

Spatiotemporal ionization dynamics in nonthermal copper plasma pumped by femtosecond relativistic laser pulse

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Understanding ultrafast ionization dynamics in high-density plasmas is essential for applications such as particle acceleration, short-pulse x-ray generation, and laser fusion. However, resolving ionization states inside optically opaque plasmas with femtosecond resolution remains challenging. Using femtosecond-resolved x-ray absorption spectroscopy, we directly measured inner-shell vacancies, overall charge states, and the nonthermal electron population responsible for ionization in solid-density copper plasma. In copper plasma generated by a 40-fs relativistic laser pulse, ionization proceeds within 0.5 ps. M- and outer-shell ionization persist for over 10 ps, whereas L-shell vacancies decay within 4 ps. The population of electrons capable of L-shell ionization decreases in picoseconds, in agreement with particle-in-cell simulations incorporating atomic processes. Our spectroscopic approach provides direct experimental insights into ultrafast ionization dynamics in high-density plasmas, supporting improvements in the modeling of laser-driven plasmas.

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I. INTRODUCTION

Irradiation of solid matter with a high-intensity femtosecond laser pulse leads to an ultrafast transition to a plasma state on femtosecond-to-picosecond timescales, a process known as isochoric heating [1–4]. The resulting plasma contains two distinct free-electron components: high-energy, nonthermal fast electrons and relatively low-energy, thermal bulk electrons. Fast electrons, generated mainly by collective processes in the laser-plasma interaction region [5], propagate into the dense plasma and transfer their energy to the bulk electrons. They also induce electron-impact ionization by colliding with bound electrons of atoms and ions, preferentially removing outer-shell electrons because of their larger cross sections [6,7]. In contrast, inner-shell ionization, though less frequent, produces energetically unstable vacancies that are rapidly filled by recombination, radiative decay, or Auger processes on femtosecond-picosecond timescales. The plasma is generally expected to approach thermal equilibrium through Coulomb collisions within a few picoseconds. However, the detailed ionization dynamics and evolution of the electron energy distribution during this nonequilibrium stage remain

incompletely understood and have been inferred largely from simulations. In particular, how rapidly electrons are removed from different atomic shells and how the corresponding electron energy distribution evolves toward thermal equilibrium remain difficult to determine experimentally.

Directly measuring the ionization dynamics of bound electrons, including inner shells, together with the time evolution of the electron energy distribution in dense plasma with femtosecond resolution remains extremely challenging. However, such measurements are essential for understanding ultrafast ionization dynamics before thermal equilibration and for improving high-intensity-laser applications such as particle acceleration [8–12], short-pulse x-ray generation [13–15], and laser fusion [16–18]. To address this challenge, one might consider using $M\alpha$ and $L\alpha$ emission to probe M- and L-shell vacancies, but in micron-thick transition metals these soft-x-ray photons cannot escape the dense plasma due to its high opacity [19]. Electron spectrometers [20] detect electrons emitted from the target surface but do not provide direct information on electron energies within the plasma interior. x-ray Thomson scattering [21] can provide an estimate of the electron energy distribution, yet it offers no information on inner-shell ionization. Therefore, a femtosecond-resolved diagnostic capable of penetrating and probing the interior of solid-density plasma is required to elucidate the ultrafast transition from solid to plasma.

Here, we demonstrate femtosecond-resolved observation of inner-shell ionization dynamics and nonthermal electron evolution in solid-density plasma using resonant x-ray absorption spectroscopy (XAS) driven by an x-ray free-electron laser (XFEL) [22,23]. An XFEL pump-probe experiment provides a powerful diagnostic of dense plasmas [24–26], owing

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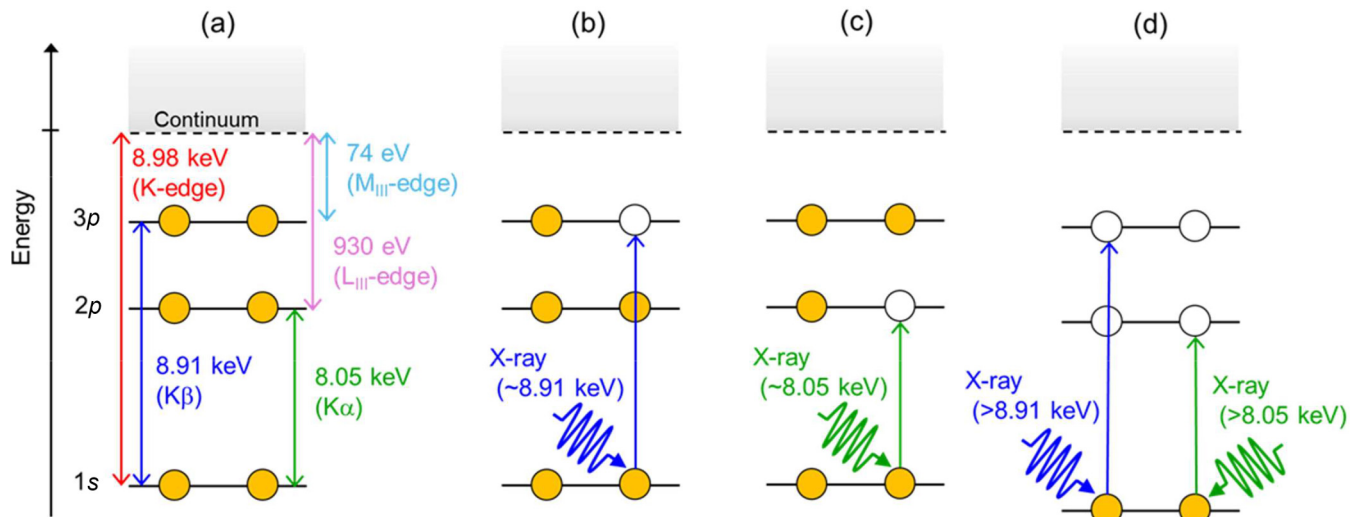


