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UPPER LIMITS ON (d,d) FUSION YIELDS UNDER CLUSTER IMPACT

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ABSTRACT :

Owing to the results of Beuhler et al [1] on cluster impact fusion we undertook experiments with accelerated $(D_2)_n^+$, $(N_2)_n^+$ and $(CD_4)_n^+$ clusters to investigate a possible enhancement of (d,d) nuclear fusion rates in cluster impact on deuterated titanium and polyethylene targets. With ($n \geq 100$) clusters containing deuterium atoms of energies below 1 keV / D, no fusion event was detected. In these conditions, the experimental background for the detection of the 3 MeV protons emitted in the fusion reaction sets an upper limit to the fusion yield of 10^{-13} per cluster impact. This is 4 orders of magnitude below the proton yields previously reported by Beuhler et al for $(D_2O)_n^+$ clusters in the same energy range. Nevertheless, the main difficulty of this kind of experiments is to avoid experimental artefacts that may result from the presence of light and fast deuterated components in the beam at the target site. Experiments with energy analysed $(CD_4)_n^+$ and $(N_2)_n^+$ clusters have evidenced that a magnetic deviation of light projectiles in front of the target is not an absolutely safe procedure. It is shown that a D^+ acceleration process might explain why the time - of - flight experiments by Beuhler et al [4] have not been able to reveal the presence of accelerated light ions at the target site.

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I . Introduction.

The first experimental evidence for cluster impact fusion was reported in 1989 by Beuhler et al [1] at Brookhaven National Laboratory. With a beam of $(D_2O)_n^+$ clusters accelerated to a total energy of ~ 300 keV and for n values between 25 and 1300, they observed 3 MeV protons from the d(d,p)t nuclear fusion reaction induced in deuterated titanium targets. The proton yield deduced from their experimental counting rates was of the order of 10^{-11} per cluster impact. This was more than 10 orders of magnitude higher than the fusion yields expected from the values of the nuclear cross section extrapolated to energies per deuteron lower than 1 keV! Moreover, fusion was also observed with $(H_2O)_n^+$ cluster beams. Although the fusion rates were then reduced to 5% of the $(D_2O)_n^+$ fusion rates, this last result suggested that the presence of heavy atoms like oxygen in the cluster was needed for the enhancement of the fusion rate up to a measurable value. The observation of cluster impact fusion was confirmed in similar experiments with $(D_2O)_n^+$ clusters performed at Stanford Research Institute by Bae et al [2] and in additional experiments by the BNL group aimed to prove the absence of artifacts [3]. The identification of the projectiles producing the fusion reaction by time - of - flight measurements [4] appeared to be the most convincing experiment.

The publication of an erratum [5], in which the BNL group admitted that their previous results were overestimated by at least two orders of magnitude, led to a better agreement with our experiments with $(D_2)_n^+$ [6] and $(CD_4)_n^+$ [7] clusters where the counting rates never exceeded the background level of the detection setup. Although the cluster beam was electrostatically and magnetically analysed after acceleration, our experiments with $(N_2)_n^+$ and $(CD_4)_n^+$ clusters have shown that beam contamination by light deuterated projectiles was not totally excluded and was responsible for the few detected events. Especially when a direct beam line is used, neither a sweeping magnet in front of the target (as in our experiment with $(N_2)_n^+$ clusters) nor a time - of - flight setup (as in the Brookhaven experiment) are able to totally eliminate artefacts originating from light projectiles.

II . Experimental

The cryogenic source of the Lyon Cockroft-Walton accelerator is designed for cluster production by expansion at given pressure and temperature of a gas through a small nozzle. The temperature is set by a first cooling enveloppe at a suited value depending on the nature of the clusters to be produced (liquid neon was used for the production of $(H_2)_n$ and $(D_2)_n$ clusters, liquid nitrogen for $(N_2)_n$ and liquid argon for methane clusters). An external reservoir is filled with liquid helium which is also used in a cryogenic pumping system at the exit of the nozzle. A fast electromagnetic valve is used to produce clusters during pulses of typically 100 ms duration every second (10% duty cycle). After passing through successive skimmers, the clusters are ionized by electron bombardment before entering the acceleration section.

