Single ionization of hydrogen molecules by fast protons as a function of the molecular alignment

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Relative cross sections for the 4 MeV $H^+ + D_2$ $(^1\Sigma_g^+) \rightarrow H^+ + D_2^+(1s\sigma) + e^-$ ionization process were measured as a function of the molecular alignment during the interaction. The angle between the molecular axis and the projectile was obtained by using a momentum imagining technique and isolating the events in which the $D_2^+(1s\sigma)$ ions are excited to the vibrational continuum and subsequently dissociate. While anisotropic cross sections have been observed in the past for a number of collision processes involving both target electrons, the one electron process investigated here is isotropic within our experimental uncertainties.

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I. INTRODUCTION

Hydrogen molecules dissociate if both electrons are excited, if both electrons are ionized, or if one electron is excited and the other ionized. Specifically, the only non-dissociative state of H_2^+ is the $1s\sigma$ electronic ground state. This tendency to dissociate is useful for measurements involving molecular alignment. For most electron- and ion-impact collisions with hydrogen, the time scale of the induced electronic activity is much shorter than the rotation time of the molecule, and the axial recoil approximation is valid [1,2]. Under this approximation, the alignment of the molecule at the time of the interaction may be deduced from the alignment of the fragments, which dissociate back-to-back along the line of the internuclear axis. Researchers have exploited this fact using a variety of experimental techniques to study the alignment dependence of collision processes between hydrogen molecules and electrons [3–6], heavy ions [4,7–12], and photons [13–19], to cite a small sample of previous experiments.

For ion impact of hydrogen molecules, a number of possible mechanisms can result in a differential cross section that is anisotropic. Dunn proposed general rules for transition probabilities between pairs of electronic states aligned parallel or perpendicular to each other [20]. These rules depend upon the symmetry of the initial and final states about the axis of the momentum transfer. For multiple ionization of larger diatomic molecules, a statistical energy deposition model [21,22] has been suggested, which predicts an anisotropic distribution, favoring molecules aligned parallel to the ion beam direction. This model, however, is unlikely to be relevant for hydrogen, where a maximum of two electrons may be removed.

In recent years, a number of experimental studies [8–10,12,23–26] have examined effects resulting from the two-center nature of the hydrogen molecule, analogous to Young's double slit experiment. These interference effects may manifest themselves either in the electron emission from the dissociating molecule or the angular cross section of the fragments. Besides the fundamental interest in this example of the wave-like behavior of particles [27,28], when induced by ultrafast laser pulses, these interference effects might be useful as a time-resolved probe of molecular dynamics [29–32].

The investigations of two-center interference effects mentioned above either focus on two-electron processes (e.g., double ionization, ionization-excitation) that result in molecular dissociation, or, as in the case of electron spectroscopy, do not distinguish between dissociative and non-dissociative final states. For projectile velocities where ionization is prevalent, these dissociative final states are only reached a small fraction of the time [33,34]. The most likely final state is non-dissociative single ionization, in which only one electron is disturbed by the incident projectile. Compared to collisions that involve both target electrons, these single ionization collisions are likely to have larger impact parameters, resulting in correspondingly lower energy electron emission [12]. These low energy electrons are not well suited for traditional electron spectroscopy [35]. Furthermore, if the molecular ion does not dissociate, straightforward measurements of alignment effects using the axial recoil approximation are not generally possible. Thus, while there is now a wealth of data for angular effects in collision processes involving both electrons of the target hydrogen molecule, angular effects in the main single ionization process are relatively unexplored.

We circumvent these experimental difficulties by exploiting the ground state dissociation (GSD) channel, illustrated in Fig. 1. In this process, a fast ion induces a Frank-Condon transition to the vibrational continuum of the H_2^+ (or D_2^+) electronic ground state. The H_2^+ subsequently dissociates into a H^+ and a $H(1s)$. The kinetic energy of the GSD fragments is typically less than 300 meV, (see the inset of Fig. 1) well separated from the higher energy fragments that occur as the result of two-electron processes. This energy gap allows the low energy GSD fragments to be isolated experimentally [36]. The result is an initial- and final-state specific measurement, isolating a pure one-electron process.

