Core-level non-resonant inelastic X-ray scattering (NIXS): Beyond the dipole limit

A. Amorese[1], B. Leedahl[1], A. Severing[2], M. Sundermann[1,2,3], L.H. Tjeng[2]

[1]Max-Planck Institute for Chemical Physics of Solids, Dresden, Germany; [2]Institute of Physics II, University of Cologne, Germany; [3]DESY / Petra III Hamburg, Germany; **andrea.amorese@cpfs.mpg.de, brett.leedahl@cpfs.mpg.de , martin.sundermann@desy.de, severing@ph2.uni-koeln.de, hao.tjeng@cpfs.mpg.de**

We would like to show how wave functions of strongly correlated *f* and *d*-electron materials can be determined to a great detail with core-level non-resonant inelastic x-ray scattering (NIXS, x-ray Raman). The key issue here is to utilize transitions that go beyond the dipole limit, in the following called *multipole transitions* [1-4]. Multipole transitions can be achieved by carrying out the NIXS experiments at high momentum transfers, e.g. of the order of $|\vec{q}| \approx 10\text{Å}^{-1}$.

In the multipole transition limit, the vector $\vec{q}$ dependence of the scattering function $S(\vec{q},\omega)$ can reveal symmetries that are not accessible by experimental methods that are based on dipole selection rules. We will show how NIXS can determine the in-plane orientation of the *4f* wave function in tetragonal Ce compounds [5,6] and the crystal-field ground state in cubic CeB_6 [7] and SmB_6 [8], with the latter being considered as *the* candidate material for a strongly correlated topological insulator.

Another important aspect of using multipole transitions is that final states with higher quantum numbers can be reached that are not accessible in a dipole experiment. These high quantum number states tend to have lower energies, making them more excitonic and less mixed with the continuum states. The atomic features will thus show up more clearly. We utilize this effect in our study of U compounds, and we were able to identify the atomic like wave functions that make up the ground state of the often highly itinerant (and certainly highly covalent) U systems [9,10].

Finally, we have explored and developed a new experimental method that directly images the active local wave function in solids. This is achieved by measuring the angular distribution of the integrated intensity of the *s*-core-to-valence NIXS. We can directly map the orbitals when using the dipole forbidden $3s \rightarrow 3d$ (M_1-edge) transitions in 3*d* transition metal compounds [11]. There is no need to do spectroscopic analysis or calculations. This provides a purely experimental and thus unbiased determination of the orbitals that actively drive the ground-state properties in many modern day solid state materials.

References

[1] - R. A. Gordon *et al.*, EPL **81**, 26004 (2008).
[2] - J.A. Bradley *et al.*, PRB **81**, 193104 (2010).
[3] - R. Caciuffo *et al.*, PRB **81**, 195104 (2010).
[4] - R. A. Gordon *et al.*, J. Phys. Conf. Ser. **190**, 012047 (2009).
[5] - T. Willers *et al.*, PRL **109**, 046401 (2012).
[6] - J.-P. Rueff et al., PRB. **91**, 201108(**R**) (2015).
[7] - M. Sundermann, *et al*., EPL **117**, 17003 (2017).
[8] - M. Sundermann *et al*., PRL **120**, 016402 (2018).
[9] - M. Sunderman *et al*., Proc. Nat. Acad. Sci. **113** (49), 13989 (2016).
[10] - M. Sundermann *et al.* , PRB **98**, 205108 (2018).
[11] - H. Yavas *et al.*, accepted with Nat. Phys.