Interference effects in double ionization of spatially aligned hydrogen molecules by fast highly charged ions

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(Received 28 May 2004; published 7 October 2004)

Cross sections differential in target orientation angle were measured for 19 MeV $F^{8+}+D_2$ collisions. Multihit position-sensitive detectors were used to isolate the double-ionization channel and determine *a posteriori* the full momentum vectors of both ejected D^+ fragments. A strong dependence of the double ionization cross section on the angle between the incident ion direction and the target molecular axis is observed with a $\approx 3.5:1$ enhancement for molecules aligned perpendicular to the projectile axis. This clear asymmetry is attributed to interference effects, analogous to Young's two-slit experiment, arising from coherent contributions to the ionization from both atomic centers. The data are compared to a simple scattering model based on two center interference.

DOI: 10.1103/PhysRevA.70.042702 PACS number(s): 34.50.Gb

I. INTRODUCTION

A fundamental result of quantum mechanics is the wavelike behavior of particles. The classic manifestation of this behavior is the interference between two particle waves (or more appropriately, a particle and itself) in the Young's two-slit experiment [1]. As early as 1966, Cohen and Fano pointed out that a simple homonuclear diatomic molecule provides a natural two-slit system for electron emission [2]. Recently, this concept has been used to explain some aspects of ionization of diatomic molecules by intense lasers, specifically that destructive interference of the outgoing electron waves from the two molecular centers suppresses ionization in some cases [3]. The same mechanism has been proposed as an ultrafast probe of molecular motion [4,5]. Similar strong fields are present in the interaction between a fast ion and a molecule, and a variety of phenomena attributed to interference in ion-molecule collisions might be expected [6,7]. However, until recently [8–11], there had been no direct evidence for interference effects in the ionization of a molecule by ion impact or other strong fields.

Interference effects have, however, been studied in the single photoionization of H_2 [2,12,13]. In this case, the electron is ejected via a dipole transition from both centers simultaneously. For ionization by ion impact, interference effects are more difficult to observe for two reasons: (1) The momentum (or energy) transfer from the projectile can vary over a wide range and is in general unknown, washing out any interference effect in the total cross section. In principle, this can be overcome with differential measurements where, for example, the momentum/energy transfer or molecule orientation is known. Furthermore, (2) the transition of the electron from the bound molecular state to the continuum is a many-body process. Therefore, not only can the energy transferred from the projectile vary, but so can the distribution of this energy to the constituent target nuclei and electrons. Furthermore, this distribution of energy is frequently classified as resulting from either binary or dipole mechanisms [8,9,14–16], and although both mechanisms may contribute to the total ionization yield, it is only the dipolelike transitions that contribute to the interference [8,17,18].

Because of these difficulties, explorations of interference effects in collisions with H_2 and H_2^+ have historically focused on electron capture [7,19–23] and related transfer ionization [6,24] channels. Here the captured electron undergoes a discrete transition that circumvents the difficulties discussed above. Only recently did electron spectroscopy measurements by Stolterfoht *et al.* [8] show evidence for interference effects in electron emission from H_2 produced by ionizing collisions. Two features of that work helped overcome the difficulties described above. First, the fast, highly charged (60 MeV/u Kr^{34+}) projectiles used were well approximated by a field of virtual photons [14–16]. Therefore, since the outgoing electron energy was measured, the effective energy of the virtual photon was known, overcoming factor (1) above. Second, the electron emission was measured at forward and backward angles far from 90° [8,16]; in these regions the dipole mechanism dominates, overcoming factor (2). Follow-up experiments have extended these measurements to include different projectiles [10] and examination of dependence on the electron emission angle [9]. Very recent results have even refined the technique enough to reveal second-order interference effects [11].

In the present article we report a strong asymmetry in the double-ionization cross section differential in the precollision angle of otherwise randomly oriented D_2 molecules. Using a momentum imaging technique, rather than electron spectros-

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copy, we have measured the emission angles of ejected D^+ fragments following collisions with 19 MeV F^{8+} ions. In this case, we have restricted the energy of one of the ejected electrons to less than 100 eV so that the dipole contribution to the ionization cross section is dominant (as verified by a simple Born-approximation calculation). The observed double ionization cross section is found to be $\approx$3.5 times stronger for molecules oriented perpendicular to the ion beam direction than for those parallel to the beam. We interpret this asymmetry as further evidence for interference effects in the ionization of hydrogen molecules.

