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Electronic energy-loss straggling of protons in an electron gas

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Abstract. The energy-loss straggling of protons in an electron gas is calculated by using the linear-response theory and dynamic local-field correction in the dielectric function. A comparison of the theoretical results obtained with the predictions of density-functional theory and experimental data is made.

1. Introduction

The energy loss of charged particles has received a great deal of attention for many years, since it plays an important role in investigating the elemental composition, depth distribution, and location of the lattice sites of atoms implanted in matter. The characterization of the distribution of the energy losses suffered by energetic ions in their interaction with matter requires two important quantities: the stopping power, and the energy-loss straggling, which is defined as the fluctuation in the energy loss of the ion beam in matter due to the statistical nature of the slowing-down process. Several authors [1–5] have calculated the energy-loss straggling by use of the linear-response theory and random-phase approximation (RPA) for the dielectric function, which is valid only for the weak-coupling limit of electron correlations, i.e. for $r_s < 1$ (where r_s is related to the density n_0 of the electron gas by $1/n_0 = \frac{4}{3}\pi(r_s a_0)^3$, where a_0 is the Bohr radius) [6, 7]. For metallic electron gases, with r_s ranging from 1.49 (Au) to 5.88 (Cs), the local-field correction (LFC) has been introduced in the dielectric function to take into account the exchange interaction and Coulomb correlations [8–15] at short range between electrons beyond the RPA, and this has been found to enhance the stopping power [16–20] and the straggling [21, 22] in the low-incident-velocity regime. The straggling involves a quadratic frequency moment, and hence the straggling depends more strongly on frequency-dependent information from the LFC than the stopping power. In the present paper, I make use of the dynamic LFC (DLFC) in the dielectric function to calculate the energy-loss straggling of protons in metallic electron gas, and investigate the effects of using the DLFC on the straggling. All of the results are expressed in Hartree atomic units ($e = \hbar = m_e = 1$). The results are clarified whenever other units are more expedient.

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2. Theory and results

In Lindhard's linear-response theory, which treats the screened potential to lowest order, and leads to energy losses proportional to the square of the ion charge, the energy-loss straggling, Ω^2, of an ion of charge Z_1 moving at the velocity v and traversing a path length dx in a spatially homogeneous electron gas is given by [1, 2]

$$\frac{\Omega^2}{\mathrm{d}x} = \frac{4\pi Z_1^2}{v^2} n_0 L$$
$$L = \frac{2}{\pi\omega_p^2}\int_0^\infty \frac{\mathrm{d}k}{k}\int_0^{kv}\mathrm{d}\omega\ \omega^2\,\mathrm{Im}\left[-\frac{1}{\epsilon(k,\omega)}\right] \tag{1}$$

where $\epsilon(k,\omega)$ is the longitudinal dielectric function for the electron gas with the electronic density n_0, and ω_p is the plasma frequency, $\omega_p^2 = 4\pi n_0$.

In generalized mean-field theory for an interacting electron gas, the dielectric function is expressed as [8–15]

$$\epsilon(k,\omega) = 1 - V(k)\chi_0(k,\omega)/[1 + V(k)G(k,\omega)\chi_0(k,\omega)] \tag{2}$$

where $G(k,\omega)$ is the (complex) DLFC factor, $\chi_0(k,\omega)$ the linear density–density response function of the noninteracting electron gas given in reference [1], and $V(k)$ the Fourier transform of the electron–electron bare Coulomb interaction, $V(k) = 4\pi/k^2$. The $G(k,\omega)$ function describes the effects of short-range correlation between electrons beyond the RPA. Neglecting its frequency dependence—that is, putting $G(k,\omega) = G(k)$—one gets the static LFC (SLFC) function. The imaginary part of $G(k,\omega)$ was proposed by Gross and Kohn [23], for small k-values, to be of the form

$$\mathrm{Im}\,G(k\to 0,\omega) = \frac{a\omega}{[1+b\omega^2]^{5/4}} \qquad a \propto k^2. \tag{3}$$

