

AAPG2023	NEXUS		JCJC
Coordinated by:	Dalila Bounoua	48 months	246k€
CE 30 Physique de la matière condensée et de la matière diluée			

NEXUS: Hidden Magnetic Texture in the Pseudogap Phase of High-Tc Cuprates

Summary table of persons involved in the project:

Partner	Name	First name	Current position	Role & responsibilities in the project (4 lines max)	Involvement (person.month) throughout the project's total duration
Laboratoire Léon Brillouin (UMR 12 CEA-CNRS)	Bounoua	Dalila	Researcher	Coordinator Tasks A.1-3, B.1-3, C1-2	39 p.month
	Bourges	Philippe	Researcher	Task B.1-3, C.1-2	8.5 p.month
	Sidis	Yvan	Researcher	Task. B1-3	8.5 p.month

Any changes that have been made in the full proposal compared to the pre-proposal / compared to the registration

The present full proposal for the NEXUS project is in fully line with the pre-proposal from the scientific point of view. The postdoc was converted into a thesis, which will help the coordinator gain experience in PhD thesis supervision. The need for consumables was also reevaluated and the budget was revised accordingly leading to a decrease of the requested amount from 270 to 246 k€.

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I. Proposal's context, positioning and objective(s)

a. State of the art

The race for room temperature superconductivity never ceased to be in full swing since its discovery in 1911. This fascinating macroscopic quantum phenomenon, where the material loses its resistivity to current flow below a critical temperature (T_c), stands out as a solution to societal challenges relevant to energy, transport, and computation fields and is already at play in several modern infrastructures.

At the current state of the art, the highest critical temperatures at ambient pressure were found to be achieved in a class of copper oxide based materials discovered in 1986, namely cuprates. The pristine cuprate materials are antiferromagnetic Mott insulators where hole/electron doping induces a superconducting state that survives, in best cases, up to 164K, above the liquid nitrogen temperature. Over the years, cuprate superconductors shook the scientific community. They have proven to be an inexhaustible well of exotic electronic instabilities that stimulated a wide range of deep experimental and theoretical investigations that lay at the heart of modern condensed matter physics.

The mechanism leading to the formation of Cooper electron pairs in the superconducting state of these systems has been a matter of 35 years continuous debate but remains elusive. Indeed, these unconventional superconductors behave as bad metals in their normal state and exhibit a d-wave superconducting gap that cannot be accounted for by the conventional electron-phonon coupling mediated Cooper pair formation, proposed by Bardeen, Cooper and Schrieffer [1]. Instead, a mechanism based on magnetic interactions is generally put forward and very recently got a strong experimental support from Scanning Tunneling Microscopy that shows a compelling correlation between the changes of the charge transfer-gap (controlling the magnetic superexchange interaction) and the Cooper pair density [2].

Phase diagram of high- T_c superconducting cuprates

Cuprate superconductors are single or multi- CuO_2 layered compounds. They exhibit a complex electronic phase diagram (**Figure.1**) where starting from an antiferromagnetic (AF) Mott insulating state, where copper spins lie within the CuO_2 [a,b] planes, increasing the hole doping quickly destroys the long-range commensurate AF order leaving only antiferromagnetic fluctuations [3,4]. The hole doping further leads to the emergence of multiple electronic instabilities as shown on **Fig.1**:

- i) The superconducting (SC) dome at low temperature defined by the T_c line and characterized by a d-wave superconducting gap, surrounded by:
- ii) The enigmatic pseudogap (PG) phase where the electronic density of states is depleted, in the underdoped region. The PG is bound by the T^* temperature line and characterized by the disappearance of large portions of the Fermi surface leaving only Fermi arcs as seen by Angle Resolved Photoemission Spectroscopy (ARPES). Its fingerprint is seen on spectroscopic or transport measurements although no sharp anomaly is seen at T^* in specific heat measurements [5–7].
- iii) The strange metal phase characterized by an electric resistivity linear in temperature and a large hole Fermi surface, beyond the PG.

Further increasing the hole doping level results in a conventional Fermi liquid state with a T^2 dependent resistivity.

Among these phases, one of the most debated is the PG phase, believed to be a key issue towards the understanding of the superconducting mechanism, either playing the role of a preemptive state for superconductivity, or of an order parameter characterized by broken symmetries that instead competes with superconductivity [3,4]. However, since its discovery [8], the origin of this mysterious electronic state of matter remains a puzzle and no obvious lattice translational symmetry breaking at T^* that could explain the occurrence of the PG was reported [6,9].

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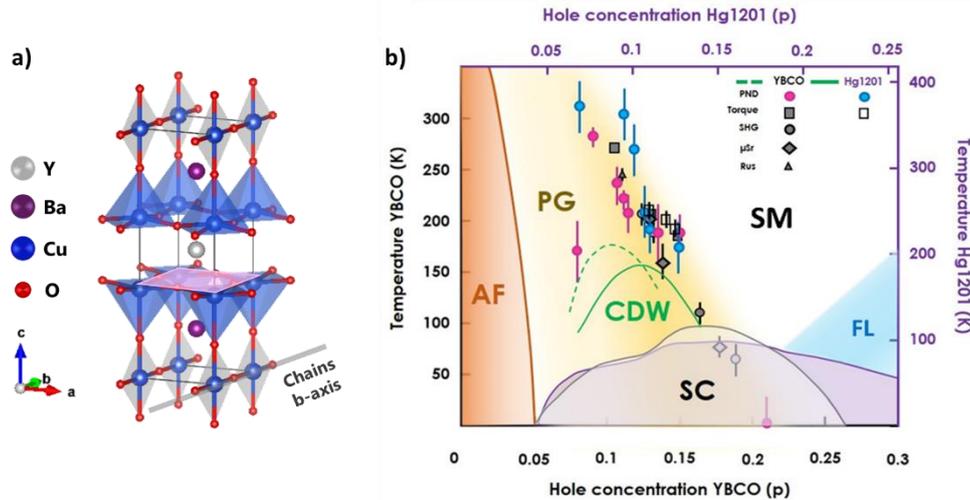


Fig.1.a) Crystal structure of the high- T_c cuprate $YBa_2Cu_3O_{6+x}$ the pink surface represents the CuO_2 planes and the arrow indicates the CuO chains. **b)** Summary phase diagram of hole-doped 2D cuprates. The symbols represent Time reversal symmetry breaking reported by PND, μ SR, Torque and Parity symmetry breaking reported by Second Harmonic Generation [12]. AF is the antiferromagnetic phase, PG the pseudogap, CDW the charge density wave, SC the superconducting state, SM the strange metal and FL the Fermi liquid. YBCO stands for $YBa_2Cu_3O_{6+\delta}$, Hg-1201 stands for $HgBa_2CuO_{4+\delta}$.

The pseudogap state

Charge density wave: At a temperature $T < T^*$, an incipient charge density wave (CDW) phase was extensively reported. The CDW modulations break the lattice translational symmetry (LT) and induce a lattice superstructure. It exhibits short-range correlations within the CuO_2 planes and lead to a uni/bi-axial response at incommensurate planar wave-vectors in reciprocal space ($q \neq 0$).

At zero magnetic field, CDW modulations are quasi-2D and weaken when entering the SC state at temperatures $T < T_c$. The CDW instability competes with SC. Indeed, upon applying either an external magnetic field a uni-axial pressure along the a-axis, 3D CDW correlations develop while SC is suppressed under magnetic field [10].

Although it breaks the lattice translation (LT) symmetry, its onset temperature T_{CDW} remains well below T^* , deep into the PG phase, and can thus not be alone at the origin of the opening of the electronic gap. Recently, a long-range CDW phase was further reported in the overdoped $Tl_2Ba_2CuO_{6+\delta}$, reinforcing the idea that the CDW phenomenon cannot alone account for the PG opening [11].

In the meantime, decades of experimental work show the occurrence of spontaneous discrete (Z_2) Ising symmetries breaking at the T^* onset temperature of the PG phase. The corresponding broken symmetries are time reversal (T), parity (P) and the C_4 rotation symmetry. Since, the PG onset exhibits no sharp anomaly in the specific heat data at T^* , these symmetry breakings are then associated to the establishment of a “hidden order parameter”.

Discrete symmetry braking at T^* : Thorough Polarized Neutron Diffraction (PND) measurements revealed the onset of a hidden intra-unit cell (IUC) **antiferromagnetic** order ($q=0$ magnetism) at T^* , breaking T and preserving LT in four distinct cuprate families: $YBa_2Cu_3O_{7-\delta}$, $(La,Sr)_2CuO_4$, $Bi_2Sr_2CaCu_2O_{8+\delta}$ and $HgBa_2CuO_{4+\delta}$ [12].

The $q=0$ magnetism is commensurate with the crystal lattice and thus appears on top of nuclear Bragg peaks. It exhibits long-range correlation lengths that are PND momentum resolution limited.

While classical interpretations in terms of dipole (spin or orbital) magnetism fail to reproduce the experimental observations, patterns involving exotic quantum magneto-electric (ME) loop currents (LC) states [13], successfully account for the PND results (Fig.2.a).

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The loop current model

The LC three-band model, originally proposed by C.M. Varma in 2006 to account for the electronic properties of the PG, predicts the existence of spontaneously circulating currents between the copper and oxygen sites within the CuO_2 sheets (**Fig.2.a**) with two counter-propagating loops per Cu site, giving each, rise to a magnetic moment perpendicular to the CuO_2 planes [13]. The LCs building blocks can correlate from site to site, leading to an IUC or $q=0$ AF response.