A characteristic of the electron spectroscopy work [8–11] is that the evidence for interference in the electron spectra is subtle, and oscillations arising from the interference effects are identified only with the help of significant theoretical efforts [8,18,25–27]. In contrast, once the double ionization channel is isolated, the asymmetric angular distribution presented here is easily visible in the raw data. Furthermore, a simple scattering picture based on two-center interference agrees nicely with our experimental results.

II. EXPERIMENTAL APPROACH

Our data were obtained using cold target recoil ion momentum spectroscopy (COLTRIMS), which by now is a well established experimental technique [28–30]. A beam of 19 MeV F^{8+} ions from the KSU tandem Van de Graaff accelerator intersected a beam of D_2 emerging from a two stage supersonic gas jet. The resulting recoiling target ions and electrons were extracted by a static transverse electric field and detected by 80 mm diameter position sensitive detectors equipped with multihit delay-line anodes [31]. A magnetic field applied parallel to the electric field radially confined the electrons to a helical trajectory, resulting in 4π collection efficiency restricted to energies below 100 eV. The electron time-of-flight was measured relative to the bunched projectile beam, and the longer recoil flight times were, in turn, associated with a particular electron using event mode data collection. We isolated the double ionization channel from the more probable single ionization and ionization-excitation events [32] by requiring a triple coincidence between both molecule fragments and one electron ($D^+ + D^+ + e^-$). Since only one of the two continuum electrons is measured, only the energy of this electron is restricted to less than 100 eV, while the second continuum electron may have any value. Transfer ionization, a competing channel that also results in two recoil ions and an electron reaching the continuum, is much less likely than double ionization at these projectile velocities and does not contribute significantly to the present result [22].

From the position and time-of-flight information recorded, the complete coincident vector momenta of one electron and both D^+ fragments ions are reconstructed. Both recoil ions are used to calculate the final fragment momentum in the center-of-mass frame. Because the collision and dissociation times are much shorter than the rotation time of the molecule, the molecule fragments are ejected along the precollision internuclear axis. Therefore, the alignment of the molecule can be inferred from the fragment momenta measurement.

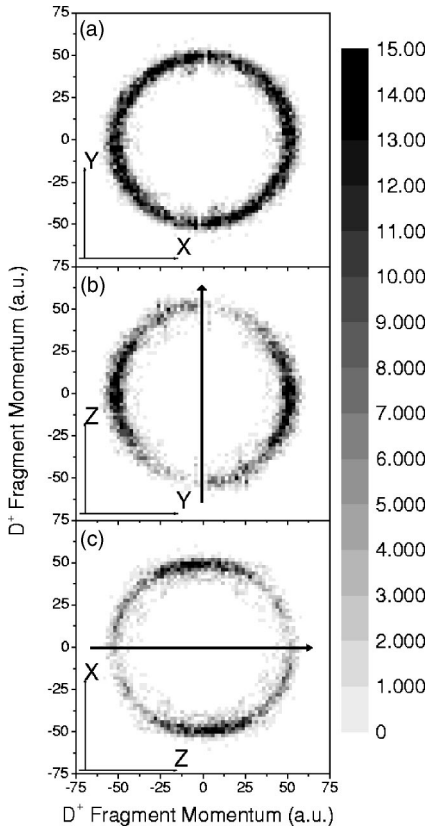

FIG. 1. Fragment momentum distributions in the laboratory frame. The ion beam is propagating in the z direction. Slices of the momentum distributions are presented along three planes: (a) P_x vs P_y, corresponding to the extraction field vs gas jet directions, (b) P_z vs P_y, corresponding to the ion beam vs gas jet directions, and (c) P_z vs P_x, corresponding to the ion beam vs extraction field directions. The long arrow through each ring represents the ion-beam direction, making clear the fragment distributions peaked perpendicular to the beam.

III. RESULTS AND DISCUSSION

The resulting momentum distributions in the laboratory frame are shown in Fig. 1. In each frame, the momentum of the D^+ fragments is shown projected into each of the three planes defined by the experiment geometry, where X, Y, and Z correspond to the field, gas-jet, and ion-beam axes, respectively. Atomic units are used throughout unless stated otherwise. In each case the momentum vectors shown are restricted to 17.4° from the associated plane. The P_x vs P_y distribution reflects the cylindrical symmetry of the collision system. Furthermore, since P_x is measured via time-of-flight and P_y via fragment position, this symmetry provides a check for the correct conversion to momentum space along both axes. However, the P_z vs P_y and P_z vs P_x distributions show a strong angular anisotropy with respect to the beam axis. This result is reflected in both slices, evidence that the effect is due to the physics of the interaction and not an instrumental artifact.

