Kilohertz sources of hard x rays and fast ions with femtosecond laser plasmas

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We demonstrate a new, stable, kilohertz femtosecond laser plasma source of hard-x-ray continuum and K_α emission that uses a microscopic liquid jet target that is continuous and debris free. Plasmas produced by ultrashort (50-fs) intense laser pulses from a fine (10–30-μm diameter) liquid Ga jet emit bright 9.3- and 10.3-keV K_α and K_β lines superimposed on a multikilovolt bremmstrahlung continuum. Kilohertz femtosecond x-ray sources will find many applications in time-resolved x-ray diffraction and microscopy studies. As high-intensity lasers become more compact and operate at increasingly high repetition-rates, they require a target configuration that is both repeatable from shot to shot and debris free. Our target provides a pristine, unperturbed filament surface at rates >100 kHz. A number of liquid metal targets are considered. We show the hard-x-ray spectrum described above. The source was generated by a 50-fs-duration, 1-kHz, 2-W, high-intensity Ti:sapphire laser. Using the same technology, we also generate forward-going sub-mega-electron-volt (sub-MeV) protons from a 10-μm liquid water target at 1-kHz repetition rates. Kilohertz sources of high-energy ions will find many applications in time-resolved particle interaction studies and will lead to efficient generation of short-lived isotopes for use in nuclear medicine and other applications. The protons were detected with CR-39 track detectors in both the forward and the backward directions up to energies of ~500 keV. As the intensity of compact high-repetition-rate lasers sources increases, we can expect improvements in the energy, conversion efficiency, and directionality to occur. The effect of these developments is discussed. As compact, high-repetition-rate femtosecond laser technology reaches focused intensities of $\sim 10^{19}$ W/cm^2, many new applications of high-repetition-rate hard-x-ray and MeV ion sources will become practical. © 2003 Optical Society of America

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1. INTRODUCTION

The many new and exciting discoveries that have recently been made in investigations of the interaction of intense femtosecond laser pulses with matter have all been made in regimes governed by the performance parameters of the lasers used in these investigations, generally the complex, low-repetition-rate, or single-shot, multistage chirped pulse amplifier, oscillator amplifier systems necessary to reach the focused intensities needed ($\sim 10^{17}$ W/cm^2). In this regime, apart from its precise alignment, little sophistication need be attached to the target system, since there is in general ample time for its manual replacement between shots. Much of the potential of these new discoveries, however, may well have practical significance only when the development of high-power, short-pulse laser technology takes this regime into the domain of compact high-repetition-rate facilities. These facilities will be the sources of relativistic electrons, ultrashort-duration x rays, and collimated mega-electron-volt (MeV) ions that will potentially open applications in time-resolved x-ray diffraction, x-ray radiography, and perhaps proton tomography and proton cancer therapy. High-repetition-rate, kilohertz, femtosecond Ti:sapphire lasers are now approaching this regime. As a conse-

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quence there is now the need to incorporate a target geometry that can (i) refresh the target material at a rate sufficient to provide a virgin target surface for each laser pulse; (ii) maintain an open, preferably ~4π, access to the target; and (iii) mitigate target debris to an extent that associated optics and diagnostic instrumentation are not damaged or destroyed by collateral particulate debris emanating from the target.

In this paper we present two investigations that operate in this regime. For what is the first time to our knowledge we have combined a high-repetition-rate kilohertz high-intensity 50-fs laser with microscopic liquid jet targets. In one case we utilize a liquid Ga jet as a metal target to create strong femtosecond K_α emission in the 10-keV region.[1] This source should find applications in time-resolved Bragg x-ray diffractions studies of structural changes in solid-state and biological media.[2,3] In a second experiment we have used a micrometer-scale liquid water jet and have detected semicollimated high-energy MeV protons emanating from the rear side of the water jet. Extrapolations of these experiments to higher intensities will, based on previous low-repetition-rate experiments,[4–8] provide higher fluxes of higher-energy protons, up to several tens of MeV in highly collimated beams. Although medical applications of these sources may be years away, pump–probe investigations of the interactions of these particles with biological and solid-state materials will be immediately possible.

