













# Electron spectroscopic study of nanoplasma formation triggered by intense soft x-ray pulses

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## ABSTRACT

Using electron spectroscopy, we investigated the nanoplasma formation process generated in xenon clusters by intense soft x-ray free electron laser (FEL) pulses. We found clear FEL intensity dependence of electron spectra. Multistep ionization and subsequent ionization frustration features are evident for the low FEL-intensity region, and the thermal electron emission emerges at the high FEL intensity. The present FEL intensity dependence of the electron spectra is well addressed by the frustration parameter introduced by Arbeiter and Fennel [New J. Phys. 13, 053022 (2011)].

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## I. INTRODUCTION

The advent of short-wavelength free electron lasers (FELs) has been providing opportunities for a wide range of novel applications such as single-particle diffractive imaging and spectroscopy with femtosecond temporal resolution. Understanding the fundamental processes in intense laser-matter interactions is a key task for further applications of the FELs.

Clusters are systems bridging between atoms/molecules and condensed matter, and present ideal models for understanding the mechanism of laser-matter interactions. Pioneering works on clusters in intense FEL fields have been carried out using several experimental techniques such as ion and electron spectroscopy,<sup>1-7</sup> fluorescence spectroscopy,<sup>8,9</sup> and X-ray diffraction.<sup>10,11</sup> In the short-wavelength region, the dominant excitation process is inner-shell photoabsorption by individual atoms in the cluster. With the

increase of the cluster charge state, outgoing electrons (photoelectrons, Auger electrons, etc.) start to get trapped in the growing Coulombic potential of cluster ions. Further photoabsorptions heat the cluster by the inner-shell ionization of the atoms and eventually lead to the formation of the so-called “nanoplasma,” which is an aggregate of electrons and ions confined in the nanometer scale. After the nanoplasma is formed, it starts to expand, emitting many fragment ions and electrons. During the nanoplasma expansion, the recombination between ions and electrons occurs, which determines the charge and kinetic energy distribution of the emitted particles. In particular, recent studies reported characteristic electronic decay processes which occur during the nanoplasma expansion.<sup>12,13</sup> These nonlocal decay processes are expected to be general and to take place irrespective of the mechanism of the nanoplasma formation. Therefore, understanding these processes is essential for being able to model the response of matter to intense FEL pulses.

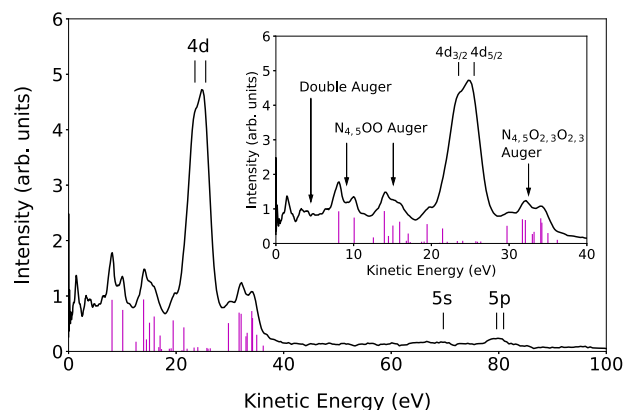
In this study, we have investigated the nanoplasma formation process with an electron spectroscopic method. We irradiated xenon clusters with intense soft x-ray FEL pulses and recorded the electron energy spectra. We systematically studied the FEL intensity dependence of the electron spectra in a wide intensity range and obtained comprehensive information on the electronic processes accompanying nanoplasma formation.

## II. EXPERIMENT

The experiments were performed at SACLA BL1,<sup>14</sup> which delivered linearly polarized soft x-ray pulses at a repetition rate of 60 Hz. The soft x-ray pulses were focused with a Kirkpatrick-Baez mirror system to about 10  $\mu\text{m}$  (FWHM) in diameter.<sup>14</sup> The intensity of the FEL beam was adjusted by the combination of thin metal foil attenuators (Zr 0.5  $\mu\text{m}$ , Zr 0.1  $\mu\text{m}$ , Al 0.2  $\mu\text{m}$ , and Al 0.1  $\mu\text{m}$ ) installed in the beamline. FEL intensities were estimated by using transmittance data.<sup>15</sup> The photon energy was set to 93 eV, corresponding to the giant resonance of Xe atoms. The pulse duration has been evaluated to be less than 100 fs from the single-shot spectra.<sup>16</sup> Intense soft x-ray FEL pulses were focused on a Xe cluster beam, and the kinetic energy distribution of the emitted electrons was measured with a velocity map imaging (VMI) spectrometer.<sup>17</sup> Two sets of light baffles, each consisting of three skimmers, were located at the entrance of the interaction chamber for removing scattered light that was specularly and nonspecularly reflected by the focusing mirrors.

