Deuterium-Deuterium Fusion Dynamics in Low-Density Molecular-Cluster Jets Irradiated by Intense Ultrafast Laser Pulses

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Following the interaction of superintense, short pulse lasers and plasmas, ions can be accelerated to velocities sufficient to drive nuclear fusion reactions, in particular, by the process of Coulomb explosion of clusters [T. Ditmire *et al.*, Nature (London) **398**, 491 (1999)]. We show here how short bursts of neutrons can be produced using a jet of low-density deuterated methane clusters. Ion velocity distributions were simultaneously measured by a Thomson parabola mass spectrometer, demonstrating deuteron energies up to 120 keV. We show that, in such conditions, nuclear fusion will occur not only in the hot plasma core, but also in the cold outer region by collision processes.

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The production of neutrons in laser plasmas by fusion of fast deuterium ions has been observed since 1970 with energetic nanosecond laser pulses [1]. With new techniques of chirped pulse amplification, such experiments formerly reserved to large scale installations now become accessible with smaller facilities. In particular, the production of neutron bursts with ultrashort titanium-sapphire lasers is a rapidly growing field of research, first, with ion acceleration produced in relativistic channels on solid targets [2,3], and, second, by explosions of cryogenic deuterium cluster jets [4]. The latter method results from recent demonstrations that cluster explosion in intense femtosecond-laser fields can produce ionic fragments with considerable kinetic energies [5,6].

Such laser-induced nuclear fusion could, in principle, be used to perform time-resolved neutronic experiments, for instance, of material damage under rapid neutron irradiation. To that aim, however, the neutron yields will have to be very strongly enhanced, which will require in turn to be able to optimize the fusion cross section, by tailoring the ion energies resulting from cluster explosions. The effect of a first important parameter, the D_2 cluster size, was investigated by Zweiback *et al.* [7]; however, the kinetic energies of the ions were estimated to only 2.5 keV per deuteron [4], whereas optimal kinetic energies are much higher, in the range of 50 to 100 keV. Indeed, in a recent theoretical work, Last and Jortner have shown interest in using clusters of heteronuclear molecules to enhance the deuteron kinetic energies [8], which may lead to a dramatic increase of fusion yields.

In this Letter, we report experimental studies of femtosecond laser-induced production of neutrons with a novel target, namely, deuterated methane clusters, which allows one to produce large deuterated clusters very easily and to reach optimal values of fusion cross sections. We present for the first time a correlated study of neutron yields and ion energy distributions, diagnosed by a Thomson parabola mass spectrometer. This permits us to investigate whether the nuclear reactions are produced in the hot core of the plasma as previously assumed, or if another mechanism is also involved.

Neutron production happens in two steps. In the first step, deuterated clusters are irradiated by an intense femtosecond laser source, which strips the clusters of most electrons, leading to cluster explosion. Under the effect of thermal pressure and Coulomb forces, ions get accelerated to keV-range energies, sufficient to drive fusion reactions in a second step, as follows:

$$\mathrm{D} + \mathrm{D} \rightarrow {}^{3}\mathrm{He}(0.82\ \mathrm{MeV}) + n(2.45\ \mathrm{MeV}). \qquad (1)$$

The corresponding cross section increases sharply for D energies beyond 5 keV, and starts saturating at about 50 keV [9]. It is hence crucial to be able to obtain large deuterated clusters in order to increase the ionic energies to at least a few tens of keV. To this aim, we have used clusters of deuterated methane (CD_4) that present major advantages over D_2 clusters: (i) thanks to the high molecular polarizability, large scale clusters are expected to be produced even at room temperature, and (ii) the molecules contain heavy carbon atoms, whose ionization may result in highly charged ions acting as an expeller with respect to D^+ ions. Experimentally, CD_4 clusters are produced by supersonic expansion with a jet throat of a 500 μm conical nozzle, of 25 mm length. The 5 mm output diameter of the cone defines the interaction length, with a beam focus 3 mm below the nozzle. At 50 bars of backing pressure, the average density at the output of the jet is estimated to be $2 \times 10^{17}/\mathrm{cm}^3$, a 100 times lower than in [4], so that no

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propagation effect is expected. From the Hagena parameters [10], we estimate an average cluster size of 2×10^5 molecules. No cryogenic cooling was performed to reach those parameters.

