

Double excitation of helium initiates interatomic Coulombic decay in helium nanodroplets

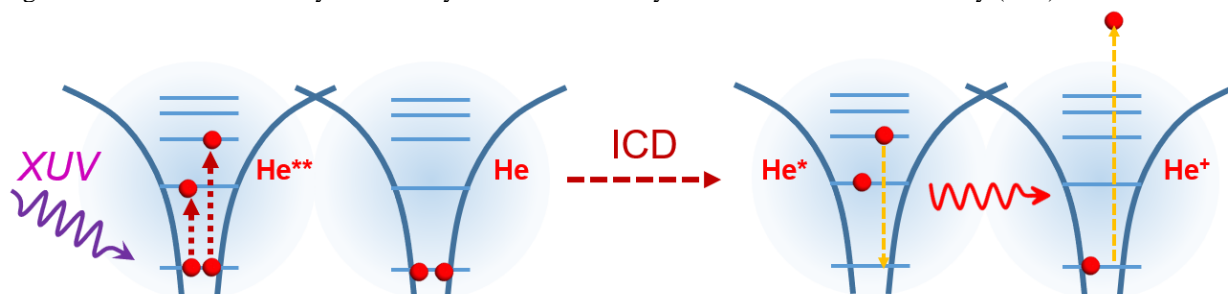
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Interatomic Coulombic decay (ICD) involving a doubly excited atom is observed for the first time. ICD is a non-local correlated electronic process which plays an important role in weakly bound complexes exposed to high-energy radiation. We use liquid helium nanodroplets (HNDs) as a well-suited model system and apply two methods for high-resolution electron spectroscopy in the extreme-ultraviolet (EUV) regime. This allows us to quantify the ICD probability and to analyze details of the process.¹

Slow electrons from inelastic scattering of photoelectrons in the nanodroplet have the same energy as the ICD electrons. This experimental challenge is overcome by fine energy scans across the $2s2p+$ resonance. We identify the ICD contribution that is shown to proceed by relaxation into $\text{He}^* \text{He}^+$ atom-pair states. The process competes with the ultrafast autoionization of doubly excited He^{**} (17.5 fs decay time) and is expected to become more efficient in denser media. This type of ICD could consequently be relevant in other systems such as water and solvated metal atoms.

Fig 1. Schematics of the decay of a doubly excited He state by interatomic Coulombic decay (ICD)



References:

1. B. Bastian, J. D. Asmussen, L. Ben Ltaief, H. B. Pedersen, K. Sishodia, S. De, S. R. Krishnan, C. Medina, N. Pal, R. Richter, N. Sisourat, and M. Mudrich, *Phys. Rev. Lett.* (2024), accepted on April 25.