In this article we report the relative probability for single ionization of hydrogen molecules by fast protons as a func-

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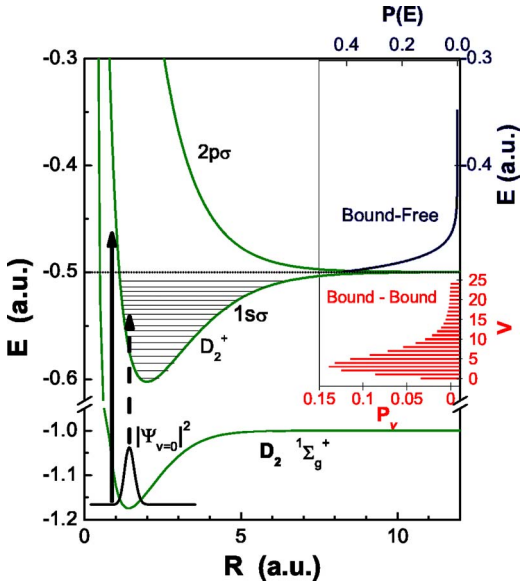

FIG. 1. (Color online). Potential energy curves of D_2 and D_2^+, illustrating the ground state dissociation process. Ionization of one electron results in a vertical transition that predominantly populates a bound vibrational state of D_2^+ (dashed arrow). The vibrational distribution is shown in the lower inset at the right. A small fraction of the time, however, the vertical transition may reach the vibrational continuum (solid arrow) and dissociate. The kinetic energy release (KER) distribution of these fragments is shown in the upper inset of the right.

tion of the angle between the incident protons and the internuclear axis. Experimentally, we measure the three-dimensional momentum vector of the dissociating GSD fragments. Since the breakup of the molecule is back-to-back, the angle with respect to the projectile can be reconstructed from the fragment momentum. Our results are compared to theoretical models based upon the symmetry of the initial and final states [20] and two-center interference effects [10].

II. EXPERIMENT

The main challenge in this experiment is to measure the momentum of a low energy fragment with high enough resolution to (a) isolate the low-energy GSD fragments, and (b) have meaningful angular resolution. Cold target recoil ion momentum spectroscopy (COLTRIMS) [37,38] is an ideal method for measuring the momentum of low energy charged particles resulting from collision processes.

In our experiment (illustrated in Fig. 2), a 4 MeV beam of protons, bunched into 1–2 ns pulses for timing purposes, was accelerated by the tandem Van de Graaff in the J. R. Macdonald Laboratory. Fast protons were used because their interaction with the target molecule is weak, producing relatively few ionization-excitation or double ionization events [34], which form part of the background for the present experiment. The beam was collimated and directed to a cold ($\approx 30 \pm 10$ K, determined by fitting a Maxwell-Boltzmann distribution to the D_2^+ KER distribution), localized target of

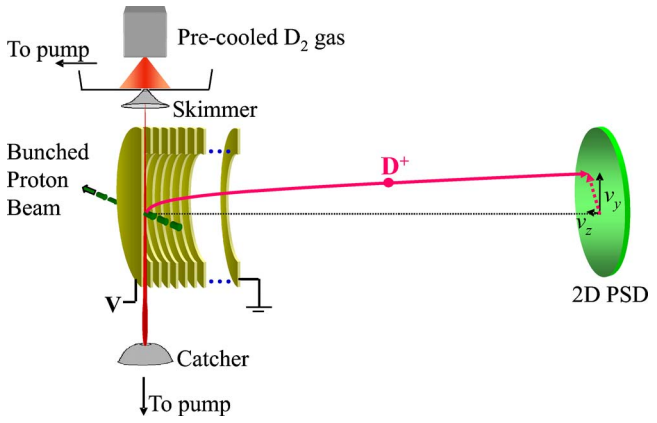

FIG. 2. (Color online) A schematic view of the experimental setup. The ion beam propagates in the z direction, the jet in the y direction, and the detector is in the x direction for our lab-frame coordinate system. The target gas is pre-cooled in a small cell mounted to a cryo-head and collimated by a single skimmer. A grounded electroform mesh (not shown) in front of the MCP detector keeps the drift region nearly field-free. The relatively long spectrometer allows the very low energy GSD fragments to spread over much of the detector surface.