FIG. 1. Schematic illustration of resonant x-ray absorption spectroscopy. (a) Energy-level diagram of cold Cu. (b) Inner-shell-ionized ion with a $3p$ vacancy. X-rays with energies corresponding to the $K\beta$ transition are resonantly absorbed, resulting in a reduction of the transmittance. (c) Inner-shell-ionized ion with a $2p$ vacancy. X-rays with energies corresponding to the $K\alpha$ transition are resonantly absorbed, leading to a decrease in the transmittance. (d) Highly charged ion with enhanced binding energies due to reduced electron screening. X-rays with energies higher than the $K\alpha$ and $K\beta$ transitions are absorbed.

to its femtosecond duration [27–30], high penetration into solid-density matter [31,32], and tunable photon energy. The principle is illustrated schematically in Fig. 1. Figure 1(a) shows the energy-level diagram of cold Cu, where the $2p$ and $3p$ shells are fully occupied, preventing resonant absorption through these transitions. Electron-impact ionization by high-energy electrons creates vacancies in the $2p$ and $3p$ shells, with electron energies exceeding the M- and L-shell ionization energies of 74 and 930 eV, respectively. Figures 1(b) and 1(c) schematically illustrate resonant $1s$ – $3p$ ($K\beta$) and $1s$ – $2p$ ($K\alpha$) transitions, respectively, which require vacancies in the $3p$ and $2p$ shells. When vacancies are created, XFEL pulses tuned to the corresponding transition energies are resonantly absorbed, reducing the transmittance [33]. In highly charged ions, reduced electron screening increases the binding energies, shifting the resonance toward higher photon energies, as illustrated in Fig. 1(d) [34,35]. If the transmittance reduction persists beyond the intrinsic lifetime of the vacancy, it indicates continuous vacancy production driven by impact ionization. The transient resonant absorption therefore provides a direct probe of the generation and relaxation of high-energy electrons with energies exceeding the respective inner-shell binding energies. Combining resonant XAS with conventional x-ray absorption near-edge structure (XANES) measurements further enables simultaneous investigation of inner- and outer-shell ionization dynamics. Because the relevant transition energies lie in the hard-x-ray regime, this method provides access to inner- and outer-shell dynamics in solid-density plasma simply by tuning the XFEL photon energy.

Our results provide the first direct experimental access to the time evolution of nonthermal electrons and shell-resolved ionization dynamics inside solid-density plasma, which has been inaccessible with previous diagnostics. We determine the characteristic timescales of inner- and outer-shell ionization and electron relaxation in solid-density plasma. Pump-probe measurements enable simultaneous tracking of M-, L-, and

outer-shell charge states together with the evolving free-electron energy distribution, providing direct evidence of high-energy electron generation and decay prior to thermal equilibration. Comparison with particle-in-cell (PIC) simulations including atomic and collisional processes provides a qualitative picture of ultrafast plasma formation and the evolution of nonthermal electrons in dense plasma. The resulting time-, spatial-, and spectrally resolved transmittance data provide useful benchmarks for the further development and validation of integrated plasma and atomic-physics models.

II. XAS MEASUREMENT USING A SPATIALLY RESOLVED DISPERSIVE SPECTROMETER

In XAS measurements, accurate determination of transmittance and absorption requires comparison between a signal spectrum and a reference spectrum. However, this comparison becomes challenging when spectra are obtained from different single-shot XFEL pulses, because self-amplified spontaneous emission (SASE) XFEL pulses exhibit significant shot-to-shot spectral fluctuations [22]. When independent XFEL pulses are used for signal and reference measurements, improving statistical accuracy typically requires accumulation over a large number of shots [36]. Such averaging is impractical in pump-probe experiments combining a high-power optical laser and an XFEL pulse, where the available number of shots is limited [25,37–40]. To overcome this limitation, we developed a single-shot XAS method based on spatially resolved dispersive spectroscopy using an analyzer crystal and two elliptical mirrors [41]. When the XFEL beam spot is significantly wider than the optical laser focal spot on the sample, spectra from the laser-pumped and unpumped regions can be recorded simultaneously within a single XFEL shot. The spectra from the unpumped regions provide an *in situ* reference that is free from laser-induced modification. By comparing the pumped-region spectrum with this shot-