Xe clusters were prepared by the adiabatic expansion of Xe gas through a 200- $\mu\text{m}$  nozzle. The Xe stagnation pressure was 2 bars. Average cluster size  $\langle N \rangle$  is estimated to be about 1000 atoms according to the well-known scaling law.<sup>18</sup> The pulsed cluster beam was cut with knife-edge slits and traveled to the reaction point located 0.8 m downstream from the nozzle.

The emitted electrons were accelerated through a VMI spectrometer in the direction perpendicular to both the FEL beam and the linear polarization axis of the FEL, toward a position sensitive microchannel plate (MCP) detector equipped with a phosphor screen. The positions of the detected electrons were recorded with a gated CCD camera synchronized with the arrival times of the FEL pulses. The three dimensional electron momentum distribution was retrieved from the measured two dimensional projection of



**FIG. 1.** Electron spectrum of xenon atoms recorded at  $F_{\text{peak}} = 6 \text{ nJ}/\mu\text{m}^2$ . Position of photolines (4d, 5s and 5p) and known energies and intensities of Auger electrons<sup>22</sup> are indicated by bars. Inset: electron spectrum of xenon atoms displayed in an enlarged scale.

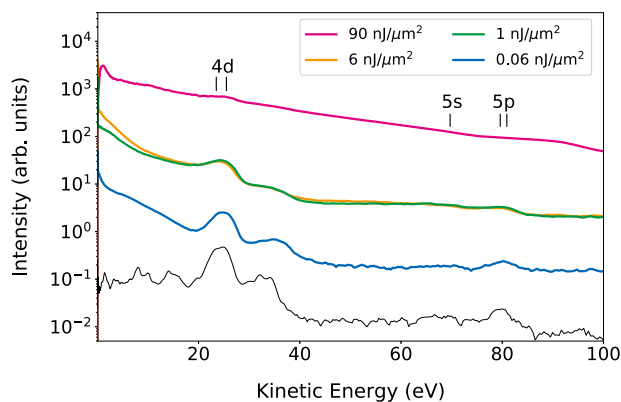
the momentum distribution using a mathematical procedure based on the Abel inversion. We used the pyAbel library<sup>19</sup> for analysis. We used the three-point method<sup>20</sup> and BASEX<sup>21</sup> for the inversion procedure and compared the results to avoid the artifacts from the analysis method. Electron signals were accumulated over more than 120 000 FEL shots and normalized by the number of shots. The background, averaged in the same way, was then subtracted. It was noted that the central part of the electron detection image recorded at high FEL peak fluences (FEL peak fluences  $F_{\text{peak}} = 6 \text{ nJ}/\mu\text{m}^2$  and  $90 \text{ nJ}/\mu\text{m}^2$ ) shows a signature of decrease in the MCP gain due to intense electron signals, i.e., the background subtracted images have an artificial asymmetric structure. We corrected the MCP gain by referring to the gain function of MCP so that the background subtracted images become symmetric. This correction affects only the low-energy part of the electron spectra at high FEL fluences. The present analysis includes the ambiguity at the low-energy part of the electron spectra ( $E_k < 3 \text{ eV}$  for  $F_{\text{peak}} = 6 \text{ nJ}/\mu\text{m}^2$  and  $E_k < 5 \text{ eV}$  for  $F_{\text{peak}} = 90 \text{ nJ}/\mu\text{m}^2$ ).

Figure 1 shows the electron spectrum of xenon atoms recorded at  $F_{\text{peak}} = 6 \text{ nJ}/\mu\text{m}^2$ . Observed peaks in the spectrum are well assigned by the photoelectrons (4d, 5s, and 5p ionization) and known  $\text{N}_{4,5}\text{O}_{2,3}\text{O}_{2,3}$  Auger electrons.<sup>22</sup> We used the spectrum for the calibration of the VMI spectrometer.

