Search for Nuclear Fusion in Deuterated Targets under Cluster-Beam Impact

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Stimulated by the recent report of Beuhler, Friedlander, and Friedman on the observation of *dd* fusion under the impact of heavy-water clusters on deuterated solid targets, we undertook a similar study with pure deuterium clusters (D_{200}^+–D_{300}^+) in the same range of incident energy per deuteron (less than 1 keV). We observed no fusion event and our upper limit for the fusion rate is more than 1 order of magnitude below the Brookhaven value. Additional measurements performed with N_n^+ projectiles were not conclusive but showed that beam-contamination problems may be very serious.

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This experimental work was motivated by the recent observation of "cluster-impact fusion" by Beuhler, Friedlander, and Friedman[1] at Brookhaven National Laboratory (BNL). Fusion was claimed to be observed when $(D_2O)_n^+$ clusters, with the number n of molecules per cluster ranging from 20 to 1000, accelerated to a total energy of the order of 300 keV, were sent onto a TiD target. The measured maximum fusion rate is more than 10^{10} times larger than that expected from isolated deuterons of the same velocity.

The cryogenic source of the Lyon accelerator cannot produce water clusters but can deliver deuterium clusters, which allowed us to test the hypothesis that D_n^+ clusters could induce *d*(beam)-*d*(target) fusion. This paper describes our experimental search for fusion events when 100–150-keV D_{200}^+–D_{300}^+ clusters, carefully mass and energy analyzed after acceleration, bombard deuterated titanium and polyethylene targets. The net result is that we found no evidence for *dd* fusion.

Moreover, we learned that the authors of Ref. 1 also observed fusion when using light-water clusters[2] as projectiles, but with a rate 20 to 50 times smaller. In that case fusion can be due only to *d*(target)-*d*(target) collisions, which suggests that the oxygen ions of the water projectiles play a major role in the energy deposition process under cluster impact. Minor modifications of our cryogenic cluster-ion source allowed us to produce and accelerate nitrogen clusters (N_n^+) depositing about the same amount of energy in the first target layers as with water clusters. Because of the large mass of the projectiles this last experiment was performed with the direct beam. Under these conditions we have not been able to draw conclusions about *d*(target)-*d*(target) fusion because of the contamination of the incident beam by fast deuterium species.

The setup of our experiment is sketched in Fig. 1. After acceleration the incident clusters are selected, first in energy, by a 74° electrostatic analyzer, and then in mass, by a 16° bending magnet, with a mass-energy product of 60 MeV u. The mass resolution ($\Delta m/m \sim 3\times10^{-2}$) is determined by the exit slit of the magnet and an opaque collimator located 44 cm upstream from the target. The distance between the exit slit and the target amounts to 185 cm.

The mass selection of the projectiles after acceleration is a major difference between the Lyon and Brookhaven facilities. At BNL the projectiles are mass selected, focused, and then accelerated towards the target along a straight direction. Identification of beam contaminants was performed by direct transmission to a solid detector, but at a much lower intensity than during the experiment. Because of the dead-layer thickness of the detector, the full-mass clusters do not enter the active zone, while the light fragments can be identified from their residual energy loss. The authors of Ref. 1 claimed that they observed no full-energy D^+ ions. The main contaminant was reported to be D_2O^+, but with a negligible contribution to the measured fusion rate.

In both experiments the beam intensity was determined from indirect measurements. The Lyon machine delivers a pulsed beam (1 Hz), with a duty cycle of 10%. Because of the stability of the beam intensity (a few percent), the target current can be normalized to the beam

