Study of the yield of D-D, D-\(^{3}\)He fusion reactions produced by the interaction of intense ultrafast laser pulses with molecular clusters

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Abstract. The interaction of intense ultrafast laser pulses with molecular clusters produces a Coulomb explosion of the clusters. In this process, the positive ions from the clusters might gain enough kinetic energy to drive nuclear reactions. An experiment to measure the yield of D-D and D-\(^{3}\)He fusion reactions was performed at University of Texas Center for High Intensity Laser Science. Laser pulses of energy ranging from 100 to 180 J and duration 150fs were delivered by the Petawatt laser. The temperature of the energetic deuterium ions was measured using a Faraday cup, whereas the yields of the D-D reactions were measured by detecting the characteristic 2.45 MeV neutrons and 3.02 MeV protons. In order to allow the simultaneous measurement of \(^{3}\)He(D,p)\(^{4}\)He and D-D reactions, different concentrations of D\(_{2}\) and \(^{3}\)He or CD\(_{4}\) and \(^{3}\)He were mixed in the gas jet target. The 2.45 MeV neutrons from the D(D,n)\(^{3}\)He reaction were detected as well as the 14.7 MeV protons from the \(^{3}\)He(D,p)\(^{4}\)He reaction. The preliminary results will be shown.

1. Introduction
The availability of high intensity laser facilities capable of delivering PW of power into small volumes has opened the possibility to use those facilities for fundamental and applied nuclear physics

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studies. In particular the Coulomb explosion of D$_2$ molecular clusters induced by the interaction with the laser pulse provides a distribution of low energy D ions in a highly ionized, electron deficient medium that might be ideal to study D-D and D-^3^He nuclear fusion reactions at very low energy. Those reactions are important in astrophysics because they contribute to the primordial nucleosynthesis. They are also important for the future design of nuclear fusion reactors for energy production. The study of two different reactions occurring simultaneously in the same environment provides a diagnostic tool to determine some properties of the plasma medium such as the temperature and the density.

The number of energetic deuterium ions and their energy distribution was measured with a Faraday cup using the Time of Flight (ToF). The ratio of the yield of the reactions $^3^He(D,p)^4^He$ and $D(D,n)^3^He$ was used to determine the average temperature of the deuterium ions involved in the reactions. Finally, the yield of 14.7 MeV protons from the $^3^He(D,p)^4^He$ reaction was used to extract the average cross section over the ion energy distribution measured by time of flight.

2. Experimental Setup

The yield of D-D and D-^3^He fusion reactions induced by the interaction of a high power laser with molecular clusters was measured at University of Texas Center for High Intensity Laser Science. Laser pulses of energy ranging from 100 to 180 J and duration about 150 fs were delivered by the Petawatt laser. D$_2$ molecular clusters are produced in the adiabatic expansion in the vacuum of high pressure ($\sim$750 psi) and low temperature ($\sim$80 K) gas, through a supersonic nozzle. When the laser hits the clusters most of its energy is absorbed by the molecules, the electrons escape first and the positively charged clusters Coulomb explode. In this process some of the ions have enough energy to drive fusion reactions. The experimental setup is schematically shown in Fig. 1. The temperature of the energetic deuterium ions was measured using a Faraday cup of 16 mm diameter, placed at 160° relative to the laser direction at a distance of about 1.07 m from the target. Fig. 2 shows a typical time of flight spectrum from the Faraday cup. The time distribution of the deuterium ions can be fitted with a Maxwell distribution to obtain the temperature $K_T$ and the number of ions as explained in 2.1.3.

The yields of the D-D reactions were determined by detecting the characteristic 2.45 MeV neutrons and 3.02 MeV protons. In order to measure the 2.45 MeV neutrons, six plastic scintillators were placed at 90° and -90° at distances of 2 m and 4 m from the target position. Four additional NE213 liquid scintillators of cylindrical shape (radius 7 cm and thickness 14 cm) were placed at 36°, 90°, -90°, 151° and distances larger than 2 m. The signals from the plastic scintillators were recorded by Tektronix oscilloscopes, while the signals from the liquid scintillators were acquired using flash ADCs.
The 3.02 MeV protons were detected using thin plastic scintillators (BC400) of thicknesses 254 μm (ρ=1.03 g/cm³) placed at 45°, 90°, 135°. Those detectors were placed at a distance of 106 cm from the target position. A schematic drawing of the detectors is shown in Fig.1. A 25.4 μm Al foil was placed in front of the detectors at a distance of about 5 cm in order to shield the detectors from the light and stop electrons of energy up to 50 keV. Two photo-multiplier tubes (PMT) Hamamatsu R1355 were collecting the scintillation light at both sides of the plastic scintillator. Home-made active bases were used to provide the high voltage to the PMTs and to get the signal. The signals were recorded using three Tektronix oscilloscopes, TDS 3052. The time duration of a pulse signal was about 10 ns FWHM. The time difference between the signals produced by x-rays and by the proton was about 40 ns. In the first part of the experiment only two proton detectors were operative (A and B). The detector C was added later.

