

Effect of coherent multiple scattering on the transmission of H_2^+ through thin foils

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Results are presented of a Monte Carlo simulation of the passage of fast H_2^+ ions through thin carbon foils, with emphasis on the transmitted fraction. An essential feature of the calculation is that the Coulomb interaction between the constituent protons and the multiple scattering of each in the foil is treated simultaneously. The agreement between previous experimental data and the simulation is excellent.

I. INTRODUCTION

Investigations of the interactions of fast molecular-ion beams with solids offer many attractive possibilities for studying atomic collision phenomena. Central to such investigations has been the Coulomb explosion.¹ For swift (\sim MeV) light projectiles (e.g., H_2^+ , HeH^+ , He_2^+) incident upon a foil, the binding electrons are stripped off within the first few angstroms of penetration into the target. This is a consequence of the characteristically large electron-loss cross sections ($\sim 10^{-16}$ cm²). There then follows a Coulomb explosion, in which the bare constituent nuclei of the projectile fly apart by virtue of their mutual Coulomb repulsion. The characteristic time for the Coulomb explosion (femtoseconds) is also generally comparable to the dwell time in the target. Inside the target the individual fragment trajectories are also influenced by multiple scattering as well as by the effects of the electron polarization "wakes" induced behind each of the fragments.²

Although most of the fragments exit the foil as individual ions or atoms, a very small fraction ($\ll 10^{-2}$) of the incoming molecules can be transmitted intact through the foil.³ It has been shown that this transmitted fraction is composed of two components: For very short dwell times, there is a finite probability for some molecules to exit the foil with their original electron(s) (the \mathcal{O} regime), while for longer dwell times, transmission is due to the reconstitution at the exit by electron capture (the \mathcal{R} regime). Capture of one or more electrons can occur at this instant into molecular orbits which can be either bonding or dissociative for different asymptotic final states. Since the cluster presents an average effective charge in some molecular orbitals (MO) that is larger than those of the isolated fragments, the probability of electron capture can be enhanced. Cue *et al.*⁴ have shown that the multiple scattering of the nuclei in the foils is a very important process that also enhances the probability for the reconstitution process by providing a final internuclear distribution which is wider than the one expected from a simple Coulomb repulsion. Thus some fragments can be sufficiently close to each other, even after long dwell times in the foil, to be trapped in the molecular potential well. A simple model using the above assumptions, in which the multiple scattering and Coulomb interaction

were treated as additive processes, reproduced (in the \mathcal{R} regime) the general trend of the observed transmitted yield of H_2^+ relative to the yield of H^0 from isotachic protons.⁴ Another approach for calculating the transmitted fraction has been described by Kononetz and Dzhamankyzov⁵ who used the Fokker-Planck equation and found good agreement with the thickness dependence of the experimental data.

The purpose of this work is to introduce a calculation of the transmitted yield of H_2^+ using a more complete model based on a simulation which treats the multiple scattering of the two protons in the cluster simultaneously with the Coulomb explosion process.

II. MULTIPLE-SCATTERING SIMULATION

The simulation is divided in two principal parts. First, the multiple scattering of each proton is generated individually, for a given number of trajectories, using a procedure developed by Moller, Pospiech, and Schrieder,⁶ which has been described elsewhere.⁷ The result is, for each trajectory, a list of scattering angles (polar and azimuthal) and the time of each collision inside the target.

Then, for the case of molecular projectiles, in addition to multiple scattering, the trajectories are modified by the mutual Coulomb repulsion of the ionic fragments. A simplified treatment of this effect is to assume that these are independent interactions producing additive small-angle deflections.² A statistical treatment using the Fokker-Planck equation to simultaneously incorporate these two effects previously demonstrated⁸ that important focusing and defocusing effects are not accounted for in the simpler model. The Gaussian approximation of the Fokker-Planck approach neglects the tails of the distribution,⁹ which are important in our case, since molecular transmission is a small effect and is strongly dependent upon these tails. An earlier simulation¹⁰ showed that incoherent treatment gives exit separation distributions shifted to much smaller values and much broader. Moreover, it is clear that the interplay between multiple scattering and the Coulomb repulsion is orientation dependent. For example, for a molecule with its internuclear axis aligned to the beam direction, the multiple-scattering deflections are perpendicular to the Coulomb explosion forces, and we expect the multiple scattering to

be independent of the dissociation process. However, for molecules aligned transverse to the beam direction, the Coulomb forces and the multiple scattering affect the same velocity components and are thus "correlated."

