

Demonstration of Femtosecond Broadband X-rays from Laser Wakefield Acceleration as a Source for Pump-Probe X-ray Absorption Studies

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We present X-ray absorption measurements near the K-edge of laser heated aluminum in a pump-probe configuration using X-rays generated in a laser wakefield accelerator. A 30 fs duration laser pulse from the HERCULES laser system was split into two beamlines, with one used to heat a 4 μm thickness Al foil and the second to drive a laser wakefield accelerator. The laser-heated plasma was probed at various pump-probe delays using the femtosecond duration X-rays generated by betatron oscillations of the electrons in the wakefield. We observe an apparent blue-shift of the K-edge occurring on a sub-picosecond timescale in the transmission spectra.

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Ultrafast X-ray probes have become indispensable across many fields of physics. Amongst other applications, they have allowed time-resolved measurements of biological processes¹, chemical reactions² and the lattice dynamics of phonons³. Within high energy density physics, the advent of X-ray free electron laser (XFEL) facilities^{4,5} has enabled unprecedented measurements of ultrafast processes, including collisional ionisation⁶ and electron-ion equilibration^{7,8}. While monochromatic XFEL sources are ideally suited to such techniques, complementary approaches can be achieved using broadband X-rays. Broadband absorption measurements play a crucial role in validating opacity calculations for laboratory and astrophysical applications^{9,10}. More detailed spectroscopic measurements of the position and structure of absorption edges yield information about the ionic structure and the electron density of states¹¹. X-ray radiography may also be used to probe the position and structure of shocks^{12,13}. While laser-driven sources can be used for such applications¹⁴, their duration cannot yet be reduced much below the picosecond level^{15,16}. Furthermore, the analysis of absorption measurements can be complicated by the presence of spectral features in the backlight¹⁷. These constraints might be surpassed with the employment of laser wakefield accelerators as an X-ray source.

The idea of using a high-intensity laser to drive a plasma based accelerator has been around for decades¹⁸ but it has barely been 10 years since the first multi-MeV, mono-energetic beams were produced^{19–21} shortly followed by exploration of the simultaneously emitted synchrotron-like X-rays^{22–25}. Due to the ultra-short nature of the laser pulses used in laser wakefield acceleration (LWFA), the resulting electron beam and X-ray bunch also have a duration on the femtosecond time-scale, making the X-rays ideal for probing ultra-fast phenomenon. One of the most appealing experiments for a LWFA, is a pump-probe design. Because LWFAs are generated by an entirely optical source, it is possible to conduct ex-

periments with a very high degree of temporal accuracy by splitting the laser pulse into a pump beamline and a probe beamline. Previous pump-probe experiments using optical sources have shown how the ultrafast nature of lasers and acute timing accuracy can allow for measurements such as the disassociation of a molecule² or appearance of ionization states as a metal is heated^{26,27}.

Here, we present transmission measurements around the aluminum K-edge using the femtosecond duration “betatron” X-rays from a laser wakefield accelerator to probe a 4 μm foil. Recent measurements using this source have successfully demonstrated femtosecond resolution X-ray absorption measurements of warm dense copper using crystal spectroscopy²⁸. In this work, single hit photon spectroscopy measurements were made using a high-energy detection, deep-depletion CCD camera with various time delays between the pump and probe beams, ranging from 50 fs to 400 fs. The single hit spectroscopy has poor resolution compared with crystal spectroscopy but enables capturing single shot spectra and has a much larger spectral range.

The experiments reported here were carried out on the HERCULES laser system at the University of Michigan. Upon entering the experimental chamber, the single, 73 ± 3 TW beam, was split using a 75/25 beam splitter with 75% of the energy reflected from an $f/20$ parabolic mirror to drive a laser wakefield accelerator. The remaining 25% was focused 1500 μm behind the foil surface by an $f/3$ off-axis parabolic mirror to uniformly heat the Al target over an area of diameter 500 μm . This results in an intensity of $(5.0 \pm 0.5) \times 10^{18}$ W/cm² for the wakefield arm and $(1.0 \pm 0.5) \times 10^{16}$ W/cm² for the heater arm. The gas target for this experiment was a 5 mm gas cell filled with a premixed gas mixture of 2.5% nitrogen and 97.5% helium, with electron number densities in the range $(0.3 - 1.0) \times 10^{19}$ cm⁻³. The cell provides stability and consistency to the electron beam²⁹. Although not measured simultaneously to the X-ray spectra in

