XUV-fluorescence of rare gas clusters in the light of FLASH

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Clusters have unique electronic and geometric properties as a function of cluster size and composition which make them intriguing study subjects of photon science. In particular rare gas clusters serve as prototypical model systems and their photophysical response to strong laser fields can be studied at short wavelengths making use of intense fs pulses from free-electron lasers (FELs). In experiments their size can be easily controlled from very few to many ten-thousands of atoms. Moreover, doped rare gas clusters containing xenon atoms either in the core or within outer shells give the possibility to study energy absorption and dissipation processes upon resonant excitation of 4d electrons (13.5 nm) at FLASH. The geometric structure of these core-shell clusters can be controlled by applying well-known co-expansion or pick-up schemes [1]. Core-shell clusters may be of interest as tamper layers in bio-imaging applications [2].
Generally speaking, a cluster reacts to a strong laser field by being destroyed. The exact way the cluster disintegrates depends very much on the wavelength of the radiation, the cluster composition and the cluster size. Previous investigations on xenon-core-argon-shell clusters showed that at a wavelength of 13.5 nm the light-matter interaction results in highly charged outer shells, while the cluster core seems to be only lowly charged or even neutral [3, 4]. The cluster 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.

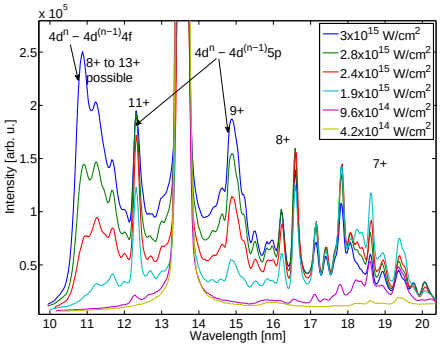

Figure 1: Fluorescence spectra of Xe clusters consisting of about 6000 atoms at varying FEL intensities. The exposure time for each spectrum was 1200 FEL shots.

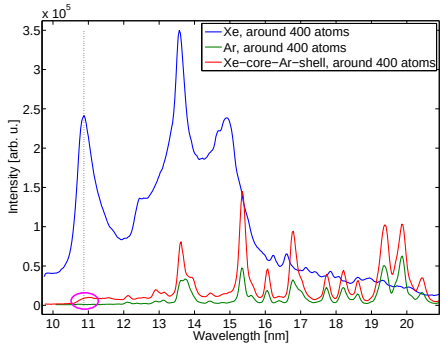

Figure 2: Comparison between pure xenon clusters, pure argon cluster and xenon-core-argon-shell clusters. The spectral line at 10.9 nm in the core-shell-clusters indicates an initial high charging of the cluster core.

The usual way to analyze the interaction is to detect the cluster fragments with ion time-of-flight (TOF) spectrometers. An inherent limitation of this technique is that only the end product of the reaction - in other words, the charge states of the cluster fragments microseconds after the reaction took place - is detected. This leaves plenty of time for higher charge states to be created and to

recombine, never to be seen in ion TOF spectra. New scattering data supports this concern [5].
In our work, we use a different method of detection, namely XUV fluorescence spectroscopy. By employing this method, we are sensitive to charge generation, charge transfer and charge recombination processes on the ultrafast fs timescale. The spectrometer is capable of detecting fluorescence in the wavelength range from 10 to 75 nm in a single image. The measured resolution at 16.6 nm is $\frac{\lambda}{\Delta\lambda} \approx 80$ (higher at higher wavelengths, since the linewidth $\Delta\lambda$ remains almost constant over the detection range). We measured spectra of xenon, argon and xenon-core-argon-shell clusters at varying FEL intensities and from atomic spectra up to cluster sizes of approximately 18000 atoms. Figure 1 shows several fluorescence spectra of xenon clusters consisting of approximately 6000 atoms. The spectra were recorded at FEL intensities from about $1 \cdot 10^{14}$ W/cm^2 to about $3 \cdot 10^{15}$ W/cm^2 and integrated over 1200 FEL shots. The FEL intensity was varied by moving the refocusing mirror (and thus the focal position) relative to the cluster beam. By comparing the measurements with theoretical data [6], one can assign charge states to the observed spectral lines. It can clearly be seen that different charge states reach their maximum at different FEL intensities. The maximum charge states of at least Xe^{11+} are similar to those found in previous experiments with ion-TOF spectroscopy [4]. There, charge states of up to 9+ were identified, at similar experimental conditions. (FEL wavelength 13.7 nm, FEL intensity $5 \cdot 10^{14}$ W/cm^2). Further conclusions about the involved charge states can be drawn from analyzing the relative changes between peaks for different FEL intensities. Even though the peaks may reach their maximum at the same FEL intensity, different intensity dependencies reveal the involvement of different charge states and excited states, respectively. Hence, the order of the underlying nonlinear interaction is another important source of information in the data analysis that is currently ongoing.
As was mentioned in the introduction, FEL-cluster-interaction at our experimental conditions ultimately results in a highly charged cluster shell and a neutral or lowly charged core. In the experiments with xenon-core-argon-shell, however, we found *direct* evidence of the cluster core being charged initially just as high as the outer shell, as can be seen in figure 2. The xenon cluster spectrum shows a clear fluorescence signal at approximately 11 nm, corresponding to charge states of at least 8+, whereas pure argon clusters exhibit no spectral line at this position. Interestingly, the xenon-core-argon-shell clusters show an evolving line at this position. Therefore, their core is initially excited to high charge states, even though this charge is redistributed later on [3].
In conclusion, the experiments provide new information on charge generation and charge migration not accessible via ion-TOF spectroscopy.

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