In the second part of the experiment, different concentrations of D₂ and ³He or CD₄ and ³He were mixed in the gas jet target to allow the simultaneous measurement of ³He(D,p)⁴He and D-D reactions. The relative concentrations of D₂ (or CD₄) and ³He in the reaction chamber were constantly monitored. The 2.45 MeV neutrons from the D(D,n)³He reaction were measured as well as the 14.7 MeV protons from the ³He(D,p)⁴He reaction. The 14.7MeV protons were detected using the same plastic scintillators used for the 3.02 MeV protons, but with a different degrader. The thin degrader was removed and replaced with a 1.1 mm thick Al degrader placed about 50 cm before the plastic scintillator. The energy of the protons was degraded down to 4 MeV so that the time difference between the x-ray signal and the protons signal was about 25 ns. Fig. 3 shows the typical signals produced by 3 MeV and 14.7 MeV protons.

![Graphs showing time-of-flight data for D ions and gamma/neutron signal production](image-url)

**Fig. 2** Left panel: time of flight of the D ions as measured by the Faraday cup. Right panel: signal produced by gamma rays and neutrons in the liquid scintillator. The first peak on the left is due to gamma rays, the second peak to the neutrons. The long tail of the neutron peak is due to the long lived characteristic fluorescence component of the light output of those detectors ($\tau_1 = 3.16$ ns, $\tau_2 = 32.3$ ns, $\tau_3 = 270$ ns [2])

![Graphs showing signals for 3 MeV and 14.7 MeV protons](image-url)

**Fig. 3** Signals produced by the γ-rays and protons on the proton detectors. The first peak on the left is due to γ-rays. Left panel: 3 MeV protons. Right panel: 14.7 MeV protons.
2.1. Detector calibration and data analysis

2.1.1. Neutron detectors. The calibration of the plastic scintillators for neutron detection was performed at UT as described in ref. [3]. They served as reference detectors. The calibration of the NE213 liquid scintillators was performed at Texas A&M using a $^{252}\text{Cf}$ source. The neutrons emitted from the Cf source were detected in coincidence with the fission fragments. In order to measure neutron time of flight and the signal amplitudes, the neutron detectors were placed at a distance of 150 cm from the source. The signal shapes were recorded using the same Flash ADCs used in the laser experiment (10 ns sampling interval). Gating on the proper times of flight, we selected the neutrons in the energy range from 2 to 3 MeV. We obtained the distribution of the peak amplitudes produced by one neutron in the selected energy range. The average of this distribution is the average signal for one neutron. In this way the number of neutrons hitting the detector during a laser shot can be obtained by dividing the measured peak amplitude by this average signal. During the calibration we observed that the light output produced by only one neutron in the detector do not show evidence of the long term fluorescence decay constant $\tau_3 = 270\text{ ns}$. In fact all the signals reach the baseline in or about 60 ns. On the other hand, when many neutrons hit the detector simultaneously this component becomes more and more evident as the number of neutrons increases. To take into account this effect we normalized to unitary peak height all the signals obtained during the laser experiment. Then we fitted the signal decay time with $y = Ae^{-t/\tau_1} + (1 - A)e^{-t/\tau_2}$. The decay constants $\tau_1 = 60\text{ ns}$ and $\tau_2 = 300\text{ ns}$ were determined averaging the values of the decay constants obtained from a fit with $A$, $\tau_1$ and $\tau_2$ free parameters. Fig 4 shows as example the values of the parameter $A$ as a function of the signal peak height for the neutron detector 3.

![Figure 4: Value of the parameter A as a function of the peak height](image)

The number of neutrons was calculated dividing the signal amplitude by $A$ and by the average signal of one neutron.

