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Transmission of Fast H_2^+ through Thin Foils

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Transmission yield of fast H_2^+ through carbon foils has been measured over a wide range of foil thicknesses for 0.4–1.2-MeV/amu H_2^+ projectiles. A model is described which gives an excellent quantitative account of this yield as well as that of the associated H^0 production.

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Evidence recently accumulated[1-3] has demonstrated that simple atoms and molecules transmitted through thin solids at velocities $V > v_0$, where v_0 is the Bohr velocity, can be the incident species with their original electron(s) as well as those reconstituted by the process of target electron capture after the loss of the incident electron(s). For convenience, we distinguish the two as the $\mathcal{O}$ (original) and $\mathcal{R}$ (reconstituted) transmission regimes, respectively. The $\mathcal{O}$ regime may be simply characterized by the survival of the incident projectile and this has been described in detail elsewhere.[1-3] The $\mathcal{R}$ regime may not be so simply understood except perhaps for the case of atomic projectiles. Indeed, data on molecular transmission in the $\mathcal{R}$ regime have been available[4-8] for a number of years and, to our knowledge, no quantitative account has previously been given for the yield of even the simplest case of H_2^+. We describe here a quantitative model for fast H_2^+ which reproduces remarkably well the observed transmitted yield of H_2^+ as well as the associated yield of H^0 breakup fragments.

The experimental features of the H_2^+ transmitted fraction at 0.4, 0.8, and 1.2 MeV/amu through carbon foils 1–8 $\mu g/cm^2$ thick are shown in Fig. 1 as a function of dwell time, t_D, in the target. These new data were obtained at Université Lyon-I using a procedure described elsewhere.[3] The $\mathcal{O}$ and $\mathcal{R}$ features can clearly be seen. There is the $\mathcal{O}$ region of $t_D \lesssim 1$ fs which is observed to follow $\exp(-t_D/\tau)$, with $\tau = 0.17$ fs, independently of the projectile velocity V. This translates into an "electron-loss" cross section $\sigma_l \propto 1/V$ which is a feature expected from binary collisions.[1] On the other hand, there is the other region of $t_D > 1$ fs, which exhibits a strong dependence on V. Such a feature can only be reasonably understood in terms of a reconstitution process after the loss of the incident electron, and it is this process that is of principal concern here.

To properly focus on the $\mathcal{R}$ regime, we subtract out the exponential $\mathcal{O}$ transmission yield from the data and display in Fig. 2(b) the $\mathcal{R}$ transmission yield $Y(H_2^+)$ relative to twice the equilibrium neutral fraction Φ_0 for incident H^+ of the corresponding velocity. In effect then we have made use of a previous observation[6] that the

