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Strong correlations
Engineering with twisted graphene platforms

Carbon is a special element: it exists as diamond, the hardest natural material, but also as graphite, which is fragile enough to slide from pencil tip to paper with minimal pressure. Graphite comprises stacks of single layers of carbon atoms, and each sheet, known as graphene, has exceptional properties [1]. When two sheets are twisted relative to each other, a plethora of phases of matter arises [2,3]. Finding such wide-ranging phenomena in a single elemental material is surprising, because such complexity is usually reserved for systems with complicated structures and composition. In this talk, I aim to discuss two proposals for engineered electronic states in twisted graphene platforms.

In the first example, I would like to discuss triple point fermions, electronic excitations that generalize Dirac and Weyl modes beyond the conventional high-energy paradigm [4]. Graphene is known for being an excellent platform to explore Dirac fermion phenomena, with two spin-degenerate Dirac cones at each K point. Engineering triple point fermions out of graphene by lifting the degeneracy of the Dirac points would require eliminating key symmetries, which usually leads to a shift of the cones or to a gapped spectrum. Introducing an additional level of complexity, twisted graphene multilayers have the potential to enlarge the degeneracy of the Dirac cones, providing new routes for degeneracy lifting. I discuss how the non-trivial interplay of the Dirac point degeneracy and the local nature of impurities decorating this material gives rise to mass generation in only one of the Dirac cones, creating a triple point at charge neutrality independently of the strength and location of the impurities [5].

In the second example, I would like to highlight how twisted trilayer graphene has been proposed as a platform to emulate heavy fermion physics [6]. This specific configuration of graphene sheets naturally hosts both extended and localized electronic modes with an electronic structure that can be controlled by interlayer bias. In the presence of interactions, the existence of localized modes leads to the development of local moments, which are Kondo coupled to coexisting extended states. By electrically controlling the effective exchange between local moments, the system can be driven from a magnetic into a heavy fermion regime, passing through a quantum critical point, allowing one to electrically explore a generalized Doniach phase diagram [7].

Exceptional van Hove Singularities
in the Pseudogap Metal

In the last few years there is increasing evidence that the behavior of several cuprates in the doping range where the pseudogap ends is rather unusual. For instance, the electronic Raman spectroscopy shows anomalous enhancement, while the specific heat 

coefficient appears to diverge.

In the same doping range a change of the Fermi surface topology, from hole to electron-like, is often expected and this originates a logarithmic divergence in the density of states, which could indeed be at the origin of an enhancement of many experimental responses. Previous studies seems however to have ruled out such a possibility for the Raman and specific heat responses in cuprates.

The Pseudogap phase is however an unconventional metal, strongly marked by strong correlations. How the strongly correlated Pseudogap is sensitive to a weak coupling feature, such as the change in Fermi surface topology, and how this could originate divergences in the density of states has remained an open question.

Here we present a phenomenological work that attempts to answer this question and to explain how the above puzzling experimental observations could be explained by the presence of an “exceptional” van Hove singularity, which is a consequence of strong coupling physics, and which is not adiabatically connected to an ordinary weak-coupling van Hove singularity.

Références :
Orbital-selective metal-insulator transitions in the presence of strong magnetic fluctuations

Materials, whose electronic spectrum has non-degenerate orbitals at the Fermi level, can exhibit intriguing orbital-selective phenomena. Electronic correlations in these materials have an intrinsic multi-orbital character, which may result in the formation of exotic phases with coexisting itinerant and localized electrons living on different orbitals. Signatures of the orbital-selective metal-insulator transitions (OSMIT) have been observed experimentally in a doped ruthenate compound $\text{Ca}_2\text{Sr}_x\text{RuO}_4$ and several iron-based superconductors. There were many attempts to explain these effects theoretically, but the microscopic mechanism of the OSMIT in realistic materials remains controversial.

An overwhelming majority of these theoretical studies is based on local approximations to electronic correlations, namely the dynamical mean-field theory and the slave-spin approach, and is thus focused on the Mott mechanism of the OSMIT. Considering effects of strong spatial collective electronic fluctuations in the multi-orbital framework is computationally challenging, which greatly limits the possibilities of studying the OSMIT that leads to a symmetry-broken magnetic state.

In this talk, I will show that a consistent consideration of the non-local magnetic fluctuations completely changes the physics of the two-orbital Hubbard model with different bandwidths of the orbitals, for which the orbital-selective Mott transition was predicted in the framework of the local theories. I will show that upon lowering temperature the considered system undergoes the Néel transition to the ordered antiferromagnetic phase before it experiences the orbital-selective Mott transition [1]. Importantly, the former occurs simultaneously for all orbitals, which eliminates the orbital selectivity from the metal-insulator transition. The possibility to realize an orbital selective Néel transition in the considered system will also be discussed [2].

Références :

Spatiotemporal characterisation of the field-induced insulator-to-metal transition

Resistive switching is a process by which the electrical resistance of a sample changes abruptly in response to a voltage pulse, often by orders of magnitude. This process is at the heart of many neuromorphic computing approaches. By doing numerical simulations on Mott resistors network model, we were able to give a theoretical background to experimental observations in order to demonstrate that filament formation is triggered by nucleation at hotspots, with a subsequent expansion over several decades in time. By comparing three case studies (VO$_2$, V$_3$O$_5$, and V$_2$O$_3$), we identified the resistivity change across the transition as the crucial parameter governing this process. Our results provide a spatiotemporal characterisation of volatile resistive switching in Mott insulators, which is important for emerging technologies, such as optoelectronics and neuromorphic computing [1].

Furthermore, in a recent follow up work we have used a combination of in-operando standard and scanning near-field optical microscopies to study the characteristic lengths of filament formation during the electrically-induced IMT. We found that, in addition to bias current, filament width is strongly dependent on base temperature and the specific material. Lower set-up temperatures yield thinner filaments, increasing current density and local temperature, leading to sharper resistive switching properties. With the aid of resistor network simulations, we discussed the material properties that control filament size, underlining the importance of the resistivity drop across the IMT as well as the substrate's thermal conductivity. Our results support recent works concerning another fundamental aspect of the electrically-induced IMT: switching dynamics [1,2]. It was pro-posed that a large resistivity ratio between insulator and metal would induce higher current focusing, increasing local Joule heating within the filament and explaining the different switching timescales observed in V$_2$O$_3$, VO$_2$, V$_3$O$_5$, NdNiO$_3$ and SmNiO$_3$. However, direct evidence of this has been lacking so far. In this work we provides a systematic study of the characteristic lengthscales of the electrically-induced IMT, unveiling a strong connection between resistivity, thermal properties, filament size and resistive switching characteristics [3].

The effect of the local moment on the metal-insulator transition in the Hubbard model

The Hubbard model exhibits different kinds of metal-insulator transitions that are driven by fluctuations, interactions, or temperature. The system’s rich phase diagram has been extensively studied using a huge collection of advanced computational methods. However, a clear identification of the effects behind each transition is missing. In this work we self-consistently combine a state-of-the-art dynamical mean-field theory description of an interaction-driven Mott transition with a diagrammatic description of the non-local collective electronic fluctuations, and revisit in detail the phase diagram of the Hubbard model. This approach allows us to identify the role the local magnetic moment plays in the metal-insulator transitions in different regimes of electronic interaction.
**Slave-spin mean field for broken-symmetry states: Néel antiferromagnetism and its phase separation in multi-orbital Hubbard models**

We introduce the generalization of the Slave-Spin Mean-Field method to broken-symmetry phases. Through a variational approach we derive the single-particle energy shift in the mean-field equations which generates the appropriate self-consistent field responsible for the stabilization of the broken symmetry. With this correction the different flavours of the slave-spin mean-field are actually the same method and they give identical results to Kotliar-Ruckenstein slave-bosons and to the Gutzwiller approximation. We apply our formalism to the Néel antiferromagnetic state and study it in multi-orbital models as a function of the number of orbitals and Hund's coupling strength, providing phase diagrams in the interaction-doping plane. We show that the doped antiferromagnet in proximity of half-filling is typically unstable towards insulator-metal and magnetic-non magnetic phase separation. Hund's coupling extends the range of this antiferromagnet, and favors its phase separation.

**Références :**

Imaging the itinerant-to-localized transmutation of electrons across the metal-to-insulator transition in V$_2$O$_3$

According to Bloch theory, metal or insulator are mutually exclusive states of matter. In insulators the highest occupied quantum-mechanical energy band is totally filled with electrons, while in metals it is partially filled. Thus, as temperature cannot change the number of electrons in a solid, it should not change either its intrinsic nature, i.e. metallic or insulating. However, V$_2$O$_3$, a metal at room temperature, shows a first-order metal-to-insulator transition (MIT) when cooling below $T_{\text{MIT}} \approx 160$ K, with an abrupt resistivity change of over six orders of magnitude. The very existence of a metal-to-insulator transition shakes the foundations of the well-tested Bloch model!

In fact in V$_2$O$_3$ the last partially filled band is formed out of $d$-orbitals, which are rather localized in space. Thus, electrons in these bands can hardly avoid each other, and are subject to their strong mutual repulsion —the electron correlations, neglected in Bloch theory. The strong repulsion between electrons can inhibit their movement and result in a “Mott” metal-to-insulator transition (MIT), a fundamental phenomenon whose understanding has remained a challenge for over 50 years. A key issue is how the wave-like itinerant electrons in the metallic state change into a localized-like state in the insulator due to increased interactions. However, observing the MIT in terms of the energy- and momentum-resolved electronic structure of the system, the only direct way to probe both itinerant and localized states, has been elusive.

In this talk, I will discuss our recent experimental studies of the MIT in V$_2$O$_3$ using angle-resolved photoemission spectroscopy (ARPES). We found that in V$_2$O$_3$ the temperature-induced MIT is characterized by the progressive disappearance of its conduction band of itinerant electrons, without any change in its energy-momentum dispersion, and the simultaneous shift to larger binding energies of a quasi-localized state initially located near the Fermi level. Only when the state of itinerant electrons crossing the Fermi level has vanished, a complete gap of about 700 meV is observed, associated to the final energy position of the quasi-localized state. Furthermore, the spectral weights of the itinerant and quasi-localized states show a clear thermal hysteresis that tracks the one observed in resistivity data across the MIT [1].

