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Electronic stopping of slow molecular ions in solids

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Abstract. Energy loss measurements were made for 12.5–130 keV per nucleon H^+ and H_2^+ on carbon and aluminium foils. For incident H_2^+ , both H^+ and H_2^+ are transmitted; the energy per nucleon of the latter being lower than that of transmitted H^+ , at low energies. The theory shows this is due to interference effects in the binary excitation of target electrons by the spatially correlated protons and suggests that transmitted H_2^+ results from di-protons travelling inside the solid with the internuclear axis aligned close to the direction of motion.

It has been known for some time that fast H_2^+ ions moving in solids do not lose energy at the same rate as two non-interacting protons (for a review, see Brandt and Ritchie 1976). The ratios of stopping power per nucleon $R = \frac{1}{2}(dE/dx)_{H_2^+}/(dE/dx)_{H^+}$ have been found to be larger than unity for projectile velocities larger than 1.73 au. This has been attributed to be due mainly to interference effects in the correlated excitation of long-wavelength plasmons in the solid which are prominent when the internuclear distance in the molecular ion, r , is much smaller than the adiabatic distance for collective excitations, v/ω_p . Here v is the velocity of the projectiles and $\hbar\omega_p$ the plasmon energy.

Determinations of energy loss of H_2^+ ions in thin foils have so far been done through measurements of the energies of the incident projectiles and of emerging protons resulting from the dissociation of the H_2^+ molecules in the foils. H_2^+ molecules have also been observed to be transmitted through thin foils (Lambert 1976) but no measurements of stopping powers on these molecules have been reported in the literature.

This Letter reports first measurements of energy losses per nucleon for H_2^+ ions incident on thin foils at low velocities (0.76–2.3 au) and emerging as protons and H_2^+ molecules. We show that interference effects in the excitation of target electrons by the correlated protons occur even at velocities where plasmon creation does not contribute significantly to the stopping power. Furthermore, the results suggest that transmitted molecular ions have travelled inside the foil with their internuclear axes aligned close to the direction of motion.

The experimental method has been described in detail before (Valenzuela *et al* 1972, Eckardt 1978). H^+ and H_2^+ ion beams were produced in the Bariloche Kevatron accelerator and sorted in mass by a 90° magnetic analyser. The energy of the ions was measured with a precision, 90° electrostatic energy analyser with its slits set to give an energy resolution of 0.4% FWHM. The energy losses were determined by measuring the energy of the beam without and with a thin foil placed in its path. The energy analyser

accepts particles transmitted through the foils within vertical and horizontal angles of 0.043° and 0.36° respectively. Under these conditions, the influence of energy losses by elastic atom-atom collisions can be neglected.

In the experiments we used three carbon foils ($\sim 150 \text{ \AA}$ thick) and five aluminium foils ($\sim 200 \text{ \AA}$ thick). The foil thicknesses were determined using the measured energy losses for protons and dE/dx data (Andersen and Ziegler 1977). About one-third of the thickness of the Al foils is estimated to correspond to surface oxide layers formed during their exposure to atmosphere. Two liquid-nitrogen traps, coaxial with the beam, were located immediately before and after the foils to prevent significant foil thickening due to the build-up of contaminant layers.

Figure 1 shows normalised energy spectra obtained for 267 keV H_2^+ incident on an Al foil. The proton peak is seen to be broader than that for transmitted H_2^+ ; this is due to the repulsion between the dissociation fragments (Gemmell *et al* 1975). Two values of

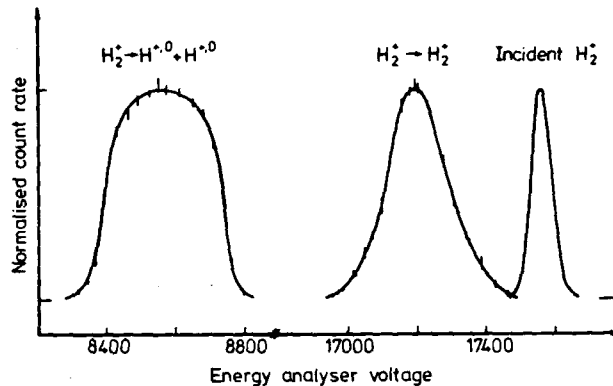


Figure 1. Normalised energy spectra for 267 keV H_2^+ incident on an Al foil.