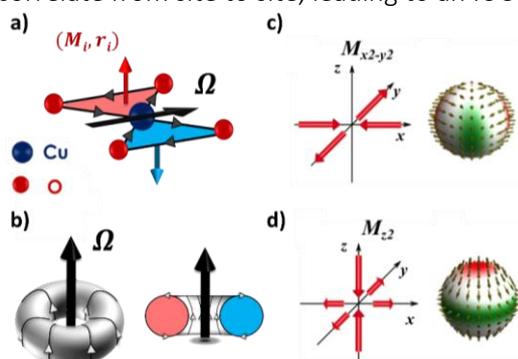


Fig.2. (a) Two counter-propagating LC between Cu and O ions, giving rise to magnetic moments (M_+ , M_-), with zero effective magnetization at each site, and an anapole, in the CuO_2 plane, corresponding to the CC-O_{II} LC pattern proposed by Varma. (b) Polar toroidal moment (black arrow Ω) associated with loop currents (LC) around the solenoid. The LC state is found by a 2D section of the solenoid [13]. (c-d) Local distribution of the magnetization density produced by magnetic dipoles (in red) leading to the formation of a magnetoelectric quadrupole. Two symmetries M_z^2 and $M_{x^2-y^2}$ are represented. Adapted from [18].

The LC quantum state can be described by an **anapole** or a polar toroidal moment (**Fig.2.b**), perpendicular to the staggered orbital moments produced by winding currents around a solenoid. It is fourfold degenerate with four possible quantum states corresponding to distinct orientations of the anapoles (**Fig.3**) [13]. LC are further expected to generate a non-zero **quadrupole** component (**Fig.2.c-d**).

The anapoles and quadrupoles appear at the second order term of the multipolar expansion series beyond the usual first order magnetic dipole (spin and orbital) moment and since the LC phase breaks both **T** and **P**, it is described as a ME multipolar phase. Quadrupoles and anapoles however involve distinct electronic distributions: centred at the Cu-atomic sites for in the former case while the delocalized over the Cu and O sites for the LCs.

In addition to the model proposed by Varma, the ME-LC state appears as an ancillary preemptive or vestigial state bound to PG phase in various theories [14–16] while a purely quadrupolar order was suggested to be at the origin of **T**-breaking in the PG [17,18]. Parent LC patterns were further proposed as an ancillary phase in square-planar spin liquids [14].

Experimental evidence

The IUC magnetism observed using PND was among the first reported order parameters in the PG state, which starts at a temperature T_{mag} that matches T^ .* Apart from PND, there is an abundant literature reporting the **T** and **P** symmetry breaking in the PG phase.

Actually, the **T**-breaking was confirmed by several other probes such as muon spectroscopy (μSR) [19], torque magnetometry [20,21] and Kerr effect experiments [22] while Optical Second Harmonic Generation (SHG) measurements give a compelling evidence for the **P**-breaking, at T^* [23], as expected for the LC state (**Fig.2.a**).

The occurrence of discrete symmetry breaking at T^* changes the perspective of the PG. Considered for a long time as a crossover phenomenon owing to the absence of a specific heat anomaly at T^* , the precise analysis of the high accuracy magnetization measurements [24], resonant ultrasound spectroscopy [25] and torque measurements [20,21] demonstrate that the PG line corresponds instead to a true thermodynamic phase transition.

It is worth emphasizing that the detection of the IUC LC (or $q=0$) magnetism signatures can be achieved using dedicated diffraction techniques, although very challenging, owing to the weak amplitude of the LC magnetic moment ($\sim 0.1\mu_B$), being further superimposed to the intrinsic response of the underlying crystal lattice, which is several orders of magnitude greater. *Its signature can thus only be revealed by dedicated (T/P)-symmetry sensitive probes equipped with polarization analysis such as PND [12,17] and Resonant X-ray Diffraction (RXD).*

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Orientation of the magnetic moment and LC fluctuations

It is noteworthy that although the $q=0$ magnetism appears static at the PND energy resolution scale (0.1 meV), Nuclear Magnetic Resonance (NMR) remains blind to the LC magnetism, unable to detect the local static fields produced by the LC orbital moment [26]. A critical slowing down of magnetic fluctuations at T^* was however reported by μ SR with a characteristic fluctuating time of (2-10 ns) [19]. The existence of such slow fluctuations with timescales laying between the PND (100 ps) and NMR ($\leq 10 \mu$ s) detection thresholds may constitute the missing link between both techniques, making the LC magnetism appearing static at the PND timescale while it evades the detection window of NMR.

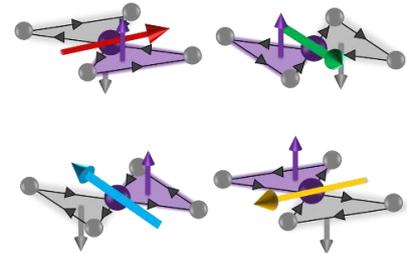


Fig.3. Four possible degenerate ground states of loop currents (LC). The red, blue, green and yellow arrows represent anapoles centered at the Cu-site.

Such fluctuations could arise from the flipping between the four possible quantum states of the LCs (**Fig.3**).

In particular, while the magnetic moments associated to LCs magnetism are expected to be strictly perpendicular to the CuO_2 planes, PND experiments reveal a systematic tilt of 45° to the CuO_2 planes (**Fig.4**) regardless of the doping level and the Cu coordination environment [12].

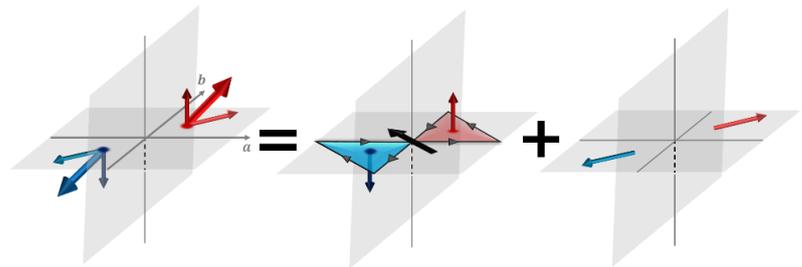


Fig.4. Representation of a possible magnetic moment arrangement compatible with the PND results. It corresponds to the superposition of an out-of-plane moments as the LC phase with an in-plane quadrupole [12].

A possible scenario is that the existence of fluctuations could lead to the appearance of a planar magnetic component yielding an apparent tilt of the magnetic moment. An alternative scenario for the interpretation of the magnetic moment tilt is the existence of a quadrupole that superimposes to the anapole at the Cu site (**Fig.4**) [12].

Beyond High-Tc cuprates

In a wider perspective, and with different physical origins, the existence of LC-like states seems now ubiquitous in a wide range of quantum materials.

For instance, the presence of a LC phase is likely to influence the anomalous magnetic properties of the quasi-2D $\text{Sr}_2(\text{Ir,Rh})\text{O}_4$ [27] and SrRuO_4 [28]. We also reported a LC-like phase in the quasi-1D spin liquid $(\text{Sr,Ca})_{14}\text{Cu}_{24}\text{O}_{41}$ [29] and a consistent **T**-breaking was shown to occur in the Kagomé Cs_3VSb_5 superconductor [18-19]. The fundamental and original property of such ME-states is that, unlike multiferroic materials where the magnetic and charge degrees of freedom are distinct order parameters, they exhibit an intrinsic coupling between magnetic and charge degrees of freedom in a single order parameter paving the way, in the long run, to applications in quantum computing. Such ME-coupling effects were recently demonstrated in $\text{Mn}_3\text{Si}_2\text{Te}_6$ [32] and CsV_3Sb_5 [33].

b. Novelty and preliminary data

While the existence of the $q=0$ magnetism and its related **T** and **P** breaking *can be taken for granted*, the fact that it preserves the LT symmetry naturally questions its relevance to the PG opening. Apart from the IUC magnetism arising from LC order, other unconventional magnetic correlations involving alternative LT-symmetry breaking charge current patterns were discussed in cuprates. In this context, while early proposals dealt with a LC phase preserving **LT**-invariance, a more recent work proposes the existence of modulated LC states breaking **LT** instead, able to lead to the PG opening [34]. Consistently, we recently discovered the existence of novel magnetic correlations within the PG phase of the high-Tc $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ that may fit within this description [34,35].

