

## Three-body effects in the fragmentation of $D_2$ by slow, highly-charged xenon

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Kinematically complete data and five-body classical trajectory Monte Carlo calculations are presented for fragmentation of  $D_2$  by slow  $Xe^{26+}$  ions. For strongly perturbing collisions, two types of three-body effects are identified. The largest, due to interactions between the undissociated molecule and the dipole field of the projectile, is compatible with a two-step fragmentation picture. A smaller effect, due to quadrupolelike and higher multipole interactions between the dissociating fragments and the projectile field, extracts internal energy from the exploding molecule and is incompatible with a two-step picture.

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Collisional fragmentation of a molecule by a fast charged particle can usually be described as a two-step process: a fast Franck-Condon electronic transition (time scale  $\sim 10^{-16}$  sec) induced during the collision, followed by dissociation of the excited molecular state (time scale  $\sim 10^{-13}$  sec) occurring after the projectile has left the scene. For such a process the projectile plays no role in the breakup dynamics beyond inducing the electronic transition. However, if the projectile is sufficiently slow or the interaction distance is sufficiently large, as is the case for electron capture by slow, highly charged ions, conditions are reached where the collision and dissociation times are comparable. In this case the molecule dissociates in the presence of the strong time-dependent electric field of the projectile, the two-step picture fails, and the final state of the molecular fragments is determined by energy and momentum exchange among all the heavy particles.

The situation is closely related to that encountered in the dissociation of molecules by short, high-field laser pulses where exchange of energy and momentum between the molecule and the radiation field is important during dissociation of the electronically excited molecule [1]. The breakup of a diatomic target by a slow ion also has many features in common with the breakup of triatomic molecular projectiles [2]. Indeed, the study of slow ion-molecule fragmentation dynamics offers another approach to studying the three-body Coulomb problem. However, unlike studies involving triatomic projectiles, slow ion-molecule interactions offer the possibility of altering the three-body, or many-body, dynamics by varying the mass, charge, and velocity of the projectile; we are not limited to studying molecular species that are inherently stable.

This work reports the observation and explanation of strong three-body effects in a kinematically complete study of double-electron capture from  $D_2$  by slow  $Xe^{26+}$  projectiles. Here the final state is particularly simple, with three positively charged ions in the continuum and no free electrons. The primary electronic transition is known to be an overbarrier process [3,4]. From well established model calculations [3] it is known that the second electron is captured when the impact parameter is large (8.2 a.u.) and well outside the  $D_2$  molecule. At this distance, the projectile applies an electric field of  $2 \times 10^{+9}$  V/cm to the molecule. The cap-

ture process produces two  $D^+$  ions, separated by 1.4 a.u., which Coulomb explode. In the absence of the projectile, the explosion is radial with each  $D^+$  ion carrying 9.5 eV of energy, or 45.3 a.u. of momentum, in the laboratory system. Because of the range of internuclear separations in the ground vibrational state of  $D_2$ , a narrow distribution of energies (momenta) results. Departure of the laboratory energy spectra of the  $D^+$  ions from this might then be taken to signal a breakdown of the usual two-step picture.

Recent theoretical studies [5,6] have predicted strong three-body effects in collisions below about 1 keV/u and for conditions where the projectile charge to velocity ratio  $q/v$  is large. Previous experimental attempts to observe three-body effects, involving the participation of the projectile during the dissociation phase, in fragmentation of diatomic molecules have been reported [7–10]. In all of these, only the laboratory momentum of one of the ionic fragments was measured and unambiguous interpretation of the data was difficult. This ambiguity is removed in the present kinematically complete experiment where the momenta of all three outgoing fragments ( $Xe^{+24}$  and two  $D^+$  ions) are determined for each event. Using this technique, we are able to identify two different three-body processes. The dominant process is the transfer of momentum to the center of mass of the  $D_2^{+2}$  system by the action of the dipole field of the projectile, where the multipole expansion is made about the center of mass of the  $D_2^{+2}$  system. Such a process does not change the internal energy of the  $D_2^{+2}$  system; it merely pushes the entire ionized molecule away. By measuring both molecular fragments, we are able to remove this dipole effect by shifting to the center of momentum of the fragmenting molecule. This enables us to identify a second three-body effect resulting from quadrupole and higher multipole components of the field. (Subsequently, use of the term quadrupole is to be understood as including higher multipole components.) Interaction with the quadrupole components is found to govern the extraction of internal energy from the  $D_2^{+2}$  system by the projectile. An important ramification of this is that the dipole effect can be accommodated within a two-step picture while quadrupole effects cannot.

