Inelastic and small angle neutron scattering study of the La$_2$-xSrxCuO$_4$ high-Tc superconductor in a magnetic field

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INELASTIC AND SMALL ANGLE NEUTRON SCATTERING
STUDY OF THE LA$_{2-x}$Sr$_x$CuO$_4$ HIGH-$T_c$ SUPERCONDUCTOR
IN A MAGNETIC FIELD

A dissertation submitted to the
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Abstract

High-temperature cuprate superconductors (HTSC) have been discovered 18 years ago and immediately attracted the interest of thousands of scientists, both experimentalists and theoreticians. An incredible number of experiments performed with different techniques revealed that all HTSC materials share a common phase diagram. Starting from an antiferromagnetic insulator, these compounds become superconducting upon the introduction of charge carriers (holes or electrons) by means of chemical doping. However, a consensus about the microscopic origin of superconductivity is still missing. In particular, the role played by antiferromagnetic correlations for the mechanism of high-temperature superconductivity is still highly debated. Since the magnetic excitations are strongly modified through the critical transition temperature $T_c$, it is natural to postulate that there exists a tight link between superconductivity and magnetism in the cuprates. However, it is not yet clear whether the magnetic excitations play an active role for superconductivity or not.

Since the theoretical approaches differ in the way the antiferromagnetic state is related to the superconducting state, the study of HTSC in external magnetic fields provides a possibility to discern between them. However, the properties of HTSC in the presence of a magnetic field are modified in a non-trivial way. From the mesoscopic point of view, HTSC are type II superconductors with strong anisotropy, giving rise to a complicated magnetic phase diagram, where the external magnetic field penetrates the HTSC in the form of quantized flux-lines (magnetic vortices). These vortices might eventually form a vortex lattice, which is susceptible to disorder and thermal fluctuations. A detailed knowledge of the magnetic phase diagram is essential for the understanding of the subtle interplay between magnetic excitations and magnetic vortices.

This thesis is devoted to the investigation of the hole-doped La$_{2-x}$Sr$_x$CuO$_4$ HTSC by means of different experimental techniques. Superconducting single crystals with different doping levels have been studied in magnetic fields up to 15 Tesla. The topics of this work are:

1. The investigation of (static and dynamic) antiferromagnetic correlations in the superconducting state as a function of doping and magnetic field

2. The investigation of the magnetic phase diagram and of the dynamics of magnetic vortices

3. The investigation of the possible connections between magnetic excitations and magnetic vortices
The magnetic phase diagram has been first studied from a macroscopic point of view by means of magnetization measurements (Chapter 2), and turned out to be strongly doping dependent. In a second step, we investigated the vortex lattice by means of small angle neutron scattering and muon spin rotation experiments (Chapter 3). In the overdoped regime, we observed a field-induced change in the symmetry of the vortex lattice (from hexagonal to square) and a temperature-induced sublimation transition to a vortex gas. In the underdoped regime, we discovered a transition to a disordered vortex glass with increasing magnetic field.

The magnetic excitations have been investigated by means of inelastic neutron scattering (Chapter 4). In the overdoped regime, the application of a modest magnetic field strongly affects the copper spin dynamics. In particular, we have some evidence that the spin gap (indirectly related superconducting gap) vanishes in the vortex gas phase, where the sample is still superconducting. In the underdoped regime, we observed no spin gap in the superconducting state due to the presence of low-energy excitations. We suggest that these excitations arise from a spin glass phase that masks the presence of a strongly reduced spin gap. We also investigated the static magnetic correlations in the vicinity of the special doping concentration \( x=1/8 \), where a decrease of \( T_c \) is accompanied by an enhancement of the spin glass phase. The magnetic correlations are strongly increased by an external magnetic field, and it has been suggested that the field-induced signal arises from antiferromagnetic regions nucleated by the vortices.

Finally, we also investigated the magnetic phase diagram of the electron-doped \( \text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4 \) by means of macroscopic, muon spin relaxation and small angle neutron scattering experiments (Appendix). For the first time, a vortex lattice could be directly observed in an electron-doped HTSC. Surprisingly, the symmetry of the vortex lattice remains square down to unusually low magnetic fields. Moreover, a field-induced crossover to a more disordered vortex glass is observed. These results are discussed in relation to those obtained in \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) and other HTSC.

Many of the results presented in this thesis have been already published in a number of articles (see Curriculum Vitae). It should be noted that these publications are a result of international collaborations (see Acknowledgments).
I superconduttori ad alta temperatura critica (ossidi di rame o cuprati) sono stati scoperti 18 anni fa e hanno immediatamente attratto l’interesse di migliaia di ricercatori, sia sperimentali che teorici. Un numero impressionante di esperimenti, effettuati con differenti tecniche, hanno rivelato che tutti i cuprati possiedono un diagramma di fase comune. A partire da un isolatore antiferromagnetico, questi materiali diventano superconduttori quando dei portatori di carica vengono introdotti tramite drogaggio chimico (*doping*). Tuttavia, a tutt’oggi, i meccanismi microscopici che portano alla formazione dello stato superconduttore nei cuprati non sono ancora chiaramente individuati. In particolare, il ruolo svolto dalle correlazioni antiferromagnetiche è altamente dibattuto. Poiché le eccitazioni magnetiche sono fortemente modificate attraverso la temperatura critica $T_c$, è naturale postulare l’esistenza di uno stretto legame fra superconduttività e magnetismo. Tuttavia, non è ancora chiaro se le eccitazioni magnetiche svolgono un ruolo attivo in favore della superconduttività.

