dd excitations in layered cuprates studied with high resolution resonant inelastic x-ray scattering at the Cu L_3 edge

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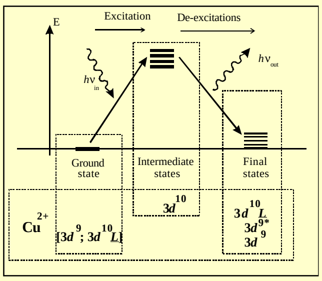
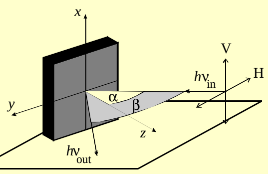
dd excitations

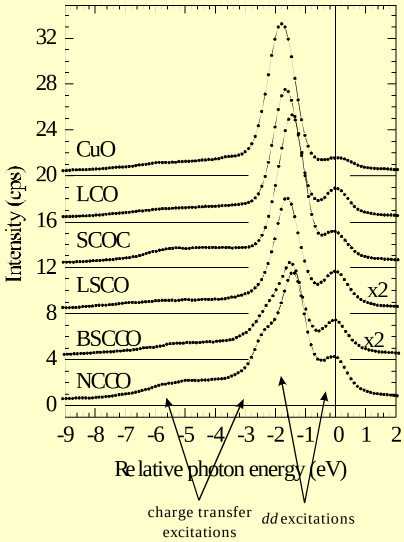
dd excitations in 3*d* transition metal compounds are the fingerprint of the electronic structure: they are influenced by electronic correlation, crystalline structure, chemical composition. And they are closely related to orbitons, charge / spin separation and other puzzling phenomena.

RIXS technique and experimental details

Resonant Inelastic X-ray Scattering is an ideal tool to study the neutral excitations in solids from several eV to (potentially) few meV. At the $L_{2,3}$ (or $M_{2,3}$) edges of 3*d* transition metals RIXS is made by two electric dipole (E1) transitions involving an incident photon (of given polarization and energy) and an emitted photon (of known direction and measurable energy). In this way the E1 forbidden *dd* excitations become accessible, in an element selective mode [1,2,3].
The main practical difficulties are energy resolution [4] and signal intensity [3], that have limited the application of RIXS to the case of *dd* excitations in the layered cuprates and high Tc superconductors (as suggested initially by Tanaka and Kotani [1]).
We have performed RIXS measurements on a variety of copper based compounds, with 0.8 eV combined energy resolution (at 931 eV), exploiting the recent improvements of our soft x-ray spectrometer (AXES [5], installed at the beam line ID08 of the ESRF, Grenoble). The samples were single crystals in most of the cases, allowing the exploration of the spectral shape dependence on the incident photon polarization and on the sample orientation. Each spectrum was measured in about 15 min.

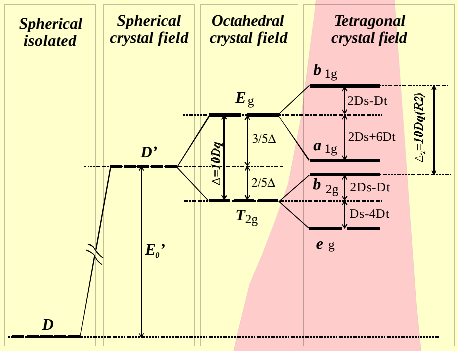


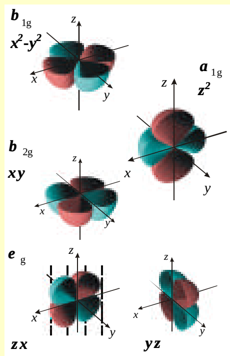


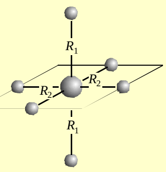


Crystal field model calculations

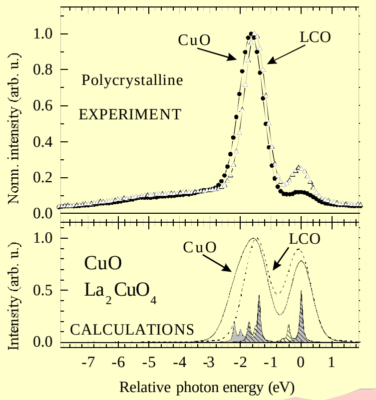
The spectra are characterized by an elastic / quasi-elastic peak, a main peak around -1.5 eV to -2.0 eV energy loss, and a tail extending to -7 eV. The features included between 0 and -2 eV are assigned to *dd* excitations, the tail is ascribed to charge transfer excitations [1,3]. The *dd* excitations can be calculated rather simply in a pure atomic model with crystal field [3]. Assuming a simple electronic structure for the Cu[2+] ground state (3*d*[9]) the transition probabilities are directly the single particle ones.
The final state energy levels are calculated with a tetragonally distorted octahedral point charge crystal field [6]. A super-exchange interaction is added afterwards (0.25 eV).
We find a set of crystal field parameters in good agreement with those already published [3]. The calculated spectra (after a 0.8 eV Gaussian broadening) are in good agreement with the experimental data. In particular they can account for the different spectral shapes of CuO and $LaCu_2O_4$, which are not compatible with the predictions made in 1993 with a cluster model calculation [1]. The elastic / *dd* peaks intensity ratio cannot be reproduced by the calculations due to the self-absorption effects decreasing the elastic part of the spectra.

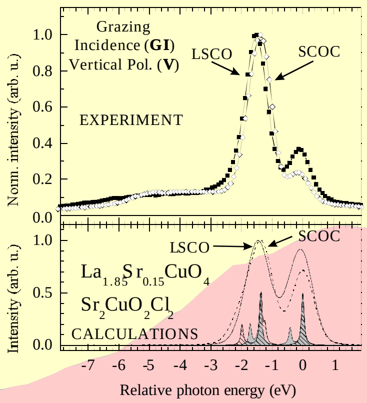


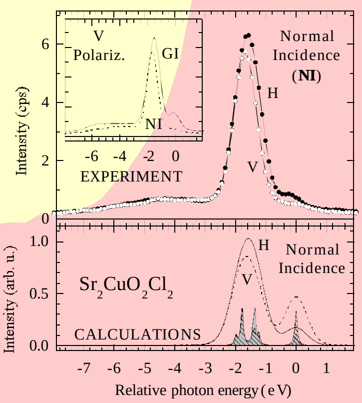




	CuO	LCO/LSCO	SCOC
$10Dq_{R2}$	1.38 eV	1.38 eV	1.36 eV
D_s	0.33 eV	0.08 eV	0.24 eV
D_t	0.79 eV	0.02 eV	0.06 eV
a_{1g} $[z^2]$	−1.73 eV	−0.41 eV	−1.23 eV
b_{2g} $[xy]$	−1.38 eV	−1.38 eV	−1.36 eV
e_g $[yz; zx]$	−1.99 eV	−1.51 eV	−1.78 eV







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Conclusions

dd excitations in the cuprates can be studied working at the L_3 edge of Cu: differences in RIXS spectra from sample to sample are seen already with 0.8 eV resolution and will be even clearer after the foreseen technical improvements of AXES. Those differences can be largely accounted for with a crystal field atomic model calculation.