2.1.2. Proton detectors. The calibration of the proton detectors was performed at Cyclotron Institute of Texas A&M University, using a low intensity 14 MeV proton beam from the K150 cyclotron. The thick aluminum degrader (1.1 mm) was placed before the plastic scintillator at the same position as in the laser experiment. The signal shapes were recorded using the same Tektronix oscilloscopes used in the laser experiment. The average signal for one proton was obtained averaging the signal amplitudes.

2.1.3. Time of flight from the faraday cup. The ToF signals from the faraday cup were fitted with a Signal + Background expression (1). The background is due to x-rays and electromagnetic noise, whereas the deuterium ions arrive later and have a Maxwellian distribution.

$$Signal = Nions \times \frac{e\Omega}{4\pi} \sqrt{\frac{2}{\pi}} \left(\frac{m}{KT}\right)^{\frac{3}{2}} \frac{d^{3}}{(t-t_{0})^{4}} e^{-\frac{d^{2}m}{4K(T-t_{0})^{2}}}$$

(1)
\[ \text{Background} = C \left( - \frac{t_{\text{rise}}}{T_1} e^{-\frac{t - t_0}{T_1}} + e^{-\frac{t - t_0}{T_1}} + D e^{-\frac{t - t_0}{T_2}} \right) \]

\( KT, N_{\text{ions}}, t_0, T_{\text{rise}}, T_1, T_2, C, D \) in (1) are parameters of the fit. \( KT \) is the ion temperature in keV. \( N_{\text{ions}} \) is the number of deuterium ions estimated assuming an isotropic distribution of the ions over \( 4\pi \). \( R \) is the 50 Ohm impedance of the circuit, \( e \) is the electron charge, \( m \) is the mass of the deuterium, \( d \) is the distance of the faraday cup from the target, \( \Omega \) is the solid angle subtended by the faraday cup. \( T_{\text{rise}} \) is the rise time of the background peak and \( T_1 \) and \( T_2 \) are two exponential decay constants, \( t_0 \) is the maximum of the background peak. The background subtracted spectra (green line in fig. 5) were further fitted with a maxwellian distribution to obtain the number of ions \( (N_{\text{ions}}) \) and the temperature \( KT \). A typical example is shown in figure 5.

![Figure 5: Red points: Faraday cup signal from the oscilloscope. Black line: fit Signal + Background. Green plot: background subtracted spectrum. Blu line: fit background subtracted signal](image)

2.1.4. Dimensions of the plasma plume. The dimensions of the plasma plume were measured at every shot from photographic images of the target area.

3. Experimental results

In the first phase of the experiment only \( \text{D}_2 \) gas was used. The yield of D-D reactions were measured by detecting the 3.02 MeV protons from the \( \text{D(D,p)}\text{Ti} \) and the 2.45 MeV neutrons from the \( \text{D(D,n)}\text{He} \) reaction. In the second phase of the experiment different concentrations of \( \text{He} \) were added to the gas mixture. The 2.45 MeV neutrons from the D-D reactions were measured. The left panel of figure 6 shows the average over many shots performed in similar conditions of the protons and neutrons yields measured at different angles. The picture shows that the reaction products are emitted isotropically in the plane of the laser direction defined as polar angle zero. Given this result, we can calculate the final yield of protons and neutrons by averaging the results of all the neutrons and proton detectors. We also note that the yield of neutron (green triangles) and protons (violet circles) measured in the first phase of the experiment is the same inside the experimental errors. The same conclusion is drawn looking at
the left panel of figure 5. In this case the yield on 3.02 MeV protons is plotted shot by shot as a function of the yield of 2.45 MeV neutrons.

Fig 6: Left panel, average yield of 2.45 MeV neutrons (triangles) or 3.02 MeV protons (circles) from D-D reactions calculated over many shots preformed in similar conditions. In the first phase of the experiment only the proton detectors A and B were working violet circles. Right panel, yield of 3.02 MeV protons versus yield of 2.45 MeV neutrons as measured in the first phase of the experiment. The neutron yield is the average of the yield measured with all the neutron detectors. The proton yield is the average yield measured by the proton detectors A and B

The yields of the D-D and D-\(^3\)He reactions can be calculated using the available parameterizations of the reaction cross-section \(\sigma\) given in the Gamow form.

\[
\sigma(E) = \frac{S(E)}{E} e^{-\sqrt{E_G/E}}
\]  
(2)

Where \(S(E)\) is the astrophysical S factor, \(E\) is the energy in the center of mass, \(E_G\) is the Gamow energy. We used the parameterizations of the S factors given in Ref. [1].

