Five-body calculations of D_2 fragmentation by Xe^{19+} impact

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(Received 9 April 1999)

A five-body classical trajectory Monte Carlo model has been developed to study fragmentation of diatomic molecules after double electron removal by highly charged ion impact. A systematic study of the final-state deuteron energy and momentum spectra has been conducted for $Xe^{19+} + D_2$ collisions at impact energies ranging from 1 eV/u to 100 keV/u. At the highest projectile energies, the fragment energies and momenta are determined by the Coulomb explosion of the doubly-ionized molecule via the known Franck-Condon transition for the isolated molecule. The deuterons are emitted back-to-back with nearly equal energies. At the lowest projectile energies, the final state behavior is due mainly to the collisional momentum transfer from the slow-moving projectile. The deuterons are strongly scattered in the direction opposite to the transverse momentum of the projectile with energies far greater than those produced in the Franck-Condon transition. At energies around 150 eV/u, both slow and fast deuterons are predicted. This is due to the vector addition of the collisional momentum transfer to the center of mass of the molecule with that due to the two-body Coulomb breakup of the dissociating ions. [S1050-2947(99)01009-4]

PACS number(s): 34.10.+x, 34.50.−s

I. INTRODUCTION

Although molecular fragmentation processes have been studied for years, recent advances in experimental and theoretical techniques have shed new light on the subject. Historically, experimental investigations of molecular fragmentation were conducted by colliding a variety of projectiles with the target. Projectiles that have been used include electrons [1,2], singly [3,4] and multiply charged ions [5–9], and photons [10]. Measurements of cross sections, yields, and energy distributions revealed the structures of molecules, how energy is transferred from the incoming projectile to the molecule, and how energy is distributed among molecular states. These data were interpreted in terms of Franck-Condon transitions between states of the isolated molecule.

Theoretically, many studies have been performed that modeled H_2 by doubling the cross sections for atomic hydrogen. In addition, a unitarized atomic orbital method has been developed to describe electron capture from H_2. In these studies a one-electron model was used for collisions involving low charge state ions [11]. Later, a two-electron atomic orbital description was developed and implemented for single electron capture using the assumption that the H_2 internuclear separation could be set equal to zero [12]. Other calculations on H_2 employed the classical trajectory Monte Carlo method with independent electrons to study state-selective electron capture and ionization [13,14]. Moreover, the continuum distorted wave model has been applied to $O^{8+} + H_2$ transfer ionization collisions to explain the cross section dependence on the alignment of the molecular axis [5].

Recently, a five-body classical trajectory Monte Carlo (CTMC) model has been developed to predict the electronic and nuclear dynamics for double electron removal by highly charged ion impact [15]. The present work is an extension of these studies and focuses on double electron removal from D_2 by Xe^{19+} projectiles. This collision system was chosen because experimental data are now available for some aspects of our theoretical study, and new fragmentation dynamics are illuminated.

These features can now be probed because of the development of slow, highly charged ion sources. These new ion sources provide the possibility to study molecular breakup at relatively low impact energies which encompasses an entirely new (non-Franck-Condon) regime [16]. The high projectile charges and low energies provide long collision times which are on the order of the femtosecond fragmentation times. This means that the molecular fragments interact not only with each other, but also with the outgoing projectile. As a result, all three nuclei (in the case of diatomic targets) participate in the energy sharing. Thus, the final energy and momentum spectra for the molecular fragments are determined by the energy and momentum transferred from the projectile, in addition to the potential energy released by the dissociation of the molecule. In contrast, for fast collisions the final kinetic energy of the deuterons is determined by isolated molecule Franck-Condon transitions.

It is the purpose of this paper to provide detailed information about the energy and momentum sharing in these reactions. The evolution of these dynamics is followed over a wide range of collision energies.

