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Citation: Applied Physics Letters **93**, 121113 (2008); doi: 10.1063/1.2991293
View online: http://dx.doi.org/10.1063/1.2991293
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/93/12?ver=pdfcov
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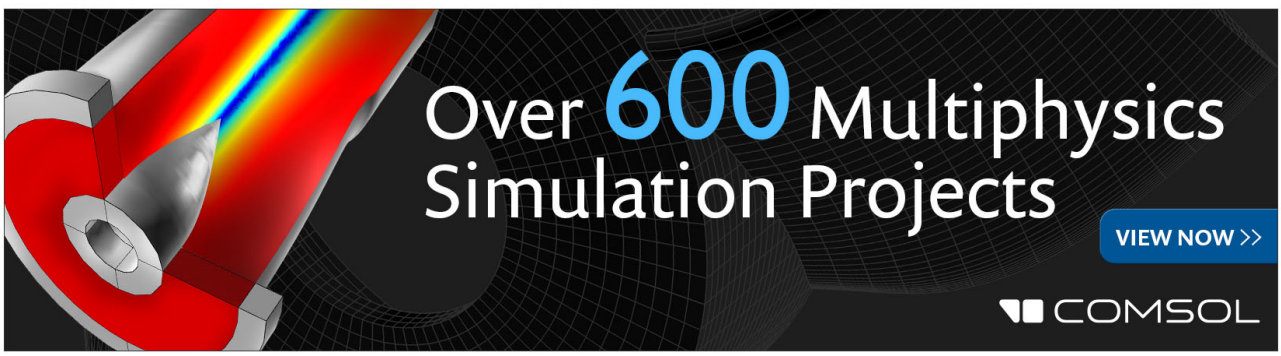

High-power 1 kHz laser-plasma x-ray source for ultrafast x-ray absorption near-edge spectroscopy in the keV range

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(Received 29 July 2008; accepted 9 September 2008; published online 26 September 2008)

A high average power broadband x-ray source is developed in the multi-keV range, based on the thermal emission of plasmas produced with a 1 kHz fs laser focused on high *Z* element target. This compact ultrafast x-ray source is used to measure the x-ray absorption near-edge spectroscopy of aluminum *K*-edge (1.559 keV) with noise lower than 1% of the absorption edge when accumulating laser shots over a few tens of seconds. That demonstrates its suitability to study atomic and electronic structures of matter during ultrafast phase transitions among solid, liquid, or higher energy density states. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2991293]

Ultrafast x-ray science grew up dramatically over the last decade, and the structural dynamics of various transient systems, ranging from molecules, crystals, and liquids to warm and hot dense plasmas can now be studied.[1] Among other x-ray techniques, near-edge x-ray absorption spectroscopy is a useful diagnostic to determine the atomic arrangement and the electronic state density of a wide variety of noncrystalline phases.[2] Such studies require broadband and ultrashort x-ray sources. Conventional synchrotrons offer routinely intense x-ray bursts but the duration is limited to about 100 ps.[3] Several experiments have been reported using the slicing technique, where the pulse duration is reduced down to subpicoseconds.[4] The price to pay is a strong reduction in the available photons. Tremendous possibilities are expected from future XFEL machines (100 fs), but the expected emission is intrinsically narrowband. Alternative compact x-ray sources have been proposed, based on the interaction of femtosecond laser pulses with various types of targets. High harmonic generation in a gas target offers a coherent and collimated beam of a few tens of femtosecond duration, but with a very low number of photons over 1 keV.[5] A recently proposed betatron x-ray source seems very attractive (a few tens of femtosecond duration in the keV range),[6] but it requires relativistic laser intensities, limiting the repetition rate and the compactness of the whole apparatus. Several recent near-edge absorption experiments using the laser interaction with a solid to produce bright incoherent emission in the XUV range have been reported.[7,8] Moreover, it has been shown that the bremsstrahlung emission of suprathermal electrons produced during the interaction could provide a useful broadband x-ray emission up to 10 keV.[9] This x-ray radiation is spectrally flat but the conversion efficiency between the x-ray energy and the laser energy remains at the 10^{-6} level. Some EXAFS experiments have been recently performed to study shock-compressed materials but the x-ray source uses 40 beams of a *kJ*-class laser in a single-shot mode.[10] Using more compact TW laser sources, it has been shown that the x-ray emission could be enhanced by several orders of magnitude, using the thermal emission from a high *Z* plasma.[11] Then, broadband *M*-shell spectra have been used in single-shot x-ray absorption experiments with *J*-class lasers.[12]