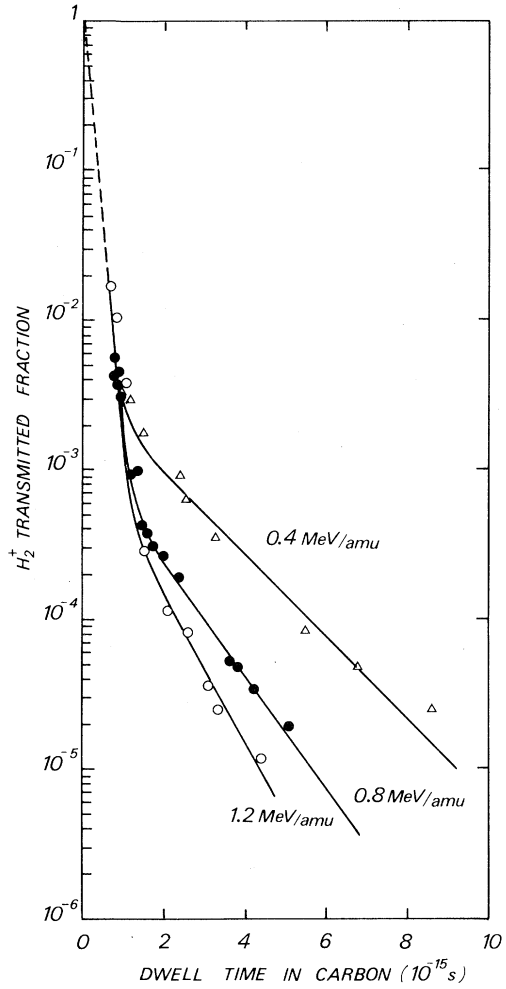

FIG. 1. Measured transmitted fraction of H_2^+ as a function of dwell time, t_D, in carbon foils for incident H_2^+. The statistical uncertainty is typically of the size of the symbol. The relative and absolute uncertainties of t_D are estimated to be ± 5% and ± 10%, respectively. The curves are the results of a least-squares fitting procedure with use of a linear combination of two exponentials and merely serve to emphasize the $\mathcal{O}$ and $\mathcal{R}$ regimes of transmission.

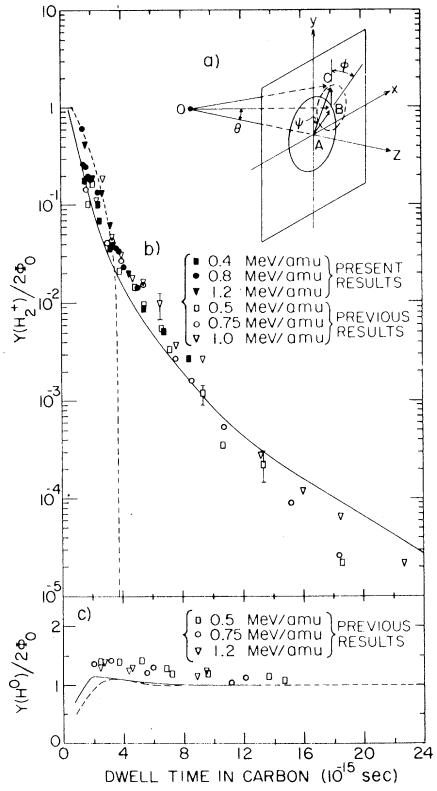

FIG. 2. (a) The effects of multiple scattering characterized by $\vec{BC}$, on the interproton separation $\vec{OB}$ oriented at an angle θ relative to the beam direction $\vec{OA}$ give a distribution to the values of the final interproton separation $|\vec{OC}|$. (b) Measured yields of transmitted H_2^+ fraction $Y(H_2^+)$ through carbon foils in the $\mathcal{R}$ regime normalized to twice the equilibrium neutral fraction Φ_0 of protons of the corresponding velocities. The previous set of transmission data (from Ref. 6) has been scaled up by a factor of 2 and the curves are the predictions of the model described in the text. (c) Yields of H^0 from H_2^+ breakup in carbon foils in the $\mathcal{R}$ regime transcribed from Ref. 1 with the curves showing the predictions of the model.

$Y(H_2^+)/2\Phi_0$ ratios cluster into a near-universal curve which is a function only of t_D. Some earlier published data[6] have also been included in Fig. 2 in order to give a wider t_D range for comparison with the predictions of the model to be described shortly. These older yield ratios were scaled upward by a factor of 2 in order to match the present set which we believe is more accurate. This is of minor consequence since the yield which we are attempting to reproduce varies by four orders of magnitude.

A previous measurement[1] of H^0 yield with incident H_2^+ has shown that the yield, per incident proton, is enhanced over that for incident H^+ of the same velocity throughout the range of t_D examined. The $\mathcal{O}$-$\mathcal{R}$ features are also apparent in that case and, we believe, correlated with the H_2^+ transmission yield. In the $\mathcal{O}$ regime, the observed exponential decay with increasing t_D of the H^0 yield can be viewed to arise from the dissociation of the incident H_2^+ (in the $1s\sigma_g$ ground state). We may thus similarly subtract out the $\mathcal{O}$ feature and focus on the $\mathcal{R}$ regime. The resulting normalized yields $Y(H^0)/2\Phi_0$ are shown in Fig. 2(c).

