Inhibition of H^- production in the foil breakup of fast H_2^+

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Comparison of emergent H^- yields for incident H^+ and H_2^+ beams of the same velocity (0.4, 0.8, and 1.2 MeV/amu) traversing carbon foils of 1–8 $\mu g/cm^2$ shows that, per incident proton, the H^- yield from H_2^+ is generally inhibited, in striking contrast to the enhanced H^0 yield previously observed. An explanation of the major inhibition effect is given in term of the Landau-Zener approximation to a "one-passage" charge transfer from the H^- to its H^+ breakup partner.

I. INTRODUCTION

A fast molecular ion (~1 MeV/amu) traversing a thin foil may be expected[1,2] to be stripped of its valence electrons in a time period short compared to a typical dwell time in the foil of $t_D \sim 1$ fs (10^{-15} s). The resulting explosion will of course increase the internuclear separation $R(t_D)$ of the breakup fragments at the foil exit, but for $t_D \sim 1$ fs and singly ionized fragments, this increase will amount to only a fraction of the original value.[1] Moreover, the comparatively much larger electron-loss cross section from a valence state over that for capture[1,2] will help to ensure that a target electron capture by an emergent cluster will most likely occur near the foil exit. This combination of factors seems tailored for the examination of the processes of capture into molecular-orbital states at various R and of electron sharing among the eventually separating fragments outside of the foil. The latter aspect is particularly interesting since, for diatomic molecules, it is intimately connected to that encountered in the charge changing atomic collisions at low energy,[3] except that here the charge-transfer probability is sampled directly because of the "one-passage" condition. We report on the manifestation of such a one-passage charge transfer in the observed inhibited production of H^- from the foil breakup of H_2^+.

II. EXPERIMENT

Our measurements consist of the emergent H^- yields from beams of H^+ and H_2^+ of 0.4, 0.8, and 1.2 MeV/amu traversing carbon foils 1–8 $\mu g/cm^2$ thick. The experimental set up is similar to that described elsewhere.[1] Briefly, the magnetically analyzed incident beam from a Van de Graaff accelerator was first collimated by a pair of apertures, 3 m apart, to a spot size of 0.15 mm diameter before striking the target. Emergent H^- were counted for a preset number of large-angle elastic scattering monitor events by magnetically deflecting them into a surface barrier detector positioned at 30° to the incident beam direction. Since each H^- run was immediately followed by a measurement of the transmitted H^+ without moving the beam spot and for the same number of monitor events, the target thickness was also determined for each run. The sensitive area of the detector is sufficiently large to intercept practically all of the H^+ from the H_2^+ breakup for the thickest foil used, and thus also all of the corresponding H^- because the latter have a narrower angular distribution.[4]

III. RESULTS

The measured yields of H^- *per proton* for incident H_2^+ molecules Φ_1^M, at the three velocities are shown in Fig. 1 for the various dwell times

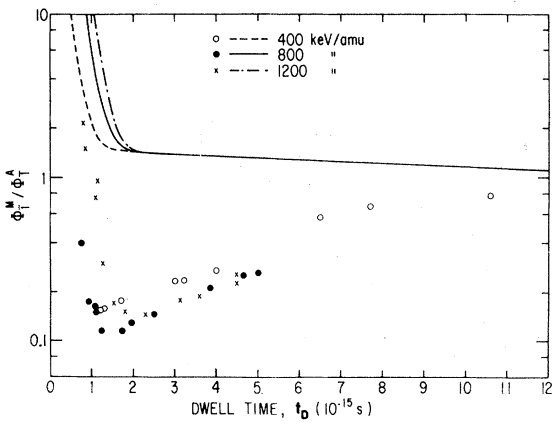

FIG. 1. Total yields of emergent H^- *per proton* Φ_1^M from H_2^+ incident on carbon foils normalized to the corresponding yields Φ_1^A from incident H^+ of the same velocity displayed as a function of the dwell time t_D in the target (symbols). For a given velocity, values of Φ_1^A measured over this t_D interval are constant to within ±5% and have a mean value of 10^{-7}, 2.9×10^{-8}, and 4.1×10^{-9}, respectively, for 0.4, 0.8, and 1.2 MeV/amu. Experimental uncertainties in the Φ_1^M/Φ_1^A ratios and t_D are estimated to be ±10% or less. The solid curves represent the corresponding ratios anticipated if the capture into the H_2 MO correlated with the $H^- + H^+$ all lead to H^- (see text).

