Formation of Fast Excited H Atoms. III. Collisional Dissociation of H_2^+ and H_3^+ on Helium*

J. C. Ford, F. M. McCoy, R. Conrads, and E. W. Thomas†
School of Physics, Georgia Institute of Technology, Atlanta, Georgia 30332
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The collisional dissociation of 75- to 700-keV H_2^+ and H_3^+ by impact on targets of He is studied. The formation of excited hydrogen is detected by quantitative measurement of collisionally induced Balmer-α emission; the contributions from $3s$, $3p$, and $3d$ levels are separated by a method that utilizes the different lifetime of the excited states. Cross sections are almost the same for each of the $3s$, $3p$, and $3d$ states; they vary slowly with projectile energy.

I. INTRODUCTION

A study has been made of the collisional dissociation of fast H_2^+ and H_3^+ on a helium target leading to the formation of excited hydrogen in the $3s$, $3p$, and $3d$ states. The processes can be described by the reaction equations

$$H_2^+ + He \rightarrow H^* \ (3s,\ 3p,\ 3d) + [H^+ + He], \qquad (1)$$

$$H_3^+ + He \rightarrow H^* \ (3s,\ 3p,\ 3d) + [H_2 + He]. \qquad (2)$$

The experiments give no information on the states of ionization, excitation, or molecular association of those post-collision products shown within the square brackets. The data are in the form of separate cross sections for the formation of the $3s$, $3p$, and $3d$ states; impact energies range from 100 to 800 keV.

The techniques of this experiment are essentially the same as those used in a preceding paper[1] for the study of how the same $3s$, $3p$, and $3d$ states are formed in charge transfer as protons traverse various targets. That preceding paper will be denoted hereafter as I. The discussion of experimental techniques is not repeated in detail here; the reader is referred to I for a complete analysis.

II. EXPERIMENTAL METHOD

Projectile ions are produced in an r.f. source—accelerated, mass analyzed, and directed through a cell containing the target gas. On emergence from the cell the projectile beam contains some excited hydrogen atoms formed by dissociation. A photomultiplier with interference filter is used to detect Balmer-α photons emitted by the excited H atoms. The Balmer-α line includes contributions for the $3s$, $3p$, and $3d$ states; each of which has a different lifetime. The intensity from one state will decay exponentially with distance from the gas-cell exit. Experimentally, a measurement is made of intensity as a function of distance from the gas-cell exit; the data is then fitted by a sum of three exponential decays corresponding to the lifetimes of the $3s$, $3p$, and $3d$ states. This procedure gives the population of the states at the exit from the cell and hence the cross sections for their formation.

The full explanation of the experimental procedures is given in I. Included there are detailed discussions of how light intensity is related to cross sections and of the methods utilized for absolute determination of the important experimental parameters. We will consider here only those matters in which the present experiments on dissociation differ from the previous work on charge transfer.[1]

Dissociation proceeds by excitation of the molecule to a repulsive state; the potential energy of the repulsive state is converted to kinetic energy of the fragments. The axis of the molecule may be inclined at any angle to the direction of motion of the center of mass. Consequently the kinetic energy released in the molecule's rest frame will cause the dissociated fragments to exhibit both a distribution of speeds and a distribution of angles when observed in the laboratory frame of reference. The procedure by which cross sections are determined from the intensity distribution requires that we know the emitting-atom's velocity component along the line of the projectile beam. We use a velocity which is calculated on the assumption that one may neglect the additional velocity due to dissociation. Thus the velocity of the excited atom produced by dissociation of an H_2^+ molecule of energy E is assumed, for the analysis, to be given by $(E/M)^{1/2}$; where M is the mass of a hydrogen atom. Similarly the velocity of a hydrogen atom produced by dissociation of H_3^+ is assumed to be $(2E/3M)^{1/2}$. One must consider whether the neglect of the released kinetic energy will cause errors. In I we have discussed a procedure for assessing the error in cross sections caused by uncertainty in particle velocity; that same procedure may be carried out here. We assume that the dissociation process releases no more than 10 eV of potential energy; this will cause no more than a 1% change in velocity component along the primary-beam direction. Following the previously discussed procedures we find that a 1% error in velocity will