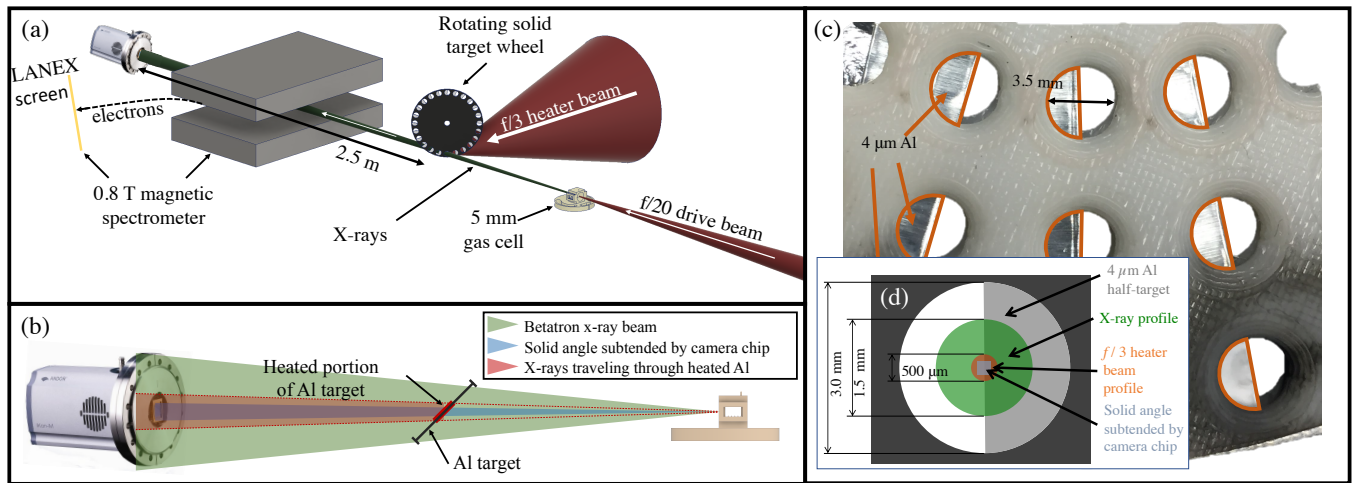


FIG. 1. (a) The experimental setup showing the orientation of the pump and probe beam lines relative to the solid Al target wheel. (b) Side view of overlap of heated region on Al foil with the betatron X-rays showing how the experiment was designed to ensure that X-rays measured on the CCD sensor would have passed through a uniformly heated region of foil. (c) Target design showing Al targets covering half of the mount holes. (d) Cartoon indicating the relative spatial overlap of the pump, probe, target and camera chip.

these experiments, electron spectra measured under similar conditions³⁰ typically had a large energy spread with a broad maximum energy peak that was at an average of 170 ± 20 MeV. Timing between the two beam paths was controlled using a delay stage on the pump beamline, allowing timing control with a temporal resolution of 10 fs. The solid target chosen for this experiment was a $4 \mu\text{m}$ aluminum foil. Aluminum was chosen because of the significant betatron X-ray flux around its K-edge for our experimental parameters and a thickness of $4 \mu\text{m}$ was chosen to ensure that a significant level of absorption can occur. The X-ray pulse had a divergence of approximately 15 mrad and a spectral intensity $d^2N/d\Omega d\hbar\omega$ of ~ 10 photons $\text{mrad}^{-2} \text{eV}^{-1}$ near the aluminum K-edge.

The pump and probe beam lines were perpendicular, with the target positioned at a 45 degree angle to both the pump and probe beam paths such that the delay across the beam was equal, as shown in Figure 1a. Because of fluctuations in X-ray flux from betatron oscillations, it is necessary to obtain data for a reference spectrum and an aluminum K-edge absorption spectrum in a single shot. In order to accomplish this, the Al targets were set up to cover half of the X-rays that hit the camera as shown in Figure 1c, with uniform heating over the full region covered by the X-rays. Figure 1b shows how the $500 \mu\text{m}$ diameter defocused $f/3$ beamline ensured that the projection of the heated target would overfill the CCD chip.