II. THEORY

The model for ion-molecule collisions using the classical trajectory Monte Carlo method is an extension of that for one-electron atoms described in Ref. [17]. For H_2 (or D_2), an additional target nucleus and electron are involved. This creates a complex five-body problem. Hamilton's equations of motion (30 coupled first order differential equations) are iteratively solved for thousands of collision events in order to obtain sufficient statistics for the reaction under study. Early CTMC studies on H_2 targets have used an independent-electron model with one electron [13] to calculate single ion-

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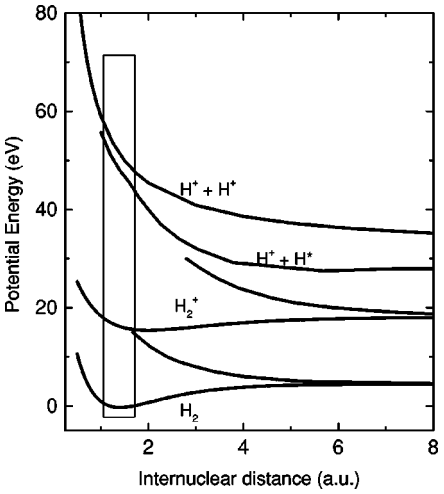

FIG. 1. Energy curves for the deuterium molecule and the ions after impact and dissociation.

ization and capture by heavy ions. Currently, two atomic centers, each with an electron, are used to enable the direct investigation of double electron removal mechanisms for D_2.

In this study, each electron is initially bound to its parent atomic center by the Coulomb force and has no dependence on the other nucleus or the other electron. The molecule is bound by a Morse potential

$$V_m(R)=D_e(1-e^{-B_e(R-R_e)})^2, \tag{1}$$

where D_e is the dissociation energy, R is the separation of the atomic centers, B_e is the curvature parameter, and R_e is the separation defined such that $V_m(R_e)$ is a minimum. The values for $D_e=4.7$ eV, $R_e=1.40$ a.u., and $B_e=0.73$ a.u. are determined from spectroscopic data [18]. With the implementation of the Morse interaction, the collisional dissociation of an isolated D_2 molecule to its ground state atoms is theoretically well described.

To extend the CTMC method to the D_2 molecule, additional quantities must be generated for the position and momentum of both atomic centers. It is assumed that the molecule is in the ground vibrational state with a separation distance selected randomly from its vibrational Gaussian squared distribution. The distribution is centered about the minimum of the potential well ($R_e=1.40$ a.u.) of the Morse potential. This implementation is necessary to obtain the correct Franck-Condon energy distribution of the dissociating deuterons for the isolated molecule. The electrons are placed on the atoms as described for this simple hydrogenlike case [17], with the ionization potential for each electron set equal to 13.6 eV. The molecular axis is randomly orientated at the start of each trajectory.

As the system evolves during a collision and one or both electrons are removed from the molecule, interactions are included to replicate the dissociative potential curves shown in Fig. 1. We dynamically model these interactions. If one electron attains a positive energy during the collision, thereby placing the molecule in the D_2^+ state, the Coulomb electron-electron interaction is included in the Hamiltonian along with the Coulomb interactions between both electrons and the other target center. If the electron remaining on the molecule then reaches an energy corresponding to the first excited state, D* ($n=2$), the Morse potential between the two centers is slowly switched off and the interactions between all particles are Coulombic. This simulates the $D^+ + D^*$ dissociating interactions which are molecular Rydbergs of D_2^{++} for $R<5$ a.u. For complete double electron removal, the Hamiltonian resorts to the five-body Coulomb problem. Since all interactions are included in the final state, the momentum of each particle can be determined to produce a kinematically complete calculation for the double electron removal process.

