Momentum spectroscopy of fragment ions emitted from Xe clusters irradiated by EUV-FEL at SPring-8

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Abstract. We have studied multiple ionization of Xe clusters by 61-nm 1011–1012 W/cm2 extreme-ultraviolet light pulses at the free-electron laser facility, SPring-8 Compact SASE Source test accelerator, in Japan. Kinetic energy distributions of Xe+, Xe2+, and Xe3+ ions emitted from Xe clusters were measured.

1. Introduction
In October 2007, a new facility, the SPring-8 Compact SASE Source (SCSS) test accelerator [1], has started operation in Japan. It provides linearly polarized EUV-FEL pulses (∼ 30 μJ per pulse, ∼ 100 fs pulse width, 10–20 Hz repetition rate) in the wavelength region 50–62 nm. This energy regime is of particular interest because all atoms in any forms of matter can be ionized by just a single photon with huge photoionization cross sections. One of the most exciting research targets of the FEL irradiation is clusters. A large amount of energy should be deposited into a single cluster in one FEL shot [2, 3, 4, 5].

In the present work, we have investigated multiple ionization of Xe clusters (average cluster sizes ⟨n⟩ ∼ 50 to 200) by 61 nm (photon energy of 20 eV) EUV-FEL pulses from this new light source, using a dedicated dead-time-free multi-particle momentum spectroscopy technique [6].

2. Experiment
2.1. Optical configuration
Figure 1 shows the optical configuration. The cluster beam is in the vertical direction, whereas the FEL beam from the SCSS test accelerator and its polarization axis are in horizontal plane.
The 61-nm FEL beam is steered by two upstream SiC plane mirrors, skimmed by a 5 mm hole 1.2 m upstream the entrance of the experimental chamber, and then introduced to the chamber placed \(\sim 26\) m downstream from the radiation source point. To focus the FEL beam onto the cluster beam of 1 mm in diameter, we use a concave mirror at normal incidence. This mirror was fabricated at Tohoku University, using a tungsten/vanadium coating on a super-polished quartz substrate with a focal length of 250 mm. The FEL beam is partially blocked by a 1.5 mm wide vertical beam stopper (see the inset of Fig. 1) so that the non-focused beam does not irradiate directly the cluster beam.

### 2.2. Ion momentum spectrometer

The design of our ion spectrometer is described in Ref. [7, 8]. A schematic of the spectrometer is shown in Fig. 2. The ion momentum spectrometer uses two acceleration regions with homogeneous electric fields and a drift tube. The source volume of the ions is the intersection of the gas beam and the FEL beam. It is 30 mm away from the extractor electrode and 25.1 mm away from the pusher electrode. The pusher and extractor electrodes are plane aluminum plates with round holes covered by flat stainless steel meshes (70% transmission each). The second acceleration region is 61.6 mm long. After this second acceleration region the extracted ions enter the 268 mm long drift tube. It is terminated by two grids to obtain a field free region inside. With three meshes of 70% transmission each and the detector of 30% detection efficiency, the overall detection efficiency is about 10%. The voltage of the electrodes are usually set to values that fulfill the space focusing condition. The mass resolution (>100) is sufficient to resolve the signals by target from those by residual gases. In this study, the electric fields are chosen to be 20.8 V/mm and 31.53 V/mm for the first and second acceleration regions, respectively.
The determination of the ion momentum is based on the measurements of the time-of-flight (TOF) and the detector hit positions for each ions [9, 10]. Here, a three-layer delay-line anode (Roentdek HEX80) was used for the readout of the microchannel plate (MCP) in order to minimize the dead-time [11].

2.3. Data acquisition system
The design of our data acquisition system is described in Ref. [6]. For typical experimental conditions, many singly charged ions are released in a single FEL shot from a single cluster. Many of them (10% in our case, due to detection efficiency) need to be recorded including their TOF and detector hit positions within a short time window of some 100 ns. Depending on the hit position the time that a pulse needs to leave the delay-line anode is 0–100 ns, therefore a substantial amount of temporal overlap of the signals from different ions occurs. A combination of the redundant information from a three-layer delay-line detector and a sophisticated logic is necessary to reconstruct all hit positions and momenta. Instead of conventional constant-fraction discriminators and time-to-digital converters an 8-channel digitizer (Acqiris DC282 × 2) is used [12, 13]. The trigger signal is derived from the master oscillator of FEL. The complete waveforms of six signals from the three-layer delay-line anode and one from the MCP are recorded by seven channels of the 8-channel digitizer and stored in the computer. The timing signals are extracted offline from the each waveform by software resembling a constant-fraction discriminator [14]. We restrict the analysis to ions whose signal pulses do not overlap on at least two layers. The detected positions and TOF of each ion are obtained from the seven different timing signals and then the 3D momentum was calculated using the position and TOF information for the individual ions. The redundancy of the data set (only 4 out of 7 readout are essential) allows us to perform a virtually dead-time free measurement for the 3D momenta of up to 100 ions produced by a single shot.

