Tender X-ray Emission Spectroscopies in Liquid Systems: New Capabilities with MOSARIX Spectrometer

Iyas ISMAIL

Laboratoire de Chimie Physique-Matière et Rayonnement, CNRS, UMR 7614, Sorbonne Université, 4 place Jussieu, 75252 Paris, France

ABSTRACT

Resonant inelastic X-ray scattering (RIXS) is a powerful technique for probing electronic structure, charge-transfer processes, and ultrafast dynamics with elemental specificity. While RIXS is well established in the soft- and hard-X-ray regimes, its extension to the tender X-ray range (2–5 keV) has remained experimentally challenging. This energy range is of particular interest because it provides access to the K edges of light elements (e.g., P, S, Cl, K, Ca) and the L edges of heavier elements (e.g., iodine). Moreover, tender X-rays offer increased bulk sensitivity compared to soft X-rays, which is advantageous for studies of liquids and complex environments.

We demonstrate that tender X-ray RIXS and X-ray emission spectroscopy are now experimentally feasible using the MOSARIX, our multi-crystal spectrometer installed at the GALAXIES beamline of the SOLEIL synchrotron. MOSARIX employs nine HAPG crystals arranged in a von Hamos geometry, combining large solid angle, high reflectivity, and high energy resolution[1]. This configuration provides unprecedented efficiency, enabling the detection of weak signals and operando measurements. Operation under ambient or helium atmosphere allows windowless detection and compatibility with liquid jets and in situ sample environments.

As a first application, we investigate radiative intermolecular decay (IRD) in aqueous solutions. In IRD, a core hole created on a selectively excited ion is filled by an electron from the surrounding solvent or ligands, resulting in X-ray emission. For aqueous Ca ions, we show that the IRD signal is sensitive to changes in the local chemical environment, including solvation structure, ion pairing, and chemical speciation. RIXS measurements across the Ca K edge provide resonant enhancement of the IRD signal, yielding additional sensitivity to ion–ligand hybridization and ultrafast electron delocalization.

We further present tender X-ray RIXS measurements of aqueous NaI at the iodine L_2 edge as a model system for ultrafast charge-transfer dynamics. The resulting RIXS maps, supported by theoretical calculations, reveal excitation into both localized and delocalized electronic states on timescales shorter than approximately 200 attoseconds.

These results demonstrate that recent instrumental advances establish tender X-ray RIXS as a robust and versatile tool, opening new opportunities for investigating electronic structure and dynamics in liquids and complex systems.

REFERENCES

1. I. Ismail et al., Rev. Sci. Instrum., 95, 53103 (2024)