Stopping power of solid targets for slow helium ions

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The energy loss of slow helium ions interacting with different solid targets is determined experimentally. By using a phase-shift sum rule for the amplitude of the dipolar backflow current, a consistent screened potential of these projectiles in an electron gas is constructed and applied to the kinetic model of stopping. The theoretical description reproduces well the experimental data over a broad range of the effective electron density including the accessible dilute limit. The combined theoretical and experimental analysis results in a consistent physical picture for the stopping of slow helium ions in solids.

The energy dissipation rate of matter for energetic particles is of recurring interest in physics. The understanding of the slowing down of these particles is of great fundamental and applied relevance. Even in the case of a specific particle with a given velocity v, it is important to know the dissipation rate for many targets accurately in order to justify our ideas for a complete description and establish a generally acceptable physical picture.

As in most areas of research a general approach to energy dissipation involves a model for the solid target. For the latter we use here a degenerate electron gas, characterized by its theoretical Wigner-Seitz radius r_s.[1] An experimental characterization of this (effective) density parameter is provided by the measured plasma frequencies $\omega_p^2=3/r_s^3$ of different (real) solid targets.[2]

When the velocity v of a heavy particle is small compared to the Fermi velocity $v_F=(9\pi/4)^{1/3}/r_s$, the energy dissipation is due to the excitations of electron-hole pairs. The rate (denoted by dE/dt) of this is proportional to v^2 and can be expressed in terms of the phase shifts generated by the spherical potential presented by the heavy particle in the following way:[3]

$$\frac{dE}{dt}=v^2 n_0 v_F \sigma_{tr}(v_F)=v^2 Q(v_F). \tag{1}$$

The transport collision frequency (Q) is proportional to the density of electrons (n_0) and to the transport cross section (σ_{tr}) at the Fermi level. The conventional stopping power (longitudinal retarding force) is $(dE/dt)/v$ for a given constant velocity.[4] Alternatively, the energy loss can be viewed in terms of the momentum transfer to a uniform current of electrons scattering from a fixed potential. This is an example of the Das-Peierls theorem which states that the total force on the impurity is proportional to the additional resistivity it causes.[5]

Of course, to implement the above-formulated approach for the case of charged external projectiles, knowledge of the scattering phase shifts and therefore the effective scattering potential is essential. The distortion of the electron density (and the determination of the screening) by a *static* external charge is a classic subject in solid-state theory.[6] It is treated in a self-consistent manner, by introducing physically reasonable approximations, within the Kohn-Sham (KS) orbital version of density functional theory (DFT). Neither the KS single-particle wave functions (stationary ''free'' electron states in a space that is empty except for an effective potential at the static center[7]) nor energies (except, in our case, the Fermi energy) have any observable meaning; one may consider the KS wave functions as *density-optimal* ones.[8]

The response of the electron system to a *moving* charge produces a velocity (v) proportional backflow current which, in the examined low-velocity limit ($v<v_F$), can be viewed as a purely reactive effect.[9] This concept is based on the particle conservation equation for the charge *and* current induced in the electron gas. The dipolar part[3,9,10] of the induced backflow current has an universal amplitude. It is given by[9,10] the magnitude of the external charge (Z) and is the result of perfect screening (it is an obvious consequence of the long-range Coulomb interaction) which requires that the induced backflow identically cancel the longitudinal part of the charged impurity current. The concept provides the natural link between the scattering phase shifts *at* the Fermi level (required in the energy dissipation rate) and an effective scattering potential $V(r)$ presented by the slow, charged projectiles. The connection is expressed by the following sum rule:[10]

$$Z=\frac{1}{\pi}\sum_l (2l+1)\sin 2\delta_l + \frac{4}{\pi}\sum_l (l+1)^2 \sin\delta_l \sin\delta_{l+1}\sin(\delta_l-\delta_{l+1}). \tag{2}$$

In a combined theoretical and experimental study we explore the capability of a description based on this rule for the problem of He ion interaction with different solid targets. It is found that the potential construction based on the concept

of the induced backflow results in a consistent explanation for measured stopping powers of the slowly moving external charge.