2. KILOHERTZ 9.25-keV X-RAY SOURCE FROM A LIQUID Ga JET TARGET

For ultrashort x-ray sources to be more widely used, they must become compact and exhibit high average power. Moreover they must operate in a continuous regime with minimal laser adjustment and target replenishment. Developments in both laser and target technology are making this a reality. The development of high-repetition-rate (kilohertz) table-top femtosecond Ti:sapphire lasers is now reaching the point where amplified systems can provide the focused intensities required.[9–11] Amplified millijoule laser pulses of several tens of femtoseconds duration focused to spot sizes of a few micrometers produce intensities in excess of 10^{16} W/cm^2. The plasmas created by these lasers need target configurations that are continuous in operation, presenting each laser pulse with a virgin target surface that remains precisely aligned within the short Rayleigh range of the high-numerical-aperture focusing optics employed (usually a few micrometers). Rotating discs or drum targets do not easily meet these requirements. Reeled tape or wire targets may be better, but all of these approaches suffer from the additional drawback that they present a target surface that is many times greater in spatial extent than the focused spot size. This not only limits the solid angle of useful x-ray emission emanating from the plasma but, more seriously, leads to the generation of large amounts of target material projectiles and debris that are ruinous to the long-term life of the associated laser and x-ray optics. The severity of this problem has led to the development of innovative approaches that bring the target size down to the size of the focus volume, ultimately to targets having a mass equal only to that of the number of x-ray radiating atoms required. These new approaches, based on 10–80-μm-diameter liquid droplet[12] or cryogenic gas jet technology,[13] have demonstrated debris-free operation in excess of 10^7 shots, even for optics located only a few centimeters from the source. Moreover, these targets present almost 4π illumination from the source, and their limited spatial extent ensures a stable, localized, point x-ray source, an important requirement for many applications. They have, however, so far been limited to only few liquids (water, alcohol, copper nitrate solution, ethylene glycol) or gases (N_2, Xe, Ar).[14]

Here we report the use of a liquid metal jet as a target for a laser plasma. For kilohertz operation the liquid metal must have a flow velocity high enough to ensure that the jet is replenished and restored after each laser pulse. This is easily achieved, for separate hydrodynamic studies[15] show that flow velocities of between 10 and 100 m s^{-1} will allow repetition rates up to 100 kHz. In addition, the jet's small lateral size, with diameters as small as 10 μm, allows for nearly perfect matching to the spatial extension of the laser focus. The narrow spatial confinement of the radiation source provided by this geometry could be favorable for the generation of very short x-ray bursts in the 100-fs time range.

The experiments reported here concentrated on laser plasmas produced from stable Ga jets. In these experiments a jet diameter $d \sim 30\ \mu$m was used. Experimentally, stable jet lengths of the order of 3.6 mm were determined with a simple HeNe laser scattering diagnostic, agreeing well with the expectations of the theory of liquid jet stability.[16] The liquid Ga jet was located vertically within a vacuum chamber, at the focus of a $f = 7$ cm lens (Fig. 1). This focused the output of the high-power, 1-kHz-repetition-rate, Ti:sapphire laser system onto the Ga jet to a spot diameter of ~10 μm at a point approximately 2 mm from the nozzle. This system employs chirped pulse amplification and consists of a mode-locked Ti:sapphire oscillator, a regenerative amplifier,[1] and a double-pass booster amplifier; the laser provides compressed 50-fs pulses at a center wavelength of 780 nm (spectral bandwidth 25 nm) with a maximum average power of 3 W, corresponding to single pulse energies of 1.8 mJ. The effective contrast ratio of the amplified pulse was ~10^4. The intensity on target was 3×10^{16} W/cm^2.

