Core-level non-resonant inelastic x-ray scattering: An extremely powerful method to determine the local ground state wave function

L.H. Tjeng*

Max Planck Institute for Chemical Physics of Solids
Nöthnitzerstrasse. 40, 01187 Dresden, Germany

hao.tjeng@cpfs.mpg.de

A prerequisite for a microscopic understanding of the physical properties of materials is the identification of quantum numbers that characterize, for example, the charge, spin, and orbital degrees of freedom of the atomic constituents. This is especially true for strongly correlated or narrow band systems. While invaluable insight has been obtained using a wide range of x-ray based spectroscopic techniques, most of these are based on dipolar electronic transitions and have as such limitations, e.g. asymmetries with higher than twofold rotational symmetry cannot be detected (unless it is accompanied by a sufficiently large energy difference).

Here we utilize the opportunities provided by a new technique, namely non-resonant x-ray scattering (NIXS). This photon-in-photon-out technique with hard x-rays has become feasible thanks to the high brilliance of modern synchrotrons and advanced instrumentation. The available large momentum transfers allows for the study of excitations that are well beyond the dipole limit. While so far most of the NIXS studies were carried out on powder samples, it becomes very clear that the directional dependence of the momentum transfer observed in experiments on single crystals has the potential to give a very detailed insight into the ground-state symmetry of the ion of interest. It is this very aspect, i.e. the direction dependence of the scattering function beyond the dipole limit, that we utilize for our study of strongly correlated systems for which the orbital symmetry remained elusive so far [1-6]. The interpretation of the spectra is straightforward and quantitative, facilitated also by the fact the multipolar excitations are more excitonic than the dipole ones. Our excitement about NIXS is further enhanced by the fact that this element specific technique is also bulk sensitive, requires only tiny samples not larger than 0.1 mm, and allows for more demanding sample environments. We now have also the first spectroscopy and imaging results from our own Max Planck inelastic scattering beamline at PETRA III in Hamburg, using analyzers and cryostat produced by the ESRF.

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