Nonlinear energy-loss straggling of slow ions in solids

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Energy-loss straggling of ions with $v \ll v_F$ in an electron gas has been evaluated from phase shifts determined from nonlinear, density-functional calculations. These results differ markedly from those obtained from linear theory and show oscillations in straggling with increasing nuclear charge which can be correlated with the scattering properties of electrons at the Fermi level.

The characterization of the distribution of energy losses suffered by energetic charged particles in their interaction with matter requires, in the simplest case, two quantities: the stopping-power and the energy-loss straggling parameter. Knowledge of these quantities is relevant in depth profiling and surface analytical techniques as well as in fusion and astrophysical studies. Nonlinear, density-functional calculations of the stopping power of an electron gas for slow ions[1–4] have provided significantly better agreement with experimental data for protons than linear theories.[5] In this paper we report the first nonlinear calculation of energy-loss straggling of slow ions in an electron gas. The straggling parameter is expressed in terms of scattering of electrons at the Fermi energy by a self-consistent, effective potential evaluated within the density-functional formalism. The results exhibit oscillations of the energy-loss straggling with the incident ion nuclear charge Z_1. We show that these oscillations are correlated with those present in the nonlinear effective charge obtained from stopping power.

The energy-loss straggling of a projectile traversing a pathlength Δx in an electron gas of density n is defined by

$$\Omega^2 \equiv \langle (\Delta E - \langle \Delta E \rangle)^2 \rangle = n \, \Delta x \, W \tag{1}$$

in terms of the straggling parameter

$$W \equiv \int d\sigma \, T^2 , \tag{2}$$

where $d\sigma$ is the cross section for energy loss T in the interval T to $T+dT$.[6,7] For a projectile moving with speed v small compared with the magnitude of the Fermi velocity v_F, with the Pauli exclusion principle taken into account, the straggling parameter is given by[8]

$$W(v) = 3(vv_F)^2 \int d\sigma_0(v_F,\theta) \sin^3\left[\frac{\theta}{2}\right] , \tag{3}$$

where σ_0 is the scattering cross section and θ the scattering angle in the center-of-mass system. The results in this paper will be presented in atomic units (a.u.) in which $\hbar = m = e = 1$.

For $v \ll v_F$, the interaction of an ion with an electron gas occurs via scattering of electrons near the Fermi surface. The cross section for scattering of electrons by the screened potential of the ion may be expressed in terms of phase shifts and leads to

$$W(v) = \frac{3\pi v^2}{4\sqrt{2}} \sum_{l=0}^{\infty} \sum_{m=0}^{\infty} (2l+1)(2m+1) \times \{1 - \cos(2\delta_l) - \cos(2\delta_m) + \cos[2(\delta_l - \delta_m)]\} J_{lm} , \tag{4}$$

where $\delta_l \equiv \delta_l(E_F)$ are the phase shifts for scattering of electrons at the Fermi energy. The quantity J_{lm} is defined by

$$J_{lm} \equiv \int_{-1}^{1} d\mu (1-\mu)^{3/2} P_l(\mu) P_m(\mu) , \tag{5}$$

where the P_l's are Legendre polynomials. This integral can be expressed as a generalized hypergeometric series[9] which reduces to

$$J_{lm} = \frac{2^{5/2}\Gamma(\frac{5}{2})\Gamma(m-\frac{3}{2})}{\Gamma(-\frac{3}{2})\Gamma(m+\frac{7}{2})} \times \sum_{k=0}^{l} \frac{(-l)_k (l+1)_k [(\frac{5}{2})_k]^2}{(\frac{7}{2}+m)_k (\frac{5}{2}-m)_k (k!)^2} , \tag{6}$$

where the symbol $(a)_k$ is defined in terms of gamma functions by $(a)_k \equiv \Gamma(a+k)/\Gamma(a)$.

Phase shifts for scattering of an electron at the Fermi energy from the spherically symmetric, self-consistent potential of a static ion have been evaluated by Puska and Nieminen[2] using nonlinear, density-functional calculations. The result for the straggling parameter for $r_s = 2$ [$r_s \equiv (3/4\pi n)^{1/3}$] is shown in Fig. 1 in the form W/v^2 as a function of Z_1. The strong Z_1 oscillations in straggling are also seen in stopping powers.[2–4] In terms of the stop-

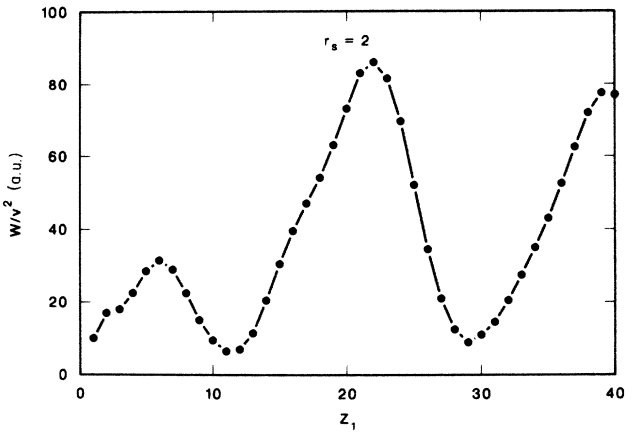

FIG. 1. Predicted variation of W/v^2 with Z_1 from density-functional calculations. These results illustrate the significant Z_1 oscillations in straggling for $r_s=2$.

