Comparative study of the ground-state dissociation of H_2^+ and D_2^+ induced by ionizing and electron-capture collisions with He^+ at velocities of 0.25 and 0.5 a.u.

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The ground-state dissociation (GSD) of H_2^+ and D_2^+ molecular ions induced by ionizing and electron-capture collisions of He^+ projectiles with hydrogen and deuterium molecules was investigated at impact velocities of 0.25 and 0.5 a.u. Momentum distributions of slow H^+ and D^+ fragments were measured and compared with calculations based on GSD theory. At the higher velocity, and for both capture and ionization, these distributions are well accounted for by a model that attributes the momenta to the vibrational wave function of the parent H_2 or D_2 molecule. For the lower velocity, this model remains successful for capture but not for ionization. For the latter case we observe a large transverse-momentum component imparted to the vibrationally excited H_2^+ or D_2^+ system beyond that predicted by the model. This feature is interpreted as being caused by momentum transferred to the motion of the nuclei within the molecule's center-of-mass frame by way of a direct interaction between the projectile and one of the molecule's nuclei.

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I. INTRODUCTION

The fragmentation of H_2 and D_2 molecules by electron and ion impact has been the subject of numerous investigations (see, for example, Refs. [1–6]). In most previous studies the emphasis was on cross-section or kinetic-energy release measurements. The development of momentum imaging techniques and, in particular, COLTRIMS (cold target recoil ion momentum spectroscopy) provided new and detailed information about the momentum of the recoiling molecular ions, fragments, and ionized electrons. Recent studies of the Coulomb "explosion" of the two charged fragments following the removal of both target electrons were carried out employing these techniques [7–10]. One point of interest was the separation of the momentum transferred by the highly charged projectile in the slow collision into (i) the momentum transferred to the molecule as a whole (i.e., to its center-of-mass motion), and (ii) the momentum of the fragments originating from the dissociation process [7]. One of the goals of this study is to find other such processes in which one may possibly separate the momentum transferred to the molecule from the momentum released upon dissociation, and even more importantly to distinguish the momentum transfer to the molecule as a whole from the momentum transferred to the motion of the nuclei within the center-of-mass frame.

In the present investigation we have studied the removal of one electron from H_2 (and D_2) targets by capture and by ionization. We have focused on electronic transitions that leave the target molecular ion in the vibrational continuum of

its electronic ground state. The overall process, referred to as ground-state dissociation (GSD), has attracted some interest recently [11–14] as a probe for studies of very slow half-collisions [14]. If the collision is fast enough that the electron removal can be described as a vertical transition (i.e., the nuclear motion during the transition is negligible), there is a small chance for populating the vibrational continuum of the $1s\sigma$ electronic ground state of H_2^+ and D_2^+ (about 1.5% and 0.5% of all transitions to the electronic ground state of H_2^+ and D_2^+, respectively [12]). This transition is immediately followed by the slow dissociation into H^+ and $H(1s)$. The charged fragments produced in GSD can be distinguished by their kinetic energy from those produced by the dissociation of the H_2^+ electronic excited states, because the former are sub-eV (their energy spectrum peaking at zero energy and falling off roughly exponentially with a full width at half maximum (FWHM) of about 0.25 eV) while the latter have typically a few eV.

The very low momentum of the GSD H^+ and D^+ fragments makes them good probes for small momentum transfers during the collision. In this paper we provide evidence that electron capture proceeds at large impact parameters relative to the size of the molecule for the collision velocities studied, resulting in small amounts of molecular-ion recoil momentum. By contrast, considerably larger amounts of recoil momentum are transferred to the dissociating fragments when the electron is promoted into the continuum (ionizing collisions), indicating that the latter process occurs at smaller impact parameters. For electron capture and for ionization at the higher velocity, we find that the recoil momentum imparted to the target molecule is primarily imparted to the molecule as a whole, i.e., to its center-of-mass (c.m.) motion. However, for the case of the lower velocity ionization we report evidence that appreciable momentum is transferred to the molecule via a direct interaction with one of the molecule's nuclei rather than with the molecule as a whole.

The experiments on which we report here, $He^+ + H_2$

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$\rightarrow$He$^+$+H$^+$+H^0+e (ionization) and He$^+$+H$_2\rightarrow$He^0+H$^+$ +H^0 (electron capture) and the equivalent collisions with D$_2$ targets, were performed at impact velocities V_p of 0.25 and 0.5 a.u. with the COLTRIMS setup installed at the CRYEBIS ion-source facility of the J R Macdonald Laboratory of Kansas State University. This setup has been used previously to study the ionization process of noble gases and H$_2$ (leaving the molecular recoil ion H$_2{}^+$ in a nondissociative state) at slow impact velocities as well as to perform state-resolved electron-capture experiments by highly charged ions [15,16].

