

## Effect of the projectile charge on the ionization and excitation of hydrogen molecules by fast ion impact

E. Wells, I. Ben-Itzhak,\* K. D. Carnes, and Vidhya Krishnamurthi†

*J.R. Macdonald Laboratory, Department of Physics, Cardwell Hall, Kansas State University, Manhattan, Kansas 66506*

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The ratio of double ionization to single ionization (DI to SI) as well as the ratio of ionization excitation to single ionization (IE to SI) in hydrogen molecules were studied by examining the effect of the projectile charge  $q$  on these processes. The DI to SI and IE to SI ratios were measured at a fixed velocity (corresponding to 1 MeV/amu) from  $q=1$  to 20. For a highly charged  $\text{Cu}^{20+}$  projectile, for example, the DI to SI and IE to SI ratios are 13% and 46%, respectively, which is a large increase from the ratios of 0.18% and 1.95%, respectively, for  $\text{H}^+$  projectiles. The DI to SI ratio initially increases much more rapidly with projectile charge than the IE to SI ratio. The measured rate of increase of both these ratios decreases for highly charged projectiles. These trends are in agreement with a simple model calculation based on the independent electron approximation. [S1050-2947(99)07011-0]

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### I. INTRODUCTION

Helium provides the most basic system to study two-electron processes in ion-atom collisions and has been the subject of much research. In particular, the ratio of double ionization to single ionization (DI to SI) of helium by both photons and charged particles has been of great experimental and theoretical interest in recent years [1–4]. The constant ratio reached at the high velocity limit (corresponding to low values of the ratio of projectile charge to velocity,  $q/v_p$ ) indicates that double ionization is caused predominantly by electron-electron interactions initiated by the ionization of one electron by the incoming projectile. At lower impact velocities, double ionization is dominated by the direct interactions between the projectile and both target electrons. McGuire has pointed out that, for intermediate values of  $q/v_p$ , interference between the two mechanisms can occur [5].

Another two-electron process is ionization excitation (IE). In this process, one electron is ionized from the target and the second is left in an excited state. The behavior of the IE process is expected to be similar to the DI process, since at small  $q/v_p$ , electron-electron interactions started by the ionization of one electron are expected to dominate, and at larger values of  $q/v_p$ , the direct interaction of the projectile with both target electrons dominates. Determining the IE yield experimentally for a helium target is difficult, however, since one must measure the emitted photons. In contrast, the hydrogen molecule offers a similar two-electron target in which IE is immediately followed by dissociation into  $\text{H}^+ + \text{H}(nl)$ . This is due to the fact that all of the potential-energy curves of the excited electronic states of  $\text{H}_2^+$  are dissociative in the Franck-Condon region. Therefore,  $\text{H}_2$  is a

two-electron target where the IE process can be identified from the measurement of the  $\text{H}^+ + \text{H}(nl)$  channel, rather than making more difficult measurements of photons. Single ionization can lead to  $\text{H}^+ + \text{H}(1s)$  in a small percentage (1.45%) of cases and must be separated from the IE channel [6].

Most of the previous studies of the DI to SI ratio for the hydrogen molecule have concentrated on the dependence on the sign and mass of singly charged projectiles. For example, Edwards *et al.* [7] measured the  $\text{H}^+ + \text{H}^+$  yield produced by intermediate velocity electron and proton impact, using a back-to-back detection system. The  $\text{H}_2^+$  yield, however, had to be measured using a different apparatus, leading to some uncertainty in the absolute magnitude of their data, as they have discussed in a later publication [8]. Bapat *et al.* [9] have measured the ratio of two-electron processes to one-electron processes in collisions between protons of 16–25 a.u. velocity and  $\text{H}_2$  and  $\text{D}_2$  targets. Kossmann *et al.* [10] have reported measurements of double- and single-ionization cross sections of  $\text{H}_2$  due to fast electron impact. Ben-Itzhak *et al.* have measured the velocity dependence of the DI to SI [11] and IE to SI [6] ratios for fast proton impact.