Various setup have been used, depending on the needed cluster mass. The experimental setup used for the $(D_2)_n^+$ and $(N_2)_n^+$ experiments is shown in Fig. 1. After acceleration at a given energy, the clusters are deflected by a 74° electrostatic analyser and then by a 16° bending magnet of a mass-energy product of 60 MeV.u. With $(D_2)_n$ clusters, the magnet allows mass selection on the beam line A. With heavier clusters, like $(N_2)_n$, the experiment was performed on the direct beam line B, the magnetic field being applied to deflect the low mass projectiles from the target area (at a total energy of 500 keV and with a magnetic field of 0.05 T, only projectiles with masses above M = 225 u can theoretically reach the target).

For the experiments with $(CD_4)_n^+$ clusters, a new beam line was built as shown in Fig. 2. With a 90° electrostatic analyser and a more powerful magnet (1500 MeV.u mass-energy product), clusters of masses up to 3000 u can be directed into the 10° beam line towards the reaction chamber. The mass resolution defined by a collimator of 14 mm in diameter at the entrance of the chamber is $\Delta M / M = 23$ % at a total energy of 520 keV. This allows to obtain beam intensities in the nA range at the target site.

The deuterated target was tilted at 45° with respect to the beam axis. The charged particles produced in the (d,d) reactions (p, t and ^3He) were detected at 90° by a 900 mm^2 Si surface barrier detector (solid angle ~ 1.5 sr). Due to the low energy background [6] only signals in a proton energy window between 2.8 and 3.2 MeV were taken as a signature of (d,d) fusion reactions. A gate signal was used in order to trigger the counting during or between the beam pulses.

III . Upper limits on fusion yields under $(D_2)_n^+$ and $(CD_4)_n^+$ cluster impact.

The background level of the detection system was measured with zero voltage at the accelerator terminal. For a net acquisition time of 2×10^4 s, no event was detected in the proton energy window. From the background recorded in the 1.4 - 7.4 MeV energy window (15 counts) and from the hypothesis of an uniform distribution of background events within this energy range, a background count rate of at most 5×10^{-5} s^{-1} can be estimated.

With 100-150 keV $(D_2)_n^+$ clusters (n = 100, 125 and 150) bombarding deuterated titanium and polyethylene targets at beam intensities of 1-2 nA and for a number of incident clusters from 7×10^{12} to 3×10^{13}, we never obtained any count in the proton energy window, even at the highest energy of 750 eV / D. For a confidence level of 95%, these results correspond to an upper limit of the proton yield of the order of 10^{-13} per cluster impact. This is illustrated in Fig. 3 for experiments with $(D_2)_{100}^+$ clusters at 100 and 150 keV on deuterated polyethylene targets, together with the detection limit calculated from the background count rate.

With $(CD_4)_{110}^+$ and $(CD_4)_{145}^+$ clusters, a more reliable comparison with $(D_2O)_n$ cluster experiments was expected due to the presence of a heavy atom (C instead of O) in the projectiles. At a total energy of 520 keV, with beam intensities ranging from 1 to 10 nA and for a number of incident clusters between 5×10^{12} and 5×10^{13}, the number of detected events in the proton energy window was zero, or only a few units. The proton yields per cluster impact, determined by summing up all the counts obtained with the same projectile, are shown

in Fig. 3 by the experimental data points (solid circles, the error bars give the upper limit for a confidence level of 95 %). As for the $(D_2)_n^+$ experiments, the upper limits are of the same order of magnitude as the detection limit fixed by the background level, which is of the order of 10^{-13} per cluster impact. That means that we did not observe any effect due to the presence of heavy atoms in the deuterated clusters using $(CD_4)_n^+$ clusters instead of $(D_2)_n^+$ clusters.

IV . Contamination by light projectiles.

A crucial point when one works with cluster beams is the purity of the beam at the target site. Such projectiles can suffer dissociation in collisions on collimator edges or on molecules of the residual gas, leading to production of neutrals and low mass ions following trajectories that are difficult to model.