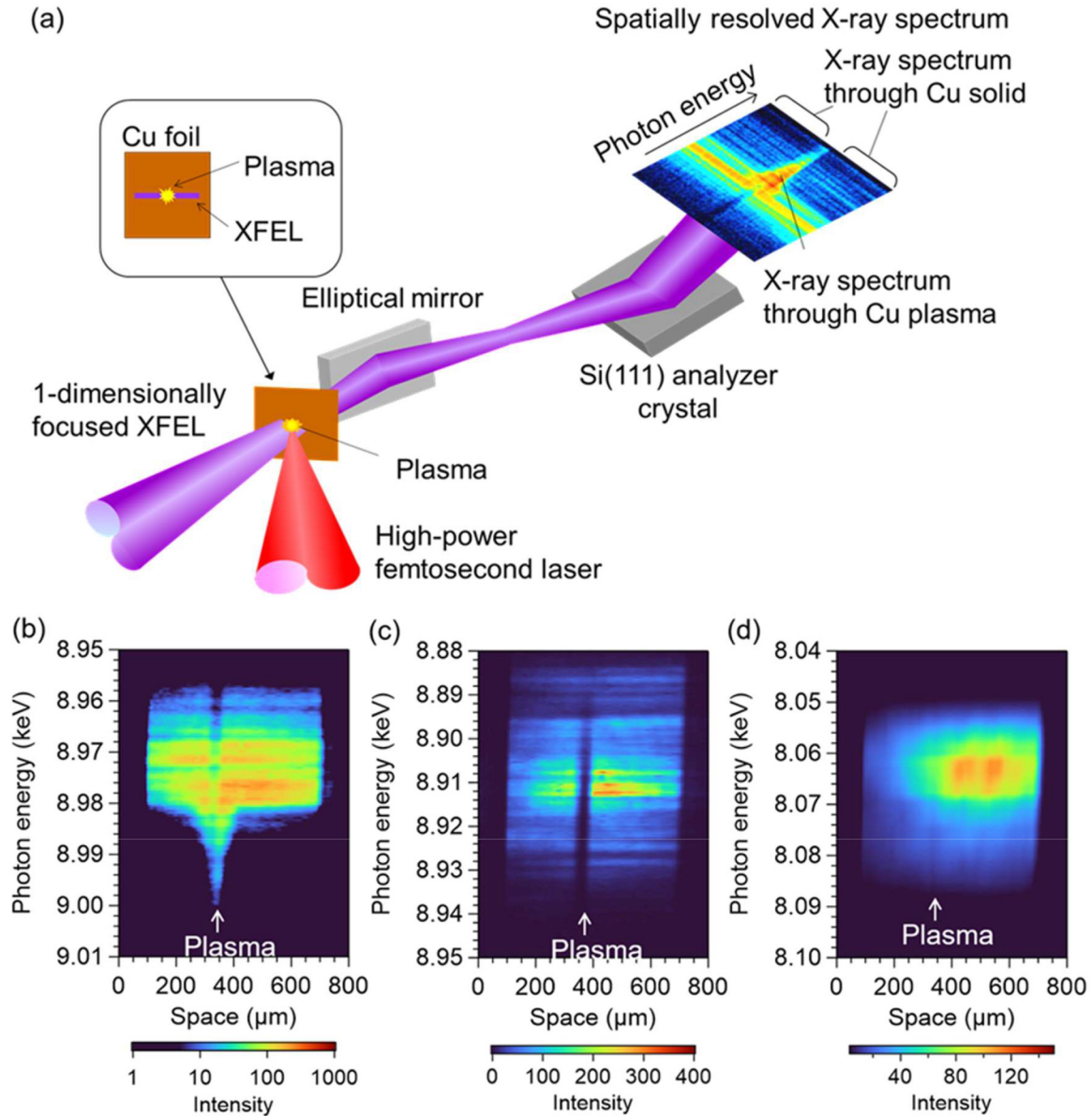


FIG. 2. (a) Schematic of femtosecond x-ray absorption spectroscopy for Cu plasma generated by high-intensity laser irradiation. A spatially resolved single-shot spectrum was obtained with a Si(111) analyzer crystal and multi-port charge-coupled device (MPCCD) detector. Because the XFEL beam is longer than the laser-heated region, spectra from both the plasma (central) and cold Cu (outer) regions are measured simultaneously. Single-shot spectra around (b) the Cu K edge, (c) K β line, and (d) K α line at 0.9 ps delay.

by-shot reference spectrum, the transmittance and absorption coefficients of the laser-irradiated region are available without averaging over multiple XFEL pulses. This approach largely cancels the effects of SASE spectral fluctuations and enables precise measurements under limited-shot conditions.

The experiment was performed at BL2 EH6 of SACLA using a combined XFEL and high-power femtosecond laser system [25]. Figure 2(a) shows the experimental layout. The sample was a 20- μm -thick copper (Cu) foil. A Ti:sapphire laser pulse was incident at 28° relative to the surface normal with a focal spot size of 30 μm (FWHM). The typical pulse energy and duration were 8 J and 40 fs (FWHM), corresponding to an intensity of approximately $1 \times 10^{19} \text{ W}/\text{cm}^2$. According to ponderomotive scaling [42], the corresponding fast-electron slope temperature is $\sim 1 \text{ MeV}$.

