Angular distribution of dissociated deuterons by impact of 2–16-MeV O^{8+}

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The angular distributions of dissociated deuterons for electron capture and ionization by bare oxygen ions with energies from 2 to 16 MeV in collision with molecular deuterium targets were measured. The dependence of the differential cross sections on the alignment of the molecular axis with respect to the beam axis was determined. The results show that in the transfer ionization and transfer excitation processes, the deuterium molecules are more likely to be aligned perpendicular to the incident beam than parallel to the beam. This feature can be qualitatively interpreted as resulting from the interference of capture amplitudes from the two atomic centers. In the double ionization and ionization excitation processes, little alignment dependence was observed.

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INTRODUCTION

The anisotropic behavior of electron impact on hydrogen molecules was recognized as early as 1935 [1,2]. It was realized that the angle between projectile electron and molecular axis can have a significant effect on quantities such as the cross sections and energy distributions of the dissociated ions. While this influence for electron impact was fairly well understood both theoretically and experimentally [3–5] by the 1960s, the same phenomenon for heavy-ion impact has received much less attention. In 1960, Tuan and Gerjuoy [6] first theoretically studied electron capture from hydrogen molecules and found that the cross section could be formulated so that the scattering amplitude could be written as a coherent sum of amplitudes for capture from the separate hydrogen-atom scattering centers. They found the relative phase of the amplitudes from the two centers to be given by $e^{-i\alpha\cdot\rho}$, where α is the projectile momentum change during the collision, and ρ is the internuclear position vector. When the molecular alignment or the projectile energy changes, this phase factor changes accordingly. Unfortunately, they did not study the differential cross-section dependence on the molecular alignment, but instead averaged over the alignment of the molecular axis. As a consequence, the dependence of the interference on the alignment of the molecule was not readily apparent. However, they did notice that the interference after the averaging generally does not cancel out. Band and Ray and Saha subsequently calculated cross sections for capture from H_2, but again averaged over molecular alignments [7–9].

In 1988 Deb, Jain, and McGuire [10], adapting the interference idea from Tuan and Gerjuoy, calculated the differential cross section for electron capture by fast protons from H_2 molecules at fixed molecular alignments as a function of the projectile scattering angle, and found an interference structure similar to the "classical" Young two-slit interference pattern. They also illustrated how this interference pattern varies with the alignment of the target molecule and with the velocity of the projectile. It would be a direct experimental test of this interference phenomenon if the differential cross sections at a fixed molecular alignment could be measured, but the scattering angles are quite small and the alignment difficult, and no experimental result of this type has been reported. Using an atomic orbital basis, Shingal and Lin [11] calculated capture from H_2 by 1–500 keV/amu H^+ and He^{2+} as a function of internuclear axis alignment and found strong interference effects which did not disappear in the total cross section. Kimura, Chapman, and Lane [12] have calculated alignment dependences in a molecular orbital basis for H^+ and Ar^+ capturing from H_2. Recently, Wang and co-workers [13,14] reformulated the problem and calculated the differential cross section for electron capture by protons and bare oxygen nuclei from H_2 as a function of the projectile energy and the molecular alignment. By assuming that the independent electron model is valid for a two-electron process, they were able to argue that their single capture theory could be compared with experimental results for electron capture accompanied by ionization or excitation.

On the experimental side, measurements of the dependence of double ionization and ionization excitation of H_2 on the alignment have been reported by Ezell *et al.* [15] and Edwards *et al.* [15] for proton projectiles between 0.5 and 2 MeV. For double ionization, nearly isotropic distributions were found, while for ionization excitation the variation in yield with internuclear axis angle was typically less than 20%. Alignment dependences were reported by Yousif and co-workers [16] for protons between 5 and 30 keV; they found nearly isotropic alignment distributions in the two-proton final channels for all these energies. Capture was not separated from ionization in this experiment. No experiment has been reported previously (to our knowledge) on the dependence of capture alone on the alignment of the internuclear axis, or on the projectile angular distribution at fixed molecular alignment, either of which might reveal features of the interference effect proposed by Tuan and Gerjuoy. In this paper, we describe an experiment which addresses this effect by measuring the dependence of capture by fast

O^{8+} ions, accompanied by incidental ionization or excitation of the other electron, on the alignment angle of the molecular deuterium molecules.