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Discovery of the $q=1/2$ magnetism within the PG phase of $YBa_2Cu_3O_{6+\delta}$

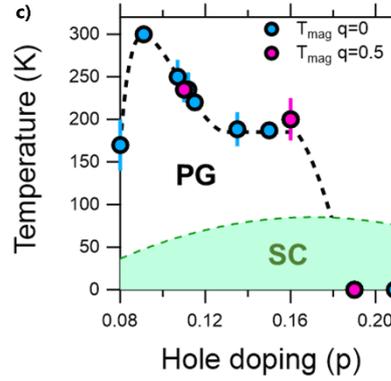
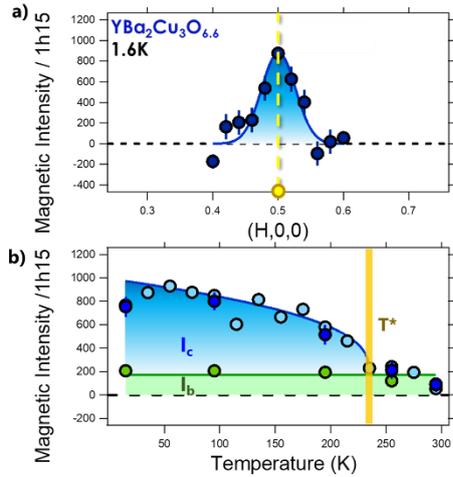


Fig.5. a) Scan along $(H,0,0)$ in reciprocal space obtained by PND showing a magnetic signal centered at $q=1/2=(0.5,0,0)$ in the PG phase of $YBa_2Cu_3O_{6.6}$. **b)** Temperature dependence of the magnetic intensity at $(0.5,0,0)$ as extracted from neutron polarization analysis. I_c and I_b are the out of plane (perpendicular to the CuO_2 plane) and in plane magnetic components, respectively [34]. **c)** Summary phase diagram of the observed onset temperature, T_{mag} , for the $q=0$ and $q=0.5$ magnetism vs doping [35,36]

Our recent PND studies in $YBa_2Cu_3O_{6+x}$ samples at different hole doping (p) levels: underdoped $p \sim 0.11$, optimally doped $p \sim 0.16$ and slightly overdoped $p \sim 0.19$, uncovered the existence of a new form of magnetism hidden in the PG phase of these high- T_c SC cuprates (**Fig.5.a**) [34,35].

Up to optimal doping, our investigations systematically show the existence of a new static magnetic response at planar wave vectors of the form $q=(0.5,0) \equiv (0,0.5)$ in r.l.u (**Fig.5.a**), hereafter labelled **$q=1/2$** , that vanish in the slightly overdoped regime (**Fig.5.c**). The **$q=1/2$** magnetism settles in, on cooling down from room temperature, at T^* (**Fig.5.b**), the PG and **$q=0$** magnetism onset temperature highlighting its intimate link to the PG physics.

Our measurements in a detwinned $YBa_2Cu_3O_{6.6}$ ($p \sim 0.16$) sample (where the a and b axis can be clearly identified) show that the $q=1/2$ magnetism arises from bi-axial correlations within the CuO_2 planes (**Fig.1.a**), leading to a doubling of the unit cell along both the a and the b-axis; and correspondingly to magnetic scattering at the planar wavevectors $q=(0.5,0)$ and $(0,0.5)$. The associated correlation lengths are short $\sim 25\text{\AA}$ in plane (about 5-6 unit cells) and $\sim 13\text{\AA}$ along the c-axis (about one unit cell).

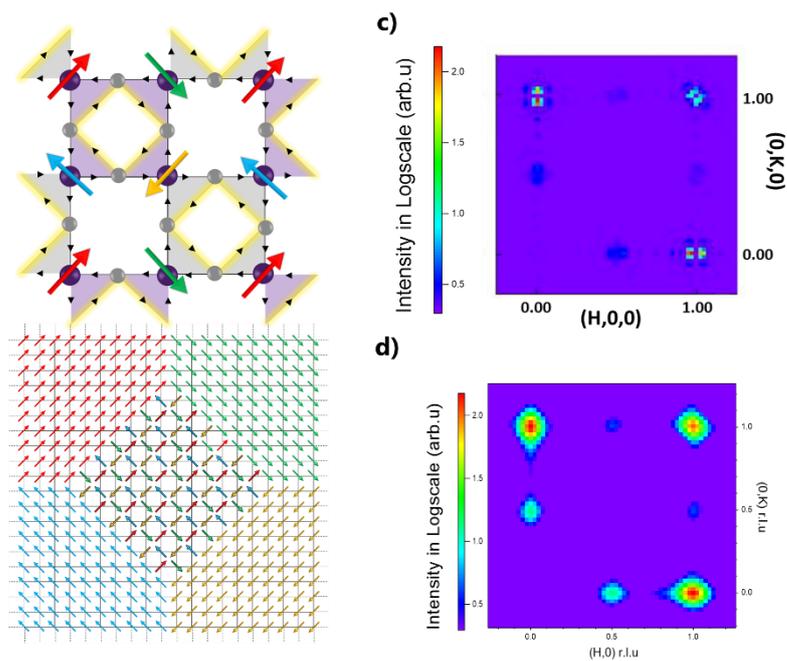
The remarkable correspondence between the **$q=0$** and **$q=1/2$** magnetism temperature dependencies and amplitudes of the magnetic moment ($\sim 0.1 \mu_B$) indicates that both phases share a common microscopic origin. Consistently, the orientation of the corresponding magnetic moment decomposes into a leading out of plane magnetic component and a smaller in-plane magnetic component (**Fig.5.b**), favoring an interpretation in terms of LCs rather than Cu spins that are expected to lay in plane owing to a strong planar magnetic anisotropy [34]. A 2×2 LCs pattern doubling the unit cell along both a- and the b-axis is able to satisfactorily reproduce the data (**Fig.6.a**).

Put together, the **$q=0$** and **$q=1/2$** magnetic scattering revealed by PND could belong to a unique complex magnetic texture of CuO_2 unit cells hosting LC (**Fig. 6.b**). Such a magnetic texture would be made of 4 large ferro-anapolar domains made of **$q=0$** LC order (with LC patterns rotated by 90° from one domain to the next) and, at their corner, an anapole-vortex like bubble of intertwined LC, doubling the unit cell along the a- and b-axis and leading to the **$q=1/2$** magnetic response. By analogy to the electronic liquid crystals where smectic modulations coexist with an IUC nematicity, one can speculate about the coexistence of a “smectic” short range (**$q=1/2$**) 2×2 LC magnetism with a “nematic” (**$q=0$**) longer range magnetism (ferro-anapolar) [36].

The existence of such a LC magnetic texture of size $2P \times 2P$ (P being the number of CuO_2 unit cells) yields an incommensurate magnetic structure. This LC supercell breaks the LT-symmetry and should lead to an incommensurate magnetic response at $q = 0 \pm \frac{1}{2P}$ that may have remained hidden (or blurred) by the low PND experimental resolution where the response appears centred at $q \sim 0$ in former reports (**Fig. 6.c-d**). Such patterns would be able to induce small displacements of the Fermi surface by a reciprocal vector k directly linked to the value of P and reproduce the Fermi arcs observed in the PG phase [37].

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Fig.6. a) 2×2 loop currents pattern that can account for the $q=1/2$ magnetism. The currents circulate clockwise (in gray) and anti-clockwise (in purple). The four states are represented by anapoles undergoing a 90° rotation between adjacent domains. **b)** Example of a 2D magnetic texture with 20×20 unit cells paved by anapoles (LC states). The central cluster describes the $q=1/2$ short-range magnetism whereas the IUC magnetic signal arises from the larger color domains. **c)** Structure factor calculation in log-scale for the LC magnetic pattern in (b), without convolution with the instrumental resolution, and **d)** convoluted with the instrumental resolution. The (H, K) intensity map in (c) shows the incommensurate magnetic scattering at $(1,0)$, $(0,1)$ and $(1,1)$ and at $(0.5,0)$ and $(0,0.5)$. The convolution by the PND instrumental resolution in d) blurs the incommensurate peaks that appear at $q=0$ [35-36].



Since our first published work in $\text{YBa}_2\text{Cu}_3\text{O}_{6.6}$, a recent Lorentz Transmission Electron Microscopy experiment reports the existence of a skyrmion like magnetic texture in the PG phase of $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$. As discussed by the authors, such a picture could correspond to the LC texture we propose (Fig.6.b) [38].

c. Objectives of the NEXUS project

The NEXUS project aims at a systematic investigation of the new $q=1/2$ magnetic correlations and their connection with the $q=0$ magnetism in different cuprate families, using state of the art neutron scattering and resonant X-ray diffraction techniques.

NEXUS relies on an interdisciplinary strategy combining material science and condensed matter physics, and targeting three cuprates families: $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and $\text{HgBa}_2\text{CuO}_{4+\delta}$ and $\text{Bi}_2(\text{La,Sr})_2\text{CuO}_{6+\delta}$.

The first objective is probing the universality of the $q=1/2$ magnetism in the PG phase of the different cuprates families and its characterization (as a function of doping, temperature, magnetic field).

The second objective is to probing the existence of incommensurate magnetic peaks in the PG phase that would be the direct proof of LT symmetry breaking and the link between the LC magnetism and occurrence of the PG.

The third objective is to probe the existence of fluctuations of the $q=0$ and $q=1/2$ LC magnetism using dedicated neutron techniques that would help bridging the gap between PND and NMR .

The work plan is divided in three work-packages (WP). This segmentation allows a linear chronological approach in order to establish a coherent and progressive schedule of tasks to meet the ambitious objectives of this project. The summary in the form of a GANTT diagram (Tab.2) specifies the timetable and the decomposition of the tasks. It appears just after the detailed description of the different tasks.

d. Methodology : Tasks and Timeline

As the coordinator of the NEXUS, I will be responsible for each of the tasks presented below. Some of the tasks will further involve collaborations with experts in PND and RXD to have the needed task force to ensure the successful achievement of the experiments. The crystal growth using the self-flux method of which I am no an expert will also involve a collaboration with experimented solid-state chemists.