Dabrowski [18] extended this representation by treating the parameters a and b for arbitrary wavenumbers k; that is,

$$\mathrm{Im}\,G(k,\omega) = \frac{a(k)\omega}{[1+b(k)\omega^2]^{5/4}}. \tag{4}$$

The real and imaginary parts of $G(k,\omega)$ are related by the Kramers–Kronig relation [10, 24]

$$\mathrm{Re}\,G(k,\omega) = \mathrm{Re}\,G(k,\omega=\infty) + \mathrm{P}\int_{-\infty}^{+\infty}\frac{\mathrm{d}\omega'}{\pi}\ \frac{\mathrm{Im}\,G(k,\omega')}{\omega'-\omega} \tag{5}$$

where P stands for the principal value. Using equation (5), the parameters $a(k)$ and $b(k)$ are determined as

$$a(k) = Ck^2 b^{5/4}(k) \tag{6}$$

and

$$b(k) = \left[\frac{\mathrm{Re}\,G(k,\omega=0) - \mathrm{Re}\,G(k,\omega=\infty)}{CDk^2}\right]^{4/3} \tag{7}$$

where $C = \frac{23}{60}(4/9\pi)^{1/3} r_s$, and

$$D = \frac{2}{\pi}\int_0^\infty \mathrm{d}\xi\ (1+\xi^2)^{-5/4} \simeq 0.763.$$

In equations (4), (6), and (7), k is in units of the Fermi wavenumber $k_f = (9\pi/4)^{1/3}/r_s$, and ω is in units of $2E_f$ ($E_f = k_f^2/2$ is the free-electron Fermi energy). From the ω^3-sum rule, $\mathrm{Re}\, G(k, \omega = \infty)$ is derived as [25]

$$\mathrm{Re}\, G(k, \omega = \infty) = I(k) - \frac{2k^2}{\omega_p^2}(\langle E_{kin}\rangle - \langle E_{kin}\rangle_0) \tag{8}$$

where $\langle E_{kin}\rangle$ and $\langle E_{kin}\rangle_0$ are the expectation values of the kinetic energy per particle for the interacting and noninteracting systems, respectively, and $I(k)$ is defined as

$$I(k) = -\frac{1}{N}\sum_{\boldsymbol{q}\ (\neq \boldsymbol{k},0)}\left[\frac{\boldsymbol{k}\cdot\boldsymbol{q}}{q^2} + \frac{\boldsymbol{k}\cdot(\boldsymbol{k}-\boldsymbol{q})}{|\boldsymbol{k}-\boldsymbol{q}|^2}\right]\frac{\boldsymbol{k}\cdot\boldsymbol{q}}{k^2}[S(|\boldsymbol{k}-\boldsymbol{q}|) - 1]. \tag{9}$$

In equation (9), N is the total number of electrons in the system, and $S(k)$ is the static form factor given in equation (2.9b) of reference [25]. $I(k)$ can be written in a form suitable for numerical computation as (see also reference [26])

$$I(k) = -\frac{1}{4\pi^2 n_0}\int_0^\infty \mathrm{d}q\ q^2[S(q) - 1]\left[\frac{5}{6} - \frac{q^2}{2k^2} + \frac{(q^2-k^2)^2}{4qk^3}\ln\left|\frac{q+k}{q-k}\right|\right]. \tag{10}$$

For $\mathrm{Re}\, G(k, \omega = 0)$, we exploit the SLFC factor given in reference [27], which is an improvement on the SLFC factor proposed by Utsumi and Ichimaru [13] for large wavenumbers.