The CD_4 clusters are irradiated by a chirped pulse amplification titanium-sapphire laser system with energy up to 800 mJ at 820 nm and 35 fs pulse width for a repetition rate of 10 Hz [11]. An off-axis parabolic mirror ($f = 2000$ mm) focuses the laser pulse to an 85 μm diameter spot. This large focal area allows us to optimize the interaction volume with the cluster jet, while keeping intensities up to several 10^{17} W/cm^2. The diagnostics are a Thomson parabola (TP) mass spectrometer for measurement of the velocity and energy distributions of carbon ions and deuterons [12], and a time-of-flight (TOF) spectrometer sensitive to neutrons and gamma rays. The TOF scintillator is constructed with a 5 cm thick and 12 cm in diameter organic liquid scintillator (NE 213) coupled to fast photomultiplier tubes, allowing for fast pulse shape discrimination (PSD) to distinguish between incident neutrons and gamma rays [13]. Some shielding against gamma rays is, however, provided by housing the detector in a lead box of 5 cm thickness. The TOF and PSD have been calibrated against a test exposure of ^{252}Cf fission source emitting 2.14 MeV neutrons and gamma rays.

TOF spectra are shown in Fig. 1 for two distances from the gas jet, 1.55 and 1.85 m. The mean arrival times exhibit a difference of 13 ± 1.0 ns, which for the 30 cm flight difference corresponds to a neutron energy of 2.76 ± 0.4 MeV, fully consistent with the 2.45 MeV energy expected for fusion neutrons. To confirm further the production of neutrons, we put a paraffin block between the source and the detector, which resulted in a scintillator signal reduction from 42 to 1 count for 100 laser shots.

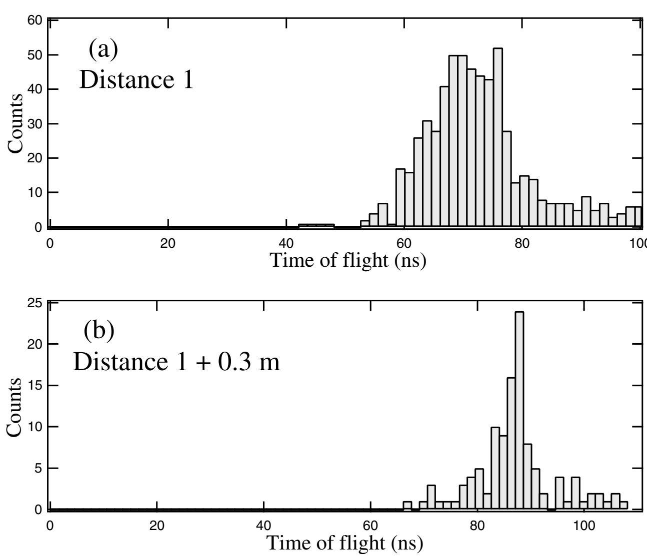

FIG. 1. Time of flight spectra of neutrons at two positions separated by 30 cm. The temporal width of the detection system is estimated to 10 ns.

Finally, replacing CD_4 by either CH_4 or cryogenically cooled D_2 suppresses completely the neutron signal. These results demonstrate that 2.45 MeV fusion neutrons are produced via reaction (1), for which the good clustering properties of CD_4 are crucial.

In order to analyze the energy distribution of the ions involved in the nuclear reaction, we use a Thomson parabola mass spectrometer [12], whose axis is oriented perpendicular to the laser axis. The ions pass through a 500 μm pinhole located 500 mm from the jet, are deflected by parallel electric and magnetic fields, and end up on a microchannel plate (MCP) associated with a charge coupled device camera. The impacts on the screen are located on a parabola shaped curve, determined by the q/m ratio. From the geometric parameters, the electric and magnetic fields values, and the deviation measured, we can deduce the absolute ion energies.

