Intense soft X-ray matter interaction: Multiple ionization of atom clusters by free-electron laser radiation

T. Möller, P. Gürtler, T. Laarmann, W. Laasch, J. Schulz,
A. Swiderski, and H. Wabnitz
Hamburger Synchrotronstrahlungslabor HASYLAB at Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany

Fourth-generation light sources based on free-electron lasers (FEL) will provide intense, short-wavelength radiation for a wide range of applications in physics, chemistry and biology. Large scale FELs are proposed that could be used to generate pulses of hard X-rays. The FEL at DESY, a proof-of-principle project [2], has begun operation at far-ultraviolet wavelengths and initial results are presented in this paper [2]. Xe clusters are illuminated with intense FEL pulses of 98 nm wavelength. Here, unexpectedly strong absorption of the laser radiation by Xenon clusters was observed, resulting in the explosion of the clusters and the ejection of high-energy, multiply charged ions. For the first time , we were able to observe such a highly nonlinear interaction between matter and soft X-rays below 100 nm. So far, most work in the field of nonlinear processes was restricted to infrared, visible and ultraviolet light from lasers [3].

The FEL at DESY is operating in the regime below 100 nm wavelength[4] and offers new scientific opportunities. The understanding of the interaction of short-wavelength, short-pulse radiation with matter is essential for all future experiments. In a first series of experiments, the ionization of Xe atoms and clusters was compared. While Xe atoms become only singly ionized by the absorption of single photons, absorption in clusters is strongly enhanced. On average, each atom in a large cluster absorbs up to 400 eV, corresponding to 30 photons. The clusters are heated up and electrons are emitted after sufficient energy is acquired. There is some evidence that the photo absorption of ionized clusters at 100 nm is too efficient to be explained by straightforward models of collision induced absorption. The results will have strong impact on our understanding of radiation damage. A key issue for future studies will be the extrapolation to short wavelengths and to identify the absorption processes in the nm to Å regime. The latter will be important because X-ray lasers could take snapshot pictures of the atomic structure of single biomolecules [5].

The experiments were performed by irradiating atoms and clusters with ~100 fs long FEL pulses at 98 nm wavelength and a power density of up to $7{*}10^{13}$W/cm^2. The resulting ions are detected with a time-of-flight mass spectrometer. Thanks to the high laser intensity, mass spectra with a high signal-to-noise ratio can be recorded in a single shot of the FEL.

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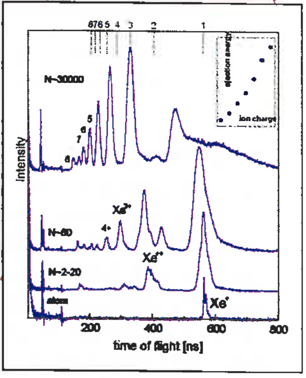
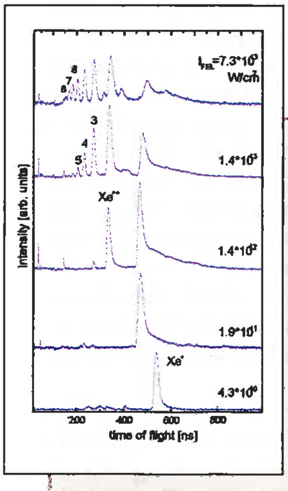
▶ **Fig. 1:** Time-of-flight mass spectra of ionisation products of Xe atoms and clusters. The spectra are recorded after ionisation with soft X-rays with 98 nm wavelength at an average power density of $2*10^{13}$ W/cm^2. After irradiation of clusters, highly charged ions are observed. The mass peaks are rather broad and displaced with respect to the calculated flight times indicated by bars in the uppermost part of the figure. This indicates that the ions have high kinetic energy. The number N of atoms per cluster is given in the figure. The kinetic energy of the ions as a function of the charge for N=1500 is displayed in the inset.