In Fig. 2, we plot the measured differential cross section for double ionization as a function of $\cos(\theta)$, where θ is the angle between the molecular axis and the projectile beam.

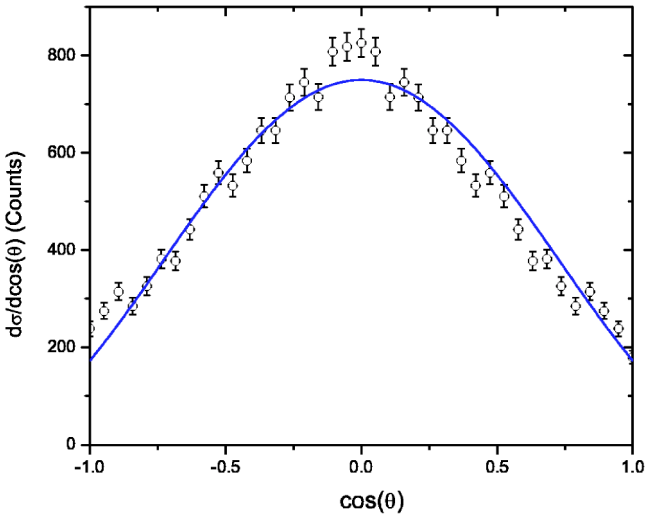

FIG. 2. (Color online) Cross section for double ionization as a function of $\cos(\theta)$, where θ is the angle between the molecular axis and the ion beam direction. The solid curve is the fit to the data as described in the text.

The cross section is about a factor of 3.5 higher at $\cos(\theta)=0$ than at $\cos(\theta)=1$, indicating a strong preference for double ionization to occur for molecules at 90° to the beam direction. The solid curve is a fit based on a simple interference model discussed later in the text.

The strong asymmetry evident in Figs. 1 and 2 is particularly interesting in that the cross section maximizes for molecules oriented perpendicular to the ion beam. This is contrary to similar past measurements and predictions. Multiple ionization of diatomic molecules does indeed frequently depend on θ, as shown by Lutz and co-workers in a series of papers focusing on ionization resulting in multiply charged fragments [33]. Their results, however, show that for some values of projectile Z/v_p, certain multiple ionization channels of N_2 and CO are peaked near $\cos(\theta)=\pm 1.0$. This is explained in the context of a statistical energy deposition model. Our results are qualitatively different, since they are peaked at $\cos(\theta)=0$ and the D_2 targets used in this work have far fewer electrons. The energy deposition model is likely better suited to molecules with multiple shells, where the ionization of many electrons happens at small impact parameters and can be attributed in part to relaxation of core vacancies via Auger decay. Furthermore, classical calculations by Wood and Olson predict a nearly isotropic dependence for the double-ionization of H_2 by a variety of projectiles [34]. These considerations suggest that the present effect might be due to the quantum-mechanical nature of the ejected electrons, resulting in interference.

There are other reasons to attribute our results to an interference effect. As has been pointed out previously [8], interference effects arise primarily from the dipole (or three body) part of the collision. Our COLTRIMS apparatus is well-suited to detection of the relatively low energy electrons that are predominately produced via dipole, or photoionizationlike collisions.

The work of Cheng *et al.* [6] examined the role of molecular alignment in double ionization and ionization excitation of D_2 by 2–16 MeV O^{8+}, finding little dependence. They did, however, observe that the cross sections for transfer excitation and transfer ionization depended on the molecular alignment. These results were interpreted using the arguments developed by Wang and McGuire [23,24] for electron capture from H_2. In this single-electron process, the amplitude for capture is the sum of the amplitudes for capture from two hydrogen atoms, to be added with relative phase $e^{i\vec{q}\cdot\vec{R}}$, where $\vec{q}$ is the projectile momentum transfer and $\vec{R}$ is the internuclear position vector. As the projectile scattering angle was not measured, the experiment and calculation both integrate over the transverse component of $\vec{q}$. The longitudinal component of $\vec{q}$ (q_z) remains, and the factor $e^{iq_z\cdot R_z}$ gives rise to the interference term. For capture,

$$q_z = \frac{Q}{v_p} - \frac{v_p}{2}, \tag{1}$$

where Q is electronic energy transfer of the reaction. For ionization

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

where ϵ_I is the binding energy of the target electron and $\vec{k}$ is the momentum of the continuum electron.

In the case of capture Q is fixed (and therefore q_z is as well), since the electron transfers from a single initial state to a single final state. Thus, Cheng *et al.* concluded that the interference was constructive for q_z perpendicular to $\vec{R}$, but not for any other angle, explaining their results for two electron processes in which one electron is captured by the projectile.

