Nuclear fusion in gases of deuterium clusters heated with a femtosecond laser*

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Recent experiments on the interaction of intense, ultrafast pulses with large van der Waals bonded clusters have shown that these clusters can explode with substantial kinetic energy. Producing explosions in deuterium clusters with a 35 fs laser pulse, deuterium ions were accelerated to sufficient kinetic energy to drive deuterium–deuterium (DD) nuclear fusion. By diagnosing the fusion yield through measurements of 2.45 MeV fusion neutrons, over \(10^6\) neutrons per laser shot were measured when 100 mJ of laser energy is used. © 2000 American Institute of Physics.

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I. INTRODUCTION

A number of experiments have been conducted in recent years examining the interactions of intense femtosecond laser pulses with large van der Waals bonded clusters. Most experiments indicate that these interactions can be very energetic, with a variety of experimental manifestations. Studies from a few years ago revealed that very bright x-ray emission in the 100 to 5000 eV range resulted from the plasmas produced from the interaction of pulses focused to intensity between \(10^{16}\) and \(10^{18}\) W/cm\(^2\) into gas jets that contained rare gas atomic clusters of a few hundred to a few thousand atoms each.\(^{1,2}\) This bright x-ray production resulted from the efficient coupling of intense laser light into the cluster gas, strong coupling that was found to be absent when irradiating a monatomic gas of similar average density at these intensities. This efficient coupling was confirmed by subsequent measurements of laser energy absorption in clustering gases.\(^{3,4}\) In fact, these experiments illustrated that nearly 100% of an intense laser pulse could be absorbed by a modest average density gas jet (\(\sim 10^{13}\) cm\(^{-3}\)) within a few mm of propagation length when clusters were present in the gas. This greatly enhanced absorption arises from the fact that the density within the clusters is high, nearly that of a solid, and can therefore exhibit the large absorption efficiency usually associated with solid targets.\(^{5}\)

The laser absorption measurements indicated that many keV of energy per atom were being deposited in the clustering gas. The mechanisms and dynamics of this energy deposition by femtosecond pulses in individual clusters has also been examined by a number of groups. These studies have included measurement of ionization charge states produced in the cluster,\(^{6,7}\) pump–probe and pulse-width experiments which have shown that the clusters disassemble on a picosecond time scale,\(^{8,9}\) and photo electron energy measurements which indicated that multi-keV electrons were ejected from large (>1000 atom) Xe clusters.\(^{10}\) Perhaps most remarkable has been the discovery that these large clusters, when irradiated at intensity above \(10^{15}\) W/cm\(^2\), also eject ions with substantial kinetic energy. In fact, ions with energy as high as 1 MeV have been seen from exploding Xe clusters.\(^{8,11}\)

This large release of kinetic energy in fast ions can be harnessed to drive nuclear fusion between deuterium ions if deuterium clusters are irradiated in a gas of sufficient average density to permit collision between ions ejected from different clusters in the gas. We recently observed these fusion reactions in gases containing deuterium clusters of roughly 5 nm diameter.\(^{12}\) In this paper we report on a detailed series of measurements of fusion yield from these deuterium cluster explosions and show that, while total fusion yield is modest, the fusion efficiency can be substantial, over \(10^7\) fusion neutrons per joule of laser energy.

The concept behind this fusion experiment is illustrated in Fig. 1. In our experiment a high intensity, ultrafast laser pulse is focused into a gas jet of deuterium clusters, rapidly heating the inertially confined clusters before they can expand. These clusters subsequently explode, ejecting energetic ions. This process creates a plasma filament with a diameter roughly that of the laser focus (\(\sim 100 \mu\m\)) and a length comparable to the extent of the gas jet plume (\(\sim 2 \mm\)). The fast deuterium ions ejected from the exploding clusters can then collide with ions ejected from other clusters in the plasma. If the ion energy is high enough (greater than a few tens of keV), D+D nuclear fusion events can occur with high probability. The well known signature of this process arises from one branch of the fusion reaction, D+D→He\(^+\)+n, in which a neutron is released with 2.45 MeV of energy.