We observed such beam contaminants in an experiment performed on a direct beam line with $(N_2)_n^+$ clusters. When the small magnet in front of the target was turned off, fully accelerated projectiles of all masses contributed to intensities in the μA range at the target site. Under these conditions, fusion events were detected with a count rate between 1 and 10 per second. However, beam contamination by deuterons being not excluded after previous experiments with $(D_2)_n^+$ clusters, we used the magnet to deflect the lightest projectiles from the target direction. With a magnetic field of typically 0.05 T (deflecting $(N_2)_8^+$ and clusters of lower masses off the target area for a total energy of 500 keV) the proton rate was reduced by 2 - 3 orders of magnitude while the intensity droped only by about one order of magnitude. The first evidence for the main contribution of light projectiles to the fusion rate can be seen from the data, reported in Table 1, obtained with and without a magnetic field, and in coincidence with the beam pulses. It must be noted that N^+, N_2^+ and N_3^+ are the predominant species in the accelerated beam, with intensities more than two orders of magnitude larger than those of the high mass clusters. Two different processes can induce fusion reaction: either direct reactions produced by contaminant deuterons in the beam, or reactions produced by target deuterium atoms recoiling in knock-on collisions with light nitrogen ions of high

velocity. Moreover the proton count rate was also measured in a time window set between the beam pulses, i.e. when a "leakage beam" exists, that results from the ionisation of the residual gas upstream from the acceleration section. As shown in Table 1, under these conditions the proton rate (per second) was almost the same. In addition, if the proton rate is related to the number of impacts, it increases by a factor of more than ten. This indicates that the ions of the leakage beam are more efficient in producing fusion reactions than the cluster beam. These results suggest the presence of contamination by fast deuterons. D^0 production by dissociation of D_2^+ ions in collisions with the residual gas in the area between the electrostatic deflector and the magnet, and also D^+ scattering leading to complicated trajectories are highly probable. Both processes may explain the limited efficiency of the magnet in preventing light projectiles in the beam from reaching the target.

Another experimental evidence for such an effect was also observed in experiments with methane clusters, although the beam line was deflected at 10°. Experiments were performed with a "leakage" beam produced when the source nozzle was at room temperature. In these conditions, the ions result from the ionisation or the breaking up of CD_4 molecules, and the main deuterated components of the beam entering the analysing magnet are then CD_4^+, D^+ and D_2^+ ions. The intensities of these various beams are listed in Table 2 (third column). With a high magnetic field set to deviate these light ions from the target direction, the target current was not measurable, as expected, but a few events were counted in the proton window. At 1.9 T, the nominal value of the field required to send $(CD_4)_{145}^+$ clusters towards the target, the measured proton count rate was $1.8 \times 10^{-2}\ s^{-1}$. With permanent magnets placed at the exit of the electrostatic deflector ((D) in Fig. 2) in order to sweep out the lightest species before the entrance of the magnet, the various beam currents and the proton count rate were reduced. The values are shown in Table 2 (fourth column). The fact that at 1.9 T the reduction factor is the same for the proton count rate and for the CD_4^+ beam current indicates that even with a high magnetic field, fusion occurs at a rate which is proportional to the number of CD_4^+ ions entering the magnet. D^+ or D^0 fragments, resulting from dissociation of CD_4^+ ions through collision processes within the magnet, are probably responsible for the detected fusion events.

In experiments with high mass $(CD_4)_n^+$ clusters, the presence of CD_4^+ ions at the entrance of the magnet can lead to a proton count rate of $(1.6 / 24) \times 10^{-3} = 7 \times 10^{-5}\ s^{-1}$ per nA of CD_4^+ ions. Since the intensity of CD_4^+ ions in the total beam is 8 times higher than the intensity of $(CD_4)_{110}^+$ and $(CD_4)_{145}^+$ clusters, the proton fusion yield resulting from contaminants of the cluster beam has been estimated to be of the order of 10^{-13} per cluster, as shown in Fig. 3 (open circles). As a consequence the few events detected in the $(CD_4)_n^+$ experiments can be attributed to high velocity components of the beam.