II. EXPERIMENTAL SETUP

The experimental technique and data reduction method have been described in detail previously in Ref. [15], so only a brief summary will be presented. The H$_2$ and D$_2$ gas targets consisted of precooled ($\approx$60 K) supersonic jets, perpendicularly intersected by the projectile beam. Electrons and charged molecular dissociation fragments were extracted from the collision region by an electric field oriented perpendicular to both the target gas jet and the projectile beam. The charged particles were collected on position-sensitive microchannelplate detectors. In the case of electron-capture experiments, the neutral postcollision projectiles He^0 were detected in coincidence with the charged fragments by another position sensitive microchannelplate detector placed farther downstream, after electrostatic separation from the main beam. In ionization experiments the ejected electrons were detected in coincidence with the charged fragments by a third-position-sensitive detector placed opposite to the fragment detector relatively close to the interaction region. In both ionization and capture experiments the flight time of the charged dissociation fragment was also recorded. The same coordinate system as that employed in Ref. [15] is used, with the Z axis coinciding with the incident beam, the Y axis with the gas target jet and the X axis with the direction of the electric extraction field. Both electron and fragment detectors are oriented parallel to the YZ plane.

Special care was taken to make sure that water-vapor contamination, which could produce H$_2{}^+$ ions indistinguishable from D$^+$ ions, was insignificant during the experiment. The precooling of the target gas eliminated possible contamination of the supersonic target jet. In addition the residual gas contained in the scattering chamber was tested for contamination in a 24 hour "dry" run without target gas. The amount of H$_2{}^+$ observed in the time-of-flight spectrum of this run was insignificant.

III. RESULTS AND DISCUSSION

In the velocity regime of the present investigation a collision between the incident projectile and the initially stationary target atom or molecule transfers momentum from the projectile mainly to the heavy constituents of the target. In the case of a collision resulting in ionization a small amount of momentum is also transferred to the ejected electron. In previous studies by us of electron spectra from ionization [15], electron momenta up to about V_p (in a.u.) were observed in He$^+$+H$_2$ collisions at impact velocities ranging

from 0.25 a.u. to 1.25 a.u. Electron momenta between zero and V_p, with a mean value of 0.5 V_p, were also observed in the present experiment. This typical electron momentum is much smaller than momenta of the heavy fragments after the collision and may be neglected in discussing the heavy-particle momentum balance. Since these electron-momentum distributions have no other bearing on the subject presented here, they will not be discussed further.

The present experiment determined all three-momentum components p_x, p_y, and p_z of the charged fragment. This momentum can be viewed as coming from two sources, namely, the collisional momentum transferred to the target from the projectile and the momentum imparted to the fragment in the dissociation process. Discriminating between these contributions does not pose a problem if both fragments are detected [9,15]. In the present case, however, one of the fragments is electrically neutral and, therefore, escapes detection. Thus the recoil momentum and the dissociation momentum of the charged fragment cannot be easily disentangled. A major point of the present paper is to suggest a model-dependent method for separating these contributions.

The longitudinal (z) component of the collisional momentum transfer to the residual target molecular ion is expected to be very narrow in distribution, as determined by the kinematics. The longitudinal momentum p_z (molecular ion) is related to the change in electronic energy (Q value of the collision) according to Eq. (3.1) or (3.2)

$$p_z(\text{molecular ion}) = \begin{cases} -Q/V_p - V_p/2 & \text{for EC,} \qquad (3.1) \\ -Q/V_p - p_z^e & \text{for } I, \qquad (3.2) \end{cases}$$

(where EC refers to electron capture, I stands for ionization, and p_z^e denotes the longitudinal momentum of the ejected electron). In the case of electron capture, the electronic transition is expected to populate a well-defined final state, namely, that of the He in its ground state and the H$_2{}^+$ or D$_2{}^+$ molecule on its ground potential-energy curve very near the dissociation limit, and thus Q is well defined. In the case of ionization, previous measurements have shown that the Q distribution is typically quite narrow due to the very small size of the ejected electron momentum. Thus the typical spread in p_z (molecular ion) is expected to be less than 1.5 a.u. for all systems shown in this paper. Because the longitudinal (z) component of the collisional momentum transfer to the residual target molecular ion is so sharp, we expect that the longitudinal momentum of the observed H$^+$ or D$^+$ fragment is almost entirely due to the GSD process. On the basis of this argument, we hereafter use the notation $p_z(d)$ to denote the momentum of the observed fragment, where the (d) indicates that this momentum is to be interpreted as due entirely from the dissociation process. No corresponding argument can be made for the transverse momentum of the fragment (p_z), however.

