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1 kHz tabletop ultrashort hard x-ray source for time-resolved x-ray protein crystallography

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We describe a compact, reliable, and high-average-power femtosecond x-ray source and its first application to diffraction on protein crystal. The setup relies on a homemade Ti: sapphire system delivering 12 mJ at a 1 kHz repetition rate, associated with a small vacuum chamber especially designed for laser-plasma interaction and x-ray applications. This device allows the generation of $5 \times 10^{10}$ photons/s/sr at 8 keV and optimized x-ray irradiation of the studied sample, which can be placed close to the source. We present the diffraction pattern of a protein crystal in a divergent beam geometry, which is a first step to a subpicosecond x-ray diffraction experiment. © 2006 Optical Society of America


The generation of ultrashort hard x-ray sources is of prime interest for a broad range of applications in biology, chemistry, and physics, since it should extend three-dimensional structure determination to the femtosecond time scale. In recent years, several experiments aimed to follow structural evolution in femtosecond time scale. In recent years, several experiments aimed to follow structural evolution in femtosecond time scale. In recent years, several experiments aimed to follow structural evolution in femtosecond time scale. In recent years, several experiments aimed to follow structural evolution in femtosecond time scale. In recent years, several experiments aimed to follow structural evolution in femtosecond time scale.

But the low flux of these sources makes diffraction experiments on more demanding diffractors such as protein crystals more difficult. Therefore laser-plasma sources offer a great advantage over other x-ray sources, since they should allow temporal resolution down to the period of atomic vibration, typically of the order of 100 fs. To reach this objective, ultrashort x-ray sources must become compact and reliable and exhibit high average power. Moreover, their potential must be better utilized by developing a new diffraction method that takes advantage of laser-plasma source divergence.

Ultrafast monochromatic x-ray pulses can be generated by the interaction of ultrashort laser pulses of adjusted intensity with condensed matter. Recently, detailed experimental and theoretical studies have characterized the dependence of the emission over laser and target characteristics. Laser parameters such as pulse duration, focal intensity, polarization, incidence angle, and intensity contrast ratio play an important role in x-ray yield. Targets have to move fast enough to present a virgin surface at each laser shot while remaining precisely aligned. In addition, the concept of a robust high-repetition-rate laser-plasma source requires particular attention to the debris generated by the laser–target interaction.

In this Letter we describe an ultrashort hard x-ray source relying on a simple and unique 12 mJ, 1 kHz Ti:sapphire amplifier and present the diffraction pattern of a protein crystal exposed to a divergent beam. The whole setup has been carefully designed to gather the key features for time-resolved crystallography, such as reliability, high average power, and optimized flux on the studied sample.

The experimental setup is shown in Fig. 1. The laser system delivers 12 mJ, 160 fs pulses at a 1 kHz repetition rate, with high spatial quality, a measured level of amplified spontaneous emission $10^9$ lower than the main pulse, and contrast with intrinsic nanosecond and picosecond prepulses measured to be $5 \times 10^6$ and $5 \times 10^4$, respectively. The system relies on a commercial oscillator and regenerative amplifier (Hurricane, Spectra-Physics), delivering 0.8 mJ and seeding a homemade amplifier pumped by three frequency-doubled diode-pumped Nd:YLF lasers (two Jades, Thales-Laser; one Evolution-30, Positive Light). To limit thermal lensing due to large pump fluence onto the amplifying crystal (Crystal Systems, 10 mm × 10 mm × 10 mm, 90% absorption at 527 nm), we have designed a simple liquid-nitrogen-cooled Ti:sapphire amplifier and x-ray diffraction experiment. © 2006 Optical Society of America


Fig. 1. Design of the experimental setup. The beams of the two 20 W Nd:YLF lasers are in two different quasi-horizontal planes. Two Glan–Laser polarizing prisms and a Pockels cell (Medox, Thales Laser) reduce the nanosecond prepulses issued from leakage in the regenerative amplifier.
cooled cryostat that yields a decrease in the focal-
length power from $f = 49$ cm at 300 K to $f = 9.6$ m at
100 K while using 63 W of total pump power.\textsuperscript{15} We
obtained a very simple and compact 4-pass amplifier
that routinely delivers 18 mJ with excellent spatial
quality. Temporal compression of the pulses is
achieved in a two-grating compressor (120 mm
$\times$ 140 mm, groove density 2000 lines/mm), leading to
pulses of 12 mJ energy and 160 fs duration.