For XAS measurements, we employed a spatially resolved dispersive x-ray spectrometer [41] mentioned above. The first elliptical mirror provided one-dimensional focusing of the XFEL beam onto the Cu foil, while the second mirror improved spatial resolution along the nondispersive direction. Both mirrors operated at a grazing incidence angle of 5 mrad, and the XFEL beam divergence after focusing was 2.5 mrad in both the horizontal and vertical directions. The XFEL beam had a width of 600 μm (FWHM) in the spatial direction and was focused to 2 μm (FWHM) in the dispersive direction, with an incidence angle of 45° relative to the surface normal. The XFEL probe intensity was sufficiently low that its influence on the plasma dynamics is negligible. Because the XFEL beam width exceeded the laser-pumped region [see inset of Fig. 2(a)], spectra from the pumped and unpumped

regions were recorded simultaneously within each XFEL shot. The beam profile was sufficiently smooth over the spatial direction, and the transmission was determined from the ratio of the pumped and unpumped regions within the same XFEL shot, minimizing the effect of spatial nonuniformity. Because the XFEL spectral bandwidth was approximately 15 eV (FWHM), two XFEL photon energies with slightly different central energies were employed for each experimental configuration to obtain sufficient spectral coverage around the Cu K-absorption edge, $K\beta$, and $K\alpha$ regions. The estimated spectral resolutions were 1.3, 1.3, and 1.1 eV (FWHM) for the K-edge, $K\beta$, and $K\alpha$ measurements, respectively, as calculated from the convolution of the analyzer crystal response and the detector pixel resolution.

The experimental data were analyzed using the same method as described in Ref. [41]. The plasma transmittance was defined as

$$T_{\text{plasma}}(E, t) = \frac{I_{\text{plasma}}(E, t)}{I_{\text{reference}}(E, t)} T_{\text{cold}}(E), \quad (1)$$

where E is the photon energy, $I_{\text{plasma}}(E, t)$ and $I_{\text{reference}}(E, t)$ are the transmitted intensities through the laser-irradiated and unpumped regions, respectively, obtained from the spatially resolved transmitted spectra recorded in each XFEL shot. The transmitted intensities were calculated as

$$I_{\text{plasma}}(E, t) = \sum_i \left(\int_{\text{width}} S_{\text{plasma},i}(E, x, t) dx \right), \quad (2)$$

$$I_{\text{reference}}(E, t) = \sum_i \left(\int_{\text{width}} S_{\text{reference},i}(E, x, t) dx \right), \quad (3)$$

where x denotes the spatial coordinate along the target surface and i represents individual XFEL shots. The spatial integration width was 15 μm , and 4–7 shots acquired under identical experimental conditions were used for each XFEL photon energy and delay condition. The spectra obtained with the two XFEL photon energies were combined to extend the spectral coverage. $I_{\text{reference}}(E, t)$ was obtained by fitting the spatial intensity profile of the cold regions on both sides of the plasma region after excluding the plasma region for each photon energy. The reference spectra were therefore determined from the unpumped regions recorded simultaneously with the plasma spectra in each XFEL shot. The ratio $I_{\text{plasma}}(E, t)/I_{\text{reference}}(E, t)$ represents the relative transmission with respect to the cold state. Because the pumped and unpumped regions are simultaneously recorded within the same XFEL shot, shot-to-shot fluctuations of the XFEL intensity are largely canceled in the relative transmission measurement, thereby reducing the uncertainty associated with XFEL intensity fluctuations. Multiplication by $T_{\text{cold}}(E)$ provides the absolute scale of the plasma transmittance. $T_{\text{cold}}(E)$ was determined from the ratio of the transmitted XFEL intensities through the Cu foil and without the sample, where both intensities were obtained by averaging over several thousand XFEL pulses without laser irradiation. This averaging reduces the uncertainty arising from XFEL shot-to-shot fluctuations in the cold-state transmission measurement. For each XFEL shot, the uncertainty associated with the spatial profile fitting of the reference intensity was evaluated, and these uncertainties were propagated when summing multiple shots to obtain the

final plasma transmittance uncertainty. The measured transmittance spectra were smoothed using a Gaussian function with a FWHM of 3 eV. The temporal evolution shown below was then extracted at selected photon energies from the smoothed spectra.

The XFEL pulse duration was 7 fs [28–30]. The pump-probe delay was scanned in steps of approximately 100 fs. The temporal jitter between the optical laser and the XFEL pulse was 29 fs (rms) [25]. Plasma self-emission may be reflected by the downstream mirror, but the corresponding solid angle is very small, so its contribution to the detected signal is negligible. Background measurements without XFEL confirmed that any signal from plasma emission within the detection window is below the noise level. Additional details of the spectrometer settings and data processing are described in the Appendix.

III. EXPERIMENTAL RESULTS AND DISCUSSION

First, we performed XANES measurements around the Cu K-absorption edge (8.98 keV). Figure 2(b) shows a single-shot spatially resolved x-ray spectrum obtained 0.9 ps after laser irradiation. The photon-energy axis was calibrated by scanning the energy of the monochromatized XFEL beam (~ 1 eV) using a double-crystal monochromator with a fixed-exit geometry. The spatial axis was calibrated at the sample position using the knife-edge method. While the K edge is clearly visible in the cold region, a pronounced increase in transmittance on the high-energy side appears in the plasma region. The plasma width is approximately 40 μm (FWHM), slightly larger than the laser focal spot. Figure 3(a) presents the time-resolved transmittance spectra. The absorption edge shifts toward higher photon energy within 0.2 ps, reaching a shift of 20–30 eV at 0.5 ps, and persisting for more than 10 ps. Figure 3(b) shows the transmittance at 9.0 keV (20 eV above the cold K edge), which increases by a factor of ~ 40 within 0.5 ps and remains high for over 10 ps. Because 9.0 keV lies on the steep slope of the K edge, the transmittance at this energy is highly sensitive to the transient evolution of the XANES spectrum. The transient spectra exhibit not only a K-edge shift but also changes in spectral shape, and therefore the transmittance at 9.0 keV is used as a representative quantity to characterize the temporal evolution of the XANES spectra. Plasma self-emission is negligible, as discussed above. These observations indicate rapid ionization of outer-shell electrons followed by a long-lived highly ionized state.