the mean energy loss per nucleon can be derived from spectra for incident H_2^+ , such as shown in figure 1. If E_0 is the mean energy of the incident H_2^+ , and E_1 and E_2 those of transmitted protons and H_2^+ respectively, we may define $\Delta E(\text{H}_2^+)_1 = \frac{1}{2}E_0 - E_1$ and $\Delta E(\text{H}_2^+)_2 = \frac{1}{2}(E_0 - E_2)$, and the ratios $R_{1,2} = \Delta E(\text{H}_2^+)_1,2 / \Delta E(\text{H}^+)$, where $\Delta E(\text{H}^+)$ is the energy loss measured with protons incident at the same velocity as the H_2^+ ions used to derive $\Delta E(\text{H}_2^+)$. Figure 2 shows R_1 and R_2 for carbon and aluminium foils as a function of projectile velocity, together with their experimental errors. The points result from averaging measurements over foils of nearly equal thickness (within 10%). Interference effects ($R \neq 1$) can be seen to persist down to the lowest velocities. It can also be observed that R_2 is smaller than R_1 at low velocities. The transmitted H_2^+ fraction was found to be more than 100 times larger for Al than for C foils, but the electronics used did not allow us to get more quantitative measurements.

The stopping power results can be analysed in the framework of the theory of the energy loss of correlated charges in an electron gas (Arista 1978). Of the targets used in these experiments, Al is the most suited for comparison since it is free-electron like (we neglect here the influence of the surface oxide layers). The energy loss of protons in Al in our energy range will be due mainly to excitation of valence electrons. The contribution of the Al L-shell can be estimated using measured values of the cross section for L-shell excitation (Benazeth *et al* 1977) and $\langle E_L \rangle$, the average excitation energy of these

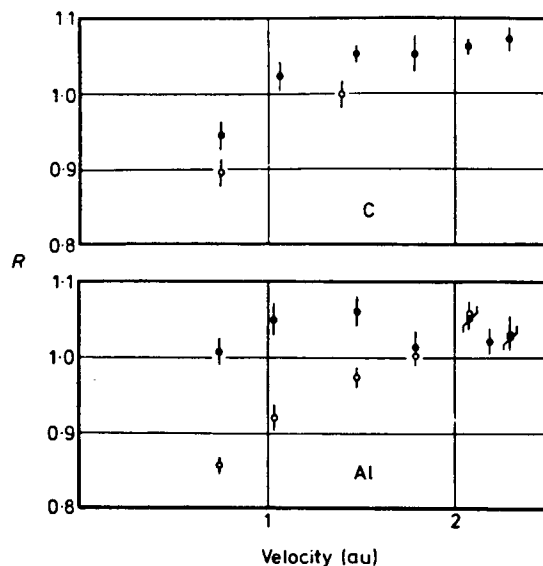


Figure 2. R_1 and R_2 as a function of projectile velocity for carbon and aluminium foils.

electrons (Haensel *et al* 1969, Powell 1974). The contribution of this shell to stopping amounts to only 1%, 3% and 10% at velocities of 0.7, 1.2 and 2 au respectively. In our energy range, the adiabatic distance for these excitations, $v\hbar/\langle E_L \rangle$ is much smaller than the interproton separation; therefore interference effects in these excitations should be negligible (Arista 1977, Lurio *et al* 1978).

Let us consider then, only the stopping due to valence electrons of the target. If we neglect the small width of the plasma resonance, there will be a threshold velocity v_{th} for plasmon excitation (Lindhard 1954) which is $v_{th} = 1.24$ au in the case of Al. At velocities smaller than this value, the electronic energy loss is due to single-particle excitations. Interference in the scattering of target electrons will occur and will be constructive or destructive (Arista 1978) depending on the ratio between the wavelength k^{-1} associated with the momentum transfer $\hbar k$ and r . The interference effects at low velocities are therefore of a different nature as those observed at high velocities which are mainly due to the coherent excitation of wakes of electron density fluctuations (plasmons) trailing the fast charged particles.

