

Simulation of the molecular recombination yield for swift H_2^+ ions through thin carbon foils

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We have calculated the recombination yield for swift H_2^+ molecular ions at the exit of thin amorphous carbon foils, as a function of the dwell time and incident energy. Our results are based on a detailed simulation of the motion through the target of the H_2^+ molecular ion (before dissociation takes place) and its constituent fragments (after dissociation), including the following effects: Coulomb repulsion, nuclear scattering, electron capture and loss, as well as self-retarding and wake forces, which provide the relative distance and velocity of the dissociated fragments at the foil exit. The recombination of an H_2^+ ion at the exit of the foil depends on the interproton separation and internal energy of the dissociated fragments, and on their probability to capture an electron. Comparison of our results with the available experimental data shows a good agreement.

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I. INTRODUCTION

The interest in the study of energetic H_2^+ beams interacting with solids lies in that H_2^+ is the simplest molecular projectile, therefore it can be used to test effects due to the correlated motion in close proximity of two charged particles through a thin foil. These effects became apparent in the transmission yield of H_2^+ molecular ions [1], as well as the charge states [2] and the energy loss [3] of the dissociated protons.

The H_2^+ molecular ion is constituted by two protons and a bound electron; after entering the foil it travels a certain time without being dissociated, until it loses its bound electron [4]; this gives two protons that move in close proximity, interacting between them and with the target electrons and nuclei. Although most of the detected projectiles at the exit of the foil are protons, there are also hydrogen atoms as well as a small fraction of hydrogen molecular ions. The transmitted molecules can be either original ones or recombined after their dissociation through the foil [1,5], depending on the dwell time.

The aim of this work is to calculate the transmission yield of recombined H_2^+ molecular ions at the exit of thin carbon foils. For this purpose we simulate the motion through the solid of the initially nondissociated H_2^+ during a short time and, after the dissociation takes place, the correlated motion of each pair of fragments. We have considered that the projectiles feel the following interactions: nuclear scattering, stopping force, wake force, and Coulomb explosion, the latter two appearing only for the dissociated fragments; the possibility of electronic capture or loss by these fragments is also included in our simulation. Therefore, when an electron is captured by a proton at the exit of the foil and if the interproton velocity and distance are favorable, a recombined H_2^+ molecular ion will result.

In what follows we present the basic ingredients of our calculations (Sec. II), whose results are compared with available experimental data in Sec. III. The conclusions of this work appear in Sec. IV. In what follows we will use atomic units, except where otherwise stated.

II. THEORETICAL AND SIMULATION BACKGROUND

The H_2^+ molecular ions incident on the foil are characterized by their interproton distance R and orientation with respect to the beam velocity v .

When a molecular ion enters the amorphous carbon foil it moves during a short time (~ 0.23 fs [6]) without dissociating, suffering an electronic stopping force and collisions with the target nuclei. Once H_2^+ dissociates into its constituents, each fragment also experiences both an electronic stopping force and nuclear scattering, plus the interactions due to the presence of its partner, which are the Coulomb repulsion (when both fragments are charged) and the electronic wake force [7].

Given a charge density, with Fourier transform $\rho(k)$, moving with velocity v through a target, the stopping force acting on it is statistical in nature and we assume that it has a Gaussian distribution whose mean value is the stopping power S_p , and its variance is related to the energy-loss straggling per unit path length, Ω^2 . In the dielectric formalism [8] these magnitudes are given by the following expressions:

$$S_p = \frac{2}{\pi v^2} \int_0^\infty dk \frac{\rho^2(k)}{k} \int_0^{kv} d\omega \omega \operatorname{Im} \left[\frac{-1}{\epsilon(k, \omega)} \right] \quad (1)$$

and

$$\Omega^2 = \frac{2}{\pi v^2} \int_0^\infty dk \frac{\rho^2(k)}{k} \int_0^{kv} d\omega \omega^2 \operatorname{Im} \left[\frac{-1}{\epsilon(k, \omega)} \right]. \quad (2)$$

The target is characterized by its energy-loss function $\operatorname{Im}[-1/\epsilon(k, \omega)]$, which contains relevant information about its response to electronic excitations with momentum k and energy ω induced by the passage of the swift charge. A procedure to describe reasonably well the energy-loss function of a realistic target has been published elsewhere [9–11] through a fitting to its optical properties at $k=0$, with a proper extension to $k \neq 0$ values.