There have been, however, comparatively few experiments investigating the double ionization and ionization excitation of a hydrogen molecule as a function of projectile charge. Shah and Gilbody [12,13] have measured the ratio of dissociative to nondissociative ionization of  $\text{H}_2$  by  $\text{H}^+$ ,  $\text{He}^{2+}$ , and  $\text{Li}^{3+}$  projectiles. Krishnakumar *et al.* [14] reported results for IE to SI for fast (3.3–7.0 MeV/amu)  $\text{C}^{6+}$ ,  $\text{O}^{8+}$ , and  $\text{S}^{16+}$  projectiles where the measured values were corrected for dissociative ionization of the  $\text{H}_2^+$  electronic ground state, and contributions from double ionization were considered negligible. Cheng *et al.* [15] measured cross sections for single ionization, double ionization, and ionization excitation of  $\text{D}_2$  by 0.5–1.25 MeV/amu  $\text{O}^{8+}$  projectiles. In this study we utilize the coincidence-time-of-flight technique described in Sec. II to directly measure the IE to SI and DI to SI ratios for projectiles ranging in charge from  $q=1$ –20 at a fixed velocity corresponding to 1 MeV/amu. In Sec. III our

\*Author to whom correspondence should be addressed. Electronic address: ibi@phys.ksu.edu

†Present address: Etec Systems Inc., 26460 Corporate Ave., Hayward, CA 94545.

results are compared to the previous measurements discussed above. In addition, since there is a lack of theoretical treatment for the hydrogen molecule compared to the helium atom, due to the complications associated with a two-center potential, we offer a simple model calculation for the DI to SI and IE to SI ratios in Sec. III. Two new points are emphasized in this paper. First, all of the charged products from each collision are measured, allowing the determination of the DI to SI and IE to SI ratios directly without assumptions about the relative yields of specific channels. Second, our model calculations based on the independent-electron approximation seem to fit the general trends of the data if both the direct and electron-electron contributions are included for the two-electron processes.

## II. EXPERIMENTAL METHOD

The major advantage of the coincidence-time-of-flight experimental technique is the identification of all recoil ion(s) produced in each collision. Many of the details of our apparatus are discussed in Refs. [16,17], and we will only discuss the main points here. A bunched beam of ( $H^+$ ,  $Li^{(2-3)+}$ ,  $C^{3+,5+,6+}$ ,  $F^{(4-9)+}$ ,  $Cu^{14+,20+}$ ) projectiles was accelerated to 1 MeV/amu by the J. R. Macdonald Laboratory EN Tandem Van de Graaff accelerator, collimated, and directed into a target cell containing a thin hydrogen-gas target. Recoil ions produced in the target cell were extracted and accelerated toward a Z-stack microchannel plate detector by a strong uniform electric field (typically 1200 V/cm) in a time-of-flight spectrometer [16]. The time of flight of the different ions was recorded by a multistop system relative to a common start signal synchronized with the beam bunch.

The  $Cu^{q+}$  beams, however, could not be bunched properly due to the mass of the projectile. In this case, a dc beam was used, and a signal from a scintillator, serving as a projectile detector, was used as the common stop. For the dc beam case, a simulation was used to correct for random stops—projectiles that preceded the true projectile by less than the time difference between the recoil ion and the true projectile (see Ref. [18] for further details).

To improve the accuracy of the measurement of the double-ionization channel we used deuterium hydride. For this isotope of the hydrogen molecule, the flight time of the two fragments is significantly different, even when using the strong extraction field needed to collect all fragments. Explicitly, the time of flight of  $D^+$  was about 100 ns longer than that of the  $H^+$  fragment, which is much longer than the time spread caused by the kinetic energy released in the dissociation for the experimental conditions used. The mass difference between the HD and  $H_2$  isotopes is not expected to affect the electronic transitions during the collision significantly, as they are the same within the Born-Oppenheimer approximation. Typical one- and two-dimensional spectra are shown in Fig. 1. In our data analysis, we have assumed that the angular distribution of fragments resulting from the double ionization of hydrogen molecules is isotropic, as was shown, for example, by Edwards *et al.* [19] and Yousif *et al.* [20]. However, if this process is not isotropic, it will have only a minor effect on our results because we have angular discrimination effects on less than 10% of the target length. Thus, the contribution from most of the target length is in-