## III. RESULTS AND DISCUSSION

Figure 2 shows the electron spectra of the  $\text{Xe}_{1000}$  cluster recorded at four FEL peak fluences  $F_{\text{peak}} = 0.06, 1, 6, 90 \text{ nJ}/\mu\text{m}^2$ . One can see the systematic change in the electron spectra with the increase of the FEL fluence. In the spectra at  $F_{\text{peak}} = 0.06, 1, 6 \text{ nJ}/\mu\text{m}^2$ , one can see distinct peaks of 4d, 5s, and 5p photoelectrons and  $\text{N}_{4,5}\text{O}_{2,3}\text{O}_{2,3}$  Auger electrons, as observed in the atomic spectrum. The electron spectrum at  $F_{\text{peak}} = 90 \text{ nJ}/\mu\text{m}^2$  shows a significant contribution of the Maxwell-Boltzmann (MB) type thermal emission, as well as the small peaks of photoelectrons.

First, we discuss the electron spectrum at the lowest FEL fluence  $F_{\text{peak}} = 0.06 \text{ nJ}/\mu\text{m}^2$ . In the spectrum, significant low-energy

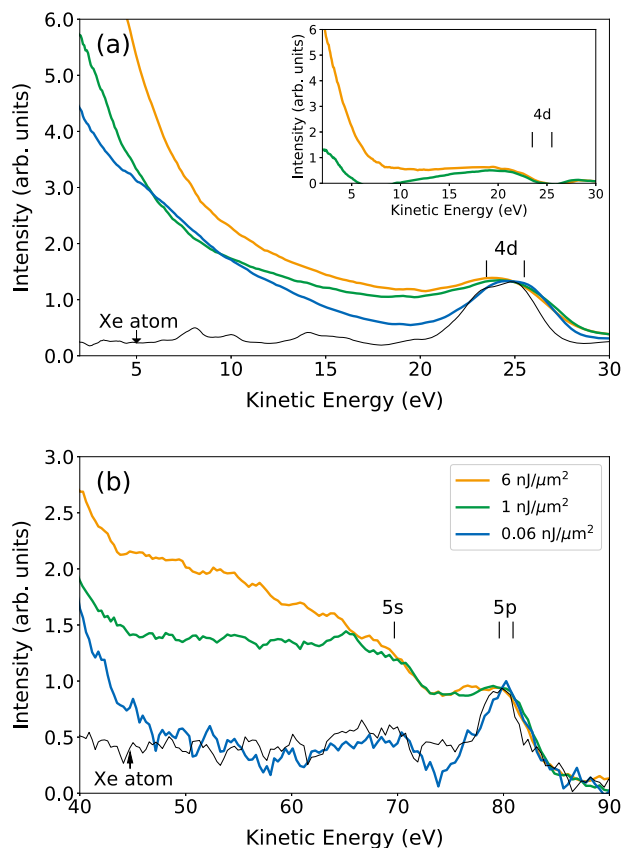


**FIG. 2.** Electron spectra of xenon clusters recorded at four FEL peak fluences ( $F_{\text{peak}} = 0.06 \text{ nJ}/\mu\text{m}^2$ : cyan,  $1 \text{ nJ}/\mu\text{m}^2$ : green,  $6 \text{ nJ}/\mu\text{m}^2$ : orange, and  $90 \text{ nJ}/\mu\text{m}^2$ : red) are plotted in the logarithmic scale. An electron spectrum of the xenon atom is plotted in black for comparison. Note that the electron spectrum of the xenon atom is offset for visibility.

electron tail between  $E_k = 0$  and 20 eV is observed. Considering the radius of the Xe clusters, the low-energy tail can be attributed to the secondary electrons resulting from the inelastic scattering of photoelectrons and Auger electrons in the clusters,<sup>23</sup> as well as from interatomic Coulombic decay (ICD)<sup>24</sup> relaxation of highly excited cationic states. The integrated yield of the observed low-energy tail is about 6 times larger than that of the 4d photoelectron peak, which is roughly consistent with the cross section for inelastic scattering<sup>25</sup> of Auger- and photoelectrons.

Second, we discuss the electron spectra at moderate FEL fluences  $F_{\text{peak}} = 1$  and  $6 \text{ nJ}/\mu\text{m}^2$ . Figures 3(a) and 3(b) display the electron spectra in an enlarged scale, where the spectra are normalized by the height of the 4d and 5p photoelectron peaks, respectively. In Fig. 3(b), we find the emergence of a plateau at the low energy side of the 5p photoline. We also find a similar structure at the low energy side of the 4d photoline in Fig. 3(a). These low-energy tails are the typical signature of the multistep ionization of clusters.<sup>26,27</sup> The energies of the photoelectrons are downshifted for each step of ionization owing to the emerging positive charge of the cluster. To clarify the intensity dependence of the features of the multistep ionization, we subtracted the normalized spectrum of  $F_{\text{peak}} = 0.06 \text{ nJ}/\mu\text{m}^2$  from the other two normalized spectra as shown in the inset of Fig. 3(a). Here, one can clearly see the emergence of the low energy tail at  $F_{\text{peak}} = 1 \text{ nJ}/\mu\text{m}^2$  and the growth of the plateau at  $F_{\text{peak}} = 6 \text{ nJ}/\mu\text{m}^2$ . We also recognize the emergence of additional low-energy electrons ( $E_k < 5 \text{ eV}$ ) and the rapid increase of the yield with the increase of the FEL fluence. The low energy electrons can be interpreted as those that escape under strong frustration of electron emissions caused by the rising charge state of the cluster.