Our model for the reconstitution process of H^0

and H_2^+ assumes the following sequence of events:

(i) The incident randomly oriented H_2^+, characterized as a rigid body with an interproton separation R_0, is stripped of its electron upon penetrating the foil; we tag this time as $t = 0$.

(ii) Inside the foil, the internuclear separation R of the diproton is altered by the mutual Coulomb repulsion and the effects of energy-loss straggling and multiple scattering.

(iii) At the exit side of the foil, $t = t_D$, $R(t_D)$ determines the probability for the diproton to capture a target electron, and this can lead either to a bound H_2^+ or to separated $H^0 + H^+$ upon emergence into the vacuum, depending upon the final state and the internal kinetic energy $\epsilon(t_D) = \frac{1}{2}\mu\dot{R}^2$ of the diproton.

The assumption in (i) that the electron is stripped at $t = 0$ is justified by the fact that the measured lifetime of H_2^+ in carbon is much shorter than the t_D values considered here. Vibrational excitation in the incident H_2^+ is known to be present and this can be characterized[8] by a distribution $D(R_0)$ in R_0 values. In (iii), the assumption that the capture event occurs at the exit surface is based on the observation[3] that, in the corresponding case of atomic projectiles, the cross section for electron loss, σ_l, is much larger than that for electron capture, σ_c, and this implies that only those reconstituted near the exit surface would be likely to emerge from the foil. Moreover, these σ_c are found[3] to be consistent with theoretical expectations for capture into a free-atom electronic state. The latter observation indeed provided the justification for our use of free molecular-orbital states (MO) of H_2^+ as the final states for electron capture by the diproton.

For a given set of $R(t_D)$ and $\epsilon(t_D)$, we consider only capture into the $1s\sigma_g$ and $2p\sigma_u$ states[9] and label their electronic energies by $U_g(R)$ and $U_u(R)$, respectively, relative to that of the $H^0(1s) + H^+$ separated atoms, which is taken to be zero. A capture into the repulsive $2p\sigma_u$ state will always lead to an H^0. In the case of the $1s\sigma_g$ ground state, an H^0 will also be produced unless $[\epsilon + U_g(R)] \leq 0$, in which case a bound H_2^+ is formed. At present there is no satisfactory theory of electron capture into MO states. We may, however, expect from a number of observations that the cross sections for capture into the two MO can be reasonably expressed as

$$\sigma_c^{g,u}(R, V) = [Z_{g,u}(R)]^5 \sigma_c^{H}(V), \quad (1)$$

where σ_c^{H} refers to that of capture by a proton of the same velocity leading to an $H^0(1s)$ and $Z_{g,u}(R)$ characterizes the effective nuclear charge of the MO. The proportionality to σ_c^{H} is based on the fact that the experimental $Y(H_2^+)/2\Phi_0$ and $Y(H^0)/2\Phi_0$ ratios are nearly independent of V (see Fig. 2) and that the ground-state σ_l is nearly the same[2,3] for H^0 and H_2^+ at the same velocity. Given the success of the hydrogenic scaling of σ_c in terms of effective charge in the atomic case,[10] it is natural to try to extend this to the present case of a simple molecule. This then accounts for the $Z_{g,u}^5$ factor. Indeed an R-dependent effective charge concept for H_2^+ arises naturally from a LCAO description as given, for example, by McCarroll *et al.*,[11] and we use their results here for $Z_{g,u}(R)$.

Considering only Coulomb repulsion inside the foil and weighting the results with the $D(R_0)$ distribution gives a reasonable simulation for the yield of H^0 but predicts no H_2^+ reconstitution products beyond $t_D \simeq 4$ fs, as illustrated by the dashed curves in Figs. 2(c) and 2(b), respectively. The predicted cutoff for the H_2^+ case is not difficult to understand. Mutual Coulomb repulsion serves always to increase R and ϵ with increasing t_D for a finite value of R_0. There will thus be a t_D beyond which the bound-state condition $[\epsilon + U_g(R)] \leq 0$ can no longer be satisfied. Since the observed H_2^+ yield shows no discernible discontinuity, we surmise that multiple-scattering effects play a critical role in the longer-t_D region since they introduce distributions in the R and ϵ values for a given R_0. Energy-loss straggling will also induce a spread in the R and ϵ values, but the effects are estimated to be comparatively small.

