Characterization of Fusion Burn Time in Exploding Deuterium Cluster Plasmas

J. Zweiback,[1] T. E. Cowan,[1] R. A. Smith,[2] J. H. Hartley,[1] R. Howell,[1] C. A. Steinke,[3] G. Hays,[1] K. B. Wharton,[1] J. K. Crane,[1] and T. Ditmire[1,*]

[1]*Lawrence Livermore National Laboratory, L-477, Livermore, California 94550*
[2]*Imperial College of Science Technology and Medicine, London SW7, United Kingdom*
[3]*Harvard University, Cambridge, Massachusetts 02138*

(Received 10 July 2000)

Exploiting the energetic interaction of intense femtosecond laser pulses with deuterium clusters, it is possible to create conditions in which nuclear fusion results from explosions of these clusters. We have conducted high-resolution neutron time-of-flight spectroscopy on these plasmas and show that they yield fast bursts of nearly monochromatic fusion neutrons with temporal duration as short as a few hundred picoseconds. Such a short, nearly pointlike source now opens up the unique possibility of using these bright neutron pulses, either as a pump or a probe, to conduct ultrafast studies with neutrons.

PACS numbers: 52.58.Ns, 52.50.Jm, 52.70.Nc

Fast neutrons are used in a wide array of applications including radiography [1] and materials science [2]. However, the possibility of ultrafast studies using neutrons, either as a pump or a probe, is usually not considered because no high flux sources of such short neutron pulses exist. Such a source needs to have not only a short initial pulse width, but also a small size and a monochromatic spectrum so that the ultrafast temporal structure is maintained over a reasonable distance from the source. High fluxes of energetic neutrons can be produced with a number of devices, including accelerators [3,4] and plasma pinches [5]. However, these devices yield neutron pulses with duration typically longer than a few nanoseconds. Neutron sources based on nuclear fusion in a hot plasma (like those produced in plasma pinches) produce nearly monochromatic neutrons (with energy near 2.45 MeV for neutrons produced from the fusion of deuterium). With a short lived fusion plasma it is possible to produce a short burst of fast neutrons.

Fusion plasmas produced by large scale inertial confinement fusion experiments produce fast fusion neutron emission ($\sim$100 ps) [6] with a fusion burn time set by the disassembly time of the inertially confined compressed plasma core (roughly equal to the plasma radius divided by the plasma sound speed). Intense, focused short pulse lasers now present another avenue to creating a short lived, hot fusion plasma [7], and a number of recent experiments have observed beam-target fusion neutrons in intense laser solid interactions [8,9] (typically produced at relativistic laser intensity with a rather broad neutron spectrum). Recent results indicate that nearly monochromatic fusion neutrons can be produced from the interaction of femtosecond laser pulses with deuterium clusters, even with laser pulses of quite modest energy ($\sim$0.1 J per pulse) [10]. In this Letter we present measurements of the neutron pulse duration from plasmas created by the irradiation of deuterium clusters with an intense, 35 fs laser pulse. We find that the fusion neutrons produced in this interaction are nearly monochromatic around a neutron energy of 2.45 MeV and exhibit a fusion burn time pulse duration as short as 500 ps. This opens the potential of using tabletop scale neutron sources in sub-ns to ps scale pump-probe experiments for probing the dynamics of neutron interactions with materials.

Nuclear fusion in a high temperature deuterium plasma will produce neutrons with 2.45 MeV of energy from the $D + D \rightarrow He^3 + n$ reaction. Ion temperatures above a few keV are needed to yield significant numbers of these fusion events in the plasma [11]. Recent experiments on the interaction of intense femtosecond laser pulses with gases of large deuterium clusters (clusters that contain many thousands of deuterium atoms each) have shown that a strong laser pulse can drive an energetic explosion of the clusters [12,13], ejecting ions with sufficient kinetic energy to fuse with ions ejected from other, nearby clusters [10]. In this experiment, the intense laser pulse is focused in a gas jet containing deuterium clusters. The clusters explode, accelerating the ions in a filament with diameter of roughly 200 μm (determined by the focused spot) and a filament length given by the absorption depth of the laser pulse in the cluster gas (about 2 mm). This cigar-shaped filament is illustrated in Fig. 1. The fusion neutrons will, therefore, originate in a small volume bounded by this heated filament.