ping power of an electron gas for an ion $(dE/dx)_{Z_1}$, the ion effective charge Z_1^* is defined operationally by

$$Z_1^* \equiv [(dE/dx)_{Z_1}/(dE/dx)_{Z_1=1}]^{1/2} . \tag{7}$$

Using the results of the nonlinear calculations[3] for stopping powers, we find $W/(vZ_1^*)^2=10.5$ to within $\sim 15\%$ for $1 \leq Z_1 \leq 40$ at $r_s=2$. That is, most of the oscillatory behavior in straggling shown in Fig. 1 is due to variation in ion effective charge as defined by Eq. (7). Predictions for W/v^2 for $r_s=1.5$ and $r_s=3$ are shown in Fig. 2. The change in W/v^2 shown in Figs. 1 and 2 as Z_1 or r_s varies can be understood in terms of screening of the ion and the formation of bound states as described earlier for stopping power.[3]

In Fig. 3 we show the variation in straggling with r_s. The nonlinear results are given by the curves labeled $Z_1=1$ for a proton and $Z_1=2$ for a helium nucleus. The straggling for a helium nucleus becomes less than that for an equal velocity proton for $r_s \gtrsim 2.8$; a similar crossover was found for stopping powers.[1,3] For comparison we show the predictions of linear theory in the random-phase

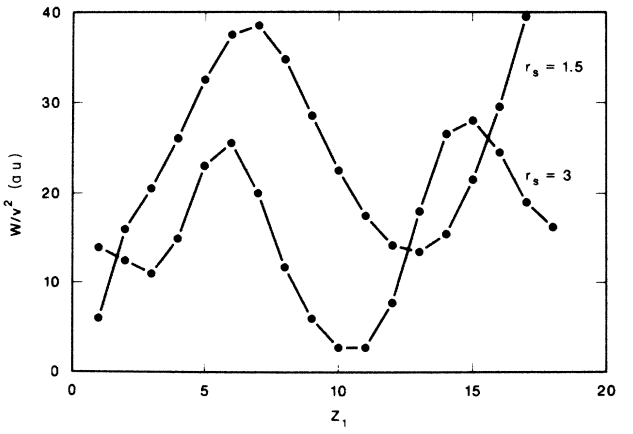

FIG. 2. Predicted variation of W/v^2 with Z_1 from density-functional calculations for $r_s=1.5$ and $r_s=3$.

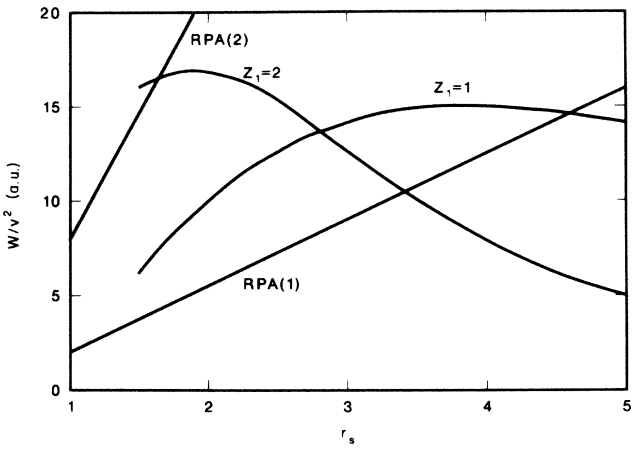

FIG. 3. Comparison of density-functional predictions for straggling with those of a linear theory for a proton [curve $Z_1=1$ vs curve RPA(1)] and a helium nucleus [curve $Z_1=2$ vs curve RPA (2)] for an electron gas with density specified by r_s.

approximation assuming a bare proton [RPA(1)] and a bare helium nucleus [RPA(2)]. These curves were calculated from[8,10]

$$W \approx 6\pi Z_1^2 (v/v_F)^2 C_2(\chi) , \tag{8}$$

where

$$C_2(\chi) \equiv \int_0^1 \frac{dz\, z^4}{(z^2+\chi^2 f_1)^2} , \tag{9}$$

$$f_1 \equiv \frac{1}{2} + \frac{1}{4z}(1-z^2)\ln\left|\frac{1+z}{1-z}\right| , \tag{10}$$

$\chi^2=0.166 r_s$, and $v_F=1.92/r_s$. The integral in Eq. (9) may also be evaluated analytically using the further approximation $f_1 \approx 1-z^2/3$. The resulting, simple form[10] for C_2 agrees well with results calculated using Eq. (10) for small r_s with the difference increasing to $\sim 8\%$ for $r_s=6$. This contradicts the assertion in Ref. 11 that very different values for C_2 result from these two different expressions (factors of 2 to 3 larger than the simple form for r_s values from 1 to 6).