The nearly isotropic measurements of the cross section for double ionization of D_2 made by Cheng and co-workers in the same work [6] was explained, at the time, as a consequence of the range of Q values inherent in an electron transition to the continuum washing out any interference. We note, however, that they were experimentally unable to separate the dominant ionization-excitation [32] channel from double ionization. Given the strong dependence of the double ionization cross section on molecular alignment in our results, where the double ionization channel can be cleanly isolated, it seems plausible that a more correct explanation of the earlier data would be that Q and q_z were, on average, too small to produce interference effects. For double ionization, which is isolated in our experiments, Q can easily approach 100–200 eV when one adds the Franck-Condon double ionization energy to the sum energy of the two continuum electrons. This value of Q, with $v_p=6.3$, results in q_z of about 1.0–1.1 atomic units, and $q_z\cdot\vec{R}$ of order unity. This is large enough to result in considerable destructive interference of amplitudes for $\vec{R}$ oriented along q_z.

We may model our situation as in the work of Bräuning *et al.* [20], in which they measured charge transfer in collisions of H_2^+ with He^{2+} and Ar^{2+}, where they adapted the result of Shingal and Lin [35]. Here it is assumed that (i) the collision time is much smaller than the rotational period of the molecule, which at $v_p=6.32$, is satisfied. (ii) Electron capture by

the projectile is negligible [22]. (iii) The wave function for D_2 is approximately the same as that for two D atoms separated by the internuclear distance R (=1.4 for D_2) [36]. By representing the one-electron H_2^+ wave function as a linear combination of atomic orbitals

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

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

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

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

$$P(\theta,\vec{b}_A,\vec{b}_B) = \frac{1}{2}|a(\vec{b}_A)|^2 + |a(\vec{b}_B)|^2 + 2\,\mathrm{Re}\{a(\vec{b}_A)a(\vec{b}_B)\cos[q_Z R\cos(\theta)]\}. \tag{5}$$

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

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

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

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

Note that $d\sigma_a$ is a maximum at θ=90 deg, regardless of the value of q_Z! If q_Z is roughly constant, we integrate $d\sigma_a$ to get

$$\sigma(\theta) = \sigma_a\{1 + \cos[q_Z R\cos(\theta)]\}. \tag{9}$$

Now we can extend the model to two electrons. In our experiment Z/v_p=1.3, and double ionization via two independent electron-projectile interactions is likely [32]. Therefore, the independent, successive ionizations are treated as a product of probabilities

$$d\sigma(\theta) \approx \int P(\theta,\vec{k}^{(1)})\{1 + \cos[q_Z^{(1)}R\cos(\theta)]\} \times P(\theta,\vec{k}^{(2)})\{1 + \cos[q_Z^{(2)}R\cos(\theta)]db\}, \tag{10}$$

where (1) and (2) denote the first and second ionization events and q_Z is related to $\vec{k}$ by Eq. (2). If we take an average value $\overline{q_Z}$, then integration over $\vec{k}^{(1)}$ and $\vec{k}^{(2)}$ yields

$$\sigma(\theta) = \sigma_{\mathrm{atomic}}\{1 + \cos[\overline{q_Z}R\cos(\theta)]^2\}. \tag{11}$$

The solid curve in Fig. 2 is a fit to the data with Eq. (11). Using the longitudinal momentum as a free parameter, we obtain a value of $\overline{q_Z}$=1.1 for the fit. Using Eq. (2), this momentum transfer corresponds to a total average energy transfer well in excess of 100 eV for each of the successive ionization events. This surprisingly large number suggests that while the above model qualitatively describes the data quite well, better quantitative calculations are necessary for a full understanding of the present results.

IV. SUMMARY

We observe a strong dependence on molecular alignment in the cross section for double ionization of D_2 by fast, highly charged ions. We attribute these results, which are apparent in the unmodified raw triple-coincidence data, to double-slit type interference effects. Our results complement earlier electron spectroscopy measurements, which also showed interference effects due to the two center nature of the target. Those data, however, required considerably more detailed theoretical interpretation than the results presented here. The data were in qualitative agreement with a simple two-center interference model.

ACKNOWLEDGMENTS

We gratefully acknowledge fruitful discussions with N. Stolterfoht and F. Robicheaux and technical assistance from I. Ali. We thank P. Richard and the J.R. Macdonald Laboratory staff for their assistance with the apparatus development and assembly. This work was supported by the Chemical Sciences, Geosciences and Biosciences Division, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy. E.W. received additional support from the Augustana Research/Artist Fund.

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