Cluster-Impact-Fusion Yields: No Collective Effect Observed for Small Water Clusters

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D+D nuclear fusion rates have been measured for 225-keV water cluster anions OD^-, $O_2D_3{}^-$, and $O_3D_5{}^-$. Contrary to a recent report for similar cations, these rates fall rapidly with cluster size and are consistent with free-deuteron rates.

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Recently Beuhler and co-workers [1,2] reported the remarkable observation that the exothermic nuclear fusion reaction $D+D \rightarrow {}^3H+p$ occurs at a rate enhanced [1,3] by a factor of the order of 10^{25} when one bombards a deuterated target with deuterated water clusters containing of the order of 100 clusters. For example, in a 300-keV cluster of 100 D_2O molecules, each deuteron has a kinetic energy of 0.3 keV, an energy more than an order of magnitude below the lowest energy for which it has been possible to measure the fusion cross section with free deuterons [4]. Extrapolation of the measured free-deuteron cross section to lower energies using a standard nuclear-reaction-rate approach [4] often employed in astrophysical calculations leads to the aforementioned enhancement factor. Fallavier *et al.* [5] have looked for fusion induced by pure deuterium clusters and did not observe an enhancement.

There is not a clear understanding of the enhancement mechanism at this time. Beuhler, Friedlander, and Friedman [1] suggested that compressional and heating effects from the rapid stopping of the cluster might be responsible. Attempts to fit the data with thermal models showed that a quite high temperature was required to account for the data [6,7]. Carraro *et al.* [8] investigated a number of mechanisms and were unable to account for the magnitude and cluster-size dependence of the yields. They also performed a molecular-dynamics calculation, as did Haftel [9] and Hautala, Pan, and Sigmund [10], and failed to account for the observed cluster fusion rates.

Very recently Bae, Lorents, and Young [11] performed an independent experimental study with water clusters. They appear to confirm the results of Beuhler *et al.* [2] for large cluster size. More surprisingly, they report large enhancements for small clusters ($n=2$–10) as well. The study of Beuhler *et al.* suffered from relatively poor cluster-size resolution and did not extend to small clusters. The present study was stimulated by the unexpected recent results of Bae, Lorents, and Young for small clusters. We have also initiated a program to investigate the possibility of enhancing fusion rates using a single, large, deuterium-containing molecule rather than a cluster of small molecules. Those results will be reported elsewhere.

Our experimental apparatus is sketched in Fig. 1. Acceleration is accomplished with the negative-ion preacceleration deck of the University of Washington Van de Graaff Booster Accelerator [12]. Water cluster anions are produced in a direct extraction ion source elevated typically to 45 kV. An arc is struck in a gas mixture of D_2O vapor and D_2. The cluster yields are enhanced by running at considerably higher pressures and lower arc currents than normally employed when producing atomic anions. The addition of D_2 gas greatly enhances the yield of $D_{2n-1}O_n$ anions compared to O_n anions.

The negative ions are then passed through a 90° bend magnetic analyzer which provides unit mass resolution for A greater than 300. During data acquisition the image slits are opened to ± 4 mm, which is sufficient, with the small object size and the high mass dispersion, to keep all beam off the slit edge without introducing any contamination from adjacent masses. Examples of mass spectra are shown in the inset of Fig. 1.

The ions are accelerated to several hundred keV and transported to the target-detector chamber several meters away. The target consisted of about 1 mg/cm^2 polyethylene $(C_2D_4)_m$ deposited on 0.0003-in. Al foil. A second 0.0003-in. Al foil further protected the detector, which was mounted in transmission geometry. The detector was a $\frac{3}{4}$-in.-diam silicon surface-barrier detector. The detector chamber region was pumped by a liquid-nitrogen–trapped oil diffusion pump and the acceleration deck by turbomolecular pumps. The pressure was typically 1 μTorr in the target chamber and acceleration tube regions.

The defining aperture was biased positively and a guard aperture (not shown in Fig. 1) was biased negatively to keep secondary electrons from the defining aperture off the target foil and secondary electrons from the target foil from leaving the target foil. Typical beam currents were 1–100 nA.