The x-ray source consists of a copper wire running
through a small vacuum chamber (see Fig. 2). The
wire, issued from a spool, is first flattened and moved
by a motorized rolling mill to expose a flat and fresh
surface to each laser shot. It then crosses the cham-
ber through two Teflon guides that maintain an air
pressure of 100 Pa and is guided by two free-rotating
bearings. The wire tension is controlled by a pair of
toothed wheels that pulled the wire out from the
chamber. The jitter of the wire motion is still reduced
by a small metallic finger positioned between the
bearings. The laser beam is focused with an 18 cm
focal-length lens and hits the wire with an incidence
angle of 60°. The laser intensity on the target is es-
timated to $3 \times 10^{16}$ W/cm$^2$, which is thought to be op-
timum for Cu-K\alpha x-ray yield.\textsuperscript{17} X-rays are collected
through a 17 mm diameter beryllium window placed
16 mm from the target. The deposition of debris on
the beryllium window is prevented by a 12.5 mm
wide plastic band continuously moving in front of it
(1 m/min). The laser beam entrance window is also
protected by a similar running band system, with a
slower speed (25 mm/h), and a band tilt that opti-
izes the laser beam transmission. The spectrum is
measured by analyzing the deposited energy of indi-
vidual x-ray photons on a thermoelectrically cooled
CCD camera (Andor Technology, DY434-FI-962),\textsuperscript{19}
that was previously calibrated with an Fe\textsuperscript{55}
source emitting K\alpha radiation at 5.9 keV. The emitted laser-
plasma x-ray spectrum consists of a broad continuum
and two narrow features at 8.05 and 8.91 keV corre-
sponding to the characteristic K\alpha and K\beta lines of
copper, respectively. If necessary, the K\beta photons can
be filtered with a thin Ni foil. The Cu K\alpha yield was
evaluated by considering the CCD efficiency at 8 keV
(given by the manufacturer), and integration of the
measured spectra over the K\alpha line and is found to be
$5 \times 10^9$ photons/s/sr. The size of the x-ray emitting
spot has been determined in the horizontal plane by
using a knife-edge technique and is 20 $\mu$m FWHM.
It is larger than the laser spot (7 $\mu$m FWHM), which may be due to either the jitter
in the wire motion or the spreading of fast electrons
in the target.

We performed an x-ray diffraction experiment on a
protein crystal with a 1 kHz laser-plasma source.
Conventional methods for single-crystal diffraction
data collection are based on a collimated beam. How-
however, collimating an x-ray beam greatly decreases the
flux available on the sample, making diffraction on
weak diffractors such as protein crystal difficult.
Here we have applied a new method first demon-
strated by Ho \textit{et al.},\textsuperscript{19} in which a stationary crystal is
exposed to a beam with a large two-dimensional con-
vergence or divergence. We recorded the diffraction
pattern of a lysozyme crystal, an enzyme widely dis-
tributed in animals and plants. The setup relies sim-
ply on a stationary crystal exposed to a divergent
beam. The sample is mounted on a goniometric head
placed at the outside of the vacuum chamber just be-
hind the beryllium window, thus being perfectly pro-
tected from the debris. The x-ray beam diameter is
reduced by a lead pinhole to the size of the crystal (a
cube with 400 $\mu$m sides), and the source angular
spread is thus 1°. This value is easily adjusted
by changing the hole. The CCD camera
(1024 $\times$ 1024 pixels of 13 $\mu$m $\times$ 13 $\mu$m) is placed
2.5 cm behind the sample, off the direction of the di-
rect x-ray beam. Acquisition consists of summing im-
ages of exposition time equal to 20 s. This exposition
time is chosen so that the probability of having more
than one photon per pixel is negligible.\textsuperscript{20} Conse-
fquently, each image is filtered to eliminate photons
whose energy is above the Cu K lines, and the final
image is then obtained by summing all the images.
This basic filtering method can be further improved
by use of event recognition techniques.\textsuperscript{21} Figure 3
shows the image obtained after only 50 min of acquisi-
tion. The resulting diffraction pattern resembles
that of a summation of precession images, while the use of a divergent beam with a stationary crystal allows simultaneous data collection over the range of the divergence angle. The main effect of beam divergence–convergence on the diffraction spots is tangential elongation, known as a Kossel line. For each spot the elongation is a function of the part of the angular spread of the source that can participate in the diffraction of the spot. Unfortunately, we observed that some high-energy photons get through our digital filtering by error, but their impacts are not larger than one or two pixels, and they do not change the evaluation of the diffraction pattern.

To collect a “complete” data set, one should record the diffraction pattern for different rotation angles of the crystal. But we underline that, for the same photon flux, the divergent beam geometry associated with the lack of x-ray optics allows us to significantly decrease the exposure time required to obtain a complete data set compared with that of the conventional oscillation method. Moreover, our objective is to study the structural modifications of a protein as it executes its function and not to determine an unknown structure. We will concentrate on specific locations in the protein, so a limited number of reflections will be necessary to determine local movements of the atoms.

In summary, we have presented an experimental setup for recording diffraction images of protein crystal with an ultrashort laser-plasma source. The whole setup lies on a single 3 m × 1.5 m optical table, which ensures the stability of a system producing 5 × 10⁹ hard x-ray photons/s/sr. The use of a divergent geometry with a sample located close to the source allows simultaneous data collection over the range of the divergence angle and optimized photon flux, decreasing the required exposure time. The signal-to-noise ratio we obtained in 50 min with a lysozyme crystal reaches 10⁵ for the most intense diffraction spots. Subpicosecond time-resolved diffraction experiments are in progress.

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