The spectrum of the x-ray emission emanating from the plasma was determined with a Si-photodiode-based energy-dispersive x-ray detector.[17] It was placed 300 mm from the liquid jet source. To avoid pileup effects, a 1.5-mm-diameter pinhole was positioned 3 cm from the detector. A 12-μm Al foil placed directly at the detector entrance acted as a cutoff filter for energies below 2.5 keV. The detector output was connected by means of an amplifier to the input of a 1024-channel multichannel analyzer. An x-ray spectrum from the plasma generated from the Ga liquid jet is shown in Fig. 2. It consists of a broad continuum and two narrow features at 9.3 and 10.3 keV. These two peaks correspond to the characteristic K_α and K_β lines of Ga. The K_α feature has two components, at 9.22 and 9.25 keV, corresponding to the $K_{\alpha 1}$ and $K_{\alpha 2}$ lines, respectively. The broad continuum is predomi-

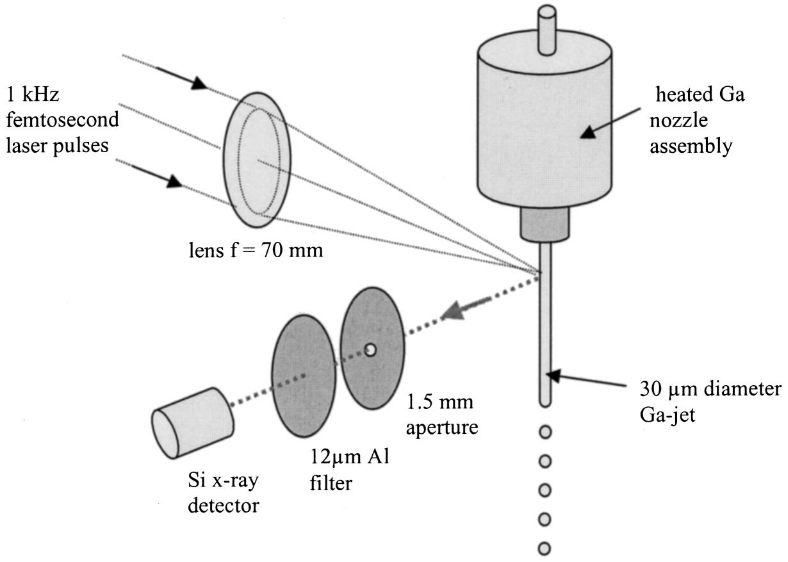

Fig. 1. Experimental arrangement of the liquid Ga jet laser plasma source.

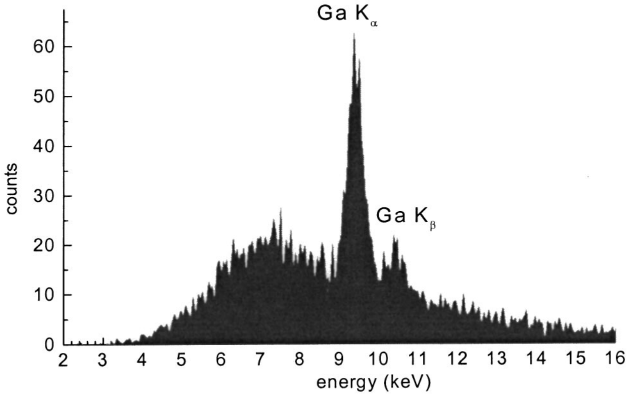

Fig. 2. Spectral emission of femtosecond laser plasma from a Ga jet target.

nantly free–free-collisional (bremsstrahlung) emission from the plasma, the falloff at the low energy end of the spectrum being due to the low-energy limit of the detector's sensitivity. The characteristic K_α and K_β lines originate from the collision of the high-energy electrons with cold target material as they propagate forward through the jet, or from those electrons that stream outward from the plasma interaction toward the laser, are captured by the Coulomb field they create, and stream back into cold jet material in the vicinity of the plasma. The energy-dispersive detector also gives an estimate of the absolute photon yield from the plasma. This was measured to be $\sim 4 \times 10^4$ photons/sr/pulse. On the assumption of isotropic emission into 4π sr, this would indicate a total emission power of x rays above the Al edge at 2.5 keV, of to $\sim 5 \times 10^8$ photons/s. Doubtlessly, the efficiency of this conversion can be improved, and as higher-powered lasers become available the hard-x-ray emission available from this type of source will increase. Moreover, the use of other materials will provide a broad spectral range of x-ray emission, either from the plasma continuum emission or from the characteristic K_α and K_β. Given its relatively debris-free nature, this ultrashort laser plasma source will therefore find wide applications in x-ray diffraction studies of solid-state and biological materials and in other time-resolved studies (radiography, fluorescence spectroscopy, etc).

