Recombination-induced autoionization process in rare-gas clusters

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Abstract: We investigate electron-ion recombination to excited states in atomic clusters exposed to intense NIR and XUV pulses, which leads to a yet undiscovered autoionization mechanism as a consequence of multiple recombination processes.

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1. Introduction

In the last two decades, a large interest was devoted to the investigation of the ionization mechanisms in clusters exposed to an ultrashort, intense light pulse in the near-infared (NIR) or extreme-ultraviolet (XUV) region. While at the early stage of the interaction with the light pulse, the ionization mechanisms is very similar in atoms and clusters, i.e. multiphoton ionization processes in the NIR regime and sequential single photon ionization for a XUV pulse, it was demonstrated that new electron emission mechanisms are observed in clusters at a later stage of the interaction. Indeed, as the electrons are stripped away by the laser field and the cluster is being charged, the increased Coulomb potential prevents other electrons from leaving the cluster. These quasifree electrons can move within the cluster and form a nanoplasma with the ions. A fraction of the electrons from the nanoplasma can eventually leave the cluster by evaporative emission, a process that only occurs in clusters. In addition, in the case of an NIR pulse, field ionization can play an important role where energy is transferred from the laser pulse to quasifree electrons in the nanoplasma, enabling them to drive electron impact ionization.

During the cluster expansion, quasifree electrons and ions in the nanoplasma may recombine. Here we demonstrate the recombination to high-lying atomic Rydberg states that are reionized by the DC detector electric field, a process known as frustrated recombination [1]. Recently, we could give experimental evidence for this effect by the detection of the corresponding electrons that have very low kinetic energies (meV to few tens of meV) [2]. In addition, we present time-resolved studies of recombination showing the formation of low-lying excited states. By using a weak 790 nm probe pulse following an intense XUV or NIR pump pulse, reionization of excited atoms from recombination (REAR) leads to additional peaks in the photoelectron spectra. The overall ion yield is significantly enhanced by REAR even for large delays in the ns range, demonstrating the significance of electron-ion recombination processes. Also, as a consequence of multiple recombination processes, new autoionization processes in the cluster can take place, where one electron relaxes to the ground state, transferring the energy to a second electron that is emitted into the continuum. Here we show evidence for such an autoionization mechanism.

2. Experiment

A Ti:sapphire laser system delivering 35 mJ, 790 nm, 32 fs pulses at a repetition rate of 50 Hz is used for the experiment. XUV pulses are obtained by high-order-harmonic generation (HHG) employing a loose-focusing geometry. Part of the NIR beam is focused by a $f = 5$ m spherical mirror into a 15 cm long gas cell that is statically filled with Ar. After blocking the residual NIR light with a 200 nm thin Al filter, the HHG pulses are spectrally filtered and focused by a spherical multilayer mirror with a focal length of 75 mm into a velocity map imaging spectrometer, where it is crossed by a pulsed cluster beam. XUV peak intensities up to 5×10^{12} W/cm^2 are achieved. The generated ions and electrons are accelerated towards a microchannel plate / phosphor screen assembly, and the 2D momentum maps are recorded with a CCD camera. A small fraction of the 790 nm beam is split before the harmonic generation, and is later recombined with the harmonic beam to perform pump-probe experiments. Upon removal of the Al filter in the XUV path, it is also possible to perform NIR-pump NIR-probe experiments.

3. Results and discussion

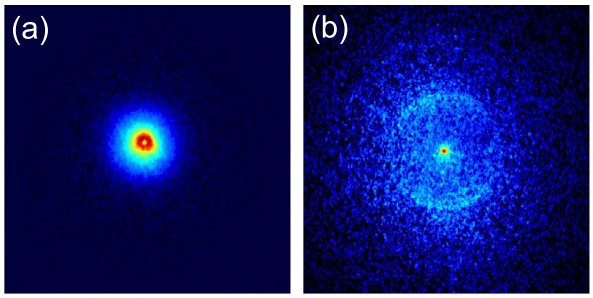

Fig. 1. (a) 2D Kr^+ ion momentum map from Kr clusters with an average size of $\langle N \rangle$=8000 exposed to a 38 nm pump pulse with an intensity of 2×10^{12} W/cm^2 and a 790 nm probe pulse at a delay of 5.2 ns with the same intensity. The signal from the XUV pulse alone is substracted. (b) Corresponding 2D electron momentum map showing ring structures attributed to reionization of excited atoms from recombination.

REAR is observed in the photoion and -electron momentum maps presented in Fig. 1, where the differences between the two-color XUV+NIR signals at a time delay of 5.2 ns and the yields obtained by the XUV pump pulse alone are displayed. In Fig. 1(a), an additional ion contribution due to the NIR pulse is visible, with the ions having non-zero kinetic energies that are therefore identified as fragments formed during the cluster dissociation. Before recombination takes place, these fragments gain kinetic energy that is preserved until their reionization. In the electron momentum maps in Fig. 1(b), clear ring structures are observed and attributed to single-photon ionization events of excited atoms formed by recombination in the nanoplasma with the weak NIR pulse. Nanoplasma formation is not limited to XUV ionization regimes, and therefore, a large number of recombination events is also observed when using an intense NIR pulse for cluster ionization.

Although the NIR probe pulse is too weak to ionize a neutral cluster, it generates a large amount of ions and electrons from REAR that can even exceed the number of ions and electrons produced by the XUV pulse only. This shows that many excited atoms are formed during the cluster expansion. Due to the large number of excited atoms with weakly bound electrons, a novel autoionization mechanism becomes possible, where one electron relaxes to the ground state of an atom, transferring the energy to a second electron, which can leave the cluster with a substantial amount of kinetic energy. These two correlating electrons can either be located in one atom leading to autoionization, or in two atoms, similar to interatomic coulombic decay. The latter case was theoretically predicted in clusters, where excited atoms were formed by resonant laser excitation [3]. In an experiment after ionization of Ar clusters with strong NIR pulses, we observe a characteristic peak in the photoelectron spectrum close to the ionization potential of atomic Ar that is attributed to the autoionization following electron-ion recombination processes. In opposite to the studies carried out in [3], the autoionization mechanism discussed here does not involve resonant excitation, but is a consequence of electron-ion recombination in the nanoplasma. Therefore, it can be regarded as a general process that is independent from the ionization wavelength and is possible at photon energies far below the ionization potential. The current findings may be important towards an improved understanding of the ionization dynamics in clusters and other extended systems.

References

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