Intracluster Coulombic decay following intense NIR ionization of clusters

XXIX International Conference on Photonic, Electronic, and Atomic Collisions (ICPEAC2015) IOP Publishing
Journal of Physics: Conference Series **635** (2015) 102004 doi:10.1088/1742-6596/635/10/102004

Intracluster Coulombic decay following intense NIR ionization of clusters

Bernd Schütte*† [1], Mathias Arbeiter,‡ Thomas Fennel‡, Ghazal Jabbari§, Kirill Gokhberg§, Alexander I. Kuleff§, Marc J. J. Vrakking*, Arnaud Rouzée*

* Max-Born-Institut, Max-Born-Strasse 2A, 12489 Berlin, Germany
† Department of Physics, Imperial College London, SW7 2AZ London, United Kingdom
‡ Institut für Physik, Universität Rostock, Universitätsplatz 3, 18055 Rostock, Germany
§ Theoretische Chemie, PCI, Universität Heidelberg, Im Neuenheimer Feld 229, 69120 Heidelberg, Germany

Synopsis We report on the observation of a novel intracluster Coulombic decay process following Rydberg atom formation in clusters ionized by intense near-infrared fields. A new decay channel emerges, in which a Rydberg atom relaxes to the ground state by transferring its excess energy to a weakly bound electron in the environment that is emitted from the cluster. We find evidence for this process in the electron spectra, where a peak close to the corresponding atomic ionization potential is observed. For Ar clusters, a decay time of 87 ps is measured, which is significantly longer than in previous time-resolved studies of interatomic Coulombic decay.

When an excited atom or molecule is located in an environment, novel decay channels can emerge that are absent for isolated particles. Relaxation of the excited species can take place by a transfer of its excess energy to an atom or molecule in the environment that gets ionized, a process known as interatomic (intermolecular) Coulombic decay [1]. Thus far, the observations of such energy transfer mechanisms have been limited to dimers [2] and clusters [3] ionized by extreme-ultraviolet (XUV) pulses.

Here we present highly surprising results on a novel intracluster Coulombic decay (ICD) process that is of universal nature, as it is not restricted to a certain system or wavelength regime. In the experiment, we observe ICD following electron-ion recombination and extensive Rydberg atom formation in clusters interacting with intense near-infrared (NIR) laser pulses [4, 5]. When a highly excited Rydberg atom decays by transferring its excess energy to a second Rydberg atom in the environment [6], an electron is emitted whose initial kinetic energy is close to the atomic ionization potential. In Fig. 1, an electron spectrum from Ar clusters ionized by intense NIR pulses shows evidence for this process, where in addition to a thermal distribution, a peak is visible just below the ionization potential of Ar that can clearly be attributed to ICD. The novel ICD mechanism was observed in all systems under investigation, including Ar, Kr, Xe, O_2 and CH_4 clusters. We have further measured the decay time of the ICD contribution as 87 ps [7], which is significantly longer than the decay time of 150 fs recently reported for ICD in Ne dimers ionized by XUV pulses [8].

Intracluster Coulombic decay is expected to be important in a large variety of systems, including biomolecules interacting with intense X-ray pulses. The novel findings offer unprecedented opportunities in the future for an improved investigation of correlated electronic decay processes.

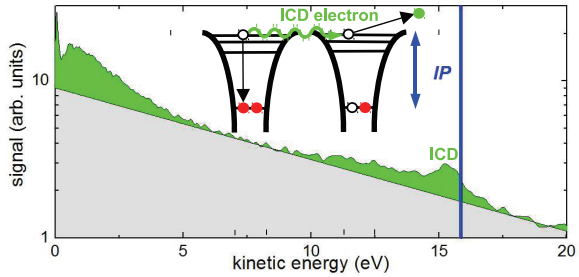

Figure 1. Electron kinetic energy spectrum from Ar clusters with an average size of $\langle N \rangle = 1000$ atoms ionized by NIR pulses at an intensity of 9×10^{13} W/cm^2. The peak just below the ionization potential of Ar (15.76 eV) is attributed to ICD between two Rydberg atoms (see inset). The gray area represents thermal electron emission.

References

[1] L. S. Cederbaum *et al* 1997 *Phys. Rev. Lett.* **79** 4778
[2] T. Jahnke *et al* 2004 *Phys. Rev. Lett.* **93** 163401
[3] S. Marburger *et al* 2003 *Phys. Rev. Lett.* **90** 203401
[4] B. Schütte *et al* 2014 *Phys. Rev. Lett.* **112** 073003
[5] B. Schütte *et al* 2014 *Phys. Rev. Lett.* **112** 253401
[6] A. I. Kuleff *et al* 2010 *Phys. Rev. Lett.* **105** 043004
[7] B. Schütte *et al submitted*
[8] K. Schnorr *et al* 2013 *Phys. Rev. Lett.* **111** 093402

[1] E-mail: schuette@mbi-berlin.de