3. HIGH-ENERGY PROTON GENERATION WITH A 1-kHz HIGH-INTENSITY FEMTOSECOND LASER

Many recent experimental investigations of laser plasmas produced by high-intensity, ultrashort pulse lasers from solid targets have shown the generation of MeV protons, predominantly from the rear-side of the target.[6,7] At sufficiently high laser intensities, $>10^{18}$ W/cm^2, these protons display distinct collimation and can contain a sizeable fraction of the initial laser energy. In fact, the intensity scaling of both the proton energy and their conversion efficiency from laser light appears to be faster than linear, prompting suggestions that at higher laser intensities such collimated beams of multi-MeV protons might be used for proton tomography, proton therapy for cancer treatment, and other similar applications. Until now, these studies have been performed with low-repetition-rate laser systems, either subpicosecond Ti:sapphire–Nd:glass hybrid laser systems or ~100-fs Ti:sapphire lasers. To exploit the potential of these high-energy particles, there is a need to take this exciting field toward operation at higher repetition rates with continuous operation, utilizing compact, operator-free laser systems; that is, toward a regime in which the laser–target system is simply regarded as a continuous source of pulsed, collimated MeV protons. Here we describe experiments that approach this regime, with a novel target geometry capable of presenting a stable target surface to the laser beam. By using a femtosecond laser system operating at a 1-kHz repetition frequency, we take, for what is to the best of our knowledge for the first time, these laser-based experiments on high-energy proton generation into a new operating domain. This regime, we believe, will open a broad range of new scientific avenues and applications.

The type of target we are using in this investigation will also contribute to the understanding of the mechanisms responsible for this intense proton generation. In the majority of previous studies the proton emission has been shown to originate from thin layers of water vapor or hydrocarbons deposited on the back of the target (generally resulting from contaminants within the evacuated vacuum chambers). In the experiments described here we use as a target a microscopic stream of water, some 10–30 μm in diameter, a target already rich in H atoms, and flowing sufficiently fast that it is unlikely to accumulate contaminants from the vacuum chamber. These experiments in addition present a different target geometry to the focused laser beam. Whereas nearly all previous investigations have used massive planar targets, in the present case we use a thin cylindrical target having a diameter similar in size to the laser focal size. This clearly defined microscopic target geometry has possible implications for the interpretation of the generation mechanisms of the protons themselves and the subsequent directionality of their emission.

The general configuration of the experiments reported here is shown in Fig. 3. The jet, of the Faubel design,[15,16] comprises basically a specially designed orifice, of diameter 10–30 μm, through which water is ejected from a pressurized reservoir that provides target material sufficient for several hours of operation. Proton emission from the target was detected with 50-mm-square CR-39 track detectors. Two detectors were located ~3 cm from the target, one monitoring the proton emission emanating from the rear of the target, subtending a total solid angle of ~40° in the forward direction of the laser beam. The second detector monitored protons accelerated from the front surface of the target on one side of the off-axis parabola, detecting particles coming from target at angles of 10°–90° from the backward direction of the laser beam. When developed for 6 h in a 6.25-mol solution of NaOH at a temperature of 70 °C, CR-39 is sensitive to protons having a minimum energy of 100 keV. The resulting circular tracks in the surface of the CR-39 are ~1–8 μm in diameter and were analyzed with a high-resolution microscope.

The angular distribution of the protons from the rear of the target in the plane normal to the jet direction had a circular symmetry and is shown in Fig. 4. These data were taken for a burst of 10^4 laser shots over 10 s. The horizontal distribution of the protons >100 keV emanating from the target has a (FWHM) angle of 40°. The total number of protons coming from the rear of the target per shot was estimated to be 3×10^3.

Spectrometric measurements were made of the energy distribution of the protons by use of various thicknesses of Mylar film. Thin strips of these films were laid across these detectors to measure both the energy spectrum of the protons and its angular dependence. A typical analysis of the energy distribution of the protons in the forward direction is shown in Fig. 5. Layers of 2 and 4 μm of Mylar selected protons having energies in excess of ~300 and ~500 keV. Protons of higher energy were not detected with thicker layers of Mylar absorbers. The scaling of the maximum proton energy measured in these experiments agrees well with previous measurements made with planar nonhydrogenic targets and larger low-repetition-rate laser systems.