In their efforts to reject any artifact for interpreting the origin of fusion events, the BNL group has performed time - of - flight experiments [4] that should be able to identify any light projectile contamination. Since they have recognized [5] that the large fusion rates measured with $(D_2O)_n^+$ clusters were mostly due to artefacts probably related to beam contamination by high velocity components, their time - of - flight results remain intriguing. According to the distances involved in their experiments (see Fig. 1 of ref. 4) and since the production of fusion protons coincides within about 1 μs with the arrival of the clusters on the target, any projectile fragmentation leading to deuteron production before acceleration to the final energy should be detected. On the other hand, any dissociation during acceleration should have induce a proton peak wider than the electron peak and shift it towards lower time of flight values. Indeed, instead of the typical 11 μs flight time of $(D_2O)_{115}$ clusters, D^+ ions will take 1 to 10 μs less to reach the target, depending whether they are produced at the end or at the entrance of the acceleration column. Processes involving fragments emitted and re-accelerated after collisions with collimators [8] or with the target itself [9] have been evoked but have not been quantified. A D^+ acceleration process resulting from collisions of clusters with the collimator edges at the end of the column is sketched in Fig. 4. Mass selected $(D_2O)_n^+$ clusters enter the acceleration column of length l_0 terminated by a defining collimator aperture O and impinge on the target T ($OT = l_1$). Both collimator and target are at the negative potential $-V_0$. As part of the cluster beam collides with the edges of the collimator, low energy D^- emission may result from the fragmentation of the deuterated clusters and from the sputtering of the deuterium - implanted material of the collimator. Emission of negative hydrogen ions is well

known in low energy ion sputtering experiments, and is more efficient when cluster ions are used [10]. These D^- ions, accelerated backwards in the column, have a significant probability to experience stripping in further collisions with the residual gas since the ($D^- \rightarrow D^+$) cross section is quite large, in N_2 gas a broad maximum of the cross section (~ 3 x 10^{-16} cm^2) is found at 200 keV [11]. At a collision location I, at a distance OI = x from the aperture O, the resulting D^+ ions are slowed down and, if $x < l_0 / 2$, are stopped at P (OP = 2x), accelerated forward, and might finally reach the target with a x dependent kinetic energy at most equal to eV_0. In the particular case, discussed in ref. 4, of 520 keV $(D_2O)_n^+$ clusters, the figures are the following: with l_0 = 1.2 m and l_1 = 0.25 m, a pressure of 2 x 10^{-7} Torr in the column, the ionisation probability of D^- is ~ 7 x 10^{-5}, and the time t needed for the path OIPOT varies from ~ 0.3 μs for x = 0.2 m to ~ 0.75 μs for x = 0.6 m. On the other hand the time needed for clusters to fly from the aperture O to the target T is ~ 1.2 μs. These values are quite small with respect to time delays measured in ref. 4 and rather close to each other: BNL time-of-flight measurements could not reveal this plausible scenario.

V . Conclusion.

In experiments with $(D_2)_n$ and $(CD_4)_n$ clusters of large n values ($n \geq 100$) accelerated on deuterated targets at energies per deuteron $E_D \leq 750$ eV, the proton counting rates never exceeded the experimental background. Then, the upper limit of the fusion yield corresponds to our experimental detection limit which is of the order of 10^{-13} per cluster impact. This is 12 orders of magnitude above the fusion yield calculated from the value of the binary (d,d) cross section at the E_D energy of 750 eV and 4 orders of magnitude below the fusion yield previously measured with $(D_2O)_n^+$ [1]. One of the main conclusions of the experimental studies on cluster impact fusion is that the risk of artefacts is high in these experiments where extremely low counting rates are expected. In particular, a total elimination of light and fast fragments in the cluster beam is very difficult to achieve.

The determined upper limit of the fusion yield does not contradict the theoretical attempts to obtain a significant enhancement of the fusion rate, since most of them were unable to reproduce the BNL experimental results. Among these theoretical approaches, acceleration processes of some deuterons by successive collisions with cluster and target heavy components, have been evidenced in various molecular dynamics simulations [12,13]. However, the predicted enhancements of the fusion rate are out of reach of the present experimental capabilities.