According to our earlier experimental results [17], when 8–20-MeV O^{8+} ions collide with D_2 molecules and capture one electron from the molecule, there is a substantial probability that the molecular ions are either left in the doubly ionized state (for transfer ionization) or left in one of the electronically excited $2p\sigma_u$, $2s\sigma_g$, and $2p\pi_u$ states (for transfer excitation). The molecular ions in such states are not stable and subsequently dissociate into ion-ion or neutral-atom–ion pairs. Since the collision time and the dissociation times are both small compared to the rotational time of the molecule, the dissociation fragments dissociate along the direction which the intermolecular axis had prior to the collision. By measuring the distribution of these fragments with respect to the beam axis in coincidence with charge-state-selected outgoing projectiles we can deduce the dependence of the selected collision process on the initial angle of the molecular axis. In the experiment we also measured the corresponding angle dependence for ionization excitation and double ionization, for which little alignment dependence is expected.

The general discussion in the following applies to the projectiles of energies between 2 and 16 MeV. Where numerical values are cited, the projectile energy of 10 MeV is assumed unless otherwise specified.

EXPERIMENT

The experimental apparatus is shown in Fig. 1. A 2–16-MeV oxygen beam from the 6-MeV Tandem Van de Graaff accelerator at Kansas State University was poststripped by a carbon foil to produce bare oxygen ions. The incoming O^{8+} beam was collimated to less than 1 mm by two successive sets of four-jaw slits. Before the ion beam collided with the target, an upstream magnet was used to clean the beam so the O^{7+} ions produced along the beamline by capture of an electron from the background gas were deflected away from the entrance slits. A turbomolecular pump was mounted immediately after this cleaning magnet to maintain a good vacuum in order to reduce the charge impurity. In the collision region, D_2 gas of a few mTorr pressure was blown towards the detector at right angles to and 1.2 mm away from the beam from a gas jet collimated by a glass capillary array in order to provide a gas flow with good directionality and target density. The D^+ and D_2^+ ions produced were extracted by a very uniform electric field established perpendicular to the beam and sent onto the face of a two-dimensional position-sensitive resistive-anode detector (2DPSD) which was mounted at 90° to the beam direction. Because the collision center was localized at the crossing of the ion beam with the jet, measurement of the position with which each fragment struck the 2DPSD and of its flight time to the detector was sufficient to allow the reconstruction of all three components of the initial fragment momentum. Thus both the energy and angle of the fragment could be calculated.

After passing through the collision region, the beam was deflected by an analyzing magnet to separate O^{8+} from O^{7+} and was detected by a position-sensitive backgammon-anode detector (PSD). During all runs involving the electron capture process, the O^{8+} ions after the analyzing magnet were moved off the PSD and only the O^{7+} ions struck the PSD, thus allowing a more intense beam to be used.

In order to eliminate events produced from interaction of the beam with the wings of the gas jet, an aperture of 6.35 mm diameter was placed between the jet and the detector to restrict its view to the central collision region. The voltage V_2 on this plate, and the overall extraction voltage V_1 (see Fig. 1) were selected to produce an electric field high enough to collect all ions from the dissociation process onto the 40-mm-diameter 2DPSD. The value of V_2 was chosen to minimize distortion effects in the region of the collision center. Such distortion effects could be evaluated on the basis of the departure from spherical symmetry of the velocity space distribution of the dissociation fragments. For our geometry, where the 2DPSD was mounted 41 mm away from the beam and the extraction region was 20.8 mm long, the optimized pusher and aperture voltages were $V_1=491$ V, and $V_2=453$ V. The position information (y_r,z_r) where the recoils hit the 2DPSD was decoded from a resistive anode by charge division.

The velocity components v_y,v_z of the recoils were calculated by $v_y=y_r/t$ and $v_z=z_r/t$, where t is the time of

