Energy loss of slow protons in solids: Deviation from the proportionality with projectile velocity

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Recent experimental determinations [J. E. Valdés, G. Martínez-Tamayo, G. H. Lantschner, J. C. Eckardt, and N. R. Arista, Nucl. Instrum. Methods B 73, 313 (1993)] of the energy loss of low-energy protons in Ag and Au show a departure from the velocity proportionality predicted by theory, while data for other metallic elements, such as Al, Sb, and Bi, are in agreement with this dependence. In this work we present measurements in Cu foils which show a similar behavior to that in Ag and Au. We give an interpretation of the differences between these cases due to the existence of small binding energies for the *d* electrons in Cu, Ag, and Au. We present a model based on transport cross section and density-functional theory, which introduces threshold effects in the calculation of the transport cross section of nonfree electrons, and explains the deviations from the velocity-proportional dependence.

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I. INTRODUCTION

Various theoretical calculations [1–8] of the stopping power of slow ions in solids (i.e., when the ion velocity is smaller than the average velocity of the valence electrons) predict a simple proportionality of the stopping power with the ion velocity v, usually expressed in the form

$$\frac{dE}{dx}=kv \ . \tag{1}$$

The origin of this velocity dependence can be traced back to basic properties of the free-electron gas. Therefore, this proportionality with ion velocity is expected to apply best for the case of metallic targets.

The most recent theoretical developments based on electron-gas theory [8] permit one to calculate with an increasing level of accuracy the values of the mean energy loss for particles transmitted without angular deflection through uniform media. New and interesting aspects of the physics of slow-ion energy losses, like nonlinear interactions and electron-ion-scattering processes, may be studied in the framework of density-functional theory (DFT) [4–8].

Experimental data in this low-energy range is rather scarce, and the available tabulations and semiempirical fits [9–11] do not agree well with more recent energy-loss measurements [12,13]. Additionally, a deviation from the velocity proportionality predicted by Eq. (1) has been observed for low-energy protons in Au [12], both for random and channeling incidence conditions. More recently, we have found that the velocity dependence in Eq. (1) is well satisfied for metallic foils of Al, Sb, and Bi, but not for Ag and Au [13], where systematic deviations were obtained for velocities below 0.4 a.u. Previous results for these elements have been compared in Ref. [13].

In a different situation—for incident protons on a He gas target—Golser and Semrad [14] found a strong deviation from the projectile-velocity proportionality at energies below 10 keV. They attributed this phenomenon to the existence of large binding energies in He, producing an electron-excitation threshold. In the case of metals, however, the well-known free-electron behavior that characterizes the stationary limit indicates that the velocity proportionality should be the correct limit of the low-velocity electronic stopping. Hence, the apparent deviations from this dependence, which was observed only for some metals, provide experimental information that we want to investigate in this paper.

From a detailed analysis of the existing low-energy data quoted in Ref. [13], we infer that the origin of this effect may be related to the electronic structure of each element. In order to clarify this phenomenon we measured the energy loss of protons in Cu, for which we expected a similar behavior to that in Ag and Au because of the similarities in their electronic structure.

In this paper we compare the results obtained for Cu, Ag, and Au, on one side, with those for Al, Sb, and Bi, on the other, and develop a model that, by including energy threshold effects in the scattering of nearly free *d* electrons (in Cu, Ag, and Au), provides a satisfactory explanation of all these results. The model is based in the transport-cross-section approach and agrees with earlier DFT calculations if the energy thresholds are neglected.

II. EXPERIMENT AND RESULTS

The equipment and the experimental method were described previously [13]. As in the previous work, we measured the energy spectra of the projectiles emerging in the forward direction inside a cone of $\pm 0.34°$. The nuclear stopping contribution within this small angular cone is found to be very small (the ratio between nuclear and electronic stopping, taking into account the restricted angular acceptance of the equipment, was estimated to be of the order of 10^{-4} for protons with energies between

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1 and 2 keV). Hence, this provides a method for *forward electronic-energy-loss* (FEEL) measurements.

The results reported here were obtained using a 268-Å Cu foil. This thickness was determined measuring the energy loss of 200-keV protons and using the stopping cross sections from recent tabulations by Paul, Semrad, and Seilinger [11]. We should note here that the fittings given by the STOP program [9] and the more recent Paul tabulation show some differences for this element, especially around the stopping maximum.