To obtain the values of S_p and Ω^2 for the nondissociated H_2^+ molecular ion we consider that it consists of two protons and a bound electron described by a linear combination

of Gaussian orbitals [12]; assuming that the H_2^+ ions enter the target with the interproton distance R randomly aligned with respect to incident velocity v , the following angular average of the squared charge density [12] must be replaced into Eqs. (1) and (2):

$$\rho^2(k) = \frac{A^2(k)}{2} \left[1 + \frac{\sin(kR)}{kR} \right] + B^2(k) - 4A(k)B(k) \frac{\sin(kR/2)}{kR}, \quad (3)$$

where

$$A(k) = 2 - C(k), \quad (4)$$

$$B(k) = e^{-\alpha R^2/2} C(k), \quad (5)$$

$$C(k) = \frac{e^{-k^2/8\alpha}}{1 + e^{-\alpha R^2/2}}, \quad (6)$$

with $\alpha = 0.43$ a.u. and $R = 2.05$ a.u. for the hydrogen molecular ion [12].

The possibility that the dissociated fragments can capture or lose an electron inside the foil implies that the charge fractions Φ_0 and Φ_+ for each charge state H^0 and H^+ , respectively, have non-null values; the H^- fraction is negligible in the velocity range studied here [13]. The value of Φ_0 is available from experimental measurements [13,14].

In order to compute the forces due to the electronic excitations induced in the target by each projectile it is convenient to work with the cylindrical coordinates z and r , parallel and perpendicular, respectively, to the projectile velocity v . Using the dielectric formalism [8,9] to calculate the force produced by a dissociated fragment, with charge density ρ_1 on the other dissociated fragment charge density ρ_2 at the relative position (z, r) , the cylindrical components of this force are given by the following expressions:

$$F_z(z, r) = \frac{2}{\pi v^2} \int_0^\infty dk \frac{\rho_1(k)\rho_2(k)}{k} \int_0^{kv} d\omega \omega J_0(r\sqrt{k^2 - \omega^2/v^2}) \times \left\{ \sin\left(\frac{\omega z}{v}\right) \text{Re}\left[\frac{1}{\epsilon(k, \omega)} - 1\right] + \cos\left(\frac{\omega z}{v}\right) \text{Im}\left[\frac{1}{\epsilon(k, \omega)} - 1\right] \right\} \quad (7)$$

and

$$F_r(z, r) = \frac{2}{\pi v} \int_0^\infty dk \frac{\rho_1(k)\rho_2(k)}{k} \times \int_0^{kv} d\omega \sqrt{k^2 - \omega^2/v^2} J_1(r\sqrt{k^2 - \omega^2/v^2}) \times \left\{ \cos\left(\frac{\omega z}{v}\right) \text{Re}\left[\frac{1}{\epsilon(k, \omega)} - 1\right] - \sin\left(\frac{\omega z}{v}\right) \text{Im}\left[\frac{1}{\epsilon(k, \omega)} - 1\right] \right\}, \quad (8)$$

where $J_{0,1}(\dots)$ are Bessel functions of the first kind.

The Fourier transform of the charge density $\rho(k)$ is chosen according to the type of fragment:

$$\rho(k) = \begin{cases} 1 & \text{for } H^+, \\ 1 - \frac{1}{[1 + (k/2)^2]^2} & \text{for } H^0. \end{cases} \quad (9)$$

Each possible charge state is taken according to its corresponding probability, which is equivalent to its charged fraction (because $\Phi_0 + \Phi_+ = 1$).

It is worth noting that Eq. (7) evaluated at the coordinates $(z=0, r=0)$ simplifies considerably to give the stopping power S_p for a given projectile when replacing $\rho_1(k)\rho_2(k)$ by the square of the corresponding charge density $\rho^2(k)$ [cf. Eq. (1)].

