Revealing secrets of exploding clusters

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An intense light pulse interacting with a weakly bound van der Waals cluster consisting of thousands of atoms can eventually lead to the explosion of the cluster and its complete disintegration. During this process, novel ionization mechanisms occur that are not observed in atoms. With a light pulse that is intense enough, many electrons are removed from their atoms that can move within the cluster, where they form a plasma with the ions on the nanometer scale, a so called nanoplasma. Due to collisions between the electrons, some of them may eventually gain sufficient energy to leave the cluster. A large part of the electrons, however, will remain confined to the cluster. It was theoretically predicted that electrons and ions in the nanoplasma recombine to form Rydberg atoms, however, an experimental proof of this hypothesis is still missing. Previous experiments were carried out at large scale facilities like free-electron lasers that have sizes from a few hundred meters to a few kilometers showing already surprising results such as the formation of very high charge states when an intense XUV pulse interacts with the cluster. However, the accessibility to such sources is strongly limited, and the experimental conditions are extremely challenging. The availability of intense light pulses in the extreme-ultraviolet range from an alternative source is therefore important to gain a better understanding of the various processes occurring in clusters and other extended systems such as biomolecules exposed to intense XUV pulses.
Scientists from the Max-Born-Institut have developed a new light source that is based on the process of high-order harmonic generation. In the experiment, an intense pulse in the extreme-ultraviolet range with a duration of 15 fs (1fs=10-15s) interacted with clusters consisting of argon or xenon atoms. In the current issue of Physical Review Letters (Vol. 112-073003 publ. 20 February 2014) http://journals.aps.org/prl/abstract/10.1103/PhysRevLett.112.073003 Bernd Schütte, Marc Vrakking and Arnaud Rouzée present the results of these studies, which are in very good agreement with previously obtained results from free-electron lasers: the formation of a nanoplasma was inferred by measuring the kinetic energy distributions of electrons formed in the cluster ionization process, showing a characteristic plateau up to a maximum kinetic energy given by the kinetic energy resulting from photoionization of an individual atom. In collaboration with the theoreticians Mathias Arbeiter and Thomas Fennel from the University of Rostock, it was possible to numerically simulate the ionization processes in the cluster and to reproduce the experimental results. In addition, by using the velocity map imaging technique, a yet undiscovered distribution of very slow electrons was observed and attributed the formation of high-lying Rydberg atoms by electron-ion recombination processes during the cluster expansion. Since the binding energies of the electrons are very small, the DC detector electric field used in the experiment was strong enough to ionize these Rydberg atoms, leading to the emission of low energy electrons. This process is also known as frustrated recombination and could now be confirmed experimentally for the first time. The current findings may also explain why in recent experiments using intense X-ray pulses, high charge states up to Xe26+ were observed in clusters, although a large number of recombination processes is expected to take place. Moreover, the opportunity to carry out this type of experiment with a high-order harmonic source makes it possible in the future to perform pump-probe experiments in clusters and other extended systems with a time resolution down to the attosecond range.
Full citation:
Bernd Schütte, Mathias Arbeiter, Thomas Fennel, Marc J. J. Vrakking and Arnaud Rouzée, "Rare-gas clusters in intense extreme-ultraviolet pulses from a high-order harmonic source", Physical Review Letters 112, (2014)

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