In Fig. 1 we show the transverse-momentum (p_x) distributions for H$^+$ fragments from both ionization and electron capture for He$^+$ incident on H$_2$ at a projectile velocity of 0.25 a.u. The p_x spectra, derived from the flight times of fragments, served to discriminate the GSD process from other fragmentation processes yielding fragments with larger
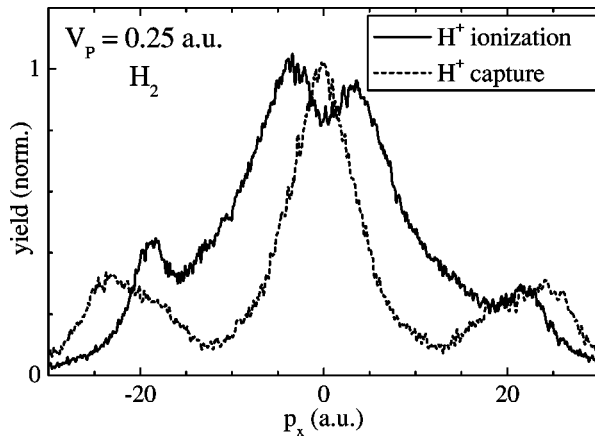
FIG. 1. Distributions of one (p_x) of the transverse-momentum components of H^+ from ionizing (solid line) and electron-capture (dashed line) collisions of He^+ with a H_2 target at a projectile velocity of 0.25 a.u. These distributions were derived from the flight time of H^+. While the central peaks correspond to ground-state dissociation (GSD), the lateral structures are caused by the presence of dissociation channels yielding "fast" fragments not discussed in the present paper. It should be stressed that no absolute cross sections were determined in the present paper, thus the experimental yields in this and all subsequent figures are normalized to unity.

FIG. 2. Experimental p_x (open circles) and $p_z(d)$ (solid line) momentum distributions for electron capture (a)–(c) from and ionization (d)–(f) of H_2 and D_2 targets at impact velocities of 0.25 a.u. and 0.5. a.u.

momentum values ("fast" fragments). GSD fragments (the center peak) are separated from "fast" fragments, which appear as lateral structures. This separation is due to the fact that only a small number of fragments with larger momentum values are collected on the fragment detector because of the large distance from the collision region and the relatively weak extraction field used. Only those "fast" fragments are detected that are predominantly emitted towards (negative momenta) or away from (positive momenta) the fragment detector. "Fast" fragments emitted predominantly parallel to the detector (i.e., possessing small p_x values) escape detection, thereby, creating the central "void" where the GSD peak is located. All fragment p_z spectra discussed hereafter in the paper were generated by selecting only events whose associated charged fragment possessed a p_x value within a gate placed on the respective GSD peak.

It becomes evident from the figure that the width of the GSD capture peak is considerably smaller than that of the GSD ionization peak even though in both cases the momentum gained from the dissociation is the same (see below). This indicates that a smaller collisional recoil momentum is transferred to the target in a capture collision than in an ionizing collision. The GSD peak for H^+ fragments (ionization) appears "split." It will become apparent during the discussion of this "splitting" that it should not be interpreted as a GSD double peak, but rather as a feature superposed on the "main" GSD distribution, where the latter is peaked at zero momentum [see Fig. 5(c)]. This additional feature will be attributed to a different momentum-transfer mechanism leading to GSD, whereby the projectile interacts directly with one of the target molecule's nuclei.