Poiché le varie teorie differiscono nel modo in cui l’antiferromagnetismo è collegato con la superconduttività, lo studio dei cuprati in presenza di campi magnetici esterni fornisce una possibilità per discernere fra loro. Tuttavia, le proprietà dei cuprati in presenza di un campo magnetico sono modificate in modo non banale. Dal punto di vista mesoscopico, i cuprati sono superconduttori di tipo II caratterizzati da una forte anisotropia, dando vita ad un diagramma di fase magnetico molto complicato. Il campo magnetico esterno penetra sotto forma di linee di campo magnetico quantizzate (chiamate anche vortici). Questi vortici possono formare una rete periodica e ordinata, che è però suscettibile a disordine e a fluttuazioni termiche. Una conoscenza dettagliata del diagramma di fase magnetico è essenziale per la comprensione dell’interazione fra le eccitazioni magnetiche ed i vortici.

Questa tesi è dedicata all’indagine del superconduttore ad alta temperatura $La_{2-x}Sr_xCuO_4$ tramite diverse tecniche sperimentali. Cristalli con differenti livelli di drogaggio (dove le cariche aggiuntive sono buchi lasciati liberi dopo che alcuni elettroni sono stati rimossi) sono stati studiati in campi magnetici fino ad un massimo di 15 Tesla. I temi di questo lavoro sono:

1. Lo studio delle correlazioni magnetiche (statiche e dinamiche) nello stato superconduttore in funzione del livello di drogaggio e del campo magnetico
2. Lo studio del diagramma di fase magnetico
3. L’indagine sui possibili collegamenti fra le eccitazioni magnetiche e la dinamica dei vortici
In primo luogo il diagramma di fase magnetico è stato studiato da un punto di vista macroscopico per mezzo di misure di magnetizzazione (Capitolo 2), risultando fortemente dipendente dal livello di drogaggio. In seguito, abbiamo studiato il reticolo di vortici per mezzo di diffrazione di neutroni a piccoli angoli (small angle neutron scattering) e di esperimenti di rotazione di spin del muone (muon spin rotation) (Capitolo 3). Nel regime sopradrogato abbiamo osservato un cambiamento nella simmetria del reticolo di vortici (da triangolare a quadrata) indotto dal crescente campo magnetico, e una transizione di sublimazione verso un gas di vortici indotta dalla crescente temperatura. Nel regime sottodrogato, abbiamo scoperto una transizione verso un reticolo disordinato di vortici con l’aumento del campo magnetico.

Le eccitazioni magnetiche sono state studiate per mezzo di diffusione anelastica dei neutroni (inelastic neutron scattering) (Capitolo 4). Nel regime sopradrogato, l’applicazione di un modesto campo magnetico influenza fortemente la dinamica degli spin degli atomi di rame. In particolare, abbiamo scoperto che il gap di energia degli spin (spin gap) scompare nella fase gassosa dei vortici. Nel regime sottodrogato, non abbiamo osservato alcuno spin gap nella fase superconduttrice, a causa della presenza di eccitazioni a bassa energia. È possibile che queste eccitazioni siano dovute a una fase in cui gli spin sono disordinati (spin glass), mascherando quindi la presenza di uno spin gap. Inoltre abbiamo studiato le correlazioni magnetiche statiche nelle vicinanze della particolare concentrazione di drogaggio $x=1/8$, dove una diminuzione della temperatura critica $T_c$ è accompagnata da un aumento delle correlazioni magnetiche. L’applicazione di un campo magnetico esterno aumenta in modo sensibile le correlazioni magnetiche, ed è stato proposto che il segnale indotto dal campo magnetico sia dovuto a regioni antiferromagnetiche create attorno ai vortici.

Inoltre abbiamo studiato il diagramma di fase magnetico del superconduttore Nd$_{2-x}$Ce$_x$CuO$_4$ tramite esperimenti di small angle neutron scattering, di rilassamento di spin del muone (muon spin relaxation) e misure di magnetizzazione (vedi Appendice). A differenza di La$_{2-x}$Sr$_x$CuO$_4$, in questo cuprato i portatori di carica sono elettroni e non buchi. Per la prima volta, un reticolo di vortici ha potuto essere osservato in un cuprato drogato con elettroni. Sorprendentemente, la simmetria del reticolo rimane quadrata anche a campi magnetici insolitamente bassi. Inoltre, a campi magnetici più elevati il reticolo di vortici diventa più disordinato. Questi risultati sono discussi in relazione a quelli ottenuti in La$_{2-x}$Sr$_x$CuO$_4$ e in altri cuprati.

Molti dei risultati presentati in questa tesi sono già stati pubblicati in numerosi articoli scientifici, frutto di collaborazioni internazionali (vedi Curriculum Vitae e Ringraziamenti).
Chapter 1

Introduction

In this chapter a broad overview of the problematic of high-$T_c$ superconductivity is given. After introducing the (generic and magnetic) phase diagrams, the $La_{1.8}Sr_xCuO_4$ compound, on which most of the measurements have been performed, will be presented. The goals and organization of the thesis, together with a short description of the employed experimental techniques, will also be discussed.