The program includes the recruitment of a PhD student and three master trainees that will all be actively involved in the synthesis, characterization and large-scale facilities experiments following the timeline of the project (Tab.2).

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WPA: Single crystal growth and characterization

The crystal growth represents the **cornerstone** of the NEXUS project. Indeed, **WPB & C** strongly depend on the success in obtaining single crystals of the three targeted cuprates: $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and $\text{HgBa}_2\text{CuO}_{4+\delta}$ and $\text{Bi}_2(\text{La,Sr})_2\text{CuO}_{6+\delta}$ since the timeline of the project will be rhythmmed by the availability of the materials (Tab.2). The choice of these cuprate families stems from the fact that the PG line in their corresponding electronic phase diagram is clearly identified. Additional arguments are detailed case-by-case in **Task.A.1**.

The main idea is to achieve, for each family, at least two underdoped, one optimally doped and one overdoped single crystals to cover the electronic phase diagram. The single crystal growth will be carried using two techniques: the travelling solvent floating zone (TSFZ) technique using a four mirrors image furnace and the self-flux method.

✓ **Task. A.1. Growth of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and $\text{Bi}_2(\text{La,Sr})_2\text{CuO}_{6+\delta}$ single crystals**

The travelling solvent floating zone (TSFZ) growth technique (Fig.7) offers the ability to grow centimetric oriented crystals in a reasonable time (1 to several days depending on the growth rate) perfectly suitable for both neutron and RXD measurements. It is particularly adapted to the growth of compounds with complex thermodynamic phase diagrams such as the incongruently melting $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and $\text{Bi}_2(\text{La,Sr})_2\text{CuO}_{6+\delta}$ (Fig.7.b-d). An advantage of the TSFZ technique is the absence crucible contact that significantly reduces external sources of pollution and allows a precise control of dopant amounts.

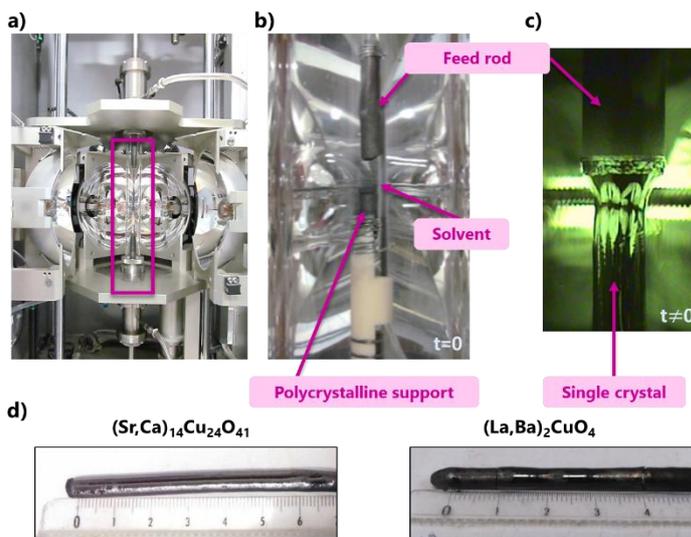


Fig.7. Crystal growth by the optical floating zone (OFZ). a) Four mirror image furnace setup, similar model to the one purchased by LLB/SPEC. The pink frame shows the growth chamber (zoomed in in b) surrounded by a quartz tube allowing the conditioning of the growth atmosphere. b) Starting growth setup comprising a polycrystalline feed rod, a polycrystalline support and a solvent pellet in between (for the travelling solvent floating zone procedure TSFZ) at $t < t_s$. c) Screenshot during the growth, at the stationary state t_s , including the feed rod, the grown single crystal and the molten zone in between. d) Examples of centimetric size single crystals of incongruently melting compounds grown using TSFZ.

Case of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$: The motivations for the growth of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ samples are multiple. First, the $q=0$ magnetism is well documented in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ [12] and the $q=1/2$ magnetism starts to be well characterized [34,35] in this compound and gives solid starting grounds towards the study of the interplay between both phases. The second motivation is having a unique source of samples grown by the same TSFZ method owing to the advantages offered by this technique in terms of crystal quality, chemical homogeneity and sample size.

Indeed, single crystals of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, are usually synthesized using the self-flux method or equivalent top-seeded solution growth due to the instability of the TSFZ route. The well-known problem with stability of the $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ TSFZ growth is the low viscosity of the molten zone and the porosity of the feed rod. The stabilization of the floating zone crystal growth process was however achieved recently [39]. I will use these same starting conditions and extend the growth experiments to the overdoped region once suitable conditions are isolated to achieve $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ compositions.

The growth of large single crystals would further allow shortening the experiment preparation time, with no necessary co-alignment of several single crystals from different batches (up to 60 single crystals in our previous experiments) as when self-flux grown.

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Case of $\text{Bi}_2(\text{La,Sr})_2\text{CuO}_{6+\delta}$: The reasons for choosing this family of compounds are multiple. First, La-doping allows to tune the hole doping, covering almost the entire electronic phase diagram as shown by Fig.8.a and reaching the overdoped regime through the adjustment of the La-content [40].

The second motivation is that neither the $q=0$ or $q=1/2$ magnetism were ever investigated in this compound although the $q=0$ magnetism was reported in the parent Pb doped $\text{Bi}_2\text{Sr}_2\text{CuO}_{6+\delta}$ and $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ [12]. The aim will be then to search for both $q=0$ and $q=1/2$ magnetisms as seen in the related cuprates families and probe their evolution as a function of hole (La) doping.

The crystal growth route of $\text{Bi}_2(\text{La,Sr})_2\text{CuO}_{6+\delta}$ is well documented and controlled although of its being challenging due the incongruent melting issues which requires attentive monitoring.

I have already initiated the work of solid-state powders synthesis of this compound through the hiring of an apprentice (O. Haimoud). This successful preliminary work lead to the synthesis of single-phased materials with appropriate La-doping.

The first year of the NEXUS project will be dedicated to the $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ samples growth that will be carried by the coordinator (Q1-Q4). Once the suitable conditions identified, a PhD student hired starting from year 2 (Q5) will join the team and be involved in the growth of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and $\text{Bi}_2(\text{La,Sr})_2\text{CuO}_{6+\delta}$, supported by two Master 2 trainees. The first trainee (hired in Q2) will proceed to the characterization of the $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ single crystals and the solid-state synthesis and characterization of the needed powders of $\text{Bi}_2(\text{La,Sr})_2\text{CuO}_{6+\delta}$. The second trainee (Q6) will be involved in the growth and characterization of the $\text{Bi}_2(\text{La,Sr})_2\text{CuO}_{6+\delta}$ single crystals.

✓ Task. A.2. Growth of $\text{HgBa}_2\text{CuO}_{4+\delta}$

The motivation for the growth of $\text{HgBa}_2\text{CuO}_{4+\delta}$ is that the $q=0$ LC magnetism is well characterized in this material while the $q=1/2$ magnetism was never probed so far.

The growth of $\text{HgBa}_2\text{CuO}_{4+\delta}$ samples will be carried in collaboration with Dr. D. Colson and A. Forget (SPEC laboratory) who developed an original and efficient method for the synthesis of high quality single crystals of $\text{HgBa}_2\text{CuO}_{4+\delta}$ using a self-flux method. The accessible doping range, covers the electronic phase diagram, reaching the overdoped regime through oxygen doping or gold substitution [41].

The obtained crystals are square shaped with typical size of few mm suitable for RXD and were already user in our pilot experiments (see Task.C.2). The use of the single crystals for neutron scattering requires the co-alignment of several crystals to have a sufficient volume of material.

This task will involve the coordinator, Dr. D. Colson and A. Forget and will be supported by a Master 2 trainee hired at (Q10) for the growth, characterization and the co-alignment of the samples.

✓ Task. A.3. Doping and characterization of the single crystals

Doping the samples: Through sintering under an appropriate atmosphere of oxygen. To ensure the efficient doping on single crystals, which require long diffusion times, thermogravimetric analysis will be carried on pieces of single crystals to optimize the efficiency of the doping procedure.

Structural characterizations: will be carried on the SPEC XRD powder diffractometer to check the phase purity of powders and single crystals and determine the different structural parameters (lattice parameters, atomic positions and sites occupancy). In $\text{Bi}_2(\text{La,Sr})_2\text{CuO}_{6+\delta}$, the site occupancy will allow determining the La-effective amount and since the compound exhibits structural transitions upon doping, it will bring a crosscheck information about the hole doing level. This characterization is further important for $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ where the c-axis parameter strongly depends on the doping level.

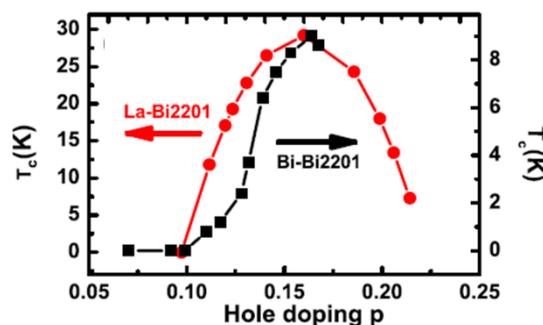


Fig.8.a. a) Low temperature electronic phase diagram of La-doped $\text{Bi}_2\text{Sr}_2\text{CuO}_{6+\delta}$ compared to oxygen doped compound [40].