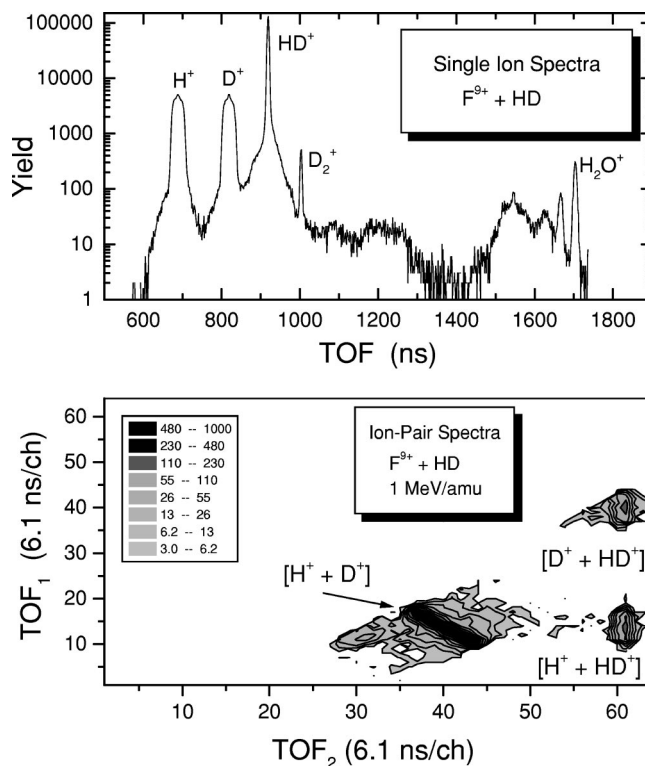


FIG. 1. Representative raw coincidence-time-of-flight data for 1 MeV/u  $F^{9+}$  + HD. Top: Singles spectra. Bottom: Ion-pairs spectra. The vertical axis is the time of flight of the first fragment; the horizontal axis is the time of flight of the second fragment.

dependent of the angular distribution of the fragments.

The ionization excitation of hydrogen molecules is determined from the contribution of single  $H^+$  and  $D^+$  fragments. All the excited states of  $HD^+$  are dissociative within the Franck-Condon region, and the fragments typically have a few eV ( $>2$  eV) of kinetic energy. Thus, the dissociation of the excited states can be separated from the very slow ( $<0.5$  eV)  $H^+$  and  $D^+$  fragments resulting from the dissociation of the electronic ground state of  $HD^+$ , which takes place if the vertical transition populates the vibrational continuum. The contribution of the ground-state dissociation channel was measured using a weak extraction field as previously described in detail by Ben-Itzhak *et al.* [6]. For high projectile charge, however, it became difficult to evaluate the ground-state dissociation contribution due to the increased amount of fast fragments from the IE channel. In these cases, the value of ground-state dissociation by fast proton impact was used (assuming that ground-state dissociation is due to the overlap between the initial and final vibrational wave functions, and, thus, is independent of  $q$ ). To determine the yield of the IE channel, the contribution of slow  $H^+$  and  $D^+$  fragments was subtracted from the yield of the  $H^+$  and  $D^+$  fragments measured with a strong extraction field.

The DI to SI ratio is evaluated by comparing the number of  $[H^+ + D^+]$  ion-pair events ( $H^+$  in coincidence with  $D^+$ ) to the number of  $HD^+$  recoil ions. The ratio of IE to SI is evaluated by comparing the sum of  $H^+$  and  $D^+$  single events to the number of  $HD^+$ . Three corrections must be made to the raw data in addition to dividing by the proper detection efficiency of each channel: (i) Random ion pairs are subtracted from the DI yield. These are events in which both a

