DD nuclear-fusion reactions with small D_2O and H_2O clusters impacting heavy ice

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The large DD-fusion-yield enhancement reported earlier by our group [Phys. Rev. Lett. **63**, 1292 (1989); J. Phys. Chem. **94**, 7665 (1990)] in the bombardment of deuterated targets by large heavy-water clusters was shown by postacceleration magnetic and electrostatic filtering to be due to small-ion impurities produced in the acceleration column. With the filtering arrangement in place we have carefully studied DD-fusion rates with small $(D_2O)_nD^+$ and $(H_2O)_nH^+$ ions ($n \leq 10$) which produced detectable fusion rates. Formation of carbon films on the polydeuteroethylene surfaces caused the observed fusion rates to decrease rapidly with time. This problem was solved by using D_2O ice targets which are "self-cleaning." No enhancement was observed for the fusion rates of the small D_2O clusters after the oxygen knock-on corrections were made. However, the fusion yields for knock-on processes produced by $(H_2O)_nH^+$ clusters ($4 \leq n \leq 10$) showed approximately a twofold enhancement over the yields for H_3O^+ ions at the same velocity. The stopping power of clusters was measured and shown not to be responsible for this enhancement.

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I. INTRODUCTION

Studies of rates of DD-fusion reactions generated by impacts of accelerated cluster ions were initiated several years ago [1] to search for "molecular" effects (collective interactions of atoms in the projectile with target atoms) on nuclear-fusion processes. The question raised was whether the local high-energy density produced by the penetration of an energetic polyatomic cluster projectile into a solid target could perturb the nuclear-fusion rates. The original experimental results gave surprisingly high rates which could not be readily explained by theory based on individual atomic collision processes [1,2]. A diligent search for artifact ions finally revealed evidence for traces of atomic or molecular deuterium ions sufficient to account for most of the observed fusion events [3].

In this paper we present a summary of the evidence responsible for our misinterpretation of the original experimental results. This is followed by a description of the improved experiment with special attention to targeting problems with cluster ions. Finally results of studies of fusion yields obtained with small D_2O clusters striking solid D_2O targets and for knock-on reactions with H_2O cluster projectiles on similar targets are reported. In these studies no evidence outside of limits of experimental error was found for enhanced D-D reaction rates induced by small D_2O cluster projectiles. But with the knock-on processes and with small H_2O clusters impacting solid heavy-water targets, small enhancements of fusion rate were observed. Small light-water cluster knock-on fusion rates, calculated on a per oxygen atom basis, were approximately a factor of 2 larger than values obtained with H_3O^+.

II. EVIDENCE FOR CLUSTER FUSION AND ARTIFACTS

The hazard of higher-velocity, low-molecular-weight beam impurities giving rise to spurious results was recognized from the outset and beams were carefully mass analyzed prior to acceleration to minimize this problem. The energy dependence of the observed fusion rates was clearly inconsistent with that of rates of fully accelerated D^+-ion beams. This observation established confidence in the quality of the beams prior to acceleration. Furthermore, an optimum cluster size appeared to be required for the fastest fusion rates [1,2]. The observation that higher-velocity clusters of lower molecular weight were not so effective as larger clusters supported the argument that beam contamination by low-molecular-weight ions was not responsible for the detected fusion events. The attempt to reproduce our findings by Fallavier *et al.*, with D_n^+ cluster ions failed [4], but this result was interpreted to reflect the importance of the presence of oxygen atoms for the observed fusion enhancement [2].

The apparent inconsistency between the results of various theoretical models [5–10], except the one by Kim *et al.* [11], and the experimental results stimulated a diligent search for evidence of artifact ions. Experiments with targets covered with thin gold films proved that D^+ ions with more than half of the acceleration energy were not responsible for the observed fusion rates. Time-of-flight studies [12] which compared the arrival times of ions at the target with times of fusion events eliminated the possibility of low-molecular-weight oxygenated ions as artifact impurities in the cluster ion beams. These time-of-flight studies also showed that D^+ ions generated in the first half of the acceleration process could not significantly contribute to the experimental results.

An independent experimental confirmation of our earlier results [1,2] with the use of an almost identical experimental setup was reported by Bae, Lorents, and Young [13], strengthening confidence in our results. Further experiments not reported earlier showed the production of small numbers of particles with energies close to 3 MeV when accelerated heavy-water cluster ions struck light polyethylene targets containing only the natural abundance of D. The fact that the energy was slightly

($\sim 10\%$) below 3 MeV was interpreted to be due to the deposited polyethylene film on the solid-state detector from target sputtering by cluster impact. Although very low, the yields in these experiments were still much higher than expected from interactions of projectile D atoms with deuterium of natural abundance in the polyethylene target. This experiment was taken as support for an exotic energy-amplification mechanism in the cluster impact process in which energetic deuterons of the clusters interact with each other.

