Hidden Charge States in Soft X-ray Laser Produced Nanoplasmas Revealed by Fluorescence Spectroscopy

A. Przystawik [1], L. Schroedter [1], M. Müller [2], A. Kickermann [1], S. Toleikis [1], M. Adolph [2], L. Flückiger [2], T. Gorkhover [2], L. Nösel [2], M. Krikunova [2], T. Oelze [2], Y. Ovcharenko [2], D. Rupp [2], M. Sauppe [2], D. Wolter [2], S. Schorb [3], C. Bostedt [3], T. Möller [2], T. Laarmann [1]

[1] *Deutsches Elektronen-Synchrotron DESY, Notkestrasse 85, 22603 Hamburg, Germany*
[2] *Institut für Optik und Atomare Physik, TU Berlin, Hardenbergstr. 36, 10623 Berlin, Germany*
[3] *SLAC National Accelerator Laboratory, 2575 Sand Hill Road, Menlo Park, CA 94025, USA*

We use rare gas clusters as prototypical model systems and their response to strong laser fields can be studied in the limit of short wavelengths making use of intense fs pulses from free-electron lasers (FELs). In experiments their size can be easily controlled from a very few to many ten-thousands of atoms. FLASH offers the possibility to study energy absorption and dissipation processes upon resonant excitation of Xe 4d electrons at 13.5 nm. The structure of core-shell clusters can be controlled by applying well-known co-expansion or pick-up schemes. Core-shell clusters may serve as a model system for tamper layers in bio-imaging applications. Of interest is the exact way the cluster disintegrates and what processes play a role in the initial nanoplasma formation. Previous investigations on Xe-core-Ar-shell clusters using ion time-of-flight (TOF) spectrometers show that at 13.5 nm wavelength the interaction results in highly charged outer shells, while the cluster core seems to be only lowly charged or even neutral [1, 2]. The shell then disintegrates by Coulomb explosion, while the behavior of the core can be described by a hydrodynamic expansion. However, these studies have to resort to theoretical predictions when making statements about the initial FEL cluster response on the ultrafast timescale. In our work [3], we circumvent a crucial limitation of TOF studies, namely that this technique only detects the ionic fragments long after the interaction, leaving plenty of time for charge states to be created and to recombine, never to be seen in ion TOF spectra. By employing XUV fluorescence spectroscopy, we are sensitive to charge generation, charge transfer and charge recombination processes on the ultrafast fs timescale.

Our spectrometer detects fluorescence in the wavelength range from 10 to 76 nm in a single image with a resolution of about 0.2 nm over the whole range. We measured spectra of xenon, argon and xenon-core-argon-shell clusters at varying FEL intensities and cluster sizes up to approximately 19000 atoms. Fig. 1 shows typical fluorescence spectra of xenon and argon clusters integrated over 1200 FEL shots as well as the size dependent fluorescence yield per atom. Fluorescence is predominantly emitted from a shell of highly-charged ions on the surface of the cluster during the disintegration when the number density has already significantly decreased. The fraction of surface atoms decreases with increasing cluster size as can be seen in the reduced fluorescence yield per atom in Fig. 1c, while the spectrum hardly changes aside from small shifts in the ionic charge distribution. The assignment of charge states is possible by searching for correlations in the FEL intensity dependence of fluorescence line intensities in comparison with available data from atomic spectra [4, 5, 6]. The maximum charge states of at least Xe^{11+} are similar to those found in previous experiments with ion-TOF spectroscopy [2]. There, charge states of up to 9+ were identified, at a somewhat lower FEL intensity of $5 \cdot 10^{14}$ W/cm^2).

The well-defined radial distribution of different elements in Xe-core-Ar-shell clusters allows to disentangle directly the radiative decay of the Coulomb exploding cluster ion surface (Ar) from fluorescence that stems from the center of the Xe nanoplasma core because the characteristic short-wavelength fluorescence appears in different spectral ranges. While the vast majority of the spectrum consists of fluorescence from Ar ions, which were situated in the cluster shell, we can gain direct insight into the charge states that are initially created upon resonant excitation of the Xe core by the FEL. This information is hidden and not observable in TOF ion spectra. Figure 2 shows a comparison of spectra of pure Xe and Ar clusters with a spectrum of a core-shell-cluster as well as