Subsequent to the explosion of the clusters in this heated filament (the clusters explode on a sub-ps time scale) the fast ions ejected from clusters in the filament can fuse with each other. However, the burn time of this hot deuterium plasma is limited by the time it takes the fast ions to escape the heated filament. As illustrated in Fig. 1, the fast ions will exit this filament on a time scale roughly given by the diameter of the filament (comparable to the laser focal spot diameter) divided by the ion velocity. Assuming 10 keV ions traversing a 200 μm plasma filament, the fusion burn time will be on the order of 200 ps. Furthermore, the emitted neutrons will have an energy centered at 2.45 MeV with a narrow energy spread arising from ion Doppler broadening.

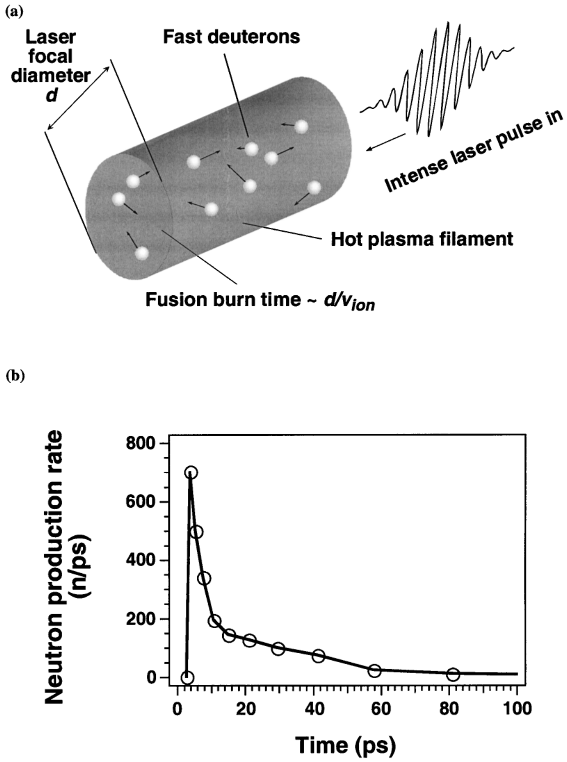

FIG. 1. (a) Experimental concept for an ultrafast neutron source. (b) Simulation showing the predicted neutron pulse width. The total pulse width is under 100 ps.

We have conducted simulations of the expected fusion neutron pulse expected from the situation illustrated in Fig. 1(a). In these simulations we have assumed that the ions stream freely out of the plasma (i.e., that their collisional mean free path is much longer than the plasma dimensions, a good assumption for multi-keV deuterons in plasmas of density below 10^{20} cm^{-3}). We have integrated over all possible ion trajectories assuming a Gaussian initial energy deposition profile in radius (reflecting the Gaussian profile of the focused heating laser pulse). We then integrate over an ion energy distribution measured from the exploding clusters in our experiments. The results of this simulation are illustrated in Fig. 1(b). In this calculation, the peak average ion energy was 12 keV, the average ion density was 2×10^{19} cm^{-3}, and the filament dimensions were a diameter of 200 μm and a length of 1 mm, corresponding to a deposited laser energy of about 0.1 J. This calculation confirms that the neutron pulse is likely below 100 ps. The two-pulse shape seen in the calculation of Fig. 1(b) arises from the difference in transit times of ions exiting the filament radially and ions exiting along the long, axial dimension of the plasma.

To experimentally examine the neutron pulse duration, we conducted a series of experimental studies with high time resolution neutron detectors. The fusion neutrons were produced by focusing 820 nm laser pulses with 120 mJ of energy per pulse and 35 fs pulse duration into a jet of deuterium. The focused intensity of the laser at the cluster jet was $\sim 10^{17}$ W/cm^2. A reservoir of deuterium was held at high pressure (70 atm) and low temperature (100 K) and expanded into vacuum through a pulsed jet valve, producing a spray of large deuterium clusters. For the experiments presented here, we estimate (from Rayleigh scattering measurements [14]) that the average cluster diameter was 5 nm (a few thousand atoms per cluster) and the average density in the gas was 2–4 $\times 10^{19}$ atoms/cm^{-3}. This interaction produced roughly 10^4 neutrons per shot, emitted into 4π sr. These neutrons are emitted uniformly into all directions, indicating that we do not produce fusion from high energy, directional ions.