produce an error in the $3s$, $3p$, and $3d$ cross sections of 1% or less. It is therefore concluded that, to within an accuracy of 1%, the velocity distribution produced by dissociation does not influence the validity of the analysis procedures. The angular distribution of the dissociated fragments might cause errors if any appreciable fraction of the particle flux were to be intercepted by the exit aperture of the target cell. If one again assumes that the released energy is 10 eV or less then the angular distribution of the dissociated fragments cannot exceed $\pm\frac{1}{2}^\circ$ from the direction of the primary beam. The exit aperture of the gas cell subtends an angle of 1° at the entrance aperture of the cell; thus dissociation fragments produced at any point in the cell will be able to pass through the exit. This situation was confirmed by measuring the flux of ions and neutrals to an annular ring placed just inside the exit aperture. That flux was less than 0.01% of the incident beam flux when there was no gas in the cell; the flux then represents the divergence of the primary beam. With target gas in the cell, the flux remained essentially unchanged, indicating that the interception of projectile fragments was negligible.

For the velocities of excited H atoms used here the decay length for the short lived $3p$ state becomes very short, typically of the order 1 cm. Consequently, the separation of the $3p$ contribution from the $3s$ and $3d$ must be based on perhaps only one good measurement of intensity. Moreover, corrections due to the spatial pressure gradient at the exit of the target cell (see I) will be very large. It follows that the accuracy of the $3p$-state measurement is very poor, and in fact, no reliable estimate of $3p$ cross section could be achieved at energies below 300 keV.

Of course, the experiments were carried out under single-collision conditions where the collisionally induced light intensity varied linearly with target pressure. For the dissociation experiments this required the target pressure to be maintained below 2.5×10^{-4} Torr.

III. CASCADE

The process under study is the direct formation of the excited atom by dissociation. Population of the excited states by secondary-collision mechanisms is excluded by the use of sufficiently low target pressures. It is impossible, however, to exclude the population caused by cascade transitions from higher levels. Cascade has two important effects. First, it does of course complicate the relationship between the measured population and the cross section to be derived from it. Secondly, it introduces an additional term into the decay scheme characterized by the lifetime of the cascading state. This latter consideration is most important because a substantial cascade contribution will invalidate the whole basis of the analysis. In I the influence of cascade on the decay scheme was discussed in some detail.

In the present work the analyses for $3s$-, $3p$-, and $3d$-state populations were carried out on the assumption that cascade could be neglected—essentially the same procedure as in I. Tests for influence of cascade were carried out using a cascade-decay equation [Eq. (9) of I] instead of the normal equation [Eq. (4) of I]. Simple analysis suggests that cascade from the $4f$ state into $3d$ will be most serious; this is because the branching ratio for decay into the $3d$ state is unity and the lifetime of the $4f$ state lies between that of the $3s$ and $3d$ levels. Analysis of the data with a term accounting for cascade from the $4f$ level did not significantly change the cross sections derived for the $3s$, $3p$, and $3d$ levels. The small differences (of the order 5% or less) between cross sections determined with and without cascade are considered to be a contribution to the uncertainty of the experiment; this is included in our over-all estimate of possible error.

In the previous measurements on charge transfer (see I) the population of high-angular-momentum states is small and cascade is negligible. In dissociation there is a substantial population of high-angular-momentum states and cascade is detectable; however the influence on the cross-section measurements is still small.

IV. RESULTS

In Figs. 1 and 2 are shown the cross sections for formation of H ($3s$) and H($3d$) by dissociation of H_2^+ and H_3^+ on helium. The measurements for the $3p$ state are of rather poor quality and are not shown here. Within a rather large experimental uncertainty the cross section for formation of H($3p$) are the same as that for formation of H($3s$) and H($3d$).

It is observed that all three cross sections are

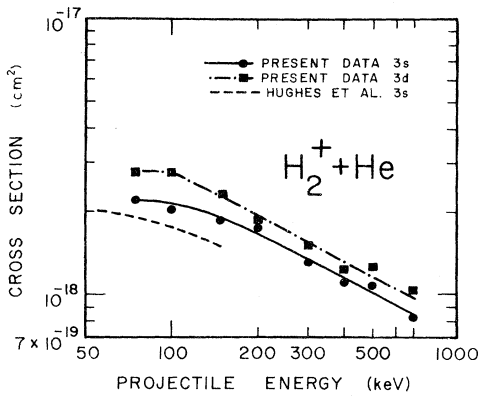

FIG. 1. Cross sections for the formation H($3s$) and H($3d$) atoms by dissociation of H_2^+ on a target of He. Present measurements are shown along with previous measurements for the H($3s$) state by Hughes *et al.* (Ref. 2).