t_D in the target. These yields have been normalized to the corresponding H^- yields for incident H^+, Φ_1^A, whose values at a given velocity were measured to be independent of t_D in the range examined here. That molecular effects are important is clearly demonstrated by the deviation from unity of the Φ_1^M/Φ_1^A ratios and by the fact that this deviation is nearly independent of the velocity, but depends only on t_D, which in turn determines R and its time derivative $\dot{R}$ at the foil exit. Such nonequilibrium features of atomic fragment yield have of course been previously observed[1] for H^0 from H_2^+ breakup under the same experimental conditions, but there is one striking contrast. Whereas the yield of H^0 *per proton* for incident H_2^+ relative to that for incident H^+, Φ_0^M/Φ_0^A, is observed to be always larger than unity and to approach unity from above for large t_D, the Φ_1^M/Φ_1^A ratio here is significantly less than unity, except for the shortest t_D, and tends to approach unity from below for large t_D. Such an inhibition of H^- would not be surprising if the H^- can be viewed as a point charge. This is because the H^- is almost always accompanied by a nearby H^+ for incident H_2^+, which is never the case for incident H^+, and the Coulomb attraction between H^- and H^+ can surely cause the H^- to lose an electron before the two separate to a large distance. However, H^- is not a point charge and, for the range of R concerned here, an explanation must take the molecular aspects explicitly into account.[5]

IV. DISCUSSION

A. Electron capture—enhancement only

In order to produce a two-electron H^- from a one-electron H_2^+, a capture of at least one electron must have occurred inside the target. Since the lifetimes of these "bound" states inside a solid are known[1,2] to be $\tau \lesssim 0.2$ fs and the t_D range examined here is $>3.5\tau$, a two-electron cluster must have been formed in the last few atomic layers of the target if it were to survive and emerge into the vacuum. We may thus speak of capture into the usual molecular-orbital states (MO) of H_2. For simplicity we consider only those MO(H^-) that correlate with the $H^-(1s^2)+H^+$ channel at $R=\infty$, namely, the $\bar{H}^1\Sigma_g$ and $\bar{B}^1\Sigma_u$ orbitals.[6]

There are two possibilities for forming these MO(H^-). If the incident H_2^+ molecule survived with its original electron intact, an event which is less likely the longer is t_D,[2] the capture of only one target electron is required. Otherwise the capture of two electrons by a diproton cluster is necessary. Although the cross section for the capture of two electrons σ_{2c}^M may be expected to be much smaller than that of one electron σ_{1c}^M, the preponderance of the number of diproton clusters with increasing t_D will give rise to an increasing dominance of the two-electron capture process. If we denote by σ_{2c}^A the H^- formation cross section from a single incident H^+, we will have $\sigma_{2c}^M \geq 2\sigma_{2c}^A$ because the spatial correlation of the two protons at finite R will increase the capture probability over that for $R=\infty$. Since we also have $\sigma_{1c}^M \gg \sigma_{2c}^M$, we may expect the capture processes to lead only to an enhanced H^- yield in the absence of channel coupling and provided that the trapping into a bound H_2 state in the attractive part of the MO(H^-) potentials can be neglected. Indeed we may surmise from the analogous case[1] of H^0 yield from H_2^+ that this trapping effect cannot account for the magnitude of inhibition observed here. It is thus reasonable to conclude that the major inhibition process occurs outside of the foil and the most likely one is charge exchange at the pseudocrossings of the H_2MO (Ref. 7).