In spite of the accumulated body of circumstantial evidence supporting our conclusion, we continued to search for experimental evidence for artifact ion-beam impurities. Studies with beams deflected off the axis of ion acceleration and with magnetic and electrostatic filtration were undertaken to address this issue further. A decrease in fusion rates by more than two orders of magnitude with use of the filtration arrangement was observed and reported by us [3]. This observation convinced us that small-ion impurities, probably D^+ and D_2^+, which were not fully accelerated, and thus could not be detected in the time of flight experiment, contributed significantly to the observed fusion rates for large clusters [3]. When we succeeded in improving the vacuum in the acceleration column from 1×10^{-6} to 1×10^{-7} Torr, the fusion yields decreased almost proportionally to the pressure. This was clear indication that the small-ion impurities were formed in the latter part of the acceleration column by collisions of cluster ions with residual gas molecules.

Just after our completion of these critical experiments, Lo, Petrasso, and Warren [14] pointed out that proton widths reported by us reflected reactant deuteron kinetic energies of the order of 50 keV ($\frac{1}{6}$ of the full acceleration energy), whereas the deuterons in our cluster projectiles had kinetic energies of only several hundred eV. Rejecting the idea of energy amplification [1,2],[11] in cluster impact, Lo, Petrasso, and Warren [14] correctly concluded that undetected artifact D^+ ions were present in our cluster beams. It was then recognized that the significance of the widths of fusion proton energy distributions was overlooked and misinterpreted in our original experiments.

More careful investigations showed that the misleading results obtained with heavy-water clusters on light polyethylene targets resulted from the $^{12}C(d,p)$ reaction, giving protons with energy within 10% of the 3-MeV DD-fusion protons. The steep energy dependence of the observed fusion reactions may reflect the energy dependence of the rate of very rare collision-induced dissociation reactions of cluster ions in the acceleration column giving atomic D-ion products. The hypothesis that deuteron "splash-back" [6] from impacts on deuterium-containing targets was responsible for the beam impurities is inconsistent with the off-axis studies.

With magnetic and electrostatic filtering of the cluster beams and with significantly improved vacuum conditions, the search was continued for evidence of molecular effects on rates of fusion reactions. With these refinements the fusion rates of $(D_2O)_nD^+$ clusters with $n>10$ incident on polydeuteroethylene (PDE) targets were below our detection level at available acceleration energies up to 750 keV. These negative results are in agreement with the findings of Fallavier *et al.* [15], who observed no enhancement of fusion yields with $(CD_4)_n^+$ clusters.

III. EXPERIMENT

The experiments were carried out in an apparatus similar to the one used in earlier experiments [2]. A sketch of the apparatus is shown in Fig. 1. Cluster ions were generated by supersonic expansion of weakly ionized mixtures of water in He carrier gas. The gaseous ion mixture contained a sufficient excess of neutral water molecules to react with all atomic and small molecular ions to generate cluster-ion distributions. These distributions were subjected to high-resolution mass analysis using a quadrupole mass analyzer. The mass-selected cluster ion beams were accelerated to energies as high as 750 keV. The typical pressure of the acceleration column was $\sim 1\times10^{-7}$ Torr.

The principal change from the earlier experimental setup [2] is the addition of beam deflection plates and magnetic filter in the target region. This arrangement serves for energy and mass filtration of cluster ions as well as off-axis rejection of neutral species generated in the acceleration column. The magnetic filter was a cylindrical soft iron yoke which contained up to three pairs of samarium cobalt permanent magnets. With three magnet pairs the magnetic field strength around the beam axis was 0.25 T. Beam intensities were monitored by copper cross-hair wires or mesh grid, which transmitted 95% and 90% of the beam, respectively, onto a target containing deuterium. As before, fusion events were detected by measurement of 3-MeV protons in a solid-state semiconductor detector placed near the target.

Polydeuteroethylene targets, bombarded with cluster-ion beams, showed rapid decay of target sensitivity as a function of irradiation time. Because the surface concentration of deuterium in PDE targets after ion bombardment is difficult to establish and rapidly decreasing, and because the cluster projectile D atoms lose their energies significantly through the dead target surface layers, accurate measurements of fusion rates became very difficult. Furthermore, with low fusion rates, targets deteriorated before measurements of statistical significance could be made.

