Double ionization of H_2 by fast H^+ and D^+ projectiles

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The cross sections for the double ionization of H_2 by 0.08—1.0-MeV/amu D^+ and 0.40—3.5-MeV/amu H^+ are reported for the H_2 internuclear axis oriented at 90° and 30° relative to the ion-beam direction. There is no apparent difference between the cross sections at the 90° orientation and at the 30° orientation. Bethe theory does not give qualitative agreement with the experimental results.

I. INTRODUCTION

The double ionization of helium by fast projectiles has been studied by several experimental groups[1–5] as a test of collision theories and of models of double ionization. Helium is a simple two-electron system and is a beginning point for the study of electron-correlation effects in double ionization. The H_2 molecule is another simple two-electron system, but has obvious structural differences. Few detailed studies have been reported on it.

In the shakeoff model of double ionization, the incident projectile removes one electron from the target and leaves the other electron in a continuum state. In the two-step model, the incident projectile interacts directly with one electron and then the other producing double ionization. This latter process requires a second Born term in the Born approximation. The first Born approximation has been applied to the problem of double ionization of helium by Byron and Joachain[6,7] and by Oldham[8]. Byron and Joachain investigated the importance of electron correlation in the initial-state wave function for the double-ionization process. More recently, Haugen *et al.*[5] investigated the multiple ionization of noble gases and discussed the limitations of the first Born approximation. In all of these cases the first Born approximation has not been successful in describing the experimental results.

Prior to these studies Mittleman[9] applied the sudden approximation to the problem; again, theory did not match experiment. Manson *et al.* [10] have suggested that the sudden approximation may not work even at very high collision energies. Kaminsky and Popova[11] have assumed a double-collision model for the ionization process and have done an impact-parameter calculation of the cross section for proton collisions. These results, displayed in a log-log plot, are in qualitative agreement with the data.

For the helium target the ratio of the cross section for double ionization to the cross section for single ionization has become a standard for comparing experiment and theory. A particularly interesting experimental result is that this ratio for electron impact[3,4] on helium is approximately twice that for proton impact.[1,2] There is an indication that these ratios may be converging to a common value at very high projectile velocities[5] ($v = 50$ a.u.). McGuire[12] has presented the argument that the difference of these ratios is due to the interference of the amplitude for the shakeoff process and the amplitude for the direct double-collision, or two-step, process. More recently, Reading[13] has suggested that the difference is due to the mass difference of the projectiles.

We have developed a technique that measures the dissociative states of doubly ionized molecules[14] and applied it to the double ionization of H_2. The molecule H_2 is a two-electron system as is helium and, therefore, electron correlation should play a similar role in the double-ionization process. However, H_2 does not have the spherical symmetry of He and an additional factor is introduced: the double ionization of H_2 as a function of the orientation of the internuclear axis.

Also, H_2 has only one strongly bound singly ionized state, the $1s\sigma_g$ state. The others are either repulsive or have only very shallow potential wells, which means that much of the $H_2{}^+$ formed dissociates. Therefore, the ratio of double- to single-ionization cross sections cannot be measured in the same way as in the helium case. In the present work the ratio of the double-ionization cross section to the excitation of the $H_2{}^+2p\sigma_u$ cross section is estimated. The excitation of the $2p\sigma_u$ state is a two-electron process, also. One electron is removed and the other is excited to a higher state.

The cross sections for double ionization of H_2 by 0.08—1.0-MeV/amu D^+ and 0.40—3.5-MeV/amu H^+ are measured for the H_2 internuclear axis oriented at approximately 90° and 30° relative to the ion-beam direction. Both D^+ and H^+ ion beams were used in order to extend the range of collision velocities. Shah and Gilbody[15] have measured the dissociative ionization of H_2 by fast projectiles in this energy regime, but they did not separate out the double-ionization process.

II. EXPERIMENTAL PROCEDURE

The doubly ionized state of H_2 was detected by observing in coincidence the two protons from $H_2{}^{2+}$ produced at an energy of about 9.7 eV. Figure 1 illustrates the collision region. A pulsed beam of projectiles passes through

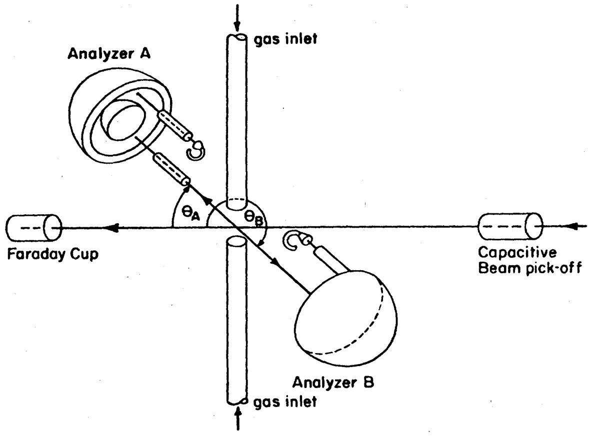

FIG. 1. Schematic of the experimental apparatus. The pulsed ion beam passes through a capacitative-beam pickoff unit and the target gas, and is then collected in a Faraday cup. Both analyzers lie in the horizontal plane.