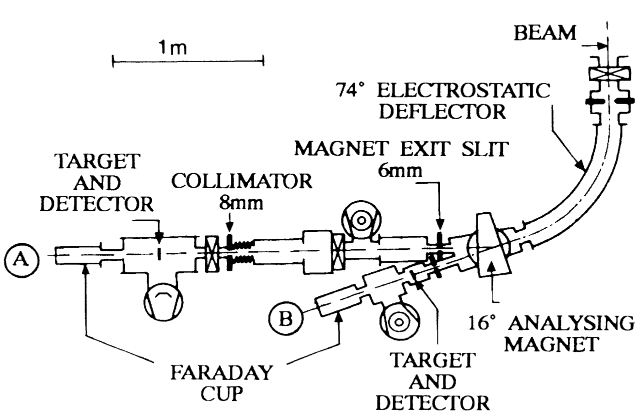

FIG. 1. Experimental setup for the D_n^+ (line *A*) and for the N_n^+ (line *B*) experiments.

current measured with a specially shielded and biased Faraday cup placed behind the removable target. Each current pulse was displayed on-line on a digital oscilloscope and integrated for dose measurement. At BNL, relative values of primary-beam intensities are calculated from the measured yield of secondary electrons, using data obtained previously.[3] This procedure is also used to check the integrity of the cluster beam, since the secondary-electron multiplicity distribution, at a given velocity, is related to the cluster mass.[4]

The nitrogen-cluster experiment could not be performed on the same beam line, due to the limited bending power of the magnet. We used the direct beam line *B* (Fig. 1) where a small magnetic field can be applied to prevent low-mass projectiles from reaching the target. On this beam line the distance between the exit port of the electrostatic sector and the target is only 82 cm.

The target-detector assemblies are quite comparable in the BNL and Lyon experiments. The target was tilted at 45° with respect to the beam axis and the 900-mm^2 Si surface barrier detector was placed at 90°, 2 cm from the beam spot on the target. A thin absorber (20 μg cm^{-2} Formvar) was used to protect the detector against the low-energy scattered atomic fragments. In order to check the calibration of the detection system, low-mass $D_n{}^+$ clusters could be directed onto the deuterated target and the fusion products (^{3}He,t,p) could then be easily observed when necessary. As an example we show in Fig. 2(a) a calibration spectrum obtained with 100-keV $D_3{}^+$ ions. Two types of deuterated targets have been used. The TiD target was elaborated[5] by deposition of 320 μg cm^{-2} Ti on a copper backing followed by high-pressure D loading up to the $TiD_{1.7}$ stoichiometry. We also used deuterated polyethylene disks, 0.7 mm thick, because of possible D depletion at the very surface of the TiD target. Moreover, huge sputtering effects arising with nitrogen clusters prevent the use of thin TiD layers.

Conventional electronics was used to inhibit transient stray signals generated in the environment of our pulsed accelerator. Moreover, a gate signal can be applied in order to trigger the counting during or between the beam pulses.

A crucial point of the experiment is the survival probability of the mass-analyzed clusters. Two mechanisms can lower the mass of the projectiles, the collisional dissociation in the residual gas and the evaporation induced by the ir photons emitted from the beam-line tube.

The collisional-dissociation probability of clusters in gas targets has been previously studied[6] for 100–600-keV $H_n{}^+$ clusters up to $n=23$. It has been found that the dissociation cross section tends to be the geometrical cross section. This approach leads to a dissociation rate of 15% for $D_{200}{}^+$ clusters under our experimental conditions (pressure 7×10^{-7} hPa, length 185 cm).

Thermal evaporation rates can be estimated from the Stephan-Boltzmann law. First, one can assume that the $D_n{}^+$ clusters entering line *A* are very cold, the evaporation time of the internal energy of the clusters produced by the ion source being much smaller than the time spent by the projectiles to reach the magnet. Then, after mass analysis, the incident clusters are plunged for a few μs into a 300-K blackbody chamber. The photon flux through the cluster external surface is of the order of 10^{22} m^{-2}s^{-1}, which corresponds during the flight time to an energy smaller than 1 eV for a $D_{250}{}^+$ cluster. The most pessimistic hypothesis, assuming that the radiation energy is entirely absorbed by the projectile, leads therefore to a mean energy of a few meV per D_2 molecule in the cluster. Even in that case this energy is much smaller than the typical evaporation energy of D_2 molecules, calculated from the macroscopic value of 13 meV for the latent heat of vaporization (this value is quite comparable to measured bond energies of D_n clusters[7]).