A magnetic field following the acceleration tube was used to sweep away any D^- ions produced upstream or within the acceleration tube which might be accelerated to higher velocities and contaminate the result. With the open slit geometry employed, however, we did not observe contamination effects even with this sweeping field turned off. Also, the mean proton energy observed with molecular beams exhibited the expected kinematical downshift as compared to that observed with high-energy deuterons.

We first compare our yields with the data of Bae,

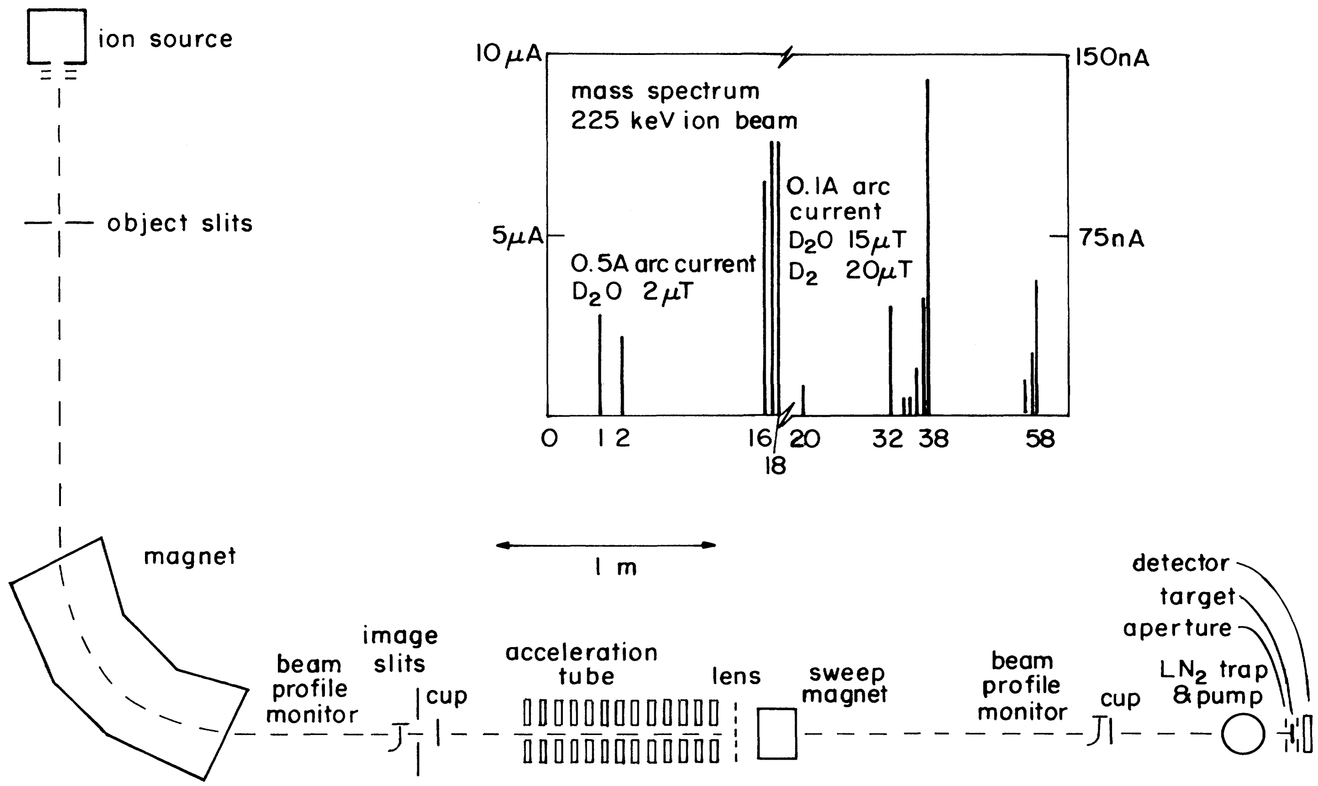

FIG. 1. Diagram of experimental apparatus. Inset: Two mass spectra. The left part of the inset shows the low mass spectrum when there was still appreciable residual ^{1}H, and the right part shows the mass spectrum after optimization of $n=2$ and 3 clusters by addition of D_2 gas.