The previous experiments on cluster explosions\(^{8}\) indicated that large deuterium clusters ionized at high intensity should produce the high ion energies required for fusion. When large clusters (>1000 atoms/cluster) are ionized by the intense laser pulse, a plasma-like sphere is formed with optically and collisionally ionized electrons undergoing rapid collisional heating for the short time (<1 ps) before the clus-
fter disassembles in the laser field. Through various collective and nonlinear processes, the laser rapidly heats the electrons to a nonequilibrium state (with mean energies of many keV). The escape of these hot electrons from the cluster produces a strong radial electric field which accelerates the cluster ions; therefore, the deposited energy is transferred from the light electrons to the more massive ions. The consequence is that the very efficient absorption of laser energy in the cluster is ultimately released in ion kinetic energy when the heated cluster explodes isotropically. Though large, high $Z$ clusters typically exhibit a “plasma-like” hydrodynamic explosion due to the fact that space charge forces confine many of the photoionized electrons to the body of the cluster; low-$Z$, medium size ($\sim$1000 atom) clusters can be stripped of all of their electrons during the laser pulse, before the cluster explodes. In this case, essentially a pure Coulomb explosion of the deuterium cluster results. With deuterium cluster sizes of greater then a few tens of angstroms, fully stripping the cluster with the laser field will result in the explosion of ions with energies greater than a few keV.

II. EXPERIMENTAL APPARATUS

The layout of our experiment with the suite of diagnostics is illustrated in Fig. 2. Our experiment uses a 10 Hz, Ti:sapphire, chirped pulse amplification laser delivering 120 mJ of laser energy per pulse with 35 fs pulse width and wavelength of 820 nm. This laser was focused into the exit of a deuterium gas jet with an f/12 lens. Because the van der Waals forces between deuterium molecules are weak, the gas jet was cryogenically cooled by flowing cooled nitrogen through a jacket surrounding the gas jet body to $-170^\circ$C, producing large clusters in the D$_2$ gas. The laser spot size within the gas jet varied from roughly 20 to 130 $\mu$m depending on the focal spot position, with respect to the gas jet nozzle. Consequently the peak laser intensity was between $5 \times 10^{17}$ W/cm$^2$ and $2 \times 10^{16}$ W/cm$^2$ for all of the experiments described here.

To estimate the deuterium cluster size in our gas jet, we conducted Rayleigh scattering measurements in the jet by passing low energy (<0.1 mJ) 532 nm pulses from a frequency doubled, Q-switched Nd:YAG laser (pulse width 10 ns) through the plume of the gas jet. The Rayleigh scattered light from cluster formation was imaged 90 deg from the laser propagation axis. From the observed onset of Rayleigh scattering, we can determine the onset of large cluster formation in the jet. In addition, using independent measurements of the average gas density (see below), estimates for the throughput and detection sensitivity, and the Rayleigh scatter cross section, we are able to estimate the average cluster size in the jet. Measured scattering as a function of gas jet temperature (monitored with a thermocouple mounted near the jet nozzle) is illustrated in Fig. 3(a) when the gas jet backing pressure is 70 atm. From these data it can be seen that the scattering signal is a strong function of the jet temperature and that no significant cluster formation is observed until the jet temperature decreases below $-150^\circ$C. A strong variation with gas jet backing pressure is also observed. Scatter signal as a function of backing pressure when the jet is cooled to $-170^\circ$C is plotted in Fig. 3(b). Here, it is clear that backing pressure above 20 atm is needed for good deuterium cluster formation.