The conversion efficiency of laser energy to protons in these experiments is fairly low, $10^{-5}\%$. However, we can expect this efficiency to rise as the laser intensity is increased. With continued improvements to high-repetition-rate femtosecond laser systems, intensities approaching 10^{19} W/cm^2 can be expected in the near future. In this regime proton generation will be much more efficient, and their maximum energy will be in the multiple-MeV range. Whether these particle fluxes will be sufficient to stimulate interest in their use for proton tomography or proton cancer therapy is not sure. However, they will without doubt be enough to initiate studies of the effect of short bursts of protons on solid-state and biological matter. The availability of synchronized femtosecond optical pulses, either at the laser wavelength or at other wavelengths in the IR, visible, or UV ranges derived from the laser wavelength by nonlinear optical methods, will provide many ways for time-resolved pump–probe determinations of reversible and nonreversible structural changes. Further, one can envisage these methods being augmented with time-resolved x-ray diffraction studies facilitated by a high-repetition-rate femtosecond x-ray source of the type described here.

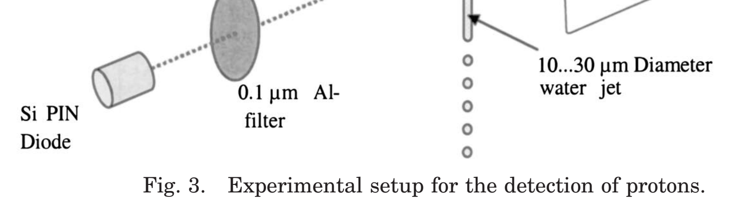

Fig. 3. Experimental setup for the detection of protons.

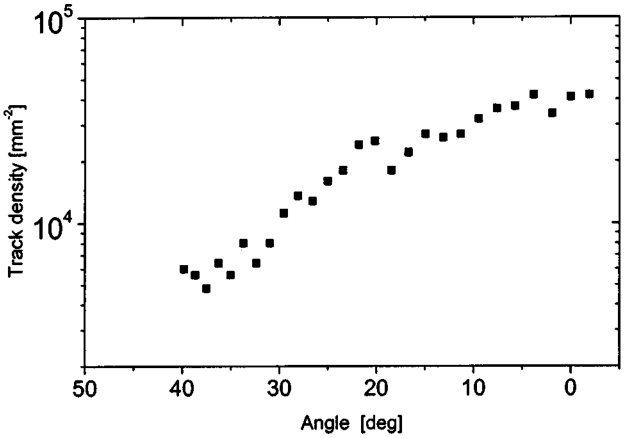

Fig. 4. Horizontal angular distribution of proton tracks on CR 39 detection plate counted up to the center of a circular distribution. Exposure time was 10 s (10^4 shots).

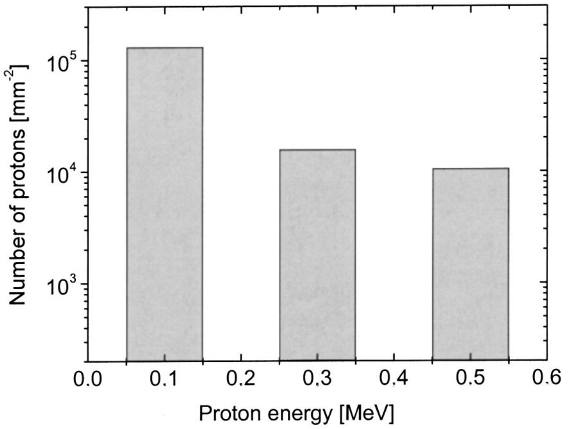

Fig. 5. Proton energy distribution counted on CR 39 with no Mylar, 1 layer, and 2 layers of 2 μm Mylar.

In summary, we have briefly described two important outcomes of the marriage of high-intensity, high-repetition-rate femtosecond laser technology with a new, microscopic liquid jet target design. We expect to see rapid development in this interaction regime in the future. At the Max-Born-Institute we are currently extending the performance of the laser system to higher powers and adapting the liquid jet technology for other target materials.

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