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Chemical characterization Quantitative chemical analysis for over (hole-) doped $\text{Bi}_2(\text{La,Sr})_2\text{CuO}_{6+\delta}$ samples (low La-doping) can be carried on the ICP (Inductively Coupled Plasma Spectroscopy) apparatus of Département de Mesures Physiques at Université Paris Saclay. It will aim at checking the effective incorporation and the precise amount of La within the as grown crystals.

Magnetic and specific heat measurements

The MPMS platform of LLB is a modular installation able to perform series of physical characterizations such as VSM and AC magnetization and specific heat measurements under different conditions of temperature (1.7 to 100K) and field (0 to 9T).

Magnetization measurements will first aim at determining the T_c of the powders and single crystals to check the doping level and its homogeneity. Additionally, magnetization and specific heat constitute sensitive probes. These measurements will be systematically conducted on single crystals to rule out the existence of impurity phases that may fall below the threshold of detection of XRD.

The aforementioned characterizations will be subsequent to all the solid-state synthesis and crystal growths. They will be carried by the coordinator, the PhD student and the master trainees and will follow the timeline of (Tasks A.1-2).

WP2. Systematic study of the LC magnetism using neutron scattering

Neutron scattering equipped with polarization analysis (XYZ-PA) is a state-of-art tool that proved to be efficient in probing the $\mathbf{q}=0$ and $\mathbf{q}=1/2$ LC magnetism, at the heart of the NEXUS project.

The principle of a polarized neutron scattering experiment relies on the preparation of an incident beam of neutrons with a given neutron spin polarization and analysing the final state of the neutron beam spin polarization after interaction with the sample. Indeed, any source of magnetism within the sample is able to flip the neutron spin. The neutron beam can be polarized along different directions defined within the experimental setup coordinates. Usually, one refers to (X,Y,Z) such X and Y lay within the scattering plane and Z perpendicular to the scattering plane. Relying on the dipolar nature of the neutron interaction and the Pauli matrices that describe the nature of the interaction along the polarization direction, the intensity sum-rule over the cross sections collected along different directions then allows the extraction of the purely magnetic scattering with no assumption on the background.

The power of XYZ-PA is that it allows the precise determination of the orientation matrix of the measured magnetic moments and unambiguously discriminating the magnetic signal from a nuclear signal when both superimpose in reciprocal space as is the case of the $\mathbf{q}=0$ magnetism.

✓ **Task B.1. Hidden magnetic texture in the PG phase of High- T_c cuprates**

The aim of this task is investigating the existence of a hidden LC magnetic texture in the PG phase of three cuprates families: $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and $\text{HgBa}_2\text{CuO}_{4+\delta}$ and $\text{Bi}_2(\text{La,Sr})_2\text{CuO}_{6+\delta}$, using PND.

The experimental work will consist in characterizing the evolution of the $\mathbf{q}=1/2$ magnetism onset temperature, correlation lengths, amplitude and magnetic moment orientation as a function of temperature and hole-doping and check its disappearance in the overdoped regime that would confirm its direct link with the PG. The characteristics of the $\mathbf{q}=0$ and $\mathbf{q}=1/2$ magnetism will be connected and put into comparison to describe the corresponding magnetic texture.

A particular focus of this task will be the precise determination of the structure factor of the $\mathbf{q}=1/2$ magnetism of which knowledge is still missing. Although this novel magnetism does not superimpose to nuclear Bragg peaks, rendering the measurement of the associated magnetic signal much easier than at $\mathbf{q}=0$, the amplitude of the corresponding magnetic moment remains small and will benefit from the use of XYZ-PA to disentangle it from the nuclear and incoherent background.

A second focus of this task will be the investigation of the $\mathbf{q}=0$ LC magnetism in $\text{Bi}_2(\text{La,Sr})_2\text{CuO}_{6+\delta}$. The magnetic signal will be probed on nuclear Bragg peaks of the form (1,0,0)/(0,1,0) based on the comprehensive experimental work carried by PND in earlier studies [12].

A third focus will be the determination of the magnetic moment orientation evolution versus doping. Indeed, although the orientation of the $\mathbf{q}=1/2$ LC magnetism remains mainly out-of-plane at all doping levels in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, the ratio of the in-plane to the out of plane components decreases upon increasing

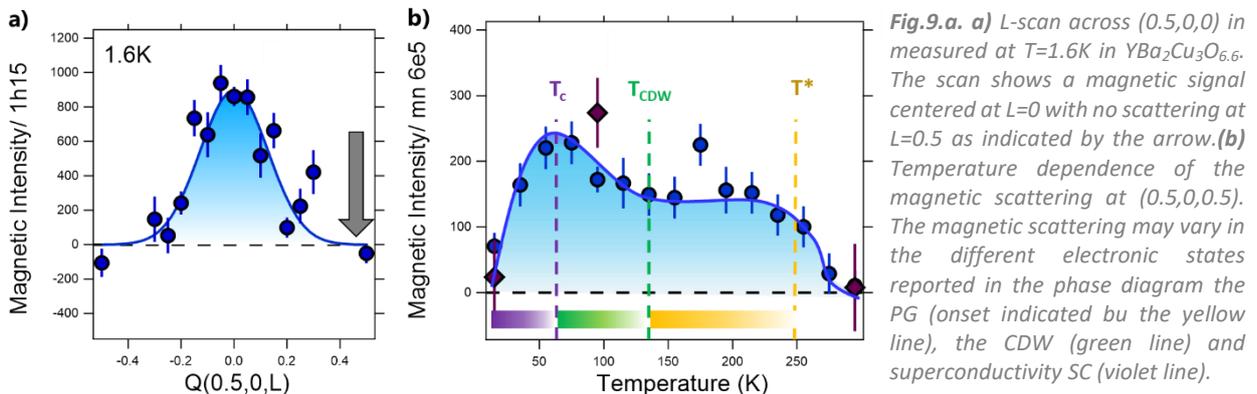
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the hole-doping [34,35]. This point needs to be systematically established in other cuprates to gain insight into the doping dependence of the planar component of which origin remains unclear. An interesting scenario would be that the planar component gets systematically enhanced at optimal doping, as approaching a quantum critical point at the ending point of the T^* line, beneath the SC dome [5]. In this scenario, the planar component would be a signature of LC fluctuations [19]. These investigations will provide a set of data that will allow generalizing the existence of the $q=1/2$ magnetism involved in a magnetic texture hidden within the PG phase of high- T_c cuprates across the full electronic phase diagram.

This task will involve the Coordinator, the PhD student and Dr. P. Bourges and Dr. Y. Sidis following the timeline presented in Table 2.

✓ **Task B.2. Interplay of the $q=1/2$ magnetism with other electronic instabilities**

Another interesting question that this project will address is the existence of an interplay between the $q=1/2$ magnetism and other electronic instabilities within the phase diagram of high- T_c cuprates. While the link with the $q=0$ magnetism seems natural and unavoidable, our preliminary data (unpublished) indicate that the c -axis correlations exhibit interesting features. Indeed, an additional magnetic signal is observed at momentum positions of the form $(0.5,0,0.5)$ in both underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{6.6}$ and optimally doped $\text{YBa}_2\text{Cu}_3\text{O}_{6.9}$, further doubling the unit cell along the c -axis with a peculiar temperature dependence that may be affected at key temperature corresponding to the onset of the CDW and superconductivity (**Fig.9.a-b**). This peculiar behaviour hints at an intriguing interplay between the different electronic instabilities existing in the PG and was only observed in a detwinned single crystal up to date. In particular, it seems to affect correlations within the $[a,c]$ plane, while only a smooth temperature dependence was observed in twinned crystals [35].



To establish the possible interaction between the LC magnetism, the CDW and the SC, these investigations need to be pushed by pursuing two directions. The first one will naturally consist in investigating the evolution of the c -axis correlations at different doping levels, in particular, at optimal doping where the CDW is suppressed. The second direction we will pursue is the study of the magnetic field dependent behavior of the $q=1/2$ magnetism. Indeed, while the $q=0$ magnetism superimposes to the strong nuclear peaks that would make such a measurement a complex task, the $q=1/2$ magnetism offers a unique opportunity to address this question.

In the same spirit as the extensive studies carried on the CDW electronic instability [10], the idea is to explore the field, doping and temperature dependence of the correlation lengths, intensity, and any wavevector changes of the $q=1/2$ magnetism. In particular, in the picture we propose of a magnetic texture involving both kinds of magnetic phases, a field dependent study will bring insight into the interplay between the $q=0$ and $q=1/2$ magnetism and the behavior of the domain wall boundaries. This study will also allow complementing the field-dependent phase diagram of the electronic instabilities reported in the PG so far.

This task will involve the Coordinator, the PhD student and Dr. P. Bourges and Dr. Y. Sidis following the timeline presented in Table 2.

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✓ **Task.B.3. $q=1/2$ magnetism fluctuations**

LCs fluctuations at T^* were already detected near $q=0$ by PND in nearly optimally doped $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ and in $\text{HgBa}_2\text{CuO}_{4+\delta}$ [12]. This task aims at investigating the existence of fluctuations related to the $q=1/2$ magnetism. Indeed, owing to the short-range correlation lengths of the 2×2 magnetism clusters (about 25 \AA planar correlations); the $q=1/2$ magnetic correlations are inevitably fluctuating at a time-scale that needs to be determined. The corresponding fluctuations will be probed in two ways:

First, using classical inelastic neutron spectroscopy (INS) on triple axis spectrometers and time of flight spectrometers at the corresponding reciprocal space positions, to determine their momentum and energy dependence. The resolutions that can be achieved on INS spectrometers is of the order of 1 meV giving access to characteristic time-scales at the order of 100 ps .