1.1 High temperature superconductors

Superconductivity (SC) has been discovered in 1911, when Onnes \(^1\) measured zero electrical resistance in mercury below a threshold temperature $T_c = 4.2 \text{ K}$ \([1]\). The second characteristic of SC, i.e. the expulsion of an external magnetic field from the superconductor, was discovered later in 1933 by Meissner and Ochsenfeld \([2]\), therefore establishing that the superconducting state forms a new thermodynamic phase. A convincing microscopic theory of SC was developed by Bardeen, Cooper and Schrieffer in 1957 \(^2\) \([3]\). In the BCS-theory, the electron-phonon interaction results in the formation of pairs of electrons (Cooper pairs) below a critical temperature $T_c$. The different pairs are strongly coupled to each others and form a SC condensate consisting of a considerable fraction of the total number of conduction electrons. Because of the coupling between all the electrons, one cannot break up a single Cooper pair without perturbing all the others. As a consequence, the amount of energy to break a Cooper pair must exceed a critical value, the so-called energy gap.

For many years the maximum $T_c$ was limited to very low temperatures ($\leq 23 \text{ K}$) until the revolutionary discovery in 1986 \(^3\) of new superconducting materials based on two-dimensional copper oxide planes \([4]\). Since the value of $T_c$ (up to 164 K for $HgBa_2Ca_2Cu_3O_9$ under pressure \([5]\), see also Fig.1.1) can be much larger than the temperature at which nitrogen liquefies, these cuprate superconductors are called high-temperature superconductors.

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\(^1\)H.K. Onnes won the Nobel Prize in Physics in 1913 "for his investigations on the properties of matter at low temperatures which led, inter alia, to the production of liquid helium".

\(^2\)J. Bardeen, L.N. Cooper and J.R. Schrieffer won the Nobel Prize in Physics in 1972 "for their jointly developed theory of superconductivity, usually called the BCS-theory".

\(^3\)J.G. Bednorz and K.A. Müller won the Nobel Prize in Physics in 1987 "for their important breakthrough in the discovery of superconductivity in ceramic materials".
superconductors (HTSC).

While conventional superconductors are *good* metals in their normal state and are well described by the Fermi liquid theory, HTSC are highly correlated electron systems and *bad* metals, with normal state properties that do not resemble at all those of a Fermi liquid. HTSC are better thought of as doped Mott insulators, rather than as strongly interacting versions of conventional metals. Mott insulators are predicted to be paramagnetic metals by band theory, due to the odd number of electrons per unit cell. However, due to the strong Coulomb repulsion that inhibits double site occupancy and charge conduction, they are insulating. The cuprates also exhibit numerous types of competing orders (e.g. antiferromagnetism (AF), stripes, ...) which can coexist with superconductivity. Apparently, none of these complications modifies the fundamental character of superconductivity and Cooper pairs are still formed as in conventional superconductor. However, many experiments have shown that the superconducting gap is anisotropic and has $d$-wave instead of $s$-wave symmetry [6]. The failure of Fermi liquid theory to describe the normal state and the presence of competing orders necessitates an entirely different approach to understand much of the physics and the mechanism of high temperature superconductivity. Almost 20 years after their discovery and after $\sim 10^5$ scientific papers on this topic, an unchallenged theory for HTSC is still missing.
1.2 Generic phase diagram

All HTSC cuprates have similar properties and structural peculiarities which differ strongly from classical superconductors. These materials are all built of a stacking of CuO$_2$ planes separated by different kinds of layers (the charge reservoirs) and it is generally believed that these copper oxide planes are essential for the occurrence of high-$T_c$ superconductivity. This is supported by the simple observation that $T_c$ increases with the number of CuO$_2$ planes per unit cell [7].

The parent (non-superconducting) compound of each family of the HTSC’s is an antiferromagnetic Mott insulator, which is transformed into a metal by introducing a concentration $x$ of doped charge carriers into the CuO$_2$ planes. Depending on the type of charge carriers, electrons or holes, one distinguishes between electron-doped (n-type) and hole-doped (p-type) HTSC. The doping is usually done by chemical substitution: for example one can induce holes by substituting La$^{3+}$ ions with Sr$^{2+}$ ions in La$_{2-x}$Sr$_x$CuO$_4$, or by increasing the oxygen content in YBa$_2$Cu$_3$O$_{6+x}$ and Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$, while electrons can be induced by substituting Nd$^{3+}$ ions with Ce$^{4+}$ ions in Nd$_{2-x}$Ce$_x$CuO$_4$.

As a function of increasing $x$, the Néel temperature $T_N$ is suppressed to zero, and long-ranged antiferromagnetism is replaced by a "strange" metal (see Fig.1.2). At low-temperatures the system becomes eventually superconducting with a transition temperature $T_c$ which first increases (underdoped region), then reaches a maximum value at an optimal doping level, decreases (overdoped region) and finally vanishes. Although the phase diagram appears similar for both p-type and n-type cuprates, it is in fact not truly symmetric. In electron-doped HTSC, the AF insulating state survives over a larger doping range, whereas the superconducting phase exists over a much narrower doping range and has lower values of the maximum $T_c$ relative to the hole-doped cuprates. Electron doping is believed to occur in the d-orbitals of copper, therefore creating spinless Cu ions that dilute the 2D antiferromagnetic spin system. On the contrary, it has been suggested that doped holes reside primarily on oxygen atoms [8]. This creates a significant spin frustration, since the exchange interaction between the hole on the oxygen and the two neighboring copper holes is the same and therefore requires the Cu spins to be parallel. As a consequence, a small quantity of holes is able to destroy long-range AF order [9, 10].