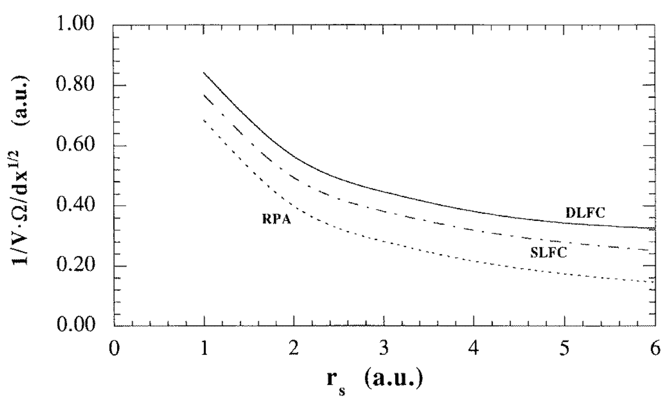

Figure 1. Predictions of the linear-response theory for $\Omega/(\mathrm{d}x^{1/2}\, v)$ for protons in an electron gas with the density specified by r_s, which is determined by using the DLFC (solid curve), SLFC (chain curve), and RPA (short-dashed curve), respectively.

For low velocities of protons $v \ll v_f$ (v_f is the free-electron Fermi velocity), by making a low-frequency-limit approximation, we find (see the appendix)

$$\frac{\Omega^2}{\mathrm{d}x} = 8\chi^2 E_f^2 C_2(r_s)\left(\frac{v}{v_f}\right)^2 \tag{11}$$

where χ^2 is defined as $\chi^2 = (4/9\pi)^{1/3}(1/\pi)r_s \approx 0.166 r_s$ and $C_2(r_s)$ is given in equation (A4) of the appendix. Figure 1 shows the variation of $\Omega/(\mathrm{d}x^{1/2}\, v)$ for protons with r_s. The results for $\Omega/(\mathrm{d}x^{1/2}\, v)$ based on the DLFC are apparently larger than those based on the SLFC in the low-velocity limit. Similarly, obvious increases over the SLFC predictions were also found for the stopping power [18]. For comparison, the predictions of density-functional theory (see also reference [28]), in which the screened potential is determined

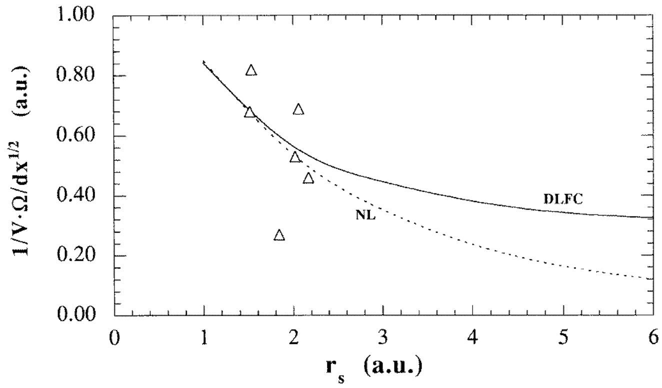

Figure 2. Comparison of the theoretical predictions for $\Omega/(dx^{1/2}\,v)$ based on linear-response theory and the DLFC (solid curve) with those of the density-functional theory (dashed curve) for a proton in an electron gas. Empty triangles represent experimental data as quoted in reference [28].

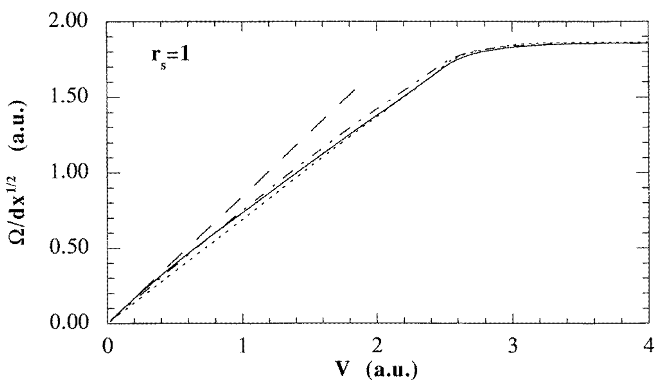

Figure 3. $\Omega/dx^{1/2}$ for protons in an electron gas with $r_s = 1$ calculated from equation (1) using the DLFC (solid curve), SLFC (chain curve), and RPA (short-dashed curve), respectively. $\Omega/dx^{1/2}$ in the low-velocity limit determined from equation (11) using the DLFC is represented by a long-dashed line.