We determined an upper bound to the foil roughness by analyzing the energy spectra of transmitted He ions and protons at 200 keV [15]. Assuming a Gaussian thickness distribution we obtained upper bounds for the standard deviation of the thickness values which were always less than 10% of the mean foil thickness. The spectra of transmitted protons had a nearly symmetric Gaussian-like shape, which allowed an accurate determination of the mean energy loss. The errors—not including the possible error in the foil-thickness determination—did not exceed 2%.

The measured values are quoted in Table I, in the format of Ref. [13]. Figure 1 shows the results of the FEEL measurements for protons in Cu vs the mean velocity of the protons inside the foil, $\langle v \rangle = (v_0 + v_1)/2$, where v_0 and v_1 denote the (mean) incident and emergent proton velocities (this takes into account the effect of finite energy losses in the foil, cf. Ref. [13]). The experimental results obtained by Arkhipov and Gott [16] are also shown in this figure. For comparison we also include the theoretical predictions based on the density-functional theory [4–8], the semiempirical electronic-energy-loss values resulting from the STOP program [9], and the total semiempirical energy-loss values from Janni [10].

The experimental points in Fig. 1 show a deviation, with respect to the proportionality with ion velocity, similar to that found earlier for Ag and Au targets [13]. One should notice that this deviation is smaller for Cu than for Ag and Au, which is in fact what should be expected based on the physical interpretation given below.

This phenomenon cannot be explained either by nuclear-stopping contributions or by secondary effects, such as path length enlargement due to multiple scattering [17], or effective foil-thickness variation due to foil roughness [18]. As already noted, the nuclear stopping contribution was estimated to be very small for protons emerging within the small angular acceptance of the present experiments (in addition, it could be noted that this effect would lead to an increase, instead of decrease, of the energy loss). The second above-mentioned effect would also lead to a negligibly small increase of the energy loss at lower velocities. As for the possible effect of foil roughness or inhomogeneities, the analysis based on multiple-scattering theory [18] shows that it can be represented using an *effective* foil thickness $\langle \tau \rangle$—the thickness probed by the detected particles—which may be smaller than the mean thickness $\bar{\tau}$ determined using absolute techniques. However, it can be shown that [18] (i) for particles emerging with very small angular deflections the ratio $\langle \tau \rangle / \bar{\tau}$ does not depend on the beam energy (i.e., it does not affect the velocity dependence), and (ii) if the thickness is calibrated using the ion beam technique (as in the present case) no corrections for this effect are necessary. Moreover, it could be noted that these effects cannot explain even qualitatively the difference between the two groups of elements.

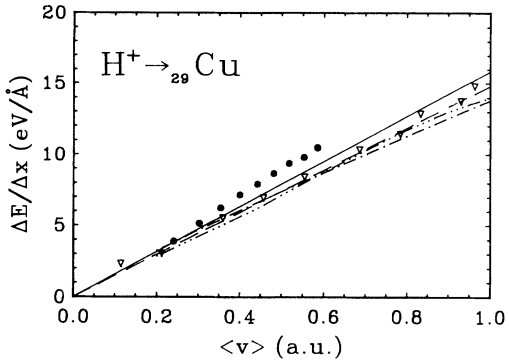

FIG. 1. Energy loss of protons in Cu vs the average velocity in the foil $\langle v \rangle$. Solid points, this work; triangles, data from Arkhipov and Gott [16]; dot-dash line, values from the STOP program [9]; dot-dot-dashed line, values from Janni tables [10]. The long-dashed and solid lines show the DFT calculations [6,7] (see the text for comments).

TABLE I. Measured values: incident energy E_0 and exit energy E_1, in kev, and energy loss $\Delta E/\Delta x$ in units of eV/Å, for protons in a Cu foil characterized by an average foil thickness $\Delta x = 268$ Å and an upper bound to the roughness coefficient $\rho_{max} = 0.10$.