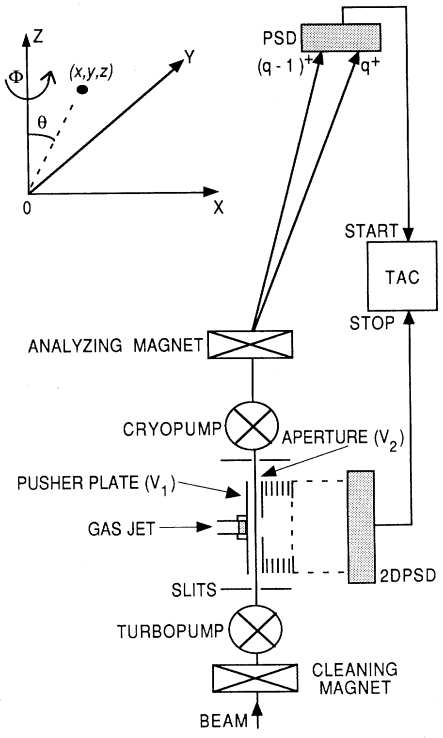

FIG. 1. Schematic of apparatus used to study the molecular alignment for the O^{8+} on D_2 collision system. The z axis is along the beam, the x axis towards the recoil detector, and the y axis, directly into the paper. TAC denotes a time-to-amplitude converter.

flight (TOF) of the recoils. The v_x velocity component was calculated by a more complicated formula, including the pusher voltages and the pusher geometry as parameters, and is implicitly a function of t. A table look-up method was used to find v_x for each t to ensure both the accuracy and the processing speed.

Once v_x, v_y, an v_z were known, it was straightforward to convert them to the equivalent v, θ, and ϕ (see Fig. 1 for the definition of θ and ϕ):

$$v=(v_x^2+v_y^2+v_z^2)^{1/2} , \tag{1}$$

$$\theta=\cos^{-1}\left[\frac{v_z}{v}\right] , \tag{2}$$

$$\phi=\cos^{-1}\left[\frac{v_x}{(v_x^2+v_y^2)^{1/2}}\right] , \tag{3}$$

and the recoil energy

$$E_k=\tfrac{1}{2}mv^2 . \tag{4}$$

In Fig. 2 we show an isometric display of events versus vector velocity, where the velocity distribution is projected onto the yz plane. These data are for electron capture plus ionization or excitation for 10-MeV O^{8+} on D_2. It is immediately apparent from this figure that more events occur when the molecule is perpendicular to the beam than when it is parallel to it.

Before arriving at final angular distributions, it was necessary to take into account that the gas target jet was not an ideal point source but rather extended slightly along the beam. This not only spread the image slightly in the yz plane, but, through, the presence of the aperture, influenced the angular distribution by causing a nonuniform event detection efficiency along the beam direction. Because the full width at half maximum (FWHM) of the jet profile itself was 2 mm, a substantial fraction of the 6.35-mm-diameter of the aperture, a nonnegligible fraction of collisions occurred near the aperture periphery. The dissociation fragments from these collisions have a substantial chance to be blocked by the aperture, and this chance is greater for explosions parallel to the beam than for those perpendicular to it.

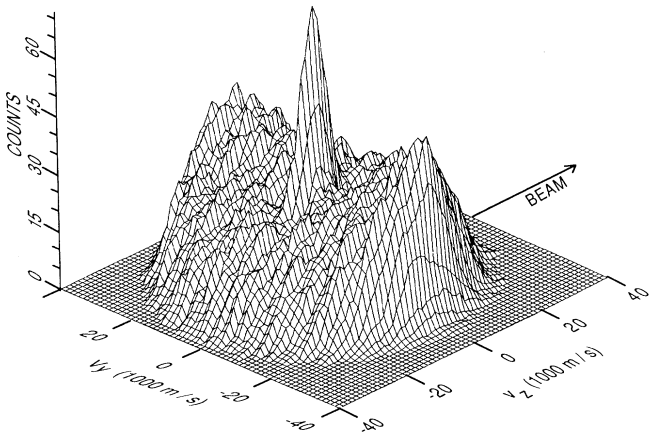

FIG. 2. The yz projection of a three-dimensional velocity spectrum of recoiled D^+ for electron capture accompanied with ionization or excitation. The central peak is the D_2^+ ions. The outer ring is the D^+ ions. It is apparent there are more D^+ ions in the direction perpendicular to the beam axis (v_x axis).

These two influences on the distributions of θ and ϕ were closely related. Their importance was quantitatively evaluated and corrected for using a computer simulation procedure, whereby a stream of isotropically distributed dissociation events was randomly generated with the jet profile and beam geometry as source distribution. The trajectories of the dissociation events were followed numerically through the geometry of the experimental apparatus, including the aperture block, to find their arrival locations on the 2DPSD. These computer generated events were then sorted by the same data acquisition program which was used to evaluate the actual data. Any departure from isotropy which appeared in the resulting θ and ϕ distributions was then labeled instrumental. The departure from isotropy, typically less than 15%, was then removed from the data by multiplying the data by the same function which would have been necessary to return the simulation results to isotropy. This procedure is not rigorously valid, but was found to be quite adequate for the relatively small correction needed here.