Next, resonant XAS was performed at 8.91 keV (Cu $K\beta$, $1s-3p$) to probe M-shell ionization. After laser irradiation, a clear reduction in transmittance appears in the plasma region [Fig. 2(c)]. The decrease at 8.91 keV in Fig. 3(c) indicates the presence of inner-shell-ionized ions with a $3p$ vacancy. The absorption feature extends up to ~ 8.94 keV, suggesting a distribution of highly charged ions with upward-shifted orbital energies. These two observations suggest the coexistence of inner-shell-ionized ions (with $3p$ vacancies) and highly charged ions in the plasma. As shown in Fig. 3(d), the transmittance at 8.92 keV decreases to approximately one-third of the cold value immediately after irradiation and persists for more than 10 ps. Because the lifetime of a $3p$ vacancy is shorter than 10 fs, this observation indicates that $3p$

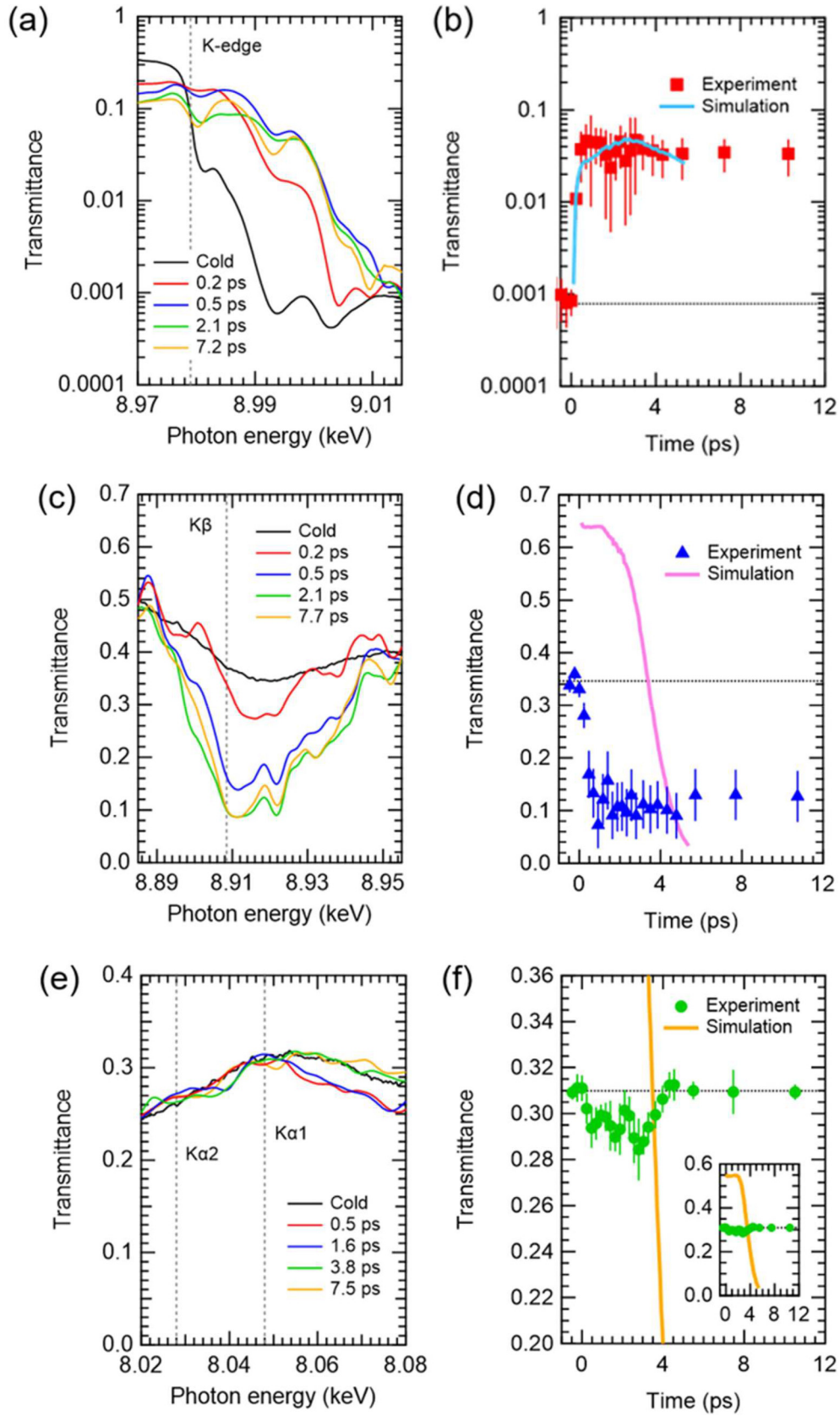


FIG. 3. X-ray transmittance spectra around (a) Cu K edge, (c) K β line, and (e) K α line, obtained by averaging over 15- μ m width for several shots. Time evolution at (b) 9.0 keV, (d) 8.92 keV, and (f) 8.06 keV, averaged over ± 2.5 eV. Dotted lines indicate cold Cu transmittance. Panels (b), (d), and (f) include simulation results from PICLS combined with FLYCHK. The inset in panel (f) shows the measured and simulated transmittance over an extended vertical range.