Références :
Spin dynamics seen by Inelastic Neutrons Scattering in an 1D Fe superconducting compound

Since recent years and the discovery of their high superconducting Tc under high pressure, pnictides have been an intense field of research to understand the necessary ingredients and the origin of pairing mechanism for superconductivity, both experimentally and theoretically. In this context, we have studied the quasi-1D Fe spin ladder BaFe$_2$Se$_3$ characterized by a (quasi 1D) “block” magnetic order suggesting frustrated interactions at ambient pressure, different from usual stripe or classical antiferromagnetism, and its quasi-unidimensionality, it becomes superconducting under pressure. This reshape the common usual ingredients for inducing superconductivity in 3d systems: magnetic fluctuations are expected to be responsible of cooper pairs formation [3]. To try out this hypothesis, it is important to determine the magnetic exchange couplings at play, and in turn to estimate the strength of magnetic fluctuations, and the stability of the superconducting phase.

To address this issue and find out exchange couplings in BaFe$_2$Se$_3$, we performed Time of Flight inelastic neutrons scattering on single crystal; data show nice dispersions and acoustic and optical modes at low and high energy. We also performed spin wave calculations [4,7] and Monte Carlo simulations to find a model that could support our results, and be compatible with magnetic transition under pressure [6,7]. This seminar will show the results and our analysis of this study, and discuss research directions planned for the future.

Références :

31 May 2023
Quantum Magnetism
Emergent Symmetry and Quasi-Particles

Emergent phenomena in which many body interactions drive the appearance of a higher symmetry than that of the Hamiltonian or of degrees of freedom at the microscopic level are of major importance in condensed matter physics. Two classic examples are the emergence of U(1) fields in two dimensional planar systems [1,2] and in the frustrated magnets known as spin ice [3,4]. I will review this physics, highlighting a remarkable number of similarities between the two examples. I will show how the emergence leads to a simplified grand canonical description in which topology and topological phase transitions play a vital role. In each case I will extend the problem to include emergent fields of higher rank [5,6] and discuss the effects of quantum fluctuations. Experimental consequences will be extensively reviewed.

Références :
Phase transitions in topologically constraints kagome and pyrochlore lattices

An antiferromagnetic Ising model on pyrochlore lattice (a corner sharing tetrahedron on diamond lattice) presents a ground state called spin ice which is characterized by a 2 spins in and 2 spins out rule on each tetrahedron[1]. This phase is famous for presenting excitations acting like magnetic monopoles that locally breaks this ice rule. Therefore, a spin ice with a non-zero defect concentration is called Coulomb phase[2].

When a strong field is added in the direction pointing by tetrahedron, all spins will align and one will have a non-degenerate ground state respecting a 3-in-1-out rule[3]. If this constraint is enforced, the system is in a crystallized monopoles phase, excitations won’t be single spin flip but loops spawning through boundaries conditions, and the system will present a Kasteleyn phase transition between the low temperature fully ordered phase and high temperature topological liquid[4]. This will be extensively discuss during this seminar.

A very close issue is the kagome lattice under a 2-in-1-out constraint with antiferromagnetic interactions and a magnetic field, which have exactly been solved by Kasteleyn[5]. We’ll discuss the issue occurring when a plaquette term is added, counting the number of flippable small loop in the system.

Références :

Degeneracies in achiral topologically-ordered phases

Topological order appears in strongly interacting systems where different phases cannot be distinguished by local symmetries. In two dimensions, it is characterized by point-like quasiparticles known as anyons that interact through self- and mutual- Aharonov-Bohm phases, and a ground-state degeneracy that depends on the surface’s topology (sphere versus torus, e.g.). Only recently, experiments have brought evidence for the presence of anyons in the fractional quantum Hall effect [1,2]. Topological order is also expected to be present in spin liquids. Motivated by the dream of using topological order for quantum computation, other recent experiments have led to the observation of anyons in a quantum processor made of superconducting qubits [3,4].

On the theoretical side, one of the most studied toy models for topologically ordered phases is the string-net model of Levin and Wen [5]. This model makes it possible to describe a large class of (achiral) topological quantum phases. While finding its energy spectrum is straightforward, determining the degeneracies of the excited states is challenging.

In this talk, after a short introduction to the string-net models, I will explain how to compute their energy-level degeneracies. Our results are valid for any achiral topological quantum phase and any surface topology. The degeneracies give access to the partition function of the string-net models, opening the door to the study of finite temperature properties.

Références:
2D quantum magnetism: when theory meets experiments

Two-dimensional (2D) systems are of unique importance to many-body quantum mechanics, as attested, e.g., by superconductivity in the cuprates and at the LAO/STO interface, as well as graphene mono-layers. Part of this importance stems from the Mermin-Wagner theorem, which precludes any long-range order arising from the spontaneous breaking of a continuous symmetry in two dimensions, but leaves room for a topological transition at finite temperature, named after Berezinski, Kosterlitz, and Thouless (BKT), e.g., in superfluid thin films or 2D easy-plane magnets. However, experimental realizations of purely 2D magnets are rare, and the observation of BKT criticality remains a very challenging task. In order to make some progress, a better theoretical understanding of 3D residual couplings is clearly necessary [1].

Here I propose to discuss some recent works where 2D magnetism has been explored for a few systems [2-5]. I will start with the so-called Han Purple pigment, BaCuSi$_2$O$_6$, which upon applying high magnetic fields displays an unconventional quasi-2D criticality [2], as also recently revealed by neutron scattering up to 25.9 T [3]. Interestingly, we have discovered that Sr doping helps to restore a more conventional 3D critical behavior in the parent material Ba$_{0.9}$Sr$_{0.1}$CuSi$_2$O$_6$ [4]. In a second part, I will discuss another class of systems: the monolayer halides CrCl$_3$ that we have theoretically predicted [5] to achieve (upon applying a compressive strain) a relatively high BKT transition temperature, around 50 K, due to their singular dependence on the weak easy-plane anisotropy of the material.

Dimensional modulation of spontaneous magnetic order in quasi-2D quantum antiferromagnets

Multiple Magnetic Bilayers and Unconventional Criticality without Frustration in BaCuSi$_2$O$_6$

Investigating field-induced magnetic order in Han purple by neutron scattering up to 25.9 T

Revealing three-dimensional quantum criticality by Sr substitution in Han purple

Monolayer CrCl$_3$ as an Ideal Test Bed for the Universality Classes of 2D Magnetism
Superconductivity
Spin triplet superconductivity in bulk strongly correlated materials.

Spin triplet superconductivity appears to be a rather rare state of matter. However, it came back in the limelight with the development of quantum engineering, thanks to its potential topological properties. Presently, all well-established candidates for spin triplet superconductivity are found among uranium-based systems.

After a general introduction to spin-triplet superconductivity, we will discuss the main properties arising from the coexistence of ferromagnetism and superconductivity in UCoGe and URhGe, and notably the field dependence of the pairing mechanism.

This will be compared to the case of UTe₂, a paramagnetic spin-triplet superconductor presenting a flurry of pressure [1] or field-induced superconducting phases [1,2,3], with record high critical fields (70T) for a rather low critical temperature (1.6K).

Beyond this stunning “endurance” to very large magnetic fields, a major interest of this system is the interplay between magnetic properties or more generally electronic correlations, with superconductivity: UTe₂ is probably a unique case, where different superconducting phases appear thanks to competing pairing mechanisms [1].

Références :
[1] Aoki et al., Unconventional superconductivity in UTe₂, JPSJ, 2022, 34, 243002
[2] Rosuel et al., Field-Induced Tuning of the Pairing State in a Superconductor, PRX, 2023, 13, 011022
Superconductivity is a peculiar property of materials characterized by zero resistance to direct current and the expulsion of magnetic flux. It is explained by the formation of bound electrons called Cooper pairs. The most widely studied superconductors are the perovskite cuprates which have the highest critical temperature ($T_c$) at ambient conditions. In the search for compounds analogous to cuprates, nickel oxides were long proposed to host superconductivity [1,2]. This research crystallized in 2019 with its discovery in infinite layer nickelates $R_{1-x}Sr_xNiO_2$ [3] and lately in a reduced Ruddlesden-Popper $Nd_6Ni_5O_{12}$ [4] with similar critical temperatures ($T_c = 9-15$ K), albeit without clarifying the pairing mechanism. Here we use Density Functional Theory (DFT) to show that superconductivity is tightly related to a charge and bond ordered state that produce a significant electron-phonon coupling. We reveal the existence of an intrinsic instability of $\text{Ni}^{1.5+}$ cations ($3d^{8.5}$) in the half-doped case that prefers to transform to more stable $\text{Ni}^+$ ($3d^9$) and $\text{Ni}^{2+}$ ($3d^8$) cations in the ground state. It results in a bond disproportionation of $\text{NiO}_2$ square complexes, a charge ordering and an insulating state. Starting from this half doped situation, doping is shown to suppress disproportionation effects, ultimately resulting in a metallic regime. Once this charge ordered state is suppressed, the related bond disproportionation mode is still highly coupled to the electronic structure and the resulting electron-phonon coupling is sufficient to explain the $T_c$ dome versus doping content observed experimentally. It suggests that the vicinity of a charge ordered state is a prerequisite for superconductivity in the nickelates. Hence, these compounds appear similar to non-magnetic bismuthates [5].

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References:

Measurements of the superconducting properties of aluminum thin films by Point Contact Spectroscopy

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Point contact spectroscopy (PCS) is an experimental technique allowing to measure the elementary fermionic excitations of a metal and their coupling to bosonic degrees of freedom, such as phonons or magnons [1]. In superconducting material, PCS allows to measure the gap amplitude and electron-phonon coupling constants [2] by measuring the successive derivatives of the current-voltage characteristics.

Bulk Aluminum has a critical temperature (Tc) of 1.2K that increases in thin films, on the contrary to most superconductors. The cause of this behaviour is not clearly established yet, due to the various degrees of freedom involved in this system (microstructure-structure relation, electron correlations, changes in the electron-phonon coupling, etc).

In this work, we have measured the energy-resolved electron-phonon coupling function (known as Eliashberg function) in aluminum thin films (grown on Si) of various thicknesses. A systematic change of the Eliashberg function is observed, resulting surprisingly in a decrease of the e-ph coupling constant in thinner films (with the higher Tc). Moreover, the thinnest films exhibit spectroscopic signatures of local superconductivity at temperatures one order of magnitude higher than the critical temperature deduced from transport measurements. Those results suggest that the inhomogeneities of Al thin films and the resulting superconducting fluctuations [3] are a key ingredient to understanding superconductivity in these systems.