Finally, we focus on the spectrum at the highest fluence,  $90 \text{ nJ}/\mu\text{m}^2$ . At this FEL intensity, one observes clear thermal emission. The electron temperature was estimated to be  $T_e = 30 \text{ eV}$  from a Maxwell-Boltzmann fit to the spectrum. This value roughly agrees with the  $T_e$  reported in the previous work.<sup>28</sup> In addition, the estimated electron temperature of 30 eV is close to the average between

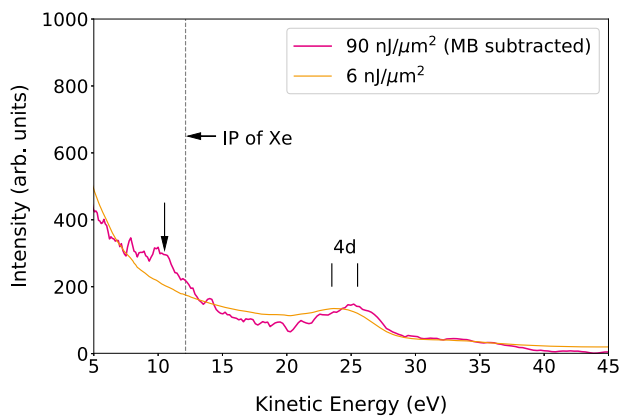


**FIG. 3.** Electron spectra of xenon cluster displayed in an enlarged scale. (a) Electron spectra in the low kinetic energy region ( $<30 \text{ eV}$ ). The electron spectra are normalized by the height of the 4d photoelectron peak at 24 eV. The inset shows the difference spectra (see the text). (b) The electron spectra in the kinetic energy region ( $>40 \text{ eV}$ ). The electron spectra are normalized by the height of the 5p photoelectron peak at 80 eV.

the excess energy of 4d photoelectrons (27 eV) and the kinetic energy of Auger electrons (32 eV). This agreement suggests the ionization heating mechanism where the initial electron temperature of the nanoplasma is determined by the excess energy of the photoelectrons and the subsequently emitted Auger electrons<sup>27</sup> from the inner shells.

Subtracting the component of the thermal emission from the spectrum in Fig. 2 and comparing it with the electron spectrum at  $F_{\text{peak}} = 6 \text{ nJ}/\mu\text{m}^2$  in Fig. 4, we find that the subtracted spectrum surprisingly agrees with the electron spectrum at  $F_{\text{peak}} = 6 \text{ nJ}/\mu\text{m}^2$ . This result is well understood with the assumption that the thermal emission from nanoplasma occurs after the multistep ionization process. The assumption was previously confirmed in the molecular dynamics simulation of nanoplasma formation.<sup>27,28</sup>

We also find a slight broad peak around  $E_k = 10 \text{ eV}$  in Fig. 4 that is not observed in the electron spectra at lower FEL fluences. This peak could be attributed to the correlated electronic decay (CED) of excited atoms/ions created in the expanding nanoplasma.<sup>12</sup> The mechanism of the CED is as follows. Once a nanoplasma is



**FIG. 4.** The electron spectrum recorded at  $F_{\text{peak}} = 90 \text{ nJ}/\mu\text{m}^2$  (red line) from which the thermal electron contribution is subtracted. The thermal electron contribution is estimated by a Maxwell-Boltzmann (MB) fit. The electron spectrum at  $F_{\text{peak}} = 6 \text{ nJ}/\mu\text{m}^2$  is plotted in orange. Note that the spectrum at  $F_{\text{peak}} = 6 \text{ nJ}/\mu\text{m}^2$  is scaled for comparison. The ionization potential of the xenon atom is indicated by the vertical dashed line.