a differently pumped H_2 target region and is collected in a Faraday cup. The H^+ fragment ions resulting from dissociation of the target molecules are focused onto the entrance slit of a hemispherical electrostatic analyzer set to transmit ions of 9.7 eV. By pulsing the incident ion beam it is possible to measure the time of flight (TOF) of the 9.7-eV H^+ fragment ions from the collision region to the detector. This TOF measurement separates the H^+ fragments from equal energy fragments of background gases such as N^+ and 0^+. Two hemispherical analyzers are placed 180° relative to each other on opposite sides of the incident ion beam. Each analyzer is free to rotate independently about a vertical axis through the target region.

The beam pulses have a time width of about 100 ns and are spaced at intervals of 8 μs. The target gas pressure in the interaction region is estimated to be 2 mTorr. The effective solid angle subtended by the analyzer is 5×10^{-3} sr. In the horizontal plane the half-angle of acceptance of the analyzer system is 1.2°. Prior to entering the analyzer the ions to be detected are accelerated by an electrostatic lens system to seven times their initial kinetic energy. This yields a transmission function for the analyzers that is triangular in shape with an energy width (full width at half maximum) of about 7% of the initial kinetic energy. Upon leaving the analyzer, the ions are accelerated to a high velocity by a 2800-V potential applied to the front of the channel-electron multipliers. These detectors have been in use for many years and their efficiencies are estimated to be 50%.[16]

Pulses produced by detectors A and B are amplified and sent to separate time-to-amplitude (TAC) converters as *start* pulses. A signal produced by a beam pulse is used as a *stop* pulse for both TACs. The output of the TACs are processed by single-channel analyzers to select the H^+ ion signals. The output of the two single-channel analyzers are sent to a coincidence gate that yields a signal upon occurrence of both true coincidence events and accidental coincidence events. The two inputs to the coincidence gate are also sent to a second coincidence gate, except in this case the A detector signal is delayed by 8 μs. This means that the two inputs to the second coincidence gate could not have occurred as the result of the same projectile beam burst. Thus, only accidental coincidence events are recorded by the second gate. The difference between the counts measured by the two coincidence gates is the yield of the true events.

The total number of H^+ counts for each of the A and B detectors were recorded along with the true-plus-accidental and accidental events. The incident ion beam is integrated and a signal is generated to stop the scalars once a preset amount of charge is collected.

In the center-of-mass frame of the H_2 molecule the two H^+ dissociation products are constrained to travel along the internuclear axis with equal energies. However, in the laboratory frame a spread in energy and angle are introduced because of thermal motion. A computer program was developed[17] to simulate the dissociation process of molecules whose centers of mass are moving because of thermal energy. The program predicts the fraction of fragment ions detected in detector B given that its partner was detected by detector A. Weighting factors are introduced for the angular acceptances and the energy

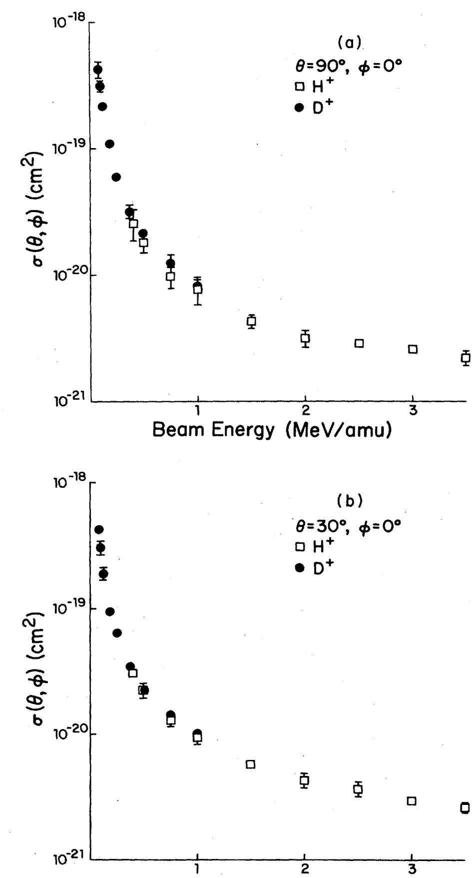

FIG. 2. (a) and (b) The estimated cross sections for the double ionization of H_2 by H^+ and D^+ projectiles. The internuclear axis of the H_2 molecule lies in the horizontal plane ($\phi=0°$) and is oriented at 90° and 30°, respectively, to the beam axis.

transmission functions of both analyzers.