Références :

Non-reciprocal transport in superconducting Nb

Certain asymmetric conductors exhibit non-reciprocal transport properties, characterized by the fact that the resistance for current flow is different in opposite directions. The most well-known example of such a system is the p-n junction diode. The phenomenon of non-reciprocal transport is quite general and may be observed in systems lacking inversion symmetry. A broad range of solid-state systems, including both semiconductors and superconductors, are known to exhibit such properties. A prominent signature of non-reciprocity is the generation of a voltage signal having d.c. and even harmonic components when an a.c. current is applied.

In superconducting devices, non-reciprocity may arise due to non-centrosymmetric crystal structure [1], Rashba-type spin orbit interaction [2], interface with a different material [3,4], and the presence of asymmetric pinning arrays [5,6]. In most instances, non-reciprocal behaviour is seen in the presence of an applied magnetic field, therefore requiring the absence of both inversion and time reversal symmetries. Accordingly, the voltage observed has an antisymmetric field dependence. Some of these systems are capable of rectifying environmental electromagnetic fluctuations into a d.c. voltage, leading to the spontaneous generation of d.c. electricity [7,8].

We have conducted experiments with superconducting Nb films and observed the spontaneous generation of d.c. voltage resulting from the rectification of environmental fluctuations. The devices used in our experiments are not designed to be asymmetric. The rectification effect observed here can be attributed to an unconventional mechanism of non-reciprocal transport resulting from a spontaneous breaking of inversion symmetry at the superconducting transition. The unconventional nature is further confirmed by the fact that the rectified signal is symmetric with respect to the magnetic field, in stark contrast to most other systems known.

Références :
On the stabilisation of superconductivity in infinite-layer praseodymium nickelate thin films

Araceli Gutiérrez-Llorente, Dongxin Zhang, Laurent Divay, Christophe Galindo, Manuel Bibes, Lucia Iglesias

The first observation of superconductivity at relatively high temperature, around 15 K, in single-crystal thin films of infinite-layer nickelates upon hole doping was a significant breakthrough in the field of unconventional superconductivity (D. Li et al., Nature 572, 624 (2019)). Thus, this result generated enormous enthusiasm, but experimental replication of superconducting nickelate thin films with infinite-layer structure has proved to be hugely challenging.

Superconductivity emerges in infinite-layer nickelate thin films through hole doping and oxygen deintercalation by means of topotactic reduction using CaH₂ of the perovskite phase grown by pulsed-laser deposition. Therefore, the major difficulties of this system lie first in stabilising epitaxially the hole-doped precursor perovskite phase and secondly, in turning that phase into the superconducting infinite-layer phase in single-crystalline film form. Thus, the growth parameters and the reduction conditions should be thoughtfully studied. We have grown praseodymium strontium nickel oxide, Sr₀.₂Pr₀.₈NiO₃, films up to 10 nm in thickness on different substrates by pulsed-laser deposition and characterised their structure, cation stoichiometry and electrical transport properties. Upon reducing annealings, we have successfully synthesized Sr₀.₂Pr₀.₈NiO₂ films that exhibit superconducting transitions reaching a zero-resistance state with onset transition temperature around 9K. From these results, we discuss the impact of epitaxial strain on the stabilisation of the infinite-layer nickelate superconductors, which at the present remains unclear but whose understanding is essential to improve sample quality and get some insight into the role of the interface between substrate and nickelate thin films in the emergence of superconductivity.
The topotactic fluorination of LaFeSi emphasizes the 2D character of the dominant Fe-3d single band and LaFeSiF$_x$ becomes superconducting around 10K [1]. The comparison of Fermi surface of LaNiO$_2$ and La$_2$NiO$_3$F (T’ structure analogous to Nd$_2$CuO$_4$) show the remarkable 2D character of 3d$_{x^2-y^2}$-Ni in La$_2$NiO$_3$F and the absence of La-5d electron pockets as found in LaNiO$_2$ [2]. The presence of Fluorine in Fluorite ‘LaF’ sheets contributes to increase the ionicity of this block, then by inductive effects, to enhance the covalent or the metallic character of Ni-O or Fe-Si bonds. In both these systems, nickelates and silicides, additional 5d-rare earth bands also contribute to the Fermi surface, giving rise to electron pocket which dissappear after fluorination or by enhancing the rare-earth block ionicity in tri-layers RE$_4$Ni$_3$O$_8$ compared to RENiO$_2$.

In numerous cuprates and nickelates, transition metals are stabilized in square planar or elongated octahedral sites of oxygen whereas in iron arsenates or silicides, iron atoms occupy tetrahedral sites of arsenic or silicon. These materials families exhibit superconducting properties at ambient pressure.

The strong H-S bonding in H$_3$S (Im-3m space group) at high pressure, leading to high Tc superconductivity, produces a strong density of states at the Fermi level with a perfect electron-phonon coupling (80 % H and 20% S vibrational mode contributions). The H-s orbital are predominant at E$_F$, each H atoms forming a square planar with other H and a dumbbell with two S atoms.

The charge transfer between metal and first neighbors (the smallest in high Tc cuprates), the long range hopping to nearest neighbors hopping and finally the splitting of transition metal (e$_g$ or t$_{2g}$) orbitals are key features. The strong electron phonon coupling in cuprates involving Cu d$_{x^2-y^2}$ and O 2p$_{x,y}$ bands at the Fermi level, the single Ni d$_{x^2-y^2}$ band crosses also the Fermi Level with Rare-earth d electron pockets suggesting Kondo interactions in nickelates and finally Fe-3d bands domination at the Fermi level in silicide-fluoride, have to be considered. In other superconducting silicides with the same Fe-based, the hole band of p(Si) orbital with high frequency phonons are predominant at the Fermi level. The strongest (O)p-(Cu)d hybridization linked to the lowest charge transfer energy with a larger N(E$_F$) density of states at the Fermi level is specific to high Tc cuprates.

The topotactic reduction of RE$_{1-x}$Sr$_x$NiO$_3$ 3D perovskite to RE$_{1-x}$Sr$_x$NiO$_2$ (RE = La, Nd) 2D-infinite layers allows showing the strong influence of rare-earth nature on the structural features with the occurrence of stacking faults. The electronic and magnetic properties will be discussed considering metallic character, electronic correlations as well as short-range magnetic correlation and spin-glass behavior. Contributions from the crystal field splitting of Nd$^{3+}$ with a two level Schottky anomaly at low temperature, the phonons and the occurrence of long-range AFM order in infinite layers nickelates as observed in parent cuprates could be invoked.

References :


Topology Enabled Unconventional Superconductivity in a Time-Reversal Symmetry Breaking Bulk Superconductor

Unconventional superconductors have Cooper pairs with lower symmetries than in conventional superconductors. In most unconventional superconductors, the additional symmetry breaking occurs in relation to typical ingredients such as strongly correlated Fermi liquid phases, magnetic fluctuations, or strong spin-orbit coupling in noncentrosymmetric structures. In this presentation, I will show that the time-reversal symmetry breaking in the superconductor LaNiGa$_2$ is enabled by its previously unknown topological electronic band structure, with Dirac lines and a Dirac loop at the Fermi level [1,2]. Two symmetry related Dirac points even remain degenerate under spin-orbit coupling. These unique topological features enable an unconventional superconducting gap in which time-reversal symmetry can be broken in the absence of other typical ingredients. Our findings provide a route to identify a new type of unconventional superconductors based on nonsymmorphic symmetries and will enable future discoveries of topological crystalline superconductors. I will discuss how a symmetry guided discovery of topological superconductors can be realized.

Références :
A theoretical description of the interplay of collective fluctuations in strongly correlated systems

Correlated fermion systems often display complex phase diagrams with different competing orderings. A theoretical description of competing instabilities remains one of the major challenges of modern condensed matter physics. We introduce a multi-channel extension of the recently developed fluctuating field approach to tackle this problem, based on a variational optimisation of a trial action that explicitly contains the leading fluctuations channels [1]. Application of the approach to the extended Hubbard models captures the interplay of competing charge density wave, antiferromagnetism, s-wave superconductivity, and phase separation fluctuations [1,2]. For the case of the attractive model, our approach has allowed us to identify a novel phase that is characterised by the coexistence of s-wave superconductivity and phase separation [2]. Our findings resonate with previous observations of interplaying phase separation and superconducting phases in electronic systems, most importantly in high-temperature superconductors.

Références :

2 June 2023
Spectroscopy (neutron, ARPES, Raman ... )
Quantum materials: Angle-resolved Photoemission spectroscopy of topological insulators and semi-metals

In the last two decades, quantum materials exhibiting new quantum phases associated with non-trivial topological electronic structures have been discovered [1]. Topological insulators have been first identified. This new kind of insulators is characterized by an insulating bulk electronic structure and a topological metallic edge state. The metallic character of the edge state is very robust due to its topological character protected by fundamental symmetries (like for example time reversal symmetry) [2]. More recently, non-trivial topology has also been discovered in semi-metals (Dirac and Weyl semi-metals) with Dirac cones at (or close to) the Fermi surface [3-5]. Weyl nodes reminiscent to Weyl fermions of particle physics are characterized by their chiralities induced by time reversal or inversion symmetry breaking. The Weyl nodes appear by pair of opposite chiralities and they are source and sink of Berry curvature. Angle-resolved photoemission spectroscopy (ARPES) is a technique perfectly adapted to highlight the topological aspects of the electronic structure in particular the correspondence between bulk states and topological edge states [6].

After having presented the main concepts of differential geometry (connection and curvature) and topology (topological Chern numbers and topological Z2 numbers), I will discuss the ARPES signature of topological edge states of 3D topological insulators. I will then present ARPES results on Dirac and Weyl semi-metals which evidence the bulk nodes and the corresponding topological edge states. The singular spin texture of these edge states can be probed by spin resolved ARPES.

Références:
Crystal-Electric-Field excitations of CeCoSi unveiled by Raman spectroscopy

CeCoSi is a Kondo system which has been observed to form an unusual “hidden-order” state [1-3]. Furthermore, this state has been proposed to occur due to local inversion symmetry breaking between the two Ce sites in this otherwise globally inversion symmetric system [4], which makes it similar to the recently discussed CeRh$_2$As$_2$. It is also predicted that this hidden-order state is intimately connected to excitations between a trio of Kramers doublets that form from the splitting of the Ce$^{3+}$ $J=5/2$ multiplet under the action of a tetragonal crystal-electric-field (CEF) [1-4]. However, the symmetries of these multiplets are currently disputed [3,4], but they are vital for predicting the ordering of this system.