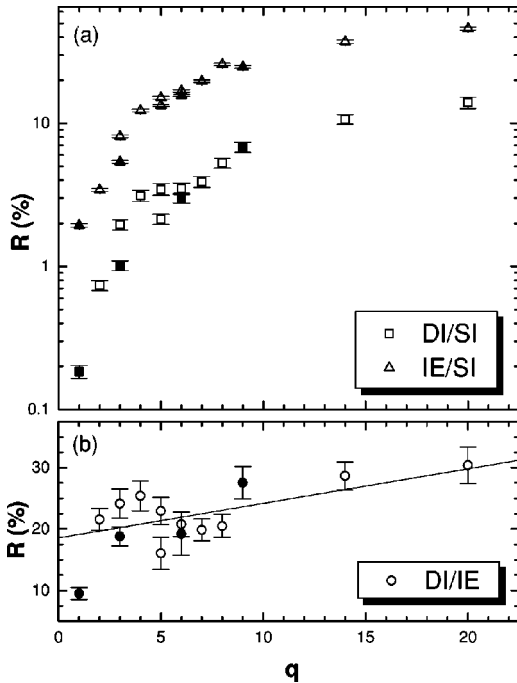


FIG. 2. (a) The ratios  $R$  of DI to SI ( $\square$ ) and IE to SI ( $\triangle$ ) measured in this experiment. (b) The ratio of DI to IE ( $\circ$ ) measured in this experiment. The solid line in (b) is a linear fit of the data for  $q \geq 2$ . In both plots the solid symbols represent bare projectiles.

$D^+$  and a  $H^+$  recoil ion are recorded for the same beam bunch, but they do not originate from the same molecule. This channel can be estimated by using the purely random [ $H^+ + HD^+$ ] and [ $D^+ + HD^+$ ] ion-pair channels, which must come from double collisions in the same beam bunch [21]. (ii) “Lost fragments” from ion-pair events must be subtracted from the IE yield. These are double-ionization events in which only one of the two recoil ions was detected, because the detection efficiency is smaller than one [21]. (iii) The yield of slow  $H^+$  and  $D^+$  fragments from ground-state dissociation, as described above, has to be subtracted from the IE channel and added to the SI channel [6].

### III. RESULTS AND DISCUSSION

The measured IE to SI and DI to SI ratios are shown in Fig. 2(a) as a function of projectile charge, for  $q = 1 - 20$ , at a fixed collision velocity corresponding to 1 MeV/amu.

The DI to SI ratio begins to increase rapidly as a function of the projectile charge from an initial value of 0.184% for proton impact, but slows its increase somewhat for  $q > 4$ . For  $Cu^{20+}$ , the value of the DI to SI ratio reaches 13% [see Fig. 2(a)]. The IE to SI ratio shows a similar rapid increase with projectile charge, increasing from 1.95% for proton impact to about 25% for  $F^{9+}$  projectiles. The rate of increase then slows somewhat, reaching a value of 46% for  $Cu^{20+}$ . The ratio of DI to IE also increases slowly with projectile charge, approximately linearly for  $q > 2$  as can be seen in Fig. 2(b). Note the rapid change in the DI to IE ratio between  $q = 1$  and  $q = 2$ .

The influence of projectile electrons can be seen in the data for  $q = 3, 5$ , and 6 where we have measurements with

two different projectiles. The DI to SI and IE to SI ratios were consistently higher for the projectiles with the higher  $Z$ . Even when neither projectile was fully stripped ( $F^{5+}$  and  $C^{5+}$ ), the projectile with higher  $Z$  had higher DI to SI and IE to SI ratios. Cocke and Montenegro [22] have discussed the enhancement of ionization cross sections in collisions between dressed projectiles and light atomic targets. In a collision between a two (or more) electron target and a bare projectile, ionization (or excitation) by an electron-electron interaction is a second-order process. If the projectile is dressed, however, ionization (or excitation) of both collision partners is possible by first-order electron-electron interactions. Some DI and IE processes in the collisions under study, therefore, happen at a small enough impact parameter that the projectile cannot be treated as a point charge. The fact that the DI to SI and IE to SI ratios increase with the number of projectile electrons might be evidence that DI and IE processes are more strongly affected by this “antiscreening” effect than the SI process. We have not conducted a careful study of this matter and only wish to point out that the effect of the projectile electrons can account for some of the scatter in our data, in particular with the fluorine projectiles, which ranged from  $F^{4+}$  to  $F^{9+}$ .

While the contribution of double ionization to the dissociative ionization channel is negligible for proton impact, it is clearly not negligible for more highly charged projectiles at this velocity. Any attempt to deduce the amount of IE from the dissociative ionization channel based on the assumption that DI is a negligible contribution to the dissociative ionization channel [14] would yield results with only a limited accuracy for our measurements, since DI accounts for 20% to 30% of the dissociative ionization for  $q \geq 2$ . Of course, this assumption is not needed in our case, since the DI is determined directly from the [ $H^+ + D^+$ ] ion-pair yield. Furthermore, in previous experiments using  $H_2$  as a target, the contribution of DI to the measured  $H^+$  yield might be even larger, because the detection efficiency of at least one fragment out of two, in those cases, might be close to twice the detection efficiency of one fragment out of one, depending on the efficiency of the detection system used.