E_0	E_1	$\Delta E/\Delta x$
2.017	0.987	3.84
3.024	1.657	5.10
4.019	2.357	6.20
5.027	3.123	7.10
6.026	3.918	7.87
7.026	4.714	8.63
8.026	5.523	9.34
9.026	6.407	9.77
10.025	7.242	10.38

III. PHYSICAL INTERPRETATION AND THEORETICAL MODEL

We propose here a physical interpretation of these results and develop an alternative model to describe the electronic energy loss in the low-velocity limit ($v < 1$ a.u.). First, we shall distinguish between the really *free* electrons in the metal [19] (i.e., those that determine the metallic conductivity, or the density of states at the Fermi surface) and other *nearly free* electrons that participate in electron-hole and bulk plasmon excitations, as obtained, e.g., from the maxima in electron-energy-loss experiments [20,21], although in some cases (like in the transition metals) the energy-loss function shows a complicated structure, mainly due to a superposition of interband transitions [20]. In Table II we show some characteristic electron-gas quantities: number of free electrons

TABLE II. Number of free electrons per atom N_f (from Ref. [19]); effective number of electrons per atom N_{eff} (from Ref. [21], using the free-electron gas formula, $\omega_P=(4\pi n_{\text{eff}}e^2/m)^{1/2}$, with $N_{\text{eff}}=n_{\text{eff}}/n_a$); and the corresponding Wigner-Seitz radii, r_s and r_s^{eff}; k and k_{eff} are the stopping constants calculated for each r_s value from the DFT (Ref. [7]). The outer electronic configuration of each element is also indicated.

Element	N_f	r_s	N_{eff}	r_s^{eff}	k_f	k_{eff}	Electronic configuration
Al	3	2.07	2.83	2.12	0.281	0.275	$3s^23p$
Cu	1	2.67	3.14	1.83	0.210	0.308	$3d^{10}4s$
Ag	1	3.02	7.76	1.53	0.177	0.329	$4d^{10}5s$
Sb	5	2.14	5.57	2.06	0.272	0.282	$4d^{10}5s^25p^3$
Au	1	3.01	8.21	1.49	0.177	0.330	$5d^{10}6s$
Bi	5	2.25	5.57	2.17	0.259	0.268	$5d^{10}6s^26p^3$

per atom, N_f, and electron gas parameter r_s (calculated from the free-electron density n, by $r_s^3=3/4\pi n$, where $n=n_aN_f$, and n_a is the atomic density of the metal). The first two columns show the values of N_f and r_s taken according to standard free-electron gas parameters of each metal [19]; the next two columns show the *effective* values N_{eff} and r_s^{eff} determined from experimental data of electron-energy-loss peaks, $\Delta E_P=\hbar\omega_P^{\text{eff}}$, assuming a free-electron gas formula for the effective frequency, $\omega_P^{\text{eff}}=(4\pi n_{\text{eff}}e^2/m)^{1/2}$, with $N_{\text{eff}}=n_{\text{eff}}/n_a$ [21].

These effective values can be used in a phenomenological way to represent the stopping power at energies above or below the stopping power maxima [22,23]. By comparing the values of N_f and N_{eff} in Table II one can observe that these values agree reasonably well for the three cases where the velocity proportionality was observed (Al, Sb, and Bi), while for the other cases (Cu, Ag, and Au) one finds much larger values of N_{eff}. Obviously in these latter cases the values of N_{eff} include a number of electrons that participate in interband transitions, mixed in some cases with collective excitations, although they cannot be considered as free electrons. From a study of the density of states of each of these metals [24,25] one can conclude that these are d electrons pertaining to a band that is separated in energies from the metal Fermi surface (with energies between -2 and -7 eV); we will call them *nearly free* electrons in a rather general way.

Therefore, it could be expected that at low enough projectile velocities these electrons will cease to contribute to the stopping power, due to the finite energy threshold, and thus a departure from the simple velocity dependence of Eq. (1) should occur. By contrast, the binding energies of the d electrons in Sb and Bi are in the range of 30–40 eV, so that their behavior does not overlap with the free electrons in these metals. The analysis based either on the N_f-N_{eff} differences, or in the comparison of the density of states, indicates also that the effect should be smaller for Cu than for Ag and Au, as observed here.

The most accurate approach currently available to describe the stopping power of slow ions in metals is the one provided by the density-functional theory [4–8], which incorporates nonlinear effects through a self-consistent quantum description of screening and scattering of electrons in the intruder ion, and calculates the stopping force in terms of the transport cross section (TCS) of electrons at the Fermi surface. The values of k and k_{eff} in Table II are the values of the stopping coefficient in Eq. (1), which were recalculated here according to the DFT-TCS approach, for the corresponding r_s and r_s^{eff} given in the same table. As expected, a significant discrepancy between the k and k_{eff} values is obtained for the cases of Cu, Ag, and Au.