Here, we report the optimization of such a thermal broadband x-ray source, which uses a small-scale high repetition rate laser. The capability of this source is illustrated by clean Al *K*-edge XANES measurements performed over a short accumulation time (a few tens of seconds). The experimental setup is schematically presented in Fig. 1. At CELIA, our 1 kHz Ti:Sapphire laser delivers laser pulses with the central wavelength at 800 nm and an adjustable duration from 30 fs to 3 ps full width at half maximum (FWHM). The energy goes up to 5 mJ on target. Laser pulses are focused by a $f/4$ lens onto a rotating solid target with 25° incident angle and *P*-polarization. Due to the lens, the minimal duration at focus is estimated to be 45 ± 15 fs FWHM. The x-ray radiation produced from the laser-target interaction is emitted quasi-isotropically in the 2π sr on the target front side. A broadband x-ray spectrometer (not represented), based on a charge coupled device (CCD) camera operating in photon counting mode, has been set at 45° from the laser axis, in front of the target. It allows one to measure emission from 2.5 to 20 keV with 150 eV of spectral resolution. A conical KAP Bragg crystal (potassium acid phtalate with interplanar parameter $2d=26.64$ Å) has been set to disperse the x-ray emission in the 1.5–1.75 keV range with high spectral resolution [~1 eV (Ref. 13)]. In the present configuration, the crystal collects x rays at 90° from the laser axis onto a CCD, 3 cm shift from the x-ray focal line, in order to get spatial resolution along the crystal aperture. Various samples (Al,

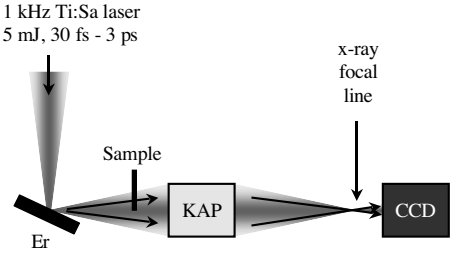

FIG. 1. Experimental setup. The laser-produced x rays are spectrally dispersed by a conical KAP Bragg crystal. Transmitted x-ray spectra through different samples (Al, CH, Al_2O_3) are recorded onto a cooled 16 bit CCD.

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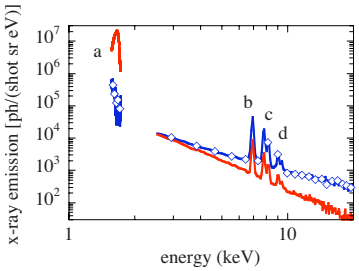

FIG. 2. (Color online) X-ray source spectra obtained when focusing 5 mJ laser pulses on erbium target. Blue line with white squares: 45 fs laser pulse duration. Red line: 2.7 ps laser pulse duration. *M*-shell spectra (a) are measured with the KAP crystal spectrometer. The energetic part of spectra is recorded with the broadband x-ray spectrometer that resolves the characteristic *L*-lines of erbium: L_α (b), L_β (c), and L_γ (d).

Al_2O_3, and CH) have been set between the x-ray source and the KAP crystal. Lens, rotating target, sample, and x-ray devices are set in a vacuum chamber below 10^{-4} mbar.

The erbium target has been chosen to optimize the *M*-shell x-ray radiation ($4f$-$3d$ transitions) in the spectral range of interest for Al *K*-edge XANES. X-ray emitted spectra are plotted in Fig. 2, respectively, obtained with 45 ± 15 fs and 2.7 ± 0.3 ps FWHM laser durations. With the shortest duration, the laser intensity reaches 10^{17} W/cm^2 on target, leading to a significant suprathermal population in the electron distribution. That induces efficient hard x-ray generation in bremsstrahlung (continuous part of the broadband spectrum) and characteristic core-excited Er *L*-lines. When increasing the laser duration (2.7 ps), the laser intensity decreases (10^{15} W/cm^2) and the laser coupling with suprathermal electrons becomes less efficient, as clearly pointed out by the lowering of the hard x-ray part of the spectrum. In contrast, the laser energy absorption into thermal electrons is enhanced, leading to a large increase in the thermal *M*-shell x-ray emission (about two orders of magnitude).

The x-ray source high resolution spectrum near the Al *K*-edge is displayed in Fig. 3 (2.7 ps laser pulse duration). An intense emission is observed over a quite large spectral range (200 eV). Besides this signal, a background noise is observed over the entire CCD chip. It is also plotted in Fig. 3, demonstrating a signal-to-noise ratio (SNR) of about 10^3:1. When decreasing the laser duration down to 45 fs, the SNR is reduced down to 10:1 due both to the *M*-shell x-ray emission decreasing and to the noise increasing. The background noise is understood as the KAP crystal fluorescence when illuminated by hard x rays emitted by the source.