D_2 molecules. The D_2 is introduced to the chamber (typical base pressure of 10^{-7} Torr) using a single-stage supersonic jet. The gas is collimated by a skimmer to about 1 mm diameter and is collected after the interaction region in a differentially pumped catcher region. D_2 is used instead of H_2 since H^+ from residual water in the experiment chamber would contaminate the channel of interest. However, ionization of water can induce bond rearrangement, producing H_2^+ [39], which has the same mass to charge ratio as D^+. The effect is small but not insignificant. Cold traps filled with liquid nitrogen are added to the vacuum chamber to further reduce residual water vapor. The remaining residual gas contributions, mainly the H_2^+, are removed by subtracting a properly normalized background data set obtained with the jet off.

As shown in Fig. 2, the recoil ions are directed toward a position sensitive detector (PSD) by a weak electric field ($\sim$40 V/cm). The ions are position and time focused by an electrostatic lens incorporated into the spectrometer making the size of the jet-ion beam interaction region insignificant. The low energy ions ($<$1 eV) are collected with 4π (solid angle) efficiency. The ions are detected by a micro-channel plate (MCP) detector in a z-stack configuration located 860 mm from the target. Timing signals are taken from the back plate of the MCP. A two-dimensional resistive anode PSD was employed for decoding the position information. From the time and position data of each ion the complete three-dimensional momentum vector of the dissociating ion is obtained on an event-by-event basis. The angle at the time of dissociation is then reconstructed from the measured momentum vectors of each ion,

$$\cos(\theta) = \frac{P_z}{|\mathbf{P}|}, \tag{1}$$

where z is the direction of the ion beam, x is directed toward the detector, y is in the direction of the jet flow, and θ is the

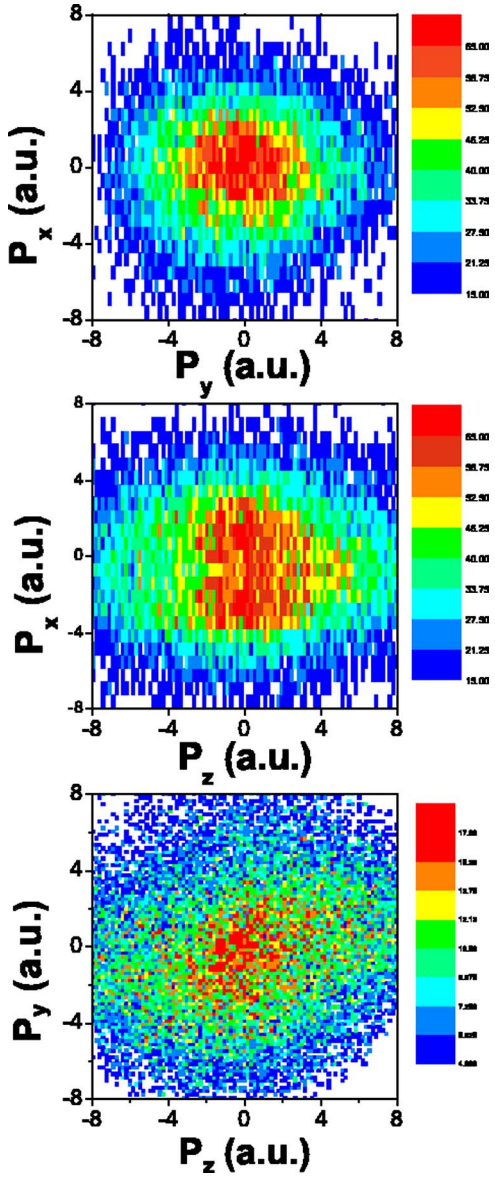

FIG. 3. (Color online) Fragment momentum distributions in the laboratory frame. The ion beam is propagating in the z direction. Slices of the momentum distributions are presented along three planes: (top) P_x vs P_y, corresponding to the extraction field vs gas jet directions, (middle) P_x vs P_z, corresponding to the extraction field vs ion beam directions, and (bottom) P_y vs P_z, corresponding to the gas jet vs ion-beam directions. In each distribution, the momentum vector in the third dimension is restricted to $|\mathbf{P}_i| \leqslant 1.5$ a.u., i.e. the P_x vs P_y plot shown is restricted to -1.5 a.u. $\leqslant P_z \leqslant 1.5$ a.u.

angle between the molecular axis and the projectile beam. This calculation assumes an unperturbed two-body breakup, which is valid for our collision system. The other angle in spherical coordinates, ϕ, lies in the xy plane around the beam direction. The events taken into account in the analysis were limited to those for which $0.15 \leqslant \mathrm{KER} \leqslant 0.70$ eV. The upper limit was imposed in order to isolate the events due to the GSD mechanism, while the lower limit was imposed because the resolution in $\cos(\theta)$ becomes poor when KER is small.