The recoil detector used to record the (y,z) position information of the recoil ions was tested carefully to extract key parameters such as spatial resolution, linearity, overall efficiency, and so on. The overall efficiency across the detector anode surface was found to be flat with a 5% error bar, the spatial resolution was about 0.11 mm, and the angular resolution was about 2.5°. The linearity was checked by comparing a detected image against a well-defined pattern placed in front of the detector.

Additional corrections made before arriving at the final distributions included a correction for random coincidences, a correction for double collisions, and a correction for detecting two recoils within the resolving time. The correction for double collisions is necessary because ions can capture an electron from the residual or target gas in one event and, in a separate collision, ionize or excite a deuterium molecule, thus producing a charge changed projectile which appears in coincidence with a deuteron produced by dissociation which is indistinguishable from the true two-electron process which would produce the same products. Contributions from this double scattering were evaluated as described in Ref. [17] and subtracted from the data. The correction for the detection of two recoils in one event is required because the resistive anode is a slow device. The $D^+ + D^+$ breakup in the intermolecular direction near the beam direction produces two deuterons which have very similar flight times. In this case, the amplifier used for the position signal cannot generate an output without a pileup, and the position information is rendered false. The size of this effect was again evaluated using the simulation program, and the corresponding correction made.

Since the data analysis involved a number of corrections and relied on simulation results, we performed a test of the whole data taking and processing system by measuring the angular distribution of protons produced by the bombardment of methane molecules by oxygen

ions. The angular distribution of these protons should be isotropic because CH_4 molecules are highly symmetrical [18,19]. The measurements showed, after performing the corrections described above, isotropic angular distributions, indicating that the data taking and processing apparatus was not introducing an angular preference into the results.

RESULTS

The same operations were done for the electron capture spectra at all energies. For the ionization process, almost identical operations were performed except that the correction for double collisions was found to be negligibly small for this channel and was not made.

All final results for projectile energies of 2–16 MeV, both transfer ionization plus transfer excitation and double ionization plus ionization excitation, are shown in Figs. 3–6. Several general remarks can be made based on these figures. For the $dN/d\cos\theta$ distributions of the transfer ionization and transfer excitation, there is no molecular alignment θ dependence at 2 MeV. Starting from 4 MeV, however, $dN/d\cos\theta$ begins to peak at $\theta=90°$. The peak continues to grow until the projectile energy is close to 10 MeV, then declines when the energy goes higher. A very interesting thing happens when projectile energy reaches 16 MeV where $dN/d\cos\theta$ shows a weak indication of minima at angles other than 0° or 180°. If the peak in the $dN/d\cos\theta$ distribution can be explained by the interference between the two scattering amplitudes from the different atomic centers, these minima may be evidence for the onset of destructive interference.

For double ionization and ionization excitation processes, $dN/d\cos\theta$ has no molecular alignment dependence for all 2–16-MeV energies. For any process at any energy, the $dN/d\phi$ distribution has no ϕ dependence.

COMPARISON WITH THEORY

The theoretical results for 10- and 16-MeV O^{8+} impact have been calculated based on the previous work of Wang and co-workers [13,14] in which they formulated the impact parameter treatment of single electron capture by bare ions at high velocity from the hydrogen molecules. In their formulation for the single electron process, the amplitude for electron capture emerges as the sum of amplitudes for capture from two hydrogen atoms, to be added with relative phase $e^{i\alpha\cdot\rho}$, where α is the projectile momentum transfer. They integrate over the transverse component of α, as does the experiment, since no projectile scattering angle was measured, but the factor $e^{i\alpha_z\rho_z}$ remains, and gives rise to the interference effect which