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Figure Captions

FIG. 1 : Experimental setup for the $(D_2)_n^+$ (line A) and for the $(N_2)_n^+$ (line B) experiments.

FIG. 2 : Experimental setup for the $(CD_4)_n^+$ experiments. (A) reaction chamber, (B) and (C) collimators, (D) permanent magnet location (see text).

FIG. 3 : Proton fusion yield per cluster impact for $(CD_4)_{110}^+$ and $(CD_4)_{145}^+$ at a total energy of 520 keV and for $(D_2)_{100}^+$ at a total energy of 100 and 150 keV. Experimental data points (solid circles), detection limit deduced from the background count rate (dashed bars) and proton yields from light contaminants (open circles). Only upper limits are indicated when no count was obtained. (all evaluations with 95% confidence level).

FIG. 4 : Proposed D^+ acceleration process in BNL experiments.

Table Captions.

Table 1 : Number of detected protons in various experimental conditions with $(N_2)_n^+$ clusters, for a given number of impacts and for a net acquisition time, respectively.

Table 2 : Experimental results obtained with the molecular methane beam. Effect of permanent magnets on the current of the various beams, and on the proton count rate measured with 1.9 T (see text).

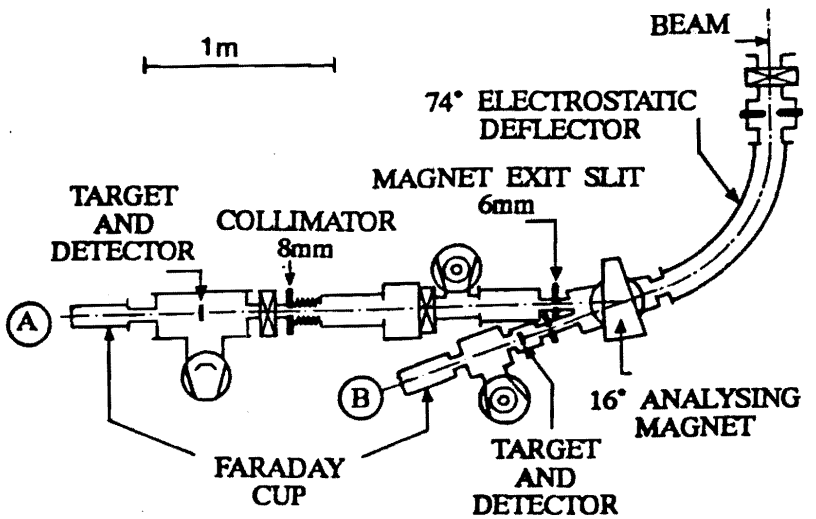
1m
BEAM
74° ELECTROSTATIC DEFLECTOR
MAGNET EXIT SLIT 6mm
TARGET AND DETECTOR
COLLIMATOR 8mm
A
B
16° ANALYSING MAGNET
FARADAY CUP
TARGET AND DETECTOR