2.4. Cluster beam source
The cluster beam is produced by expanding the sample gas with a stagnation pressure from 0.5 to 1 MPa through a nozzle with a pinhole of 30 µm in diameter and 250 µm in thickness. The nozzle can be cooled and temperature stabilized between room temperature and 95 K. The average cluster sizes are estimated by the scaling low published in [15, 16].

3. FEL power effect
Kinetic energy distributions (KEDs) of Xe$^+$ ions emitted from Xe cluster taken at three different FEL power, 1.3, 5.2, and $13 \times 10^{11}$ W/cm$^2$ [4], are shown in Fig. 3 (a). The averaged cluster size was $\langle n \rangle \sim 150$. Our 3D momentum-resolved measurement allows to obtain kinetic energies and angular distributions of fragment ions. For the spectrometer setting described above, we could collect all Xe$^+$ ions emitted into $4\pi$ sr with kinetic energies up to $\sim 20$ eV and their angular distributions were found to be isotropic. Therefore, we restricted the analysis to a 1.5 sr detection cone towards the ion detector so that we could extract the kinetic energies of the Xe$^+$ ions up to 50 eV. Fig. 3 (a) clearly shows that the Xe$^+$ ions get more energy as the power density increases. Such a trend might be expected because an estimate based on the known atomic photoionization cross sections indicates that several photons per FEL shot are absorbed by a cluster, as we will discuss below. Hence, more photons will be absorbed when increasing the incident FEL power density, resulting in a higher charged cluster ion that fragments into atomic ions by Coulomb explosion. Consequently, fragment ions get more energy as the power density increases.

In order to discuss the findings more quantitatively we estimate the number of photons absorbed by a single cluster from the power density of the FEL beam and the photoabsorption
Figure 3. Kinetic energy distributions of $\text{Xe}^+$ ions emitted from Xe clusters, (a) measured for $(n) \sim 150$ at three different FEL power densities. $1.3, 5.2, \text{and } 13 \times 10^{11} \text{ W/cm}^2$; (b) simulated for $\text{Xe}_{150}^{Z+}$ by a classical MD calculation.

cross section of xenon atoms (see, e.g., [3]). The maximum laser power density irradiating the cluster beam was estimated to be $\sim 1.3 \times 10^{12} \text{ W/cm}^2$ with uncertainty of a factor of 2 [4].

Chan et al [17] determined the photoionization cross section for atomic Xe at $\sim 20 \text{ eV}$ as 34 Mb. We obtain values of 0.12, 0.41, and 0.73 for the photoionization probabilities of a single atom by laser pulse of 100 fs width and power densities of $1.3 \times 10^{11}, 5.2 \times 10^{11}, \text{and } 1.3 \times 10^{12} \text{ W/cm}^2$, respectively; close to saturation, the photoionization probability is no longer proportional to the power density. It should be noted that, though single-photon absorption coincides with single-photon ionization for an isolated atom, single-photon absorption of an atom inside the ionic cluster may cause inner ionization in which an electron is not emitted but promoted to the conduction band within the cluster ion. The outer ionization potential increases with growing the charge state of the parent cluster because of the increase in the Coulomb attractive force that the electron feels outside the cluster. Thus, once the outer ionization potential of the cluster ion becomes larger than the photon energy, the charge state of the parent cluster does not coincide with the total number of absorbed photons. This phenomenon is sometimes called frustration of the cluster photoionization [3].

To estimate the average charge state of the parent cluster ion, we have calculated the KEDs of $\text{Xe}^+$ emitted from $\text{Xe}_{150}^{Z+}$ by a classical molecular dynamics (MD) calculation [4]. Figure 3(b) shows some results of this simulation for several different values of $Z$. The experimental KED at $1.3 \times 10^{11} \text{ W/cm}^2$ by the simulation with $Z = 12$. Let us recall that the probability for the photoionization of a Xe atom at $10^{11} \text{ W/cm}^2$ is 0.12. Thus, the cluster $\text{Xe}_{150}$ is expected to absorb $\sim 18$ photons, slightly larger than the estimated charge state $Z = 12$ of the parent cluster ion $\text{Xe}_{150}^{Z+}$. This indicates that the ionization takes place atom by atom and that the cluster structure remains intact during this sequential ionization. We note that, for $\text{Xe}_{150}$ with a radius of 1.7 nm, direct cluster photoionization will be frustrated already at a charge state of $Z \sim 8$. The peak positions of the experimental KEDs at $5.2 \times 10^{11} \text{ W/cm}^2$ and $1.3 \times 10^{12} \text{ W/cm}^2$ suggest that the charge states $Z$ of the parent clusters were 18 and 25, respectively. Apparently these numbers are far smaller than the expected numbers of photons absorbed, i.e., $\sim 62$ and $\sim 110$, respectively. The ratio of the estimated charge $Z$ and number of the absorbed photons, $\sim 0.3$ and $\sim 0.2$, decreases rapidly with increasing FEL power density. This trend may
be understood as due to the frustration of the cluster photoionization described above. Once the direct cluster ionization by a single photon becomes energetically forbidden, inner ionization proceeds further by sequential single-photon absorption by the individual atoms. The energy stored in the cluster by inner-ionization will be electronically relaxed by emitting electrons but the number of emissions is no more equal to the number of photons absorbed.