As an example of the many data sets obtained, the integrated fusion proton counts per 400 sec are shown as a function of the beam exposure time of a PDE target in Fig. 2. The results were obtained with $(D_2O)_5D^+$ cluster

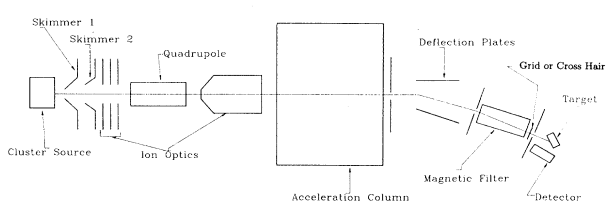

FIG. 1. Schematic diagram of the experimental apparatus (not to scale).

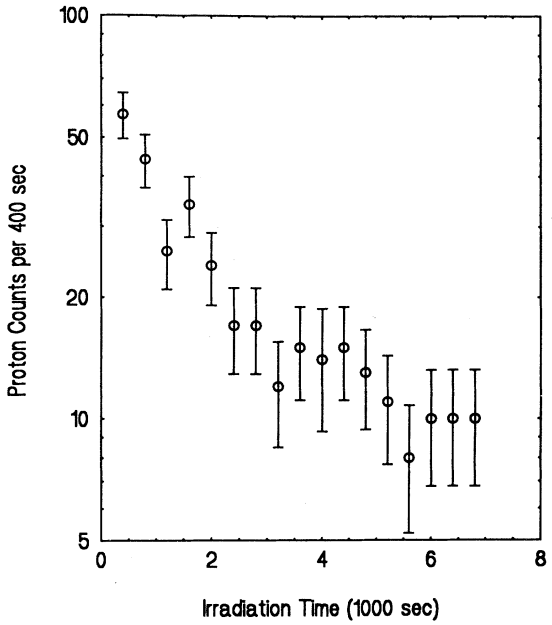

FIG. 2. Integrated fusion proton counts per 400 sec as a function of beam exposure time of a PDE target. The results were obtained with a 1-nA $(D_2O)_5D^+$ cluster-ion beam accelerated at 638 keV.

ions at 638 keV (12.5 keV/D). Fusion rates dropped by approximately a factor of 5 in 3000 sec with 1-nA beams of clusters, then reached a steady-state value. The corresponding integrated cluster flux for reaching the steady-state value was 2×10^{14} clusters/cm^2. The beam-exposed target showed a visible black carbon film on what was originally a white insulating plastic surface. Presumably these films are formed because the rate of dehydrogenation of polyethylene is higher than the rate of polymer or carbon sputtering when the surface is subjected to ion bombardment.

Fusion yields on a fresh surface, uncontaminated by a radiation-induced carbon film, are calculated by extrapolating back to zero time of irradiation with the cluster beam. The extrapolated yield shown in Fig. 2 is 2.5 ± 0.3 times the thick-target yield calculated for 12.5-keV deuterons in the 638-keV $(D_2O)_5D^+$ cluster. The thick-target yield (i.e., the probability of a fusion reaction of slowing down deuterons of the clusters with deuterons in the target) was calculated using the relation [5]

$$Y_{TT}=n_D\int_0^{E_D} dE_{lab}\frac{\sigma_f(E_{c.m.})}{\left|\frac{dE_{lab}}{dx}\right|}, \quad (1)$$

where n_D is the number density of deuterons in the target, σ_f is the fusion cross section, E_D is the initial kinetic energy of the deuterons, $E_{c.m.}$ is the center-of-mass energy for DD collisions, $E_{lab}=2E_{c.m.}$, dE/dx is the total stopping power of D in polydeuteroethylene. The experimental values of the fusion yield include fusion events that result from reactions of energetic deuterons produced by oxygen knock-on collisions, and the knock-on yields will be discussed later. Values of extrapolated DD-fusion yields corrected for knock-on yields obtained with H_2O clusters are approximately 1.5 ± 0.6 times the calculated thick-target yields.

The gross yields observed after several thousand seconds before correcting for knock-on contributions are roughly half the calculated thick-target yields. These low yields reflect energy loss of projectile atoms in carbon films generated in a steady state on the target surface. From the decrease of the fusion rates the steady-state film thickness was estimated to be ~1000 Å.