The aluminum target was placed in a circular, rotatable, plastic mount with 90 holes of 3.5 mm diameter to hold the targets. The Al was placed so that it covered half of each 3.5 mm target hole as shown in Figure 1c. The spatial alignment of the beams was performed by centering a $50 \mu\text{m}$ cross hair on each of the beam lines using a high magnification system. By doing so, we en-

sured that the spatial overlap of the beam centers were set to within $50 \mu\text{m}$ of one another. To achieve temporal overlap, it was necessary to have both beamlines imaged onto the same high magnification system. A piece of Mylar $2 \mu\text{m}$ thick was placed into one of the 3.5 mm holes in the target wheel to be used for temporal alignment. The timing was set by interfering the reflection of the $f/20$ focused pulse with the transmitted $f/3$ beamline through the Mylar. A delay stage on the $f/3$ beamline was adjusted until strong interference fringes appeared, indicating temporal overlap. The fringes existed over roughly one pulse duration and so we were able to confidently set the timing of the beams to within the 30 fs pulse duration. The $\Delta t = 0$ timing was set in vacuum at the start of the experiment and adjusted before each shot series.

X-ray absorption data were obtained using an Andor iKon-L deep-depletion X-ray CCD camera. The CCD images were analyzed to generate spectra by using a single photon counting algorithm³², by converting the signal read from each pixel into a specific photon energy. This is possible because there is a linear relationship between produced photoelectrons and incident photon energy over a large energy range³³. In an ideal situation when the photon flux is sufficiently low, we are confident that the signal in each pixel is produced by only one photon hit, so taking a histogram of the hits across the CCD reveals the spectrum. If the flux of X-rays is too high, two or more photons can hit the same pixel, adding together to be registered as a higher energy photon hit. Additionally, the X-rays often undergo multiple collisions as they lose energy in the depletion layer of the CCD chip or the electron charge diffuses away from the original site. These multiple collisions often lead to a primary pixel of high signal along with several of the neighboring pixels containing residual energy from the subsequent collisions,

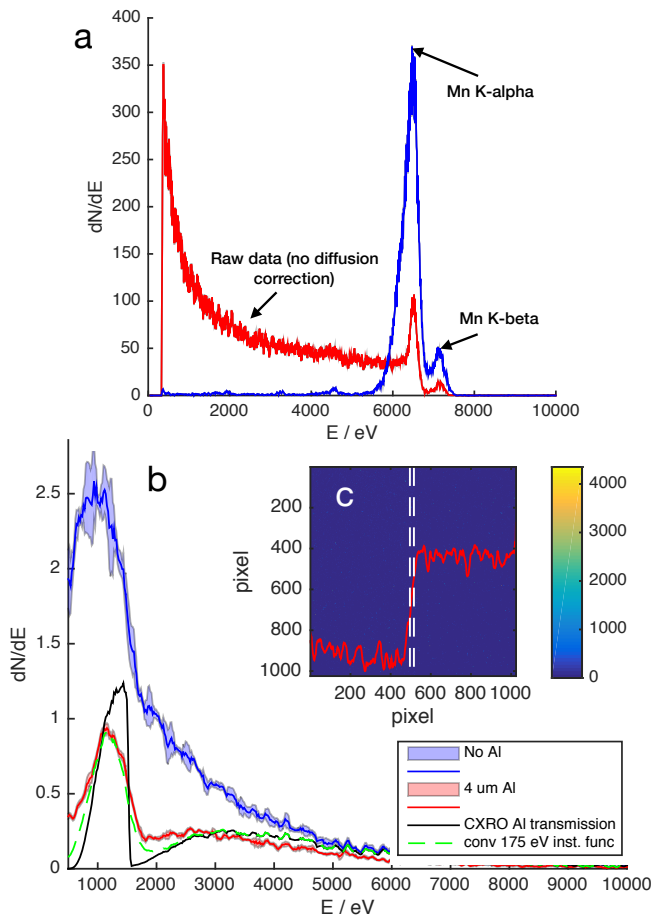


FIG. 2. Single hit spectroscopy. (a) Fe-55 source. The red line is the raw spectrum with each pixel being analyzed as a single photon. The blue line shows the spectrum after applying an algorithm to account for charge spreading and double hits. The low energy shoulder from the red line is removed and clear K-alpha and K-beta peaks result, with a FWHM of $\Delta E \approx 200$ eV. (b) Reference and unheated Al absorption spectra calculated from each half of the image in (c), along with the reference spectrum multiplied by a tabulated transmission curve for $4 \mu\text{m}$ of Al (CXRO³¹) and also the transmission curve convolved with a 175 eV width Gaussian instrument function. The shaded regions represent the statistical error though there is likely additional systematic error. (c) The raw CCD image with a region of interest selected by identifying the edge in the X-ray transmission (the red line shows the vertically integrated profile).

which will be recorded as lower energy photons.