The charge dependence for the DI to IE ratio is different for a change of  $q$  from 1 to 2 than it is for changes starting above  $q = 2$ . This could indicate that while at  $q = 1$ , most DI is the result of electron-electron processes, by  $q = 2$ , there is an onset of a significant contribution from direct interactions between the projectile and both target electrons. For IE, however, the change between  $q = 1$  and  $q = 2$  is less severe, indicating that the direct interactions between the projectile and both target electrons are already a factor in IE at  $q = 1$ .

We have employed the independent-electron approximation [23,24] to formulate a model for the DI, IE, and SI processes we have measured for interactions with bare projectiles. The following impact-parameter ( $b$ )-dependent quantities are defined:  $P_I(b)$  is the probability for ionization of the active electron, and  $P_E(b)$  is the probability for excitation of the active electron to all possible final states. The cross sections for the DI, IE, and SI processes can then be written as follows:

$$\sigma_{DI}^d = 2\pi \int_0^\infty P_I^2(b) b db, \quad (1)$$

$$\sigma_{IE}^d = 2\pi \int_0^\infty 2P_I(b)P_E(b)b db, \quad (2)$$

$$\sigma_{SI}^d = 2\pi \int_0^\infty 2P_I(b)[1 - P_E(b) - P_I(b)]b db. \quad (3)$$

Using these three equations all of the measured ratios are determined. We have used the common technique of choosing  $P_I(b)$  and  $P_E(b)$  to be exponential functions (see, for example, Refs. [25–27]), since values of  $P_E(b)$  for hydrogen molecules are not readily available in the literature. For proton impact,  $P_I(b)$  and  $P_E(b)$  are

$$P_I(q=1,b) = P_I(0)e^{-\alpha_I b}, \quad (4)$$

$$P_E(q=1,b) = P_E(0)e^{-\alpha_E b}. \quad (5)$$

The parameters  $P_I(0)$ ,  $P_E(0)$ ,  $\alpha_I$ , and  $\alpha_E$  were determined by fitting to the experimental data. The probabilities for proton impact were then scaled with projectile charge such that for a projectile of charge  $q$  they are

$$P_I(b) \equiv P_I(q,b) = q^2 P_I(q=1,b), \quad (6)$$

$$P_E(b) \equiv P_E(q,b) = q^2 P_E(q=1,b) \quad (7)$$

(see, for example, Hansteen, Johansen, and Kocbach [28]). While this approximately agreed with the observed ratios of DI to SI and IE to SI, it did not fit the DI to IE ratio well at all, particularly the rapid change from  $q=1$  to  $q=2$ , which was not reproduced. This result is not surprising, since the independent electron approximation is inadequate for dealing with collisions in which the electron-electron mechanisms are important, as they are for low  $q/v_p$ . To address this problem, the cross sections for IE and DI had to be written as the sum of an electron-electron part, which dominates at low  $q/v_p$ , and a direct part, which dominates the contribution at high  $q/v_p$ ,

$$\frac{\sigma_{DI}}{\sigma_{SI}} = \frac{\sigma_{DI}^d + \sigma_{DI}^{e-e}}{\sigma_{SI}}, \quad (8)$$

$$\frac{\sigma_{IE}}{\sigma_{SI}} = \frac{\sigma_{IE}^d + \sigma_{IE}^{e-e}}{\sigma_{SI}},$$

where we have defined  $\sigma^{e-e}$  as the part of the cross section that comes from electron-electron interactions (in the target), and  $\sigma^d$  as the part of the cross section that is due to the direct interaction between the projectile and both target electrons. The possibility of interference between the two mechanisms has been omitted for simplicity. The values ( $\sigma_{IE}^{e-e}/\sigma_{SI} = 0.0195$  and  $\sigma_{DI}^{e-e}/\sigma_{SI} = 0.0013$ ) for the electron-electron part were taken from our previous measurements of proton impact at the high velocity limit [6,11].

