Freedom of electrons is short-lived

June 27 2014

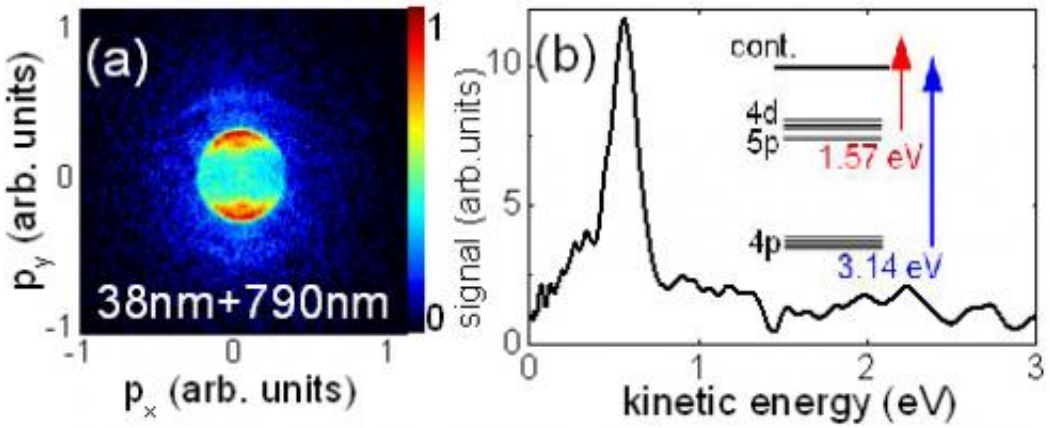

Fig. 1: (a) Two dimensional electron momentum map, showing the momentum distribution of the ejected electrons along (vertical) and perpendicular (horizontal) to the XUV/NIR laser polarization, after XUV ionization and NIR probing of argon clusters with an average size of 3500 atoms. The ring structure corresponds to the ionization of excited atoms by the NIR pulse. (b) The corresponding kinetic energy spectrum shows a peak at an energy of 0.6 eV that results from single-photon NIR ionization of the 4d and 5p excited states of argon.

During the interaction of an intense extreme-ultraviolet (XUV) laser pulse with a cluster, many ions and free electrons are created, leading to the formation of a nanoscale plasma. In experiments using XUV/X-ray free electron lasers (FELs) it was previously demonstrated that only a small fraction of these electrons can leave the cluster, while the majority of the electrons remain trapped within the cluster and may therefore recombine with ions. In a novel approach using a laboratory-scale XUV

source, we have now measured the time scale of these electron-ion recombination processes leading to a strong formation of excited atoms, which is in the picosecond range. The results show that it is even possible to follow the laser-induced cluster expansion process up to nanosecond times.

The formation of a large number of charges in a cluster by an intense laser pulse can lead to the generation of a transient nanoplasma consisting of free electrons and ions. In the past, fascinating processes could already be observed in nanoplasmas, including nuclear fusion or the creation of neutral atoms with very high kinetic energies. While nanoplasmas are routinely generated during the interaction of clusters with intense XUV pulses from free-electron lasers, a detailed understanding of the processes inside the plasma is challenging. Theoretical models have predicted that the majority of electrons remains trapped in the cluster and may eventually recombine with ions such that both transient species cannot be observed in usual experiments. However, an experimental investigation of these dynamics is crucial, since processes in clusters are complex and manifold, and their detailed prediction is difficult. A promising route towards a better understanding of the different mechanisms in nanoplasmas is the development of time-resolved experiments. In this context, intense high-order harmonic generation (HHG) sources that can deliver light pulses down to the attosecond regime are very interesting. This laboratory-scale XUV sources provide a straightforward way to carry out pump-probe experiments on clusters and can significantly improve the possibilities for the understanding of cluster dynamics.

In an international collaboration led by researchers from the Max-Born-Institut, the first pump-probe experiment on clusters using an intense HHG source was now performed. In the current issue of *Physical Review Letters* [112, 253401 (2014)] Bernd Schütte, Marc Vrakking and Arnaud Rouzée and their colleagues Filippo Campi from the University of Lund

and Mathias Arbeiter and Thomas Fennel from the University of Rostock present the results of these investigations.

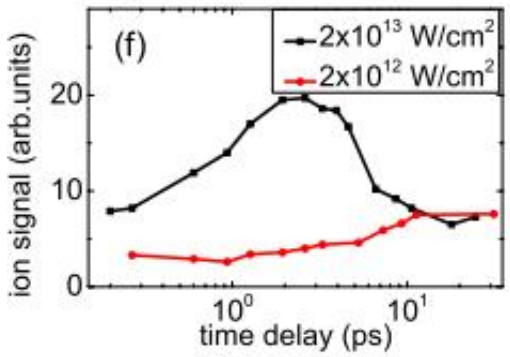

Fig. 2: Time-dependent Xe+ ion yield after XUV ionization of mixed clusters consisting of a xenon core and an argon shell. An NIR pulse at two different intensities is used for probing. At an intensity of 2x1013 W/cm2, the Xe+ ion yield has a maximum at a delay of appr. 3 picoseconds, due to a well-known plasma resonance effect. At the lower intensity of 2x1012 W/cm2, the signal monotonically increases during the first 10 picoseconds, which is identified as the time scale of electron-ion recombination.

The development of a technique allowing the Reionization of Excited Atoms from Recombination (REAR) makes it possible for the first time to infer information on ion charge states prior to recombination. By using near-infrared (NIR) probe pulses, a surprisingly extensive formation of excited atoms was observed and could be shown to originate from recombination between electrons and ions. It was demonstrated that in the nanoplasma electrons released by means of photo-ionization only remain quasi-free for a short time up to 10 picoseconds before they undergo a recombination process with the surrounding ions.

More information about these processes was obtained by generating special clusters that consist of a xenon core and an argon shell. These

investigations showed that recombination preferentially takes place in the xenon core of the cluster. The wavelength of the ionizing pulse interacting with the cluster was shown not to be important: excited atom formation attributed to recombination processes was also observed when using NIR or blue pump pulses instead of XUV pulses. This demonstrates the general implications of the current findings for the explanation of previous experiments carried out in different wavelength regimes. Moreover, the cluster expansion dynamics could be traced up to the nanosecond range by using the REAR technique.

Our results show the remarkable versatility of intense HHG pulses for the study of dynamic processes in clusters. In the future, the investigation of other extended systems like biomolecules will benefit from the availability of these laboratory-scale XUV light sources.

More information: Bernd Schütte, Filippo Campi, Mathias Arbeiter, Thomas Fennel, Marc J. J. Vrakking and Arnaud Rouzée: "Tracing electron-ion recombination in nanoplasmas produced by extreme-ultraviolet irradiation of rare-gas clusters", *Physical Review Letters* 112.253401,(2014)

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