4. Cluster size effect

![Figure 4. Kinetic energy distributions of Xe$^+$, Xe$_2^+$, Xe$_3^+$, and Xe$_4^+$ emitted from Xe clusters measured for $\langle n \rangle \sim 50$ and $200$ at $5.2 \times 10^{11}$ W/cm$^2$. Note that the full scales of the kinetic energy for Xe$_2^+$, Xe$_3^+$, and Xe$_4^+$ are 1/2, 1/3, and 1/4 of that of Xe$^+$, respectively. In these plots, the distributions of different ions would appear at the same positions if the momentum distributions were the same for these ions.](image)

The number of photons absorbed by a single cluster is also expected to increase with the cluster size $n$. Figure 4 shows the measured KEDs of the Xe$^+$ ions for different cluster sizes, $\langle n \rangle \sim 50$ and $200$, at a FEL power density of $5.2 \times 10^{11}$ W/cm$^2$ [4]. As expected it shows that the average kinetic energy of the Xe$^+$ ions increases with the cluster size, i.e. with the number of absorbed photons.

5. Other fragments

Besides the Xe$^+$ ions, we found Xe$^{2+}$, Xe$^{3+}$, Xe$^{4+}$, and larger fragment ions. Xe$^{2+}$ ions have very low energy and are thus attributed to double ionization of monomers in the cluster beam. We could not record enough fragment ions larger than Xe$^{3+}$ to extract KEDs. KEDs of the Xe$^{2+}$, Xe$^{3+}$, and Xe$^{4+}$ fragment ions at the same experimental conditions as Fig. 3 (a) were extracted and shown in Fig. 5. For these ions, KEDs are depicted using ions emitted into $4\pi$ sr because kinetic energies of these ions are small. Assuming that all ions receive the same momentum at average suggests that the kinetic energies of Xe$^{2+}$, Xe$^{3+}$, and Xe$^{4+}$ should be half, 1/3, and 1/4 of that of Xe$^+$, respectively. Compared with KEDs of Xe$^+$ at the same experimental conditions, however, we notice that the average kinetic energies of the Xe$^{2+}$, Xe$^{3+}$, and Xe$^{4+}$ ions are even
Figure 5. Kinetic energy distributions of Xe$^+$, Xe$^+_2$, Xe$^+_3$, and Xe$^+_4$ emitted from Xe clusters measured for $\langle n \rangle \sim 150$ at 1.3, 5.2, and 13 $\times 10^{11}$ W/cm$^2$. Note that the full scales of the kinetic energy for Xe$^+_2$, Xe$^+_3$, and Xe$^+_4$ are 1/2, 1/3, and 1/4 of that of Xe$^+$ in Fig. 3, respectively. In these plots, the distributions of different ions would appear at the same positions if the momentum distributions were the same for these ions.

lower. Interestingly, we notice that the yield of Xe$^+_2$, Xe$^+_3$, and Xe$^+_4$ does not increase as much with an increase in the power density as that of the Xe$^+$ ions and KEDs for Xe$^+_2$, Xe$^+_3$, and Xe$^+_4$ ions have no clear FEL power dependence as that of the Xe$^+$ ions. The KEDs of Xe$^+_2$, Xe$^+_3$, and Xe$^+_4$ emitted from Xe clusters measured for $\langle n \rangle \sim 50$ and 200 at a FEL power density of 5.2 $\times 10^{11}$ W/cm$^2$ are also shown in Fig. 4. Again, the average kinetic energies of Xe$^+_2$, Xe$^+_3$, and Xe$^+_4$ are lower than half, 1/3, and 1/4 of that of Xe$^+$. In contrast with FEL power dependence shown in Fig. 5, the KEDs of these ions have similar trend with that of Xe$^+$, i.e., KEDs of Xe$^+_2$, Xe$^+_3$, and Xe$^+_4$ increase with $\langle n \rangle$. The slow-down effects for the heavier fragment ions than Xe$^+$ may be understood by dynamical considerations. The Xe$^+$ leave the cluster quicker and, therefore, are repelled by the full charge inside the sphere behind them, while the slower Xe$^+_2$, Xe$^+_3$, and Xe$^+_4$ ions experience repulsion from the further out-going Xe$^+$ and are decelerated.

6. Summary
KEDs of fragment ions emitted from Xe clusters irradiated by EUV-FEL have been measured. KEDs of Xe$^+$ increases when increasing the laser power and the cluster size. KEDs of larger fragment, Xe$^+_2$, Xe$^+_3$, and Xe$^+_4$, increase with cluster size, but have no dependence on laser power.

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