Observation of a Striking Departure from Velocity Proportionality in Low-Energy Electronic Stopping

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By time-of-flight spectrometry we have measured the energy loss of hydrogen projectiles in helium gas. We show that for projectile velocities v_1 below the Bohr velocity the presence of an appreciable minimum excitation energy leads to a significant deviation of the electronic stopping cross section from the v_1 proportionality generally assumed; at 4 keV a dependence on roughly the third power of v_1 is found.

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It is commonly assumed, e.g., in published tabulations,[1-4] that at low projectile velocities v_1 the electronic stopping cross section for ions

$$\varepsilon(v_1) = -\frac{dE_1}{N\,ds} = \int_{T_{\min}} T\,d\sigma(T,v_1) = \bar{T}\sigma(v_1) \qquad (1)$$

is proportional to v_1. Here, E_1 is the kinetic energy of the projectile, N is the atomic target density, and s is the path length. T is the energy transferred to the target and $d\sigma$ denotes the cross section corresponding to T; integration runs over all possible values of T, starting from a minimum excitation energy $T_{\min}$, the threshold energy; $\bar{T}$ is the mean transferred energy. However, it is well established[5,6] that in the case of excitation or ionization of target electrons via Coulomb interaction the cross section σ is not proportional to v_1. For nonzero $T_{\min}$ this should apply to ε as well when this energy-loss channel dominates over losses due to charge exchange, since for small values of v_1, $\bar{T}$ is close to $T_{\min}$ and almost constant.[7] For the special case of protons (at fixed charge state) in H_2 a similar behavior of ε has been recently derived by Phelps[8] from individual collision processes (see also the discussion in Ref. 9). The influence of a nonzero $T_{\min}$ was first considered by Fermi and Teller[10] and worked out in Refs. 11–13. We have found by theoretical arguments[11] that $\varepsilon(v_1)$ should be modified for projectile energies up to $500T_{\min}$.

In the following we provide experimental evidence that the stopping cross section of helium gas for hydrogen isotopes strongly deviates from v_1 proportionality. Helium has been chosen because of the largest possible $T_{\min}$ (19.8 eV) and because of the large mismatch in the ground-state levels of projectile and target (H, −13.6 eV; He, −24.6 eV); this will reduce energy-loss processes made possible by charge exchange (the Massey criterion, see, e.g., Ref. 14).

To measure ε of gases for light ions we have developed a time-of-flight (TOF) facility. Here, only a short description of the apparatus and of the evaluation procedure is given; details will be published in a forthcoming paper.

The beam of hydrogen ions is produced in a duoplasmatron ion source and accelerated to energies from 6 to 22 keV. After collimation, the beam is swept over the chopper baffle in a rectangular pattern, using two electrical deflectors working in orthogonal transverse directions. So, the beam passes the 0.25-cm-diam hole in the chopper baffle only once per cycle (this defines the start signal for TOF) and therefore allows us to optimize the deflecting-voltage change for speed without compromise for the return phase.[15] Neutral projectiles are removed from the beam by a 60° bending magnet downstream. The chopper baffle also acts as the entrance into the target gas cell; differential pumping provides a pressure reduction by 4 orders of magnitude within a distance of 5 cm. The total length of the gas cell is 210 cm; fifty equally spaced antiscattering baffles (diameter 6 cm) absorb projectiles scattered towards the wall. The exit baffle (diameter 0.4 cm) is covered with a carbon foil of about 4 $\mu g/cm^2$ in order to keep the vacuum for the stop detector [microchannel plate (MCP)] below 2×10^{-6} mbar. As the carbon foil is only 3 cm from the stop detector, the additional energy loss of the projectiles (between 1 and 2 keV, depending on velocity) gives only a minor change in the total TOF, which has been properly corrected for. This energy loss puts a limit of about 4 keV on the target-exit energy for protons, below which TOF spectra need correction due to MCP efficiency.[16] Energy-loss straggling in the foil increases the total system resolution to about 1 ns. The pressure of the target gas is measured by a capacitance manometer and electronically regulated within 1×10^{-4} mbar.