self-consistently to all orders in Z_1, and the straggling obtained includes full nonlinear effects, are exhibited in figure 2. The nonlinear results for the straggling of protons are very close to the linear-response results based on the DLFC for the range $r_s \lesssim 2$. For $r_s > 3$, the nonlinear results are much lower than the linear ones. As shown in reference [29], at low electronic densities the nonlinear effects become important, and cause significant reduction in the stopping power and the straggling. Figure 2 also shows some experimental data [30] for the low-velocity range. Comparison of these experimental data with our calculation results makes it apparent that they agree quite well for some cases but differ for others. As pointed out in references [4, 28, 31], there are large discrepancies, not only between experimental data and theoretical predictions, but also between the different measurements for energy-loss straggling reported so far; the film thickness nonuniformity, and large crystallites frequently yield additional contributions to the energy-loss straggling, and make it difficult to compare the experimental results and the theoretical ones.

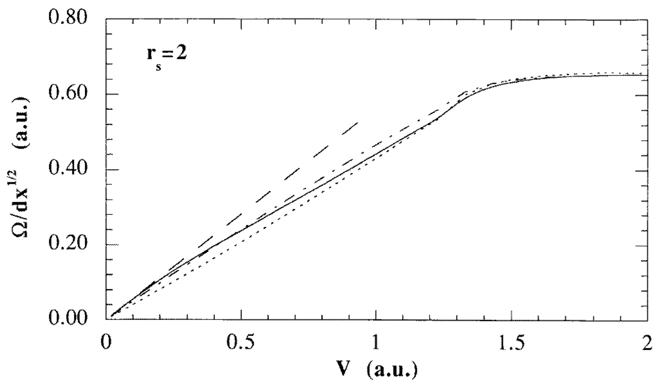

Figure 4. As figure 3, but for $r_s = 2$.

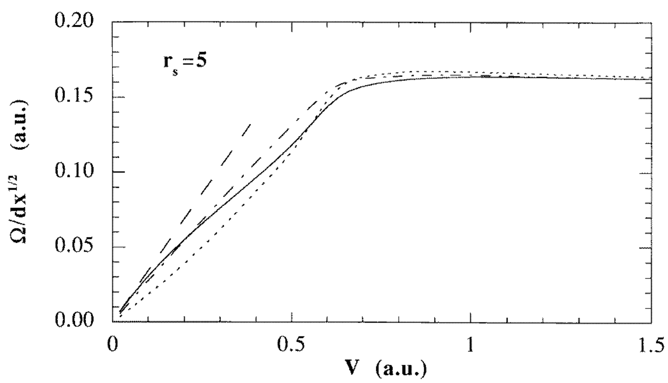

Figure 5. As figure 3, but for $r_s = 5$.

In order to show the differences among the DLFC, SLFC, and RPA dielectric theories over a wider velocity range, the electronic energy-loss straggling for protons in a uniform electron gas with $r_s = 1$, 2, and 5 is calculated by using these three dielectric functions, and integrating equation (1) numerically; the corresponding numerical results are plotted in figures 3–5. For very low velocities, the straggling determined from the DLFC is higher than that from the SLFC, but as the incident velocity increases, the former becomes lower than the latter. This is because the real part of the DLFC factor, $\mathrm{Re}\, G(k, \omega)$, decreases with the frequency ω (see equation (5)), and its contribution to the straggling is less than that obtained from the SLFC (see equations (1) and (2)). Furthermore, when the projectile velocity increases to some extent (for example, $v > 3$ for $r_s = 1$, $v > 1.5$ for $r_s = 2$, and $v > 0.7$ for $r_s = 5$), the results for the straggling based on the LFC (both DLFC and SLFC) become slightly lower than that based on the RPA. This characteristic of the straggling is unlike that of the stopping power [16, 19]. For high velocities of protons, $v \gg v_f$, one can use the plasmon-pole approximation to the dielectric function [32–34, 16], and derive (see the appendix)