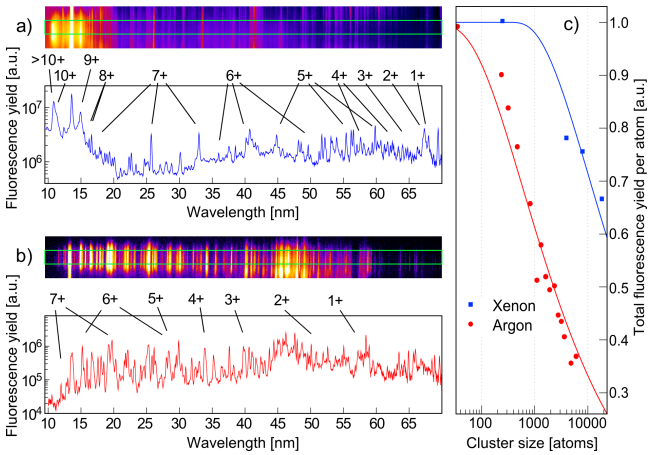

Figure 1: Typical fluorescence spectra of pure Xe clusters (a) and pure Ar clusters (b) comprising 4100 and 400 atoms, respectively. The data is recorded with an x-ray camera at an FEL intensity of approximately $2 \cdot 10^{15}$ W/cm^2 and plotted in logarithmic scale. The same line out of the CCD chip (green rectangle) is evaluated for each spectrum. Charge states have been assigned to more than 100 fluorescence lines ($\sim$80%) and a characteristic selection is given. Total fluorescence yield per atom as function of cluster size is shown in (c).

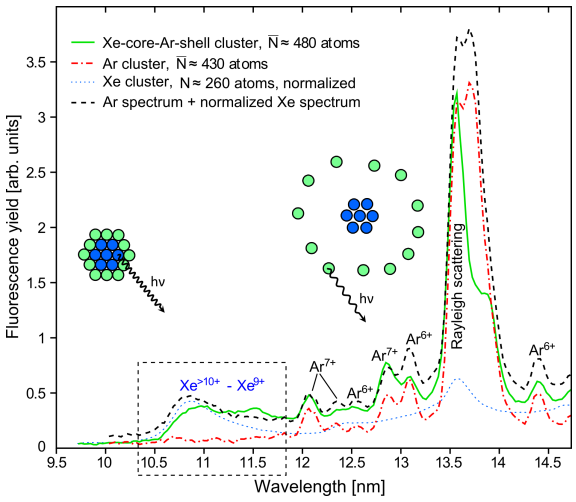

Figure 2: Comparison of measured spectra of Xe-core-Ar-shell and pure Ar clusters with a spectrum synthesized from normalized Ar- and Xe spectra. The Xe-core-Ar-shell and the synthesized spectrum show reasonable agreement at short wavelengths with Xe charge states of at least 11+. As the pictographs illustrate the timeintegrated spectra provides information on the highly charged Xe core from early times of the interaction (left), as well as information on the Ar-shell upon Coulomb explosion (right).

one synthesized as a weighted sum of spectra of Ar and Xe clusters. There is a clear similarity in the structures between 10.5 and 12 nm of the measured core-shell spectrum and the emulated one. While the interaction between the two constituents cannot be replicated by this method, the result is clear evidence of the transient presence of highly charged $Xe^{>10+}$ ions in the core-shell cluster. The detected plasma fluorescence must occur fast, before it is efficiently suppressed in the center of the nanoplasma due to non-radiative recombination on a sub-ps timescale [7].

In summary, the ionization dynamics of Xe clusters coated with an Ar tamper layer exposed to FEL pulses at 10^{15} W/cm^2 has been studied with XUV fluorescence spectroscopy. The experiments give direct evidence that – in addition to the multiply charged Ar surface which Coulomb explodes and finally leads to fluorescence from individual ions – also the resonantly excited Xe core is initially highly charged. A sacrificial tamper layer provides an efficient electron source for partial neutralization of highly charged ions created in the center.

References

[1] M. Hoener, C. Bostedt, H. Thomas, L. Landt, E. Eremina, H. Wabnitz, T. Laarmann, R. Treusch, A. R. B. de Castro and T. Möller, J. Phys. B 41, 181001 (2008)

[2] H. Thomas, C. Bostedt, M. Hoener, E. Eremina, H. Wabnitz, T. Laarmann, E. Plönjes, R. Treusch, A. R. B. de Castro and T. Möller, J. Phys. B 42, 134018 (2009)

[3] L. Schroedter *et al.*, submitted to Phys. Rev. Lett. (2014)

[4] N. Böwering, M. Martins, W. N. Partlo and I. V. Formenkov, J. Appl. Phys. 95(1) 16 (2004)

[5] Brühl, Ph.D. thesis, Universität Hamburg (1996)

[6] A. Kramida, Yu. Ralchenko, J. Reader *et al.*, http://physics.nist.gov/asd (2012)

[7] E. Ackad, N. Bigaouette, S. Mack *et al.*, New J. Phys. **15**:053047 (2013)