Molecular fingerprinting of f-electron molecules using a soft x-ray, HHG light Source

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While bonding and electronic structure are well understood for most elements in the periodic table, basic unanswered questions remain for lanthanides and actinides containing f-electrons, which do not always follow clear trends due to a large number of electronic states and competing potentials. X-ray spectroscopy, typically done at synchrotrons, has proven to be a powerful tool for measuring electronic structure in f-electron systems and for distinguishing subtly different molecules. Recently, tabletop light sources based on high harmonic generation (HHG) driven by femtosecond lasers, which produce extreme ultraviolet to soft x-ray femtosecond pulses, have emerged as a complementary tool for x-ray absorption spectroscopy. Here we describe a table-top, soft x-ray light source based on High Harmonic Generation (HHG) for static and potentially time-resolved spectroscopic investigations of f-electron systems and we demonstrate how x-ray absorption with this light source leads to a detailed molecular fingerprint. Our system generates photon energies from 150 eV to 330 eV, making it well-suited for studying lanthanide molecules from the perspective of multiple elemental transitions within the molecule, the lanthanide N-edge, the chlorine L-edge and the carbon K-edge. Here we will show our recent results applying to distinguish different lanthanide molecules based on their electronic structures.

Keywords [optional]: Organic semiconductors, electronic excitations, machine learning, density functional theory, materials discovery..

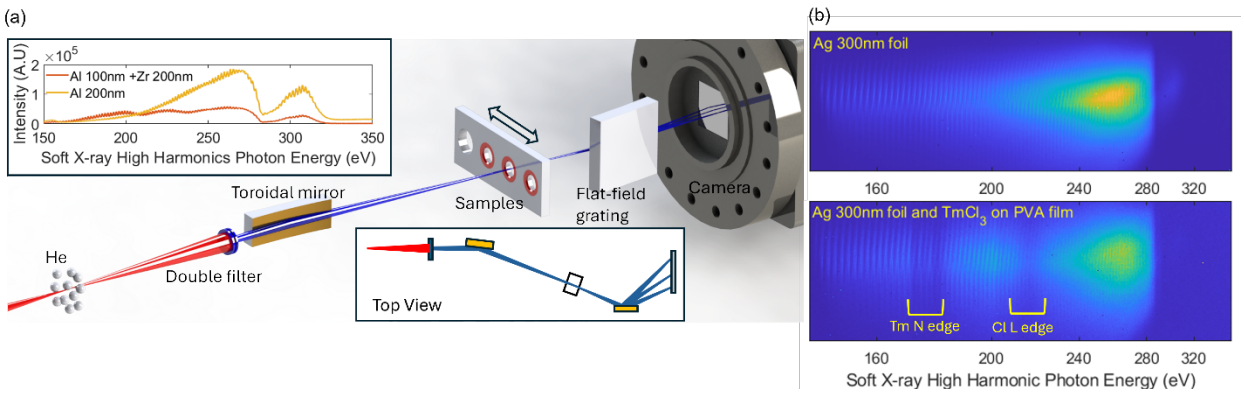

Fig. 1 A broadband HHG-based soft x-ray spectrometer for characterizing f-electron molecules: (a) Soft x-rays are produced by HHG of ~2.2 mJ, 40 fs, 1 kHz pulses at 1400 nm in a 10-mm helium-filled gas cell. Residual IR laser light is filtered out by two metal filters (double filter), and the soft x-rays are focused onto the sample using a toroidal mirror (TM). A flat-field grating (2400 lines/mm) is employed to diffract and refocus the beam onto the CCD camera. The top inset displays the soft x-ray spectrum, while the bottom inset provides a top-view schematic of the experimental setup. (b) Camera image measured (top) without and (bottom) with a $TmCl_3$ thin film sample.