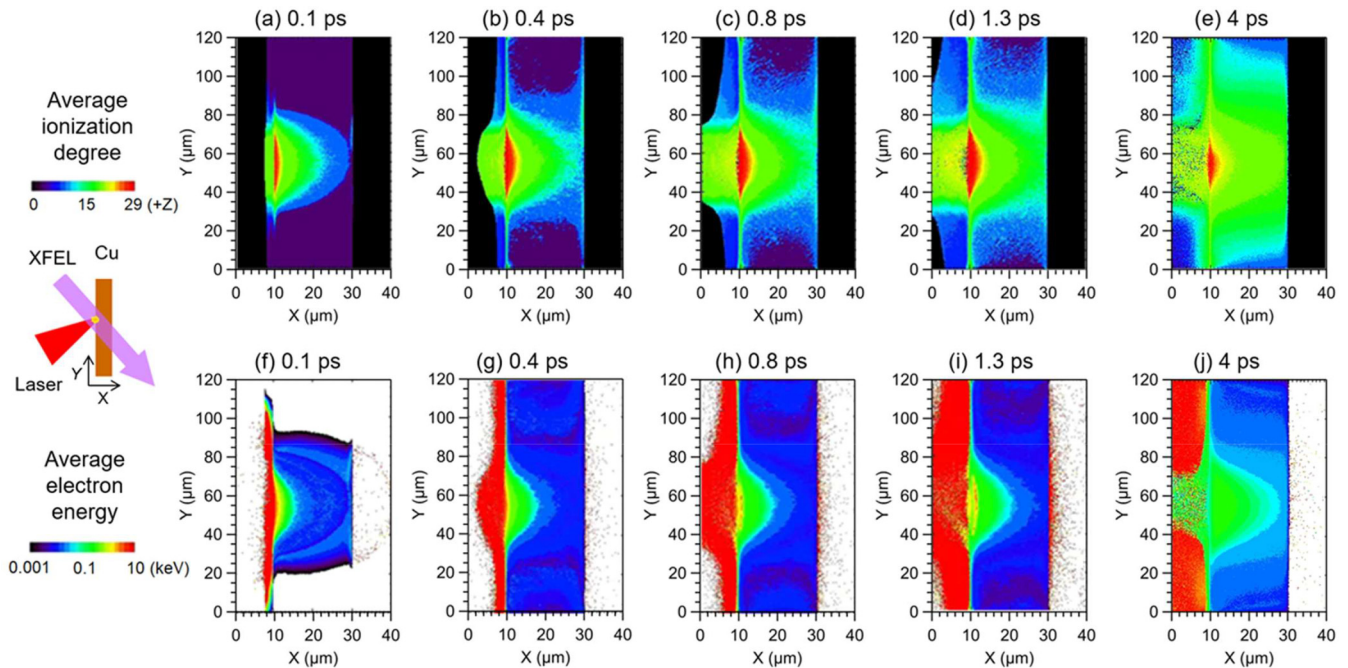


FIG. 4. Calculated average ionization degrees at (a) 0.1 ps, (b) 0.4 ps, (c) 0.8 ps, (d) 1.3 ps, and (e) 4 ps, and average electron energies at (f) 0.1 ps, (g) 0.4 ps, (h) 0.8 ps, (i) 1.3 ps, and (j) 4 ps. Laser, XFEL, and Cu foil configuration is the same as in the experiment. The initial solid-density Cu target is located at 10–30 μm . The high average electron energy regions outside the laser-irradiated front surface of the target are associated with electron expansion from the target.

vacancies are continuously regenerated. This behavior suggests the presence of electrons with energies exceeding the ionization threshold of the $3p$ electrons (M_{III} absorption edge: 74 eV), which continuously regenerate ions with $3p$ vacancies through collisional ionization.

Finally, L-shell ionization was investigated using resonant XAS at 8.05 keV (Cu $K\alpha$, $1s-2p$). Under cold conditions, the lifetime of a $2p$ vacancy is shorter than 2 fs, and the ionization threshold is approximately 930 eV (L_{III} absorption edge). Figure 2(d) shows a slight decrease in transmittance in the plasma region. As seen in Fig. 3(e), a weak reduction appears on the high-energy side of the $K\alpha$ line immediately after irradiation. Although only the tail of the absorption is visible, its position indicates contributions from highly charged ions with shifted $1s-2p$ transition energies. In contrast, no pronounced absorption is observed at the cold $K\alpha$ energy, suggesting that inner-shell-ionized ions with a $2p$ vacancy are not significantly populated along the line of sight. Figure 3(f) shows that the transmittance at 8.06 keV recovers to its initial value within ~ 4 ps. This transient behavior suggests that electrons energetic enough to ionize the $2p$ shell (>930 eV) contribute to the generation of $2p$ vacancies during the early stage of plasma formation, with their contribution decreasing rapidly within several picoseconds.

In summary, the shell-resolved XAS measurements reveal distinct temporal behaviors for outer-, M-, and L-shell ionization. During the first few hundred femtoseconds, multi-shell ionization proceeds rapidly, producing a highly ionized nonequilibrium plasma. Within ~ 4 ps, the contribution of nonthermal electrons capable of ionizing the L shell decreases substantially, whereas electrons above the M-shell threshold remain sufficient to sustain M-shell ionization for more than

10 ps. These observations indicate a progressive relaxation of the electron energy distribution while maintaining sufficient energy to sustain outer- and M-shell ionization.