$$\frac{\Omega^2}{\mathrm{d}x} \approx \frac{16}{3}\chi^2 E_f^2 \left[1 + \left(\frac{\beta'^2}{v_f^2} + \frac{\omega_p}{2E_f}\right)\left(\frac{v_f}{v}\right)^2 \ln\frac{v}{v_f}\right] \tag{12}$$

where

$$\beta'^2 = \frac{3}{5}v_f^2 - \gamma\frac{\omega_p^2}{k_f^2}.$$

By using equation (12) and noting that $\gamma = 0$ for the RPA, it is found that, for higher velocities, the straggling based on the LFC is slightly lower than that based on the RPA. However, for very high velocities, $(v_f/v)^2 \ln[v/v_f]$ approaches zero, and these two results are both consistent with the Bohr theory result $\Omega_B^2/\mathrm{d}x = \frac{16}{3}\chi^2 E_f^2$ [35].

In conclusion, we have evaluated the energy-loss straggling by using the linear-response theory and the DLFC dielectric function, and investigated the effects of using the DLFC on the straggling. The results based on the DLFC differ significantly from the predictions of the RPA and SLFC dielectric theories. It has been found that for higher densities, $r_s < 2$, the linear straggling based on the DLFC coincides well with the nonlinear one. Comparisons have been made with some of the limited, existing experimental data for the low-velocity regime, but, clearly, more accurate data are needed.

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Appendix

On introducing the dimensionless variables $z = k/2k_f$ and $u = \omega/kv_f$, $\chi_0(k,\omega)$ can be expressed, in terms of z and u, as

$$\chi_0(z,u) = -\frac{1}{\pi^3\chi^2}[f_1(z,u) + \mathrm{i} f_2(z,u)] \tag{A1}$$

where $f_1(z,u)$ and $f_2(z,u)$ are given in reference [1]. For very low velocities $v \ll v_f$, $u \leqslant v/v_f \ll 1$, by making a low-frequency-limit approximation:

$$\begin{aligned} f_1(z,u) &\approx f_1(z,0) \\ f_2(z,u) &= \pi u/2 \end{aligned} \tag{A2}$$

we have

$$L = 3E_f C_2(r_s)\left(\frac{v}{v_f}\right)^4 \tag{A3}$$

and

$$C_2(r_s) = \int_0^1 \frac{z^4\,\mathrm{d}z}{\{z^2 + \chi^2[1 - G(z)]f_1(z)\}^2} + \frac{4}{\pi}\chi^2\int_0^\infty \frac{a(z)f_1^2(z)z^3\,\mathrm{d}z}{\{z^2 + \chi^2[1 - G(z)]f_1(z)\}^2} \tag{A4}$$

where $f_1(z) = f_1(z,0)$, and $G(z) = G(z,0)$. For the RPA and the SLFC, the second term on the right-hand side of equation (A4) vanishes. Then, by taking the long-wavelength-limit approximation $f_1(z,0) = 1 - z^2/3$ and $G(z) = 4\gamma_0 z^2$, the analytical expressions for $C_2(r_s)$ are obtained as

$$C_2(r_s) = \left(1 - \frac{1}{3}\eta\chi^2\right)^{-2} \times \begin{cases} 1 + \dfrac{t_0}{2(1+t_0)} - \dfrac{3}{2}\sqrt{t_0}\,\arctan\dfrac{1}{\sqrt{t_0}} & \text{if } t_0 > 0 \\ 1 + \dfrac{t_0}{2(1+t_0)} - \dfrac{3}{4}\sqrt{|t_0|}\,\ln\left|\dfrac{1+\sqrt{|t_0|}}{1-\sqrt{|t_0|}}\right| & \text{if } t_0 < 0 \end{cases} \tag{A5}$$

$$C_2(r_s) = 1/5\chi^4 \qquad \text{if } t_0 = 0 \tag{A6}$$

where t_0 is defined as $t_0 = \chi^2/(1 - \frac{1}{3}\eta\chi^2)$ with $\eta = 1 + 12\gamma_0$.