It is important to note that the energy required to remove both electrons from D_2 is the sum of the ionization energies for the two electrons (27.2 eV), the 4.7 eV needed to break the ground-state D_2 molecular bond, and the approximately 19 eV required to place the two deuterons on the repulsive Coulomb curve at the equilibrium distance. Our model reproduces the true energy of 50.9 eV required to remove both electrons in a vertical Franck-Condon transition. If the D_2 molecule is displaced from its equilibrium position during the collision, the corresponding energy required for double

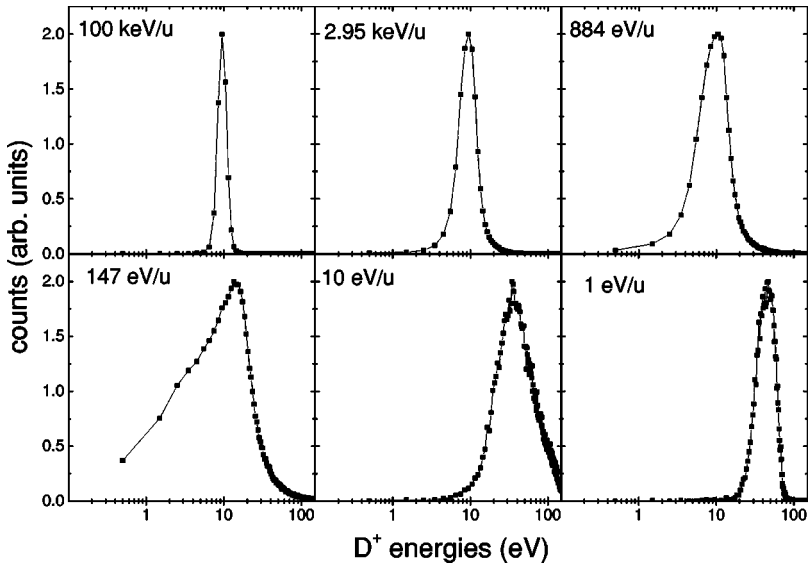

FIG. 2. Energies of the deuterons after double electron removal in $Xe^{19+}+D_2$ collisions.

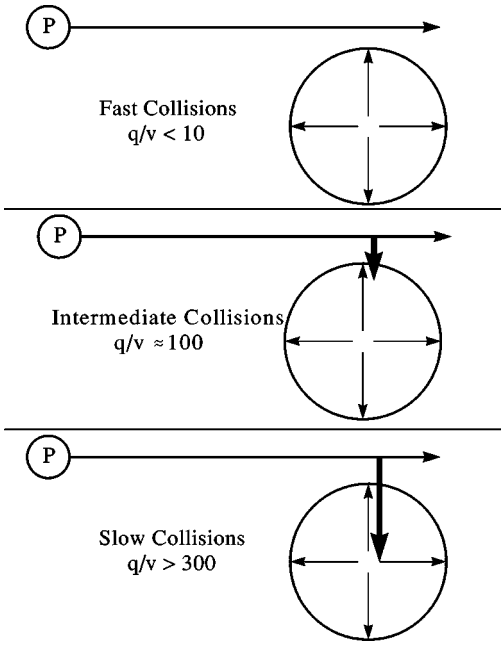

FIG. 3. Illustration of vector addition of momentum for $Xe^{19+} + D_2$ collisions. The bold arrows correspond to collisional momentum transfer while the light arrows correspond to momentum acquired via fragmentation.

electron removal will differ from the vertical Franck-Condon transition and be portrayed by the changes in the Morse interaction for the ground state and the internuclear position of fragmentation on the repulsive $D^+ + D^+$ Coulomb curve.

III. RESULTS AND DISCUSSION

The removal of both electrons from a D_2 molecule results in the dissociation of the molecule. To investigate the properties of the energy and momentum spectra of the target nuclei, simulations have been made for the $Xe^{19+} + D_2$ collision systems at projectile energies of 100 and 2.95 keV/u and 884, 147, 10, and 1 eV/u. There are two contributions to the final kinetic energy of the target ions. One is the energy transferred from the projectile during the collision, and the other is from the Coulomb repulsion between the deuterons after the electrons have been removed (see Fig. 1). At low collision energies, double capture and transfer ionization are the dominant double electron removal mechanisms. At the highest energy, impact ionization of both electrons also contributes to the two-electron removal process.