In the underdoped regime of p-type HTSC there is a crossover phenomena observed at temperature $T^* > T_c$ in which the low energy spectral weight of various quantities (magnetic susceptibility, electronic density of states, ...) are apparently suppressed. These phenomena are associated with the opening of a so-called pseudogap, which has been observed in NMR [11], tunneling [12] and angle resolved photoemission spectroscopy (ARPES) [13, 14] experiments (for a review, see [15]). The pseudogap formation is also seen in other measurements such as resistivity [16, 17], specific heat [17], and neutron crystal field spectroscopy experiments [18, 19, 20]. Since the scale of energies and the momentum dependence of the pseudogap are very reminiscent of the d-wave superconducting gap observed in the same materials at temperatures well below $T_c$ [21], it is highly attempting to identify the pseudogap with some form of local superconducting pairing. Superconductivity is the result of two distinct quantum phenomena: pairing and long range phase coherence. In conventional homogeneous superconductors, these two phenomena occur simultaneously. In HTSC it is possible that Cooper pairs are formed above $T_c$, whereas...
phase coherence (and therefore true superconductivity) is established only below $T_c$. However, the origin of the pseudogap and its relationship to high-$T_c$ superconductivity is still strongly debated. One of the unresolved issues is whether the pseudogap crossover line merges with the superconducting transition line in the overdoped regime or goes to zero under the SC dome at a quantum critical point (QCP) [22] (see Fig.1.2). In the first case, the pseudogap and superconducting phases are closely related, whereas in the second scenario they are independent and in competition. Moreover, in electron-doped HTSC, the presence of a pseudogap is still under discussion [23], therefore questioning the universality of the pseudogap phenomena.

Between the underdoped and overdoped regimes, the system behaves in the normal state as a strange metal, with a striking linear temperature dependence of the in-plane resistivity. This led Varma and co-workers to propose a marginal Fermi liquid phenomenology to explain many of the anomalous behaviors of cuprates [24, 25]. In the overdoped region, on the other hand, the system is well described within the Fermi liquid theory, as indicated by the observation of a Fermi-liquid like $T^2$ dependence of the resistivity [26].

The link between antiferromagnetism and superconductivity remains one of the hottest issues in the field of high-$T_c$ superconductivity. While $\mu$SR experiments on La$_{2-x}$Sr$_x$CuO$_4$ revealed that a spin-glass phase extends into the SC state [27, 28], inelastic neutron scattering experiments have shown that spin excitations coexist with superconductivity and survive even in the overdoped regime of La$_{2-x}$Sr$_x$CuO$_4$ [29, 30]. These experimental works motivated the theoretical study on the significance of antiferromagnetic correlations and spin fluctuations for the mechanisms of high-temperature superconduc-


tivity. A spin-fluctuation-induced pairing mechanism, which naturally explains the \( d \)-wave nature of the order parameter \[31\], and a unified theory of superconductivity and antiferromagnetism based on SO(5) symmetry \[32\] have been proposed. Alternative scenarios consider the possibility that the charge carriers are inhomogeneously distributed in the CuO2 planes, involving periodic spatial modulations of charge and spin. The \textit{static} version of this scenario would imply the segregation of the charge into \textit{stripes} that act as domain walls between antiferromagnetic domains \[33\].

In order to test the different theoretical scenarios, several experimental investigations have been recently undertaken in the presence of an external magnetic field \( H \). The interpretation of the experimental results is complicated by the fact that the magnetic field distribution inside HTSC is not homogeneous, and requires the understanding of the magnetic phase diagram.

\section*{1.3 Magnetic phase diagram}

The application of a magnetic field strongly affects the properties of superconductors. The magnetic (\( H \) vs \( T \)) phase diagram has been extensively investigated, both from the theoretical and experimental point of views. In this section we want to address the most important general aspects, which are relevant to the present thesis. For further reading one can find in the literature a number of review articles (see e.g. Ref.\[34, 35, 36, 37\]).

The superconducting response to an external magnetic field is determined in the phenomenological Ginzburg-Landau (GL) theory by the GL parameter \( \kappa = \frac{\lambda}{\xi} \). \( \lambda \) and \( \xi \) are the two relevant lengthscales for superconductivity: the penetration depth \( \lambda \) is the typical scale over which the electromagnetic potential varies, whereas the coherence length \( \xi \) represents the correlation length of the Cooper pairs and gives the scale for variations of the order parameter.

If \( \kappa < 1/\sqrt{2} \) (Type-I) the magnetic phase diagram contains only two phases: for \( H < H_c(T) \) and \( T < T_c \) the external magnetic flux is completely expelled from the sample, which behaves like a perfect diamagnet (\textit{Meissner state}). Cooper pairs of electrons are formed, resulting in zero electric resistivity. Above \( H_c(T) \) the normal state is recovered, the diamagnetic properties are lost and finite resistivity appears (see Fig.1.3a).

If \( \kappa > 1/\sqrt{2} \) (Type-II) an additional phase is present between the Meissner phase (below the lower critical field \( H_{c1}(T) \)) and the normal phase (above the upper critical field \( H_{c2}(T) \)) (see Fig.1.3b). In this mixed phase the magnetic field can penetrate the sample in form of flux-lines (vortices) each carrying a flux quantum \( \Phi_0 = \frac{hc}{2e} \). These vortices consist of a normal conducting core region of radius \( \xi \) surrounded by superconducting currents \( j_s \) which create a magnetic field distribution extending over distances comparable to \( \lambda \) (see Fig.1.4). Due to repulsive interactions, the vortices were predicted to arrange themselves in a lattice \footnote{Alexei A. Abrikosov won, together with V.L. Ginzburg and A.J. Leggett, the Nobel Prize in Physics in 2003 "for pioneering contributions to the theory of superconductors and superfluids"}, and indeed long-range ordered vortex lattices have been directly observed experimentally in a number of systems. Even though the lowest energy configuration is \textit{hexagonal}, in many compounds \textit{square} vortex lattices have been observed.