FIG. 1

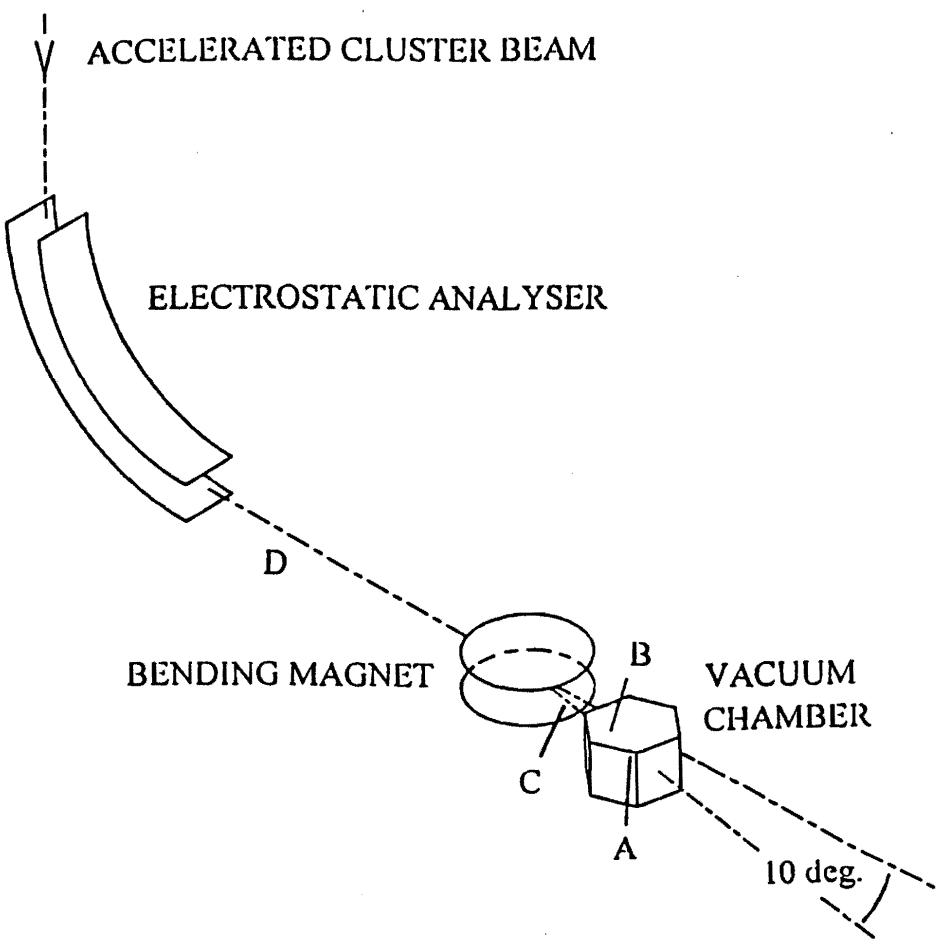
ACCELERATED CLUSTER BEAM
ELECTROSTATIC ANALYSER
D
BENDING MAGNET
B
VACUUM CHAMBER
C
A
10 deg.

FIG. 2

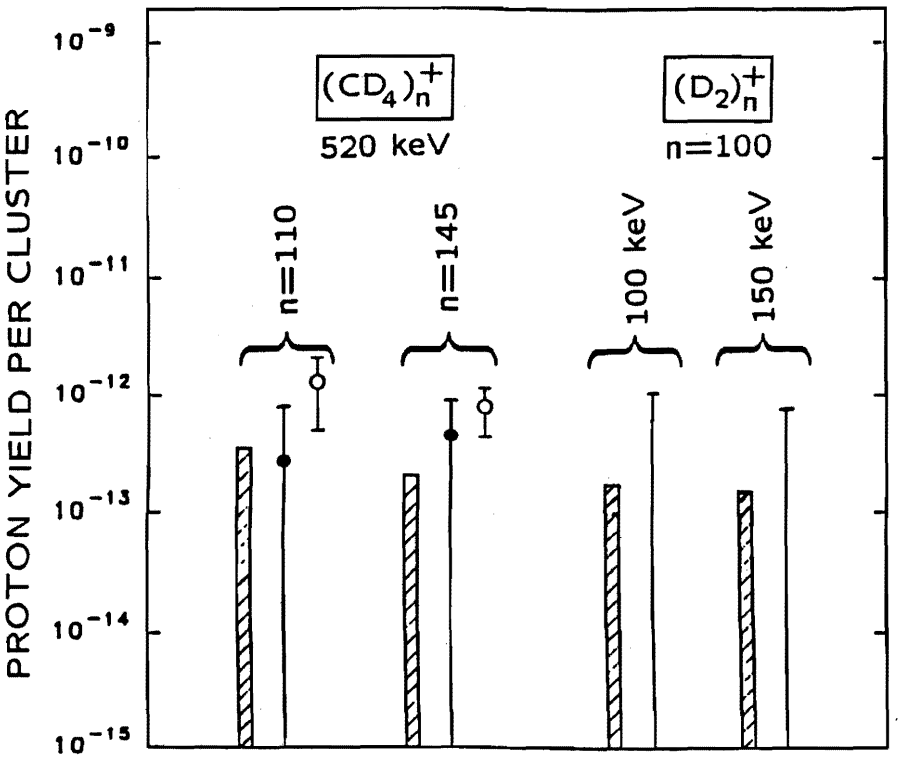
PROTON YIELD PER CLUSTER
10^{-9}
10^{-10}
10^{-11}
10^{-12}
10^{-13}
10^{-14}
10^{-15}
$(CD_4)_n^+$
520 keV
$(D_2)_n^+$
n=100
n=110
n=145
100 keV
150 keV

