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Ultrafast Dynamics And Electronic State - Lifetime Interferences In Chlorine-Containing Molecules

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Synopsis In order to have access to fine phenomena such as ultrafast nuclear dynamics and electronic state - lifetime interferences (ELI) at high energies (few keV), one has to rely on great experimental resolution. The new experimental HAXPES endstation at the GALAXIES beamline of the French synchrotron SOLEIL provides photons in the 2-12 keV energy range along with a total instrumental resolution below 500 meV. In this work we show experimental evidences of ultrafast nuclear dynamics and ELI on HCl and CH_3Cl excited around the Cl 1s resonance. We show also simulations that allow to disentangle the contribution of nuclear dynamics and ELI in our experimental spectra.

Ultra-fast photodissociation in chlorine-containing molecules has been known for more than twenty years [1,2].

While the first observations were made after photoexcitation of the Cl L shell, the study of nuclear motion in these molecules has been recently extended to deep shells by means of Resonant Inelastic X-ray Scattering (RIXS) in the tender x-ray region (2–12 keV) [3,4,5].

A new experimental station [6] dedicated to HArd X-ray PhotoElectron Spectroscopy (HAXPES), has been mounted on the GALAXIES beamline of the synchrotron SOLEIL.

Using this setup, we have studied the electronic relaxation of HCl and CH_3Cl [7] after Cl 1s excitation under resonant Raman conditions, i.e. with total instrumental resolution below the natural line width of the decay features.

In particular, we were able to observe nuclear dynamics on the 1-femtosecond time scale using the 'core-hole clock approach' [8].

These measurements also allowed us to unravel subtle phenomena as electronic state-lifetime interferences, which were also observed in RIXS [9] and in Argon using HAXPES [10]. We present a 'simple model' simulation, initially based on a theoretical model [11], which reproduces well the electronic state-lifetime interferences in both HCl and CH_3Cl.

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