We have performed He energy loss measurements on 17 elements (Ag, Al, Au, Bi, Ca, Cd, Cr, Fe, Ge, Mg, Pb, Rb, Si, Sn, Sr, Y, and Zn) with the effective[2] r_s values ranging from 1.6 (Ag) to 5.5 (Rb). In addition, our targets show very different electron configurations in the outermost shells; among them are simple metals, transition metals, and semiconductors. The high reactivity of some of these elements requires an *in situ* production[11] and an *in situ* calibration[12] of the target layer. For each element a minimum of two targets has been produced. The pressure during evaporation extended from 1×10^{-5} mbar (Sr), 6×10^{-6} mbar (Al), and 1.5×10^{-6} (Ca) to 7×10^{-9} mbar (Cr); most of the evaporation processes took place at a pressure in the intermediate 10^{-8} mbar range. To absorb that gas which is released during the evaporation process we have mounted a liquid-nitrogen-cooled baffle above the evaporation source: the condensed material effectively acts as selective getter. The base pressure in the target chamber was 3×10^{-10} mbar. Depending on the sticking probability, the adsorbtion rates on the targets were in the order of one monolayer per hour. Therefore critical targets had to be evaporated and measured the same day.

Basically, there are two experimental layouts to measure stopping power: we favor backscattering experiments over experiments in transmission geometry. The advantages are[13] a greater variety of useful backing materials, a more simple target preparation and calibration, and insensitiveness to surface contamination. Shortcomings are the more elaborate evaluation procedure[14] and, due to plural and multiple scattering, a rather high lower limit of the available energy range. For light target elements this limit is roughly at 50 keV and for heavy elements at 100 keV, respectively. So we need for some of the elements a prescription as to how to extrapolate our data towards smaller velocities. On the one hand, it is well accepted that in metals below the Fermi velocity v_F the energy loss of helium projectiles is strictly proportional to the velocity.[4,15] On the other hand, Janni[16] stated for protons that the linear behavior extends almost to the Bohr velocity (corresponding energy for He ions: 100 keV) quite independent of v_F; this may also apply to helium with its smoother stopping function.[14]

The effective screened potential required in our theoretical model is constructed in the following way. Within the framework of an electron-gas description of a real solid we shall use a one-parametric (α) trial potential

$$V(r)=-\frac{1}{r}e^{-2\alpha r}(1+\alpha r) -\frac{1}{r}e^{-\alpha r}\left(1+\frac{3}{4}\alpha r+\frac{1}{4}\alpha^2r^2+\frac{1}{8}\alpha^3r^3\right). \quad (3)$$

The form in Eq. (3) would correspond to a *formal* electrostatic solution for a ''neutral atom,'' with $1s$- and $2s$-type normalized wave functions for the screening electron density. The ground-state energy (E_{gr}) of this ''atom,'' in an independent electron approximation, is $E_{gr}=-(5/4)\alpha$. With $\alpha=2.3$ one gets a very *acceptable* characterization of

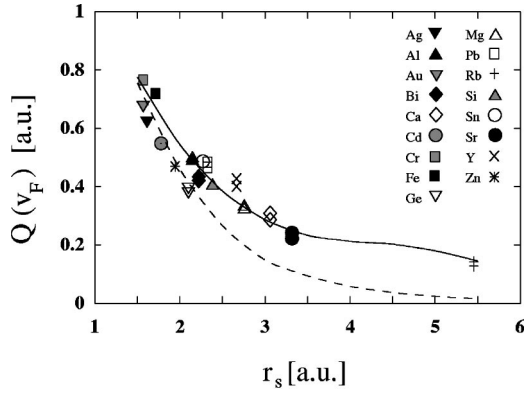

FIG. 1. The transport collision frequency $Q(v_F)$, as a function of the density parameter r_s. The theoretical curves are based on the consistent [Eq. (3) with Eq. (2)] and rigid [Eq. (3) with $\alpha=2.3$] potentials and plotted by solid and dashed curves, respectively. The experimental data are denoted by different symbols and the target-symbol correspondences are given in the inset.

density-optimal static DFT results for the energy[17] and with the corresponding $V(r)$ for the transport collision frequency[4,17] (see Fig. 1) for the whole metallic range. Note that the ground-state DFT (Refs. 4 and 17) calculations for the *static* embedded ''atom'' with filled KS bound state satisfy the Friedel sum rule of extra states[7] generated below the Fermi level.