Lorents, and Young [11]. In Fig. 2 we show the total proton fusion yield per cluster as a function of cluster size n. On the basis of the interpretation of our results which we present below, the data from the two experiments should be further scaled by the number of deuterons per cluster and by a smaller factor for the difference in deuteron energies for $(D_2O)_n{}^+$ and $(D_{2n-1}O_n)^-$. The net effect is less than a factor of 2 and is not included in this figure. Although our results for $n=1$ are in reasonable agreement with those of Bae, Lorents, and Young and also with the thick-target model of Carraro *et al.*, for larger n our yields fall rapidly with n while those of Bae, Lorents, and Young fall much more slowly. Our yields are approximately 1 order of magnitude lower than theirs at $n=2$, and our $n=3$ yield is 2 orders of magnitude lower than the interpolation of their $n=2$ and 4 points. We have also attempted a measurement with $n=4$. We only obtained 0.5 nA of beam, and in several hours of running observed two counts in the peak region (taken here to be a window which includes 95% of the observed protons for smaller-n clusters). We show this result as an upper limit in Figs. 2 and 3.

We believe that our present results indicate the absence of any special enhancement due to cluster impact for $n<5$ and can be understood simply in terms of the free-deuteron thick-target excitation function. An attractive feature of our experimental setup is that we can measure the thick-target fusion yield as a function of energy for D^- ions. The results of these measurements, taken interspersed with the cluster measurements, are shown in Fig. 3. Note that these thick-target yields have been measured down to energies approaching that of the lowest energies for which thick-target measurements have been made for astrophysical purposes [4].

If there is no special enhancement over the free-deuteron cross sections due to the other atoms in the cluster, one would expect the cluster yields to be intimately related to the free-deuteron thick-target yields. One could think of a D_yO_z cluster as giving rise to a yield given by y times the deuteron yield at $E_d=(m_d/m_{\rm cluster})\times E_{\rm cluster}$. Thus we would expect $1/y$ times our cluster yield (yield/deuteron) plotted at E_d as defined above to fall on our experimental free-deuteron yield curve. We have plotted our data in this manner in Fig. 3. We find that this simple expectation is satisfied within our uncertainties due to beam integration. We have included in this plot additional data from ions less directly comparable with those of Bae, Lorents, and Young, namely,

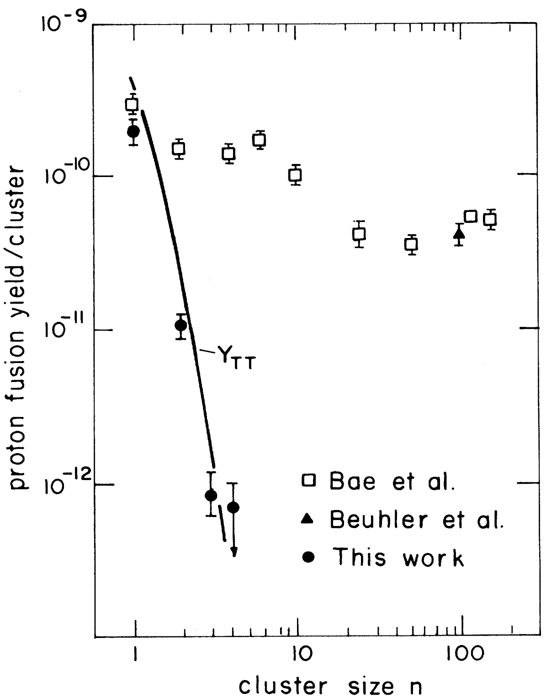

FIG. 2. Proton fusion yield for 225-keV clusters incident on $(C_2D_4)_m$ targets. The clusters in the work of Bae, Lorents, and Young are $(D_2O)_n$ and in the present work are $D_{2n-1}O_n$. The full curve is the calculated thick-target yield (Y_{TT}) as discussed in the text.