In this work, we use polarised Raman spectroscopy to identify the energies and symmetries of the CEF transitions and the electronic continuum of the system. At low temperatures and amongst all of the phonons that are expected for the structure, we observe multiple excitations that agree well with previous inelastic neutron scattering measurements [3]. By extracting the symmetries of the excitations, we clearly identify the CEF scheme of the Ce$^{3+}$ site in CeCoSi (in order of increasing energy from the ground state) to be $\Gamma_7$ – $\Gamma_7$ – $\Gamma_6$ in agreement with Nikitin et al. [3]. Furthermore, we observe that we have more CEF excitations than expected for a trio of doublets which is indicative of one of several scenarios, however the most likely scenario seems to be local-symmetry breaking between the two Ce.

Références:

Superconductivity and Kondo lattice effects in La$_{1-x}$Ce$_x$FeSiH: An interdisciplinary approach to a strongly correlated electron system

Strongly correlated electron systems refer to a broad class of materials in which exotic phases of matter such as unconventional superconductivity are observed. Very often, the strong electronic correlation is associated with atomic Coulomb repulsion in d or f orbitals.

I will present in this talk the work of my PhD thesis, which was the occasion for an interdisciplinary work between solid state chemistry and theoretical physics. Starting from the iron based superconductor LaFeSiH, we studied the family of intermetallic hydrides La$_{1-x}$Ce$_x$FeSiH (0 ≤ x ≤ 1), in which rich physical properties might emerge from the interaction between cerium 4f and iron 3d electrons.

On one side, we realized the synthesis of those compounds, together with structural analysis. The physical properties measurements permits to identify superconductivity, incoherent Kondo effect and coherent Kondo lattice regimes. With the help of different temperature scales, those results are summarized in a phase diagram temperature vs cerium concentration x.

On the other side, we studied some effective model Hamiltonians adapted to the correlated electrons present in those compounds. Considering a non local Kondo hybridization between the cerium 4f orbital and the iron 3d orbital, we showed pocket selective doping effect of iron by cerium, related to the symmetry of the cerium low energy crystal field Kramers doublet.

Key words: strong correlations, heavy fermions, iron based superconductors, non local Kondo coupling, ZrCuSiAs compounds
Doping and temperature evolution of the electronic properties of electron-doped Sr₂IrO₄ seen by ARPES

Sr₂IrO₄ is a layered perovskite isostructural to the high T_c cuprate superconductor La₂CuO₄. In Sr₂IrO₄, the strong spin-orbit coupling of the 5d Ir⁴⁺ ions lifts the degeneracy of the t₂g orbitals, resulting in a single narrow half-filled band described by pseudospin J_{eff}=1/2 degrees of freedom. Consequently, and despite a much lower Coulomb interaction in 5d elements than in 3d, Sr₂IrO₄ is a correlated insulator that orders antiferromagnetically below 240 K adding striking similarities with cuprates. However, Sr₂IrO₄ has a non-interacting electron-like Fermi surface rather than the hole-like one of cuprates. This suggests a particle-hole conjugate doping phase diagram consistent with the prediction of d-wave superconductivity in electron doped Sr₂IrO₄ [1].

Here, I will present new angle resolved photoemission (ARPES) results on electron doped Sr₂IrO₄ compounds with Sr²⁺/La³⁺ substitution. Previous ARPES work on 5% doped Sr₂IrO₄ revealed an unusual metallic state emerging from the breakdown of the Mott insulating state of the undoped compound. This metallic state is characterized by a large Fermi surface broken into disconnected Fermi arcs and a pseudogap near the antinodal direction. However, no superconductivity was observed down to 100 mK [2]. After a brief summary of these results on the undoped and lightly doped compounds, I will discuss the evolution of the Fermi surface volume and the antinodal pseudogap in newly synthetized crystals with La doping as high as 10%. A particular emphasis will be placed of the temperature dependence of the electronic properties.

References:
Magneto-elastic coupling in KTb$_3$F$_{10}$

In recent years, the study of the coupling between the 4f electronic density and phonon modes has aroused a lot of attention. Such interactions are suspected to play a key role in a number of issues, ranging from highly frustrated magnetism to the design of smart materials in optics. In the celebrated Tb$_2$Ti$_2$O$_7$ frustrated magnet, for instance, interaction between 4f quadrupoles and lattice vibrations may be responsible for the lack of long-range order, as well as for the rise of new magneto-elastic modes, which remain unexplained so far [1, 2,3]. Even if such hybrid modes have been reported for a long time [4,5], they are still topical as they may also be at the origin of the newly discovered “phonon Hall effect” or “thermal Hall effect”, which consists in the appearance of a sizeable thermal conductivity perpendicular to both the applied magnetic field and heat flux [6,7]. In close analogy with the classical Hall effect in conductive materials, the phonon Hall effect could result from an “anomalous” phonon velocity due to the remarkable topological properties of phonon bands. Actually, several theoretical works advocate that these properties may be due to the coupling between CEF and lattice degrees of freedom [8].

Terbium potassium fluoride KTb$_3$F$_{10}$ is another promising novel model system in which to investigate this physics. Our first investigations by means of neutron scattering show that the Tb ions occupy the vertices of a network of corner-sharing octahedra (cubic $Fm\overline{3}m$ space group). Furthermore, our study shows that ground state is a crystal field singlet, with a peculiar field induced structure resulting from site dependent easy plane anisotropies (Figure 1 et Figure 2). The Tb$^{3+}$ CEF scheme encompasses an excited doublet at 2.7 meV which is suspected to play a key role in the anomalous low thermal conductivity.

References :
Phase transitions and spin dynamics of the quasi-one dimensional Ising-like antiferromagnet BaCo$_2$V$_2$O$_8$ in a longitudinal magnetic field

BaCo$_2$V$_2$O$_8$ is a realization of a spin-$1/2$ Ising-like quasi-one dimensional antiferromagnet (AF) with fascinating static and dynamical behaviors. In zero-field, the weak interchain interactions stabilize below 5.6 K a peculiar Néel ordering, characterized by magnetic moments aligned along the chain $c$-axis and dressed with confined two-spinon excitations. We have explored the influence of a longitudinal external magnetic field ($H \parallel c$) both on the AF ordering and on the spin dynamics by means of single-crystal neutron scattering combined with numerical simulations [1].

When the field closes the excitation gap arising from the magnetic anisotropy, at $H_c \sim 4$ T, the Néel ordering turns into a longitudinal incommensurate one, due to the Ising character of the system. Concomitantly, a fundamental reconfiguration of the spin dynamics occurs, with a strong modification of the spectral weight and an incommensurate nature above $H_c$. This phase has raised a strong interest as it is a unique example of the Tomonaga-Luttinger liquid (TLL) physics experimentally accessible under moderate magnetic field. If the applied field is increased further, another transition into a transverse antiferromagnetic (TAF) ordering takes place at $H^* \sim 9$ T due to the competition between longitudinal and transverse correlations. The spin dynamics remains nevertheless essentially incommensurate in the low field region of the TAF phase, as determined experimentally up to 10 T.

Numerical simulations allowed to demonstrate that the competing longitudinal and transverse fluctuations result in a contrasting nature of the phases, below and above the critical field $H^*$, concerning the spin dynamics and the magnetic order stabilized by the weak interchain coupling. The modelling of BaCo$_2$V$_2$O$_8$ could also be refined by including 4-site periodic terms into the Hamiltonian in order to account for the anticrossing observed in the inelastic scattering spectra and for higher field results unexplained in the framework of the Tomonaga-Luttinger liquid theory.

Référence :

Study of magnetic epitaxial thin films using neutron diffraction

The field of antiferromagnetic spintronics has been the object of active research over the past decade for next generation of spin-based devices [1]. A particular focus has been on the design of new materials, specifically thin film preparation and detailed understanding of their magnetic properties.

Determination of the magnetic structure of these films is still a challenge and neutron diffraction technique is a good candidate for such a determination. The D10 instrument at ILL has been increasingly used for measurements on such films [3,4]. This challenging technique is still under development and the future commissioning of D10+ at the ILL will be a very good opportunity.

In this framework, we use neutron diffraction to solve nuclear and magnetic structure of epitaxial thin films. We are interested in La\textsubscript{2/3}Sr\textsubscript{1/3}MnO\textsubscript{3} (LSMO) thin films deposited on Si substrate with a SrTiO\textsubscript{3} (STO) buffer layer. We have employed SQUID magnetometer, electron diffraction, X-ray diffraction and ab-initio calculations to complement the ND data. Our results show that these films can be treated as single crystals to measure diffraction peaks and determine the nature of magnetic spin configuration in the system. Then we have applied the same technique to LSMO-BTO (BaTiO\textsubscript{3}) superlattices in which the magnetization can be maintained up to room temperature thanks to the strain induced by the BTO layers which controls the orbital ordering of the LSMO layers.

In this talk, I will present our most recent results obtained on these manganese oxide thin films.

This work is the Himanshu PhD works made in Institut Laue Langevin.

Thanks to the team : Elisa Rebolini, Marie-Bernadette Lepetit, Bachir Ouladdiaf, Ketty Beauvois, Stéphane Grenier, Bernard Mercey, Bernadette Domenges.

Références :

Magnetization reversal and Non CentroSymmetric character in α-CrPO₄ family

The so-called α-CrPO₄ structure consisting of [Cr₂O₁₀] units (edge-sharing octahedra) interconnected through corner sharing [Cr₁O₆] octahedra [1] has attracted our attention. The framework is illustrated in the bottom right of figure with [Cr₁O₆] octahedral in blue, [Cr₂O₁₀] units in grey sharing corners and edges with PO₄ tetrahedra in pink.

Following a charge balance \( M^{3+} \leftrightarrow M^{2+} + Na^+ \) methodology, an Imma to Imm2 phase change is highlighted in α-CrPO₄ type structure from NaCoCr₂(PO₄)₃ to NaCuCr₂(PO₄)₃ respectively. In the top, peaks splitting around 35° and 80° and 117° in the Neutron Powder Diffraction patterns underline Jahn-Teller effect consequences found in Cu²⁺ cation. The lattice symmetry decrease is unambiguously seen by Electron Diffraction experiment on the bottom left. The red circles emphasis the presence of forbidden reflections in the Imma space group. The red arrows underline the presence of diffusion.

Up to now, all α-CrPO₄ reported structures crystallized in the Imma centrosymmetric space group. The chemical approach followed is first a sodium intercalation in the channels resulting in a charge balance \( M^{3+} \leftrightarrow M^{2+} + Na^+ \) [3]. Then, contrary to Co²⁺ and Ni²⁺ cation, using the Jahn-Teller cation Cu²⁺, we recently reported the first noncentrosymmetric phosphate related to the α-CrPO₄ type [2] at RT even found (figure 1). Single crystal growth is underway to better understand the studied physical properties.