Our computer code, whose details appear elsewhere [7], numerically solves Newton's equations of motion for each projectile: the molecular ion or its dissociated fragments (either a proton or a hydrogen atom), taking into account all the forces that act on each particle. Only when the charge state of both fragments is nonzero will they feel a Coulomb repulsion along the interproton direction. We have used a Monte Carlo algorithm [15] to account for binary collisions with the target nuclei, which contribute mainly to deviations in the trajectory direction and in a smaller amount to energy losses; the universal interatomic potential [16] was used for this purpose. For the case of the dissociated fragments we neglect coherent scattering because it is a small effect in the velocity range discussed in this work [17].

The distribution of R values for the incident molecular ions is chosen according to the population of vibrational levels of H_2^+ (which was calculated using the Frank-Condon principle); the orientation of the interproton axis was taken randomly from a uniform distribution. Given the positions and velocities of the particles as well as the forces acting on them at each time t , the simulation provides the corresponding new positions and velocities after an elapsed time Δt . In this manner, the relative positions and velocities when the fragments leave the target can be known.

Once the fragments reach the rear surface of the foil, H_2^+ molecular recombination can only take place when the three following conditions are fulfilled: (i) One of the fragments has captured an electron; (ii) the internal energy E_{int} of the system is smaller than the maximum of the effective potential energy for dissociation:

$$E_{\text{int}} < U_{\text{eff max}}(R, V), \quad (10)$$

where

$$E_{\text{int}} = \frac{1}{2} \mu V^2 + U_{\text{eff}}(R, V) = \frac{1}{2} \mu V^2 + U(R) + \frac{L^2}{2\mu R^2}; \quad (11)$$

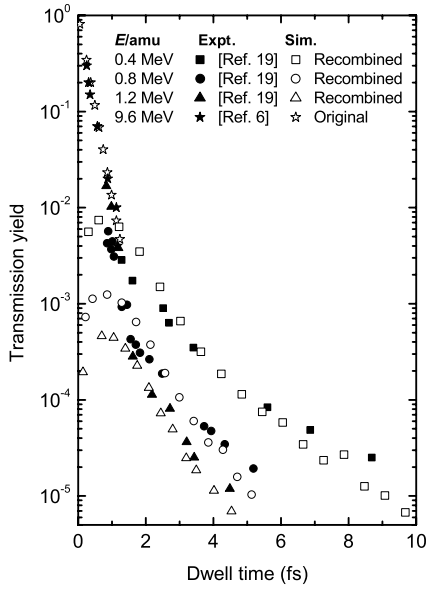


FIG. 1. Yield of transmitted H_2^+ molecular ions through amorphous carbon foils, as a function of the dwell time. Solid symbols represent experimental data for several incident energies ($t < 1$ fs, originally transmitted [6]; $t > 1$ fs, recombined [19]) and empty symbols are the results of our simulations for recombined molecular ions.

(iii) the value of the interproton distance R is comprised between the classic turning points of the effective potential well U_{eff} . In the above expressions μ , V , and L are, respectively, the reduced mass, the module of the relative velocity, and the module of the relative angular momentum ($\vec{L} = \mu \vec{R} \times \vec{V}$), corresponding to the two fragments of the H_2^+ molecular ion; $U(R)$ is the $1s\sigma_g$ ground-state potential energy [18].

III. RESULTS

Figure 1 shows the fraction of transmitted H_2^+ ions through thin carbon foils, as a function of the dwell time. The solid symbols correspond to experimental data [19] for recombined molecular ions with incident energy in the range 0.4–1.2 MeV/amu; for comparison purposes, also the fraction of transmitted original molecular ions is depicted for the case of higher incident energy [6], 9.6 MeV/amu, corresponding to very small dwell times. The two regimes (recombined and original transmission) can be clearly appreciated through the different slopes and the ordinate intercept at $t \rightarrow 0$. The results of our simulation for the recombined molecules (0.4–1.2 MeV/amu incident energy) are depicted by open symbols, showing a rather good agreement with the experimental data; the symbols appearing at small dwell times show how the small contribution of recombined molecular ions is masked by the rather larger number of molecular ions that were transmitted without dissociation.