Neutrons were detected with two classes of high resolution neutron time-of-flight detectors. The first set of detectors consisted of square 12 $\times$ 12 cm^2, slabs of scintillator of 0.7 cm thickness, coupled, via a light pipe, to fast photomultiplier tubes. We measured the fast time response of these detectors by producing fast, hard x-ray pulses from irradiation of solid copper with the femtosecond laser pulse. We varied the laser intensity to keep the x-ray calibration signal to levels approximately produced by single 2.45 MeV neutrons in the scintillators. The 0.7 cm thick detectors exhibited an intrinsic time resolution of roughly 1 ns. (This measurement does not account for the neutron transit time through the scintillator slab, which should be about 300 ps for 2.45 MeV neutrons.) These detectors were used to measure the neutron pulse width at distances greater than 30 cm from the source, where the neutron pulse width was longer than 1 ns. The second class of detector was a similar device, optimized to yield very high time resolution (with lower detection efficiency) by utilizing a thin scintillator coupled directly to a fast photomultiplier tube. The scintillator was a 1.5 mm thick, 50 mm diameter disk (with a neutron transit time of 70 ps). The acquisition electronics of this device were optimized using fast positron pulses produced by a pelletron accelerator. This detector exhibited an intrinsic time response of 250 ps. All detectors were triggered off of the rising edge of a fast photodiode monitoring the laser pulse.

Each detector was operated in a regime in which a single neutron was observed every few laser shots. The neutron pulse width at a variety of separations between detector and source was measured. Each pulse width measurement represents the integration of between 5000 and 10 000 laser shots, so as to build up complete time history. Two examples of measured pulse widths are illustrated in Fig. 2. The first, Fig. 2(a), shows data from the fast detector, located 9 cm from the neutron source. The time response function of this detector, measured with an x-ray flash, is also shown in this plot. The neutron pulse width is determined from these data by fitting a Gaussian pulse convolved with the measured time response function (and estimated neutron transit time through the scintillator). This best fit is overlaid on the data and indicates a neutron

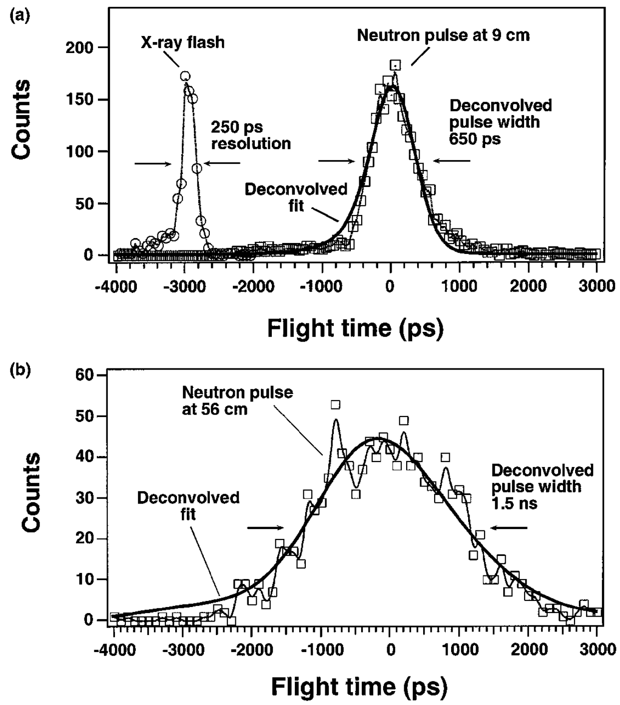

FIG. 2. (a) Measured neutron pulse width 9 cm from the target. The deconvolved pulse width is 650 ps. The detector response (measured by producing hard x rays from a solid target) is shown to the left. (b) Measured neutron pulse width 56 cm from the target. The pulse width is 1.5 ns. The detector resolution was 1.1 ns.

pulse of 650 ps at this distance. Data from one large area detector located 56 cm from the fusion plasma are shown in Fig. 2(b). The best fit neutron pulse convolved with the measured time response of this detector (which exhibited a width of about 1.1 ns) is also shown. The deconvolved pulse width here is 1.5 ns.