Figure 2 shows an example of a raw Thomson parabola image. Three separate parabolic lines appear, corresponding to D^+, C^+, and C^{2+} ions. The parabola origin point arises from neutral particles or x rays that form a pinhole image of the active plasma region. The C^{2+} yield is much weaker than the C^+ yield. Moreover, no carbon ions of higher charges are detected, whereas the carbon atoms from CD_4 would be expected to be largely stripped, resulting, in principle, in large populations of all charge states between C^+ to C^{4+}. This apparent paradox can be resolved by examination of the energy spectra of the ion species.

Figure 3 thus presents the measured energy distributions of (i) D^+ and (ii) C^+ and C^{2+} ions, obtained for a laser energy of 400 mJ, and considers a flat response in ion energy of the MCP [14]. The deuteron spectrum exhibits a maximum around 45 keV, then presents a rapid decay down to the noise level at 120 keV. In contrast, C^+ energy distribution is maximum at about 110 keV, much higher than D^+ energy. If C^+ was directly originating from the cluster

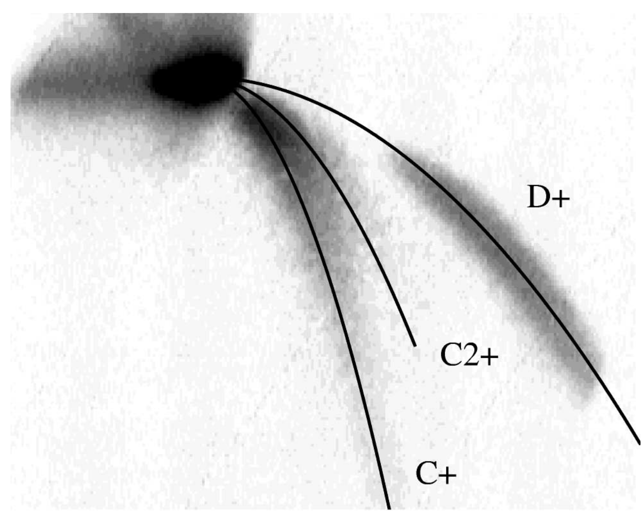

FIG. 2. Thomson parabola image of the ions resulting from the interaction. D^+, C^+, and C^{2+} ions are clearly visible.

Coulomb explosion, its energy would be about identical to that of D^+, as they have the same charge state and share the same electrostatic potential at the initial step of the explosion. The higher values of C^+ energies indicate that C^+ was mostly formed by electron recapture on ions of higher charge states that originated from the cluster explosion. Note that electron recapture cross sections show indeed a maximum of about 20 keV [15]. However, the ratio of C^+ to D^+ kinetic energies tends to indicate that the spatially averaged initial charge state of carbon at the onset of Coulomb explosion reaches only 2 to 3, below the expected value of 4 at saturation of L-shell electron ionization. This may reflect a geometrical averaging: the TP collects all ions created both on axis and in the wings of the focal spot, at which position the intensity is too low to saturate L-shell ionization.

The dashed line in Fig. 3 indicates a Maxwellian fit to the D^+ energy spectrum. From the fit, an effective temperature of 45 keV is inferred. The sudden drop in the distribution below 25 keV is an experimental artifact, due to the physical edge of the MCP; the high energy tail is close to a Maxwellian, with a slightly faster decay. Note that, even if thermalization occurs locally in the plasma, the experimental result is averaged over the whole focal region, so that the measured distribution is a weighted summation of the spatially varying distributions. Assuming a transverse Gaussian profile of effective temperatures to mimic the Gaussian laser intensity profile, it is possible to fit the measured distribution with a central temperature of 90 keV. For our cluster parameters, the scaling laws derived by Last and Jortner [8] predict an energy of 115 keV, in good agreement with our data. Of course, a more rigorous comparison should take into account the wide span of cluster sizes. Moreover, any elastic collision between the fast ions and colder atoms will tend to decrease the ion energy, so that the final average energy may be slightly underestimated.