To constrain the parameter space somewhat when fitting, we have used the following procedure: First, we note from Eq. (3) that if the  $[1 - P_E(b) - P_I(b)]$  term is close to 1, as it is expected to be for 1-MeV proton impact, where  $P_E(b)$  and  $P_I(b) \ll 1$ , we can write Eq. (3) as follows:

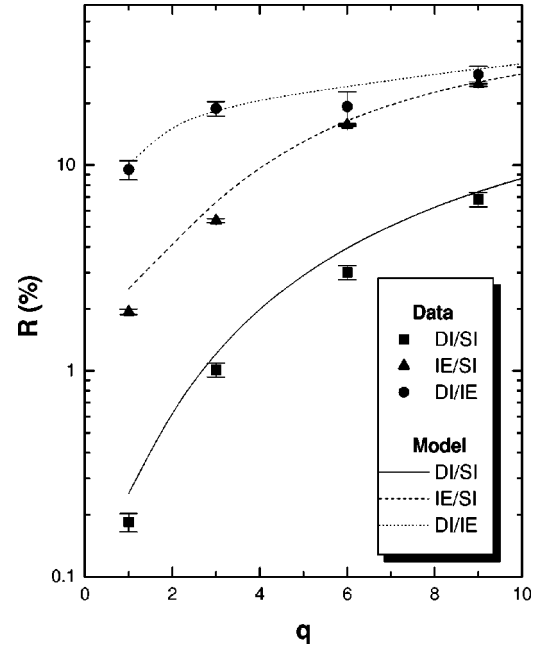


FIG. 3. Comparison between experimental data for the DI to SI ( $\square$ ), IE to SI ( $\triangle$ ), and DI to IE ( $\circ$ ) ratios, from impact on hydrogen molecules by bare projectiles ( $H^+$ ,  $Li^{3+}$ ,  $C^{6+}$ , and  $F^{9+}$ ) and our model calculations.

$$\sigma_{SI} = 2\pi \int_0^\infty 2P_I(b)b db = \frac{4\pi P_I(0)}{\alpha_I^2} = 1.45 \text{ a.u.}^2 \quad (9)$$

The value of  $\sigma_{SI}$  was taken from Shah and Gilbody's measurement of the nondissociative ionization cross section of  $H_2$  [12]. (There is another consistent measurement by Edwards *et al.* [29].) To convert their measurement from the cross section for nondissociative ionization to the cross section for single ionization, we must divide by 0.985 in order to account for the dissociative single-ionization channel [6]. As an additional constraint, we consider the DI to SI ratio,

$$\frac{\sigma_{DI}^d}{\sigma_{SI}} = \frac{\sigma_{DI}}{\sigma_{SI}} - \frac{\sigma_{DI}^{e-e}}{\sigma_{SI}}, \quad (10)$$

where  $\sigma_{DI}/\sigma_{SI}$  is the measured ratio for 1-MeV proton impact. Therefore, substituting Eq. (4) into Eqs. (3) and (1) and again using the fact that  $[1 - P_E(b) - P_I(b)] \approx 1$ , we find that

$$\frac{P_I(0)}{8} = 0.00184 - 0.0013. \quad (11)$$

Solving Eqs. (9) and (11) yields  $P_I(0) = 4.32 \times 10^{-3}$  a.u. and  $\alpha_I = 0.1935$ . Using these values as a starting point for our fit, we scanned the parameter space looking for the best least-squares fit. With the correction to the independent electron model given in Eq. (8), we were able to fit the experimental data quite well, using the exponential form given in Eqs. (4) and (5) for the direct part of the interaction. The best values for the parameters are  $P_I(0) = 0.0078$ ,  $P_E(0) = 0.0476$ ,  $\alpha_I = 0.205$ , and  $\alpha_E = 0.378$ . The results of our fit can be seen in Fig. 3. Varying the parameters by up to 5% does not affect the quality of the fit significantly.



