

Internship and thesis proposal

Laboratory name: **LPS** (UMR 8502) and Synchrotron SOLEIL (GALAXIES beamline)

Internship/thesis director: **Victor BALÉDENT**

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Internship location: **Laboratoire de Physique des Solides, Université Paris-Saclay, Orsay, and SOLEIL, Saint-Aubin**

Thesis possibility after internship: **YES** Funding: **Doctoral school, ANR or Synchrotron SOLEIL**

Imaging orbitals in quantum materials

Why orbitals?

In strongly correlated electron systems such as transition-metal oxides, charge, spin and orbital degrees of freedom are intimately coupled. The **shape and occupation of the atomic orbitals** set the symmetry of the system, the hybridization with the ligands and the strength of electronic correlations; they therefore govern doping behaviour, transition temperatures and, ultimately, **all key properties: superconductivity, magnetism, metal-insulator transitions, electronic order**. Orbitals are central to the most active topics of condensed matter, and above all to unconventional superconductors. In **cuprates**, the CuO_2 plane long described by a single band is now recognised as multi-orbital, with a significant role of oxygen orbitals. In **iron pnictides**, nematicity arises from a delicate balance between the occupations and correlations of the d_{xz}/d_{yz} orbitals. In **nickelates**, the relative occupation of the $d_{x^2-y^2}$ and d_{z^2} orbitals is a central open question. In all these families, doping or pressure modifies hybridization and thus the orbital occupation, with a direct impact on superconductivity. Yet the ground state is still debated, because the orbitals involved have never been observed directly.

Experimental access to orbitals and electronic structure relies mainly on:

- ARPES; transport and thermodynamic measurements; optical spectroscopies; neutron scattering;
- X-ray spectroscopies (XAS, RIXS); Compton scattering and core differential Fourier synthesis.

These techniques share common limitations: they are **surface-sensitive** (ARPES) or **macroscopic and indirect** (transport, optics); they are **not selective of the few electrons and orbitals that matter** for the physics; and the orbital information is obtained only through **model-dependent analysis or complex data reconstruction**, which can lead to contradictory conclusions (as in cuprates). Several of them are also hardly compatible with extreme sample environments such as high pressure. A bulk-sensitive and direct probe of orbitals is therefore needed.

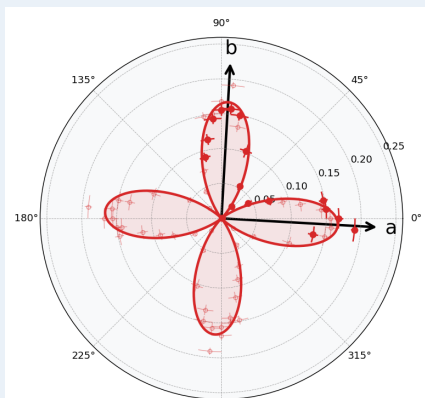


Fig. 1 | Orbital imaging in a cuprate. Cu M_1 ($3s \rightarrow 3d$) XRS intensity of LSCO at 300 K as a function of the direction of the momentum transfer $\mathbf{q}$ in the ab plane (full 360°). Filled points: data; open points: 4-fold symmetrized data; line: fit showing the $d_{x^2-y^2}$ orbital.

Measured by our team on GALAXIES @ SOLEIL.

A new probe: orbital imaging by X-ray Raman scattering (XRS)

XRS uses hard X-rays to excite shallow core levels (here the $3s \rightarrow 3d$ M_1 edge) through inelastic scattering at high momentum transfer, where quadrupolar transitions couple directly to the empty d orbitals. It is therefore **bulk-sensitive** and compatible with complex sample environments. By measuring the momentum-transfer dependence of the signal while rotating the sample, with a multi-analyzer spectrometer (Fig. 2), we obtain **orbital selectivity** (element and orbitals of interest) and a direct **real-space image of the orbital shape**, hence of its occupation. Changes of the electronic structure and the active orbitals responsible for a given property can thus be identified **directly, with minimal model assumptions**. Our team, at the Laboratoire de Physique des Solides and the SOLEIL synchrotron, is **one of only two in the world** able to perform and analyse such measurements, and has already imaged orbitals in nickelates, cuprates, vanadates and sulfides such as BaCoS_2 (Figs. 1–3).