To minimize computation, we treat the multiple-scattering processes for the two protons in the cluster as uncorrelated with one another[12] and with the Coulomb explosion process. If multiple-scattering effects on each proton result in a Gaussian profile of width σ_p in a lateral direction, the resulting distribution of interproton separations projected onto the same lateral direction for a pair initially collinear in a longitudinal track will also be a Gaussian but with a width $\sigma_s = \sqrt{2}\sigma_p$. More generally the distribution of $R(t_D)$ for a given R_0 may be obtained by considering the coordinates shown in Fig. 2(a). Coulomb explosion from an initial $\vec{R}_0$ directed at an angle θ relative to the beam direction $\overrightarrow{OA}$ results in the vector $\vec{R}_c = \overrightarrow{OB}$. Its lateral component $\vec{r}_1 = \overrightarrow{AB}$ is spread out by multiple-scattering effects $\vec{s} = \overrightarrow{BC}$, giving rise to a distribution in $\vec{R}_1 = \overrightarrow{AC}$ and thus

in $\vec{R}=\overrightarrow{OC}$. Since $\vec{s}$ is randomly oriented in the lateral plane with a distribution

$$P(s,\varphi)s\,ds\,d\varphi = (2\pi\sigma_s^2)^{-1}\exp(-s^2/2\sigma_s^2)s\,ds\,d\varphi, \tag{2}$$

the distribution in the magnitude of $\vec{R}=\vec{R}_c+\vec{s}$ can be written as

$$Q(R;R_c,\theta)dR = (R/\sigma_s^2)\exp[-(R_1^2+R_c^2\sin^2\theta)/2\sigma_s^2]I_0(z)dR, \tag{3}$$

where $I_0(z)$ is the zeroth-order modified Bessel function, $z=(R_1R_c\sin\theta)/\sigma_s^2$, and $R_1=(R^2-R_c^2\cos^2\theta)^{1/2}$. In going to Eq. (3) from Eq. (2), we have first made a transformation from the (s,φ) to the (R_1,ψ) space and integrated over ψ from 0 to 2π. A similar consideration applied to the relative velocity $v=\dot{R}$ of the two protons results in a distribution $Q(v;v_c,\theta)dv$ identical in form to Eq. (3) but with a corresponding multiple-scattering width σ_w.

From the above considerations, the probability that a diproton, from a randomly oriented population, will have a separation R and relative velocity v at foil exit is then

$$S(R,v;R_c,v_c) = \tfrac{1}{2}\int_0^\pi d\theta\,\sin\theta Q(R;R_c,\theta)Q(v;v_c,\theta). \tag{4}$$

With this, the normalized H_2^+ yield follows as

$$Y(H_2^+)/2\Phi_0 = \tfrac{1}{2}\int_0^\infty dR_c[D(R_0)dR_0/dR_c]\int_0^\infty dR Z_g^5(R)\int_0^{v'} dv S(R,v;R_c,v_c), \tag{5}$$

where $v'=[-2U_g(R)/\mu]^{1/2}$ (μ being the reduced mass of the diproton) is the largest value of v for which the bound-state criterion is satisfied.

Numerical evaluation of Eq. (5) for 0.8-MeV/amu projectiles that uses multiple-scattering widths σ_s and σ_w deduced from the simplest multiple-scattering theory[13] results in the solid curve shown in Fig. 2(b). The agreement with the H_2^+ data is remarkable in view of the fact that the data span four orders of magnitude and no adjustment was made in any parameters of the model. Use of widths based on a more recent theory[14] alters the predicted yields slightly but not the overall agreement with the data. The clustering of the data for different V into a near-universal curve of t_D is also reproduced. For example, calculated yields for 0.4-MeV/amu H_2^+ are reduced only by 40% at $t_D=20$ fs and less with decreasing t_D. Such differences are well within the range of scatter of the data points. The predictions were also found to be relatively insensitive to reasonable changes in the $D(R_0)$ distribution. For the H^0 case, multiple-scattering effects are expected to be unimportant because of their comparatively much larger yields and the calculated results confirmed this expectation.