We have calculated the energy loss of a pair of protons in Al using, for the dielectric constant ϵ , the expression given by Lindhard (1954). The energy loss per nucleon,

$\frac{1}{2}(dE/dx)_{H_2^+}$ is given by

$$\frac{1}{2}(dE/dx)_{H_2^+} = (e^2/\pi^2 v)(L + I) \quad (1)$$

with

$$L = \int d^3 k [(k \cdot v)/k^2] \text{Im} | -\epsilon^{-1}(k, k \cdot v) |$$

$$I = \int d^3 k [(k \cdot v)/k^2] \cos(k \cdot r) \text{Im} | -\epsilon^{-1}(k, k \cdot v) |$$

where v is the velocity of the protons, k the momentum transfer and r the vector joining the two protons. In equation (1), L is proportional to the stopping number for a proton and I represents the interference in the stopping of the two protons. The ratio $R(\theta, r) = 1 + I(\theta, r)/L$ has been calculated for $\theta = 0^\circ, 45^\circ$ and 90° , where θ is the angle between the internuclear axis and the direction of motion, i.e. between r and v . We have also calculated the angular averaged value $\langle R(r) \rangle$. The results are shown in figure 3, for several projectile velocities.

The distance r will be a function of particle velocity and depth of penetration in the foil. An H_2^+ ion with average internuclear separation r_0 impinging on the solid will lose its valence electron within a few atomic layers. At low particle velocities, the screening by target electrons will be strong and so the two protons will experience little repulsion. The mean value of r will be determined by the combined effect of the forces between particles modified by the polarisation of the medium, the different stopping of each particle, multiple scattering and energy straggling.

In order to compare theory and experiment, the results shown in figure 3 should be integrated over r which in turn is a function of position in the foil. The correct inclusion of all the effects enunciated above is very complicated and falls outside the scope of this work. As a first approximation we will estimate r as given only by the screened Coulomb repulsion between the protons, using the simple potential $V(r) = e^2/r(1 + r^2/a^2)$ which neglects distortion of the screening cloud, and where a , the screening length, is taken as v/ω_p for $v > v_F$ and v_F/ω_p for $v < v_F$ (Brandt 1975). We thus obtain for the separation at exit, in our case, the values 9.5, 7.6, 6.2, 4.9 and 4.4 au at $v = 0.71, 1, 1.41, 2$ and 2.28 au respectively; the corresponding values for the average internuclear separation being 5.4, 4.5, 4, 3.3 and 3.2 au. This indicates the values of r over which the results of figure 3 should be integrated. For our estimates, we used the value (Brandt and Ritchie 1976) $r_0 = 2.44$ au and assumed that H_2^+ ions are ionised on entrance to the foil.

The anisotropy of the polarisation cloud around each moving ion, neglected above, is expected to modify the initial orientation of the molecular axis. The net effect will

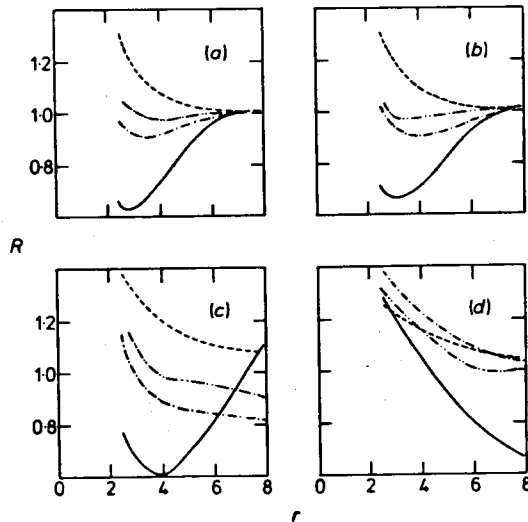


Figure 3. Plot of R versus r for various projectile velocities: (a), $v = 0.63$; (b), $v = 0.90$; (c), $v = 1.26$; (d), $v = 2.25$.

be an alignment of this axis with the direction of propagation, which will be larger the longer the transit time in the foil and the larger the alignment forces. Alignment effects have been beautifully demonstrated by Gemmel *et al* (1975) at high energies (0.3–2 MeV per nucleon). At the lower energies used in this work, multiple scattering and energy straggling will smear out this effect for long transit times.

In spite of the fact that the correct dependence of r with depth in the foil is unknown and that the theory was made for an unbounded free-electron gas whereas our thin Al target inevitably had surface oxide layers, some qualitative conclusions can still be derived from figures 2 and 3. First we notice that interference effects subsist at low velocities where plasmon excitations are unlikely and that the low values of R_2 for transmitted H_2^+ at these velocities can be explained if these molecules result from proton pairs which have travelled inside the solid with their internuclear axis aligned close to the beam direction. On the other hand, transmitted protons will result from di-protons for which multiple scattering and energy straggling have overruled the alignment forces. These di-protons will travel on the average with larger separations than in the other case and the stopping ratio should be given approximately by $\langle R \rangle$.

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