We have improved the single photon counting method to account for these effects. For two-photon hit events, we make use of the fact that the number of photons per pixel will obey Poisson statistics. The spectrum of two photon events will be proportional to the autocorrelation of the real photon spectrum, weighted by the Poisson probability distribution³⁴. An auto-correlation of the measured spectrum will be an estimate of the two-photon hit event spectrum, which can then be subtracted from

the original spectrum, i.e. we consider the two-photon hit events as a perturbation. This technique was verified with Monte Carlo simulations to determine the accuracy of the convolution and its limits, which translate to 30 % occupancy corresponding to an $\mathcal{O}(0.01\%)$ error. Charge spreading is commonly modeled as a diffusive process³⁵. We may therefore reconstruct the original pattern of hits by solving the diffusion equation with time in reverse, $\partial f/\partial(-t) = \kappa \nabla^2 f$, which is equivalent to solving the diffusion equation with a negative diffusion coefficient $-\kappa$. Defining $s(x, y) = f(x, y, t = 0)$ we seek the solution of $\partial f/\partial t = -\kappa \nabla^2 f$ at $t = \infty$. This is because $a(x, y) = f(x, y, t = \infty)$ is a stationary distribution of this equation. The removal of a finite time renders the precise value of κ irrelevant. To solve this numerically, we first find the finite difference gradient of the charge in the x and y directions, then use it to transport the charge to the adjacent cells in the direction of the gradient and iterate. This method is simple, conserves charge, ensures positivity and has a stationary solution for isolated single pixel values. The spectral retrieval method, including charge spreading and double hits correction was tested by applying it to an Fe-55 source spectrum recorded with the same CCD as used in the experiment, as shown in Figure 2. The algorithm recovers Gaussian peaks around the Mn K-alpha and K-beta lines as well as the fluorescence and escape peaks which could not be achieved with 3×3 cell charge collection methods.

For pump-probe absorption measurements, the X-ray flux on the camera was limited by placing it 2.5 meters away and using $20 \mu\text{m}$ of beryllium as a light filter. Figure 2c shows an example of the half-covered raw images that were obtained for various time delays. The dark half of the square is from X-ray absorption in the aluminum and the light half is X-rays passing through vacuum. The red line shows a vertically integrated lineout, showing how the edge of the foil could be identified. Each data image like the one in Figure 2c were analyzed to produce two separate spectra; one reference spectrum with no Al between the source and camera and one data shot with a heated or cold Al foil present between the source and camera. An example set of two spectra is shown in Figure 2b. The blue curve is the reference spectrum, the red curve is the spectrum through $4 \mu\text{m}$ of Al (unheated). The black curve is the reference spectrum multiplied by the tabulated transmission for Al in the CXRO dataset³¹, and the green curve is the transmission convolved with a 175 eV width gaussian instrument function.

To obtain the transmitted spectra $T(\hbar\omega)$ for different timing delays between the pump and probe beamlines, we divided the spectrum after absorption by the laser-heated Al, $A(\hbar\omega)$ by the reference spectrum $R(\hbar\omega)$, $T = A/R$. Figure 3b shows several of these transmission curves superimposed for various time steps. The lines show the spectra averaged over typically 2-3 shots and then smoothed, the shaded regions show the error, which is a combination of both the noise in a spectrum before smoothing and the statistical error. Although typically

10 shots per timing were taken, most had to be rejected because the photon flux was too high such that the spectrum could not be reconstructed accurately. In all of the data shots used, the flux of photons was less than 30 % of the pixels filled on the *A* side. Shots that did not satisfy this criterion, or for which a clear sharp gradient (i.e. foil edge) could not be determined were rejected. Evidently, the *A* side always had a lower occupancy than the *R* side meaning the reference spectra reconstruction was less reliable. The increased occupancy on the *R* side results in more double hit events, thus decreasing the reliability of the reconstructed spectrum. In addition, the X-ray beam was not uniform across the left and right sides of the CCD image³⁶ for some shots, which had to be taken into account when generating a transmission spectrum.