Finally we mention one additional support for the model. Our measurements at Argonne National Laboratory of the 0° energy distribution of H^+ from the foil breakup of 1-MeV H_2^+ showed that the two Coulomb explosion peaks lie closer together when the H_2^+ beam is composed of those H_2^+ which had survived a previous passage through a 330-Å carbon foil. Our calculations of the H^+ energy distribution for this case reproduced the data well. The smaller explosion effect is essentially due to the larger R of the reconstituted H_2^+.

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Time Correlations between the Two Sidebands of the Resonance Fluorescence Triplet

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A new type of time correlation analysis of resonance fluorescence is presented. A strontium atomic beam is excited by a 28-Å–off-resonance laser. The photons of the two sidebands of the fluorescence triplet are shown to be emitted in a well-defined time order. A simple interpretation of this effect is given which implies a quantum jump of the atom from the lower to the upper state through a multiphoton process.

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Resonance fluorescence (i.e., scattering of radiation by free atoms irradiated by a resonant or quasiresonant laser beam) has been extensively studied during the last few years. First, it has been predicted[1] and observed[2] that, for two-level atoms and at high laser intensities, the fluorescence spectrum consists of three components (fluorescence triplet). More recently, the distribution of time intervals between photoelectric counts recorded on the scattered light has been measured, giving evidence for an antibunching of the fluorescence photons originating from a single atom.[3]

These two types of experiments emphasize, respectively, the frequency or time features of resonance fluorescence. One can also consider the possibility of a mixed analysis dealing with the time correlations between fluorescence photons previously selected through frequency filters[4] (the frequency resolution $\Delta\nu$ introduces, of course, an uncertainty $\Delta t = (\Delta\nu)^{-1}$ in the determination of the emission time). If, for example, the three components of the fluorescence triplet are well separated (their splitting Ω being much larger than their widths γ), one can use filters centered on any one of these components and having a width $\Delta\nu$ such that $\gamma \ll \Delta\nu \ll \Omega$. With such filters, it is possible to determine which components of the triplet the detected photons are coming from and, simultaneously, to study the statistics of the emission times with a resolution Δt better than the atomic relaxation time γ^{-1}.

In this Letter, we report the first experimental investigation of time correlations between frequency-filtered fluorescence photons. In this experiment, the detuning $\delta = \omega_L - \omega_0$ between the laser and atomic frequencies ω_L and ω_0 is much larger than the Rabi nutation frequency ω_1 (off-resonance excitation) so that the splitting $\Omega = (\omega_1^2 + \delta^2)^{1/2}$ is simply equal to the detuning δ. The three components of the triplet are therefore located at ω_L for the central component (Rayleigh scattering) and $\omega_A = \omega_L + \Omega \simeq 2\omega_L - \omega_0$ and $\omega_B = \omega_L - \Omega \simeq \omega_0$ for the two sidebands. The experiment hereafter described shows that the photons of these two sidebands, selected by two filters centered at ω_A and ω_B, are correlated and emitted in a well-defined order (ω_A before ω_B).

We use a strontium atomic beam ($^1S_0$-$^1P_1$ resonance line; $\lambda_0 = 460.7$ nm) irradiated by the 28-Å–off-resonance blue line of an argon-ion laser ($\lambda_L = 457.9$ nm). The multimode-laser light (1 W power) is focused onto the atomic beam (laser-beam waist less than 10 μm) and focused back by a spherical mirror in order to double the laser intensity in the interaction region. In these conditions, the Rabi nutation frequency ω_1 is much smaller than the detuning δ ($\omega_1/2\pi = 80$ GHz and $\delta/2\pi = 4000$ GHz) and the central line of the fluorescence triplet is about 10^4 (i.e., $4\delta^2/\omega_1^2$) times