Second, using neutron spin-echo spectroscopy equipped with XYZ-PA that allows scanning Fourier times lying in the range of (1 ps - 20 ns). Indeed, according to μSr measurements, a critical slowing down of magnetic fluctuations occurs at T^* , the PG onset, leading to the T breaking. The corresponding magnetic fluctuations are slow with a time-scale of about 10 ns .

The aim of the neutron spectroscopy experiments will be the determination of the time-scale of fluctuations and probe its evolution as a function of doping and temperature down to below T_c , and their fate in the strange metal phase above T^* .

This task will involve the Coordinator, Dr. N. Martin, Dr. P. Bourges and Dr. Y. Sidis following the timeline presented in Table 2.

These PND experiments will be carried on triple axis spectrometers and diffractometers equipped with XYZ-PA worldwide. The INS measurements will be performed on triple axis/time-of-flight spectrometers or spin-écho spectrometers. The experiments will be performed at ILL (France), SINQ (Switzerland), FRMIII (Germany), ORNL (USA) and the upcoming ESS spallation source (Sweden, by 2026).

WP3. Search for the $q=1/2$ and $q=0$ and magnetism using RXD

The first aim of this WP is probing the existence of the $q=1/2$ magnetism using a different T/P sensitive advanced diffraction technique, namely, RXD equipped with polarization analysis. The second objective is the investigation of incommensurate magnetic peaks at the shoulders of the nuclear Bragg peaks that would attest for the effective breaking of the LT symmetry.

RXD represents a powerful asset towards the detection of the LC multipolar phases through their anapole/quadrupole. The excitation of the electrons of a selected ion, at the resonance edge, leads to the enhancement of the magnetic signal and the use of linear or circular polarization analysis of the scattered beam allows disentangling the magnetic and nuclear contributions. RXD is complementary to PND since it offers the opportunity of precisely determining the magnetic structure factor owing to the absence of limitation by the magnetic form factor. We will perform the measurements at the K and L absorption edges of Cu as described in tasks (C.1-2), in order to access the terms corresponding to the anapoles and quadrupoles appearing in the dipolar-quadrupolar interference term (E1-E2) [42]. Our experiments will include azimuthal (ψ) scans, in different polarizations. Indeed, the presence of a magnetic signal arising from the (E1-E2) term is expected to lead to modulation of the polarization as a function of ψ . These experiments will be guided by PND data, theoretical predictions [17] and pilot experiments that we already performed in cuprates.

✓ **Task C.1. $q=1/2$ generalization**

These RXD measurements will be performed at the Cu L_3 -edge ($\sim 930 \text{ eV}$). The first objective is to confirm the observation of $q=1/2$ magnetism in $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ samples where the signal is well characterized from PND measurements. The next objective is to highlight the nature of the objects involved in this magnetism by determining the azimuthal dependence of the corresponding magnetic signal.

These studies will then be extended to the two other cuprates families guided by PND results to demonstrate the universality of $q=1/2$ magnetism to the PG phase of high T_c cuprates.

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In collaboration with the group of Dr. R. Arpaia (Chalmers University of Technology, Sweden) and Dr. G. Ghiringhelli (Politecnico di Milano, Italy), we have already performed a pilot RIXS experiment at the ID32 beamline of ESRF on $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ thin films grown by the group of Dr. Arpaia. The first successful experiment show consistency with the PND data with the occurrence of the $q=1/2$ magnetism in the underdoped regime. The idea is going further probing the hole-doping dependence of these magnetic correlations especially on the same bulk single crystals as the ones used for PND.

The experiments will be carried on ID32 (ESRF, Grenoble), I21 (Diamond Light Source, UK) that allow reaching the corresponding Q-wavevectors at the L-edge.

✓ Task C.2. LT symmetry breaking

The high Q-resolution of RXD will allow assessing the existence of incommensurate magnetic peaks near $q=0$ that would be the direct signature of LT symmetry breaking in the PG phase. We will perform RXD measurements at the Cu-K edge (8990 eV) which grants access to the large wavevectors corresponding to the Bragg peaks $(1,0,0)/(0,1,0)$ where the signal is expected from PND. $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ and $\text{HgBa}_2\text{CuO}_{4+\delta}$ constitute the priority of this task owing to a simpler nuclear structure than $\text{Bi}_2(\text{La,Sr})_2\text{CuO}_{6+\delta}$. RXD investigations at the K-edge will probe the existence of incommensurate magnetic peaks at the shoulders of the Bragg peaks as inferred from PND, their evolution as a function of temperature and doping.

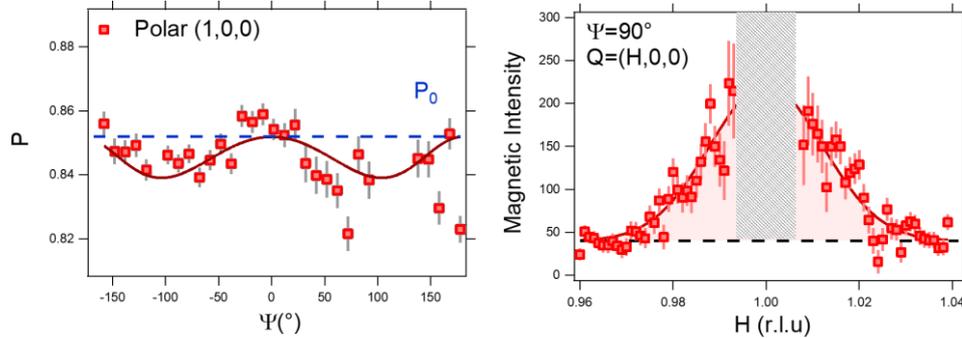


Fig.10. Preliminary RXD results Cu K-edge in $\text{HgBa}_2\text{CuO}_{4+\delta}$. **a)** Evolution of the polarization versus the azimuthal angle. A modulation can be observed such that $P=P_0-\Delta P \sin^2(\psi)$. P_0 is the instrumental polarization at the Cu K-edge while ΔP might correspond to the intrinsic polarization expected from LC or quadrupole contributions [17,42]. **b)** H-scan in the rotated channel at $\psi=90^\circ$ where ΔP is maximum. The signal (red area) appears broader than the experimental resolution and might correspond to short range magnetic signal with finite correlation lengths or incommensurate peaks located at $Q=(1\pm\delta, 0, 0)$ from the Bragg peak $Q=(1,0,0)$. The central dashed area is obscured by the strong Bragg nuclear peak.

We have already performed pilot experiments at the Cu- K absorption edge in an underdoped $\text{HgBa}_2\text{CuO}_{4+\delta}$ sample on the I16 beamline of the Diamond light Source. The experiments were carried in collaboration Victor Balédent (LPS-Orsay) using samples of Dorothée Colson (SPEC). The measurements were performed using linear polarization and collecting the scattered intensity in two channels $\sigma\sigma$ (unrotated) and $\sigma\pi$ (rotated). The analysis of the azimuthal dependence of the polarization $\mathbf{P}=(I_{\sigma\sigma} - I_{\sigma\pi})/(I_{\sigma\sigma} + I_{\sigma\pi})$ reveals a modulation (**Fig.10.a**) that may correspond to a multipole contribution (anapoles or quadrupoles). Our results hint at a possible magnetic signal lying at the shoulders of the Bragg peak (**Fig.10.b**), which may have two interpretations: **i)** the magnetic correlation length of the $q=0$ magnetism is finite or **ii)** the multipole contribution incommensurate with the lattice exhibiting a long period that may correspond the existence of a LC magnetic texture breaking LT in the PG. The broad nature of Bragg peak shoulder could suggest as well a distribution of such super-cells but experimental work is needed to draw a clear picture.

RXD experiments will be made on I16 (DLS, UK), XMaS, D2AM and P09 (Petra, Hamburg) beamlines.

This task will involve the coordinator, Dr. P. Bourges, Dr. Y. Sidis, and Dr. V. Balédent.