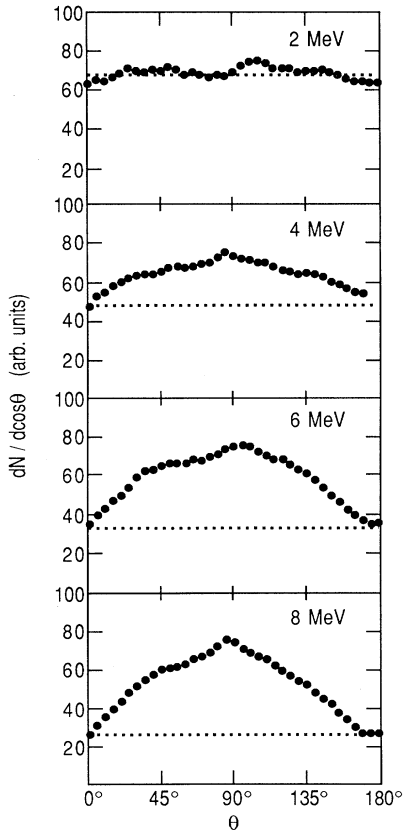

FIG. 3. The final results of the $dN/d\cos\theta$ distribution for the transfer ionization and transfer excitation for projectiles of energies from 2 to 8 MeV.

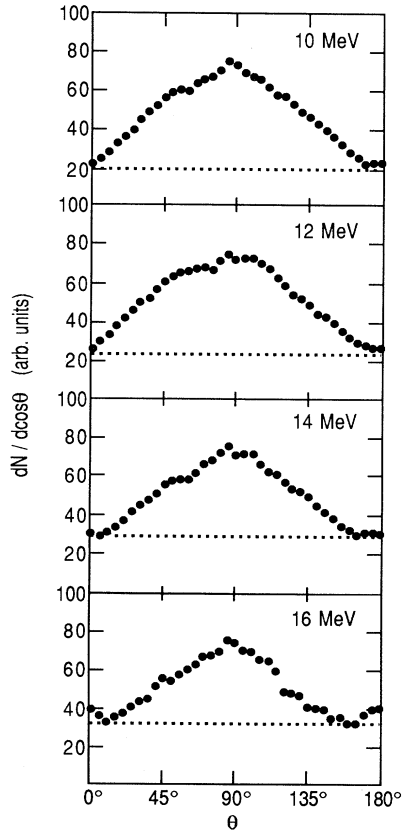

FIG. 4. The final results of the $dN/d\cos\theta$ distribution for the transfer ionization and transfer excitation for projectiles of energies from 10 to 16 MeV.

they calculate. From their Eq. (6), the probability for scattering from a diatomic molecule has a general form

$$P_M(\theta,b)=a_{11}(b)+a_{12}(b)\cos(\alpha_z\rho_z)$$
$$=a_{11}(b)+a_{12}(b)\cos[\alpha_z\rho\cos\theta]\ , \quad (5)$$

where b is the projectile impact parameter, α_z is the projectile longitudinal momentum change, ρ_z $(=\rho\cos\theta)$ is the projection of internuclear vector onto the z axis, θ is the molecular alignment, a_{11} is the direct term due to the individual centers, and a_{12} is the interference term from two centers.

The interference effect comes from the second term in Eq. (5) and depends on the quantity $\alpha_z\rho$ as well as the alignment angle θ. Since $\rho=1.4$ for deuterium molecules and $\cos\theta$ is between [0,1], the magnitude of α_z determines whether there is a strong or weak interference. For single capture, α_z is related to the electronic energy gain Q of the reaction by

$$\alpha_z=\frac{Q}{v}-\frac{v}{2}\ , \quad (6)$$

where v is the projectile velocity. Q in turn depends on the principal quantum number n to which the capture goes, and is given by

$$Q=\frac{Z^2}{2n^2}-V_{\rm ion}\ , \quad (7)$$

where $V_{\rm ion}$ is the ionization potential of atomic deuterium, 15.4 eV. Wang and McGuire use the Oppenheimer-Brinkman-Kramers [20] approximation to estimate the n distributions for the captured electron, which center at $n=5$ for 2-MeV O^{8+} impact and at $n=3$ for 16 MeV. Inserting these values into the above equations results in values of α_z of -0.77 and -1.5 for these two energies. It is seen that the lower energy α_z is too small for $\boldsymbol{\alpha}\cdot\boldsymbol{\rho}$ to generate a large phase difference, but that as the energy rises, α_z becomes larger and above 8 MeV one would expect to see effects.

