the separation between the particles is unobservable, even for the closest collisions. Using now the limit $F(x) \to (1-\gamma) + \ln(1/x)$, for $x \to 0$, we get

$$I \cong \ln(2mv^2/\hbar\omega_p) = L\,, \tag{29}$$

and the factor between square brackets in Eq. (8) becomes $(Z_1+Z_2)^2 L$. This corresponds to the case of a simple charge of value $(Z_1+Z_2)e$.

c. Mixed case. We finally consider the following condition: $\hbar/2mv \ll r_0 \ll v/\omega_P$. Using the appropriate limits for $F(x)$ we now get[4]

$$I \cong (1-\gamma) + \ln(v/r_0\omega_P)\,, \tag{30}$$

where $\gamma = 0.577$ is Euler's constant. This limit corresponds to the important case in which the charges behave as if they were separated with respect to the closest individual excitations (because $r_0 \gg \hbar/2mv$), but they act as united charges in the excitation of the longest wavelength plasmons (because $r_0 \ll v/\omega_P$).

In Fig. 5 we show the results for the relation I/L as a function of $\omega_P r_0/v$ for different values of the velocity v. Here the continuous lines correspond to the high-velocity analytical results of Eqs. (26) and (27), whereas the circles are the results of numerical integrations using Lindhard's dielectric constant. Small discrepancies exist for $v = 1.5$ a.u., whereas for larger velocities the agreement is excellent. The dashed curve is the result of numerical integrations of Brandt *et al.*[3] on the basis of a high-velocity approximation; their curve shows a plateau for $\hbar/2mv \ll r_0 \ll v/\omega_P$, which is in disagreement with the results presented here.

We notice that, in particular, the value of $F(2mvr_0/\hbar)$ may be neglected for the case of swift molecular ions, as long as the condition $r_0 \gg \hbar/2mv$ is fulfilled, so that we can express the interference term I as a function of the parameter $r_0\omega_P/v$ alone: $I = F(r_0\omega_P/v)$.

We finally observe that the value of the ratio I/L is an appropriate quantity to describe the relation between the energy loss of correlated charges and the energy loss of independent ones; in fact, the energy loss of independent charges is simply

$$\sum_i \frac{dW_i}{dt} = \frac{4\pi n e^4}{mv}(Z_1^2+Z_2^2)L\,, \tag{31}$$

and thus the energy-loss ratio becomes

$$R = \left\langle \frac{dW}{dt} \right\rangle \Big/ \sum_i \frac{dW_i}{dt} = 1 + \left(\frac{2Z_1Z_2}{Z_1^2+Z_2^2}\right)\frac{I}{L}\,. \tag{32}$$

VI. ENERGY LOSS AND PARTITION RULES

A simple argument to estimate the energy loss of a cluster of fast charges has been given by Brandt *et al.*[3,6] on the basis of partition rules for the stopping and with the additional assumption that, if the separation r_0 between the charges Z_1e and Z_2e is such that

$$\hbar/2mv \ll r_0 \ll v/\omega_P\,, \tag{33}$$

they will act independently with respect to single-particle excitations ("close collisions") and as a single charge, of value $(Z_1+Z_2)e$, with respect to plasmon excitations ("distant collisions"). For $Z_1 = Z_2$ they give the following formula for the energy-loss ratio R:

$$R \cong 1 + L_r/L\,; \tag{34}$$

whereas according to Eq. (32) we get $R = 1 + I/L$. Using in Eq. (34) Bohr's equipartition rule for the energy loss, which states that the energies lost in close and distant collisions are approximately equal ($L_r \sim L_s \sim \frac{1}{2}L$), they find $R \cong 1.5$. On the other hand, the partition rule of Lindhard and Winther

states that $L_s = L_r + C$, where the partition constant C is a positive quantity independent of the velocity. Using this latter rule in Eq. (34) more appropriate values of R were found,[6] always smaller than 1.5.