The distinction between Type-I and Type-II superconductors is due to the surface
Figure 1.3: Schematic view of the magnetic ($H$ vs $T$) phase diagram in a) Type-I and b) Type-II superconductors. In c)-e) the Meissner, mixed and normal states are shown schematically.

Figure 1.4: Schematic view of the superfluid density $n_s(r)$, the internal field distribution $B(r)$ and the supercurrent density $j_s(r)$ through a cross-section of a single vortex.
Figure 1.5: Schematic behavior of a vortex-line in the cage potential due to neighboring vortices perturbed by point disorder. a), b) At low fields below the vortex glass transition the pinning energy $E_{\text{pin}}$ due to disorder is much smaller than the elastic energy $E_{\text{el}}$. c), d) With increasing field $E_{\text{pin}}$ becomes relatively more important resulting in a rougher potential landscape, therefore enhancing the wandering of the vortices and giving rise to the formation of dislocations (from Ref.[38]).

The mixed state provides an excellent system for both fundamental research and applications. From the fundamental point of view, the vortex lattice (VL) provides a unique system to study a crystal, in which one can change the density (the lattice spacing) by simply varying the magnetic field. Moreover, since the "magnetic crystal" (vortex lattice) is grown on a real atomic crystal with a much smaller lattice constant, it can be submitted to various perturbations such as disorder. This provides the unique opportunity to investigate the combined effects of disorder and thermal fluctuations on a crystal. From a more practical point of view, one would like to have a superconductor which can operate at high temperatures and high magnetic fields, and capable to transport large currents [39]. However, if an external current is applied perpendicular to the VL, the magnetic vortices will feel a Lorentz force and start to move, inducing an electric field parallel to the direction of the transport current which therefore experiences a resistance (flux-flow resistivity). In order to have bulk resistance-free transport current, it is crucial to decrease the displacement of the vortices. This can be achieved by pinning them to inhomogeneities.
Figure 1.6: Schematic view of the magnetic phase diagram in HTSC. Due to disorder/pinning, the vortex lattice is transformed into a Bragg glass at low fields and a vortex glass at high fields. Because of thermal fluctuations the Bragg glass undergoes a melting transition to a vortex liquid at high temperatures. (from Ref.[35]).

of the underlying crystal structure. Crystalline defects or impurities locally reduce the order parameter. Vortices are therefore attracted to them, because of the smaller loss of condensation energy in the vortex core.

Within an elastic description of the vortices, the VL can be considered as a system of elastic strings coupled together by elastic forces. The VL can also be pinned if individual vortices bend elastically to be accommodated by pinning sites (see Fig.1.5). The effect of pinning is described by the Larkin-Ovchinnikov (LO) theory of collective pinning [40]. In the presence of a random array of weak pins, long range order of the vortex lattice is always destroyed over large distances but preserved within some smaller correlation volume. Due to the interaction between the single vortices, the vortex lattice is deformed collectively in the same unit volume, whose size depends on the balance between the energy gain of the pinning sites and the increase in elastic deformation. The concept of collective pinning may be applied for weak pins, where the local displacements of the vortex lines is small. In this case, the vortex lines are pinned by an ensemble of weak pinning sites rather than individual strong pins.

A direct consequence of pinning is that the long-range translational order of the VL is destroyed [41, 40] giving rise to a sort of glassy state. As long as the elastic properties of the VL are preserved (absence of dislocations, see Fig.1.5) the VL remains quasi-ordered.
and Bragg peaks are still observed in neutron scattering experiments. Such a Bragg glass is a glass nearly as ordered as a perfect crystal, with quasi long range order (algebraically divergent Bragg peaks) and perfect topological order (absence of defects such as dislocations) [37, 42, 43]. Recently, the power-law decay of the crystalline order characteristic of a Bragg glass has been observed in (K, Ba)BiO₃ by neutron scattering [44]. By increasing the effect of disorder, the Bragg glass will undergo a transition to a more disordered vortex glass phase (or entangled solid phase). Such a phase is expected at high magnetic fields and low temperatures, when pinning of vortices by point defects competes with elastic vortex interactions, and the Bragg glass becomes unstable to the formation of the vortex glass \(^5\) [43, 38, 35, 45]. This is schematically shown in Fig.1.6.

Taking into account the effect of thermal fluctuations (important especially for HTSC), the Bragg glass is expected to undergo a first-order melting transition into a vortex liquid [46, 47, 48]. The melting of the VL can be extracted from the elastic description. A detailed theory is still lacking, but one can use the simple Lindemann criterion [49] that states that a crystal melts when the thermally induced displacements \(u\) in the crystal become a sizeable fraction of the lattice spacing \(a\):

\[
\langle u^2 \rangle = c_L^2 a^2
\]

where \(c_L\) is determined empirically to be about 0.1-0.2.