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Table 2. Project plan and timeline

Task	Description	Year 1				Year 2				Year 3				Year 4			
		Q1	Q2	Q3	Q4	Q5	Q6	Q7	Q8	Q9	Q10	Q11	Q12	Q13	Q14	Q15	Q16
Management																	
1	PhD student	[Timeline bar from Q4 Year 1 to Q4 Year 4]															
2	Master trainees	[Timeline bar from Q1 Year 1 to Q4 Year 3]															
WPA Crystal growth and characterization																	
A.1	Growth of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and $\text{Bi}_2(\text{La,Sr})_2\text{CuO}_{6+\delta}$ crystals	$\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$				$\text{Bi}_2(\text{La,Sr})_2\text{CuO}_{6+\delta}$											
A.2	Growth of $\text{HgBa}_2\text{CuO}_{4+\delta}$ crystals					$\text{HgBa}_2\text{CuO}_{4+\delta}$											
A.3	Doping and characterization of the single crystals	[Timeline bar from Q1 Year 1 to Q4 Year 3]															
WPB Study of the LC magnetism using neutron scattering																	
B.1	Hidden magnetic texture in the PG phase of High-Tc cuprates					$\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$				$\text{HgBa}_2\text{CuO}_{4+\delta}$ & $\text{Bi}_2(\text{La,Sr})_2\text{CuO}_{6+\delta}$				$\text{HgBa}_2\text{CuO}_{4+\delta}$ & $\text{Bi}_2(\text{La,Sr})_2\text{CuO}_{6+\delta}$			
B.2	Interplay of the $q=1/2$ magnetism with other electronic instabilities									$\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$				$\text{HgBa}_2\text{CuO}_{4+\delta}$			
B.3	$q=1/2$ magnetism fluctuations									$\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$				$\text{HgBa}_2\text{CuO}_{4+\delta}$			
WPC Search for the $q=1/2$ and $q=0$ and magnetism using RXD																	
C.1	$q=1/2$ generalization									$\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$				$\text{HgBa}_2\text{CuO}_{4+\delta}$			
C.2	LT symmetry breaking									$\text{HgBa}_2\text{CuO}_{4+\delta}$				$\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$			

- Milestone** [Blue bar] Staff hiring
- Milestone** [Brown bar] Crystal growth, doping and characterization achieved
- Milestone** [Green bar] Publication of the neutron scattering results
- Milestone** [Purple bar] Publication of the RXD results

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e. Risk management

All of the tasks described above call for many techniques, both at LLB, within the framework of collaborations or on large facilities. High quality single crystals, beamtime access on large-scale facilities and man-power represent the main requirements to successfully carry the ambitious investigations proposed in the NEXUS project.

The first risk is the unsuccessful synthesis of single crystals of the different cuprates families. This risk is however balanced by the coordinator experience in the TSFZ growth of incongruently melting compounds (ranging from 1D to High Tc cuprates [29,43]).

For $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$, the conditions of growth of single crystals using the TSFZ method are already provided by a pioneer study that will serve as starting conditions in our experiments. As a backup, crystals at different doping levels are available from worldwide collaborations (Group of Dr. B. Keimer, MPI and Pr. X. Yao, University of Shanghai) and are already characterized and used in our earlier PND experiments.

In particular, the crystal structure twinning of the crystals obtained by the TSFZ is an unknown parameter. This question is relevant for the **(Task.b.2)** and will be addressed since the very beginning of the single crystal characterizations. As a backup, crystals are available within the framework of a collaboration with the group of Dr. B. Keimer who developed a technique for the detwining of self-flux grown crystals. For $\text{Bi}_2(\text{La,Sr})_2\text{CuO}_{6+\delta}$, the preliminary work done by an apprentice already led to the successful synthesis of powders of wanted compositions.

For $\text{HgBa}_2\text{CuO}_{4+\delta}$, the group of Dr. D. Colson has a long time expertise in the self-flux growth of these cuprates. If the size of the single crystals is however detrimental to PND measurements, back-up single crystals from the group of Dr. M. Greven will be used within the framework of a collaboration that already lead to several publications around the $q=0$ LC magnetism [44].

The second risk is related to the need of work force to handle the growth, the experiments and the data analysis. This is why the timeline of the project places the growth of materials as the early milestone of the project and the requested fund will be principally engaged in the hiring of a PhD student and trainees that along with my experimentalist collaborators will guarantee having the critical mass for the accomplishment of the different tasks.

The third risk comes from the need for beamtime access on large facilities. Indeed, although the measurement protocol for the $q=0$ and $q=1/2$ LC magnetism are under control, the small amplitude of the magnetic signal requires long measuring times.

As a regular user of European neutron and more recently synchrotron facilities, I developed an expertise in writing proposals for beam time requests and achieved a high success rate over the last 5 years. My beam time access amounts to about 6 weeks neutron (ILL, PSI and ISIS without counting Orphée) experiments and 3 weeks synchrotron experiments per year (ESRF, Soleil and the DLS).

With the upcoming ESS spallation source by year 3 of the project, this time will be increased, especially since the LLB is involved in the building of several instruments at ESS. Among those instruments, a polarized single crystal neutron diffractometer MAGIC and a time of flight neutron spectrometer BIFROST include the LCs magnetism within their scientific case. This will benefit to this project since experiments will be possible starting from the hot commissioning period of the instruments.

II. Organisation and implementation of the project

a. Scientific coordinator and consortium / team

The coordinator

I completed my PhD research at Institut de Chimie Moléculaire et des Matériaux d'Orsay (ICMMO). My PhD project concerned the investigation of the microscopic mechanisms at the origin of ballistic magnetic heat transport of low-dimensional spin liquids using neutron scattering and, in particular, INS to probe the fractional magnetic excitations (spinons) at the origin of the magnetic heat conduction. The studies were carried single crystals of incongruently melting materials that I grew using the TSFZ.

My first postdoc at Laboratoire Léon Brillouin concerned the study of the hidden $q=0$ LC magnetism in the PG phase of high-Tc superconducting cuprates, low dimensional spin liquids and iridates using state

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of the art PND and RXD. My second postdoc at ICMMO concerned the crystal growth of High Tc superconducting cuprates using the TSFZ method and the stabilization of new High Entropy Oxides.

I thus acquired a technical expertise in neutron and X-ray scattering along with solid-state synthesis and crystal growth.

I was hired at LLB in 2021 in the “Nouvelles Frontières dans les Matériaux Quantiques” group as a permanent researcher where I am in charge of the development of a joint LLB-SPEC (CEA-Saclay) floating-zone crystal growth platform and whom I will be the scientific responsible in the solid state chemistry group Dr. D. Colson-SPEC, CEA Saclay.

My research topics concern unconventional magnetism in high-Tc superconducting cuprates and iridates (5 publications), spin liquids (6 publications), the study of the magnetic properties of high entropy oxides (perovskites, pyrochlores) with a paper under preparation and multiferroics (2 publications).

My expertise in crystal growth lead to several collaborations, for instance, with the group pf Dr. P. Roy (Soleil Synchrotron), and the group of Pr. P. Foury (LPS, Paris Saclay University). I have further collaborated with several groups worldwide on neutron scattering experiments (for instance, Pr. M. Greven University of Minnesota, Pr. N. Dragoe at Université Paris-Saclay). My scientific experience and skills are thus in perfect agreement with the needed requirements to the success of the NEXUS project.

Experience in coaching and coordination

I supervised several trainee students on low dimensional spin liquids and the synthesis and growth of high-Tc cuprates (License 3 student: M. Roussel in 2017, Master 1 student: M. Isard in 2015, 4 Master 2 students: C. Arico in 2015, T.A. Nguyen in 2017, D. Msika in 2018 and O. Demortier in 2022). I am the apprenticeship manager of O. Haimoud (Master student 2021-2023). I participated to the supervision of the PhD student D. Msika (2019-2022).

I supervised several tutorials in the training programs on neutron scattering : Fans du LLB (2015-2018) and Hercules (2018-2023). I trained master and PhD students during experimental sessions within the framework of collaborations with other groups (F. Vayer, PhD student at ICMMO, O. Demortier PhD student at LLB, Z. Anderson PhD student at Minnesota University).

I was chair of the “Journées de la Diffusion Neutronique” (2022), “Workshop on time of flight neutron diffraction at the ESS” (2022) and I am chair of the “Gordon Research Seminar on Neutron Scattering” (2023).

The team around the young researcher

In addition to the skills of the principal investigator covering a large part of the needs of the project, I will be able to rely on various collaborations within the laboratory and outside:

Collaborations at LLB with Dr. Philippe Bourges and Dr. Y. Sidis, world-recognized experts in the field of neutron scattering and high-Tc superconducting cuprates and the first team that has reported the occurrence of **T** breaking in the PG phase of high-Tc cuprates using PND. The neutron spin-echo measurements will be carried in collaboration with Dr. N. Martin, expert in this technique and already involved in pilot experiments on the subject.

Collaboration outside the laboratory The solid-state synthesis and growth of single crystals will be carried at the SPEC laboratory in collaboration with Dr. Dorothée Colson and A. Forget, experts in solid-state synthesis and crystal growth using the self-flux method.

RXD measurements will be carried in collaboration with Dr. V. Balédent (LPS, Université Paris-Saclay), expert in X-ray scattering experiments, who was already involved in earlier experimental sessions.

I have also developed collaborations on the theoretical side with Dr. Catherine Pépin (IPHT, CEA-Saclay) and Dr. A. Meszaros (LPS). In particular, Dr. Pépin proposes a theory for the PG based on the concept of a multi-component (or vectorial) order parameter. Within the concept of intertwined orders, $q=0$ ME-LC magnetism can occur as an ancillary state, resulting from combinations of higher order(s) of the primary parameters [16]. Discussions are already ongoing around the a $q=1/2$ magnetism.

Leadership skill improvement The NEXUS project will allow giving a strong impulsion to my research activity through the recruitment of a PhD student and the constitution of my own team. This will bring

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new expertise to LLB based on my skills in crystal growth and RXD, complimentary to PND. The supervision of Master trainees is expected to lead to a thesis supervision by the end of the project. The supervising experience gained through this project will allow me passing the Habilitation à Diriger des Recherches and apply to European funding programs to carry my own scientific research on a timely project supported by a team and a solid collaboration network. The NEXUS project will allow strengthening the existing collaborations around a high potential project combining solid-state chemistry, experiments and theory and drawing an extended fruitful network of collaborations.

b. Implemented and requested resources to reach the objectives

Requested resources

The requested financial contribution for the NEXUS project amounts to **246 k€**. The table below summarizes the requested budget before each item is detailed.