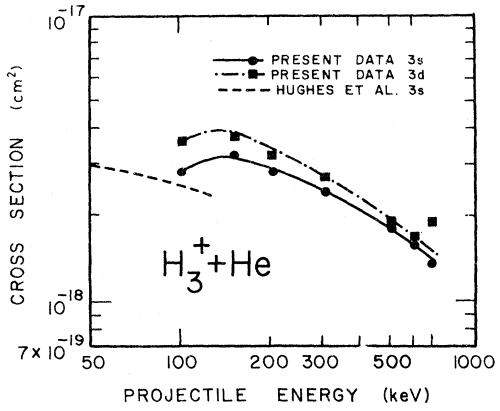

FIG. 2. Cross sections for the formation of H(3s) and H(3*d*) atoms by dissociation of H_3^+ on a target of He. Present measurements are shown along with previous measurements for the H(3s) state by Hughes *et al.* (Ref. 2).

about equal for each collision combination and that these cross sections vary only slowly with incident projectile energy. Also shown on the figures are previous measurements of H(3s) formation carried out by Hughes *et al.*[2]; these data were obtained in a similar manner to those of the present experiments. Within experimental error, the data by Hughes *et al.*[2] agree with the present work.

The error limits of these data are estimated by the same arguments used in I. Possible systematic errors are again assessed as being ± 16.5%. Random uncertainties in the data are generally less than those for the corresponding charge-transfer reaction because in dissociation the light intensities are higher. It is estimated that the possible random error in the dissociation cross sections for the 3*s*, 3*p*, and 3*d* states does not exceed ± 10%. No account was taken of the possible polarization of emission from the 3*p* and 3*d* states.

When carrying out studies with H_2^+ and H_3^+ projectiles, one must consider the possible influence of vibrationally excited ions in the primary beam. It is expected that an ion source such as the one used in the present experiments will produce molecular ions in various states of vibrational excitation; the various states will exhibit different cross sections. Consequently, a dependence of cross section on ion-source operating conditions might be observed. Williams and Dunbar[3] observed substantial source effects on cross sections for dissociation of H_2^+ at 6 keV but only very small effects at 50 keV. McClure[4] also finds large effects at low energies (10 keV), while Barnett and Ray[5] find negligible effects between 40- and 200-keV impact energy. Within the present experiments a search was made for any systematic dependence of cross section on ion-source operating conditions. No such systematic changes were found, leading us to conclude that excited-state content of the primary beams is unimportant. Clearly such tests are not conclusive, and there are other conclusions that may explain this negative observation.

There are no detailed theoretical formulations with which this work may be compared. Each of the excited-atom states can be formed by dissociation via a number of repulsive states. It follows therefore that the problem is quite complicated. The various repulsive molecular curves which terminate with a free H(3*s*), H(3*p*), or H(3*d*) atom do have almost the same energy. It follows that there are no differences in energy defect for the possible dissociation channels, and it is, therefore, not surprising that the behavior for the formation of the 3*s*, 3*p*, and 3*d* states is almost the same. There is a slight preference for population of the 3*d* state; this might be related to the greater statistical weight for this state.

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†Temporary address: FOM Institute for Atomic and Molecular Physics, Kruislaan 407, Amsterdam, The Netherlands.

[1]J. C. Ford and E. W. Thomas, first preceding paper, Phys. Rev. A 5, xxx (1972).

[2]R. H. Hughes, D. B. Kay, C. A. Stigers, and E. D. Stokes, Phys. Rev. 167, 26 (1968).

[3]J. F. Williams and D. N. F. Dunbar, Phys. Rev. 149, 62 (1966).

[4]G. W. McClure, Phys. Rev. 130, 1862 (1963).

[5]C. F. Barnett and J. A. Ray, *Atomic Collision Processes* (North-Holland, Amsterdam, 1964), p. 743.