Multiphoton Multiple Ionization of Rare-Gas Atoms and Clusters by X-Ray Free-Electron Laser Pulses from SACLA

H. Fukuzawa[1,2], K. Motomura[1], T. Tachibana[1], Z. Jurek[3], S.-K. Son[3], P. Johnsson[4], K. Nagaya[2,5], M. Siano[6], S. Wada[2,7], S. Mondal[1], M. Kimura[1], Y. Ito[1], T. Sakai[5], K. Matsunami[5], H. Hayashita[7], J. Kajikawa[7], X.-J. Liu[8], E. Robert[8], C. Miron[8], R. Feifel[9], E. Kukk[10], B. Rudek[11,12], B. Erk[11,12], L. Foucar[11,13], J. Marangos[6], K. Tono[14], Y. Inubushi[2], T. Hatsui[2], M. Yabashi[2], B. Ziaja[3,15], R. Santra[3,16], M. Yao[5], and K. Ueda[1,2]

[1] *Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, Sendai 980-8577, Japan*
[2] *RIKEN SPring-8 Center, Sayo, Hyogo 679-5148, Japan*
[3] *Center for Free-Electron Laser Science (CFEL), DESY, 22607 Hamburg, Germany*
[4] *Department of Physics, Lund University, P.O. Box 118, 22100 Lund, Sweden*
[5] *Department of Physics, Kyoto University, Kyoto 606-8502, Japan*
[6] *Blackett Laboratory, Imperial College London, London SW7 2AZ, United Kingdom*
[7] *Department of Physical Science, Hiroshima University, Higashi-Hiroshima 739-8526, Japan*
[8] *Synchrotron SOLEIL, L'Orme des Merisiers, Saint-Aubin, BP 48, FR-91192 Gif-sur-Yvette Cedex, France*
[9] *Department of Physics and Astronomy, Uppsala University, P.O. Box 516, SE-751 20 Uppsala, Sweden*
[10] *Department of Physics and Astronomy, University of Turku, 20014, Finland*
[11] *Max-Planck Advanced Study Group at CFEL, 22607 Hamburg, Germany*
[12] *Max-Planck-Insitut für Kernphysik, 69117 Heidelberg, Germany*
[13] *Max-Planck-Insitut für medizinische Forschung, 69120 Heidelberg, Germany*
[14] *Japan Synchrotron Radiation Research Institute (JASRI), Sayo, Hyogo 679-5198, Japan*
[15] *Institute of Nuclear Physics, PAS, Radzikowskiego 152, 31-342, Kraków, Poland*
[16] *Department of Physics, University of Hamburg, 20355 Hamburg, Germany*
fukuzawa@tagen.tohoku.ac.jp

Abstract: We have investigated multiphoton multiple ionization dynamics of rare-gas atoms and clusters by 5 keV and 5.5 keV x-ray free-electron laser (XFEL) pulses provided by a new XFEL facility, SACLA in Japan.

OCIS codes: (020.4180) Multiphoton processes; (300.6350) Spectroscopy, ionization

1. Introduction

X-ray free-electron lasers (XFELs) provide extremely intense femtosecond x-ray pulses that promise access to high-resolution structure determination, while outrunning any radiation damage. Achievement of this goal, however, requires the ability to describe the dynamics of samples containing heavy atoms in high-intensity x-ray pulses. In early 2012, a new XFEL facility, the SPring-8 Angstrom Compact free electron LAser (SACLA) started user operation in Japan [1]. As one of the first experiments at SACLA, we have studied the ionization dynamics of heavy rare gas atoms [2,3] and rare gas atomic clusters. Understanding of ionization dynamics of these systems exposed to the intense XFEL pulse provides useful input for future dynamic imaging using XFELs.

2. Experiment

The experiment has been carried out at the experimental hutch 3 (EH3) of the beamline 3 (BL3) of SACLA [4]. The XFEL beam is focused by the Kirkpatrick-Baez (KB) mirror system to a focal size of ~1 μm (FWHM) in diameter. The sample gas was introduced as a pulsed supersonic jet to the focal point of the XFEL beam. After crossing the gas jet at right angle, the XFEL beam exits the vacuum chamber via beryllium window. The relative XFEL pulse energy was measured shot-by-shot by a PIN photodiode.

In the experiment with atoms, we used an ion time-of-flight (TOF) spectrometer equipped with microchannel plates (MCPs) and a delay-line anode [3]. Signals from delay-line anode and MCPs were recorded by the digitizer and analyzed by a software discriminator. The arrival time and arrival position of each ion were determined. The position information was used to compensate the TOF that varied depending on the departure positions perpendicular to spectrometer axis [3]. On the other hand, in the experiment with clusters, we used a velocity map imaging (VMI) spectrometer [5] equipped with MCPs and phosphor screen. The two-dimensional projection of the velocity distribution imprinted on the phosphor screen was recorded using a CCD camera. The three-dimensional velocity distribution was reconstructed from the projection using an Abel inversion procedure. Electrons or ions emitted into 4π sr with kinetic energies up to ~1 keV could be collected using our VMI spectrometer. The TOF and VMI spectrometers were placed face-to-face with the focal point of the XFEL beam in between. Thus we could switch between the spectrometers without opening the chamber.