formed, it immediately starts to expand, and the temperature of the nanoplasma decreases quickly. Then, the recombination of electrons and ions takes place in the nanoplasma, which results in the formation of many excited atoms in the nanometer-scale volume. In CED, an electron in a Rydberg state de-excites and transfer its energy to an electron that is bound to a neighboring excited atom<sup>29</sup> or to a quasifree electron in the environment.<sup>12</sup> In addition, a quasifree electron can recombine with an ion, transferring the energy to another quasifree electron or to a bound electron.<sup>30</sup> These decay processes can give rise to additional peaks in the electron spectrum.<sup>13</sup> Here, we estimate the energy of emitted electrons by assuming a decay process between two highly excited Rydberg atoms, where one excited atom goes to the ground state by transferring the excess energy to the neighboring excited atom. It gives the electron energy close to the ionization potential of xenon ( $=12.3 \text{ eV}$ ), which is close to the observed peak position of  $10 \text{ eV}$ . Such phenomena should be general in the decay process of nanoplasma, independent of the excitation photon energy. Similar spectra have been reported in near infrared laser induced nanoplasma,<sup>12</sup> as well as FEL induced nanoplasma.<sup>31</sup>

Finally, we discuss the onset of nanoplasma formation in connection with the evolution of the electron spectra. Here, we employ the frustration parameter  $\alpha$ , introduced by Arbeiter and Fennel,<sup>32</sup> which is defined as the ratio of the total number of Auger- and photoelectrons  $N_{\text{tot}}$  and the critical cluster charge state for full frustration  $q_{\text{full}}$ ,

$$\alpha = \frac{N_{\text{tot}}}{q_{\text{full}}}. \quad (1)$$

When  $\alpha \lesssim 1$ , the direct emission of Auger- and photoelectrons is dominant, which coincide with the multistep ionization regime. When  $\alpha \gg 1$ , strong frustration of emitted electrons leads to the formation of dense nanoplasma. We calculated  $N_{\text{tot}}$  under the

assumption that 4d photoionization is always followed by one Auger electron

$$N_{\text{tot}} = 2 \times \frac{F_{\text{peak}} \sigma N}{h\nu}, \quad (2)$$

where  $\sigma$  is the 4d photoabsorption cross section of Xe.  $q_{\text{full}}$  was calculated by assuming a uniformly charged sphere

$$q_{\text{full}} = \frac{E_{\text{ex}} r_s N^{1/3}}{14.4 \text{ eV} \text{ \AA}^{-1}}, \quad (3)$$

where  $E_{\text{ex}}$  is the highest energy of the Auger- and photoelectrons ( $=33 \text{ eV}$ ) and  $r_s$  is the Wigner-Seitz radius ( $2.56 \text{ \AA}$  for Xe). In the present experiment, the lowest FEL fluence  $F_{\text{peak}} = 0.06 \text{ nJ}/\mu\text{m}^2$  corresponds to  $\alpha = 0.15$ . We note that even though the condition is well below the full frustration, the emission of low-energy electrons takes place due to the inelastic collisions with ions. The electron spectrum recorded at  $F_{\text{peak}} = 1 \text{ nJ}/\mu\text{m}^2$  ( $\alpha = 2.5$ ) shows a plateau at the low energy side of the 5p and 4d photoelectron peaks [Figs. 3(a) and 3(b)], which is consistent with the multistep ionization mechanism. As the FEL fluence increases, the low energy tail in the electron spectra grows ( $F_{\text{peak}} = 6 \text{ nJ}/\mu\text{m}^2$ ,  $\alpha = 15$ ), and evident thermal electrons appears at  $F_{\text{peak}} = 90 \text{ nJ}$  ( $\alpha = 230$ ). Recently, the onset of plasma formation of Xe clusters was observed by fluorescence spectroscopy.<sup>8</sup> The authors concluded that the plasma formation occurs near  $\alpha = 100$  based on the observation of the 4f-4d fluorescence yield, which is in good agreement with the present results of electron spectra.

#### IV. CONCLUSION

In conclusion, we carried out an electron spectroscopic study of rare-gas clusters excited by intense soft x-ray pulses. When the FEL fluence is sufficiently low, the dominant electron emissions are the Auger-/photoelectrons and the secondary low-energy electrons, produced either by the inelastic collisions of the fast electrons or by the ICD of highly excited ions. As the FEL fluence increases, the photoelectron peaks are systematically downshifted, and a plateau emerges at the low energy side of the photoelectron peaks. The dense nanoplasma formation is characterized by the Maxwell-Boltzmann type electron emission and the following decay processes between excited atoms generated by electron-ion recombination. The obtained electron spectra have offered comprehensive information on the electronic processes, ranging from the frustration in the multistep ionization regime to the formation and disintegration of dense nanoplasma.

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