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Dead-time-free ion momentum spectroscopy of multiple ionization of Xe clusters irradiated by euv free-electron laser pulses

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We have investigated multiple ionization of Xe clusters by 61-nm $10^{11}-10^{12}$ W/cm^2 extreme-ultraviolet light pulses at the free-electron laser facility, SPring-8 Compact SASE Source test accelerator, in Japan, using a dead-time-free three-dimensional momentum spectrometer. It was found that the average kinetic energy of atomic Xe^+ ions increases when increasing the laser power density and the cluster size. For these experimental conditions significant frustration of the cluster photoionization occurs but no indication for heating mechanisms other than sequential photoabsorption by individual atoms in the cluster was found.

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Investigations of the interaction of intense short-wavelength laser pulses with matter are currently one of the most exciting topics in fundamental research [1]. Free-electron lasers (FELs) based on self-amplified spontaneous-emission (SASE) in the extreme-ultraviolet (euv) region below $\lambda = 100$ nm have proven to be a very powerful tool to explore the interaction of strong euv laser pulses with atoms [2–5], molecules [6–8], and clusters [9–12]. One of the most striking findings was the large amount of energy deposited into a single cluster in one FEL shot under certain conditions. In their pioneering work using the TESLA test facility (TTF) at Deutsches Elektronen-Synchrotron (DESY) Wabnitz *et al.* [9] investigated multiple ionization of Xe clusters by 98-nm FEL radiation and found complete Coulomb explosion and ions with charge states up to 8+. These unexpected results triggered many theoretical investigations worldwide [13–17], indicating that various plasma heating processes are the important energy deposition mechanisms. Because of the lack of other light sources, all experimental studies of clusters with intense euv light pulses so far have been performed at the TTF and its upgraded facility free-electron laser in Hamburg (FLASH) at DESY [9–12].

Very recently, a facility, the SPring-8 Compact SASE Source (SCSS) test accelerator, has started operation in Japan [18]. It provides linearly polarized euv-FEL pulses (~30 μJ per pulse, ~100 fs pulse width, and 10–20 Hz repetition rate) in the wavelength region of 51–61 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, much larger than those at shorter wavelengths currently available at FLASH.

In the present work, we have investigated multiple ionization of Xe clusters (average cluster sizes $\langle n \rangle \sim 50-200$) by 61-nm (photon energy of 20 eV) euv-FEL pulses from this light source, using a dedicated dead-time-free multiparticle momentum spectroscopy technique. This technique, developed as one of the key techniques for a single-shot analysis of nanoclusters and biomolecules by x-ray FELs (X-FELs) available in near future, allows us to determine three-dimensional (3D) momentum of more than 100 charged particles generated by a single shot of FEL pulses. One of the key questions that may be answered with this technique is the determination of the threshold for plasma heating processes by the euv-FEL currently available. Using power densities of $1.3-13 \times 10^{11}$ W/cm^2 we have not found any indication for heating mechanisms other than sequential photoabsorption by individual atoms in the cluster [12] followed by Coulomb explosion.

Figure 1 shows the experimental configuration. The cluster beam was in the vertical direction, whereas the FEL beam from the SCSS test accelerator and its polarization axis were in horizontal plane. The 61-nm FEL beam was 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 ~26 m downstream from the radiation source point. To focus the FEL beam onto the cluster beam of 1 mm in diameter, we used a concave mirror at normal incidence. This mirror was fabricated at Tohoku University, using a tungsten-vanadium coating on a superpolished quartz substrate with a focal length of 250 mm. The FEL beam was partially blocked by a 1.5-mm-wide vertical beam stopper (see the inset of Fig. 1), so that the nonfocused beam did not irradiate directly the cluster beam.

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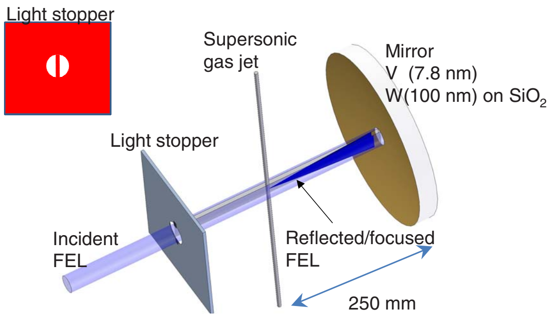

FIG. 1. (Color online) Experimental configuration.

The Xe clusters were produced by expanding Xe gas at stagnation pressures of 0.5, 0.9, and 1 MPa at room temperature through a nozzle with a pinhole of 30 μm diameter and 0.25 mm thickness. The corresponding average cluster sizes were estimated to be $\langle n \rangle \sim 50$, 150, and 200, respectively, by the scaling law published in [19,20].

Using electrostatic fields, ions were projected onto a 80-mm-diameter microchannel plate (MCP) in front of a delay-line anode. The determination of the ion momentum is based on the measurements of the time of flight (TOF) and the detector hit position for each ion (see, e.g., [21]). Here, a three-layer-type delay-line anode (Roentdek HEX80) was used to minimize the dead time. The design of the ion spectrometer with a two-stage acceleration section is very similar to the one described in [22]. The lengths and fields are chosen to be 30 mm, 20.8 V/mm and 61.5 mm, 31.53 V/mm, respectively. The acceleration is followed by a 268-mm-long field-free drift tube with the ion detector mounted at its exit.