The nonlinear results for straggling of protons, curve $Z_1=1$ in Fig. 3, predicts values ~ 1.6 to ~ 1.8 larger than linear theory, curve RPA(1), for $1.5 \lesssim r_s \lesssim 3$. Similarly large increases over linear theory predictions were also found for stopping powers[1,3,4] in this region of r_s values corresponding to solids used in experiments. As discussed in Ref. 3, for large electron-gas densities screening of the ion is very strong and predictions based on the nonlinear, density-functional theory tend toward agreement with those of the linear theory. This requires values of r_s much smaller than those shown in Fig. 3.

Experimental information on energy-loss straggling in the energy region considered here is not very extensive. Eckardt has measured stopping power and straggling for six materials for $Z_1=1,2$ for an energy range that extends into the region of interest here.[12] The quantities $(1/v)dE/dx$ and $\Omega/v\sqrt{\Delta x}$ are shown in Table I in atomic units. The columns labeled "expt" are from Eckardt's

TABLE I. Comparison of nonlinear, density-functional calculations of stopping-power and energy-loss straggling with experimental data from Ref. 12. All numerical quantities are in atomic units (a.u.). Conversion from a.u. to other systems of units can be made through the relations E (keV/amu)$=25v^2$ (a.u.), dE/dx (eV/Å)$=51.4\,dE/dx$ (a.u.), and $\Omega^2/\Delta x$ (eV^2/Å)$=1400\Omega^2/\Delta x$ (a.u.).

		$Z_1=1$				$Z_1=2$			
		$(1/v)dE/dx$		$\Omega/v\sqrt{\Delta x}$		$(1/v)dE/dx$		$\Omega/v\sqrt{\Delta x}$	
		expt	theor	expt	theor	expt	theor	expt	theor
Ge	$r_s=2.02$	0.23	0.26	0.53	0.54	0.42	0.41	0.98	0.69
Se	$r_s=1.84$	0.20	0.28	0.27	0.58	0.34	0.51	0.54	0.81
Pd	$r_s=1.51$	0.34	0.31	0.68	0.66	0.86	0.74	1.4	1.1
Ag	$r_s=1.53$	0.32	0.31	0.82	0.65	0.66	0.72	2.0	1.0
Sb	$r_s=2.06$	0.23	0.26	0.69	0.53	0.44	0.40	1.3	0.67
Bi	$r_s=2.17$					0.41	0.35	1.1	0.62

data[12] (Tables I and II) for $v<v_F$ and correspond to $E_0=20$ keV for $Z_1=1$ and to an average of five values for $E_0\leq 80$ keV for $Z_1=2$. The linear dependence on v for both dE/dx and Ω was confirmed[12] for $Z_1=2$. Conversion to the values in Table I assumes the bulk densities apply for the films used in the experiment.

The electron-gas densities corresponding to the materials listed in Table I are determined from effective values of r_s as tabulated by Isaacson.[13] The theoretical values (columns labeled "theor") for stopping powers are from the density-functional results of Refs. 1 and 3. Reasonably good agreement is found for all materials for both $Z_1=1$ and $Z_1=2$. Comparisons of experimental data with the results of our new calculations for straggling, Table I, show values which agree quite well for some cases but differ by a factor of 2 for others. With the exception of Se, for which both stopping power and straggling fall below our theoretical estimates, and Ge for $Z_1=1$, the calculated straggling gives results smaller than the data.

The significant contribution of foil roughness to straggling in Cu foils was described by Nomura and Kiyono.[14] No foil-roughness corrections were made to obtain the values given in Table I. Eckardt assumed such corrections were not needed since the energy spread due to foil roughness is proportional to stopping power and the straggling data for protons does not show the curved behavior seen in the stopping-power data. However, if reasonable values are taken for foil thickness and foil roughness for Ge and Eckardt's straggling data for protons (his Table I) are corrected as described in Ref. 14 using measured stopping powers, a plot of the "corrected" data shows mainly an increase in slope with no more evidence of curvature than that shown in the original data. This suggests that the straggling results in Ref. 12, and the derived values in Table I, may contain significant contributions due to foil roughness, particularly at low energies. Not enough information is given in Ref. 12 to allow a systematic reevaluation of the data to obtain the electronic contribution to straggling for a more meaningful comparison with theoretical estimates. The electronic contribution to $\Omega/v\sqrt{\Delta x}$, extracted from the data of Ref. 14 using a foil-roughness correction, yields 0.56 for protons and 1.1 for helium in reasonable agreement with theoretical values of 0.58 and 0.81 for Cu assuming $r_s=1.83$.

In conclusion, we have evaluated for the first time the nonlinear straggling of slow ions in an electron gas. The results show Z_1 oscillations correlated with the Z_1 oscillations in stopping power and differ significantly in magnitude from linear theory predictions. Comparisons have been made with some of the limited, existing experimental data but clearly more accurate data at smaller ion velocities is needed. We hope our work will stimulate new experiments in this area.

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