A quantitative assessment of the relative contributions of the trapping and charge-exchange effects can be facilitated by introducing a quantity called the H^- survival probability, defined as

$$S = \Phi_1^M/\phi_1^M, \tag{1}$$

where ϕ_1^M is what the corresponding yield would be if the capture into the MO(H^-) all lead to H^-. Values for S implied by the data in Fig. 1 can be deduced by assuming that the enhancement in the capture process scales according to that observed[1] for the H^0 case as

$$\phi_1^M/\Phi_1^A = \Phi_0^M/\Phi_0^A \tag{2}$$

for each incident velocity V and t_D. The ϕ_1^M/Φ_1^A so constructed are shown as curves in Fig. 1. Equation (2) is consistent with the process in which the H_2MO correlated with H^-+H^+ are formed by a sequential two-electron capture into a cluster in which the two protons are moving away from each other and the intermediate states are the H_2^+MO correlated with H^0+H^+. It attributes the enhancement entirely to the first step of the two-step capture process and thus may lead to underestimates of the ϕ_1^M. On the other hand, the deduced values for S based on Eqs. (1) and (2) and shown in Fig. 2 are seen to cluster into a trend conforming with the expectation that S should not depend on V, and thus suggests that such a characterization of the MO(H^-) formation may not be unreasonable. Strictly speaking, these deduced S values already include some correction for the trapping effect since we have used experimental values for the Φ_0^M/Φ_0^A ratios in Eq. (2). As we shall see shortly, this is of minor consequence as the trapping effect is small except for very

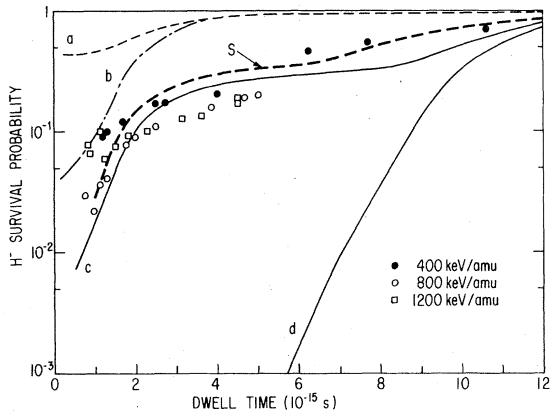

FIG. 2. Experimentally deduced probabilities for H^- surviving the trapping effect and a charge-transfer process (symbols), and the various theoretical predictions for S discussed in the text (curves).

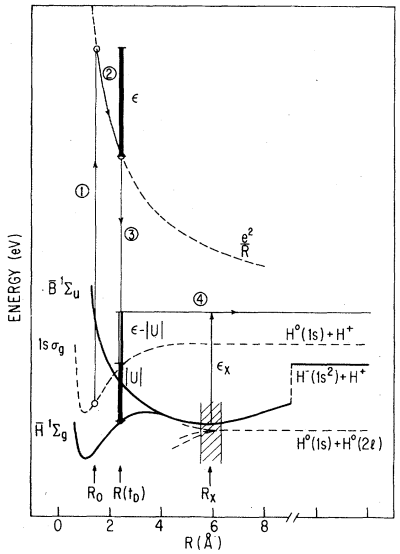

FIG. 3. Schematic diagram for the various MO's relevant to the inhibition mechanisms discussed in the text.

short t_D. In any event, the deduced values in Fig. 2 do define an upper bound for S in that t_D range.