In this oral/poster, I would like to first present the nuclear structure found on α-CrPO₄ type structure and the underlined AFM to FM phase transition in Na₄₋ₓCo₁₊ₓCr₂₋ₓ(PO₄)₃ materials [4]. Then the methodology allowing us to lower the symmetry from Imma to Imm2 will be discussed comparing NaCoCr₂(PO₄)₃ to NaCuCr₂(PO₄)₃. Finally, a brief description of the Multiphos Project [5] for « Polyanionic frameworks for Multiferroic properties » will be given.

Références :
[5] https://anr.fr/Projet-ANR-21-CE08-0051
Materials and artificial intelligence
From Crystal Structure Prediction to Materials Discovery: the case of metastable sulfide compounds

Crystal structure is a key piece of information about a material since it determines many of its properties. In material design, being able to predict the crystal structure of a compound based only on its structural formula is a key step, as one can then compute many of its properties before it being synthetized. It can also be of valuable help when developing new metastable materials where experimental characterization is problematic and only provides partial information on the crystal structure.

In this talk we will address this problem, called Crystal Structure Prediction (CSP), for crystalline inorganic compounds. For long only heuristic rules prone to the nature of the compound were the only available guideline. Global optimization techniques now routinely give access to the stable and metastable states of bulk compounds, and thrived for many years mostly in the extreme pressure domain. After describing their ingredients and potentialities, we show that they are also particularly interesting in identifying new metastable compounds, especially in soft chemistry, where chemists often come to grips with tough problems in reaching large monocrystalline domains. We explore in particular the case of new metastable sulfide compounds obtained by topochemical reactions, that have been successfully predicted and synthetized in our group for the last years [1,2].

Références :
**Le projet DIADEME : accélérer la découverte de nouveaux matériaux grâce à l'Intelligence Artificielle.**

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Dans le contexte actuel, le déploiement de nouveaux matériaux doit répondre à trois contraintes :
- le temps limité fixé par l’urgence des enjeux sociétaux rappelés ci-dessus
- la complexité chimique, structurale et morphologique au sein de multi-matériaux
- la nécessité absolue de respecter les contraintes environnementales en termes de recours aux matières premières critiques, de durabilité des composants et de recyclabilité

L’objectif principal du projet DIADEME(*) est d’accélérer la découverte de matériaux en respectant ce triptyque temps/complexité/contraintes environnementales.

Dans la première phase de DIADEME qui est en cours, 17 projets ciblés ont pour tâche principale de mettre en place et de valider l’efficience de plateformes. Ces plateformes concernent la synthèse et la mise ne forme à haut débit, les caractérisations haut débit, la numérisation des matériaux et procédés et l’Intelligence Artificielle.

Parmi ces 17 projets, quelques exemples en lien avec les thématiques du GDR MEETICC seront détaillés :
- approche micro-fluidique pour la synthèse haut-débit de matériaux fonctionnels
- exploration de diagrammes de phase complexes sur couches minces d’oxydes fonctionnels par PLD et PVD combinatoires [1]
- laboratoires autonomes pour la synthèse haut débit et la caractérisation structurales de poudres [2]. Dans ce cas, l’apprentissage profond permet de distinguer automatiquement une phase perovskite sur la base de diagrammes de diffraction
- instrumentation de lignes SAXS à l’échelle du laboratoire et sur synchrotron pour la détermination structurale haut débit in situ sur nanoparticules.

**Références :**

(*) Dispositifs Intégrés pour l’Accélération du Déploiement de Matériaux Émergents


Resistive switching in (Cr$_{1-x}$V$_x$)$_2$O$_3$ Mott insulator for data storage and neuromorphic applications

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Resistance random-access memory (RRAM) is considered as a promising candidate to replace the flash memory technologies that approach their limits due to miniaturization and short economical payback [1]. In that context, canonical Mott insulators such as (Cr,V)$_2$O$_3$, or AM$_4$Q$_8$ have recently attracted a considerable attention thanks to the electric-field-driven insulator to metal transitions (IMT) that could be used in memory and neuromorphic applications [2-4]. Previous studies have demonstrated competitive memory performances in Mott insulator thin films, but so far these studies overlooked the variability of the devices [5]. Here we investigate the resistive switching in Mott insulators thin films of (Cr$_{1-x}$V$_x$)$_2$O$_3$ ($x = 0.7$) integrated in micro-sized memory cells and report for the first time the cycle to cycle, device to device variability and state retention of the high resistive state (HRS) and low resistive state (LRS). To this end, thin films were deposited and annealed in order to reach the expected stoichiometry and a good crystalline quality. The electrical behavior was investigated on MIM structures using 2 µm via memory cells made of 50 nm thick (Cr$_{0.30}$V$_{0.70}$)$_2$O$_3$ Mott insulator thin films. Preliminary experiments on these devices using short electric pulses for the SET and RESET show an endurance of more than 2000 cycles (Figure 1.a), with very narrow cumulative distribution function (CDF) curves (see Fig 1.b). Moreover, both HRS and LRS exhibit a very good temporal stability at room temperature, as well as a state retention that can be extrapolated up to 10 years (Fig 1.c). This work shows therefore the potential of (Cr$_{0.30}$V$_{0.70}$)$_2$O$_3$ Mott insulator thin films for integration into the next generation of non-volatile memories and neuromorphic devices.

![Figure 1](image_url)

**Fig1.** (a) Cycling endurance obtained on a device of 2 µm via and 50 nm thick (Cr$_{0.30}$V$_{0.70}$)$_2$O$_3$ Mott insulator thin films demonstrating 2400 resistive switching cycles. (b) Resistance distribution for the high (HRS) and low (LRS) resistive states. (c) Data retention at room temperature extrapolated to 10 years for HRS and LRS.

Title

The application of a strong electric field to Mott materials like vanadium oxides induces an insulator-to-metal transition which is spatially inhomogeneous and characterized by the formation of metallic filaments within the insulating bulk. The surge in the electronic current that follows the collapse of the resistance endows these materials with a spiking property, which enables the implementation of biologically plausible neurons. Such devices may achieve better that state-of-the-art energy efficiency thanks to the miniaturization potential of quantum materials, which goes beyond the limits of CMOS technology. However, the characterization of these materials proves challenging; in particular, it has been observed experimentally that the transition becomes increasingly unpredictable as the voltage is lowered, questioning the notion of a well defined voltage threshold. Moreover, it is not yet clear if the resistive collapse may be attributed to thermal effects only, or if electronic effects also play a role. Understanding these features not only would further our knowledge of Mott materials, but is also instrumental in the realization of Mott-based neuromorphic devices. In this presentation we use numerical simulations of a resistor network model to study the incubation process of the metallic filaments in Mott materials under an applied voltage. We show that this process is affected by both thermal and electronic effects, in the form of Joule heating and current density concentration respectively, and that it is intrinsically stochastic. Remarkably, we establish a quantitative similarity between the probably distribution of filament formation in Mott materials and of spike emission in biological neuronal models with an exponential escape rate, concluding that both processes may be characterized as Poissonian. We support the results of our numerical simulations with VO2 experiments, which are in agreement with the predicted Poissonian behaviour. The work that we present shows that Mott materials are suitable candidates for developing energy-efficient biologically plausible neurons with a stochastic spiking behaviour.

Références :
2 June 2023
New materials
High-mobility ferroelectric two-dimensional electron gases based on strain-engineered SrTiO$_3$ thin films

Two-dimensional electron gases (2DEGs) based on the quantum paraelectric SrTiO$_3$ display fascinating properties such as large electron mobilities, superconductivity and efficient spin-charge interconversion owing to their Rashba spin-orbit coupling.\(^1\)\(^-\)\(^3\) However, such 2DEGs have almost exclusively been generated in SrTiO$_3$ single crystals, with the few attempts to replace crystals by heteroepitaxial SrTiO$_3$ thin films leading to low carrier mobilities. This is limiting the potential to integrate SrTiO$_3$ 2DEGs in future devices as well as the possibility to introduce additional functionalities specific to SrTiO$_3$ thin films, such as strain-induced ferroelectricity. Here, we use oxide molecular beam epitaxy to grow high quality strain-engineered SrTiO$_3$ films that are ferroelectric up to 170 K. We then generate a 2DEG by sputtering a thin Al layer and demonstrate an increase in both the low and room temperature mobilities by up to factor of four compared to earlier literature. Furthermore, through Raman spectroscopy and magneto-transport measurements, we show that the ferroelectric character is retained after 2DEG formation. These results thus qualify our samples as ferroelectric 2DEGs up to temperatures well above previous results based on Ca-SrTiO$_3$ substrates (~30 K)\(^4\), opening the way towards ferroelectric 2DEGs operating at room temperature.

**Références :**

The CRISTECH network [1], created in 2006, aims at identifying and federating the crystal growth community in France. It is hosted and funded by the “Mission for Transversal and Interdisciplinary Initiatives” (MITI) of the CNRS. The network is open to all people interested in crystal growth and crystallization processes, whatever their scientific field of interest (physics, chemistry, biology, geology, material sciences, ...) or their organization (CNRS, University, EPIC, industry). Thus, the network gathers users of many techniques of single crystal growth (high temperature, solution, micro-fluidic, thin films...) whether they are devoted for inorganic, semiconducting, organic, molecular or biological compounds.

In particular, crystal growth is often associated with inorganic bulk crystals (oxides, metal halides ...) [2-4]. A review of the main crystal growth techniques available in France, from a molten bath or from a liquid solution at high temperature, is presented.

Références :
Molecular magnetism deals with the design of spin-carrying molecules for either individual or collective magnetic behaviours. This covers the investigation of diamagnetic – paramagnetic transitions, magnetic ordering, magnetochirality, superparamagnetism… Such investigations imply a multidisciplinary work between chemists, physicists and theoreticians in a multi-instruments and multi-scales approach. Thanks to the versatility of coordination chemistry, various kind of applications are envisioned like molecule-based electronics, quantum computing, switches and sensors, MRI contrast agent, information storage, barocaloric materials, actuators, among others.