For high velocities $v \gg v_f$, we can employ the plasmon-pole approximation to the dielectric function [32–34, 16]:

$$\epsilon(k,\omega) = 1 + \frac{\omega_p^2}{\omega_p^2 + \beta^2 k^2 + (k^2/2)^2 - \omega(\omega + \mathrm{i}\delta)} \tag{A7}$$

where $\beta^2 = \frac{3}{5}v_f^2$, and δ is an infinitesimally small positive quantity. Noticing that equation (A7) corresponds to the RPA, and applying the long-wavelength approximation to the LFC factor, it is found that the LFC dielectric function with the plasmon-pole approximation can also be expressed by equation (A7) if β^2 is simply replaced by $\beta'^2 = \beta^2 - \gamma\omega_p^2/k_f^2$, where

$$\gamma = \begin{cases} \gamma_0 & \text{(SLFC)} \\ G(k, \omega = \omega_p)/(k/k_f)^2|_{k\to 0} & \text{(DLFC).} \end{cases} \tag{A8}$$

For $\delta \to 0$, we get

$$\mathrm{Im}\,\frac{-1}{\epsilon(k,\omega)} \approx \frac{\pi\omega_p^2}{2\omega_k}[\delta(\omega - \omega_k) - \delta(\omega + \omega_k)] \tag{A9}$$

with $\omega_k^2 = \omega_p^2 + \beta'^2 k^2 + (k^2/2)^2$. By the use of equation (1), it follows that

$$L = \int_{k_{min}}^{k_{max}} \frac{\omega_k}{k}\,\mathrm{d}k = \frac{E_f}{2}\int_{k_{min}}^{k_{max}} \frac{\sqrt{(\omega_p/E_f)^2 + 4(\beta'^2/v_f^2)(k/k_f)^2 + (k/k_f)^4}}{(k/k_f)^2}\,\mathrm{d}[(k/k_f)^2]. \tag{A10}$$

Finally, we get an analytical result:

$$L = \Bigg\{\sqrt{a + b(k/k_f)^2 + (k/k_f)^4} - \sqrt{a}\ln\frac{\left[2a + b(k/k_f)^2 + 2\sqrt{a}\sqrt{a + b(k/k_f)^2 + (k/k_f)^4}\right]}{(k/k_f)^2} + \frac{b}{2}\ln\left[2\sqrt{a + b(k/k_f)^2 + (k/k_f)^4} + 2(k/k_f)^2 + b\right]\Bigg\}\Bigg|_{k=k_{min}}^{k=k_{max}} \tag{A11}$$

where

$$a = (\omega_p/E_f)^2 \qquad b = 4\beta'^2/v_f^2 \tag{A12}$$

and

$$\begin{pmatrix} k_{max}^2 \\ k_{min}^2 \end{pmatrix} = \left(\frac{v^2 - \beta'^2}{v^2} \pm \sqrt{\left(\frac{v^2 - \beta'^2}{v^2}\right)^2 - \left(\frac{\omega_p}{v^2}\right)^2}\right)\Bigg/(1/\sqrt{2}v)^2. \tag{A13}$$

By expanding equation (A11) as a series, and retaining the leading terms down to the order $(v_f/v)^4$, one finds

$$\frac{L}{(E_f/2)} = 4(v/v_f)^2 + 2A\ln\frac{v}{v_f} + [(3\ln 2 - 1)A - A\ln A] - \frac{3}{8}A^2(v_f/v)^2 - \frac{5}{64}A^3(v_f/v)^4 + \cdots \tag{A14}$$

where $A = 2\beta'^2/v_f^2 + \omega_p/E_f$.

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