We note that condition (33) is the one corresponding to the mixed case of Sec. V, and furthermore, that the behavior of separated or united charges in this case is rigorously correct only if we consider, respectively, high-k individual excitations, such that $k \gg r_0^{-1}$, or low-k resonant excitations in the sense that $k \ll r_0^{-1}$. However for intermediate values of k, which also contribute to the stopping, the variation of the interference factor $\sin(kr_0)/kr_0$ gives place to a more complicated behavior [see, for instance, Eqs. (10), (15), and (27)]. On the other hand, when condition (33) is fulfilled, Eq. (30) gives a quite satisfactory analytical approximation for the interference term I at high velocities. This analytical result corresponds to internuclear separations such that the charges behave as separated with respect to violent individual excitations [$r_0 k_2 = r_0(2mv/\hbar) \gg 1$], and as united charges for long-wavelength collective excitations ($r_0 k_1 = r_0 \omega_P / v \ll 1$), and where the interferences are adequately integrated over the whole intermediate range of k. As the internuclear separation r_0 increases (for constant velocity v), the contribution from the collective excitations to the interference term I decreases in a continuous manner, as our results show (Fig. 5).

In conclusion, the behavior described above does not support the existence of a general relation between the partition rules for the energy loss of a single charge, and the energy loss of a cluster of charges with internuclear distances in the range $\hbar/2mv \ll r_0 \ll v/\omega_P$. This conclusion also raises some question on the use of experimental data for the energy-loss ratio between molecular and atomic ions, in order to get a quantitative evaluation of the partition rule.[9]

VII. DISCUSSION AND CONCLUSIONS

We have calculated the energy loss of a pair of charges in correlated motions through a degenerate electron gas, within the linear response approximation, and using Lindhard's dielectric constant to describe collective and single-particle excitations. For sufficiently low velocities the energy loss of correlated charges depends on the relation between the internuclear distance r_0 and the wavelength of the electrons at the Fermi surface $\lambda\!\!\!^{-}_F$. For $r_0 \gg \lambda\!\!\!^{-}_F$ the interference effects on the energy loss become negligible, as it is physically plausible.

Numerical calculations of the energy-loss terms ($L_{r,s}$ and $I_{r,s}$), have been performed over a wide range of velocities, and the results compared with simple analytical expressions for the case of high velocities. We conclude from this comparison that the analytical expressions give an accurate approximation to the energy loss for $v/v_F \gtrsim 2$. The results of this work do not support a proposed relation between the partition rules for the energy losses of a single charge and the energy loss of a cluster of charges.

We will finally make a few comments in relation with recent studies of the energy loss of molecular ions traversing thin solid films.[3-6] In the velocity range that has been studied experimentally, both plasmon and short-range excitations of valence band electrons give the most important contribution to the energy loss. Clear evidences of molecular effects on the energy-loss ratio R have been presented, and they were interpreted as arising from collective resonance excitations.

Tape *et al.*[5] have remarked that the experimental results are clearly lower than the value $R = 1.5$, which might be expected from an application of the equipartition rule; they are instead in better agreement with estimations[6] using the partition rule of Lindhard and Winther. It must be admitted that one should not expect an accurate description of the energy loss in real media from a direct application of the results for a free electron gas; however this procedure may give a reasonable estimation for the energy loss of swift charges due to the excitation of valence electrons.[2] Using the results of our work it is quite straightforward to get an estimation of R for fast H_2^+ ions incident on carbon foils; we take $r_0 = 3$ a.u., $v = 7$ a.u., and $\omega_P = 0.9$ a.u., which are appropriate values for the experimental situation.[5,10] Using these values in Eqs. (26) and (30), we get[11] $R = 1 + I/L \cong 1 + (1.4)/(4.7) \cong 1.3$, in fairly good agreement with the measurements.

For a more consistent evaluation of the energy loss of swift molecular ions in a film, one must integrate over internuclear distances which increase, due to the mutual Coulomb repulsion between the ions, during their transit through the film. This can be readily accomplished when the analytical expressions (26) and (27) are used. Smaller effects in the case of carbon targets are the excitation of the inner shell and the width of the plasma resonance, which however, may be taken into account in an approximate manner. After performing these calculations, for H_2^+ incident on a carbon foil, good agreement was obtained with the results of Tape *et al.* within experimental uncertainties. However, from a quantitative point of view, we believe that more accurate experimental results are necessary to enable a conclusive comparison.

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