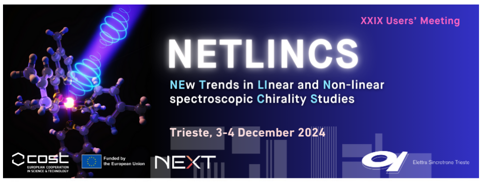

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Probing the vibronically coupled decay of intermediate states with multiphoton PECD: Theory and Experiment

Vibrationally resolved picosecond laser (2+1) multiphoton ionization of fenchone, pumped via a partially resolved manifold of Rydberg states, indicates strong vibronic branching in the decay channels with extensive vibrational redistribution. Associated to the vibronic structures are dramatically fluctuating photoelectron circular dichroism (PECD) chiral asymmetries.

The strong sensitivity of PECD to structure, conformation, and orbital localisation is well established, and dramatic vibrational effects have previously also been reported.

A theoretical MP-PECD model has been developed that combines calculation of the two-photon transition tensors for the resonant intermediate states with a reliable modelling of the subsequent one-photon PECD. Applying established models for ground state molecular photoionization demands special consideration when extended to treat ionization of Rydberg states as their greater spatial extent emphasises self-interaction errors inherent in a one-electron scattering potential.

Comparison of this model with selected experimental results permits the identity of the intermediate state and coupled states connecting with a final decay channel to be inferred. The experimental and theoretical results, and their comparison, fully demonstrate the power of PECD as a probe of molecular dynamics.

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