The aim of this presentation is obviously not to cover all the aspects of this field but to give some current evolutions of this domain on the search for single molecule magnetic bistability, room temperature magnets, molecular electronics, switches…

Références:


**Exotic Compounds with Frustration between low-D units.**

Magnetic frustration and lattice dimensionality are two key ingredients that harbor exotic phenomena and unconventional states of matter, such as quantum spin liquids (QSLs),\(^1\) spin ice,\(^2\) superconductivity,\(^3,4\) and topological states.\(^5\) In most one or two spatial dimensions, the Mermin-Wagner theorem states that long-range ordering does not occur at finite temperatures, otherwise that assisted by magneto-crystalline anisotropy. However, in some inorganic materials, it exists original intermediate topologies where strongly inner-coupled low-D topologies (chains, layers) are weakly coupled together via magnetic ions, with very ambiguous roles towards the 3D-ordering.

I will present new compounds studied at the UCCS laboratory in which this “idle pin” like situation hesitates between a spin-liquid state or very specific magnetic orders. For instance in \(\text{Sr}_2\text{Cr}(\text{PO}_4)_2\) (a rare example of \(\text{Cr}^{2+}\) compound, an unequalled J.T cation), the magnetic ordering between layers leaves \(1/3\)rd of quasi-paramagnetic “idle” S=2 spins, between them while their participation in the 3D magnetic ordering is mandatory. On further cooling, their subsequent ordering at lower temperature requests a lowering of the magnetic symmetry, to overpass the frustration. In \(\text{Cu}_3\text{Te}_2\text{O}_5(\text{OH})_4\), a similar frustrated situation was encountered between chains arranged in an original “magnetic-raft like” topology. The frustrated knots between them, hamper magnetic ordering and also leaves a significant part of nearly-paramagnetic ions into a QSL state down to 1.8K.\(^6\)

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High-pressure ilmenite-type MnVO$_3$: crystal and spin structures in the itinerant-localized-covalent regimes.

Systems with partially filled valence electrons may suffer electronic and structural changes due to instabilities, for instance the Peierls transition (cation dimerization) in VO$_2$ accompanied by a metal-insulator transition (MIT). In here, MnVO$_3$-I in its ilmenite polyform will be presented with such instabilities.

MnVO$_3$-I was reported long time ago by Syono et al.$^{[1]}$. However, the triclinic symmetry ($P\overline{1}$) precluded the authors for a proper structural determination. We have prepared MnVO$_3$-I via high-pressure high-temperature synthesis at 4 GPa and 1100 °C with a Walker-type multi anvil apparatus. The crystal structure was solved from an isolated small single crystal. At 300 K, MnVO$_3$-I possess $P\overline{1}$ symmetry with $a = 5.0177(7)$ Å, $b = 5.0513(7)$ Å, $c = 5.5210(8)$ Å, $\alpha = 116.679(6)$ °, $\beta = 90.044(6)$ ° and $\gamma = 118.924(5)$ ° unit cell parameters. The polymorph represents a distorted version of the ilmenite-type structure with alternating Mn-V honeycomb-layers, Fig. 1a, similar to the recently reported MgVO$_3$ and CoVO$_3$.$^{[2,3]}$

The triclinic distortion in MnVO$_3$-I arises from V-V dimerization with a short bond of 2.85 Å as shown in Figure 1. It also shows an AFM transition at $T_N = 77$ K with a Curie-Weiss behaviour above 250 K and presents a $|\theta_W| = 233(1)$ K and a $\mu_{\text{eff}} = 5.7(1)$ $\mu_B$. The latter is close to the theoretical value of 5.92 $\mu_B$ for Mn$^{2+}$.

I will also present our results on the structural evolution at high-temperature along with the MIT transition and the low-temperature magnetic structure with respective refined moments of 3.7(1) $\mu_B$ and 0.45(1) $\mu_B$ for Mn and V at 1.5 K.

Referénces:
Construction of room-temperature magnets from redox-active building blocks

Magnets derived from molecule-based precursors have been viewed as emerging materials for next-generation technologies.¹ These materials offer several commercial advantages compared to their inorganic counterparts such as reducing device fabrication costs (e.g., low-energy production, high abundance of elemental sources) and combining magnetic properties with other physical properties (e.g., conductive, mechanical). To realize their potential, careful consideration must be given to the design of magnetic architectures with strong and directional exchange interactions between spin carriers. In this regard, metal-organic frameworks derived from redox-active building blocks have shown to be promising candidates.²,³ This presentation will focus on the importance of molecular design and synthetic strategies to overcome the performance limitations in molecule-based magnets through the post-synthetic reduction of the ferrimagnetic network Cr³⁺(pyz)(pyz)⁺Cl₂ (pyz = pyrazine). The resulting transformation affords the material Li₀.7[Cr²⁺(pyz⁻)₂]Cl₀.7•0.25(THF) (THF = tetrahydrofuran), which shows a dramatic increase of the critical temperature (i.e., from 55 K to 515 K) and a 7500 Oe room-temperature coercivity.

Références :
Eutectics of (Al$_2$O$_3$-YAG):Ce for Phosphor Converters of White Solid-State Lighting Driven by Laser Diodes

Currently, a trend in white solid-state lighting with phosphor conversion (PC-WSSL) is to use a scheme in which a laser diode (LD) with a wavelength of 450 nm irradiates a remote phosphor converter. Such a scheme allows the production of a high-intensity flux but also requires new converters that can withstand a high irradiation load of tens of Watts.

Eutectics of (Al$_2$O$_3$-YAG):Ce meets all requirements for the phosphor converters and due to “Chinese script” morphology possesses advantages in thermal conductivity and light scattering among other materials like ceramics, single crystals, and phosphors in glass (PIG).

In this work, the comprehensive study of the (Al$_2$O$_3$-YAG):Ce eutectics for phosphor converters of PC-WSSL has been carried out. The study consists of both theoretical modeling and experimental investigations. It was theoretically predicted and experimentally shown that the materials with scattering centers increase twofold the light output [1].

(Al$_2$O$_3$-YAG):Ce eutectics of different morphology were grown by the Horizontal Directed Crystallization of the melt of the mixtures of Al$_2$O$_3$, Y$_2$O$_3$, and CeO$_2$ powders. An effect of the growing parameters and Ce concentration on the morphology of the eutectic as mechanical properties was established. A deviation from the Jackson-Hunt model was revealed.

A study of luminescence and decay kinetic revealed emissions that correspond to 1) the emission of the Ce$^{3+}$ ions in the YAG environment at $\lambda_{em}$=500-650 nm, a decay time $\tau$= 62 ns, 2) Ce$^{3+}$ ions in the Al$_2$O$_3$ environment at $\lambda_{em}$=398 nm, $\tau$= 35 ns, 3) F$^+$ centers $\lambda_{em}$=395 nm, $\tau$= 2 ns that disappeared under the air annealing [2].

It has been established that the chromaticity coordinates (CC) and correlated color temperature (CCT) were independent of the LD power up to 3 W. Only minor dependence of these parameters on LD power was revealed for the samples of different morphology. CC and CCT can be changed by the thickness of the samples. The quantum yield of the blue light conversion doesn't depend on the LD power. The converter efficiency reaches more than 200 lm/W of the LD power.

The indicatrix of the transmitted beam has Lambertian distribution.

References:
Poster
"Dynamique hors équilibre des matériaux quantiques sondée par diffusion Raman résolue en temps"

Dans ce poster, nous présentons un nouveau dispositif de diffusion Raman résolue en temps capable de sonder la dynamique hors-équilibre induite par une impulsion lumineuse dans les matériaux quantiques. Le dispositif est basé sur une source laser Yb amplifiée émettant à 1.03 micron couplée à un compresseur externe permettant d’obtenir des impulsions de 60fs à des taux de répétition de 125kHz. Les faisceaux de sonde et de pompe sont séparés spectralement par génération de second harmonique sur la sonde permettant ainsi de réaliser une configuration à deux couleurs de type pompe proche-infrarouge (1.2 eV) sonde Raman visible (2.4eV). Les impulsions de sonde Raman sont formées spectralement afin d’obtenir le meilleur compromis entre résolutions temporelle et énergétique. Les résultats obtenus avec ce dispositif sur les effets photo-induits dans l’isolant excitonique présumé Ta2NiSe5 et dans la phase isolante antiferromagnétique du cuprate Bi2Sr2CaCu2O8 seront présentés.

Référence :

**AC calorimetry in diamond anvil cells: method comparison using the heavy-fermion superconductor CeCoIn$_5$**

A useful method to characterize magnetic transitions or bulk superconductivity in diamond anvil cells (DAC) is AC calorimetry. The sample is thermally excited at a specific frequency, and a value for the specific heat is then retrieved from the thermal response of the sample. At a suitable frequency range, the sample is effectively decoupled from the cell, but not from the heating device and the thermometer.

Several ways are available to setup our heating device. For example, optic heating using laser pulses have proven to be very efficient in a lot of cases. However, we might want to place our DAC in a magnetic field and allow *in situ* rotation of the cell, which forces us to heat with an electric current. In this case, we can use either a thermocouple by directly welding Au and AuFe wires to the sample [1], or we could try passing a less-disturbing weaker current through a resistive resin.

In this poster, we compare both methods by analysing the superconducting transition of the well-known heavy fermion CeCoIn$_5$ ($T_c \approx 2.3$K) [2], due to its highly noticeable specific heat jump. The results show that using a resistive resin (made from epoxy and conductive carbon paste) seems to yield the best results.

**Références :**


Advanced Transmission Electron Microscopy Study of The Metallic and Mott Insulator Phases of \((V_{1-x}Cr_x)_{2}O_3\)

Mott insulators are a class of material which are predicted to be metal according to the conventional band theory, but behave as insulators due to the strong electron-electron repulsion that is not taken into consideration in the conventional band theory. The insulator to metal transition (IMT) in the prototypical Mott insulator \((V_{1-x}Cr_x)_{2}O_3\) has attracted enormous attention either from the fundamental point of view or for its applications in microelectronics. Several mechanisms can lead to such an IMT in this compound, such as manipulating the temperature and the pressure, or chemical doping. Besides these conventional ways of driving an IMT, another stimulus that has recently emerged in Mott insulators is the application of electric pulses. An electric pulse above a certain threshold leads to creation of nanometer-sized filamentary metallic paths in the Mott insulator matrix, yielding to a low resistance state. This low resistance state can relax back to the high resistance state by application of an electric pulse [1]. Hence, this material can switch between different resistance states, paving the way for memory and artificial synapse applications. The nature of the electric-pulse-induced metallic state is currently hotly debated.