For the simulation, the initial distribution of the internuclear distance of the molecular ions was taken to be that measured by means of Coulomb explosion experiments.¹¹ In such experiments the initial beams come from a rf ion source, and it has been demonstrated that many vibrational states are populated by the ionization process. We used a Gaussian distribution with a mean value of $r_0 = 1.17 \text{ \AA}$ and a width of $\sigma = 0.3 \text{ \AA}$ (compared to $r_0 = 1.08 \text{ \AA}$ and $\sigma = 0.13 \text{ \AA}$ obtained for the ground state alone). For each trajectory, the initial distance was taken from this distribution, and the internuclear axis rotated randomly in space, to represent the different orientations of the molecules present in the beam.

The method of computation consisted of numerically integrating the equations of motion for the different "free paths" between each collision of either proton. At each collision the velocity vector of each proton was rotated, as given by the polar and azimuthal angles computed in the first part of the simulation, to account for multiple scattering. The screened potential between the two ions inside the solid was given by

$$V(r) = \frac{Z_1 Z_2 e^2}{r} e^{-r/\alpha}, \quad (1)$$

where r is the distance between the two ions, Z_1 and Z_2 are the nuclear charge, and the exponential term reflects the screening due to target electrons. For slow projectiles, i.e., $v < v_0$ (v_0 is the Fermi velocity), the screening distance α does not depend on the projectile velocity v and is always smaller than internuclear distances for the H_2^+ molecule (0.4 \AA in carbon). As a consequence, the Coulomb forces between fragments of a slow incident molecular ion are negligibly weak in a solid medium. But for projectiles with velocities $v > v_0$, one has to consider dynamic screening in which α is proportional to the cluster velocity and given by $\alpha = v/\omega_p$, where ω_p is the plasma frequency of the solid ($\hbar\omega_p = 25 \text{ eV}$ for carbon). The result of this procedure produces the final internuclear distance \mathbf{R} and relative velocities \mathbf{v}_r of the protons at the exit of the target foil.

III. CALCULATION OF TRANSMITTED YIELDS

For H_2^+ the probability for transmission is related to electron loss (in the \mathcal{O} regime) and electron capture (in the \mathcal{R} regime). Let us first consider a capture event.

Let E_k be the internal kinetic energy for the diproton with an internuclear distance R , and let $U(R)$ be the electronic energy of a given MO relative to the energy of the $\text{H}^+ + \text{H}$ at $R = \infty$, which is taken to be zero. Capture of an electron into an antibonding state will eventually lead to a proton and a hydrogen atom (in its ground or excited state). Capture into a bonding state can also lead to atomic hydrogen, unless $E_k + U(R) \leq 0$, in which case a bound H_2^+ is formed. This occurs mainly for capture into the $1s\sigma_g$ state,¹² and thus we consider only that MO.

The final ingredient is to calculate the electron-capture cross section into the $1s\sigma_g$ state. Cue *et al.*⁴ suggested the following scaling rule:

$$\sigma_c^g(R, v) = Z_g(R)^5 \sigma_c(v), \quad (2)$$

where $\sigma_c(v)$ is the electron-capture cross section by a proton of the same velocity v leading to capture into the $1s$ state. The $Z_g(R)^5$ factor is a natural extension of the hydrogenic scaling of σ_c in terms of an effective charge in the case of atomic projectiles.¹³ For the molecular case, Z_g is the R -dependent effective charge appropriate for the $1s\sigma_g$ MO as given by McCarroll, Piacentini, and Salin.¹⁴