The energy loss is derived from the difference in TOF between evacuated ($<5\times10^{-6}$ mbar) and pressurized gas cell. As the target gas is distributed along the whole flight path, the energy-loss spectrum can be derived from the measured time spectrum only via the corresponding transport equation. However, by numerical integration of this transport equation we can show that the *first moment* of the TOF spectrum can be converted into the corresponding *first moment* of the energy spectrum by simply integrating the equation of motion, assuming the

continuous-slowing-down approximation. The result of this evaluation procedure deviates from that of the transport equation by less than 1% for all projectile energies and target pressures considered in this work.

As a basic test of our TOF apparatus we have measured ε of amorphous carbon for hydrogen isotopes, using a set[17] of four carbon foils with calibrated[18] thicknesses between 2.7 and 7.3 $\mu g/cm^2$, located 140 cm upstream from the MCP. Our result is in excellent agreement with a forthcoming report[19] (within 3.6% at 4 keV and 1.4% at 10 keV). This semiempirical fit interpolates between the only experimental data below 3 keV, by Overbury *et al.*,[20] and the energy region above 10 keV, where a number of measurements exist (Ref. 2 and references therein).

To guard against errors inherent to gaseous targets we furthermore measured ε of H_2 and D_2. The close matching of projectile and target ground-state energies strongly enhances charge-changing processes, which will show up in a completely different dependence of ε on v_1, compared to that expected for helium. We find $\varepsilon \propto v_1^{0.5}$ at 20 keV and $\varepsilon \propto v^{0.9}$ at 4 keV. The use of different projectile isotopes (p,d) at the same velocity as well as different target isotopes and a variation of target density by more than 1 order of magnitude have little influence on ε. As an example, we cite all four combinations at 6-keV equivalent proton energy, i.e., at the energy of a proton having the same velocity as the projectile (ε in units of 10^{-15} eV cm^2): p on H_2, 3.10; d on H_2, 3.16; p on D_2, 3.07; and d on D_2, 3.14. We consider this a strong confirmation that there are no essential flaws in the experimental procedure. A comparison to the only other experimental data[21] again yields agreement within 4% between 7 keV (lowest energy in Ref. 21) and 20 keV. The corresponding prediction for the velocity dependence by Phelps[8] for neutral H projectiles in H_2 up to 10 keV is also in fair agreement; his absolute values are significantly lower, 20%, which is still within the accuracy claimed by the author.

The errors taken into account in the measurement and evaluation of the energy loss for helium are those due to the drift of the beam energy, changes of the time shift caused by the exit foil under gas load, and uncertainties of the first moment of the time spectra: These errors add to 7% at 4 keV and to 2% at 20 keV. Furthermore, we assume the random error of the target areal density to be 5% at 4 keV (where smaller gas densities had to be used) and 2% at 20 keV. All errors mentioned so far represent standard deviations and are summed geometrically. Systematic errors considered are the uncertainties in the effective length of the flight path and in the time calibration, which will contribute about 1.5%, the manometer accuracy (2%, inclusive of the transpiration[22] effect), and possible impurities (less than 1%) in the target composition; these contributions are added algebraically.