In the current experiment, a large number of Xe^+ were released in a single FEL shot from a single cluster. Many of them hit the detector within some 100 ns and have to be recorded including their individual momenta in order to extract information about the energy deposition mechanism. Depending on the hit position of individual ions on the detector, the time that a signal 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 obtaining redundant information from a three-layer delay-line anode and a sophisticated logic is necessary to reconstruct time and positions of all "hits." Instead of conventional constant fraction discriminators and time-to-digital converters, an eight-channel digitizer (Acqiris DC282 $\times 2$) was used. The complete wave forms of six signals from the three-layer delay-line anode and one from the MCP were recorded by seven channels of the eight-channel digitizer and stored in the computer. The timing signals were extracted off line from each wave form by a software resembling a constant fraction discriminator. This software can potentially be modified to deal even with overlapping pulses. Here, we restrict the analysis to ions whose signal pulses do not completely overlap on at least two layers. The detected positions and TOF of each ion were 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 four out of seven readouts are necessary) allowed us to perform a virtually dead-time-free measurement for the 3D momenta of over 100 ions produced by a single FEL pulse.

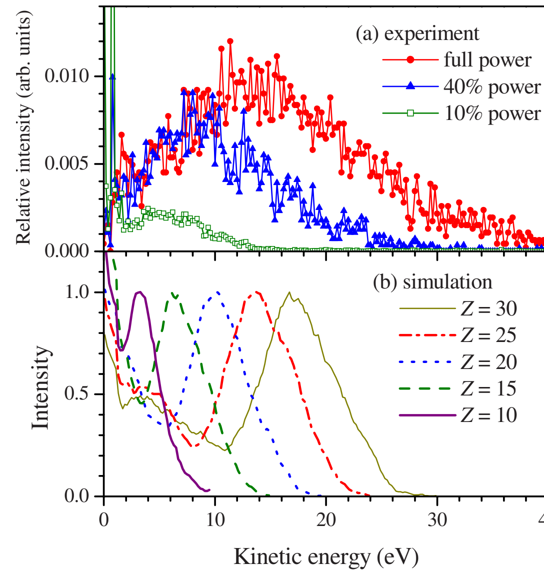

FIG. 2. (Color online) Kinetic-energy distributions of Xe^+ ions emitted from Xe clusters, (a) measured for $\langle n \rangle \sim 150$ at three different FEL power densities of 1.3, 5.2, and 13×10^{11} W/cm^2 and (b) simulated for Xe_{150}^{Z+} by a classical molecular-dynamics (MD) calculation.

To investigate the FEL power density dependence, the measurements were performed by reducing the power to 40% and 10% from the full power by a variable horizontal slit upstream the beam stopper. The relative intensities were estimated from ion yields of residual gas impurities ionized by the unfocused direct FEL beam, because these yields were proportional to the FEL power. The relative uncertainties were less than a factor of 2.

In the TOF ion mass spectra, we found Xe^{2+}, Xe^+, Xe_2^+, and Xe_3^+ ions. Xe^{2+} ions have very low energy and are thus attributed to double ionization of monomers in the cluster beam. Here, we present the results for kinetic-energy distributions (KEDs) of only Xe^+. Figure 2(a) shows KEDs of Xe^+ thus taken at three different FEL powers. The averaged cluster size was $\langle n \rangle \sim 150$. Our 3D momentum-resolved measurement allows us to obtain kinetic energies and angular distributions of fragment ions. For the spectrometer setting described above, we could collect all Xe^+ ions emitted into 4π sr with kinetic energies up to ~ 20 eV and their angular distributions were found to be isotropic. Therefore, we restricted the analysis to a 1.5 sr detection cone toward the ion detector, so that we could extract the kinetic energies of the Xe^+ ions up to 50 eV. Figure 2(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.

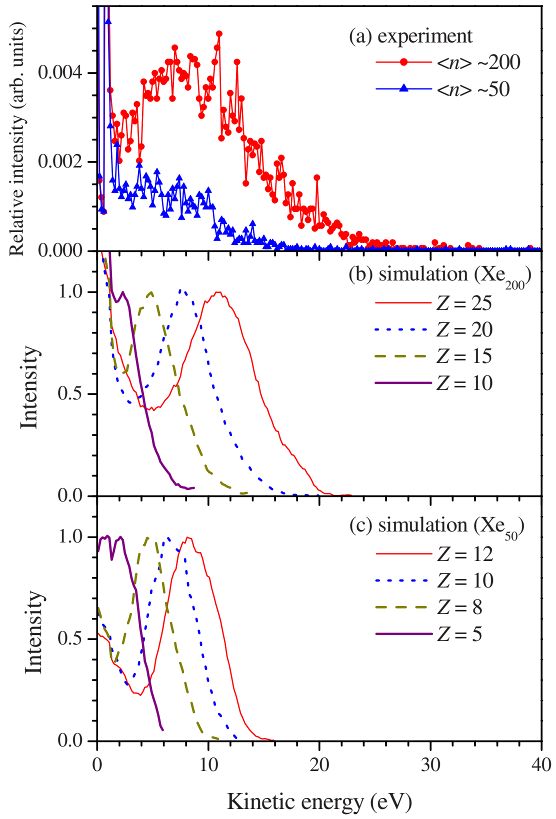

FIG. 3. (Color online) Kinetic-energy distributions of Xe^{+} from Xe clusters, (a) measured for $\langle n\rangle \sim 50$ and ~ 200 at 5.2×10^{11} W/cm^2, (b) simulated for Xe_{200}^{Z+}, and (c) simulated for Xe_{50}^{Z+}.