There is very little data available that clearly distinguishes the SI, IE, and DI channels. The ratio of dissociative to non-dissociative ionization, however, has been measured by other groups [12–15]. For our purposes, the more interesting ratio is the ratio ( $R^*$ ) of two-electron to one-electron processes,  $R^* = (\sigma_{IE} + \sigma_{DI}) / \sigma_{SI}$ . This ratio differs from the ratio of  $H^+ / H_2^+$  only because of the dissociative single-ionization channel. We have used our previous results [6] to convert data from some previous measurements [12,13,15] of the ratio of  $H^+ / H_2^+$  to  $R^*$ . A scaling law suggested by Knudsen *et al.* [30] for the DI to SI ratio in helium has been adapted to a hydrogen molecule by Krishnakumar *et al.* [14]. This scaling law has the form

$$R^* = A + Bq^2 / v_p^2 \ln(9.18v_p) \quad (12)$$

where  $q$  is the projectile charge, and  $v_p$  is the projectile velocity in atomic units. Krishnakumar *et al.* [14] found the constants, by fitting the available data, to be  $A = 0.0277$  and  $B = 0.407$ . We have compared our data for bare projectiles to that of the groups cited above in Fig. 4 and generally found them to be in good agreement with previous results and the scaling law. In order to compare our results to the other measurements of  $R^*$ , we have added our separate measurements in the following manner:

$$R^* = \frac{\sigma_{IE}}{\sigma_{SI}} + \gamma \left( \frac{\sigma_{DI}}{\sigma_{SI}} \right), \quad (13)$$

where  $\gamma$  is the ratio of the probability of detecting at least one out of two recoil ions to the probability of detecting one recoil ion. For our experimental setup, this ratio was found to be 1.625. As a general trend, the measurements made using bare projectiles agree more closely with the previous measurements, which were all made with bare projectiles. For our measurements of nonbare projectiles, the projectile electrons play a role, and one would need to calculate an effective charge for each of the projectiles in order to compare it with the data presented in Fig. 4.

#### IV. SUMMARY

In summary, we have measured the IE to SI and DI to SI ratios in hydrogen molecules as a function of projectile

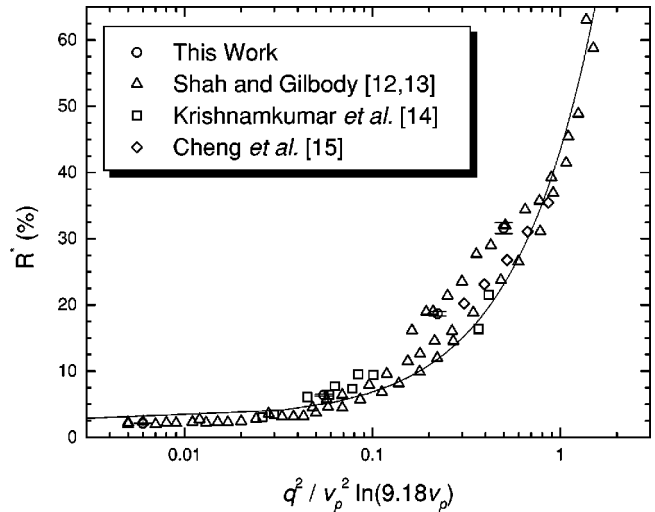


FIG. 4. The ratio  $R^*$  of two-electron to one-electron processes as a function of  $v_p^2 \ln(9.18v_p)$  for impact on hydrogen molecules by bare projectiles. Shown is our data ( $\circ$ ), Ref. [14] ( $\square$ ), Ref. [15] ( $\diamond$ ), and Refs. [12] and [13] ( $\triangle$ ). The curve is from the scaling law of Krishnakumar *et al.* [14] given in Eq. (12) with  $A = 0.0277$  and  $B = 0.407$ .

charge, from  $q = 1$  to 20, at a fixed velocity (1 MeV/amu) using the coincidence time-of-flight technique. The contribution from dissociation of the electronic ground state of molecular hydrogen was accounted for, and the IE to SI and DI to SI ratios were measured simultaneously. The contribution of double ionization to dissociative ionization is negligible for proton impact, but as the projectile charge increases, it rapidly becomes significant. For a highly charged  $Cu^{20+}$  projectile, the DI to IE ratio is about 30%. Both the DI to SI and IE to SI ratios increase with projectile charge, but the rate of increase with  $q$  decreases for higher projectile charge. The model calculation we have performed using the independent electron approximation compares reasonably well to the experimental data if the electron-electron contribution to the two-electron processes is included in the model.

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