Presented in Fig. 2 are the calculated energy distributions of each deuteron. At 100 keV/u, distinct Franck-Condon behavior is observed. The distribution peaks at 9.5 eV and has a full width at half maximum of approximately 2.5 eV. At an impact energy of 884 eV/u, several changes in the distribution are noticed. Although the energy distribution maximizes at roughly the same energy as before, the distribution is much broader. In addition, there are more slow (sub-5 eV) ions present. At a projectile energy of 147 eV/u, the energy distribution changes dramatically. The peak moves to a higher value of ≈ 13 eV. However, of greater interest are the large numbers of both slow and fast ions. In the 10 and 1 eV/u collisions, there are no slow ions present and the peak of the distribution is in the 30–50 eV range.

One can understand these energy spectra by considering the vector addition of momentum (see Fig. 3). In the figure, collisional momentum transfer from the projectile to the molecule is indicated by a heavy arrow and momentum acquired via fragmentation is indicated by a light arrow. In fast collisions (top), the momentum transfer from the projectile is negligible. Since the projectile velocity is much greater than the dissociation velocity of the deuterons, it does not participate in the post-collision momentum sharing. It simply strips the electrons from the molecule and leaves the molecule to dissociate via the known Franck-Condon transition. In intermediate speed collisions (middle), the momentum transfer from the projectile is comparable to the fragmentation momentum. When one adds the two momentum vectors, one can see how both fast and slow deuterons can be produced in the lab frame, as was seen for the 884 and 147 eV/u cases. In slow collisions (bottom), the momentum transfer from the projectile dominates the collision. The projectile speed is comparable to the fragmentation speed of the deuterons which causes all three heavy particles to actively participate in the post-collision momentum sharing. As a result, large

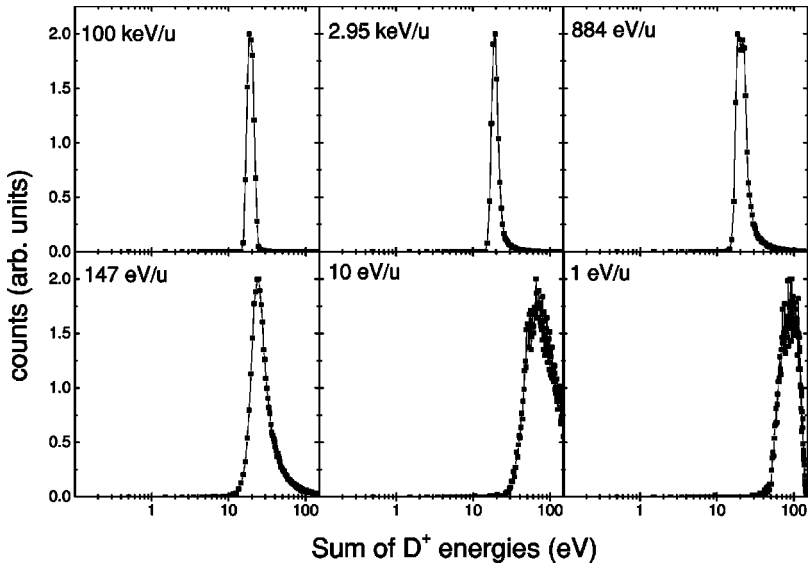

FIG. 4. Sum of the energies of the two deuterons after double electron removal in $Xe^{19+} + D_2$ collisions.

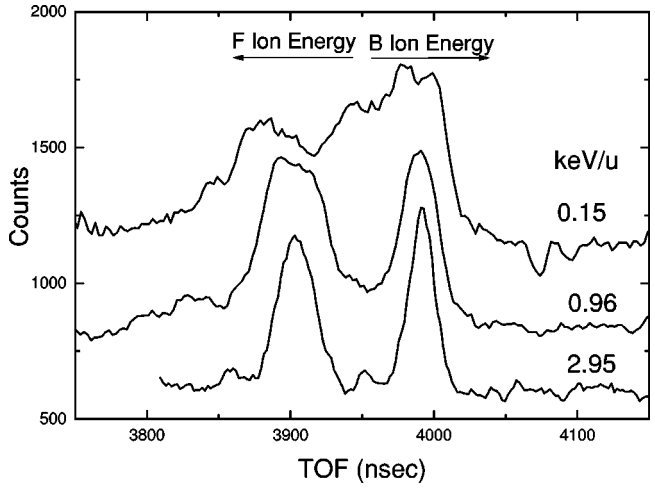

FIG. 5. Time-of-flight spectra for D^+ ions produced after double electron removal from D_2 by Xe^{19+}. The spectra have been arbitrarily scaled in height and offset vertically for display purposes.

amounts of momentum are transferred to each deuteron and only fast deuterons are seen, as was the case for the 10 and 1 eV/u collisions.