We introduce now a simple modification to the standard DFT-TCS approach, to incorporate in an approximate way the binding-energy effects in the excitation of nearly free electrons. In the DFT-TCS description, the low-velocity stopping power can be written in terms of the transport cross section $\sigma_{\text{tr}}(v_F)$ for electrons with Fermi velocity v_{F} as follows [4]:

$$\frac{dE}{dx}=nmv_1v_F\sigma_{\text{tr}}(v_{\text{F}}) , \tag{2}$$

where v_1 is the ion velocity.

The transport cross section includes all electron scattering events allowed by the exclusion principle, with energy transfers (in the laboratory system) ranging from 0 to a maximum value $\Delta E_{\text{max}}=2mv_1(v_1+v_F)$. It may be shown from a simple analysis of binary collision events [26] that the energy transfer is given by

$$\Delta E(\vartheta)=\Delta\mathbf{p}\cdot\mathbf{v}_{\text{c.m.}}\cong mv_{\text{c.m.}}v_r(1-\cos\vartheta) , \tag{3}$$

where $\Delta p=mv_r(1-\cos\vartheta)$ is the momentum transfer, in terms of the *relative* velocity $v_r=|\mathbf{v}_1-\mathbf{v}_2|$ (with $\mathbf{v}_1$ and $\mathbf{v}_2$ denoting the ion and electron velocities), and the c.m. scattering angle ϑ. In our case the c.m. velocity can be approximated by the ion velocity, $v_{\text{c.m.}}\cong v_1$, while the electron velocity v_2 can be taken either as $v_F=1.919/r_s$ or $v_F^{\text{eff}}=1.919/r_s^{\text{eff}}$, using the values in Table II. The relative velocity will be approximated, for $v_1\ll v_2$, by $v_r\cong v_2$.

We will account now for the relatively small binding energy of the d electrons in Cu, Ag, and Au, by introducing a minimum energy transfer U, which represents the shift in the position of the d band relative to the Fermi energy. Therefore, we can now write the modified transport cross section, by considering only excitation processes with energy transfers $\Delta E(\vartheta)>U$, through a minimum scattering angle $\vartheta_0\equiv\vartheta_0(v_1)$, as follows:

$$\sigma_{\rm tr}(v_1,v_2)=\int_{\vartheta_0}^{\pi} d\Omega(1-\cos\vartheta)|f(\vartheta)|^2 \times[1+Cg(r_s)h(\vartheta)] . \quad (4)$$

Here $f(\vartheta)$ is the scattering amplitude, given by the usual expansion

$$f(\vartheta)=\frac{1}{2v_2}\sum_{l=0}^{\infty}(2l+1)P_l(\cos\vartheta)[\exp(2i\delta_l)-1] \quad (5)$$

in terms of the Legendre polynomials $P_l(x)$, and with phase shifts $\delta_l(v_F)$ which are evaluated according to the DFT [5] at the corresponding Fermi velocity. The term inside the brackets in Eq. (4) takes into account the fluctuations in the spectrum of electron-hole pairs due to local-field corrections, in the form deduced by Nagy, Arnau, and Echenique [7].

We have introduced here the minimum scattering angle $\vartheta_0(v_1)=\arccos(1-U/v_1v_2]$ [from Eq. (3), with $v_{\rm c.m.}\cong v_1$ and $v_r\cong v_2$], so that only energy transfers $\Delta E(\vartheta)$ larger than U will be included in our calculation. Obviously, if we set $U=0$, then $\vartheta_0=0$, and the transport cross section assumes the expression for a free-electron gas [7]. In particular, if local-field corrections were also neglected, we would retrieve the well-known result [8]

$$\sigma_{\rm tr}=\frac{4\pi}{v_2^2}\sum_{l=0}^{\infty}(l+1)\sin^2(\delta_l-\delta_{l-1}) . \quad (6)$$

Using Eqs. (4) and (5), and assuming typical binding-energy values U, we have calculated the stopping power of protons in each of these metals. We show in Fig. 2 (solid lines) the values obtained using the $r_s^{\rm eff}$ values of Table I, for binding energies $U=2$ and 4 eV. For comparison, the dashed and dotted lines show the values calculated without energy threshold ($U=0$) and using the