To interpret the molecular alignment dependence observed in transfer ionization plus transfer excitation, we regard it as a two-electron process and assume an independent electron approximation is valid. In this model, the projectile first captures one electron from the target D_2 and subsequently excites the other electron to a final dissociative D_2^+ state. In the impact parameter treatment, the combined two-electron process is written as [14]

$$P_f(B)=P^c(B)P_f^{\rm ex}(B)\ , \quad (8)$$

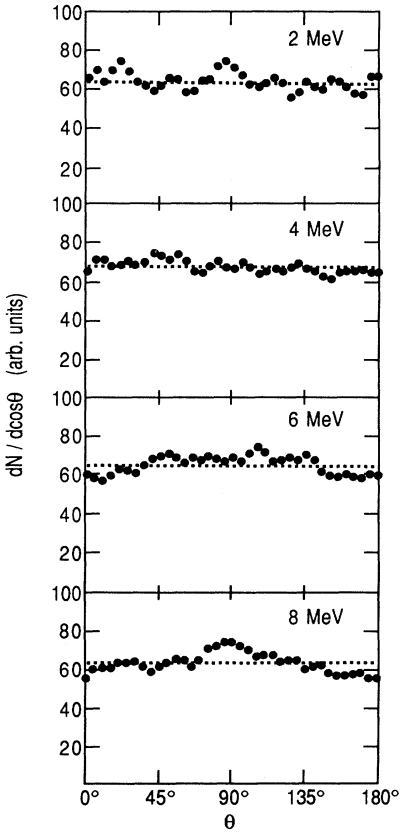

FIG. 5. The final results of the $dN/d\cos\theta$ distribution for the double ionization and ionization excitation for projectiles of energies from 2 to 8 MeV.

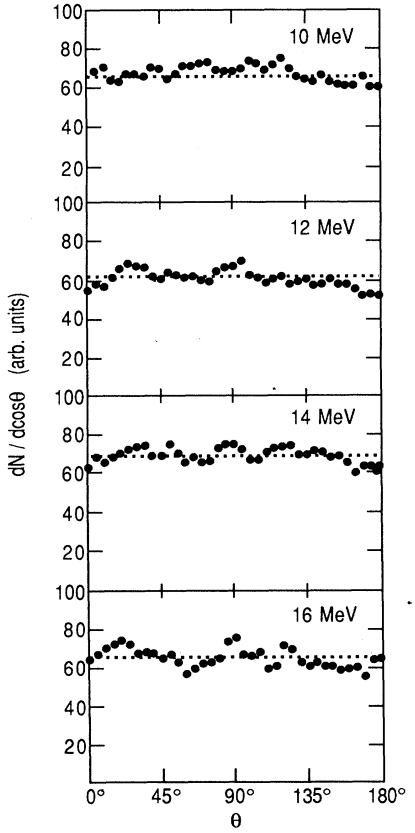

FIG. 6. The final results of the $dN/d\cos\theta$ distribution for the double ionization and ionization excitation for projectiles of energies from 10 to 16 MeV.

where P^c and P_f^{ex} stand for capture and excitation, respectively. Using a closure relation, we can sum contributions from all final D_2^+ states f (i.e., including transfer excitation and transfer ionization),

$$P(B)=\sum P_f(B)=P^c(B)[1-P_0(B)] , \quad (9)$$

where P_0 is the probability for elastic scattering between the projectile and the ground-state D_2^+ ion which is nondissociative. A further approximation is made regarding the elastic scattering probability $P_0(B)$. We assume that molecular orientation dependence in Eq. (9) is dominated by $P^c(B)$ alone and the elastic probabilty $P_0(B)$ does not contribute to the overall molecular orientation dependence. Thus the calculated molecular orientation dependence for electron capture process alone should approximately determine the observed angular distributions of transfer excitation and transfer ionization.

An explicit comparison between data and theory for 10 MeV is shown in Fig. 7. The theoretical results have been summed over n. The theoretical calculation agrees in shape with this experimental results. The peak at $\theta=90°$ suggests the interference is constructive at this angle and is in accordance with the "classical" double-slit interference, where the phase difference is zero. As the θ moves away from the 90°, the phase difference increases and the interference effect decreases. Our experimental data have minima at $\theta=0°$, 180° while the theoretical result is rather flat at $\theta=0°$ with a little decline as the θ increases until a minimum is reached at an angle somewhere between 0° and 45°. However, weak evidence for a minimum at an angle other than the angle $\theta=0°$ is seen in the experimental results for 16-MeV O^{8+} impact. The calculation for 16-MeV O^{8+} impact (Fig. 8) shows a rather similar interference pattern to the 16-MeV data but it differs in relative height. The qualitative agreement between the theory and the experiment suggest we have observed the interference effect described by Tuan and Gerjuoy in 1960.