It is possible for neutrons scattered from nearby experimental equipment to contribute to the observed neutron pulse. To ascertain the magnitude of this effect in our experiments we modeled neutron scattering via the Monte Carlo radiation transport code COG [15]. Using this code, we simulated neutron transport from the plasma to the detectors. The neutron source was approximated by a cylinder 200 μm in diameter and 2 mm long, with a Gaussian energy distribution at the D-D fusion energy of 2.45 MeV and an energy spread of 10% (with an instantaneous burn time). The geometry of hardware surrounding the plasma and detector was placed into the model. We find at the closest separation (9 cm) over 85% of neutrons striking the detector arrived in a prompt pulse with a time FWHM of $\sim$200 ps. The remaining intensity is distributed in a small tail that stretches out to $\sim$6 ns. Since over 70% of neutrons arrived unscattered in this simulation, these results were insensitive to assumptions about source position and material geometry. From this, we concluded that neutron scattering does not contribute significantly to the measured pulse width.

As a result of the finite energy spread of the emitted neutrons about their center energy of 2.45 MeV, the pulse duration of the neutron burst broadens with increasing distance from the plasma. The measured neutron pulse width as a function of distance away from the source is plotted in Fig. 3. From this plot it is possible to extrapolate back toward the source to estimate the fusion burn time. Since the neutron pulse width variation will be due to a combination of the initial burn time and the spreading due to finite neutron energy spread, we fit these data with a function $(\alpha x^2 + \delta t^2)^{1/2}$ where δt is the initial burn time, x is the distance from the source, and α is a fitting parameter that is related to the velocity spread of the neutron pulse. The best fit of this function is plotted on the data in Fig. 3. This fit implies a neutron pulse duration of 500 ps at the source with a standard deviation of the fit varying from 210 to 620 ps (illustrated as a thick bar on the plot).

Clearly an accurate measure of the pulse width at the source requires detectors with higher time resolution than those used in our experiments. However, these data confirm that the neutron pulse is well under one nanosecond and is likely to be in the few hundred picosecond regime. We also note that the data of Fig. 3 indicate that the neutron pulse is nearly monochromatic. The velocity spread of the neutrons is 5% implying an energy spread of about 10%. Such an energy spread can be used to gain information about the mean ion temperature because the spread is principally caused by Doppler broadening from finite deuterium ion energies. If the ion distribution is Maxwellian, the neutron energy spread is related to the ion temperature as $\Delta E = 83 T_i(\mathrm{keV})^{1/2}$ (in keV) [16]. This formula implies that our mean ion temperature is around 8 keV (average ion energy of 12 keV), consistent with other ion energy measurements [14]. Because the ions in our experiment are produced from Coulomb explosion of clusters, it

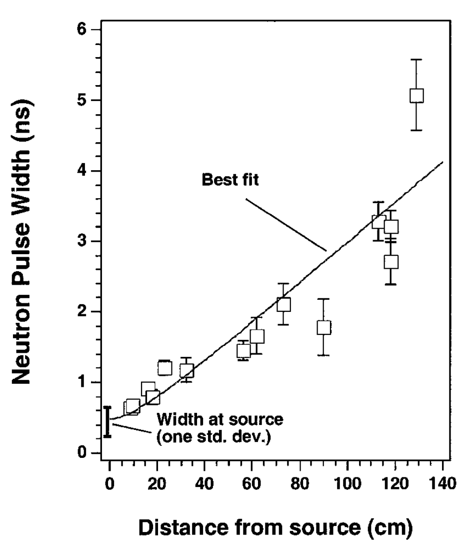

FIG. 3. Measured neutron pulse widths at different distances from the target. The extrapolated pulse width at the target is $\sim$500 ps.

is unlikely that the ion distribution is that of a Maxwellian. Nonetheless, ion time-of-flight measurements indicate an ion energy distribution near that of a Maxwellian, likely the result of a broad distribution of cluster sizes, yielding a broad range of Coulomb explosion energies. Thus, the above approximation likely yields a reasonable estimate of the ion temperature, at least in the hot tail of the ion distribution where most of the fusion reactions occur.