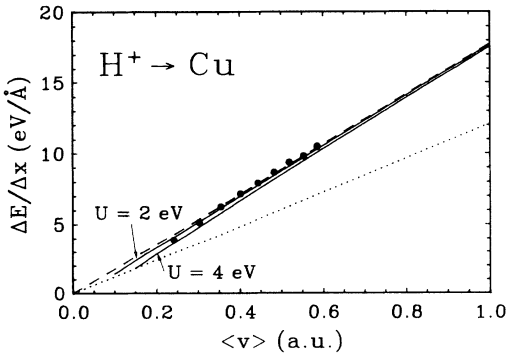

FIG. 2. Experimental energy loss values for protons in Cu (●), and calculations according to the usual DFT-TCS approach assuming $N_{\rm eff}=3.14$ ($r_s^{\rm eff}=1.83$), dashed line, and $N_f=1$ ($r_s=2.67$), dotted line, cf. Table I. The solid curves show the results obtained with the present model for $U=2$ and 4 eV. The theoretical values have been scaled by a factor of 1.12 (see text for comments).

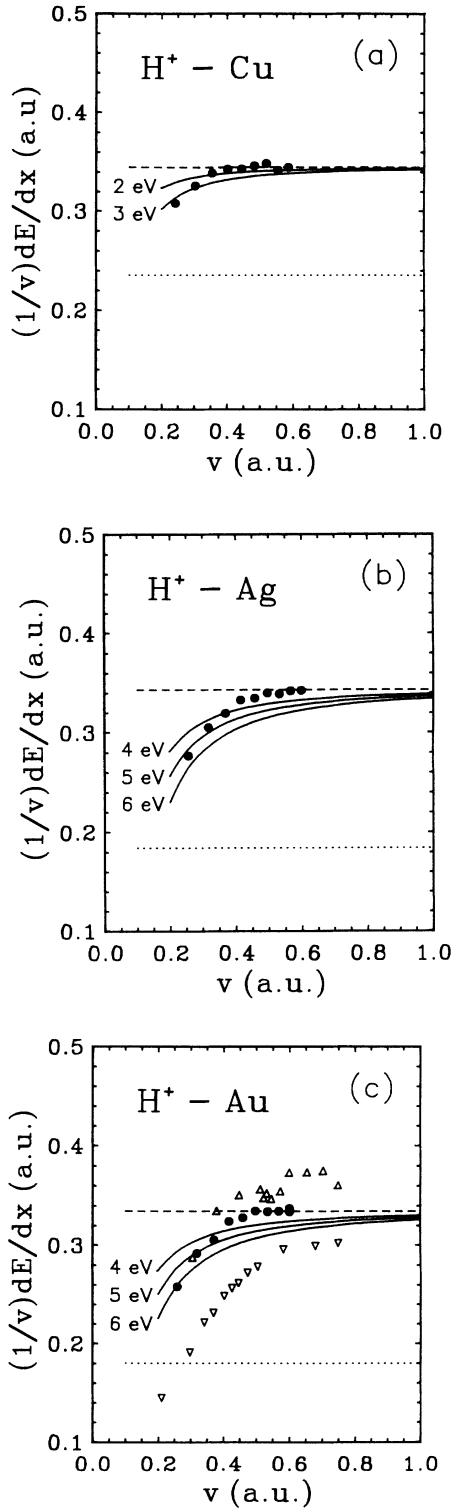

FIG. 3. Forward energy loss of protons in Cu, Ag, and Au divided by the average ion velocity $\langle v\rangle$ vs the velocity $\langle v\rangle$. ●, experimental values for Cu (this work), Ag, and Au (from Ref. [13]); the data by Blume *et al.* (Ref. [12]) for channeling (∇) and random incidence (Δ) in Au are included in part (c). The dashed and dotted lines show the standard DFT values (with $U=0$) for the $r_s^{\rm eff}$ and r_s values given in Table II. Solid curves, present model for threshold energies U in the range of 2 to 6 eV, as indicated for each element. The theoretical values have been normalized to the highest experimental points.

r_s^{eff} and r_s values, respectively. To illustrate more clearly the velocity dependence, the calculations have been normalized here as described below. The results using r_s^{eff} are in good agreement with the velocity dependence of the data at higher energies, while those using r_s give a theoretical lower limit to the friction force at very small velocities. The data points seem to indicate a transition between these two limiting values. The deviation below the velocity proportionality at lower velocities is in fair agreement with calculations including small threshold energies ($U \cong 2-4$ eV).