The two-dimensional character of the superconducting CuO₂ planes and the associated \(c\)-axis anisotropy further complicate the situation. For highly anisotropic HTSC (such as Bi₂Sr₂CaCu₂O₈₊ₓ), it is expected that the vortex line structure breaks into a stack of two dimensional pancake vortices connected to each other by electromagnetic (Josephson)

\(^5\)For vortices, increasing the field is like increasing disorder. In fact the effective disorder is proportional to the average density of vortices, and at large enough magnetic fields the disorder term becomes stronger than the elastic term.
coupling (see Fig. 1.7). A dimensional crossover from 3D (vortex lines) to 2D (pancakes) vortex systems is expected around \( H_{2D} = \Phi_0/(\gamma s)^2 \), where \( s \) is the interlayer spacing and \( \gamma \) is the out-of-plane anisotropy [50]. Such a transition has been indeed observed in a remarkable small angle neutron scattering experiment [51]. The combination of low-dimensionality and thermal fluctuations led to the proposal of a sublimation theory [52, 53]. Within this scenario, the melting of the VL is concomitant to the formation of pancake vortices decoupled between layers (see Fig. 1.7).

Due to the high complexity of the magnetic phase diagram of HTSC, numerous aspects are still controversial. It is therefore important to collect more experimental data. In particular it is crucial to compare results obtained by different techniques to theoretical predictions.

1.4 The \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) compound

1.4.1 Why \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \)?

Most of the measurements presented in this thesis have been performed on superconducting \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) (LSCO) single crystals. This hole-doped compound has a maximum \( T_c \) of less than 40 K. However, it has the advantage of having a simpler structure than other higher-\( T_c \) compounds. For example \( \text{YBa}_2\text{Cu}_3\text{O}_{6+x} \) (YBCO, \( T_c \sim 90 \) K) has double \( \text{CuO}_2 \) layers and \( \text{CuO} \) chains which often complicate the interpretation of experimental data. LSCO is also interesting because large high-quality crystals can be grown over a wide doping range \(^6\) [10] covering all the phase diagram shown in Fig. 1.2. On the contrary, \( \text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x} \) (BSCCO) crystals can be produced only in small quantities and in a limited doping range. Moreover, in LSCO, the out-of-plane anisotropy \( \gamma \) depends on the Sr content \( x \) and allows the study of HTSC over a wide range of anisotropy which lies inbetween the values for \( \text{YBa}_2\text{Cu}_3\text{O}_{6+x} \) and \( \text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x} \) [53]. Because of these intermediate values of \( \gamma \), the LSCO compound represents also a system with different and unique mesoscopic properties of the vortex lattice. For example, the large anisotropy of BSCCO results in essentially weakly coupled strings of 2D pancake vortices (see Fig. 1.7b). Due to this weak coupling, thermal or pinning induced disorder effectively leads to states characterized by large fluctuations along the field direction. On the other side, in YBCO, the pancake vortices are extremely strongly coupled by the Josephson currents flowing between the conduction planes, forming rigid vortex lines (see Fig. 1.7a). YBCO is much less susceptible than BSCCO to disorder, so the melting line occurs very close to \( T_c \) and the disorder crossover occurs at fields above those currently accessible in neutron scattering and \( \mu \)SR experiments (in excess of 10 Tesla). In contrast, the materials parameters of LSCO, and in particular in the underdoped regime, give rise to a system of fairly rigid vortex lines which are nonetheless highly susceptible to transverse fluctuations. This system is therefore a good candidate for investigating experimentally the transition from a Bragg glass to a vortex glass, due to the changing scale of disorder as the separation of the vortices decreases in an increasing field.

\(^6\)The experimental results presented in this thesis have been obtained on high-quality LSCO single crystals grown by the traveling solvent floating zone (TSFZ) technique by the group of Prof. Oda, Hokkaido University, Japan (see e.g. [54]).
1.4 The $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ compound

Figure 1.8: Structure of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$: the arrows indicate the AF ordered Cu spins in the undoped compound ($x=0$). A CuO$_6$ octahedra is also shown. In the smaller panel, a CuO$_2$ plane is shown: the unit cell in the orthorhombic notation (red square) is $\sqrt{2}$ larger and $45^\circ$ rotated compared to the unit cell in the tetragonal notation (blue square).

1.4.2 Structure and phase diagram of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$

The structure of LSCO consists in a stacking of CuO$_2$ planes separated by La(Sr)O-blocks, as shown in Fig. 1.8. LSCO is tetragonal or orthorhombic depending on doping and temperature. The room-temperature lattice constants of undoped $\text{La}_2\text{CuO}_4$ are $a \approx 5.36$ Å, $b \approx 5.40$ Å, and $c \approx 13.16$ Å [55].

The second-order phase transition from the high-temperature tetragonal (HTT) to the low-temperature orthorhombic (LTO) structure is caused by the tilting of the CuO$_6$ octahedra, which is related to the softening of the respective phonon-modes [56]. The transition temperature $T_0$ decreases with increasing doping and vanishes at $x \approx 0.22$ (see Fig. 1.9). This structural phase transition can be easily observed in neutron scattering experiments [55, 56, 57, 58, 59], by measuring the additional reflections present in the LTO phase. As an example we show in Fig. 1.10 elastic neutron scattering measurements on LSCO ($T_c=31$ K, $x=0.20$) of the (104) orthorhombic reflection, which appears below $T_0 \approx 70$ K. It is interesting to notice that below $T_c$ the continuous increase of the orthorhombicity and the related tilt of the CuO$_6$ octahedra are stopped. This result is consistent with previous measurements [60, 58] and might indicate that superconductivity is tightly
related to lattice deformations. The symmetry reduction at the phase transition leads to the formation of twin domains corresponding to different tilt axes for the CuO$_6$ octahedra [57]. In the LTO phase one has therefore to deal with twin boundaries, which represent planar defects in the crystal. These defects can be observed in SANS experiments and are more pronounced in the underdoped regime, as shown in Fig. 1.11.