We use the above one-parametric trial form in the present backflow-mediated construction, too. We have determined the phase-shift values from the numerical solutions of the Schrödinger equation with $v_F^2/2$ scattering energy using $V(r)$. By forcing the one-parametric form to satisfy the sum rule of Eq. (2) we obtained the consistent[9,10] screening, via the parameter $\alpha(r_s)$, of slowly *moving* (for the need of a proper treatment, see Ref. 18) ions as a function of the density parameter r_s. The phase shifts obtained in such a way are used in the basic equation (1), to calculate energy dissipation rates. We add, for completeness, two remarks. The rigid trial potential ($\alpha=2.3$) gives, by the corresponding numerical phase shifts, nearly zero values for the right-hand side of Eq. (2) in the lower density ($3<r_s<6$) range. For a high-density electron gas ($r_s<1$, i.e., the Sommerfeld parameter $Z/v_F<1$, for $Z=2$) one can obtain an analytical result for $\alpha(r_s)$. In this, first-order Born case Eq. (2) has the following form:

$$Z=\frac{v_F}{\pi^2}V(q=0), \quad (4)$$

in which $V(q)$ is the Fourier transform of Eq. (3). Simple calculation gives the $\alpha(v_F)=(15v_F/\pi)^{1/2}$ Thomas-Fermi-like, perturbative result.

Now, we turn to the presentation of our experimental and theoretical results obtained for helium ($Z=2$) interacting with different solid targets. Figure 1 shows the transport collision frequency $Q(v_F)$ as a function of the density parameter r_s. The experimental data are denoted by different symbols and plotted at the effective[2] Wigner-Seitz radius.

To obtain the gradient of the stopping power from experiment, we made use of both (see above) extrapolation procedures, giving us in some cases two slightly different values

for the experimental low-velocity stopping power. In Fig. 1 *both* extrapolated data are shown. To estimate the overall accuracy we have to add geometrically a 2% error for the determination of energy and energy loss, and 2% for the evaluation procedure;[14] we get a random error of 2.8%. This has to be summed up arithmetically with systematic errors: 1%–2% for the areal mass density of the target and 1%–4% for target impurities, mainly for highly reactive target elements. The total error is 5% for elements stable in ambient air and going up to 9% for rubidium.

The theoretical results obtained via the present consistent model with variable $\alpha(r_s)$ screening parameters and via a rigid-potential approximation ($\alpha=2.3$) are exhibited in the figure by solid and dashed curves, respectively. The effective potential, based on a nontrivial constraint for scattering phase shifts, Eq. (2), results in transport collision frequencies which are in very nice agreement with the present experimental data (for other, confirmative, recent ones, see Ref. 19). Especially, the agreement for the nearly free electron materials Al, Mg (see Ref. 15, too), and Rb is gratifying.

Note that to obtain enhanced stopping powers from intermediate to lower electron densities (i.e., from Al to Rb) when using a rigid-potential (dashed curve) DFT-like approximation *within* the kinetic theory particular smaller r_s values would be needed (cf. Ref. 15). On the other hand, it is well known that for positive unit charges (proton,[4,20] muon[21]) such an extra ''metallization'' is not needed; indeed the theoretical results were in nice agreement with corresponding experiments, at standard effective[2] r_s values. Consequently, the DFT calculations may need modifications beyond the ground-state concept to allow *ionic* charge states in real metallic targets for moving[18] projectiles, practically for the corresponding $2<r_s<6$ range of the Wigner-Seitz radius.

In conclusion, the energy loss of slow helium ions interacting with different solid targets is investigated in a combined theoretical and experimental study. Using a nontrivial sum rule of scattering phase shifts, based on the backflow concept, a consistent effective potential is constructed and applied to the kinetic theory of the energy loss. The obtained theoretical and experimental values for the transport collision frequencies are in good agreement. The effective r_s values determined from measured plasma frequencies provide a solid base to the quantitative interpretation of the stopping power. The main physical conclusion, similarly to the one in Ref. 21, is the following: our calculation shows that for *moving* ions in real solid targets the problems of charge-state and related screenings need a consistent attempt.

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[1] Atomic units (a.u.) $e^2=\hbar=m_e=1$ are used throughout.
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