Investigation of environmental effects in prototypical noble gas clusters using fluorescence spectroscopy

Andreas Hans*[1], Xaver Holzapfel*, Philipp Schmidt*, Christian Ozga*, Uwe Hergenhahn†, Till Jahnke‡, Reinhard Dörner‡, Arno Ehresmann*, and André Knie*

* Institut für Physik und CINSaT, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany
† Leibniz-Institut für Oberfächenmodifzierung e. V., Permoserstr. 15, 04318 Leipzig, Germany
‡ Institut für Kernphysik, J. W. Goethe-Universität, Max-von-Laue-Str. 1, 60438 Frankfurt am Main, Germany

Synopsis The investigation of environmental effects in weakly bound clusters has evolved as a comprehensive field in atomic physics during the last two decades. Recently it was shown that fluorescence spectroscopy can be utilized as complementary tool to track such processes. Here, we report on progress in fluorescence spectroscopy experiments on prototypical noble gas clusters.

Recently, it was demonstrated, that non-local relaxation processes in noble gas clusters can be identified and quantitatively characterized by fluorescence measurements [1, 2]. The detection of photons therefore provides an advantageous method for the investigation of environmental effects in weakly bound systems. Using electron and ion spectroscopy, often in coincidence experiments, numerous variants of interatomic Coulombic decay (ICD), electron transfer mediated decay (ETMD) and radiative charge transfer (RCT) were discovered and extensively investigated [3]. Noble gas clusters are regarded as prototype systems for the investigation of these process, because they can easily be produced in a controlled way.

Here, we report on the progress and perspectives on fluorescence measurements concerning these environmental effects. In a proof-of-principle experiment, we show that a resonant variant of ICD triggered by resonant 2s-*n*p photoexcitation of neon clusters leads to characteristic fluorescence emission in the vacuum-ultraviolet (VUV) spectral range, which is absent in isolated atoms [1]. This is the first unambiguous observation of an ICD process using photon detection. The complete decay cascade of this path is revealed by dispersed VUV photon detection and simultaneous measurement in the ultraviolet/visible spectral range [4].

Using atomic benchmarks in a partially condensed jet, the cross section of the VUV emission is calibrated on an absolute scale [2]. No absolute cross section of any ICD process following photon absorption was reported before. Complementary to the population of radiative states after resonant excitation, ICD can quench atomic radiative decay, e.g. after neon 2s ionization. This effect is observed by comparing ion yield and photon yield in the vicinity of the neon 2s edge [4].

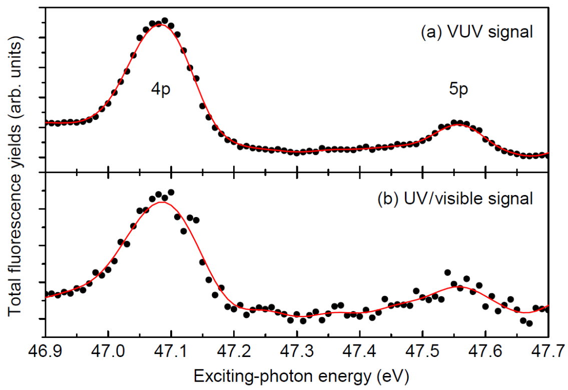

Figure 1. VUV and UV/visible fluorescence yield after 2s-*n*p photoexcitation of neon clusters. [4]

Time resolved photon detection, enabled by the bunched nature of the exciting synchrotron radiation, reveales significant differences in the lifetime of radiative states in clusters compared with monomers [1]. For various species it is shown that signals from both condensed and non-condensed parts of a typical experimental cluster jet can be discriminated very efficiently by proper time gating. This is particularly useful, if excitation energy and/or emission wavelength overlap and cannot be separated by other means.

References

[1] A. Knie *et al.* 2014 *New J. Phys.* **16** 102002

[2] A. Hans *et al.* 2016 *J. Phys. B* **49** 105101

[3] U. Hergenhahn 2011 *J. Phys. B* **184** 78

[4] A. Hans *et al.* 2017 *Chem. Phys.* **482** 165

[1]E-mail: hans@physik.uni-kassel.de