Nanoplasma formation in rare-gas clusters ignited by intense X-ray free-electron laser pulses from SACLA

In early 2012, a new X-ray free-electron laser (XFEL) facility, the SPring-8 Angstrom Compact free electron LAser (SACLA) [1], started user operation. As one of the first experiments at SACLA, we performed an experiment to observe multiphoton multiple ionization of heavy Xe atoms [2]. We found that the ionization of heavy atoms irradiated by intense XFEL pulses rapidly progresses by repeated inner-shell photoionization and subsequent Auger/Coster-Kronig decay within the XFEL pulse duration. What will happen to heavy atomic clusters exposed to intense XFEL pulses? It is known that a nanoplasma is formed when clusters are irradiated by intense lasers with longer wavelengths. Rare-gas clusters exposed to an intense near infrared (NIR) laser field are probably the best-studied system. In the NIR laser field, field ionization takes place. A nanoplasma is formed when the electrons escaping from their mother atoms still remain within the Coulomb potential of the multiply charged cluster ion (i.e., inner ionization). The trapped electrons further acquire energy from the laser field via the inverse bremsstrahlung (IBS). When the electrons acquire sufficient energy, they escape from the cluster (i.e., outer ionization). When the laser field is turned off and the nanoplasma expands, electrons trapped in the nanoplasma are mostly recombined with the individual atomic ions. Rare-gas clusters exposed to intense extreme ultraviolet (EUV) laser pulses have also been studied extensively since the advent of EUVFELs. In the EUV regime, the main mechanism of the cluster ionization and heating is single-photon absorption of the individual atoms in the cluster. The outer ionization proceeds first if the photon energy is higher than the ionization potential of the individual atoms. With the increase in the charge of the cluster, the photoelectrons are decelerated and eventually trapped, although inner ionization still proceeds. As the number of trapped electrons increases, a nanoplasma is eventually formed. The following question now arises: is nanoplasma also formed by XFEL irradiation? We have investigated this question by electron spectroscopy using Ar and Xe clusters as target species [3,4].

The experiment was carried out at experimental hutch 3 (EH3) of beamline **BL3** of SACLA [5]. The XFEL beam is focused by a Kirkpatrick-Baez mirror system to a focal size of ~1 μm (FWHM) in diameter. Rare-gas (Ar or Xe) clusters were prepared by adiabatic expansion of the rare gases through a 25 μm nozzle at room temperature. The cluster beam was skimmed by two skimmers placed 20 and 400 mm from the nozzle. The inner diameters of the first and second skimmers were 0.5 and 2 mm, respectively. The distance between the second skimmer and the reaction point was 250 mm. The cluster beam at the reaction point was estimated to be ~2 mm (FWHM) in diameter. Thus, the source volume of the electrons was roughly cylindrical, ~1 μm in diameter and ~2 mm along the XFEL beam. The XFEL pulse energies were measured by a beam-position monitor located upstream of the beamline. The relative X-ray pulse energy passing through the interaction point was also measured shot-by-shot by a PIN photodiode.

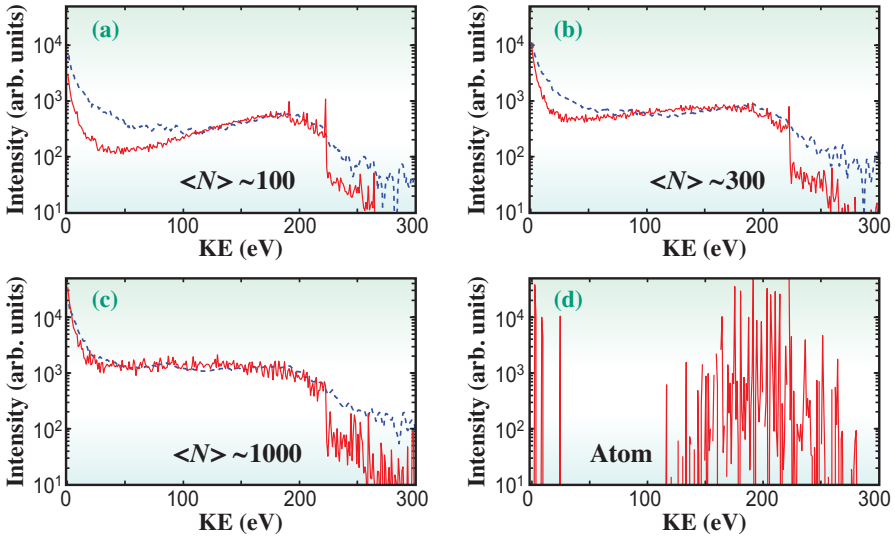

Fig. 1. Electron spectra of Ar clusters at a photon energy of 5 keV [3]. The peak fluence was ~50 μJ/μm^2. Dashed lines are the experimental results and solid lines are corresponding theoretical results.