In Fig. 2 we present fragment p_x distributions from electron-capture collisions [left column, Figs. 2(a)–2(c)] and ionizing collisions [right column, Figs. 2(b)–2(f)] as well as
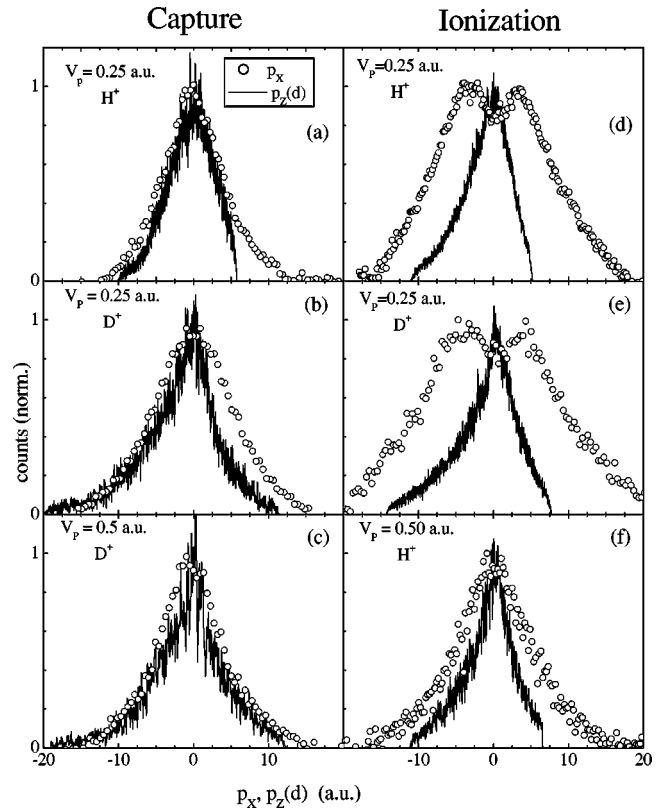
the associated $p_z(d)$ distributions. Keeping in mind that the latter contain no recoil component we conclude, from the similarity of the p_x and $p_z(d)$ capture distributions, that indeed little recoil momentum is transferred to the molecular ion during a capture collision. In stark contrast to this conclusion are the p_x and $p_z(d)$ distributions for ionizing collisions. A considerable amount of recoil momentum is present in p_x, in particular, so at the lower impact velocity of 0.25 a.u., where the aforementioned GSD peak "splitting" appears. The presence of a large recoil-momentum component suggests that ionization is taking place at smaller impact parameters than capture.

Figure 3 compares fragment $p_z(d)$ spectra for ionizing and capture collisions at $V_p = 0.25$ and 0.5 a.u. For a given projectile velocity, ionization, and capture spectra have very similar shapes, indicating that (i) indeed the longitudinal momentum distributions do not contain any additional recoil momentum component besides the one related to the Q value of the collision as assumed previously and (ii) the same dissociative state of the molecular ion is populated in both ionization and capture, i.e., the electronic ground state of the H_2^+ (D_2^+) in its vibrational continuum as expected for the GSD process.

In order to place the above interpretations on a more quantitative foundation, we evaluate a definite model calculation of the expected fragment momentum spectra from the GSD process. The first step of this process is a vertical tran-
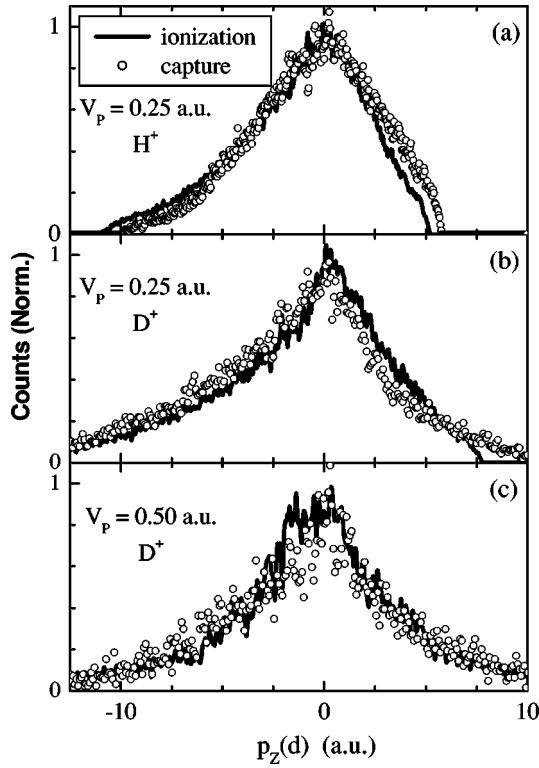
FIG. 3. Longitudinal dissociation momentum $[p_z(d)]$ distributions for H^+ and D^+ fragments at $V_p = 0.25$ a.u. (a), (b) and D^+ fragments at $V_p = 0.5$ a.u. (c) originating from electron capture (open circles) and ionizing (solid line) collisions.