In this context, advanced characterizations are clearly required, ideally at the (sub)nanometer scale. Thanks to the recent advances in transmission electron microscopy (TEM), probing this IMT in Mott insulators is now feasible at atomic scale. Before engaging an in-situ study of the electric-pulse-induced IMT, we have characterized the main spectroscopic and structural features of the \((V_{0.95}Cr_{0.05})_{2}O_3\) Mott insulator and \(V_2O_3\) paramagnetic metal at room temperature. In this poster, we will present the result of electron energy loss spectroscopy (EELS) and HAADF-STEM atomic resolution images of both phases. In particular, we unveil a difference in the energy of the plasmon oscillation between the metallic and insulating phases that provides a spectroscopic criterion to distinguish between these two phases as a consequence of the change in the electronic structure. In addition, our high-resolution images allow us to detect a structural differences between the Mott insulator and metallic phases, in particular a detectable change in the Vanadium-Vanadium distance in the V-V dimer.

Références :
Bulk layered non superconducting (Ln\textsubscript{1-x}A\textsubscript{x})NiO\textsubscript{2+y} nickelates

(Ln = Nd, Pr; A = Sr, Ca)

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The discovery of superconductivity below $T_c \sim 15$ K in thin films epitaxially grown on SrTiO$_3$ (STO) of doped infinite layer (Nd\textsubscript{1-x}Sr\textsubscript{x})NiO$_2$ (Nd,Sr) nickelate mid-2019 has generated an enormous interest in the condensed matter community because of their great similitude with cuprates [1]. Then, superconductivity was also reported in films with other rare earth elements and Ca\textsuperscript{2+} instead of Sr\textsuperscript{2+} as dopant, i.e. in La,Sr (or Ca) [2] and Pr,Sr [3] based systems, and finally in thin films of Pr\textsubscript{1-x}Sr\textsubscript{x}NiO$_2$, not grown on STO, but on (LaAlO$_3$)$_{0.3}$(Sr$_2$AlTaO$_6$)$_{0.7}$ (LSAT) substrates [4] with maximal $T_c$ up to 20 K.

At Néel Institute, we have synthesized and characterized good quality polycrystalline samples of the infinite layer (Pr\textsubscript{1-x}Sr\textsubscript{x})NiO$_2$, (Nd\textsubscript{1-x}Sr\textsubscript{x})NiO$_2$, and (Nd\textsubscript{1-x}Ca\textsubscript{x})NiO$_2$ series doped with Sr or Ca up to $x$(Sr) = 0.3 and $x$(Ca) = 0.5 respectively. So far, no superconductivity was found in any of these compounds but a spin-glass like and insulating behavior at low temperature is observed. The temperature dependence of the crystal structure of the (Nd\textsubscript{1-x}Ca\textsubscript{x})NiO$_2$ compounds with $x$=0.1, 0.2, 0.3 were investigated by high resolution synchrotron x-ray and neutron diffraction down to 4 K.

We also synthesized a new non-superconducting polycrystalline partially reduced form of NdNiO$_3$: NdNiO$_{2+y}$. Although its XRD pattern looks very similar to the one reported by Moriga et al. [7], the crystal structure we found by Transmission Electron Microscopy is totally different. The Selected Area Electron Diffraction (SAED) evidenced a modulated structure in a tetragonal subcell with the parameters $a \approx 3.8$ Å and $c \approx 3.5$ Å. Due to the commensurate nature of the modulation, the structure can be described in a monoclinic supercell with the parameters $a \approx 16.4$ Å, $b \approx 7.8$ Å, $c \approx 5.5$ Å and $\beta = 105^\circ$. A first structural model determined using 3D ED crystallography is currently refined thanks to synchrotron XRD and neutron diffraction data.

References:
In this poster, we consider the repulsive Hubbard model in the strong coupling limit $t \ll U$ and perform a mean field decoupling called composite operator method (COM). Two sets of self-consistent equations are considered: the first one enforces Pauli principle and the second imposes charge-charge, spin-spin and pair-pair correlations through a decoupling scheme formalized by L. Roth. Two distinct solutions named « COM1 » and « COM2 » can be extracted from the self-consistency imposing Pauli principle. In addition, one unique solution can be obtained from the self-consistency with the Roth decoupling. We review these three solutions with nearest-neighbours hopping on square lattice. For each solution we study the validity of particle-hole symmetry and Luttinger theorem. Finally, we extend the initial basis to study superconductivity. We conclude it is induced by the Van Hove singularity.

Composite operator treatment of the Hubbard model in the strongly correlated limit and superconductivity

In this poster, we consider the repulsive Hubbard model in the strong coupling limit $t \ll U$ and perform a mean field decoupling called composite operator method (COM). Two sets of self-consistent equations are considered: the first one enforces Pauli principle and the second imposes charge-charge, spin-spin and pair-pair correlations through a decoupling scheme formalized by L. Roth. Two distincts solutions named « COM1 » and « COM2 » can be extracted from the self-consistency imposing Pauli principle. In addition, one unique solution can be obtained from the self-consistency with the Roth decoupling. We review these three solutions with nearest-neighbours hopping on square lattice. For each solution we study the validity of particle-hole symmetry and Luttinger theorem. Finally, we extend the initial basis to study superconductivity. We conclude it is induced by the Van Hove singularity.
Fluctuation-Dissipation relation in spin ice

Geometrical frustration in magnetism has become a challenge in modern condensed matter physics. Frustration is source of exotic ground states that are still a problem for physicists. Spin ice is an example of this kind of state where the magnetism is governed by local rules giving rises to a degenerate ground state. The excitations of this ground state can be described as magnetically charged quasiparticles, called magnetic monopoles[1]. In this system, at very low temperature, spins can only flip by quantum tunneling [2]. In addition to geometrical considerations, this results in stringent constraints to the motion of magnetic monopoles and gives rise to far from equilibrium metastable states [3].

In order to probe this out-of-equilibrium state in spin ice we developed a measurement technique based on the fluctuation-dissipation relation. The experiment use an ultra-sensitive SQUID magnetometer which can measure magnetic noise and AC susceptibility in the same setup down to very low temperature (200mK). This allow the precise determination of deviations from the fluctuation-dissipation relation in spin ice for the first time, and thus to characterize quantitatively the out-of-equilibrium state and the monopole dynamics at very low temperature.

The aim of the poster is to present the current results we obtained from this experiment and how we can go further to investigate this mysterious metastable state.

Références :
Fluoride-based magnetic metal-organic frameworks for CO$_2$ capture and sequestration

With carbon dioxide (CO$_2$) being the largest contributor to greenhouse gas emissions, there exists a crucial need in today’s society to effectively capture and sequester atmospheric CO$_2$ from both environmental and industrial perspectives. Existing carbon capture technologies are simply too expensive to be practical. Meanwhile, the development of new technologies that imprison CO$_2$ at low concentrations from gas mixtures in an efficient method is still a major challenge faced by researchers. [1,2] Regarding this matter, metal-organic frameworks (MOFs) have been viewed as emerging materials that can be chemically engineered to possess high thermal robustness and appealing physisorption properties in order to greatly improve the energy efficiency of small molecule gas separation. [3] Following this avenue, novel three-dimensional MOFs of the unit formula Cu(py2)$_2$MF$_6$ (M = Ti, Ir, Os) with critical pore sizes for CO$_2$ capture have been synthesized and structurally characterized by single crystal X-ray diffraction. By incorporating paramagnetic MF$_6$ units, the promotion of magnetic coupling between the spin carriers was achieved, which lead to the stabilization of magnetically ordered phases. This presentation will discuss the facile and efficient methodology towards developing these magnetic MOFs and CO$_2$ absorption studies using these materials.

![Figure: View of the porous 3D structure of [Cu(py2)$_2$MF$_6$] compounds with imprisoned CO$_2$ molecules.](image)

Références :


Investigating the Rashba effect at the interface of 2D transition metal dichalcogenides and ferroelectrics

2D crystals have emerged as a new class of materials that can exhibit a large range of quantum phenomena such as superconductivity, 2D magnetism, and non-trivial topological phases [1], [2]. Of these quantum phenomena, the Rashba effect has gained popularity recently as a result of its applicability to create charge-to-spin interconversion for next-generation spintronics devices. Highly stable PtSe$_2$ is one such 2D material which has a centrosymmetric crystal unit cell and a band-structure that evolves drastically as a function of the number of monolayers deposited, evolving from a high band-gap semiconductor to a semi-metal [3]. Interfacing a 2D material with a ferroelectric (FE) crystal applies a strong local electric field at the interface which breaks the inversion symmetry and triggers the interfacial Rashba effect in the 2D layer. Importantly, the sign of the Rashba spin-orbit coupling depends on the direction of the FE polarisation, such that the spin texture chirality should reverse upon a change in direction of the FE polarisation [4].

Here, we use 2D layers of inversion-symmetric PtSe$_2$ on top of ferroelectric LiNbO$_3$ and BiFeO$_3$ surfaces to investigate the interfacial Rashba effect and corresponding spin-to-charge conversion. Differences between 2D layers on up and down polarization are highlighted.

Références :

Magnetostriction and thermal expansion in the superconducting state of UTe$_2$

Magnetostriction and thermal expansion, respectively the length change of a material under the application of an external magnetic field or a temperature change is a very powerful thermodynamic bulk measurement technique. The latter is sensitive to entropy changes occurring under pressure and necessary to calculate the Grüneisen parameter. Magnetostriction reveals magnetization changes under pressure and opens another possibility to study the irreversible magnetization due to the vortex lattice pinning in type II superconductors.

Here we report magnetostriction and thermal expansion studies performed on the superconducting state of the novel heavy fermion superconductor UTe$_2$ [1] up to 30 T for magnetic fields along the b-axis. We observe different irreversibility lines between the low field and high field reinforced superconducting phase, together with a detailed study of the specific heat, leading to the assumption of two different pairing mechanism along the b-axis in UTe$_2$ [2]. Furthermore, we present the hysteretic behavior of the low field irreversible magnetostriction, revealing a complicated critical current behavior in UTe$_2$.

Références:
Modelling Oxygen and Rare-Earth atomic impurities in Nickelates.