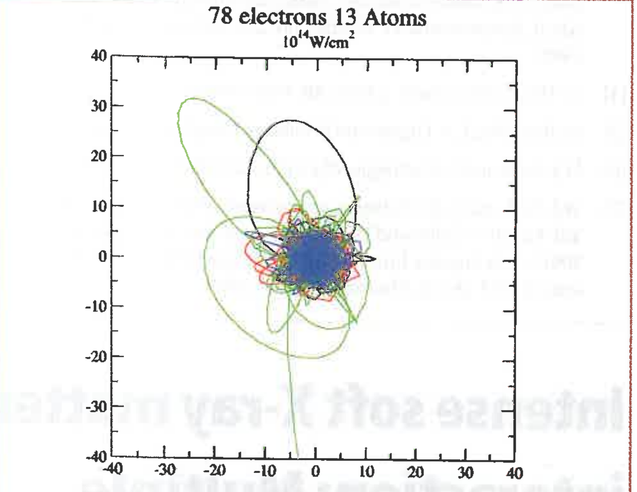
TOF mass spectra for different clusters sizes recorded at $2*10^{13}$ W/cm^2 are shown in Figure 1. The most striking result is the surprisingly different ion signal from atomic and cluster beams. While only singly charged ions are observed after irradiation of isolated atoms, atomic ions with charges up to +8 are detected if clusters are irradiated. The clusters absorb many photons and completely disintegrate into singly and multiply charged ions. The mass peaks are very broad, indicating that the ions have a high kinetic energy. This can be understood in terms of a Coulomb explosion process. The population of different ion states and their kinetic energy strongly depends on the power density. This is shown in Figure 2 for clusters comprising 1500 atoms. At the highest power level of $7*10^{13}$ W/cm^2, charge states up to +8 are detected. The strong dependence on the power density is a clear sign that optical non-linear processes dominate the ionisation of the clusters at the power levels used.

The average kinetic energy per ion strongly varies with its charge state and the cluster radius. For Xe^{7+} kinetic energies more than 2 keV were observed. The high energies are a clear signature of a Coulomb explosion [6]. From the results it is concluded that an energy of up to several hundred eV per atom is taken from the FEL beam. Coulomb explosions of clusters are not a new phenomenon [7,8] and have been already induced with infrared (IR) light. It is agreed that the experimental findings are explained by field ionisation of the atoms and the clusters by the strong electric field of the IR laser [9,10]. The dramatic effects observed when clusters are exposed to short-wavelength radiation are somewhat surprising because the Coulomb explosion starts at 10^{11} W/cm^2, which is much lower than the power density needed to induce a Coulomb explosion in the IR. Classical model calculations for the movement in charged clusters show that the ionization process at short wavelength is due to the absorption of many photons heating the cluster and subsequent electron emission (see Figure 3). The experimentally determined absorption of the clusters is 3-5 times larger than predicted by the classical calculation. The steps of the ionization process are illustrated in Figure 4.



◀ **Fig. 2:** Time-of-flight mass spectra recorded after irradiation of Xe clusters comprising 1500 atoms as a function of the power density. The power density is given in the figure. The spectrum at the bottom is recorded at a reduced gain of the FEL. The intensity of highly charged ions increases with increasing power density. Experimental details: The intrinsic pulse energy of the ~ 100 fs long SASE-FEL pulses [1] typically varies between 1.5 and 25 µJ. The spectra with a power density of $1.9*10^{11}$- $7*10^{13}$ W/cm2 are taken with pulses of 25 µJ energy. The power density could be lowered by moving the cluster beam out of the focus to 10^{10} W/cm^2 at 1.5 µJ.

The absorption of short-wavelength radiation and subsequent ionisation differs considerably from that in the optical spectral range. A absorption and ionization start by single photon absorption as described by quantum mechanics. After many unbound electrons are created, a plasma is formed. There is evidence that already at 100 nm quantum mechanical modelling of the absorption processes including resonant electronic transitions becomes important. Going to shorter wavelengths will enhance this trend. The results show that the VUV-FEL has opened up a new and exciting field of non-linear physics and they are a first step towards experiments in the nm and Å range.



▲ **Fig. 3:** Classical simulations of the electron motion and ionization for Xe_{13} clusters with soft X-rays. Electron trajectories were calculated for 100 fs long pulses of constant intensity. The position of the ions is that of the neutral cluster and kept fixed during the simulation. The scale is in Å.

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