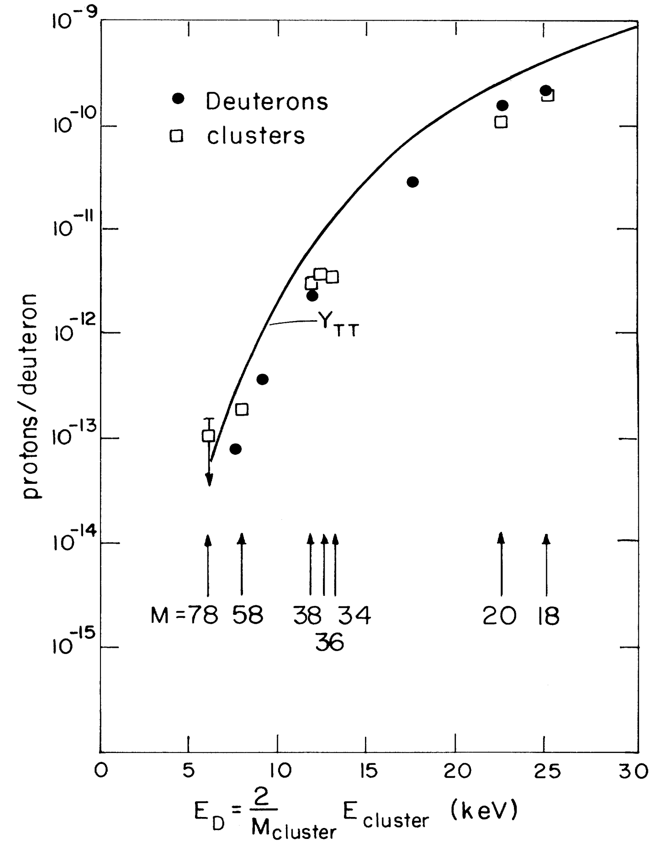

FIG. 3. Proton fusion yield per cluster divided by the number of deuterons per cluster plotted as a function of the kinetic energy of each deuteron. Also plotted are measured yields for free deuterons. The full curve is the calculated thick-target yield based on literature values of cross sections [4] and stopping powers [13].

$A=20$, 34, and 36. The $A=20$ ion was demonstrated to be ^{18}OD by magnetic analysis of breakup products from the tandem Van de Graaff terminal stripper. The $A=34$ and 36 ions are attributed to O_2D^- and $O_2D_2^-$ on the basis of a similar analysis. These tandem Van de Graaff stripper studies also provide an important demonstration that the clusters retain their integrity during acceleration and transport to the target, as the tandem analysis requires transport of this beam 10 m past the target position to the stripper of the tandem.

The accelerating structure available to us for this work required that we use negative ions rather than positive ions and, therefore, species having fewer deuterons per oxygens than in the positive-ion studies. The fact that O_2D^-, $O_2D_2^-$, and $O_2D_3^-$ have yields that scale as expected suggests that the D/O ratio does not have any unanticipated effect on the fusion rate.

We have also calculated the expected $(C_2D_4)_m$ thick-target proton yield following the method of Carraro *et al.* [8] and using stopping powers given by Andersen and Ziegler [13]. The result is shown by the full curves in Figs. 2 and 3. The agreement in shape is excellent. The absolute magnitude of the calculation is somewhat higher than our data, perhaps due to uncertainties in stopping powers in the calculation and in the absolute detection efficiency in the experiment.

For experimental reasons, the dependence of the fusion yield on cluster size has been presented [1,2,11] for *fixed cluster energy*. However, we suggest that the determination of the size dependence for *fixed cluster velocity* is less ambiguous. If there is indeed a collective enhancement over the basic thick-target yield curve derived from the standard D+D fusion cross section, one might expect this enhancement to increase monotonically with cluster size for *fixed cluster velocity* and perhaps saturate at some value corresponding to an infinite sheet cluster geometry. On the other hand, with a fixed degree of enhancement (e.g., at saturation) and *fixed cluster energy* one would expect the fusion yield to decrease with increasing cluster size (due to the decrease in energy of each atom). An interplay of these two effects could produce the peak in fusion yield observed by Beuhler *et al.* [2] near $n=200$ with higher-energy (300-keV) clusters. We conclude from our measurements that any fusion yield enhancement for clusters must occur for $n>4$. The present results do not contradict the measurements of

Beuhler *et al.* [2] for much larger n.

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