In Figure 3b, the red line shows the transmission spectrum reconstructed from unheated Al, while the other curves indicate the transmission at various delays, as indicated by the legend. For the negative delay, the X-ray pulse arrives before the heater beam and so the absorption spectrum should be the same as the red curve. We attribute the observed increase in transmission at just below 1500 eV to K-shell fluorescence from the laser heated foil. This emission contribution to the absorption spectrum is apparent for all delays.

In addition to the fluorescence there is also an observed time dependent shift of the K-edge. Figure 3a shows the position of the edge, calculated as the midpoint between the peak and valley of the curve, as a function of delay, indicating a time dependent shift of the edge to higher photon energy. Heating of the target at 10^{16} Wcm⁻² is expected to generate hot electrons with $\mathcal{O}(8$ keV) energies³⁷⁻³⁹ that will propagate into the target on a fast timescale, ionizing and heating through collisions and Ohmic effects. Blue shifts of the K-edge transmission have been observed on previous experiments where the heating is rapid and relatively isochoric⁴⁰⁻⁴⁴. Blue shifts of the K-edge may be due to collisional ionization of inner shell electrons by the hot electron population of the plasma. The shift we observe here is consistently observed for positive delays. Due to the limited spectral resolution here, we are not able to distinguish between a shift and the appearance of fine scale absorption structure above the K-edge due to the local atomic structure near the absorbing atom¹¹. Nevertheless, rapid changes in the absorption spectrum are resolved by laser-wakefield generated X-ray probe.

In conclusion, we have demonstrated proof-of-principle single shot time resolved absorption spectroscopy of rapidly heated dense plasma using ultra-fast X-rays from a LWFA. Other effects such as hollow atom formation^{45,46}, continuum lowering^{40,41} and warm dense matter opacities could also be studied using these techniques. The spectral resolution of the single photon counting technique used in this work was very limited and not appropriate for studying fine edge structure. For future work, it would be beneficial to use single photon counting in conjunction with crystal spectroscopy to ob-

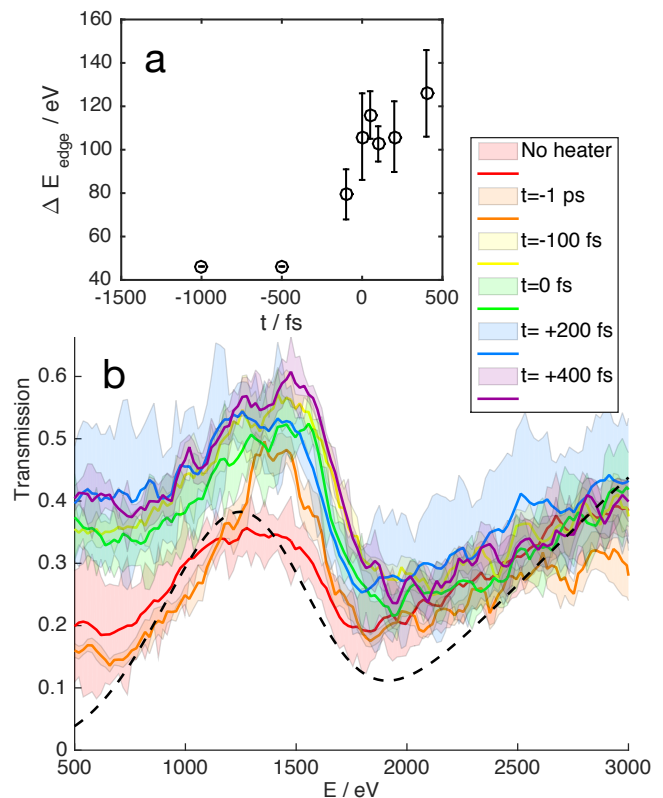


FIG. 3. (a) The shift in the K-edge measured from the transmission spectra at different pump-probe timing delays. (b) Transmission spectra of Al at various delays of the probe beam in reference to the pump (or heater) beam. Shaded regions represent the statistical error of the measurements. The dashed black curve shows the cold aluminum transmission convolved with the 175 eV width Gaussian instrument function for reference.

tain higher resolution transmission curves⁴⁷.

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