$Xe^{26+}$  ions were extracted from the Kansas State University CRYEBIS ion source and accelerated to energies rang-

ing from 3.9 MeV to 26 keV (approximately 30–0.2 keV/u, Sommerfeld parameters,  $q/v$ , from 24–292 a.u.). As a control, 50-keV protons ( $q/v$  of 0.71) were also used. These ions passed through a recoil-ion spectrometer where they interacted with a jet of  $D_2$ .  $D^+$  and  $D_2^+$  ions were extracted from the interaction region using transverse electric fields from 62 to 250 V/cm. The extraction fields were sufficiently strong to direct all ions onto a large position-sensitive detector. The ions were counted using multihit electronics capable of 0.5-mm position resolution and resolving ions if their arrival times differed by at least 15 ns.

All data shown here required the detection of two  $D^+$  ions, thereby isolating the double capture channel. Projectile ions were charge-state analyzed and detected 2 m downstream from the interaction region. Both  $Xe^{24+}$  and  $Xe^{25+}$  ions were allowed and only the direct beam was rejected. We did not distinguish between radiative and nonradiative stabilization of the excited Xe ion formed in the collision since this is expected to play a little role in the  $D_2^{2+}$  fragmentation. The data were taken in “event mode” and the full momentum vectors of the two  $D^+$  ions were determined from their impact positions and flight times to the recoil-ion detector. This is sufficient to fully determine the three-body final state for each interaction. Further experimental detail will appear in a forthcoming publication.

Theoretical calculations, based on a five-body classical trajectory Monte Carlo (CTMC) model [5,6], were performed using statistics and sorting parameters comparable to those used experimentally. These calculations were used to verify and help interpret the experimental results, as well as to extend outside the range of parameters experimentally accessible. In the theoretical model, the  $D_2$  molecular center is bound by a Morse potential with parameters obtained from spectroscopic observations and the molecule is placed in its ground vibrational state. In the initial channel, all Coulomb interactions are incorporated between the projectile and the four-body target and between the molecular electrons and their parent nuclei. If, during a collision, one target electron is removed and the other is excited to the  $n=2$  level, the Morse interaction is dynamically replaced by Coulomb interactions between all five centers. This simulates the  $D^+ + D^*$  dissociating states that are molecular rydbergs of  $D^+ + D^+$  for  $R < 5$  a.u. and provides the complete description of the final state. The final-state products are tracked in time sufficiently long in order to establish their final energies and spatial distributions.

Figure 1 shows  $x$ - $y$  slices through the center of the Coulomb explosion spheres for three collision systems. (Throughout this paper the  $z$  axis is along the beam and the  $x$  axis is along the extraction field; thus the  $xy$  plane is transverse to the beam direction.) The first column presents the transverse spectra of  $\mathbf{k}_1$  (or  $\mathbf{k}_2$ ), where  $\mathbf{k}_1$  and  $\mathbf{k}_2$  are the transverse momenta of the individual deuterons in the laboratory coordinate system. Data are shown for three systems, ordered according to increasing  $q/v$ : (a) is for the least perturbing projectile studied, 50-keV protons ( $q/v = 0.71$  a.u.); (b) is for an intermediate case, 2-keV/u  $Xe^{26+}$  impact ( $q/v = 92$ ); (c) represents a highly perturbing case, 200-eV/u  $Xe^{26+}$  ( $q/v = 292$ ). As  $q/v$  is increased, the  $\mathbf{k}_1$  (or  $\mathbf{k}_2$ ) spec-