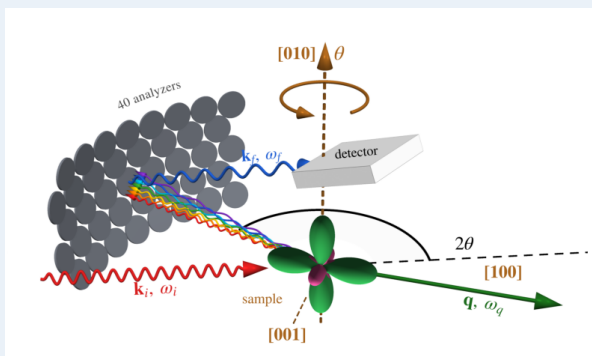


Fig. 2 | Experiment. The sample is rotated about θ while 40 crystal analyzers collect the inelastically scattered X-rays at momentum transfer $\mathbf{q}$, giving the orbital shape in real space (GALAXIES @ SOLEIL).

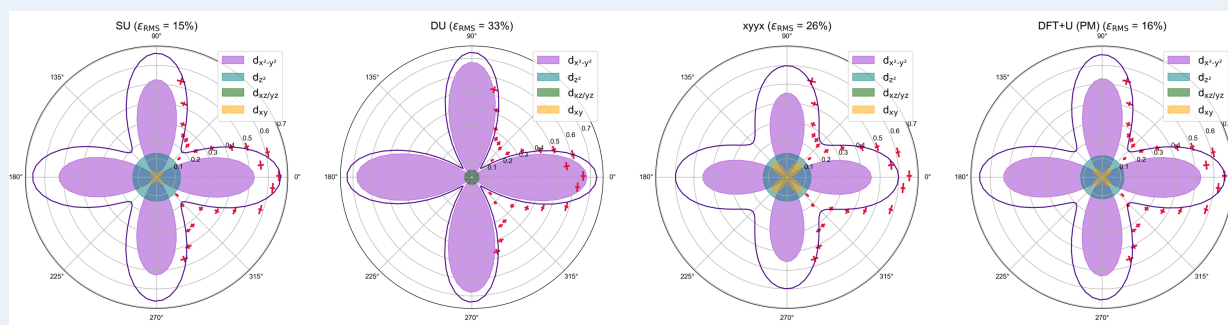


Fig. 3 | Orbital imaging in BaCoS₂ and comparison with theory. Co M₁ XRS hole-orbital angular profile (symbols), measured by our team on GALAXIES @ SOLEIL, compared with DFT+DMFT calculations with static U (SU) and dynamic U (DU), the orbital-ordered xyxx solution and paramagnetic DFT+ U (lines; colored areas: individual d -orbital contributions; ϵ_{RMS} : deviation from the data). Collaboration with the theorists of IMPMC (M. Casula and B. Lenz).

Scientific objectives

Unconventional superconductivity

Which orbitals ($d_{x^2-y^2}$ vs d_{z^2} , transition-metal vs ligand) are active in cuprates, iron-based pnictides and the new nickelate superconductors, as a function of doping, temperature and pressure?

Mott transitions

Orbital reconstruction and occupation changes across the metal-insulator transition, e.g. in (Cr-doped) V₂O₃ or BaCo_{1-x}Ni_xS₂.

Nematicity and orbital ordering

Direct imaging of orbital order and of the rotational-symmetry breaking of the orbital occupation (electronic nematicity, e.g. d_{xz}/d_{yz} in iron-based compounds).

Spin-orbit coupling

Orbital mixing and its evolution in strongly spin-orbit-coupled systems.

The project

The student will take part in synchrotron XRS experiments, develop the Python analysis of multi-analyzer image data (NeXus files) and extract orbital images and occupations, in comparison with ab initio (DMFT) calculations carried out in collaboration with the theorists of IMPMC, M. Casula and B. Lenz. A background in solid-state physics and an interest in experiments and data analysis are expected.

- **Internship:** analysis of existing XRS data sets (cuprates, nickelates, vanadates), orbital fits and first comparison with theory, with participation in a beamtime at SOLEIL.
- **Thesis:** extension to unconventional superconductors under extreme conditions (low temperature, pressure, uniaxial strain), development of the corresponding sample environments, and quantitative comparison of orbital occupations with (cluster) DMFT.