IV. ANALYSIS WITH PIC SIMULATION

To interpret the measurements, we performed simulations using the particle-in-cell code PICLS [43–45], which incorporates ionization processes and Coulomb collisions. The detailed simulation conditions are described in the Appendix. Figures 4(a)–4(j) show the calculated average ionization degree and average electron energy, illustrating the propagation of ionization and heating from the laser-irradiated surface into the target. For comparison with the experiment, line-integrated electron spectra [Fig. 5(a)] and the temporal evolution of the average electron energy [Fig. 5(b)] were extracted along the XFEL axis. During the laser pulse (0.03 ps), the electron spectrum contains multi-keV components, which rapidly decrease after the pulse, while sub-keV electrons become dominant. The average electron energy increases on the picosecond timescale as the high-energy component diminishes. This trend indicates a reduction of electrons with energies exceeding the L-shell ionization threshold, which is qualitatively consistent with the observed evolution of the $K\alpha$ absorption feature.

The simulation predicts an average ionization degree exceeding +25 near the surface and approximately +17 in the interior [Figs. 4(a)–4(e)]. This spatial distribution helps explain the characteristic transmittance behavior near the $K\alpha$ energy. Because ionization generally proceeds from outer shells inward, the $2p$ shell remains fully occupied up to Cu^{19+} . The first $2p$ vacancy appears in Cu^{20+} , where the electron

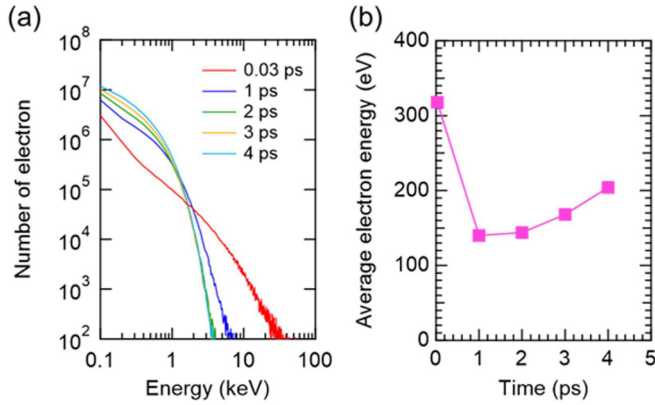


FIG. 5. (a) Time-dependent electron spectra in Cu plasma, obtained by integrating PICLS simulation results along the XFEL axis; 0.03 ps corresponds to the laser pulse duration. (b) Time evolution of average electron energy, calculated along the XFEL axis from PICLS simulations.

configuration becomes $1s^2 2s^2 2p^5$. Thus, in the interior region ($\sim +17$), absorption at the cold $K\alpha$ energy (8.05 keV) is weak. Near the surface ($> +25$), partial depletion of $2p$ electrons shifts the $1s$ – $2p$ transition toward higher photon energies [34]. Line-of-sight integration of these regions yields negligible absorption at the cold $K\alpha$ energy and finite absorption on the higher-energy side. For the M shell, the simulated ionization degree exceeds the threshold for outer-shell $3p$ removal ($+12$), consistent with the strong reduction in transmittance around the $K\beta$ line and its broadening toward higher energies.

Figure 3(b) compares the measured and simulated transmittance near the Cu K-absorption edge, obtained using the PICLS-derived ionization degrees and electron energies combined with opacity data from FLYCHK [46]. The rapid increase in transmittance (>40 -fold) within 0.5 ps and its persistence on the picosecond timescale are well reproduced. While good agreement is achieved near the K edge, the simulated transmittance evolutions around the $K\beta$ and $K\alpha$ energies, shown in Figs. 3(d) and 3(f), respectively, do not quantitatively reproduce the measured absorption changes. These deviations likely arise from limitations in the combined PIC and atomic-physics modeling—such as the treatment of transient inner-shell processes, opacity data, and the evolving electron energy distribution—rather than from experimental uncertainty. These comparisons highlight the need for further development of integrated plasma and atomic-physics modeling. At the same time, the present measurements provide valuable experimental information for further development and validation of plasma and atomic-physics models by offer-

ing simultaneous temporal, spatial, and spectral information on transient absorption in dense plasmas.

V. SUMMARY

In this study, we observed pronounced ultrafast changes in x-ray transmittance in Cu plasma using resonant XAS and XANES, clarifying the time evolution of inner- and outer-shell ionization and electron energy. These measurements reveal distinct relaxation timescales for inner- and outer-shell ionization and electron energy, indicating progressive relaxation toward a more thermalized plasma state. The measurements show good quantitative agreement with plasma simulations for the evolution of transmittance near the K-absorption edge, whereas the simulated transmittance evolutions around the $K\alpha$ and $K\beta$ lines do not reproduce the measured absorption changes. The measurements also provide useful information for improving integrated plasma and atomic-physics models, including the treatment of transient absorption around the $K\alpha$ and $K\beta$ lines. Such advances are essential for achieving accurate radiation-transport simulations and for developing advanced plasma diagnostics and ultrafast x-ray control schemes based on rapid transmittance changes. Furthermore, resonant XAS can be extended to access higher charge states through wider photon-energy scans, while multilayer targets incorporating thin tracer layers could reduce spatial-gradient effects along the XFEL line of sight, enabling more precise spatially resolved measurements. Overall, this work demonstrates the capability of high-resolution XFEL experiments to probe ultrafast ionization dynamics in dense plasma, paving the way for future advances in high-energy-density physics and related applications.