For the D(D,n)\(^3\)He reaction the S factor is given by

\[
S(E) = a_1 + E \left(a_2 + E \left( a_3 + E (a_4 + E a_5) \right) \right)
\]  
(3)

For the \(^3\)He(D,p)\(^4\)He the S factor is given by the expression (4) where the parameters \(b\) account for a broad resonance at about 250 keV.

\[
S(E) = \frac{a_1 + E (a_2 + a_3 E)}{1 + E (b_1 + E (b_2 E + b_3))}
\]  
(4)

Figure 7: Reaction cross-sections for the reactions D(D,n)\(^3\)He (left panel) and \(^3\)He(D,p)\(^4\)He (right panel)

The values of the parameters \(a\) and \(b\) are listed in Ref. [1] for both reactions. Figs. 7 show the calculated cross-sections for the D(D,n)\(^3\)He and \(^3\)He(D,p)\(^4\)He reactions as a function of the center of mass energy.
In order to calculate the yield of D-D reactions, we have to consider two possibilities. D-D fusion reactions may happen inside the plasma plume between two energetic D ions coming from the Maxwellian distribution of measured KT or between an energetic D ion and a D at rest outside the plasma plume. In this case the yield of 2.45 MeV neutrons, \( Y_n \), can be written as:

\[
Y_n = N_{ions} \rho_D \left( \frac{1}{2} \int \frac{1}{E} \left( \frac{1}{KT} \right)^{3/2} e^{-\frac{E}{KT}} \sigma(E) dE \right) + \frac{1}{2} \frac{<\sigma v> T}{v_{av}} \rho_D \left( \frac{1}{2} \int \frac{1}{E} \left( \frac{1}{KT} \right)^{3/2} e^{-\frac{E}{KT}} \sigma(E) dE \right)
\]  

Where \( N_{ions} \) is the number of energetic D ions, \( \rho_D \) is the atomic density of deuterium, \( r \) is the radius of the plasma plume (varying between 0.5 to 1.2 mm), \( R \) is the average radius of the cluster cone (2.5 mm), \( E \) is the energy of the D ions in the center of mass, \( T \) is their temperature, \( v \) is the velocity, \( \sqrt{v} \) is the average velocity and \( m_r \) is the reduced mass of the system.

The first term in formula (5) accounts for the reactions between energetic D ions and deuterons at rest outside the plasma plume. The average cross-section \( <\sigma>_T \) is calculated in this case for the corresponding temperature in the center of mass \( T/2 \). The second term gives the yield of fusion reactions occurring between energetic D ions in the plasma plume. In this case the reactivity \( <\sigma v>_T \) is calculated at the center of mass temperature \( T \).

Figs.8 show the values of \( <\sigma v>_T \) and \( <\sigma>_T \) calculated for the reaction D(D,n)\(^3\)He as a function of KT using the formulas above. Our calculation of \( <\sigma v>_T \) agrees with the values tabulated in the NACRE database [4]. This is expected since both calculations are based on the same parameterization of the cross section.

Since \(^3\)He atoms do not form clusters at temperatures near the liquid nitrogen, \(^3\)He ions are not accelerated during the coulomb explosion of the D\(_2\) clusters. Therefore D-\(^3\)He reactions only occur between fast D ions and \(^3\)He at rest. The yield of 14.7 MeV protons is given by
\[ Y_p = N_{ions} \cdot \rho_{^{3}He} \cdot \frac{<\sigma>_{^{3}He}}{<\sigma>_{D}} \cdot R \]  
with 
\[ <\sigma>_{T} = \int_{0}^{\infty} 2 \sqrt{\frac{E}{\pi}} \left( \frac{1}{KT} \right)^{3/2} e^{-\frac{E}{KT}} \sigma(E) dE \]

The atomic density of \( ^{3}He \) \( \rho_{^{3}He} \) and can be calculated as \( \rho_{^{3}He} = \rho_{^{3}He}/[D] \), where \( [^{3}He] \) and \( [D] \) are the concentrations of \( ^{3}He \) an D in the experimental chamber. The average cross-section \( <\sigma>_{^{3}He} \) is calculated at the corresponding temperature in the center of mass \( 3T/5 \).