3. Results and discussion

We have obtained charge state distributions of Ar and Xe atoms irradiated by XFEL pulses. The absolute fluence of the XFEL pulse has been determined using two-photon processes in the Ar atom with the help of benchmark *ab initio* calculations. Average peak fluence at the full XFEL power condition is ~50 μJ/μm^2 for both 5 keV and 5.5 keV photon energies.

Figure 1 shows charge state distribution of isolated Xe ions measured at the photon energies of 5 keV and 5.5 keV. The charge states up to +22 are observed at the photon energy of 5 keV, whereas the charge states up to +26 are observed at the photon energy of 5.5 keV. Theoretical charge state distributions are also shown in Fig. 1. In the theoretical model [6], relativistic effects and shake-off are not included. In spite of these limitations, the experimental and theoretical results are in reasonable agreement at 5.5 keV. In the charge state distribution at 5 keV, noticeable discrepancy between experiment and theory appears above the charge state of +16 and increases with increasing charge state. This discrepancy is interpreted by the resonance-enabled x-ray multiple ionization mechanism, where transient resonant excitation enhances the ionization process beyond the limit expected by the sequential ionization model [7].

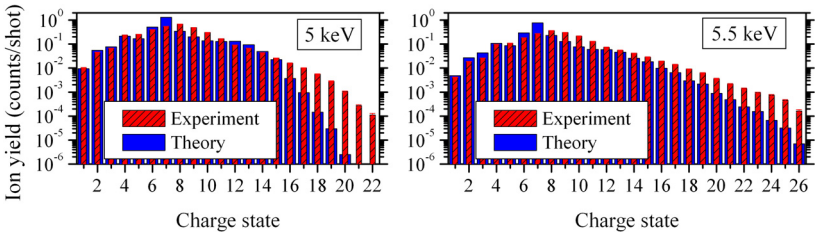

Fig. 1. Experimental and theoretical charge state distribution of Xe at the photon energy of 5 keV and 5.5 keV.

We have obtained electron energy spectra of Ar clusters irradiated by XFEL pulses at cluster sizes 100-1000. In the spectra for the small cluster sizes, we can identify the LMM Auger electrons that are emitted following the KLL Auger emission and form the broad peak in the range 150-200 eV. The intensity of continuum electron emission with kinetic energy below 150 eV increases with the increase in the cluster size. When the intense XFEL pulse irradiates the cluster, atoms in the cluster are ionized by photoionization and sequential Auger decay. The cluster gets more charge with the increase in number of ionized atoms. As a result, the LMM Auger electrons are decelerated by the Coulomb attractive force from the highly-charged cluster ion. The decelerated electrons are eventually trapped by the cluster ion. The LLM Auger electrons and the secondary electrons produced by the low-energy electron impact will be also trapped. As a result, one can expect that nanoplasma is formed. In the electron spectra, we found also thermal electrons that form the peak at zero energy. This thermal electron emission is also enhanced with the increase in the cluster size. This thermal emission is the evidence of nanoplasma formation. Our theoretical calculations [8] that fully reproduce these observations support the above described scenario and predict that the thermal emission from nanoplasma occurs after the XFEL pulse duration.

Acknowledgements

This study was supported by the X-ray Free Electron Laser Utilization Research Project and the X-ray Free Electron Laser Priority Strategy Program of the MEXT, by JSPS, by the Proposal Program of SACLA Experimental Instruments of RIKEN, and by the IMRAM project.

References

[1] T. Ishikawa *et al.*, "A compact X-ray free-electron laser emitting in the sub-ångström region" Nat. Photonics **6**, 540-544 (2012).

[2] H. Fukuzawa *et al.*, "Deep inner-shell multiphoton ionization by intense x-ray free-electron laser pulses" Phys. Rev. Lett. **110**, 173005 (2013).

[3] K. Motomura *et al.*, "Sequential multiphoton multiple ionization of atomic argon and xenon irradiated by x-ray free-electron laser pulses from SACLA" J. Phys. B **46**, 164001 (2013).

[4] K. Tono *et al.*, "Beamline, experimental stations and photon beam diagnostics for the hard x-ray free electron laser of SACLA" New J. Phys. **15**, 083035 (2013).

[5] A. T. J. B. Eppink and D. H. Parker, "Velocity map imaging of ions and electrons using electrostatic lenses: Application in photoelectron and photofragment ion imaging of molecular oxygen" Rev. Sci. Instrum. **68**, 3477-3484 (1997).

[6] S. K. Son and R. Santra, "XATOM–an integrated toolkit for X-ray and atomic physics" (CFEL, DESY, Hamburg, Germany, 2011).

[7] B. Rudek *et al.*, "Ultra-efficient ionization of heavy atoms by intense X-ray free-electron laser pulses" Nat. Photonics **6**, 858-865 (2012).

[8] Z. Jurek, B. Ziaja and R. Santra, "XMDYN" (CFEL, DESY, Hamburg, Germany, 2013).