sition from the ground vibrational state of the neutral molecule into the vibrational continuum of the $1s\sigma$ electronic ground state of the molecular ion. Thus, the transition probability to a specific vibrational continuum state, i.e., a specific energy above the dissociation threshold, is given by the Franck-Condon factors

$$P(E) = \left| \int_0^\infty \psi_{\nu=0}^*(R)\phi_E(R)dR \right|^2, \qquad (3.3)$$

where R is the internuclear distance, $\psi_{\nu=0}(R)$ is the vibrational wave function of the H_2 ground state and $\phi_E(R)$ is the continuum vibrational wave function of the $H_2^+(1s\sigma)$ state for an energy E above the dissociation threshold. This energy E is carried away as the kinetic energy of both fragments after dissociation. The wave functions were computed numerically as described in detail previously [11,16], then the overlap integrals were evaluated. The transition-probability peaks at the threshold and falls off approximately exponentially with increasing energy above threshold with a FWHM of about 250 meV (see Fig. 5 of Ref. [16], for example). The total fraction of transitions to the continuum was evaluated by integrating over the energy in the continuum and compared to the same quantity calculated using closure, i.e., subtracting the sum of all transitions to the bound vibrational states from unity. Finally, the momentum distribution of the dissociating fragments was generated for the computed $P(E)$ distribution assuming that the dissociation is isotropic. In what follows we frequently compare experimental results with those of calculations. It should be kept in mind, however, that no absolute cross sections were measured in the present work. Therefore data and calculation are always normalized for the sake of comparison.

We compare the results of this calculation with the experimental spectra of $p_z(d)$ in Fig. 4. The distributions from electron capture and ionizing collisions at $V_p = 0.25$ and 0.5 a.u. are in good agreement with the theory for all cases, confirming our interpretation of the longitudinal fragment momenta as due entirely to the GSD process.

The transverse-momentum case is more complex. In Fig. 5 we compare the simulated p_x distributions obtained from the above GSD calculation with the data for three selected collision systems. This calculation clearly fails, increasingly so as one proceeds from capture to ionization and from high to low projectile velocity. We interpret this to mean that the collisional momentum is not at all negligible in the transverse direction. In order to deal with this contribution, we extended the model calculation by folding into the GSD momentum distribution the transverse collisional momentum
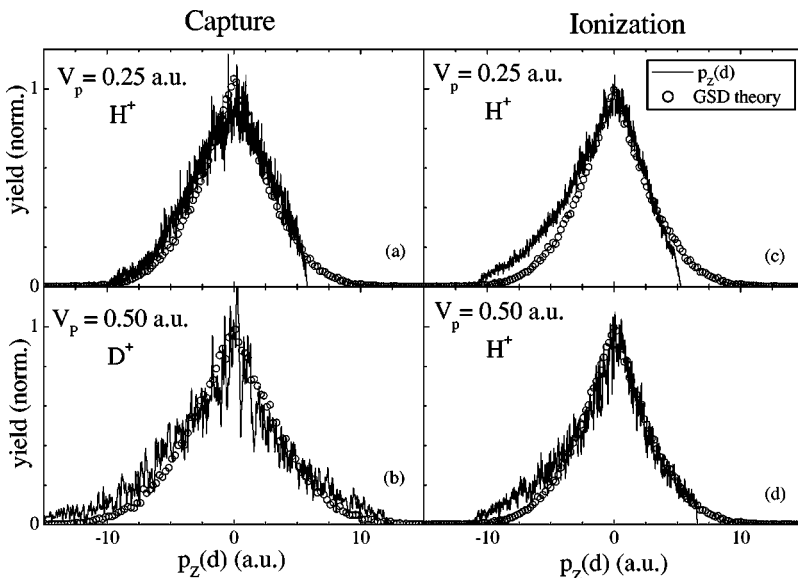