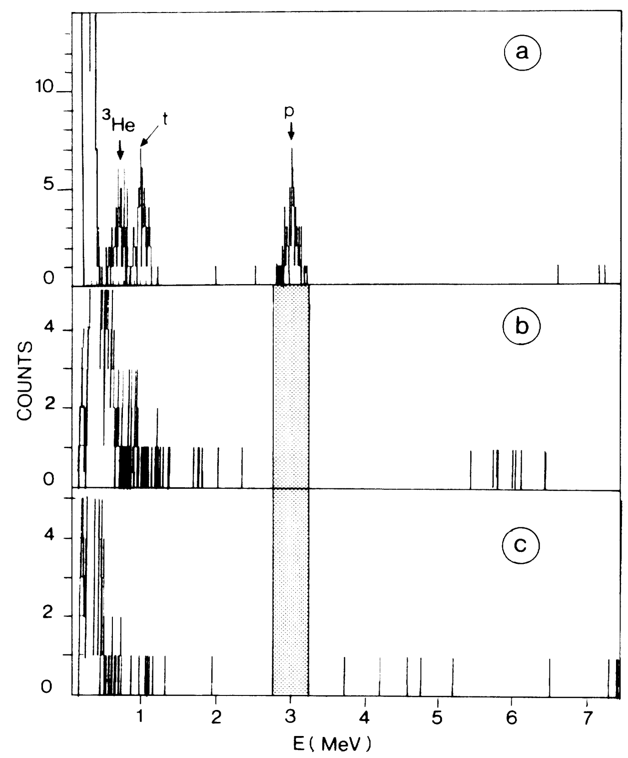

FIG. 2. Energy spectra obtained for the following conditions: (a) 100-keV $D_3{}^+$ ions on TiD (beam intensity 3.3 nA); (b) without accelerator voltage (acquisition time 2×10^4 s); (c) summation of four spectra corresponding to the data of Table I (total accumulation time 8400 s). The dotted area corresponds to the proton energy window.

These estimates show that in-flight dissociation must result mainly from collisions with the residual gas. Accordingly, under our experimental conditions, most of the projectiles survive up to the target.

Our search for fusion events was restricted to the detection of protons in the energy window ranging from 2.8 to 3.2 MeV [see Fig. 2(a)]. We measured the background level of the detection system with zero voltage at the accelerator terminal [Fig. 2(b)]. The shape of this spectrum explains why we did not use t and ^{3}He particles to sign fusion events. For a net acquisition time of 2×10^4 s, there are fifteen background counts from 1.4 to 7.4 MeV, but *none* in the proton window. In the most pessimistic hypothesis of a uniform distribution of background events from 1.4 to 7.4 MeV, there is no more than *one* background event in the proton window. This corresponds to a counting rate of 5×10^{-2} per 1000 s. Under these conditions we should be able to measure fusion rates 2 orders of magnitude below those reported in BNL experiments.

In Table I, we present all our experimental results obtained with 100-150-keV $D_n{}^+$ clusters ($n=200$, 250, and 300) bombarding deuterated titanium and polyethylene targets. In order to allow a direct comparison between the Lyon and BNL data we compare clusters of the same velocity. The BNL data have been normalized to the same number of incident deuterons as in our experiment. Our relatively low effective acquisition time, 4200 s for the longest measurement, results from the beam duty cycle of 10% and the limited running-time length of the Lyon accelerator (60 h per week, due to the operating mode of the cryogenic ion source).