As for the other HTSC, the phase diagram of LSCO is characterized by an antiferromagnetic insulating phase at low doping. The electron configuration of copper in the undoped compound is 3d$^9$, the one of oxygen is 2p$^6$. O$^{2-}$ is therefore non-magnetic, and there is a hole with spin 1/2 in the d-shell of Cu$^{2+}$. By substituting divalent Sr$^{2+}$ for trivalent La$^{3+}$ charge carriers are induced and, to maintain charge balance, electrons are removed from the CuO$_2$ planes. At large enough concentration of holes the superconducting phase appears (0.05 < $x$ < 0.27), see Fig. 1.9.

Above $T_c$ a pseudogap has been observed in the underdoped regime [17]. However most of the results have been obtained by indirect methods such as resistivity, susceptibility, spe-
1.4 The La$_{2-x}$Sr$_x$CuO$_4$ compound

Figure 1.10: a) Raw data of the (104) reflection of LSCO ($x=0.20$) at different temperatures, fitted by Gaussians. b) Temperature dependence of the integrated intensity. The gradual increase of the (104) reflection in the LTO phase below $T_0 \approx 70$ K is due to the continuous tilt of the CuO$_6$ octahedra, which is stopped below $T_c$.

specific heat and neutron crystal field spectroscopy [18, 19, 20] measurements. Reliable and detailed tunneling and photoemission investigations of the pseudogap are still missing, mainly because of the difficulty to obtain good cleavage planes in the LSCO compound. At low doping ($0.02 < x < 0.05$), between the insulating and the superconducting phase, LSCO is characterized by a frozen magnetic state (referred in the literature as the spin glass phase [63]) which has been observed in $\mu$SR, NMR and NQR experiments [28] as well as in magnetic susceptibility measurements [64, 65]. This magnetic phase survives in the underdoped (superconducting) region, as shown in the inset of Fig.1.9. Interestingly, the freezing temperature $T_{sg}$ is enhanced around $x=1/8$ and coincides with a slight suppression of $T_c$. This phenomena is referred in the literature as the "1/8 anomaly". To notice is that in LSCO the exact doping level of the anomaly is $x=0.115$, therefore slightly lower than $x=1/8=0.125$ (see ref. [28] and references therein).

1.4.3 Annealing procedure

The single crystals used in the experiments presented in this thesis were all grown by the TSFZ method, which has the advantage that no crucible is needed, therefore avoiding contamination from crucibles. The demerit of the TSFZ technique is that the crystal can contain defects and oxygen (and therefore doping) inhomogeneity. Post-annealing of the crystal in an oxygen atmosphere at 850 °C for several days is therefore needed in order to relax the distortion of the crystal and to get a more homogeneous oxygen distribution inside the sample. In order to have a controlled oxygen distribution inside the sample and to protect the crystal from the humidity, which degrades the sample strongly and steadily, we always annealed our samples prior to experiments.
Figure 1.11: SANS diffraction pattern at $T=40$ K in underdoped and overdoped LSCO (logarithmic color scale). A large signal arising from the twin boundaries is clearly seen in the underdoped sample, while in the overdoped sample the scattering from crystal defects is more isotropic. The direct beam is masked by a beam stop in the middle of the 2D detector.

1.5 Goals and organization of the thesis

This work is devoted to the investigation of the magnetic and superconducting properties of LSCO single crystals at different doping levels. Several complementary experimental techniques have been employed (see next section).

In Chapter 2, the doping dependence of the magnetic phase diagram is investigated from a macroscopic point of view.

Chapter 3 reports on a mesoscopic study of the mixed phase in both underdoped and overdoped LSCO by means of small angle neutron scattering. Some complementary muon spin rotation measurements will also be presented.

Chapter 4 presents a microscopic investigation of the spin excitations as a function of temperature and magnetic field. The differences between the overdoped and underdoped regime will be discussed in relation to muon spin relaxation results and numerical calculations of the spin susceptibility.

In Chapter 5 a conclusion of the work is given. In particular I will try to give an unified picture of the phase diagram of LSCO on the basis of results obtained in Chapters 2-4.

Finally, in the Appendix, macroscopic, muon spin relaxation and small angle neutron scattering measurements in electron-doped $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$ (NCCO) will be presented and discussed in relation to the results obtained in LSCO.

The Chapters 2-4 and the Appendix have been conceived so that they are stand-alone

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7Muon spin rotation experiments have been performed at PSI in collaboration with the group of S.L. Lee (University of St. Andrews, UK) on the same LSCO crystals used for macroscopic and neutron scattering measurements. These supplementary results have been included in this thesis where they are important.

8Muon spin relaxation experiments have been performed by Ch. Niedermayer (Paul Scherrer Institute, Switzerland) on the same LSCO crystals used for macroscopic and neutron scattering measurements. These supplementary results have been included in this thesis because they are relevant to our neutron scattering results.
and can be read independently from each others. At the beginning of each chapter, a short abstract summarizes the content. After an introduction to the respective experimental techniques, the experimental results are described in detail. The data are discussed on the basis of proposed theoretical models. Special emphasis is given to the comparison of results obtained by the different experimental techniques.

The scheme of the thesis is summarized in Fig.1.12.