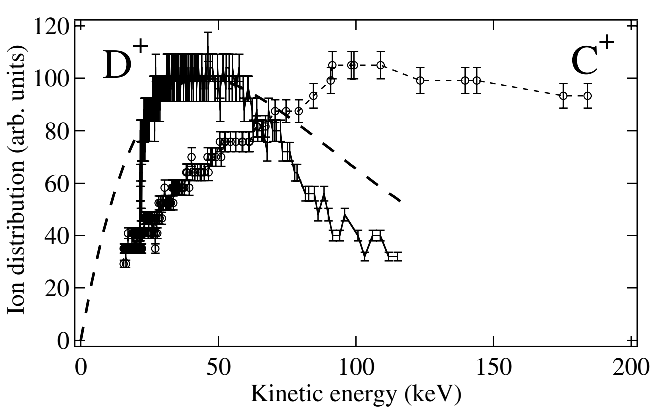

FIG. 3. Energy distributions of D^+ and C^+ ions measured by the Thomson parabola. The dashed line indicates a fit of the D^+ distribution by a Maxwellian profile.

Figure 4(a) shows the intensity dependence of the neutron yield measured, scaled to the full 4π solid angle, to be compared with the most probable energies measured for C^+ and D^+ versus intensity, displayed in Fig. 4(b). The neutron yield increases continuously, with a power law $I^{2.2}$ fitting best to the last three points. The estimated number of neutrons generated per shot at maximum intensity is 7000. The energy distribution of D^+ is almost linear, from 10 to 50 keV, while the C^+ distribution is steeper, this behavior resulting from both an increase in kinetic energy with increasing laser intensity for each charge state and an increase in the average charge state.

Surprisingly, the neutron yields measured here are almost identical to those reported in Ref. [4], although our jet density was lower by two decades, implying collision probabilities 10^4 times smaller. However, the ion kinetic energies are also much higher, implying larger fusion cross sections. To weigh both effects, we display as a dashed line in Fig. 4(a) the neutron number N expected by collision within the plasma [4], according to

$$N = n^2 \langle \sigma v \rangle_T V \Delta t, \qquad (2)$$

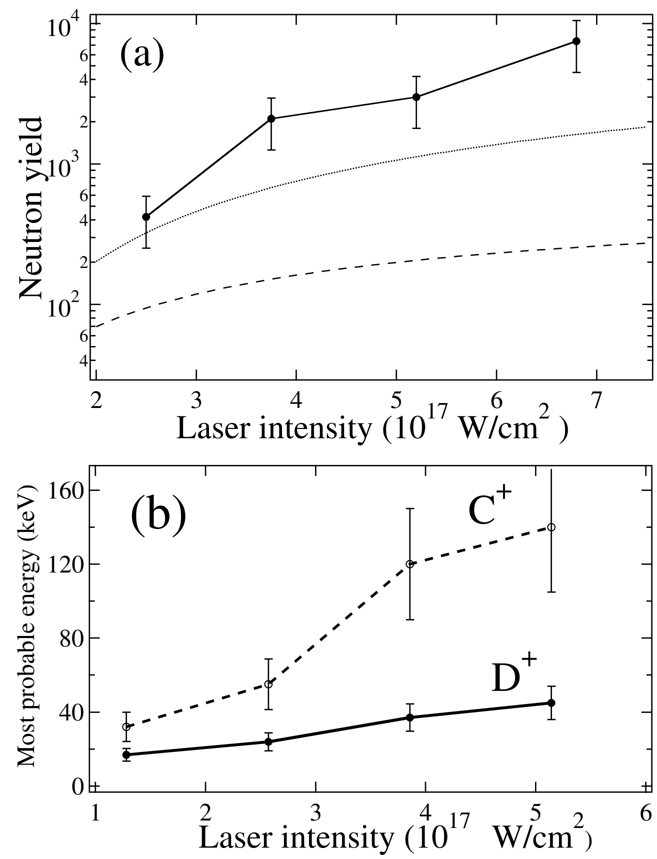

FIG. 4. (a) Intensity dependence of the total neutron yield over $4\pi sr$; theoretical estimates assuming central plasma model (dashed line) and the lateral collision model (dotted line). (b) Most probable energies of the measured distributions of C^+ and D^+.