If the original electron of the molecule is not lost, then we can consider the molecule to be still bound [providing that $E_k + U(R) \leq 0$]. The transmission in the \mathcal{O} regime is then directly related to the process of electron loss. The molecular cross section for this process has been found to be, to a good approximation, equal to the electron-loss cross section σ_l for atomic hydrogen.^{15,16}

The probability for transmission of a molecule after traveling a distance z in the target is then given by

$$F(z) = \begin{cases} \frac{\sigma_c^g}{\sigma} + \left[1 - \frac{\sigma_c^g}{\sigma}\right] \exp(-n\sigma z) & \text{if } E_k + U(R) \leq 0 \\ 0 & \text{otherwise,} \end{cases} \quad (3)$$

where $\sigma = \sigma_c^g + \sigma_l$ and n is the target density.

The atomic cross sections for electron capture and loss employed were those of Bohr:¹⁷

$$\sigma_l = \pi a_0^2 Z_t^{2/3} Z_p^{-1} \frac{v_0}{v}, \quad (4)$$

$$\sigma_c = 4\pi a_0^2 Z_t^{1/3} Z_p^5 \left[\frac{v_0}{v}\right]^6, \quad (5)$$

where Z_t and Z_p are the target and projectile atomic number, respectively, and a_0 is the Bohr radius.

For each trajectory, the probability of transmission $F(z)$ is calculated and compared to a random number ξ , $0 \leq \xi \leq 1$. If $E_k + U(R) \leq 0$ and $\xi \leq F$, the two protons are considered bound and that H_2^+ is considered to be transmitted.

IV. RESULTS

The experimental transmission yield¹⁶ $Y(\text{H}_2^+)$ at 0.4, 0.8, and 1.2 MeV/amu through carbon foils $1-8 \mu\text{g}/\text{cm}^2$ thick are shown in Fig. 1 as a function of dwell time t_d in the target. The \mathcal{O} and \mathcal{R} regime can be clearly seen. The \mathcal{O} ($t_d \leq 1 \text{ fs}$) regime follows a simple exponential law, independently of the projectile velocity. For longer dwell times, the transmission yields exhibit a strong dependence on v . The simulation results are shown in the same figure as solid symbols. The agreement with the experimental data is excellent in the two different regimes, especially considering the fact that the simulation has no adjustable parameters.

Figure 2 shows the transmission yield^{16,18} $Y(\text{H}_2^+)$ in the \mathcal{R} regime relative to twice the equilibrium neutral

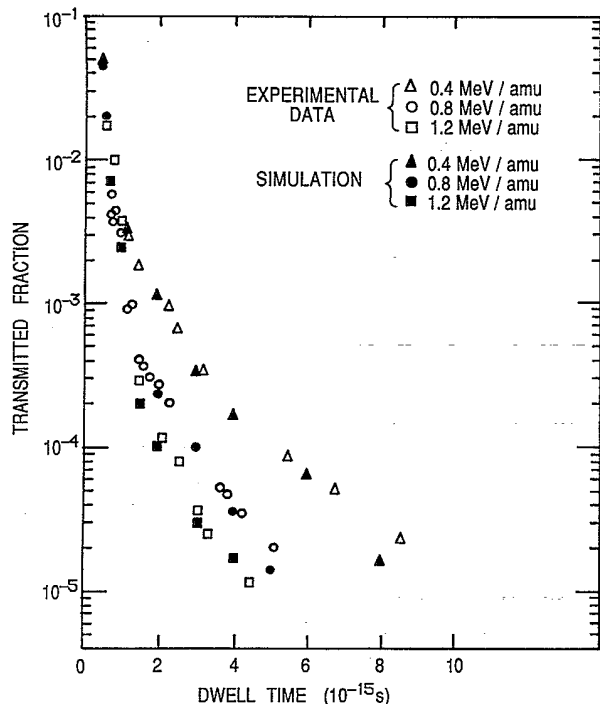


FIG. 1. Transmitted fraction of H_2^+ as a function of dwell time t_d in carbon foils for different incident energies. Open and solid symbols represent the experimental (Ref. 16) and simulated results, respectively. The statistical uncertainty is typically of the size of the symbol.