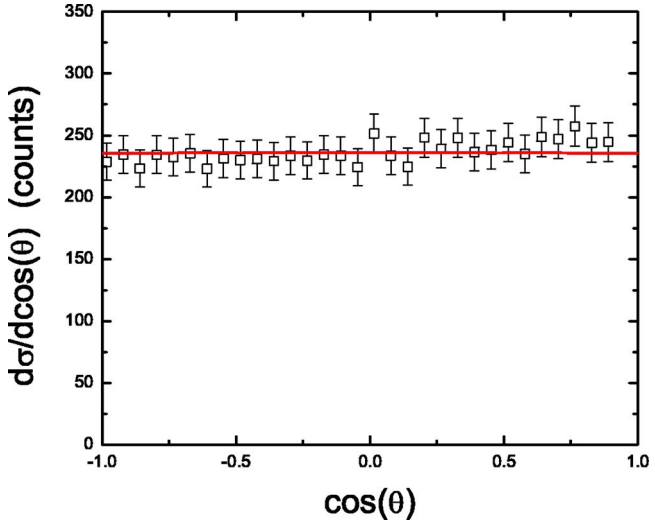

FIG. 4. (Color online) The measured differential cross section for single ionization as a function of $\cos(\theta)$, where θ is the angle between the molecular axis and the projectile beam. The curve is a fit to the data as described in the text. The flat distribution indicates the single ionization of D_2 is independent of the molecular alignment.

III. RESULTS AND DISCUSSION

The main results of our measurement are shown in Figs. 3 and 4. The momentum distributions in the laboratory frame are shown in Fig. 3. Each panel in the figure shows the momentum of the D^+ fragments projected into each of the three planes defined by the experiment geometry. Atomic units are used throughout unless otherwise specified. The plot of P_x vs P_y reflects the cylindrical symmetry of the collision system. Since P_x is measured from the time-of-flight and P_y from the position of the ions on the detector, this symmetry provides a check for the correct conversion into momentum space along each axis. From the momentum vectors we can calculate the angles of interest for this study. Figure 4 shows the measured differential cross section for single ionization as a function of $\cos(\theta)$.

From Fig. 4 it is clear that, unlike the results for double ionization [10], the single ionization cross section shows very little, if any, angular dependence. This result may be understood within the framework of both Dunn's symmetry arguments and two-center interference. First, Dunn [20] has shown that, within the Born approximation, ionizing transitions to some final states of the dissociating molecular ion may have anisotropic cross sections. This result follows from an analysis of the relative symmetry of the initial and final states about the axis of the momentum transfer. For the GSD process being studied here, both the initial and final state are specified, and transitions both parallel and perpendicular to the symmetry axis are predicted to be non-vanishing, so no anisotropy is expected. This result depends upon the momentum transfer to the electron being essentially transverse, which is consistent with the recent experimental results of Dimopoulou *et al.* [11].

Second, within the context of two-center interference, the isotropic result may be understood qualitatively by consider-

ing that the values for the electronic energy (Q) and projectile momentum ($\mathbf{q}$) transfers are both smaller, on average, for single ionization compared to double ionization. This decreases the amount of destructive interference one would expect. More quantitatively, we may model this one-electron process in much the same way as double-ionization [10,40]. Essentially, for the one-electron process, the amplitude for ionization is treated as the amplitude for ionization from two hydrogen atoms, to be added with a relative phase $e^{i\mathbf{q}\cdot\mathbf{R}}$, where $\mathbf{R}$ is the internuclear position vector. Since the projectile scattering angle was not measured, the experiment and calculation both integrate over the transverse component of $\mathbf{q}$. The longitudinal component of $\mathbf{q}$ (q_z) remains, and the factor $e^{iq_z\cdot R}$ gives rise to the interference term. For ionization