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An exciting new discovery has been made in the field of rare-earth nickelates, with the recent observation of superconductivity in hole-doped infinite layered nickelate $R_1-xR'xNiO_2$. This development has prompted a need to revisit the electronic structure and physical properties of rare-earth nickelates, such as $Nd_{1-x}Sr_xNiO_2$. To achieve this, researchers have employed a two-step experimental approach, involving the reduction of perovskite rare-earth nickelates to obtain layered rare-earth nickelates, followed by substitutional doping in relevant stoichiometric ratios. These systems exhibit a range of conventional and unconventional phenomena, including absence of superconductivity in bulk nickelate, limited thickness of the infinite-layer phase in thin films, absence of long-range anti-ferromagnetism, and suspect of the origin of superconductivity from the interface. Theoretical hypotheses are being put forth to explain these phenomena, including the Kondo effect, multi-orbital effect, Hund's coupling effect, and disorder effect. To shed light on the crucial ingredients manipulating superconductivity in film and bulk nickelates, researchers are simulating Friedel oscillations in the dilute impurity regime with two kinds of impurities, an extra apical Oxygen (O) impurity and/or Strontium (Sr) atom substituting Neodymium (Nd), using the Tight Binding (TB) model. The aim is to model disorder and the impact of electronic correlations from rare-earth materials, with the model parameters chosen phenomenologically and further benchmarked against previous available results.
Raman spectroscopy with uni-axial strain on iron based superconductor

Presentation of Raman spectra obtained on Co-BaFe$_2$As$_2$ under applied strain along two different directions within the (a,b)-plane (Fe-Fe and Fe-As-Fe bonds).

Références :


SIGNATURE OF SUPERCONDUCTOR HIGGS MODE IN NSN JUNCTION

All properties of superconductors can be explained by a spontaneous gauge symmetry breaking (SSB) $U(1) \rightarrow Z_2$. This SSB leads to the appearance of a massive collective mode corresponding to the oscillation of the order parameter, i.e. the superconductor gap. Recently this amplitude mode, the Higgs mode of the superconducting order parameter, has been measured in superconductors using ultra fast THz techniques [1]. After complete calculation of the response of a clean superconductor submitted to a monochromatic irradiation using the Eilenberger formalism, we study a new method to detect the mode using NSN clean junction. We show that a signature of the Higgs mode can be seen in the differential conductance of the dc current.

Références :
Spectrométrie de masse des ions secondaires : application aux couches minces d'oxydes

Cette communication s'inscrit dans la perspective d'étudier et de comprendre la supraconductivité dans des nickelates supraconductrices. Un contrôle fin de la composition chimique des échantillons y est crucial, notamment la stoichiométrie en oxygène et la quantification de l'hydrogène incorporé au cours de l'ultime étape de synthèse. L'analyse par spectrométrie de masse d'ions secondaires (SIMS) est a priori particulièrement adaptée à ces éléments légers et constitue même pour l'hydrogène l'unique technique de détection directe et résolue en profondeur dans des films minces. En atteste la confirmation en mars 2023 de la présence d'hydrogène incorporé dans des films de nickelate supraconductrice obtenue par analyse SIMS [1].

Mais l'analyse par SIMS de films minces constitue un défi technique. Les faibles vitesses de pulvérisation qu'imposent les faibles épaisseurs sondées réduisent en proportion l'intensité ionique. De plus, le mécanisme de pulvérisation résulte du transfert d'énergie des ions du faisceau primaire vers le cristal qui se produit par des collisions en cascades. Si l'énergie primaire est suffisante, l'ordre cristallographique est détruit à l'intérieur du volume dans lequel les collisions se produisent et toutes les espèces constitutives du matériau sont mélangées avec les ions primaires. Les ions secondaires détectés par le dispositif expérimental sont émis depuis cette zone chaotique, appelée zone de mélange collisionnel, qui se propage en profondeur dans le matériau au cours de la pulvérisation. En conséquence, la résolution en profondeur de l'analyse est limitée par l'amplitude du mélange collisionnel qui doit impérativement être inférieure à l'amplitude de la couche sondée. La forte dépendance de ce paramètre avec les conditions d'analyse et l'environnement chimique analysé empêchent l'établissement de lois prédictives. Pour connaître la taille du mélange collisionnel et il est donc nécessaire d'en faire directement la mesure dans les conditions d'expérience visées et sur un échantillon de composition chimique le plus proche possible.

Dans la perspective d'analyser des films minces de nickelates supraconductrices, nous avons réalisé des mesures préliminaires sur des multicouches de compositions variées, incluant nickelates et titanates. En se basant sur un modèle phénoménologique du mécanisme de pulvérisation [2], nous avons développé une procédure d'analyse numérique de nos résultats. Nous avons montré que l'élargissement des profils d'intensité ionique au franchissement d'une interface idéale est dû à l'étendue du mélange collisionnel d'une part et à la rugosité de surface d'autre part. La modélisation des profils permet de séparer ces deux contributions. La rugosité étant mesurée par AFM dans les fonds de cratères de pulvérisation, le modèle informe ainsi sur la taille du mélange collisionnel dans les conditions d'expérience.


Unconventional superconductivity (SC) and magnetism are usually mutually exclusive. However, magnetic order seems a necessary foe of unconventional SC. It is particularly the case in cuprates (AFM order in the vicinity of the SC state) or Fe-based SC (spin density wave close to the SC state). The current obstacles in the understanding of Cooper pairing mechanism are related to the dimensionality of the systems. Considering quasi-1D systems should be much simpler and should lead to the emergence of new theoretical models. Fe-based spin-ladders of the type BaFe2X3 (X=S, Se) are particularly interesting quasi-1D compounds. They are insulators at room temperature and ambient pressure. In addition, BaFe2Se3 is ferrielectric at 300K. Below TN (250 K for BaFe2Se3 and 120 K for BaFe2S3), a magnetic order develops along the spin ladders resulting in multiferroic property. The magnetic and dielectric behaviours are strongly different in the two types of spin-ladders which is not yet understood. Under pressure, a SC transition occurs in both systems leading to a very rich phase diagram as a function of pressure and temperature. The influence of other external parameters, such as magnetic/electric fields, charge doping, chemical substitution or ionic irradiation remains to be investigated with a reasonable hope to discover new phases, new properties and a better understanding of theses materials. Indeed, each of these parameters is likely to favor one or another instability among the many orders present in this family. By destabilizing the fragile balance between the different underlying order parameters, we expect to reach new remarkable ground states. This is the objectif of my PhD. To study this family, we use a large range of experimental techniques: X-ray diffraction (atomic structure), IR (phonons).

Références :
Superconductivity in monolayer and few-layer graphene: II. Topological edge states and Chern numbers

Abstract (1 page maximum)

Références:
Theoretical study of the magnetic properties of the CoCu$_2$O$_3$ compound

Julien Lévêque$^{1,2}$, Elisa Rebolini$^3$, Marie-Bernadette Lepetit$^{2,3}$, and Andrés Saúl$^1$

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Over the last decades cuprates have attracted a lot of attention following the discovery of high temperature superconductivity. Apart from superconductivity, a large number of compounds of the cuprate family have been studied for their low-dimensional quantum magnetism. Indeed, the $S = 1/2$ character of the Cu$^{2+}$ ion, and the directionality of the associated 3d magnetic orbital, are responsible for a tendency to form one- or two-dimensional magnetic systems with quantum character. This leads to high exchange integrals and thus to a high magnetic ordering temperature.

Among them, the ACu$_2$O$_3$ family ($A = $ Ca, Mg, Co) presents a 3D magnetic order associated with high transition temperatures. Although this family shows very similar structures characterized by puckered layers, the magnetic order and the associated transition temperature changes with the element A [1,2]. In particular, the compound CoCu$_2$O$_3$ presents the highest magnetic transition temperature of this family (215 K) [3].

In this presentation, we study the magnetic properties of this compound using ab-initio and classical Monte-Carlo calculation methods. The exchange integrals have been calculated with a MRCI method allowing to treat exactly the electronic correlation [4]. We find that the Co atom plays a preponderant role in the magnetic properties of this compound. Indeed, as a magnetic ion, it creates additional magnetic interactions. This results in a magnetic pattern based on coupled three-leg ladders, different from the two-leg structural ladders. Moreover, we show that the observed high transition temperature is related to its single ion anisotropy [5].

Figure: Schematic of the magnetic order of CoCu$_2$O$_3$ on the puckered magnetic layers.

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Références:
Tuning the electronic properties of two-dimensional (2D) coordination networks via 3d-metal substitution

Molecular spintronics represent an emerging field of research that bridges the quantum spin of an electron with the physical properties of molecules.\(^1\) A quintessential aspect to the design of these molecule-based materials is the ability to control the interactions between the spin-active sites in a predictable manner. Given the endless possibilities of molecular building-blocks, hybrid blends of metal ions and organic ligands offer untapped potential to tune and explore novel functionalities in future technologies that may otherwise be inconceivable to their inorganic counterparts. In this regard, a new ground has been laid in this field through the coordination of redox-active CrCl\(_2\) with pyrazine (pyz) bridging ligands affording Cr(pyz)\(_2\)Cl\(_2\), a 2D coordination network with remarkable physical properties. Below 55 K, Cr(pyz)\(_2\)Cl\(_2\) displays a ferrimagnetic ordered state, and at room temperature, the material exhibits electrical conductivity up to 32 mS cm\(^{-1}\).\(^2\) These extraordinary properties stem from an intrinsic one-electron reduction from the Cr\(^{II}\) ion to one of the neutral pyrazine ligands, which subsequently transforms the 2D network into a highly delocalized system and simultaneously promotes strong magnetic exchanges between the spin carriers. In the pursue of tuning these properties through structural modifications, we herein present the synthesis and characterization of the isostructural vanadium and titanium analogues: V(pyz)\(_2\)Cl\(_2\) and Ti(pyz)\(_2\)Cl\(_2\).\(^3\) This presentation will communicate the contrasting nature of the metal ions within these networks and the resulting impact on the magnetic and conductive properties.

\[\text{Figure: Perspective view of the 2D structure of } M(\text{pyrazine})_2\text{Cl}_2 \text{ (where, } M = \text{V and Ti) along } c\text{-direction.}\]

Références:
Investigating the magnetic properties of $\text{BaCo}_2(\text{AsO}_4)_2$: 
Magnetisation steps and Hamiltonian simulation

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We report on the observation of new magnetisation steps at very low temperature in the antiferromagnetic material $\text{BaCo}_2(\text{AsO}_4)_2$, which magnetic structure is still an enigma since the first works 40 years ago \cite{Regnault1983} and continues to be debated \cite{Zhong2020}. By combining neutron scattering measurements, spin wave and Monte Carlo simulations, we found a classical Heisenberg Hamiltonian with interactions up to the fourth nearest neighbours, which partially reproduces the spin waves dispersion observed in our experiment. We have synthesised substituted samples to test the effect of chemical pressure and the influence of the magnetic dilution on the propagation vector of $\text{BaCo}_2(\text{AsO}_4)_2$. These results are a step forwards identifying the magnetic structure of this compound.

\cite{Regnault1983, Zhong2020}
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