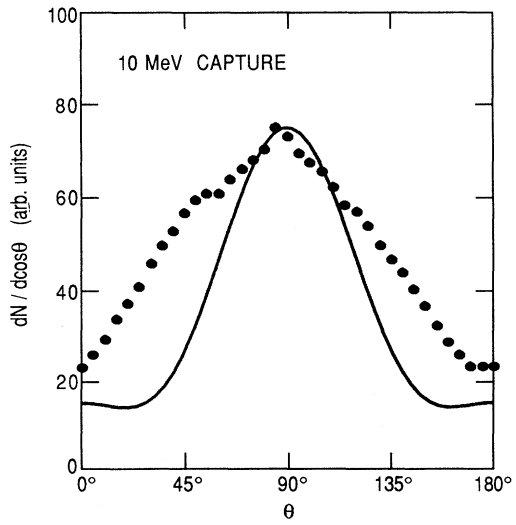

FIG. 7. A comparison of the theoretical calculation with the experimental results at 10 MeV. The theoretical results (solid line) are normalized to have the same maximum value as does the experimental result (data points).

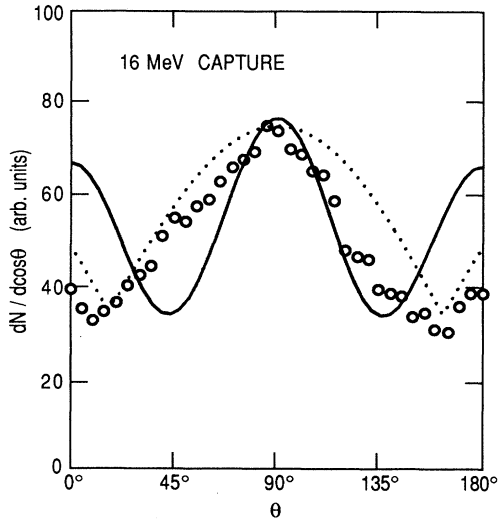

FIG. 8. The same as Fig. 6 except the projectile energy is 16 MeV. Notice that there are minima at $\theta\neq 0°, 180°$. The dotted line is a fit to the function $f(\theta)=1+c_1\cos(c_2\cos\theta)$, which is analogous to Eq. (7).

Although no calculation has been made for the alignment dependence for the ionization process (double ionization and ionization associated with excitation), qualitative explanation of our data can be made based on the general Eq. (5). For the ionization process, the parameter α_z in Eq. (5) has the form [21]

$$\alpha_z=\frac{(V_{\text{ion}}+\frac{1}{2}k^2)}{v} , \quad (10)$$

where $\frac{1}{2}k^2$, the kinetic energy of the ionized electron, is continuously distributed over a large range with typical values of the order of the deuterium atom binding energy [22]. The velocity v is in the range of 2–6 for our experiment. Therefore when integration over the continuous final states is performed there will not be much interference effect. The same argument holds for the associated excitation process. This agrees with our experimental $dN/d\cos\theta$ distributions for energies between 2 and 16 MeV (Figs. 5 and 6). This conclusion is also in qualitative agreement with the recent results of Ezell *et al.* [15], who measured the differential cross section for ionization by 1-MeV proton impact and found there is not much difference for all molecular alignments.

CONCLUSIONS

The differential cross sections for transfer ionization and transfer excitation as a function of the molecular alignment as well as the projectile energy have been measured for bare oxygen ions of energies between 2 and 16 MeV incident on deuterium molecules. We observed a significant dependence of the cross sections on the molecular alignment, usually with a peak at the alignment perpendicular to the incident beam direction. The shape and the height of the peak are observed to be changing as the

projectile energy changes. The interference between the two scattering amplitudes originating from the two atomic centers can be used to explain qualitatively the feature of the $dN/d\cos\theta$ distribution. The phase difference of the two scattering amplitudes from the two individual centers is a minimum when the molecular alignment is perpendicular to the beam, resulting in constructive interference. When the angle between the molecular alignment and the beam becomes smaller, the phase difference increases and the constructive interference decreases. The phase difference also changes when the projectile energy changes. A theoretical calculations based on the independent electron model and the two-center molecular wave function agrees qualitatively with our experimental results. As a comparison, the differential cross section for the ionization process has also been measured. No significant dependence of the cross section on the molecular alignment and the projectile energy was observed for ionization.

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