Figure 3 shows the deviation from the proportionality in the velocity dependence, for the cases of Cu, Ag, and Au. Here we show the results for the stopping "constant" k in Eq. (1); the experimental points show the values of

$$k_{\text{expt}} = \frac{1}{\langle v \rangle} \frac{\Delta E}{\Delta x} . \tag{7}$$

In the case of Au [Fig. 3(c)] we have included also the data by Blume, Eckstein, and Verbeek [12], both for channeling and random incidence. As expected, the deviation from the strict velocity proportionality becomes more important the lower the ion velocity.

The curves in Fig. 3 show the values of our model, calculated from Eqs. (4) and (5), for U values in the range of the d electron binding energies for each of these metals [24,25]. Here the theoretical values have been normalized to the highest experimental points in the figure; this normalization serves only to illustrate the behavior more clearly with respect to the velocity dependence, since the absolute values for $U=0$ and asymptotic values for $U \neq 0$ agree with the highest-energy data points within 2–12 % (cf. Figs. 1 and 2). A fairly good qualitative agreement in the deviation from the velocity proportionality in the three cases is obtained.

IV. CONCLUSIONS

We have measured the forward electronic-energy-loss of protons with energies from 2 to 10 keV in thin Cu foils, and have analyzed in particular its dependence on ion velocity. We observed a deviation of the experimental energy-loss values from the velocity proportionality predicted by theoretical models in the low-velocity range. This behavior is similar to the one observed previously for other elements of the same group in the periodic table, but of smaller magnitude (as expected from the present physical interpretation), and is distinct from the ones for Al, Sb, and Bi, which show a velocity-proportional FEEL as predicted for free-electron-like materials.

The nonproportional dependence in Cu, Ag, and Au is attributed to the existence of weakly bound d electrons in these elements, with binding energies of a few eV. These electrons can be regarded as nearly free with respect to swift ions, and produce the main contribution to the energy loss at energies close to the stopping-power maximum, via plasmon and electron-hole pair excitations. However, in the low-velocity limit studied here, electron-hole excitations are in part counterbalanced by the finite thresholds that separate the d bands from the Fermi energy (while plasmon excitations have an even more stringent threshold at velocities close to the stopping-power maximum). A simple criterion for distinguishing materials where these effects should be expected is given by the comparison between the *effective* number of electrons per atom N_{eff}, as determined experimentally from the maxima in the electron energy-loss spectra, and the number of conduction or *free* electrons per atom N_f, given by the standard tabulations for metallic elements.

We present a model based on the transport-cross-section approach and the DFT calculation of ion-electron scattering processes in a free-electron gas. The accurate nonlinear description provided by the DFT is considered here a good starting approximation for the scattering of nearly free electrons in metals. We introduce the effect of energy thresholds by excluding those scattering events where the energy transfer is smaller than a given excitation energy U.

The parameters of the model are the r_s values and the threshold energy U. To maintain a link with previous models, we used here the same values of r_s^{eff} as determined by empirical considerations on the number of electrons participating in plasmon and electron hole excitations [20–23]. The U values have been obtained from previous studies of the density of states for the d electrons in Cu, Ag, and Au [24,25].

The comparison with experiments is qualitatively good, and in particular we can notice the following: (i) the agreement of the measured FEEL values with the original DFT-TCS calculations is within 2–12 %; the discrepancies may be partly due to uncertainties in the use of r_s^{eff} values or because of limitations in the DFT-TCS model; however, the systematic deviations from the velocity proportionality in Cu, Ag, and Au are beyond the experimental uncertainties; (ii) since we are mainly interested here in the velocity dependence rather than the absolute values, we have normalized the results as in Figs. 3(a)–3(c), which shows more clearly the systematic decline of the FEEL values with respect to the velocity proportionality. We show that this change in the velocity dependence can be qualitatively explained by the threshold effect.

A more quantitative description of this effect cannot be given at this time, but we can note the following: (i) the original DFT calculations reproduced here apply to a homogeneous free-electron (jellium) system, and therefore consideration of both the energy thresholds and the different spatial distributions of the s, p, and d electrons in real metals may be important for a more accurate description; (ii) perhaps some improvement of the present model could be obtained by incorporating in a more realistic way the information on the density of states for each particular element (here represented in an average way by the parameter U). This approach may also be useful for applications to materials with larger energy gaps, where larger threshold effects should be expected. Work in this direction is currently under way.

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