FIG. 4. Comparison of the experimental $p_z(d)$ momentum distributions (solid line) for electron capture (a), (b) and ionization (c), (d) at impact velocities of 0.25 a.u. and 0.5 a.u. with results of a GSD calculation (open circles).
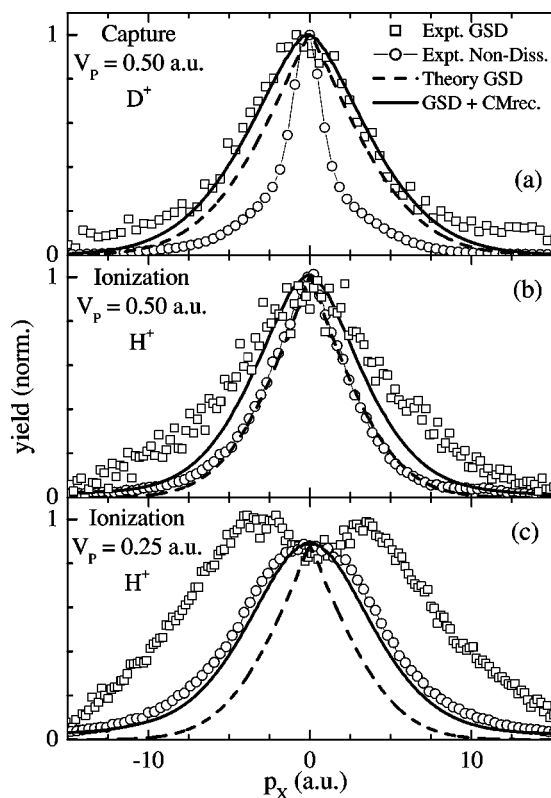
FIG. 5. Simulation of the charged fragment's transverse momentum component p_x (solid line) by a calculation based on GSD theory (dashed line) and comparison with experimental data (open squares). The molecular ion's c.m. recoil momentum distribution, which entered this simulation, was taken from the measured p_x distributions of H_2^+ and D_2^+ molecular recoil ions in nondissociative states (open circles).

measured for the same collisional systems but populating the nondissociative states of the ground H_2^+ and D_2^+ molecular ion [15]. These experimentally measured distributions are shown as open circles in the figure. The folding was performed by adding to the GSD velocity of the fragment in the c.m. frame the velocity of the c.m. of the molecule, taking the GSD velocity vector to be isotropically distributed. The distribution of the c.m. velocity vector was calculated from the experimentally determined H_2^+ (D_2^+); p_x distribution for each case. The result of this folding is shown as the solid curve in the figure.

Figures 5(a) and 5(b) show that, at $V_p = 0.5$ a.u. this model reproduces the data very well for both electron capture and ionization, suggesting that the momentum transferred during the collision goes mostly to the c.m. motion of the molecule and does not differ from the momentum exchanged during a collision leading to a stable molecular recoil ion. In contrast a ''splitting'' of the GSD peak is observed for ionizing collisions at $V_p = 0.25$ a.u. that is not

reproduced by the model. We believe that the reason for this failure of the model is that the momentum transmitted to the molecule from the projectile is partially deposited in the internal vibrational energy of the molecule and is not distributed equally between the two nuclei of the molecule. That is, the process cannot be interpreted simply as a momentum transfer to the center of mass of the molecule followed by isotropic dissociation, but instead should be viewed as a momentum transfer to one of the two nuclei of the molecule. In this scenario the projectile passes close to one of the target's protons (deuterons), imparting to it most of the transverse momentum. An estimate of the impact parameter, which would lead to the observed momentum transfers, suggests that this explanation is plausible. A Coulomb interaction between the incident He^+ (for which we choose an effective charge of 1) and one of the target's protons is assumed, for whose effective charge we adopt the value of 0.9 used in Ref. [17]. Taking 4 a.u. [see Fig. 5(c)] as a likely value for the recoil momentum imparted to one of the protons, this assumption yields a value of 1.8 a.u. for the impact parameter, comparable to the internuclear separation of 1.4 a.u.

IV. CONCLUSIONS

Momentum distributions of slow H^+ and D^+ fragments originating from GSD of the respective molecule were measured for ionizing and electron-capture collisions at impact velocities of 0.25 and 0.5 a.u. The observed longitudinal momentum distributions, for both ionizing and electron-capture collisions, are in good agreement with GSD calculations, suggesting the observed recoil-momentum spread is nearly entirely due to the GSD process rather than any collisional momentum transfer. The transverse-momentum distributions indicate that ionizing collisions at these velocities occur at smaller impact parameters than capture collisions. A model based on convoluting the GSD calculations with the measured transverse-momentum transfer to the nondissociating H_2^+ and D_2^+ molecular ions is in good agreement with the data for capture and for the faster ionizing collision. This model assumes that the collisional momentum is transferred to the molecule as a whole. However, for slow ionizing collisions this model fails, suggesting that the transverse-momentum transfer is better viewed as a direct interaction of the projectile with one of the molecule's nuclei from which it is scattered in a close collision (i.e., at impact parameters of the order of the size of the molecule or smaller).

ACKNOWLEDGMENTS

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