III. EXPERIMENTAL RESULTS

To determine the efficiency with which the 120 mJ, femtosecond laser pulse was coupled into the deuterium clusters, we measured the absorption efficiency of the laser pulse within the deuterium cluster gas jet. To do this, we measured the transmitted laser energy collected within an f/3 cone (to collect all potentially refracted light from the f/12 cone) with a large aperture lens and calorimeter. We monitored backscattered light with a photodiode (very little was observed in these experiments). We also estimated scattered light using a charge coupled device (CCD) camera and collection lens monitoring light scattered 90° from the laser propagation direction perpendicular to the laser polarization direction (the...
direction of maximum Rayleigh scatter). Extrapolating into $4\pi$, we determined that the scattered light was negligible (<1 ml).

The absorption as a function of gas jet reservoir backing pressure is illustrated in Fig. 4 when the gas has been initially cooled to $-170^\circ$C (the case in which we observe large D$_2$ cluster formation). As can be seen, when the deuterium gas jet pressure rises to $\geq$30 atm, nearly 90% of the laser energy is deposited in the plasma. This is in sharp contrast to the energy deposition efficiency of the laser in pure D$_2$ molecular gas (the case when the gas jet reservoir is at $20^\circ$C), a case also plotted in Fig. 4. In this case, very little (<5%) of the laser energy is deposited in the plasma, illustrating the importance of the laser/cluster interaction in heating the D$_2$ plasma.

To measure the volume and density of the plasma in which this laser energy was deposited, optical interferometry was performed on the plasma filament. The layout of this diagnostic is illustrated in Fig. 2. A small fraction (~4%) of the main 35 fs laser pulse was split from the beam using a thin pellicle and sent through a delay leg. The propagation distance was set such that this probe pulse traversed the plasma filament within 20 ps of the main laser pulse, a time scale after all optical ionization but faster than any hydrodynamic motion of the heated filament. This pulse then traversed a telescope, which imaged the plasma filament onto a CCD detector located in one arm of a Michelson-type interferometer. A roof prism in one arm of the interferometer caused interference between light which passed through the plasma filament and reference light which passed below the filament. Using the cylindrical symmetry of the plasma filament, the phase retrieved from the resulting interferogram could be deconvolved using an Abel inversion to derive the electron density (and thus the D$^+$ density) as a function of radius at each point along the filament.

An interferogram of the deuterium plasma when the laser is focused $\sim$2 mm below the jet aperture and 2.5 mm in front of the center of the jet gas stream is shown in Fig. 5.
The deconvolved electron density at the middle of the filament is plotted next to the raw image. This measurement indicates that the average deuterium atom density was $1.5 \times 10^{19} \text{ cm}^{-3}$. Using information derived from the absorption measurement, we can determine that roughly 5 keV of energy was deposited per atom in the filament (though, in fact, we expect that the energy deposited near the center will be higher because of the higher laser intensity). From this we can make a conservative estimate of the average deuterium ion energy by assuming that energy was equally distributed between ions and electrons. (In fact, we expect that more energy is initially transferred to the ions than the electrons in the laser driven cluster explosion\textsuperscript{18}). These data suggest that the average deuterium ion energy was at least 2.5 keV. This, however, is nothing more than a very rough estimate; non-uniform deposition of laser energy due to the intensity distribution at focus as well as absorption along the filament will give rise to a range of ion energies. Nonetheless, it is clear that the efficient absorption of the laser in the deuterium clusters can heat the deuterons to multi-keV energies, sufficient energy to drive fusion events.

To detect the production of DD fusion neutrons in these experiments we employed arrays of neutron sensitive scintillators detectors. The layout of our detectors is shown in Fig. 2. Each detector was a grouping of three plastic scintillator cylinders coupled to two photo-multiplier tubes. These detectors were located outside the vacuum interaction chamber and were shielded with 6 mm of lead. We deployed two arrays at various distances of 1, 2.5, 3.2, and 4.1 m from the plasma. The time-of-flight of particles from the time the laser irradiates the plasma is recorded. Each neutron detector was operated in a mode such that a neutron was detected every few laser shots, permitting a determination of the flight time of each detected particle with $\sim 10$ ns accuracy.