These results offer the possibility of using fast neutrons, for the first time, in ultrafast pump-probe experiments. One field of research that such a neutron source may have important application is in the study of neutron induced damage of materials [17]. Such damage studies have a broad importance including the study of materials to be used in future fusion reactors. Molecular dynamics studies suggest that fast particle damage in many materials (such as Si or most metals) is accompanied by transient local melting and recrystallization (with resulting formation of material defects) on a 10 to 1000 ps time scale (depending on material) [18]. A sub-ns neutron source like that studied here may permit time resolved experiments in which the neutron pulse drives damage in a material which is then probed by the synchronized short pulse laser. In addition, the small size of the neutron source can allow such experiments to be performed (on small samples) with high flux, even with modest neutron yield per laser pulse. For example, we estimate that neutron fluxes of $\geq 10^9$ $n/\mathrm{cm}^2\,\mathrm{s}$ would be sufficient to perform many initial radiation damage studies. With our current experiment we can produce $>3 \times 10^6$ $n/\mathrm{cm}^2\,\mathrm{s}$ within 1 mm of the source so a yield increase of about a thousand will be needed to enable such experiments.

In conclusion, we have measured the pulse width and energy spread of fusion neutrons from plasmas produced by intense laser irradiation of deuterium clusters. We find that the fast neutron pulse width is well under a nanosecond in duration near the source and that the energy spread of the neutrons is around 10%. It is likely that the pulse width of this source can be further decreased in the future if higher velocity ions can be produced in the laser-cluster interactions. Coupled with advances that would increase the neutron yield per shot, such short pulse laser produced fusion neutrons may represent a source of neutrons that will enable high repetition rate (10 Hz) neutron pump-probe experiments.

This work was conducted under the auspices of the U.S. Department of Energy under Contract No. W-7405-Eng-48. We acknowledge useful conversations with John Perkins, Tomas Diaz de la Rubia, Howard Powell, Dennis Slaughter, and Steve Zinkle. We also acknowledge the important technical assistance of Vince Tsai, Rich Shuttlesworth, and Gerry Anderson.

*To whom correspondence should be addressed.

[1] J. S. Brzosko, B. V. Robouch, L. Ingrosso, A. Bortolotti, and V. Nardi, Nucl. Instrum. Methods Phys. Res., Sect. B **72**, 119 (1992).

[2] A. Almazouzi, T. Diaz-de-la-Rubia, S. Ishino, N. Q. Lam, B. N. Singh, H. Trinkaus, M. Victoria, and S. Zinkle, J. Nucl. Mater. **251**, 291 (1997).

[3] D. W. Heikkinen, C. M. Logan, and J. C. Davis, Nucl. Sci. Eng. **106**, 228 (1990).

[4] P. W. Lisowski, C. D. Bowman, G. J. Russell, and S. A. Wender, Nucl. Sci. Eng. **106**, 208 (1990).

[5] M. Sadowski, J. Zebrowski, E. Rydygier, and J. Kucinski, Plasma Phys. Controlled Fusion **30**, 763 (1988).

[6] R. A. Lerche, D. W. Phillion, and G. L. Tietbohl, Rev. Sci. Instrum. **66**, 933 (1995).

[7] M. D. Perry and G. Mourou, Science **264**, 917 (1994).

[8] L. Disdier, J.-P. Garçonnet, G. Malka, and J.-L. Miquel, Phys. Rev. Lett. **82**, 1454 (1999).

[9] G. Pretzler *et al.,* Phys. Rev. E **58**, 1165 (1998).

[10] T. Ditmire, J. Zweiback, V. P. Yanovsky, T. E. Cowan, G. Hays, and K. B. Wharton, Nature (London) **398**, 489 (1999).

[11] E. Teller, *Fusion* (Academic Press, New York, 1981).

[12] T. Ditmire, J. W. G. Tisch, E. Springate, M. B. Mason, N. Hay, R. A. Smith, J. Marangos, and M. H. R. Hutchinson, Nature (London) **386**, 54 (1997).

[13] M. Lezius, S. Dobosz, D. Normand, and M. Schmidt, Phys. Rev. Lett. **80**, 261 (1998).

[14] J. Zweiback, R. A. Smith, T. E. Cowan, G. Hays, K. B. Wharton, V. P. Yanovsky, and T. Ditmire, Phys. Rev. Lett. **84**, 2634 (2000).

[15] R. Buck and E. Lent, *LLNL Energy and Technology Review*, 1993.

[16] D. R. Slaugher, Rev. Sci. Instrum. **60**, 552 (1989).

[17] R. S. Averback and T. Diaz-de-la-Rubia, Solid State Phys. **51**, 281 (1998).

[18] M.-J. Caturla, T. Diaz-de-la-Rubia, L. A. Marqués, and G. H. Gilmer, Phys. Rev. B **54**, 16 683 (1996).