According to Cue *et al.* [19,20] the fraction Y of detected H_2^+ molecular ions depends on the dwell time t through

$$Y(t) = (1 - B)\exp(-t/\tau) + B \exp(-bt), \quad (12)$$

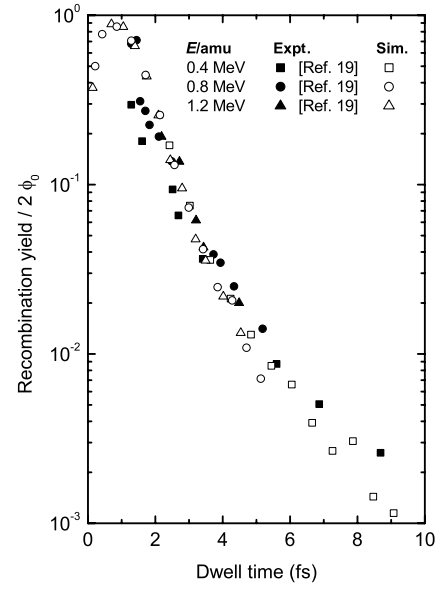


FIG. 2. Recombination yields shown in Fig. 1, but normalized to twice the proton neutral fraction for each incident energy.

where B , b , and τ are constants for a certain velocity and for a given material. The first summand of the previous equation accounts for the fraction of originally transmitted H_2^+ ions and the second summand represents the fraction of recombined H_2^+ ions. As $B \ll 1$ for high energies, τ is interpreted as the lifetime of the molecular ion inside the solid [20], and can be considered velocity independent [21]. The last term in Eq. (12) does not tend to 1 for $t \rightarrow 0$, it decays more smoothly than the first term and depends strongly on the velocity of the molecular ion; this behavior is in good qualitative agreement with the (experimental and calculated) results shown in Fig. 1. For $t \sim 2$ fs the calculated values of Y are slightly larger than the experimental data; however states having E_{int} very close to $U_{\text{eff max}}$ could dissociate, resulting in a reduction of the calculated recombination yield.

The probability that two protons exit the foil as $\text{H}^+ \text{-H}$ can be written as $P_{\text{H}^+ \text{-H}} = 2\Phi_+ \Phi_0$. For molecules having $\sim \text{MeV/amu}$ incident energies, Φ_+ is close to unity so $P_{\text{H}^+ \text{-H}} \approx 2\Phi_0$ and the molecular recombination yields can be divided by twice the neutral fraction at that velocity, $Y/2\Phi_0$. These normalized recombination yields are shown in Fig. 2, with data for Φ_0 taken from Ref. [13]; it can be seen that all the normalized recombination yields practically merge into a single curve that is a function of only the dwell time.

As it can be seen, our calculations reproduce satisfactorily well the recombination yield data for a wide range of dwell times and incident energies. From the detailed simulation we have performed, we conclude that although the recombination yield is mostly affected by the Coulomb repulsion, the proper inclusion of the other interactions felt by the dissociated fragments (nuclear scattering, wake forces, ...) is necessary to have a good agreement with experimental data.

IV. CONCLUSIONS

The recombination yield of H_2^+ molecular ions through thin carbon foils has been simulated and the results compare

fairly well with available experimental data [19].

The main ingredients in the simulation are the proper description of target energy-loss function, and the interactions felt inside the solid by the molecular ion (before dissociation) and by the molecular fragments (once they become dissociated), as well as the inclusion of the electronic-capture and -loss processes by each one of these fragments. Taking into account all these considerations, the dynamical evolution of the fragments provides their coordinates and velocities at any time in the foil; these magnitudes are used to calculate the relative position and velocity of these fragments when they leave the foil.

When two dissociated protons exit the rear surface of the foil we assume that an H_2^+ molecular ion is reconstructed if (i) one of the protons captures an electron, (ii) the internal energy of the system formed by the two protons and the

captured electron is less than the relative maximum of the effective energy corresponding to the H_2^+ ground electronic level, and (iii) the interproton separation falls between the classical turning points of the effective potential energy.

Our detailed program can be used to check how significantly the different interactions or the proposed recombination mechanism affect the final result.

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