B. Models for yield inhibition

According to Eq. (1), S should reflect both the trapping effect S_{tr} and the charge-exchange effect S_{ex} as $S = S_{tr}S_{ex}$. In estimating S_{tr}, we considered capture into the $\bar{H}\,^1\Sigma_g$ and $\bar{B}\,^1\Sigma_u$ MO of H_2 (Ref. 6) and followed the procedure used in our successful account[5] of the observed H_2^+ transmission yields. Referring to Fig. 3, the following sequence is pictured: (1) at the instant of penetration into the foil $t = 0$, the electron is stripped from the incident $H_2^+(1s\sigma_g)$ with an interproton separation of R_0, (2) the diproton then undergo mutual Coulomb repulsion inside the foil and this determines the R and $\dot{R}$ and thus also the internal kinetic energy of the diproton ϵ at the foil exit $t = t_D$, and (3) electron capture into either of the two MO(H^-) occurs at this point and this may lead to an H^-. In particular, applying a simple energy criterion, we have

$$S_{tr} = \begin{cases} 0, & \text{for } [\epsilon + U] \leq 0 \\ 1, & \text{otherwise.} \end{cases} \tag{3}$$

Here U is the MO(H^-) potential at $R(t_D)$ expressed relative to the $H^- + H^+$ separated atom energy.

In step 2 above, multiple scattering effects acquire importance with increasing t_D which, for the present purpose, may be viewed to introduce distributions in the R and ϵ values at the foil exit. These slight complications will initially be ignored in order to make clear the specific effects of the inhibition mechanisms. The results of including multiple scattering effects will be discussed later. Suffice it to note at this point that the essential conclusions to be drawn from their omission will remain unaltered.

Estimates of S_{tr} are shown as curves a and b in Fig. 2. These results have been averaged over a distribution $D(R_0)$ of initial R_0 values[8] which serves to characterize the vibrational distribution of the incident H_2^+ beam. The two curves differ only in the relative formation probabilities assumed for the two MO(H^-). For curve a, equal probabilities are used for the $\bar{H}\,^1\Sigma_g$ and $\bar{B}\,^1\Sigma_u$, while for curve b, these probabilities are assumed to scale with those[5] for the $1s\sigma_g$ and $2p\sigma_u$, respectively. In effect the $\bar{H}\,^1\Sigma_g$ is weighted much more heavily in curve b. The results in Fig. 2 clearly show that S_{tr} by itself can account for only a fraction of the observed inhibition, in agreement with our earlier inference.

For those capture events that survive trapping, charge exchange may occur before the pair separate far apart, as indicated by step 4 in Fig. 3. This can prevent H^- from emerging. The simplest description of the charge-exchange effects occurring here is provided by the Landau-Zener (LZ) model.[7] In this model, a two-state approximation is used and the electron transfer is assumed to occur at the pseudocrossing points R_x of the two MO correlated with the $H^-(1s^2) + H^+$ and $H^0(1s) + H^0(nl)$ channels. The probability that a transfer did not occur in a single passage through R_x may be written as[9,10]

$$S_{ex}=\begin{cases}\exp-\left(\dfrac{2\pi}{\hbar v e^2}|H_x|^2R_x^2\right), & R(t_D)\le R_x\\ 1, & R(t_D)>R_x\end{cases} \tag{4}$$

where H_x is the coupling energy term and v is the relative velocity of the two protons at R_x. Bates and Lewis (BL)[7] have shown that there are two well separated crossing points to consider in the $H^- + H^+$ case; at $R_{x1}=5.8$ Å and $R_{x2}=18.8$ Å corresponding to the $n=2$ and 3 levels of H^0 with $H_{x1}=0.526$ eV and $H_{x2}=9.27\times10^{-3}$ eV, respectively. In the present case for which the MO(H^-) are generally formed with $R(t_D)<R_{x1}$, the effect of the second crossing point R_{x2} is minimal since $S_{ex}=S_{ex1}S_{ex2}\simeq S_{ex1}$.