The number of photons absorbed by a single cluster is also expected to increase with the cluster size n. Figure 3(a) shows the measured KEDs of the Xe^{+} ions for $\langle n\rangle \sim 50$ and 200 at a FEL power density of 40% of the full power. 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.

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 cross section of xenon atoms (see, e.g., [12]). The maximum laser power density irradiating the cluster beam was estimated to be $\sim 1.3\times 10^{12}$ W/cm^2 from the pulse energy of $\sim 29\pm 9$ μJ, pulse duration of ~ 100 fs, and taking account of all optical components and beam skimmers and the effective focus size ($\sim 15\pm 5$ μm in diameter, including the focal volume effect). The absolute pulse energy was measured in a separate experiment using a calibrated pin diode [18]. The pulse duration was estimated from the Fourier transformation of a single-shot spectrum and the resulting value of ~ 100 fs was consistent with the measured pulse duration of ~ 150 fs of the electron bunch [18], which is known to be longer than the light pulse duration. Note that the pulse duration itself will not affect to the discussion below; only the total energy plays a role. All in all the estimates of the laser power densities are uncertain by a factor of 2.

Chan *et al.* [23] determined the photoionization cross section for atomic Xe at ~ 20 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 1.3×10^{11}, 5.2×10^{11}, and 1.3×10^{12} W/cm^2, respectively; close to saturation, the photoionization probability is no longer proportional to the power density. It should be noted that, although 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 [12].

To estimate the average charge state of the parent cluster ion, we have calculated the KEDs of Xe^{+} emitted from Xe_{50}^{Z+}, Xe_{150}^{Z+}, and Xe_{200}^{Z+} by a classical molecular-dynamics (MD) calculation. For Xe_{50}^{Z+}, we first assumed a complete icosahedron structure for the parent cluster Xe_{55} and removed five atoms randomly from the outermost shell. For Xe_{150}^{Z+} and Xe_{200}^{Z+}, we added atoms randomly to the next shell on the complete icosahedron structure Xe_{147}. We distributed the total charge Z by randomly selecting Z atoms within the cluster. Then we assume a Coulomb potential and a Lennard-Jones potential in order to describe the forces between atoms and ions and modeled their motion using classical mechanics. We did not take account of the cluster expansion during the FEL pulse of 100 fs.

Figure 2(b) shows some results of this simulation for Xe_{150}^{Z+} for several different values of Z. The experimental KED at 1.3×10^{11} W/cm^2 is best reproduced by the simulation with $Z=12$. Let us recall that the probability for the photoionization of a Xe atom at 1.3×10^{11} W/cm^2 is 0.12. Thus, the cluster Xe_{150} is expected to absorb ~ 18 photons, slightly larger than the estimated charge state $Z=12$ of the parent cluster ion 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 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 5.2×10^{11} and 1.3×10^{12} 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., ~ 62 and ~ 110, respectively. The ratio of the estimated charge Z and number of the absorbed photons, ~ 0.3 and ~ 0.2, decreases rapidly with increasing FEL power density or with increasing the charge state Z. 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 longer equal to the number of photons absorbed. We do not see additional plasma heating by which the cluster could absorb more energy than expected by sequential single-photon absorption by the individual atoms and in consequence would emit highly charged atomic ions.

The simulated KEDs are also displayed in Figs. 3(b) and 3(c). The comparison between measured and simulated KEDs indicates that the charge states are ~8 and ~20 for $\langle n \rangle \sim 50$ and ~200, respectively. The expected numbers of photons are 21 and 81 for $\langle n \rangle \sim 50$ and ~200, respectively, and thus the ratios of the estimated charge Z and number of the absorbed photons, 0.4 and 0.2, are significantly smaller than 1, again indicating frustration of the cluster photoionization.

In conclusion, using a dead-time-free ion momentum spectroscopy, we found that the kinetic energy of Xe^{+} emitted from the Xe cluster irradiated by the euv-FEL increases with increasing laser power density and cluster size. For the power densities considered here multiple ionizations of the cluster can essentially be understood as sequential photoabsorption by individual atoms in the cluster. Frustration of the cluster ionization was found but no plasma heating was noticed. It is worth noting that the dead-time-free detection technique presented here will be able to provide the compensative information about the orientation of nanoclusters and biomolecules as a target of the single-shot x-ray diffraction imaging by the X-FELs available in near future.

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