The sum of the two deuteron energies is plotted in Fig. 4. One can see that the trend is for the sum of the ion energies to increase as projectile energy decreases. This further illustrates that more energy is transferred from the projectile in the slower collisions. It is also important to note that there are no cases where the sum energy is less than 10 eV. This demonstrates that the molecular bond does not stretch prior to the electron removal, which would cause the total dissociation energy to be much less than 19 eV.

The preceeding figures illustrate that for decreasing collision energies in the few- to sub-keV/u range, the fragmenting D^+ ion energy spectra systematically change. These changes include (a) an increase in the mean energy and (b) the production of higher, and lower, energy ions than those produced via Franck-Condon transitions involving states of an isolated D_2 molecule.

These theoretical predictions are supported by experimental data obtained in a collaborative effort between the University of Missouri-Rolla and the KVI Groningen, where $2.95-0.15$ keV/u Xe^{19+} ions were used to bombard a D_2 target. An electrostatic extraction field and time-of-flight spectroscopy were used to study the fragmenting D^+ ions. By this method, time spectra with two peaks corresponding to fragment ions emitted towards (F peak) and away from (B peak), the ion exit aperture were obtained, as shown in Fig. 5. The data have been scaled and shifted vertically for display purposes.

Although the observed peak shapes and intensities are influenced by experimental parameters which must be convoluted with the fragment ion energy spectrum, the time separation between a forward and backward emitted ion is small if the fragment energy is small and large if the energy is large [19]. The relative energies are indicated by the arrows at the top of the figure. Figure 5 clearly demonstrates a systematic broadening of the peak widths with decreasing impact energy. This is due to higher and lower energy fragments being produced, just as predicted by our CTMC model. Most notable is the filling in between the peaks due to the production of low energy ions. In addition, broad tails due to the production of higher energy ion fragments can also be seen on the outside of both peaks (as predicted by our model).

The CTMC model allows us to further investigate the dynamics of the collision. The energy of one deuteron has been plotted against the energy of the other in Fig. 6. At the highest impact energy, the total energy of the deuterons is determined by the vertical Franck-Condon transition to the repulsive Coulomb potential of the isolated molecule. At this high impact energy, the molecule dissociates long after the projectile has any influence on the energy sharing. Therefore, the energy is shared equally between the deuterons, i.e., a well-defined point centered at $E_1=E_2\approx 9.5$ eV. At intermediate projectile energies, the pattern of having one fast deuteron and one slow deuteron is present. The peak shifts away from the $E_1=E_2\approx 9.5$ eV point. This indicates that the kinetic energy transfer from the projectile is beginning to have a greater influence on the collision. At the lowest impact energy, the energy transfer from the projectile dominates the collision, causing the $E_1=E_2$ line to be densely populated. This indicates that equal amounts of the energy are transferred to the deuterons from the slow-moving projectile.