With these neutron detectors, we detected the presence of substantial numbers of MeV energy particles when the gas was backed with cooled deuterium at a pressure of 65 atm and a reservoir temperature of $-170^\circ \text{C}$. The time-of-flight spectrum of particles detected on the neutron detectors is illustrated for the four plasma-detector separations in Fig. 6. These time-of-flight (TOF) spectra were acquired by accumulation of particle hits over 200–300 laser shots. For each distance, a distinct peak occurs in the detected TOF spectrum at the position expected for 2.45 MeV neutrons (which have a flight time of 46 ns/m). This measurement confirms the presence of DD fusion reactions in the cluster plasma. We also confirmed that all detected particles on the neutron detectors disappear when hydrogen clusters were made in the jet in lieu of the deuterium clusters. Furthermore, we found that the fusion neutron production disappeared when deuterium clusters where absent in the $D_2$ gas jet (when the jet was not cooled). We observed no neutron production in the gas jet with temperatures higher than $-120^\circ \text{C}$, the temperature at which we no longer observe large cluster formation via Rayleigh scattering. These data clearly indicate that the intense illumination of deuterium clusters does give rise to DD fusion.

The signal seen in these spectra after the main neutron peak arise from neutron scattering from the gas jet and vacuum chamber walls. We have conducted estimates of the expected scattered neutron signal and have confirmed that these longer time detected particles are consistent with scattered neutrons from material surrounding the fusion plasma, such as the gas jet body and vacuum chamber walls. We also see an occasional particle early in time. Though we are still unsure of the production mechanism, it is likely that these are hard x rays produced from fast electrons released in the laser/cluster interactions. These fast electrons can produce bremsstrahlung photons in the surrounding material, including the stainless-steel gas jet body. However, these gammas are rare and the vast majority of the signal observed in this experiment is from the fusion neutrons.

Under the conditions in which the highest yield was attained, we observed many multiple hits per shot on the neutron detectors within 2.5 m of the plasma. We were, however, able to estimate the fusion yield by observing the neutron count rate with 3.2 m detection separation, where the detection rate was less than one per shot. Under optimum conditions, (discussed below) we produced roughly $1 \times 10^4$ neutrons per laser shot. This efficiency, roughly $10^5$ neutrons per joule of incident laser energy, is comparable to the fusion neutron production efficiency of many deuterium laser-driven fusion experiments.

FIG. 6. Time-of-flight spectrum of particles detected on the neutron detectors for the four plasma-detector separations, $d$ (from Ref. 14).
FIG. 7. (a) Fusion neutron yield as a function of the distance between the laser best focus (in vacuum) and the center of the jet nozzle. (b) Fusion neutron yield as a function of the distance between the laser propagation axis and the gas jet output orifice when the laser is focused 2.5 mm before the jet.

To measure the change in neutron yield as different parameters were varied, an additional plastic scintillator detector was placed near the plasma in an re-entrant flange to subtend a large solid angle. It subtended an angle of 0.3 ster and was placed 20 cm from the deuterium plasma. Because the time-of-flight data indicate that there is no noticeable x-ray flash and that virtually all of the detected particles are neutrons, we obtained a shot-by-shot measure of the fusion yield from the photomultiplier tube (PMT) signal. The detection efficiency was calibrated against the other TOF detectors operating in single-particle detection mode. We also calibrated the yield by measuring the average pulse height produced when single neutrons struck the detector. Both methods yielded nearly equal values for the yield calibration.

Using this detector, we measured the yield as a function of the distance between the laser best focus (in vacuum) and the center of the jet nozzle. The results of this scan are shown in Fig. 7(a). We find that the optimum fusion yield is obtained, not when the laser best focus is placed at the center of the gas jet, but when the laser is focused before the gas jet. For a spacing between laser propagation axis and nozzle aperture of 2 mm, we find that the best yield is obtained when the laser is focused 2.5 mm before it hits the gas jet center.