Momentum transferred to the target molecule by the incident particle also results in a shift in energy and angle of the fragment ions. This effect has been reported for He^+ on N_2 collisions.[17] For the systems studied here the effects were measured and found to be negligible.

III. RESULTS AND DISCUSSION

The estimated cross sections for the double ionization of H_2 by H^+ and D^+ projectiles are shown in Figs. 2(a) and 2(b). In Figs. 2(a) and 2(b) the molecular internuclear axis lies in a horizontal plane ($\phi=0$) and is oriented at $\theta=90°$ and 30°, respectively, relative to the beam direction. The absolute cross sections are estimated to within a factor of 4. The uncertainty in the measurements is due to estimates of the target-gas density and the detector efficiencies. The error bars are the standard deviations of several measurements made at each beam velocity. In the region of beam velocities where the D^+ and H^+ projectile velocities overlap, the double-ionization cross section produced by D^+ is slightly higher than that produced by H^+. This is probably due to the presence of an ${H_2}^+$ component in the ion beam. As can be seen in Figs. 2(a) and 2(b) there is no apparent difference between the cross sections at the 90° orientation and at the 30° orientation.

In order to compare the experimental results to the Bethe theory, the data were plotted as the product of cross section times projectile energy versus the logarithm of projectile energy (Bethe-Born plot or Fano plot). According to the Bethe theory this should yield a straight line of positive slope. Figures 3(a) and 3(b) show the results of such a plot where the quantity T/R is the projectile velocity squared in atomic units.[18] The cross section has also been expressed in atomic units (a_0^2) for this graph. Not until the projectile velocity is greater than about 10 a.u. does the slope change from negative to positive. Schram *et al.*[3] also observed a negative slope for the Bethe-Born plot of the double ionization of helium by electrons.

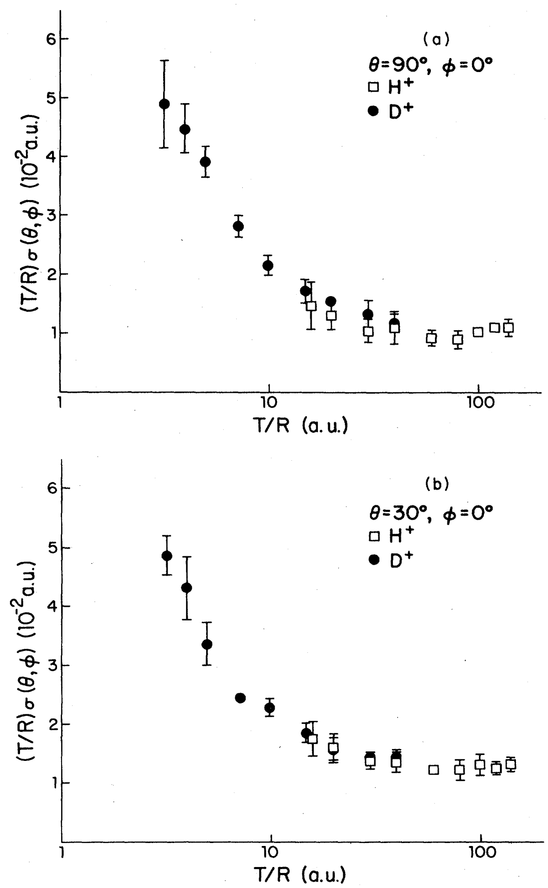

FIG. 3. (a) and (b) Bethe-Born plots of the double ionization of H_2 by H^+ and D^+ projectiles. T/R is the projectile velocity squared in atomic units. The internuclear axis of the H_2 molecule lies in the horizontal plane ($\phi=0°$) and is oriented at 90° and 30°, respectively, to the beam axis.

Not all counts recorded at 9.7 eV are due to H^+ fragment ions from the doubly ionized state. The $2p\sigma_u$ state of ${H_2}^+$ also dissociates and produces H^+ fragments at this energy.[19] In fact, compared to the doubly ionized state it produces a relatively large number of fragments.

The ratio of the cross section for double ionization to the cross section for excitation of the ${H_2}^+2p\sigma_u$ state can be estimated from the number of coincidence counts relative to the sum of the counts in detectors *A* and *B*. To make the estimate, several factors need to be considered: kinematic effects due to the thermal motion of the target molecules, the expected relative contributions from the two states at 9.7 eV as predicted by the reflection approximation,[19] factors involving detector efficiency and calculation of solid angle. Based on our measurements and consideration of these factors we conclude that the ratio of yields is approximately the same at all beam velocities studied and is about $(5\pm2)\%$. This result is in agreement with work published earlier[20] for 1 MeV H^+ on H_2 collisions.

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