$$q_z = \frac{\epsilon_I + \frac{\mathbf{k}^2}{2}}{v_p}, \tag{2}$$

where ϵ_I is the binding energy of the target electron, v_p is the projectile velocity, and $\mathbf{k}$ is the momentum of the continuum electron. We assume that: (i) the collision time is much smaller than the rotational period of the molecule, which at $v_p=12.6$, is satisfied. (ii) Electron capture by the projectile is negligible [7]. (iii) The wave function for D_2 is approximately the same as that for two D atoms separated by the internuclear distance, R [41]. In this situation, $R=1.1$ for D_2 at R's where GSD is likely to occur. By representing the one-electron D_2^+ wave function as a linear combination of atomic orbitals,

$$\Psi_i = \frac{\Psi_{1s}(\mathbf{r}_A) + \Psi_{1s}(\mathbf{r}_B)}{\sqrt{2}}, \tag{3}$$

one arrives at a one-electron amplitude for the bound to continuum transition, which is the coherent sum of individual amplitudes corresponding to emission from each center

$$a_{fi} = \frac{1}{\sqrt{2}}[a(\mathbf{b}_A) + a(\mathbf{b}_B)e^{-iq_zR\cos(\theta)}], \tag{4}$$

where $\mathbf{b}_A$ and $\mathbf{b}_B$ are the impact parameters associated with each target center. Therefore, the probability as a function of both impact parameters, the internuclear vector, and longitudinal momentum transfer is given by

$$P(\theta,\mathbf{b}_A,\mathbf{b}_B) = \frac{1}{2}\{|a(\mathbf{b}_A)|^2 + |a(\mathbf{b}_B)|^2 + 2Re[a^*(\mathbf{b}_A)a(\mathbf{b}_B)e^{-iq_zR\cos(\theta)}]\}. \tag{5}$$

Assuming that the collisions take place at impact parameters larger than the internuclear separation,

$$|a(\mathbf{b}_A)|^2 \approx |a(\mathbf{b}_B)|^2 \approx |a(\mathbf{b})|^2, \tag{6}$$

$$P(\theta,\mathbf{b}) = |a(\mathbf{b})|^2[1+\cos(q_zR\cos(\theta))], \tag{7}$$

$$d\sigma(\theta) = \int P(\theta,\mathbf{b})db = d\sigma_a[1+\cos(q_zR\cos(\theta))]. \tag{8}$$

We note that $d\sigma(\theta)$ reaches its maximum at $\theta=90°$, regardless of the value of q_z.

While the cross section is maximized for molecules oriented perpendicular to the beam direction in this model, the magnitude of the angular asymmetry may be quite small. Compared to the similar double-ionization experiment [10], this experiment typically has smaller values for both q_z and R. For GSD, the most likely R is near 1.1. As described by Eq. (2), the value of q_z depends on $\mathbf{k}$, which we do not measure. Weber and co-workers [42] have measured the momentum of the continuum electron for 1.3 MeV H^++He collisions, finding the peak of the q_z distribution near 0.14 for single ionization, suggesting a $|\mathbf{k}|$ of about 0.46. Using this number for our system gives an estimate of $q_z=0.05$. Fitting Eq. (8) to the data allowing q_z and σ_a to be free parameters yields good agreement for the q_z value estimated above, as shown by the solid line in Fig. 4. The fit quality is fairly insensitive to the specific value of q_z, as long as it does not become too large ($\leqslant 0.25$). By comparison, the value of q_z found in the double ionization experiment was 1.1 [10]. Our isotropic results, then, are in agreement with expectations based on a two-center interference model. While the maximum cross section is expected at $\cos(\theta)=0$, the momentum transfer in these single-ionization collisions does not result in a measurable anisotropy. The measurement was repeated with 1 MeV/amu F^{7+} projectiles with nearly identical results for the angular cross section.

IV. SUMMARY

By isolating the ground state dissociation channel we have successfully measured the differential cross section for single ionization as a function of $\cos(\theta)$ for collisions between fast ions and hydrogen molecules. Within the precision of our measurement, the single ionization cross section was found to be isotropic. This result is in good agreement with a model that includes two-center interference, since the longitudinal momentum transfer in these collisions is not large enough to lead to measurable anisotropy.

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