The results of combining Eqs. (3) and (4) in the form $S=S_{ex}S_{ex}$ and taking into account both crossing points and the $D(R_0)$ distribution are shown as curve d in Fig. 2. The inhibition is now overestimated. A better characterization of the experimental points can be obtained by simply reducing the H_{x1} value of BL by a factor of 3 as shown by curve c. Such a reduction of the coupling parameter may reflect either or both of the following possibilities: (1) the LZ model is inadequate and (2) the procedure used to deduce the experimental S values underestimates the electron capture cross sections for the MO(H^-). While the former is likely because of other collaborative arguments,[11] the latter cannot be ruled out definitely even though we have given in Sec. IV A a plausible argument that it may not be the case. In spite of these uncertainties, we may nevertheless conclude from the comparisons in Fig. 2 that the major inhibition effect for $t_D\gtrsim 2$ fs is due to the charge-exchange mechanism. Moreover, the LZ model with one adjustable parameter is seen to provide a reasonable characterization of the observation and this enables us to examine the effects of multiple scattering.

As mentioned earlier, multiple scattering essentially introduces distributions in both the R and ϵ values for a given R_0. When these distributions, deduced in a manner suggested in Ref. 5, is folded into a set of repeat calculations for S with the reduced H_{x1} parameter, we obtain the curve labeled S in Fig. 2. The inflection point in curve c at $t_D=8.5$ fs which is a manifestation of the R_{x1} crossing point is now smoothed out and this brings the predicted results in closer agreement with the experimental points in this t_D region, and thus adding confidence to our overall description. We mentioned for completeness that the effects on the S_{tr} (curves a and b) are minor, while on the S corresponding to the original set of BL parameters (curve d), although more noticeable, still do not raise the predicted S significantly closer to the experimental points. For example, the value at $t_d=6$ fs is raised only by a factor of 6 as compared to the required 200.

C. Comparison with binary collisions

The same charge-transfer process discussed here is encountered in the mutual recombination phenomenon observed in low-energy $H^- + H^+$ collisions.[12,13] In the collision case, because the partners first coalesce and then recede from each other, the charge-transfer probability $P=1-S_{ex}$ enters[3] as $2P(1-P)$ for a given impact parameter b. An integration over all b is required to obtain the charge-transfer cross section σ_{ex}. As a consequence, the effects of the different crossing points are weighted differently than the present one-passage case. Nevertheless, the observed[13] discrepancies between the collision data and the predictions of the LZ model using the BL parameters corresponding to the present range of v, do not contradict the suggestion here that the H_{x1} value be reduced. There the predictions lie below the data nearly uniformly by a factor ~4 over the range of 0.2–20 eV in collision energy. Reducing H_{x1} below the BL value will raise the predicted σ_{ex} as long as the R_{x1} crossing point is accessible. Whether or not the nearly uniform factor can be reproduced for that energy range by this single-parameter adjustment can only be answered by a complete calculation for that case.

Questions have been raised[11] concerning the applicability of the LZ model. The main objection lies in the drastic approximation of a sharp transition region inherent to the model. Our analysis here suggests that the study of one-passage case offers perhaps the most direct means for examining the shape of this transition region, particularly if the incident molecules can be prepared with a narrower R_0 distribution.

V. SUMMARY AND CONCLUSION

Yields of H^- from the breakup H_2^+ molecules in carbon foils have been measured over a wide range of thicknesses at three incident velocities. Compared to those for incident H^+ at the corresponding conditions, the yields from H_2^+ are consistently lower for the thicker targets when normalized to the same number of incident protons. This marked the first time that such nearly velocity-independent, inhibited normalized atomic fragment yields from molecular breakup in thin foils have been noted. Inhibited yields have also been observed subsequently in our preliminary studies of H^- from H_3^+ and HeH^+ and of H^0 from HeH^+. Arguments presented show that target

electron capture by a cluster of two positive charge centers can only lead to enhanced yields and thus inhibition mechanisms are attributed to the sequence after the capture. Explicit calculations using a simple model of capture into H_2 MO correlated with $H^- + H^+$ and charge exchange at the MO pseudocrossing points indicate that the post-foil charge exchange is the dominant inhibition mechanism. These results further suggest that, because of the one-passage condition, the study of thin foil breakup of fast diatomic molecules perhaps provide the most direct information on the charge-transfer process between atoms in close proximity to each other.

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