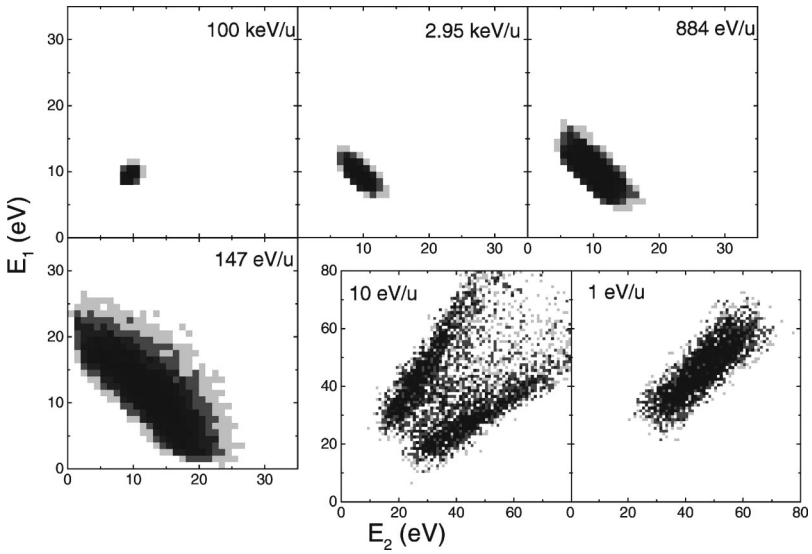

FIG. 6. Correlation of the energy sharing between the deuterons after double electron removal in $Xe^{19+}+D_2$ collisions.

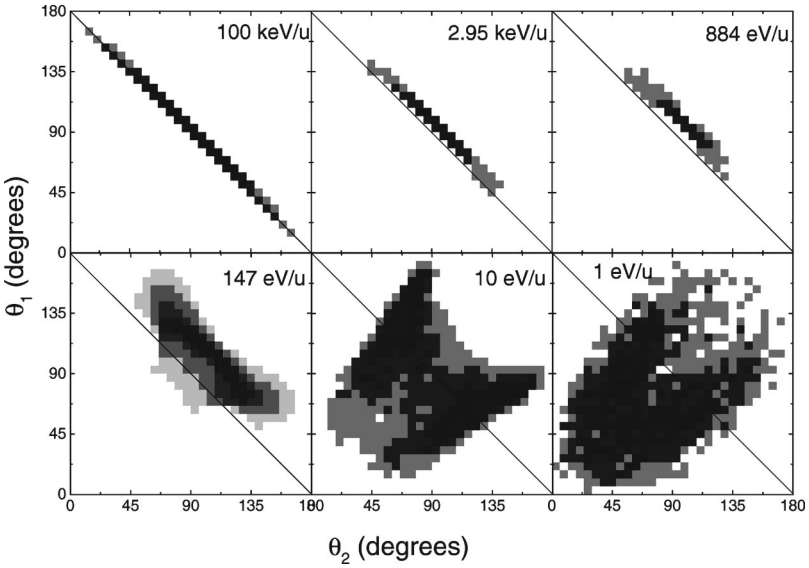

FIG. 7. Correlation of the emission angles of the deuterons after double electron removal in $Xe^{19+} + D_2$ collisions. The diagonal line indicates back-to-back collisions.

To further demonstrate the vector addition of collisional and fragmentational momentum, we have plotted in Fig. 7 the polar emission angle of one D^+ against that of the other. Zero degrees is defined as the initial projectile direction. As before, the initial molecular orientation was randomized at the start of each trajectory. At 100 keV/u, one can see what would be expected of an isolated molecule. The deuterons are emitted back-to-back, that is, on the diagonal line where the sum of the emission angles equals 180°. At the next three energies, this back-to-back emission is still dominant, but the process is accompanied by a slight push in the backward direction. This is due to competition between the energy loss Q, which causes the longitudinal momenta to be shifted by Q/v, and that due to the forward emission of the two electrons. Since most of the deuterons end up in the backward direction, these plots indicate that the repulsive Coulomb force of the projectile acts on them mainly after the collision. This demonstrates that for these impact energies the electrons are not stripped from the molecule until the projectile nears the distance of closest approach. At 10 eV/u, the collisional and fragmentation momenta are approximately equal. The graph for the 10 eV/u collision contains two ''wings.'' These wings basically indicate collisions in which there is a large transfer of energy to the deuterons. Since the projectile has a velocity comparable to the dissociating deuterons, all three heavy particles actively participate in the energy and momentum sharing. The large Coulomb repulsion of the projectile tends to push the deuterons in the same general direction. This is emphasized in the plot for the 1 eV/u collision, in which nearly all of the deuterons are emitted in the forward direction. Since the deuterons are pushed in the forward direction, the electrons are removed from the molecule before the projectile reaches the distance of closest approach.

