



12th December 2018 - 2:00 p.m.
CFEL-bldg. 99, seminar room IV

Michael Odelius

Department of Physics, Stockholm University, AlbaNova University Center,
Stockholm, Sweden

Dynamical information in static and time-resolved core-level spectra resolved with multi-configurational quantum chemistry

X-ray spectroscopy has only recently been explored to probe of structure and dynamics in hydrogen bonding solutions. Since these probes are sensitive to structural and electronic degrees of freedom, it is natural to combine electronic structure calculations and molecular dynamics simulations to interpret the experimental data. In this presentation of a few case studies, I discuss how multi-configurational quantum chemistry (particularly RASPT2) complemented with density functional theory (DFT) can be used as a basis for quantum dynamical and molecular dynamics simulations of X-ray spectra. In particular, we investigated structural, electronic and dynamical information in both static and time-resolved resonant inelastic X-ray scattering (RIXS). Scalar relativistic RASPT2 calculations complemented with spin-orbit coupling allows for an accurate treatment of scattering against different core-excited states where the initial and final states of the RIXS process may include ground and valence-excited states for simple molecules and transition metal complexes.

In a series of studies of gas phase water, we have established a quantitative analysis of the excitation energy dependence of ultra-high resolution oxygen K-edge RIXS measurement. The framework is employed to investigate hydrogen bonding in liquid water. I will also discuss our contributions to rationalize time-resolved iron L-edge and nitrogen K-edge RIXS and get insight into valence excited state dynamics in solution. In particular, the interpretation of L-edge spectra is complicated and relies on accurate theoretical modeling and spectrum simulations to take into account effects of multiplet effects, spin-orbit coupling, chemical interactions, dynamics in the spectroscopic process. The multi-configurational restricted active space method (RASPT2) is an efficient approach which can target the states of interest also along chemical reaction, but it is current very limited by computation demands and further development is necessary for efficient interpretation of time-resolved X-ray spectra