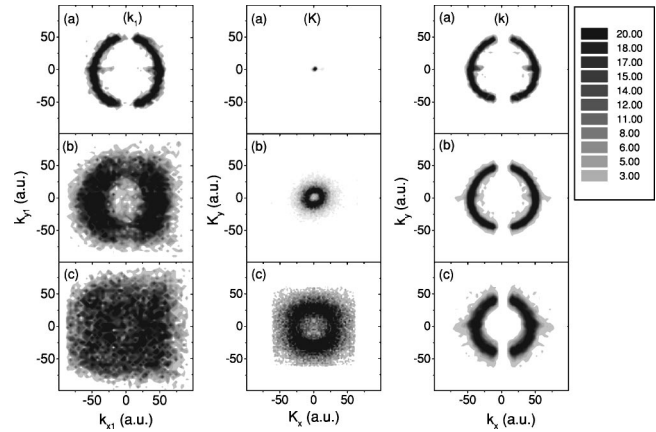


FIG. 1. Momenta, transverse to beam direction, of  $D^+$  fragments following double electron removal from  $D_2$  by 50-keV  $H^+$  impact, row (a), 2-keV/u  $Xe^{26+}$  impact, row (b), and 0.2-keV/u  $Xe^{26+}$  impact, row (c). The first column shows the transverse momenta in the laboratory coordinate system, the middle column shows the momentum transfer to the  $D_2^{2+}$  system, and the last column shows the momentum of  $D^+$  fragments, measured in the center-of-momenta coordinate system. All data shown are for slices through the center of the appropriate three-dimensional momentum sphere, with the  $z$  component being restricted to  $\pm 2$  a.u. from its center value.

tra (column 1) show an evolution from a narrow Coulomb explosion ring of radius near 45 a.u. in momentum to a highly diffuse distribution for slow  $Xe^{26+}$  impact. Similar observations have been reported in other recent experiments [9,10].

Because we measure both  $\mathbf{k}_1$  and  $\mathbf{k}_2$  for each event, we can also plot spectra in the Jacobi coordinates  $\mathbf{K} = (\mathbf{k}_1 + \mathbf{k}_2)/2$  and  $\mathbf{k} = (\mathbf{k}_1 - \mathbf{k}_2)/2$ , shown in the middle and right-hand columns, respectively.  $\mathbf{K}$  is equal to half the center-of-mass momentum of the  $D_2^{2+}$  molecule after the collision, while  $\mathbf{k}$  describes the internal or relative motion of the two deuterons as measured in the center-of-mass system of the  $D_2^{2+}$  molecule. The major capture and dissociation processes become transparent when the data are presented in these coordinates.

The  $\mathbf{K}$  spectra form annuli in momentum space with radii given by the transverse momentum imparted to the  $D_2^{2+}$  system as a result of the double electron transfer process. These spectra demonstrate that for fast proton impact, virtually no transverse momentum is transferred, whereas for slow  $Xe^{26+}$  impact nearly 30 a.u. is transferred. In the  $z$  direction (not shown), the  $\mathbf{K}$  spectra are centered at  $-(Q/v - v)$ , where  $Q$  is the electronic energy change in the capture reaction and  $v$  is the collision velocity. For our data the transverse momenta and values extracted for  $Q$  are consistent with predictions of the overbarrier model of Niehaus [3] for double electron capture.

The  $\mathbf{k}$  spectra reveal directly the internal energy distributions of the  $D_2^{2+}$  molecule and how it is influenced by the time-dependent strong electric field of the slow, highly charged projectile. As seen in the third column, the internal energy remains remarkably stable as  $q/v$  increases by more than two orders of magnitude. Thus, columns 2 and 3 show

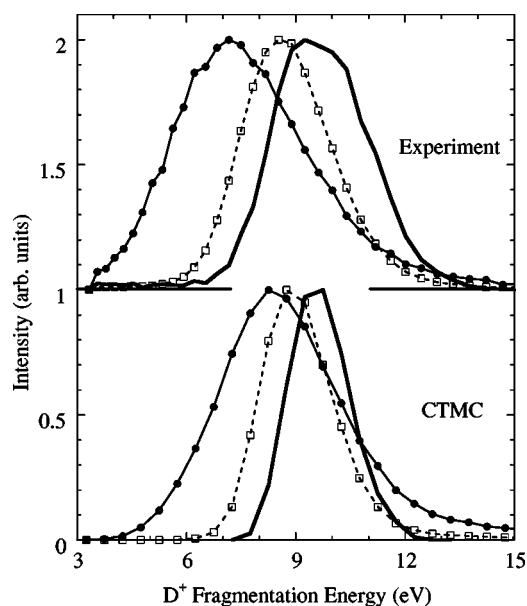


FIG. 2. Kinetic energies for  $D^+$  fragments following double electron removal from  $D_2$ . Energies are in the center-of-mass coordinate system of the exploding molecule. All spectra are normalized to unity at their maxima. Upper portion, experimental data; lower portion, CTMC calculations; solid curve, 50-keV  $H^+$ ; dashed curve with open squares, 0.8-keV/u  $Xe^{26+}$ ; solid curve with filled circles, 0.2-keV/u  $Xe^{26+}$ .

that the major broadening effect observed in the laboratory spectra (column 1) is due simply to the vector addition of the recoil velocity of the doubly charged molecule and the “normal” Coulomb explosion velocity of each fragment ion. This vector addition is in accordance with CTMC predictions [6] and agrees with the conclusions of Fremont *et al.* [10]. The two-step picture seems to hold over a wide range in collision strength.

