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Molecular-frame angular distributions of spectator resonant interatomic coulombic decay electrons in neon dimers

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Synopsis The spectator resonant interatomic Coulombic decay (SRICD) following the $2s \longrightarrow 5p$ excitation of Ne dimers has been examined experimentally and theoretically. The molecular frame angular distributions (MFADs) of the SRICD electrons depend strongly on the direction of the polarization vector of the exciting photon with respect to the molecular axis. We demonstrate that this effect is caused by the scattering of the low-energy SRICD electron on the density of the spectator electron.

It is widely accepted that the creation of a core hole by photoionization and its subsequent Auger decay can be described, in a good approximation, independently. This approximation is known as the two-step model [1], in which the electron emitted by the Auger decay is not influenced by the initial excitation or ionization process [2]. For instance, within the two-step model, the MFADs of the Auger electrons are independent of the polarization properties of the ionizing/exciting photons. For a resonant Auger decay, the two-step model holds if the excited electron only witnesses the decay process as a spectator.

Here, we demonstrate that, for low-energy electrons, the generally-admitted two-step approximation breaks down. For this purpose, we performed a joint experimental and theoretical study of SRICD after inner-valence excitation of the neon dimer into the 5p state. Experimentally, the momenta of all charged particles created after the absorption of the photon and subsequent SRICD were coincidently measured using the COLTRIMS technique [3]. We observe a strong dependency of the SRICD electron angular emission distribution on the polarization direction of the exciting photons with respect to the dimer axis (cf. Fig. 1b and 1c), which is in contradiction with the two-step model.

*E-mail: abir.mhamdi@uni-kassel.de †E-mail: demekhin@physik.uni-kassel.de For a deeper understanding of these observations, we performed electronic structure and dynamics calculations by employing the Single Center method [4]. The present observations are explained by the substantial scattering of the escaping low-energy SRICD electron on the density of the excited spectator electron [5] (see Fig. 1a).

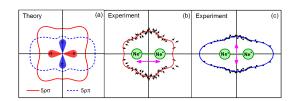


Figure 1. MFADs of SRICD electrons after $2s \longrightarrow 5p$ excitation. Panel (a): Theoretical MFADs for $5p\sigma$ and $5p\pi$ excitations of Ne₂. Panels (b) and (c): Corresponding experimental results, where the polarization direction of the exciting light is oriented in parallel or perpendicularly to the dimer axis, as indicated by the double arrows.

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