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TABLE I. Parameters of single-shot spectrometer for each experimental condition.

Objectives of measurement	K-absorption edge	$K\beta$ line	$K\alpha$ line
Analyzer crystal	Si(111)	Si(111)	Si(111)
Bragg angle (deg)	12.72	12.82	14.22
Pixel resolution (eV/pixel)	0.47	0.46	0.37
Overall spectral resolution (eV, FWHM)	1.3	1.3	1.1
Observation range (eV)	100	98	79

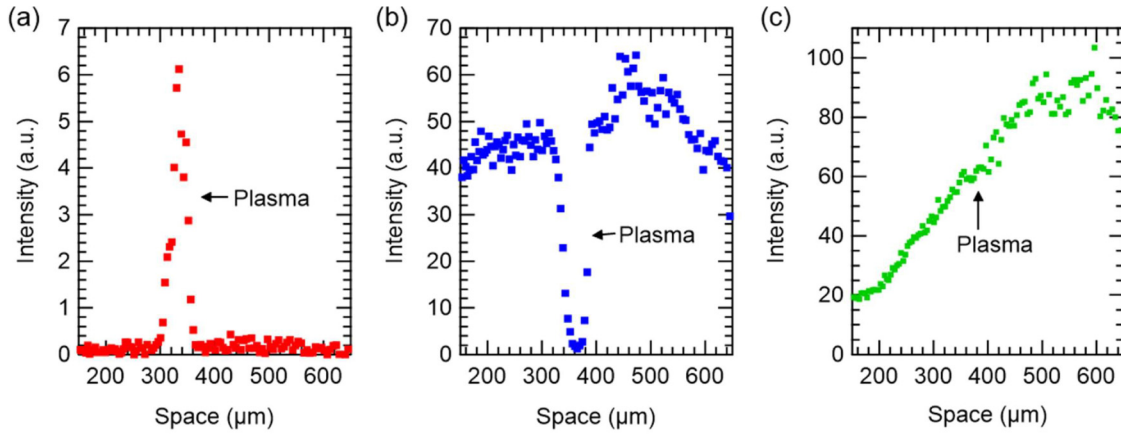


FIG. 6. Representative spatial profiles of the transmitted XFEL intensity at a delay of 0.9 ps. (a) Spatial profile at 9.00 keV near the Cu K-absorption edge. (b) Spatial profile at 8.92 keV near the Cu $K\beta$ line. (c) Spatial profile at 8.06 keV near the Cu $K\alpha$ line. Each spatial profile was obtained by integrating the transmitted spectrum over a 3-eV-wide photon-energy range centered at the indicated photon energy.

DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

APPENDIX

1. Spectrometer conditions for each photon energy

A spatially resolved spectrometer equipped with an MPCCD detector [47] with a pixel size of 50 μm was utilized. The design parameters and spectral resolutions of the single-shot spectrometer for each experimental condition are summarized in Table I. The overall spectral resolution was estimated from the convolution of the analyzer-crystal resolution and the detector pixel resolution. To obtain sufficiently wide spectral coverage around the target absorption edge or emission line, two slightly different XFEL photon energies were used for each experimental configuration. This was necessary because the XFEL pulses had a relatively narrow spectral width of approximately 15 eV (FWHM).

2. Spatial profile of the transmitted x-ray intensity

Figure 6 shows representative spatial profiles of the transmitted XFEL intensity across the laser-irradiated region at a pump-probe delay of 0.9 ps. The profiles were obtained by integrating the transmitted spectra over a 3-eV-wide photon-energy range centered at 9.00 keV near the Cu K-absorption

edge, 8.92 keV near the $K\beta$ line, and 8.06 keV near the $K\alpha$ line, as shown in Figs. 6(a)–6(c), respectively.

3. Particle-in-cell simulation and FLYCHK calculation

The laser-target interaction was self-consistently simulated in 2D-3V using the PIC code PICLS [43], which includes atomic and radiation processes such as Coulomb collisions, field ionization, impact ionization [44], and radiation transport [45,48] of energetic photons emitted via bremsstrahlung [49]. The target was a 20-μm-thick Cu foil with an initial charge state $Z = 2$. A small preplasma is placed in front of the target, characterized by an exponential density profile with a scale length of 0.4 μm over a total thickness of 2 μm. A laser pulse with the same parameters as in the experiment is focused on the target surface with 30° incidence angle. The total number of grids used in the simulation is 2000×6000 , with a square grid $\Delta x(y) = 20$ nm and time step $\Delta t = 0.066$ fs. We use 2 Cu ions and 58 electrons per cell in the region of plasmas. The total number of PIC particles is 200 million and the simulation was run up to 4 ps. We use the atomic code, FLYCHK, to compute the opacities of photons in the XFEL. As shown in Fig. 5(a), the electron energy distributions were thermally equilibrated, and they evolved much slower than the femtosecond timescale of the XFEL. Using the ion density and the local electron temperature evaluated in $0.25 \mu\text{m} \times 0.25 \mu\text{m}$ box, the opacities were computed with FLYCHK, and the x-ray transmittance in the laser-heated Cu target was evaluated.

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