Table.1. Requested means by item of expenditure

	Laboratoire Léon Brillouin (UMR 12) <i>Dalila Bounoua</i>
Staff expenses	130 000 €
Instruments and material costs	46 000€
Building and ground costs	0 €
Outsourcing / subcontracting	12 500 €
Overheads costs (including missions expenses, general and administrative costs & other operating expenses)	28 000€
Administrative management & structure costs** (13.5%)	29 500€
Sub-total	246 000 €
Requested funding	246 000 €

Staff expenses (130 000€) NEXUS relies on extensive experimental work. In particular, the crystal growth activity (WPA) from solid-state synthesis to the growth process and characterization of the single crystals will represent an important part of the time. Thus, a PhD student along with three (3) Master trainees will be hired at LLB following the timeline presented in (Tab.2), with the aim of creating a team with the needed critical mass to tackle all the aspects of this project, making the best use of the available research equipment. The staff costs decompose as follow:

Recruitment of a PhD student: in charge of the crystal growth using the floating zone furnace (70% of the time) and carrying neutron experiments on large scale-facilities (30% of time). The PhD student work will concern tasks (A.1,3 and B.1-2). The PhD student will develop complementary skills in crystal growth and scattering techniques on large-scale facilities. The ideal environment of Paris-Saclay will allow the interaction with senior scientists either in the field of crystal growth or condensed matter physics. The PhD student financial support represents a total cost of 115 k€.

Recruitment of 3 Master trainees for 6 months each: the Master trainees will allow complementing the team. Their arrival will be staggered in time (a year shift). The first Master student will be in charge of the preliminary solid-state synthesis of La doped $\text{Bi}_2\text{Sr}_2\text{CuO}_{6+\delta}$ powders and their structural and magnetic characterization (task A.1,3) in support to the PhD student. The second Master trainee will grow and characterize single crystals of La doped $\text{Bi}_2\text{Sr}_2\text{CuO}_{6+\delta}$ and participate to the neutron scattering studies (A.1,3 and B.1-2). The third Master trainee will come in support to the synthesis of the $\text{HgBa}_2\text{CuO}_{4+\delta}$ single crystals using the self-flux method and their characterization (tasks A.2-3). The Master trainees represent a cost of 15 000€.

Instruments and material costs (46 000€)

Consumables represent starting oxides, mortars and crucibles needed to perform the solid-state synthesis and small supplies (as sample holders) to carry the neutron and MPMS measurements. The budget further comprises the purchase of gas bottles for crystal growth under conditioned

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atmosphere/pressure along with replacement parts of the floating zone furnaces in case of wear (halogen lamps and quartz tubes for instance). Computers and Software Licenses for data analysis are included in the budget that represent a total of 25 k€.

Equipment The equipment cost refers to a planetary mill apparatus with its accessories (agate grinding bowls and balls) of the type on the ones supplied by Fritsch. The use of ball-milling allows guarantees the preparation of high density feed rods by reducing the powder grain size and thus the porosity to ensure a stable TSFZ growth. This is a crucial parameter for the achievement of single crystals of $\text{YBa}_2\text{Cu}_3\text{O}_{6+\delta}$ for instance. More generally, ball-milling leads to more efficient solid-state synthesis. The equipment will be used regularly for the preparation of powders and feed rods and thus must be purchased to be permanently based at the SPEC solid state laboratory.

The budget dedicated to the equipment is 21 k€.

Outsourcing / subcontracting (12 500€) dedicated to the costs of publication in open access journals.

General and administrative costs overheads & other operating expenses (28 k€) The mission expenses will cover fees of participation to:

Large scale facilities experiments: The large scale-facility experiments, on-site and travel fees, are usually supported by the neutron/synchrotron centers in Europe where up to 2 users belonging to the same team can be reimbursed. Support from the ANR within the framework of NEXUS will allow travelling to other facilities (such as ORNL) and complement the centers financial support when large teams are required, for X-ray experiment for instance. We will further ask support from national organizations as the French Neutron Federation (2FDN) to cover the fees of neutron experiments.

These expenses are estimated to 14 k€ over the four years.

(Inter)-national meetings: conferences and workshops to share our results and exchange ideas with the scientific community including schools and training for the team.

The travelling costs USA or Asia from France comprising flight, hotel, subsistence and registration fees amounts to typically 2000€/person. We will ask further support from national organizations as the 2FDN for neutron conferences. These expenses are estimated to 14 k€ over the four years.

Implemented resources (not funded by the ANR)

Instruments and material resources already available The LLB is equipped with a four mirror image furnace that will allow performing the single crystal growth by the TSFZ. The equipment was granted a joint CEA/CNRS/LabEx PALM funding as is expected to be installed and commissioned by April 2023 at SPEC in the framework of a collaboration between LLB and SPEC.

The Templin@INP "Impulse" project of which I was the coordinator allowed the acquisition of a pressure cell able to reach 2500 bars for the preparation of feed rods for the crystal growth.

The SPEC solid-state laboratory has the entire required environment to carry the solid-state synthesis (glove boxes, muffle and tubular furnaces) along with a powder X-ray diffractometer and Thermogravimetric Analysis device.

The MPMS platform of LLB allows performing magnetic, electric and specific heat measurements under different temperature, field and pressure conditions.

All of these equipment offer the perfect environment for the successful synthesis and characterization of the desired single crystals.

Human resources I plan to devote 80% of my time to the NEXUS project. I will ensure myself the crystal growth activity during Q1-Q4 (Task A.1 and 3), according to the timeline developed in **Tab.2.** prior to the arrival of the PhD student. I will also train the PhD and trainees on the different aspects of the project from the crystal growth to the large scale facility experiments and participate to the experimental sessions (Task. B1-3, C1-2).

Dr. P. Bourges and Dr. Y. Sidis (LLB-CEA Saclay) will devote 8.5 p.month of their time to neutron and RXD scattering experiments and participate to the discussion of neutron scattering data analysis.

If the project is funded, a PhD student W. Liège under the supervision of Dr. Philippe Bourges will be involved up to 18 p.months, participate to both neutron and X-ray scattering experiments as a part of his PhD project on the study of LC magnetic textures in the PG state high-Tc cuprates.

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The complimentary skills between the PhD students and master trainees will ensure a fruitful collaboration and interaction to tackle all the aspects of NEXUS.

III. Impact and benefits of the project

a. Relevance to the call and scientific impact

The origin of the mysterious PG state of high T_c superconducting cuprates remained elusive despite decades of theoretical and experimental investigations. Although leading to sizeable effects on transport and spectroscopic measurements, no obvious LT breaking that could explain the partial gapping of the Fermi surface was detected so far. However, in the recent years, several probes reported the existence of discrete symmetry **T** and **P** symmetries breaking at T^* , usually interpreted as the hallmark of a hidden IUC ME-quantum LC order and labelled as a $q=0$ magnetism.

Using PND, we recently discovered the existence of bi-axial magnetic correlations leading to a doubling of the unit cell along both the a - and the b -axis of the CuO_2 unit cell. This novel magnetism could be a game changer since, together, with the $q=0$ magnetism, it may form a hidden LC magnetic texture within the PG phase of the high- T_c cuprates that breaks the **LT** and thus could be at the origin of the depletion of the electronic density at the Fermi surface in the PG. The investigation of such textures and their relevance for the PG physics lay at the heart of the NEXUS project.

The project involves a two-pronged strategy including an important solid-state chemistry activity combined to condensed matter physics with the use of dedicated polarized neutron scattering and resonant X-ray diffraction experiments, at the forefront of refined diffraction techniques, to lift the veil on the mysterious PG state of matter.

The PND and RXD experiments are challenging owing to the small amplitude of the expected magnetic moments the measurements conditions are controlled, relying on the team expertise in probing the LC magnetism, and solid preliminary data. This project is timely and the team, being at the origin of the discovery of the $q=1/2$ magnetism, is in pole position to carry it successfully.

If the scenario of LT breaking LC patterns as the origin of the PG is true, it would thus lead to a major breakthrough in the field of unconventional superconductors calling for a novel scenario for high- T_c superconductivity.

Beyond cuprates high- T_c superconductors, there is the blooming literature around the existence of LC magnetism in a plethora of quantum materials. The LC concept gets more and more popular in the last years and the case study of cuprates offers a playground to scrutinize this exotic state of matter in a family of compounds where the state of knowledge is already well advanced to transpose it to further quantum materials where it may be involved for different macroscopic properties.

b. Communication and valorization of the results

The communication and promotion of the NEXUS project results will be done through:

- Publication in peer-reviewed and particularly open access journals.
- Sharing and diffusion of the results through oral and poster presentations in international and national (GDR) conferences and workshops in the field of superconductivity, magnetism, neutron scattering and strongly highly correlated electron systems such as: SCES, ECNS, ICNS, REXS, M^2S , ICCGE,...etc or seminars in collaborators laboratories.
- Submission of highlights to various institutions: ANR, laboratory, CEA, INP, Annual reports and highlight of Large scale facilities (synchrotrons and neutron sources).

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