Electrons produced in the source volume were extracted towards a velocity map imaging (VMI) spectrometer equipped with microchannel plates (MCPs) and a phosphor screen. Electron detection images on the phosphor screen were recorded using a CCD camera synchronized with the arrival of the FEL pulse in the interaction chamber. The measured two-dimensional projection allows us to obtain three-dimensional momentum distributions of the ejected electrons using the inverse Abel transformation.

Figure 1 depicts the electron spectra of Ar clusters with different average cluster sizes <*N*> at a photon energy of 5 keV [3]. The peak fluence of the XFEL pulse determined via the calibration using Ar atoms [2] was ~50 μJ/μm^2. In the spectrum for <*N*> ~100, we can identify a broad peak around 150–250 eV due to the LMM Auger electrons emitted after the KLL Auger process. The intensity of electron emission below 150 eV increases when the cluster size increases. This trend is interpreted as follows with the help of theoretical calculations that reproduce the experimental spectra as shown in Fig. 1 [3]. When the cluster is exposed to intense XFEL pulses, atoms in the cluster are ionized by photoionization and subsequent Auger decay(s). The charge state of the cluster increases when the number of ionized atoms increases. As a result, the LMM Auger electrons are decelerated by the positive potential from the highly charged cluster and form a plateau in the electron spectra. The decelerated electrons are eventually trapped by the cluster ions. The LLM Auger electrons and the secondary electrons produced by the low-energy electron impact are also trapped. As a result, we expect that a nanoplasma is formed. The peak at zero kinetic energy is due to thermal emission and is thus evidence of nanoplasma formation.

Figure 2(a) depicts the electron spectra of Xe clusters with different <*N*> exposed to XFEL pulses at a photon energy of 5.5 keV. The peak fluence was ~50 μJ/μm^2. In the spectrum for <*N*> ~80, there are peaks due to L_2 and L_3 photoemissions and MNN Auger emission at ~400 eV, ~700 eV, and ~500 eV, respectively. The valleys between the peaks become plateaus when <*N*> increases. This trend is also interpreted as the deceleration of the photoelectrons and Auger electrons by the Coulomb potentials from the charged cluster. A peak due to thermal emission from the nanoplasma at zero kinetic energy is also observed. Figure 2(b) depicts the peak fluence dependence of the electron spectrum of the Xe cluster with <*N*> ~10000. It is worth noting that the strong peak at zero kinetic energy is observed even at the peak fluence of the XFEL pulse of ~2.5 μJ/μm^2. In this case, only ~5% of the Xe atoms in the cluster are photoionized. This indicates that the nanoplasma is efficiently produced for the Xe clusters by Auger cascades and secondary ionization events.

In conclusion, we have investigated phenomena in rare-gas clusters exposed to intense XFEL pulses by electron spectroscopy and found that a nanoplasma is formed by trapping low-energy electrons. Note that nanoplasma formation can always occur whenever matter is exposed to intense XFEL pulses.

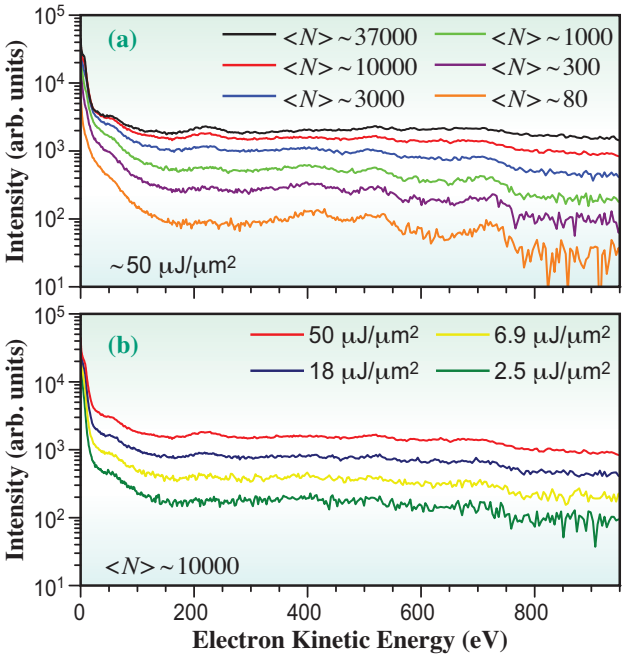

Fig. 2. Electron spectra of Xe clusters at a photon energy of 5.5 keV [4]. **(a)** Spectra for different <*N*> at peak fluence of ~50 μJ/μm^2. **(b)** Spectra for different peak fluences at <*N*> of ~10000.

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