

Tracing Interatomic Coulombic Decay in Ne₂ via XUV pump-probe Experiments at FLASH

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We present the first time-resolved measurement of Interatomic Coulombic Decay (ICD) in Ne₂ using a XUV pump-probe scheme at the free-electron laser (FEL) in Hamburg (FLASH). ICD is an ultra-fast radiationless decay mechanism where one atom relaxes via energy transfer to a weakly-bound neighbouring atom, which then emits an electron.

While a 2s vacancy in a single Ne⁺-ion can only decay via slow (\approx nanoseconds) radiative decay, the Ne₂⁺-dimer has a femtosecond relaxation pathway: ICD [1]. There, the relaxation energy of the inner-valence hole is transferred via virtual photon exchange to a 2p electron of the second neutral neon atom, which is then ionized. This process was first predicted theoretically [1], and confirmed experimentally in rare-gas- [2] and water clusters [3]. Although the exact decay time of ICD is a crucial parameter for understanding the underlying mechanism, no time-resolved investigation has been performed yet.

In our experiment the incoming FEL pulse of 58 eV with a pulse duration of \approx 60 fs is guided onto a split-and-delay mirror set-up, which creates two identical copies of the pulse with adjustable time delay and focusses them into a dilute gas target. By means of a Reaction Microscope (REMI) the kinetic energy release (KER) of each ion-ion coincidence channel is recorded as a function of the delay allowing us to trace the molecular dynamics.

The pump pulse prepares the intermediate 2s inner-valence vacancy (Ne – Ne⁺(2s⁻¹)). The system relaxes via ICD leading to a repulsive (Ne⁺(2p⁻¹) – Ne⁺(2p⁻¹)) state, which is then further ionized by the probe pulse into a repulsive (Ne⁺ – Ne²⁺) state. By measuring the yield of the coincident (Ne⁺ – Ne²⁺) pairs as a function of the delay we extract the ICD lifetime of (150 \pm 50) fs.

Classical model calculations are used to identify all possible competing pathways that lead to the creation of (Ne⁺ – Ne²⁺) pairs. Additionally, the measured ICD lifetime is compared with recent quantum-mechanical calculations.

References

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