In our five measurements we never obtained any count in the proton window, even at the highest energy of 750 eV per deuteron and for the longest acquisition time. An upper limit C_0 of the counting rate C can be calculated in that particular case where there is no count during an acquisition time T. The probability P for C to be lower than C_0 is given by

$$P=\int_0^{C_0} T\exp(-CT)dC=1-\exp(-C_0T)\,.$$

Then, for $T=4200$ s, and for a confidence level P of 95%, C_0 is calculated to be $7\times10^{-4}\ \mathrm{s}^{-1}$, 10 times below the corresponding result of BNL. Of course, this detection limit could be improved by increasing the acquisition time. However, we observed no significant differences between the background spectrum and all the spectra taken with the cluster beams, except for one run where accidental noise events have been unambiguously identified (even in that case there was no count in the proton window). This spectrum being excluded, we show in Fig. 2(c) the summation of all other spectra corresponding to an accumulation time of 8400 s. The resulting count rate in the energy range 1.4–7.4 MeV is within the error bar of the background of the detection system (measured over a time of 2×10^4 s). From this argument it is reasonable to conclude that with $D_n{}^+$ projectiles the proton yield per incident deuteron is 2 orders of magnitude below the BNL value, i.e., very difficult to measure under normal background conditions.

As mentioned above we also tried to observe *dd* fusion under impact of "deuteron-free" clusters on a deuterated target. Our negative result with incident $D_n{}^+$ clusters could be attributed to an insufficient density of the deposited energy. Consequently, $N_n{}^+$ clusters, the only heavy-atom cluster beam which could be produced within a short time, were good candidates for comparison with the $(H_2O)_n{}^+$ experiments. Here the N atoms play the role of O atoms in water clusters in the energy deposition process.

On the "direct" beam line B (see Fig. 1) fully accelerated projectiles of all masses reach the deuterated polyethylene target if the magnet is turned off. In experiments performed at 500 keV the beam pulse intensity reached values in the μA range. Under these conditions we did observe fusion, with a typical proton rate of 5 s^{-1}. However, with a magnetic field of 500 G, $N_{16}{}^+$ and lighter clusters are deflected from the target area and the intensity drops by a factor 20. Then the proton rate is reduced by a factor F of the order of 10^3. This can qualitatively be explained by an unexpected high beam contamination by deuterium (although the ion source was carefully purged after the $D_n{}^+$ experiment). Then we did measurements in which fusion events were separately counted during the cluster pulse and during a time window between the cluster pulses, i.e., at times where the incident beam is the "leak beam" composed only of atomic and molecular components. These measurements, during and between the pulses, yielded com-

TABLE I. Comparison between our experimental results for $D_n{}^+$ clusters with those of the BNL experiments. The BNL data have been normalized to the same number of incident deuterons at a given cluster velocity.

Energy per deuteron	400 eV			500 eV			750 eV	
	This work		BNL	This work		BNL	This work	BNL
Target	$TiD_{1.7}$	$TiD_{1.7}$	TiD	$TiD_{1.7}$	$(CD_2)_n$	TiD	$(CD_2)_n$	TiD
Incident cluster	$D_{250}{}^+$	$D_{300}{}^+$	$(D_2O)_{75}{}^+$	$D_{200}{}^+$	$D_{200}{}^+$	$(D_2O)_{60}{}^+$	$D_{200}{}^+$	$(D_2O)_{40}{}^+$
Cluster energy (keV)	100	120	300	100	100	300	150	300
Intensity (nA)	1.5	1.33		1.9	0.95		1.6	
Acquisition time (s)	1500	950		1800	3100		4200	
No. of incident deuterons (10^{15})	2.27	2.15	2.2	2.96	3.68	3.3	6.15	6.15
Counts in proton window	0	0	8	0	0	16	0	30

parable values of the factor F. This led us to the conclusion that the observed fusion is mainly due to beam contamination by deuterium. However, the calculation of F based on estimates of the cross sections for D^0 production, either by electron capture by D^+ or by dissociation of $D_2{}^+$, is unable to reproduce the measured F values. The closest prediction requires a $D_2{}^+$ contaminant beam (interacting with residual gas in the area between the electrostatic and magnetic analyzers), but F is still calculated to be larger than 10^4. Additional effects, related to scattering, could also play a role in measurements with such a short beam line. This shows that beam contamination is not easy to prevent in a direct beam line. Its contributions to the measured cluster fusion rate may be rather difficult to evaluate.