Figs. 9 show the values \( <\sigma>_{T} \) and \( <\sigma>_{T} \) as a function of \( KT \) calculated using the formulas (6) and (7) and compared with the evaluations from the NACRE database (blue line).

If the atomic concentration of D and \( ^{3}He \) is the same, the ratio of the yields of D-\( ^{3}He \) and D-D reactions is independent of the atomic density and depends only on the temperature of the system.

\[ \text{Ratio}_{D-^{3}He/D-D} = \frac{<\sigma>_{T}^{3}He}{<\sigma>_{T}D-D} \cdot \frac{R}{R} \]  

Figure 9: Same as Figure 8 but for the reaction \( ^{3}He(D,p)^{4}He \) using the parameters in [1]. See the text for details.

Figure 10: Ratio of the yield of the reaction \( ^{3}He(D,p)^{4}He \) and D(D,n)\( ^{3}He \) as a function of the D ions temperature measured from the time of flight. Note that for temperatures around and larger than 20 KeV the Faraday cup is at the limit of his detection capability.

The experimental ratios as a function of the temperature of the energetic D ions measured form time of flight are shown in Fig. 10. The experimental yields of the D-\( ^{3}He \) reaction are normalized to
the same concentration of [D] and [^3]He]. The solid lines show the ratios calculated using formula (10) with r = 0.5 mm (blue line) or r = 1.1 mm (black line). The experimental data roughly agree with the calculated values inside the experimental errors. We note that for high temperature the fit of the time of flight spectra has larger errors.

A further check of the consistency of our data is given by the comparison of the Number of deuterium ions obtained from the fit of the time of flight spectra and the Number of deuterium ions needed to reproduce the yield of 2.45 MeV neutrons with formula (5). We only assume that all the clusters present in the plasma plume exploded so that the atomic density \( \rho_D \) of deuterium is given by \( N_{ions} \) divided by the measured volume of the plasma plume. The result of this comparison is shown in Fig. 11. Fig. 11 also shows for the same shots the values of the atomic density of deuterium \( \rho_D \).

The yield of 14.7MeV protons from the D-^3He reaction can be used to extract the experimental values of \( <\sigma>_T \) as a function of KT. The preliminary results are shown in Fig. 12. The data analysis is still in progress.

Figure 11: Left panel: Number of deuterium ions obtained from the Faraday cup (ToF spectra) compared with the number of ions calculated in order to match the measured yield of 2.45MeV neutrons from the D-D reaction Right panel deuterium ions atomic density as a function of the KT from ToF.

Figure 12: experimental \( <\sigma>_T \) for the reaction \(^3\text{He}(\text{D},p)^4\text{He} \) as a function of the D ion temperature \( KT \) in the center of mass \((3/5KT_{\text{Faraday cup}})\).
4. Conclusions
We measured the yield of D(D,T)p, D(D,$^3$He)n and D($^3$He,α)p and the ion temperature. We showed that the yield ratio can be used as a diagnostic tool to determine the temperature of the ions when the time of flight measure is not possible.

The experimental values of $\langle \sigma \rangle_{T,D-3He}$ for the reaction D($^3$He,α)p as a function of the KT in the center of mass can be calculated from the experimental yield of 14.7 MeV protons. In the next step of the analysis we will fit those data to extract a parameterization for the astrophysical S factor S(E).

We note that in many of the shots the error on the measured 14.7 MeV proton yield is pretty large due to the very low concentration of $^3$He in the gas mixture. An easy improvement of the results can be obtained by increasing the concentration of $^3$He in the gas mixture. It would be also useful to improve the experimental control over the atomic density of the deuterons with a measure of the cluster size and the electron density using Rayleigh scattering and short pulse interferometry before every shot.

References
[1] H.-S. Bosch and G.M. Hale, Nucl. Fusion 32 (1992) 611
[2] M. Moszyński et al. Nucl. Instr. and Meth. A 350 (1994) 226–234
[3] W. Bang et al. Rev. Sci. Instrum. 83, 063504 (2012)
[4] NACRE database http://t2.lanl.gov/data/astro/astro.html
[5] M. La Cognata et al., PHYSICAL REVIEW C 72 (2005) 065802
[6] A. Krauss et al., Nuclear Physics A 465 (1987) 150–172
[7] M. Aliotta et al., Nuclear Physics A 690 (2001) 790–800
[8] W.H. Geist et al., PHYSICAL REVIEW C 60 (2003) 054003