"Self-cleaning" D_2O ice targets were developed to solve this problem. Targets of heavy-water ice were formed as heavy water condensed on a flat surface of a copper rod which was attached to a copper Dewar located in the vacuum and refrigerated with liquid nitrogen. Because these targets were expected to have very high sputtering rates, the 500-l/sec turbo molecular pump used for earlier studies with PDE targets was replaced with an 8" CTI cryopump capable of pumping water vapor at a speed of 4000-l/sec. The pressure of the target chamber was monitored with an ionization gauge located approximately 50 cm from the target. The typical pressure of the target chamber was 1×10^{-7} Torr and did not show any detectable rise during beam bombardment of the ice targets. But it should be noted that the local pressure of heavy-water vapor near the impact region could be much higher.

IV. RESULTS AND DISCUSSION

The results of studies of small heavy-water clusters on the heavy-ice targets are shown in Fig. 3. These results

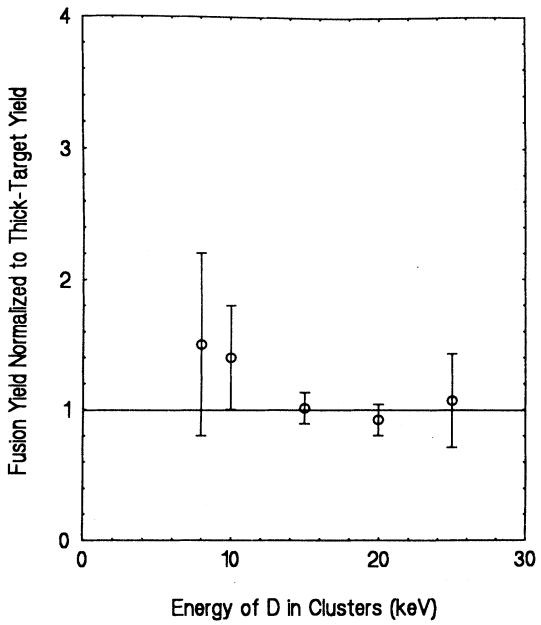

FIG. 3. Fusion yields of small D_2O clusters incident on D_2O ice, normalized to thick-target yields. The yields were measured with clusters containing 1–7 D_2O molecules and corrected for oxygen knock-on yields.

were obtained with clusters containing 1–7 water molecules, corrected for the knock-on contributions, and divided by the calculated thick-target yields. The very small contributions of energy transferred by hydrogen atoms in the $(H_2O)_nH^+$ knock-on reactions were neglected. Within experimental uncertainty, the heavy-water fusion yields did not show any cluster size dependence. The error bar of each data point was estimated from the standard deviations of several experimental runs and thus reflects both statistical errors and the reproducibility of experimental results. For deuterons with energies larger than 10 keV no fusion yield enhancement over the thick-target yields can be detected. At the lower energies, 8–10 keV/D, there is the suggestion of a small enhancement but the results are not beyond statistical error limits. These results again reflect very high cluster beam purity.

The rates of knock-on reactions were studied with $(H_2O)_nH^+$ clusters bombarding D_2O ice targets. The results are presented in Figs. 4 and 5. These results were obtained with oxygen-atom energies varying from 64 keV/atom to 280 keV/atom in clusters containing up to ten water molecules accelerated to energies up to 730 keV. Knock-on fusion yields are plotted as a function of cluster size at different energies of ion acceleration in Fig. 4. At all energies studied, the yield of $(H_2O)_nH^+$ clusters with $n \geq 4$, calculated on a per oxygen-atom basis, is a factor of approximately 2 larger than the yield for $n=1$. This enhancement can be seen more clearly in Fig. 5, which gives the ratios of fusion yields per oxygen atom for the dimer and polymer clusters to the yields of fusion with monomers.

The experimental knock-on fusion yields were estimated with the relation

$$Y_{\text{expt}} = C\,P\,\Gamma / I_{\text{CH}} , \tag{2}$$

where C is a constant, P is the number of the observed fusion protons, Γ is the secondary electron yield for cluster impact, and I_{CH} is the current registered on the cross-hair arrangement. The ratio of I_{CH}/Γ measures the cluster-ion flux. P is proportional to the theoretical knock-on yield per oxygen atom, Y_{KO} which is given by [5]

$$Y_{KO} = n_D \int_0^{E_O} dE \frac{1}{\left|\frac{dE}{dx}\right|_O} \int_0^{U_{\max}} dU\, Y_{TT}(U) \frac{d\sigma}{dU} , \tag{3}$$