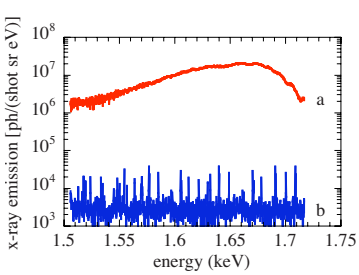

FIG. 3. (Color online) X-ray source spectrum measured by the high resolution KAP crystal spectrometer with 5 mJ, 2.7 ps laser pulses focused on erbium target. X rays are accumulated on CCD during 30 s exposure time. Er *M*-shell ($4f$-$3d$ transition arrays) x-ray emission (a). Background noise (b).

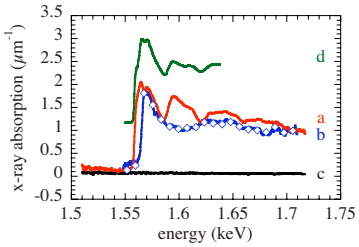

FIG. 4. (Color online) X-ray absorption spectra measured near the Al *K*-edge through different samples. Red: XANES of 1 μm Al (a). Blue with white squares: XANES of 2000 Å Al_2O_3 (b). Black: 5 μm CH (c). Green: XANES measurement of Al reported with a synchrotron at room temperature (vertically shifted) (d).

The x-ray energy integrated from 1.52 to 1.72 keV (and over 2π sr) reaches 5 ± 1 μJ per pulse. That corresponds to a conversion efficiency from laser energy of about 10^{-3}. The source geometry (quasi-point-like and isotropic) is well suited for dispersive EXAFS (Ref. 14) and allows us to record a whole spectrum in a single shot. In this experiment, the sample is set on the half of the collected solid angle. As shown in Fig. 1, the sample plane is projected on the CCD, leading to the measurement of the reference and the transmitted x-ray spectra, simultaneously. The transmission *T* is the result of their ratio. It is corrected from systematic defaults of KAP crystal reflectivity, deduced from a spectrum previously obtained without any sample. The absorption spectra μ, which are strictly speaking the linear attenuation coefficient, are finally deduced from the relation $T=\exp(-\mu d)$, where *d* is the sample thickness.

We use this method to get x-ray absorption spectra of various samples. XANES and EXAFS structures of solid Al (fcc) at room temperature are reported in Fig. 4, after accumulation of laser shots over 30 s. They look very similar to previous results obtained with a synchrotron source and reported in Ref. 15. The XANES spectrum of 2000 Å of Al_2O_3 is also presented. The difference with the pure Al absorption spectrum is clearly observed as an effect of the different atomic structures. The Al *K*-edge is shifted by 6 ± 1 eV, as expected with the higher oxidation level of Al_2O_3. We use a CH sample to estimate the residual noise on absorption measurements since no edge is expected in this spectral range. We get 0.6% and 0.3% rms compared to the amplitude of Al *K*-edge absorption, respectively, before and after this *K*-edge. This is limited by statistics of the photons detected by the CCD and could be improved by accumulating over a larger number of laser shots.

The use of high *Z* plasma *M*-shell emission can be extended typically up to the Ti *K*-edge (4.966 keV), limited both by the impossibility to use higher *Z* elements than uranium and by the plasma temperature achieved at the laser focus. The x-ray source duration is being measured and will be reported later. With solid and foil targets, a few picosecond duration is expected, as previously reported with similar experimental parameters.[16] Shorter duration (a few hundreds of femtoseconds) should be reached using high *Z* cluster target.[17] The use of different target elements makes possible the extension of such a technique up to the whole EXAFS domain, provided that an x-ray crystal is designed to offer large enough spectral range. For future time-resolved XANES experiments in a "laser pump–x-ray probe" configuration, the sample will have to be set quite close to the x-ray

source in order to probe the locally heated area of the sample. This is a standard limitation of such a dispersive EXAFS geometry.[14] It could be overcome by the use of an additional x-ray optic (e.g., polycapillary optic) that could collect and focus x rays from the source to the sample before being spectrally dispersed by a Bragg crystal.

In conclusion, a high-power broadband x-ray source has been obtained in the multi-keV range. It is based on the *M*-shell thermal emission of high *Z* plasmas produced by a high repetition (1 kHz) femtosecond laser. Measurements are presented with an erbium target. A high conversion efficiency of about 10^{-3} has been measured in a broad spectral range near Al *K*-edge (1.52–1.72 keV). X-ray absorption spectra of Al and Al_2O_3 have been measured, demonstrating the ability to resolve XANES structures with relative noise lower than 1% of the *K*-edge amplitude when accumulating x-ray source over a few tens of seconds. Besides its potential usefulness for static materials (as synchrotron sources), this compact setup is particularly suitable for future picosecond time-resolved XANES experiment aiming at studying atomic short-order range and electronic state density dynamics in a large variety of ultrafast phase transitions including noncrystalline phases.

The authors would like to thank C. Medina for her precious help on the laser as well as R. Bouillaud and A. Le Goff for their technical assistance. This work is partially supported by the Fond Européen de Développement Economique Régional and the Conseil Régional d'Aquitaine.

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