One can more fully understand the dynamics of the collision by investigating momentum spectra, as shown in Fig. 8. We have used a coordinate frame in which p_x is defined by the final transverse component of the projectile momentum and p_z is the direction of the incident projectile. The projectile's transverse momentum is in the positive p_x direction. For the 100 keV/u collision, the dissociating deuterons behave as expected for a vertical Franck-Condon transition. In such a case, the momenta are symmetric about the origin. A three-dimensional momentum picture would be a spherical

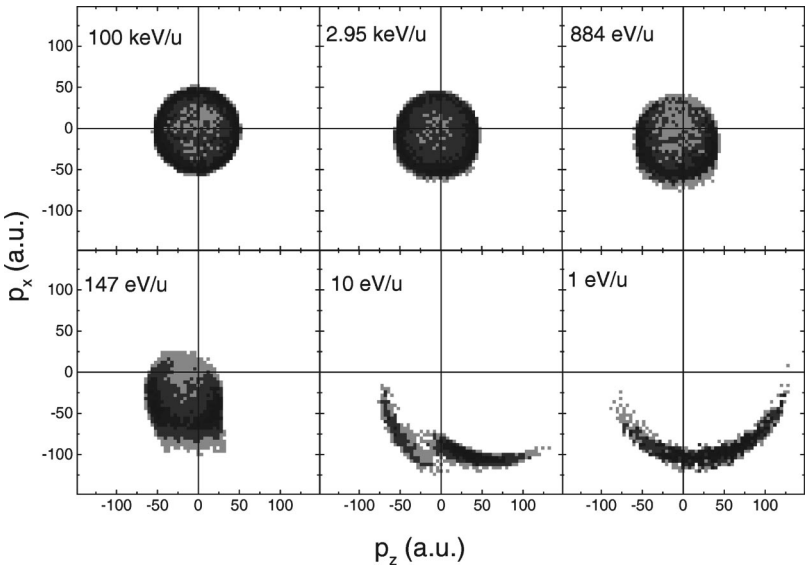

FIG. 8. Momentum spectra of the dissociated deuterons in the collision plane defined by the incident momentum vector p_z, and the transverse momentum vector p_x of the Xe^{19+} projectile.

surface of radius $p=(2mE)^{1/2}\approx 51$ a.u., when $E\approx 9.5$ eV. As the collision energy is lowered, we see a thickening on the lower half of the spherical momentum shell, coupled with a backward shift in the center of the distribution. These are both due to momentum transfer from the projectile. For the two lowest energies, the momentum transfer from the projectile is appreciable and leads to significant deviation from two-body Franck-Condon behavior. In these latter cases, all three heavy particles actively participate in the collisional dynamics and the projectile strongly scatters in the opposite direction of the deuterons.

IV. CONCLUSIONS

Five-body classical trajectory Monte Carlo calculations have been presented for double electron removal from D_2. Examination of final deuteron energies, momenta, and emission angles has shed light on the collisional dynamics. At the highest impact energy (100 keV/u), the molecule dissociates as expected for an isolated molecule Franck-Condon transition. The deuterons are emitted back-to-back with equal energies. At intermediate impact energies (147 eV/u–2.95 keV/u), the Coulomb repulsion between the deuterons and the projectile begins to play an important role. In this energy range, the exploding molecule is pushed in the backward direction and we see unequal energy sharing between the deuterons, as observed by experimental data. At the lowest projectile energies (1 and 10 eV/u), where the velocity of the projectile is comparable to that of the deuterons, the collisional transfer of energy from the projectile dominates the collision dynamics.

ACKNOWLEDGMENTS

This work was supported by the U.S. Department of Energy, Office of Fusion Energy Sciences. We also gratefully acknowledge support from the NSF under Grant No. INT-9723737, and the ''Stichting voor Fundamenteel Onderzoek der Materie'' (FOM).

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