where E_O is the incident kinetic energy of the oxygen atoms, $|dE/dx|_O$ is the total stopping power of the oxygen atom in polyethylene, $Y_{TT}(U)$ is the thick-target fusion yield given by Eq. (1), $d\sigma/dU$ is the differential energy-transfer cross section for O-D collisions, and $U_{\max} = 0.395E$. Γ is given by [16]

$$\Gamma \propto \int_0^{\infty} dx \exp\left[\frac{-x\cos\theta}{\lambda}\right] \left|\frac{dE}{dx}\right|_e , \tag{4}$$

where θ is the angle between the particle trajectory and the axis normal to the surface, $x\cos\theta$ is the depth of penetration, λ is the characteristic wavelength for elec-

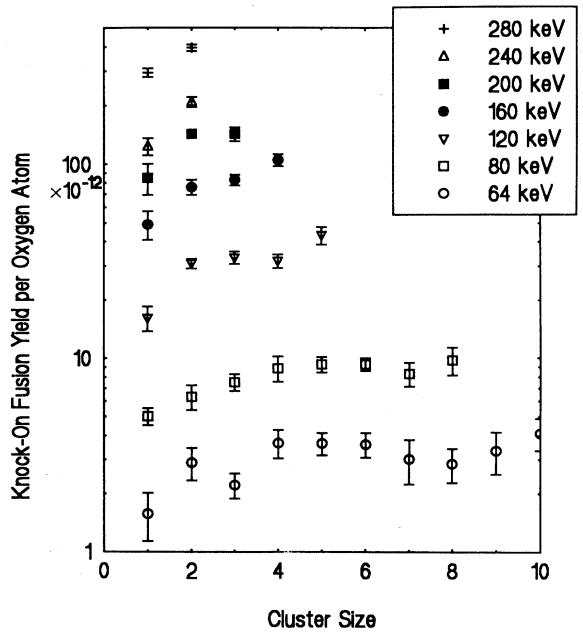

FIG. 4. Knock-on fusion yields per oxygen atom when D_2O ice is bombarded with $(H_2O)_nH^+$ clusters of various sizes and energies. Different symbols represent different oxygen kinetic energies.

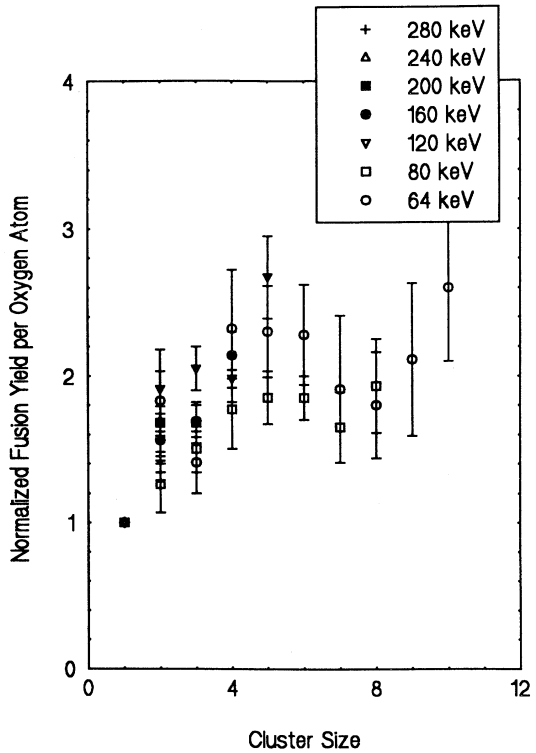

FIG. 5. Knock-on fusion yields per oxygen atom of H_2O clusters normalized to the yields of H_3O^+ at various energies per oxygen atom as a function of the cluster size.

tron diffusion, and $|dE/dx|_e$ is the electron stopping power of the cluster. For the velocity ranges studied in this work $|dE/dx|_e \simeq n|dE/dx|_O$. Since the oxygen stopping power $|dE/dx|_O$ enters into Eqs. (3) and (4) as a denominator and numerator, respectively, its effect would be cancelled.

The importance of stopping power in the calculation of thick-target-fusion yields [Eq. (1)] led us to an experimental test of the above conclusion that there would be no significant molecular effect on the stopping power of constituent cluster atoms. This question was addressed by measuring the energy loss (stopping) of clusters in a 10-$\mu g/cm^2$ carbon film placed in front of an Ortec Ultra silicon solid-state detector with a very thin window (500 Å). The detector was calibrated with very-low-intensity accelerated cluster beams without the carbon absorber.