TABLE I. Neutron yields measured at different angles A (in degrees) relative to the forward laser direction. Statistical error bars are indicated.

Angle from laser axis (deg)	Neutron yield
0	1140 ± 120
54	2331 ± 145
90	1616 ± 180

where n is the cluster gas average number density, V is the plasma volume, and Δt is the plasma disassembly time. The Maxwellian plasma reactivity $\langle \sigma v \rangle_T = (1/Z) \int \sigma[v(E)] v(E) \exp(-E/2kT)\, dE$ is computed for collisions between pairs of particles, each following a thermal velocity distribution at temperature T, while $\sigma(v)$ is tabulated for one particle at rest. As a result, the summation has to be performed with an effective thermal energy of $2kT$ [9]. The intensity dependence of the effective ionic temperature was considered linear, in order to fit Fig. 4(b). Obviously, the result of Eq. (3) comes short by 2 orders of magnitude of accounting for the experimental results. We propose to consider, therefore, a different process, in which deuterons created in the core of the plasma are ejected sideways and collide with much colder nuclei of the plasma corona or simply with nonclusterized atoms of the jet. The neutron yield expected by this process can be simply written as

$$N = n^2 \langle \sigma \rangle_T V R, \tag{3}$$

where $R = 2.5$ mm is the jet radius, and the Maxwellian average $\langle \sigma \rangle_T$ is now computed with an energy of only kT since one of the particles is indeed at rest. The dotted line in Fig. 4(a) shows the result: for temperatures higher than 10 keV, readily obtained at low laser energy, this second process overtakes the first: at very low ion energies, this "lateral collision" process is hindered by a huge difference in cross sections between kT and $2kT$, while at higher energies it is enhanced by the larger interaction volume and the quasisaturation of $\sigma(E)$ at energies beyond 50 keV [9]. The number of neutrons predicted becomes comparable to the observed value. Of course, this lateral collision model remains too crude to account fully for the complexity of the physics involved, and in particular, for the collisions between hot central deuterons and "warm" deuterons created close to the laser axis, in areas where the laser intensity reaches values about 10^{14} W/cm^2, known to be enough to induce significant cluster fragmentation [16].

The lateral collision model is qualitatively consistent with the neutron emission pattern observed. Table I presents the number of neutrons measured at three different detection angles from the laser axis, in otherwise identical conditions (detector in the horizontal plane, 300 mJ of laser energy). This result shows a small anisotropy, with a clear minimum in the forward direction. Indeed, reaction (1) is largely mediated by p waves at high energies [9], resulting in a preferred neutron emission in the axis of the bombarding particle, i.e., mostly sideways in the present case.

This small anisotropy indicates that our experiment took place in a low density interaction regime, different from that of the Livermore experiment [4], which demonstrated isotropic emission, and of the Garching and Berlin experiments [2,3], showing essentially forward neutron emission. Other new features include the reduced role of propagation effects, as the focal position could be varied over a few millimeters along both the laser and the gas flow directions, without modifying the observed neutron yields, and the much larger size of the emitting region, estimated to be 2.5 mm according to the pinhole image of the Thomson parabola diagnostic.

In conclusion, we have shown how to obtain nuclear DD reactions within a laser-produced plasma, using deutered methane clusters formed in a low-density jet. This new cluster target was shown to be ideally suited for the DD reaction, as the ion energies diagnosed through a mass spectrometer exhibit values up to 120 keV, much higher than obtained previously. The explosion dynamics of such two-element clusters is still poorly understood and will have to be investigated in detail. Moreover, the neutrons were shown to originate largely from collisions between accelerated ions from the plasma core and cold nuclei in the surrounding jet, resulting in ion emission on a nanosecond time scale. These results should open the way to a tabletop, easily implemented source of neutron bursts, to be used in time-resolved neutronic studies.

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