fraction Φ_0 for incident H^+ at the corresponding velocity. The points on this curve fall in a near-universal curve which is a function of t_d only. This is due to the similarity of the velocity dependence of the molecular and atomic cross sections for electron loss and capture. The simulation points on this graph have been calculated by the relation

$$Y(H_2^+)/2\Phi_0 = Y(H_2^+)/[2\sigma_c/(\sigma_c + \sigma_l)]. \quad (6)$$

Since most of the data is in the \mathcal{R} regime ($t_d > 1 \mu s$), we did not subtract the small contribution from the \mathcal{O} regime. The simulation results are in better agreement with the data than are the previous calculations⁴ (which are represented by the solid line). The main difference between the two calculations is the simultaneous treatment of Coulomb force and multiple scattering. To investigate this difference we have calculated the relative yield incoherently, using the same Monte Carlo routine to generate the multiple-scattering distribution of each proton, and then adding the displacements and velocity shifts due to the Coulomb repulsion only at the exit of the foil. The results of this procedure are shown in Fig. 2 by the dashed line, which is in good agreement with the previous calculation, but lower by a factor of ~ 2 relative to the experimental results. This demonstrates the importance of the interplay between the Coulomb interaction and multiple-scattering process. The fact that agreement obtained in the relative yield (Fig. 2) between the simulation and the experimental results is slightly worse than the agreement obtained in the absolute yield (Fig. 1) is

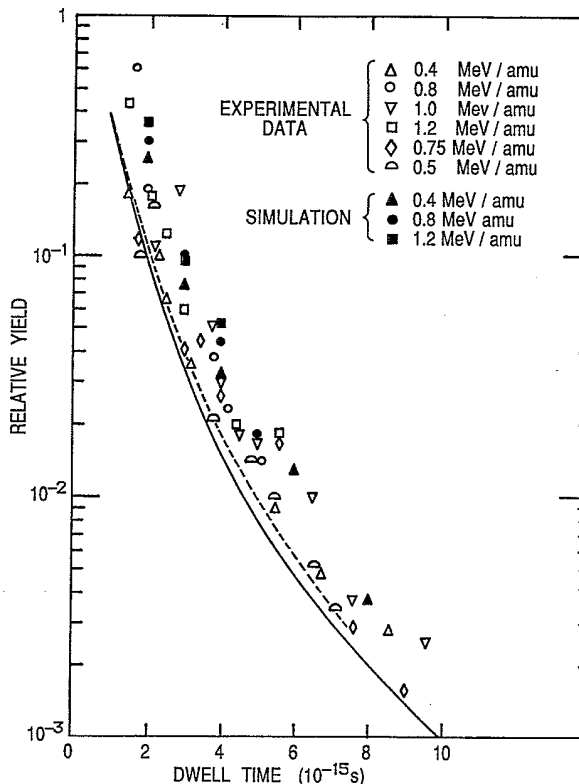


FIG. 2. Yield of transmitted H_2^+ fraction through carbon foils in the \mathcal{R} regime normalized to twice the equilibrium neutral fraction of protons of corresponding velocities. Open and solid symbols represent the experimental (Refs. 16 and 18) and simulated results, respectively. The solid curve is the prediction of the model by Cue *et al.* (Ref. 4); the dashed curve is the result of the simulation for the multiple scattering and the Coulomb interaction treated incoherently.

essentially because the neutral fraction Φ_0 from incident proton is not exactly described by the Bohr cross section in this velocity range.

It should be pointed out that data exist also for longer dwell times, but have not been included in this work due to limitation on computer time (a 3% statistical uncertainty for a transmission of 10^{-5} requires about 10^8 trajectories).

V. CONCLUSIONS

The yields of transmitted H_2^+ through thin carbon foils were calculated using a Monte Carlo simulation taking account of the correlation of the Coulomb interaction between the ions and multiple scattering. The agreement between the simulation and experimental results is very good, and demonstrates that the shape of the final internuclear and velocity distribution are very much affected by this correlation.

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