Closer examination, however, of the  $k$  spectra reveals the onset of a true three-body effect at the lowest collision velocities. Looking closely at column 3, note that the radius of the Coulomb ring becomes smaller as  $v$  is lowered. This is shown more clearly in Fig. 2, where we plot the radial distribution of  $D^+$  kinetic energies ( $k^2/2$ ) in the  $D_2^{2+}$  center-of-mass system for various collisions. The vertical scales have been normalized to unity for display purposes. For  $Xe^{26+}$  collision energies below 10 keV/u, a systematic decrease in the fragmentation energy distribution as well as increases in the width of the distribution were found for increasing values of  $q/v$ .

We attribute these effects to the action of the quadrupole components of the projectile electric field on the  $D_2^{2+}$  system. While the dipole component acts equally on both  $D^+$  ions and thus transmits momentum to the whole molecule, the quadrupole components act differently on the two fragments, depending on the locations of the fragment ions relative to the projectile. As the collision velocity is lowered, there is more time for the  $D^+$  ions to separate from their initial positions while the projectile field is still present. Thus the quadrupole effect becomes greater. For the present case, where capture occurs far outside the mean radius of the  $D_2$

molecule, the quadrupole effect removes internal energy from the molecule during the collision. This occurs because the  $D^+$  ion nearer the projectile is repelled more than the distant one, similar to a tidal process. Since the explosion motion is outward, internal energy is removed.

An explicit evaluation of this effect provided by the CTMC model is also shown in Fig. 2. Good qualitative agreement with experiment is seen. For the weakly perturbing proton collision, and indeed also for  $Xe^{26+}$  impact down to 3 keV/u, no quadrupole effects are observed. For stronger interactions, the spectra systematically shift and broaden. Quantitative differences in the widths of the experimental and theoretical distributions are partially attributed to experimental effects associated with a nonlocalized target. Likewise, the smaller energy shift predicted at the lowest impact energy may also be due to experimental uncertainties, namely in knowing the exact potential where the highly charged ions are created and, hence, the exact beam energy. Fremont *et al.* [10] have reported similar non-two-step effects for a lower-charge-state projectile for which the capture is at smaller impact parameter.

To summarize, kinematically complete data are presented for fragmentation of  $D_2$  following double electron removal by slow, highly charged xenon ions. For highly perturbative interactions, higher and lower momentum fragments than normally observed for fast ion, electron, or photon impact indicate the presence of three-body effects. The data clearly demonstrate that the major three-body effect is due to a two-step process where momentum is transferred to the entire molecule, which then fragments. In addition, at the lowest impact energies we were able to distinguish that another, “true,” three-body effect is also active. This effect influences the internal energy of the dissociating molecule and is attributed to interactions involving quadrupole components of the projectile electric field. We note that changes in the kinetic energy release in slow  $O^{7+}$ -CO collisions were also observed in a recent study by Tarisien *et al.* [11], but they observed energy increases that depended on the alignment of the molecule. This is in contrast to the present work where the internal energy decreased and, within experimental resolution, was independent of the molecular alignment.

It is interesting that the two-step process remains active under conditions where the perturbation by the projectile is so large, i.e., for  $q/v \approx 100$ , and when the collision time greatly exceeds the fragmentation time. Under these circumstances one would expect strong mutual interplay among the ionic fragments and the highly charged projectile. In retrospect, the lack of such interplay may result precisely because the projectile charge is so large. For slow, highly charged projectiles, electrons are captured when the interaction distance is large. Thus, the molecule appears as a point particle rather than two individual deuterons. This would imply that the breakdown of the two-step picture might better be observed by using low-charge-state projectiles for which the interaction distance could be made more comparable to the internuclear separation of the molecular components. As for the “true” three-body effects, these offer unique opportunities for systematically studying multibody interaction processes by varying the relative forces and masses as well as

the number of interacting particles. However, the present work demonstrates that it is essential that these investigations be made in the center-of-momentum system. Otherwise, the information sought will be completely masked.

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