In Fig. 1 we present the stopping-cross-section values

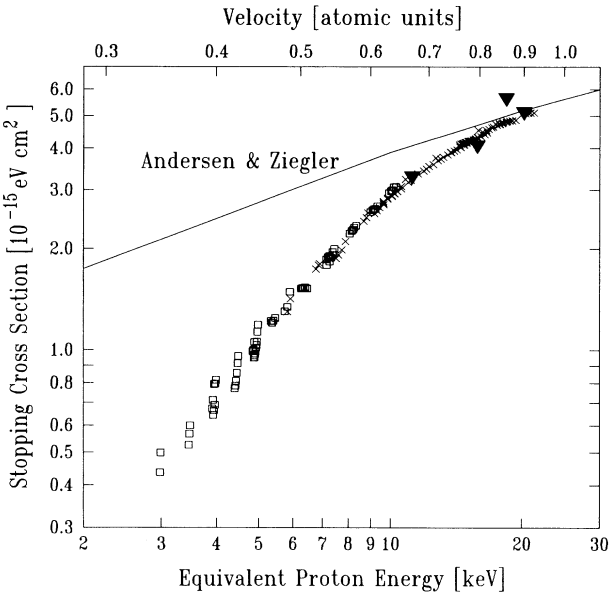

FIG. 1. Stopping cross section of He for protons (×) and deuterons (□); also shown are the data of Phillips (Ref. 21) (▼). The solid line is the fit from Andersen and Ziegler (Ref. 2), who assume v_1 proportionality below 10 keV. The x axes give the velocity in atomic units and the equivalent proton energy, respectively.

of He gas for 164 data points with protons and deuterons of initial energies between 6 and 22 keV. For each beam energy, ε is derived from a set of measurements at different target densities, and plotted at the average velocity between entrance and exit. The gas pressure has been changed between 0.01 and 0.15 mbar to detect a possible influence from the relaxation length λ of the projectile's charge state (for 10-keV protons,[23] $\lambda = 17$ cm at 0.01 mbar and $\lambda = 1.1$ cm at 0.15 mbar, respectively): No effect is found within the accuracy of the individual measurements. Nuclear stopping has not been subtracted from the data: According to Lindhard, Scharff, and Schiøtt[24] the nuclear stopping cross section of He for protons varies from 0.10×10^{-15} eV cm^2 at 5 keV to 0.03×10^{-15} eV cm^2 at 20 keV; angle-restricted nuclear stopping, which has to be applied in our case, will be even smaller.[19] Also shown in Fig. 1 are the semiempirical curve from Ref. 2, where v_1 proportionality is assumed below 10 keV, and the only other published data[21] down to 10 keV. In Table I we compare our result to frequently used compilations of ε, Refs. 2 and 3; the table also gives the error $\Delta\varepsilon$ and the exponent β of the v_1 dependence according to $\varepsilon(v_1) \propto v_1^{\beta}$.

Figure 1 and Table I indicate that at low velocities ε is proportional to $v_1^{3.3}$; we consider this a clear disproof of the assumption that electronic stopping is generally velocity proportional. The exponent β is in fair agreement with the theoretical predictions of Ref. 11 ($\beta \approx 4$). Re-

TABLE I. Comparison of our data to the tabulations of Andersen and Ziegler (Ref. 2) and Janni (Ref. 3). For three proton energies, the stopping cross section ε, its standard deviation $\Delta\varepsilon$, and the exponent β according to $\varepsilon(v_1) \propto v_1^{\beta}$ are listed. ε and $\Delta\varepsilon$ are given in units of 10^{-15} eV cm^2.

Energy (keV)		Andersen and Ziegler	Janni	Our work
4	ε	2.46	2.34	0.72
	$\Delta\varepsilon$	0.49	0.42	0.09
	β	1.0	0.88	3.34
10	ε	3.89	3.52	2.85
	$\Delta\varepsilon$	0.58	0.35	0.27
	β	0.91	0.92	2.23
20	ε	5.18	4.86	5.04
	$\Delta\varepsilon$	0.78	0.30	0.37
	β	0.77	0.92	0.86

lying on v_1 proportionality when extrapolating ε data down to low velocities can lead to values that are wrong by as much as a factor of 3.4 at 4 keV. If this $T_{\min}$ effect in electronic stopping would apply to *solid* insulators too, it would seriously affect the description of energy transfer in collision cascades; cf. Ref. 25.

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