From interferometry measurements, we know that this corresponds to a situation in which the laser best focus is located very near the edge of the jet plume (which is 2.5 mm in radius at this location below the jet nozzle). In this case, the laser spot at the center of the gas jet is nearly 130 μm in diameter, while the laser diameter is 30 μm (best focus) at the edge of the jet plume. This implies that the peak intensity at the gas jet center is well below the peak focal intensity (5 \times 10^{17} \text{W/cm}^2) and is probably \(-3 \times 10^{16} \text{W/cm}^2\), neglecting any depletion of the laser energy. This peak probably arises due to an interplay of the laser propagation in the gas plume, the dependence of ion energy as a function of laser intensity, and heated volume contributing to the fusion. A detailed analysis is underway.\textsuperscript{19}

We also find that the fusion yield increases as the distance between the laser propagation axis and the gas jet output orifice decreases. The fusion yield as a function of this separation, when the laser focus is placed 2.5 mm before the jet, is illustrated in Fig. 7(b). As the laser focus moves closer to the output orifice, the yield increases. Because of the expansion of the plume, the average gas density drops away from the nozzle. Because the total fusion yield is expected to scale roughly as the square of the density, the trend observed in Fig. 7(b) is most likely due to an increase of deuterium density closer to the orifice. However, this effect is tied to the more complex issues of laser propagation and energy deposition in the jet, so further experiments are needed to fully understand this behavior.

The fusion yield also increases rapidly with increasing laser intensity. The measured fusion yield as the laser energy is increased is plotted in Fig. 8. These data were taken with the laser focused 2.5 mm before the gas jet, under the optimum yield conditions seen in Fig. 7. The yield is plotted as

![Graph showing neutron yield vs. offset between laser and gas jet](image)

FIG. 8. Measured fusion yield as the laser energy is increased.
a function of the vacuum laser intensity directly under the jet. Here, it can be seen that the yield starts with 1000 n/s/shot at an intensity of $7 \times 10^{15}$ (30 mJ of laser energy) and increases by an order of magnitude as the intensity is increased by a factor of 3. This efficiency, roughly $10^6$ neutrons per joule of incident laser energy, is comparable to the fusion neutron production efficiency of many deuterium laser-driven colliding plasma fusion experiments.\textsuperscript{20–22}

IV. ANALYSIS

This fusion yield is consistent with some simple estimates. The expected fusion yield from our experiment can be roughly estimated from the expression

$$N \approx \left( \frac{\sigma v}{\tau_d} \right)^2 n_d dV,$$

where $\tau_d$ is the disassembly time of the plasma, $\langle \sigma v \rangle_T$, is the velocity-averaged fusion cross section, and $n_d$ is the deuterium density. These are integrated over the volume of the plasma filament. Since the exact ion energy distribution is unknown, we assume a Maxwellian distribution with an average ion energy of 2.5 keV. This implies that $\langle \sigma v \rangle_T \approx 1 \times 10^{-20}$ cm$^3$ s$^{-1}$.\textsuperscript{23} We perform the volume integral using the information from the interferogram. We can estimate the disassembly time as the time required for a $\sim$10 keV deuterium (the deuterium energies which are the main contributors to the fusion) to exit the plasma of diameter 200 $\mu$m. This means $\tau_d = 100$ ps, implying that the expected fusion neutron yield is roughly $8 \times 10^5$. This magnitude is in reasonable agreement with our measured neutron yield.

V. CONCLUSION AND ANALYSIS

In conclusion, we have observed the production of 2.45 MeV deuterium fusion neutrons when a gas of deuterium clusters is irradiated with a 120 mJ, 35 fs laser pulse. When the focal position is optimized, we have observed as many as $10^4$ neutrons per laser shot. This yield is consistent with some simple estimates for the fusion yield. We also find that the fusion yield is a sensitive function of the focal position, with optimum yield coming when the laser is focused before it enters the deuterium gas plume. Finally, we find that the yield increases quickly with increasing laser fluence. These results suggest that it may be possible to further enhance the fusion yield with an increase in laser energy. This experiment may represent a means for producing a compact, tabletop source of fusion neutrons for applications.

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