The results of the cluster-stopping measurements with clusters containing up to five water molecules at energies of 200–500 keV are presented in Fig. 6. A small increase in absolute value of energy loss with an increasing energy per water molecule is expected with the increase in electronic stopping power with projectile energy. Figure 6 shows monomers with slightly larger stopping power than polymers, but the difference is generally less than 20%. Therefore, the stopping-power effect on fusion yields would be expected to account for an increase in fusion yield of no more than 20% for polymers related to monomers.

Further evidence for the absence of low-molecular-weight impurities in our improved cluster beams was obtained from the widths of the fusion proton distributions. No detectable change in the widths as a function of cluster size was observed. This observation qualitatively supports the absence of small oxygenated molecular impurities in the cluster beam. With marginal counting statistics in individual width determinations, data were analyzed by combining results from all runs with a given energy per oxygen atoms, regardless of cluster size. The full widths at half maximum (FWHM) are listed in column 2 of Table I for four oxygen energies, and column 3 gives the deuteron energies E_D (in the laboratory system) corresponding to these widths. The relation between FWHM and E_D was calibrated with D_3O^+ beams incident on D_2O ice. The maximum deuteron energies, E_{max}, that can be produced by oxygen knock-on were calculated from the relation

$$E_{max} = \frac{4m_D m_O}{(m_D + m_O)^2} E_O \tag{5}$$

where m_D and m_O are masses of deuteron and oxygen atoms, respectively, and E_O is the initial kinetic energy of the oxygen atoms in the cluster ions. The results for E_{max} are presented in column 4. The deuteron energies deduced from the widths are seen to be in good agreement with the calculated ones from Eq. (5). This agreement strongly supports the absence of any low-molecular-weight impurities in our cluster beams.

The thick-target yields for D_2O clusters reported here are generally consistent with observations made by the research groups of Fallavier *et al.* [15] and Vandenbosch *et al.* [17]. However, we note that the molecular effect on the fusion reactions was established only for the knock-on processes, but not for thick-target processes. Our results on knock-on yields are in contrast with the results with carbon clusters by Vandenbosch *et al.* [18]. The difference might result from the use of PDE targets and frequently higher beam intensities in the experiment by Vandenbosch *et al.* [18], which would not permit the detection of the relatively small but nevertheless significant molecular effects observed in the knock-on processes with ice targets.

We have no quantitative explanation for these effects. A possible explanation might be found in the proposal of a Fermi shuttle mechanism, which in recent calculations has been shown to be theoretically capable of giving enhanced fusion rates [5–10]. Because the thick-target processes are much more sensitive to beam contamination than the knock-on process, and because experimental evidence is strongly against beam contamination, we are confident that the observed "molecular" enhancements of fusion rates are beyond reasonable estimates of experimental error. The magnitude of the observed molecular effect for larger clusters remains to be investigated.

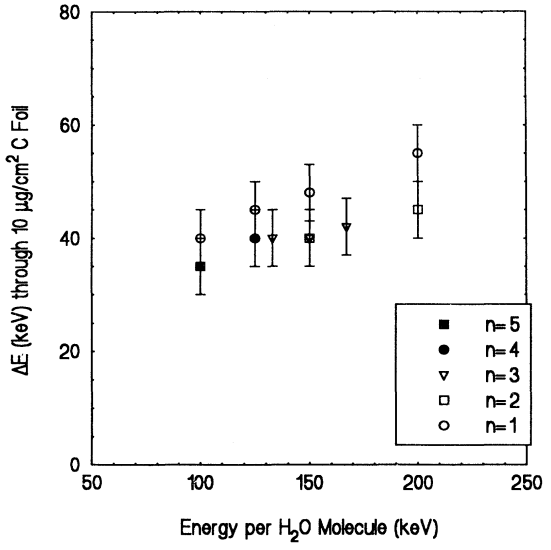

FIG. 6. Energy loss in 10 $\mu g/cm^2$ carbon films of various water clusters containing up to five molecules plotted as a function of energy per molecule. The cluster ions have the formula $(H_2O)_nH^+$.

TABLE I. Measured widths of proton energy distributions and corresponding energies of knock-on deuterons.

Energy per incident oxygen (keV)	FWHM of proton distributions (keV)	E_D (keV) from width	E_{max} (keV) from Eq. (5)
80	166±8	36±3	32
120	201±10	52±5	47
200	245±10	75±6	79
240	258±16	83±10	95

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