The possibility to induce fusion under cluster impact has also motivated several theoretical works. Basically, there are two ways for increasing the fusion rate at low incident energies (which can eventually cooperate), either the decrease of the Coulomb barrier between two colliding deuterons, or the increase of the kinetic energy of a single deuteron. The *dd* Coulomb barrier is decreased by electronic screening of the nuclear charges.[8,9] In the case of cluster impact the electron density would be increased due to the electrons brought into the target. But in our energy range one can estimate that the effect on the fusion rate remains negligible. On the other hand, various processes of high energy transfer to individual deuterons have been reviewed by Carraro *et al.*[10] Their calculations involving all possible collisions between D, O, and Ti atoms show that the fusion rate is not significantly increased compared to individual *d-d* collisions. Another scenario is the "equilibrium" approach, where it is assumed that the energy of the incident cluster is distributed over a certain number of projectile and target atoms. In the "thermonuclear model" it is assumed that fusion occurs in the hot gas during a confinement time determined by the expansion rate. The calculated fusion rate is comparable to the BNL data but does not reproduce the cluster-size dependence. Echenique, Manson, and Ritchie,[11] who also used the thermodynamical approach, have demonstrated the role of the few fast deuterons in the tail of the Maxwell-Boltzmann distribution. Their calculations support the BNL results. Another variation is the "thermal spike" model[10] where it is assumed that only a part of the incident energy is thermalized. Here the mass dependence of the fusion rate is rather well reproduced, but the absolute values are calculated to be 5 orders of magnitude lower than the BNL fusion rates. However, the validity of these thermodynamical models is questionable, particularly when a large fraction of the incident energy is dissipated into electronic excitations. Moreover, fractoemission processes have been involved to explain cold fusion,[12] but we think that such an interpretation of cluster-impact fusion is questionable, although worthwhile to explore.

As a conclusion, $D_n{}^+$ cluster impact on solid deuterated targets did not yield measurable *dd* fusion rates. If one excludes screening effects, the thermonuclear process, which is in our case the most relevant mechanism, seems to be unable to induce observable fusion rates with light-ion clusters.

Multiple knockon and thermal-spike processes are expected to play a role only with heavy-ion clusters. Any further experiment should be performed with deuterated heavy-ion clusters with an experimental configuration which permits, after acceleration, a reliable mass analysis of the beam.

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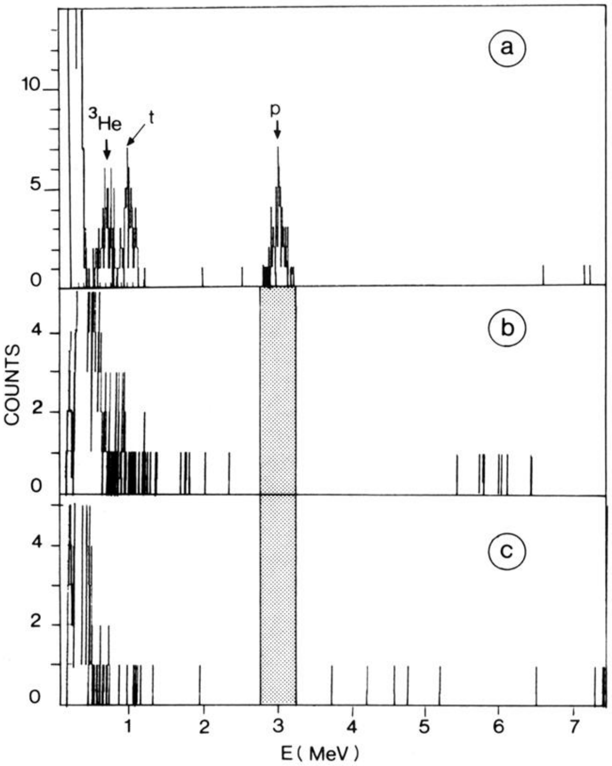

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