With the advent of intense fs laser pulses at optical frequencies and a power density larger than $\sim 10^{15}$ W/cm$^2$ a variety of fascinating effects were observed in the field of laser matter interaction [1]. One basic result of studies on atoms, molecules and clusters is the possibility of creating photons with energies much larger than that of the initial laser photon. Often molecular beams of clusters, especially rare gas clusters, were chosen to efficiently convert “low-energy” laser photons into high-energy interaction products. After irradiation of the clusters, highly charged ions with MeV kinetic energies and extremely energetic electrons in the keV range were observed [2, 3]. A lot of work has been done, both experimentally and theoretically to understand the effect of power density, pulse duration and cluster size on the dynamics of the laser cluster interaction [4]. On the other hand, only little is known about the effect of the laser photon energy, particularly in the high-energy limit.

Very recently the free electron laser (FEL) at DESY has opened the door to a new regime of strong field matter interaction in the vacuum ultraviolet spectral range (VUV) [5]. The FEL has reached a gain of $\sim 10^6$ and provides gigawatt pulses of $\sim 30$-100 fs length below 100 nm wavelength. First experiments on Xe clusters have already thrown up a surprise, namely very efficient absorption of intense fs VUV-light resulting in a Coulomb explosion of the clusters [6]. The fundamental question is the following. Which of the concepts from the optical regime to describe the absorption and ionization of matter can be taken over, or have to be modified in the case of intense VUV-radiation? A photoemission study presented in this contribution gives a fingerprint, particularly of the ionization dynamics in the short-wavelength limit.

Small Xe and Ar clusters were prepared in a supersonic expansion and irradiated with focussed FEL radiation at a power density of $\sim 10^{12}$ W/cm$^2$ and a photon energy of $\sim 13$ eV. The electrons produced during the laser cluster interaction were detected in the direction of the laser polarization with the multi-channel plate (MCP) of a time-of-flight (TOF) photoelectron spectrometer. The acceptance angle of the detector was limited to $\sim 6^\circ$ by a small entrance aperture in front of the field-free TOF drift tube. In order to increase the collection efficiency for low-energy electrons, an extraction voltage of $\sim 10$ V was applied between the grounded aperture and a grid mounted in a distance of 12 mm with the laser focus in between.

![Fig. 1: Electron kinetic energy distribution from Ar$_{270}$ clusters (a) and Ar atoms (b) irradiated with intense FEL light.](image1)

![Fig. 2: Electron kinetic energy distribution from Xe$_{70}$ clusters (a) and Xe atoms (b) irradiated with intense FEL light.](image2)
Fig. 1 shows photoelectron spectra of Ar\textsubscript{70} clusters (a) and Ar atoms (b), which were irradiated with laser pulses at approximately $4 \times 10^{12}$ W/cm\textsuperscript{2}. We would like to note that at this power density Ar as well as Xe clusters completely disintegrate into multiply charged atomic fragments by Coulomb explosion. The VUV-laser cluster interaction results mainly in a kinetic energy distribution of photoelectrons, which peaks at $\sim$5 eV and decreases almost exponentially with vanishing intensity between 20 and 30 eV. Its lineshape can be fitted with a Maxwell-Boltzmann distribution indicated by the gray-shaded area (B). In addition to these “thermal” electrons, the cluster spectra exhibit a relatively pronounced asymmetric peak (C) at $\sim$10.4 eV. By comparison with the photoelectron spectrum of a beam of Ar atoms (Fig. 1b), this peak can be assigned clearly to two-photon ionization of neutral Ar atoms which are always present in the cluster beam. It should be noticed, that the FEL photon energy is well below the first ionization potential of Ar atoms (IP=15.76 eV). Due to the extraction voltage between the grid and the aperture the transmission of the spectrometer increases by approximately three orders of magnitude for electrons with a kinetic energy below $\sim$2.5 eV. To account for these “stray” electrons with very low kinetic energy an additional contribution (A) is included in the deconvolution of the observed cluster spectrum.

Photoemission spectra of Xe\textsubscript{70} clusters and Xe atoms measured at a peak power density of approximately $4 \times 10^{12}$ W/cm\textsuperscript{2} are presented in Fig. 2a and Fig. 2b. Interestingly, the kinetic energy distribution of small Xe clusters (a) exhibit nearly the same characteristics as already described in case of Ar clusters irradiated with similar FEL pulses, namely a broad thermal distribution with a maximum at $\sim$5 eV and almost exponentially decreasing intensity up to a maximum kinetic energy of 20-30 eV. At first view, this is surprising since in case of Xe each atom inside the cluster can be easily ionized by a single-photon process (IP=12.13, see Fig. 2b), while in case of Ar at least two photons are needed.

The experimental results can be understood in the following way. At the beginning of the VUV-laser cluster interaction the “inner” ionization of the cluster occurs mainly by two-photon absorption of the Ar atoms, respectively single-photon absorption of Xe atoms inside the cluster. After a few fs a nanoplasma is formed which consists of one quasifree electron per ion. Most of the produced electrons are bound by strong Coulomb forces to the dense cluster ion core. The energy deposition is then mainly controlled by collisional heating effects of electrons and ions, such as inverse bremsstrahlung and plasma resonance absorption. Thus, the heating of the electron gas and consequently the electron emission is almost independent on the cluster species, as seen in Fig. 1a and Fig. 2a. The electrons undergo many scattering events inside the cluster before “outer” ionization. This results in a statistical redistribution of energy among the electronic degrees of freedom. Since the FEL pulse duration is $\sim$100 fs and the characteristic time scale for electron-phonon coupling is on the order of picoseconds, the electron gas becomes uncoupled from the lattice. Thus, the transient-electron temperature becomes much larger than the local lattice temperature by a transfer of laser energy into internal energy of the electron gas. A delayed release of thermally excited electrons follows, which is generally called photon-assisted thermionic emission. This behavior is in striking contrast to that at optical frequencies, namely (i) field ionization of surface electrons on the rising edge of the laser pulse and (ii) delayed emission of “hot” (keV) bulk electrons [7] We like to note that the process of thermionic electron emission is in good agreement with molecular dynamics simulation of the electron motion under the influence of the VUV-laser pulse and the Coulomb forces of electrons and ions [6].

References