FIG. 3

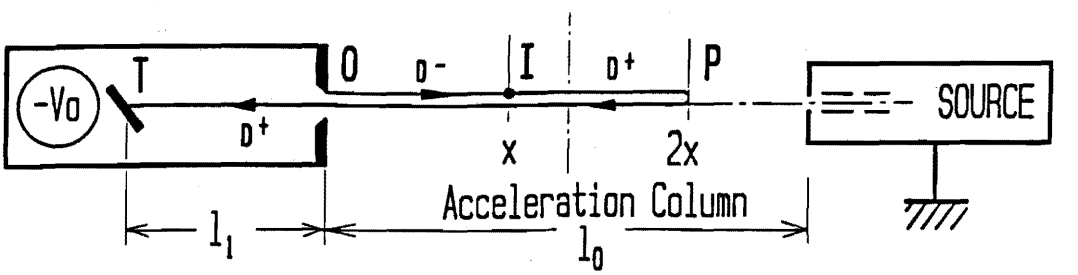
-Vo
T
O
D-
I
D+
P
SOURCE
D+
x
2x
Acceleration Column
l_1
l_0

FIG. 4

Table 1

Experimental conditions	I pulse target (nA)	p / 10^{10} impacts		p / s	
		in the beam pulses	between the beam pulses	in the beam pulses	between the beam pulses
	750	$1.8\ 10^{-2}$	$2.5\ 10^{-1}$	1.5	1.8
500 keV	6400	$1.9\ 10^{-2}$	$1.3\ 10^{-1}$	6.3	5.6
0 tesla	500	$2.0\ 10^{-2}$	$1.4\ 10^{-1}$	0.8	1.0
	2700	$1.0\ 10^{-2}$	$2.3\ 10^{-1}$	2.7	2.5
500 keV 0.03 tesla	1200	$3.5\ (\pm 0.9)\ 10^{-4}$	$4.9\ 10^{-1}$	$4.3\ (\pm 1)\ 10^{-2}$	$4.3\ 10^{-2}$
500 keV 0.035 tesla		$2.6\ (\pm 1.2)\ 10^{-4}$	$3.4\ (\pm 1.7)\ 10^{-2}$	$1.0\ (\pm 0.4)\ 10^{-2}$	$2.3\ (\pm 1.2)\ 10^{-3}$
500 keV 1.2 tesla	28	$7.7\ (\pm 7.7)\ 10^{-4}$	$2.4\ (\pm 1.0)\ 10^{-1}$	$2.3\ (\pm 2.3)\ 10^{-3}$	$1.8\ (\pm 0.8)\ 10^{-3}$
300 keV 0 tesla	2560	$3.8\ 10^{-3}$	$1.2\ 10^{-1}$	1.6	0.5
300 keV 0.05 tesla	160	$3.6\ (\pm 1.9)\ 10^{-4}$	$1.1\ (\pm 0.5)\ 10^{-1}$	$9\ (\pm 0.5)\ 10^{-3}$	$3.4\ (\pm 1.7)\ 10^{-3}$

Table 2

Magnetic field (T)	Projectile corresp. to the analysed mass	Beam currents (nA) permanent magnets out	Beam currents (nA) permanent magnets in	Reduction factor
0.05	D^+	37	0.023	1630
0.07	D_2^+	4.5	0.01	450
0.16	CD_4^+	300	24	12.5
		Proton rates (s^{-1}) permanent magnets out	Proton rates (s^{-1}) permanent magnets in	
1.9	$[(CD_4)_{145}^+]$	1.8×10^{-2}	1.6×10^{-3}	11.2

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