1.6 Experimental techniques

In this section a short description of the experimental techniques used for the study of LSCO and NCCO is presented. A more detailed discussion of the different experimental tools is available at the beginning of Chapters 2-4.

- **Physical Properties Measurement System (PPMS)**
  This system is designed to perform a variety of automated measurements of physical properties such as DC magnetization ($M$), AC susceptibility ($\chi$), heat capacity ($C$) and resistivity ($\rho$). Sample environment controls include magnetic fields up to 9 Tesla and a temperature range of 1.8 - 400 K. For more information, see http://www.qdusa.com/products/ppms.html

- **Small Angle Neutron Scattering (SANS)**
  SANS is a technique that measures the deviation to small angles (from much less than one degree to several degrees) of a neutron beam scattered by structures of mesoscopic size (between 10 Å and about 4000 Å). The data presented in this work have been obtained on different SANS instruments (SANS-I and SANS-II at the Paul Scherrer Institute (PSI), Switzerland; D22 at the Institute Laue Langevin (ILL), France). On SANS-I a horizontal magnetic field up to 11 Tesla can be applied, while on D22 the maximal field available is limited to 1 Tesla. For more information about the instruments, see http://sans.web.psi.ch/ (SANS-I), http://sinq.web.psi.ch/sinq/instr/sans2.html (SANS-II) and http://www.ill.fr/YellowBook/D22/ (D22).

- **Inelastic Neutron Scattering (INS)**
  Inelastic neutron scattering is the process of scattering neutrons from a sample, accompanied by a change in energy of the neutron. The neutron can gain (or lose) energy because of the annihilation (or creation) of an elementary excitation, such as a lattice vibration (phonon) or a magnetic excitation (magnon). INS experiments have been performed both at PSI (Rita-II), ILL (IN22 and IN14) and at the Hahn-Meitner-Institute (V2/FLEX) in magnetic fields up to 15 Tesla. For more information about the instruments, see http://rita2.psi.ch/ (Rita-II), http://www.ill.fr/YellowBook/IN22/ (IN22), http://www.ill.fr/YellowBook/IN14/ (IN14) and http://www.hmi.de/bense/instrumentation/instrumente/v2/v2.html (V2/FLEX).

- **Muon Spin Rotation/Relaxation ($\mu$SR)**
  These techniques make use of a short-lived subatomic particle (the muon), whose
Introduction

Figure 1.12: Schematic view of the thesis: the doping dependent superconducting and magnetic properties of hole-doped LSCO have been investigated both from a macroscopic, mesoscopic and microscopic point of views. The relationship between results obtained by different techniques will be a central issue of this work. In the Appendix, experimental results in electron-doped NCCO will also be presented and compared to those obtained for LSCO.
spin and charge are sensitive local magnetic and electronic probes of matter. One can distinguish between Muon Spin Rotation and Relaxation. 

Muon Spin Rotation involves the application of an external magnetic field perpendicular to the initial direction of the muon spin polarization. The muon spin precesses about the transverse field, with a frequency that is proportional to the size of the field at the muon site in the material. This configuration can be used to measure the magnetic field distribution of the vortex lattice in a type-II superconductor.

In Muon Spin Relaxation experiments one measures the time evolution of the muon polarization along its original direction in the absence of an external field (or an external magnetic field parallel to the initial direction of the muon spin polarization). This is a very sensitive method of detecting weak internal magnetism, that arises due to ordered magnetic moments, or random fields that are static or fluctuating with time.

More information about neutron scattering and muon spin rotation/relaxation can be found in the literature, see e.g. Ref.[66, 67], and in the web:
http://sinq.web.psi.ch/
http://www.ill.fr/
http://lmu.web.psi.ch/about/aboutmsr.html
http://musr.triumf.ca/intro/musr/
http://www.isis.rl.ac.uk/muons/
http://www.neutron-eu.net/
...
Chapter 2

Macroscopic measurements

DC magnetization and AC susceptibility measurements are used to characterize the mixed phase of the high-temperature cuprate superconductor La$_{2-x}$Sr$_x$CuO$_4$ over a large doping range (0.075<x<0.20) up to high magnetic fields (8 Tesla applied perpendicular to the CuO$_2$ planes). We observe a strong doping dependence of the magnetic phase diagram, which can mainly be explained by the increasing anisotropy with underdoping. These results are used to interpret SANS (Chapter 3) and INS (Chapter 4) measurements.

2.1 Introduction

Macroscopic measurements (e.g. DC magnetization, AC susceptibility, specific heat, resistivity, ...) have been extensively used to characterize the properties of high-T$_c$ superconductors (HTSC). These techniques are extremely useful for the study of the basic properties of superconductors, namely zero-resistance and diamagnetism below T$_c$, and to check the quality of the produced samples (sharp superconducting transition temperature, large superconducting volume fraction, ... ). Moreover one can extract some key parameters, such as the coherence length $\xi$, the penetration depth $\lambda$ and the anisotropy $\gamma = \sqrt{m_c^2/m_{ab}^2}$ [68, 69, 70, 52, 53]. For example, $\gamma^2$ can be defined as the ratio between the out-of-plane and the in-plane resistive components ($\rho_c/\rho_{ab}$) measured in the normal state [52, 53]. Finally, in HTSC, macroscopic measurements are helpful for the investigation of the extremely rich magnetic phase diagram up to high magnetic fields ($\sim$ 60 T)[71].

Despite belonging to the family of the first HTSC to be discovered, the magnetic phase diagram of La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) has not been so intensively investigated with respect to other cuprates such as YBa$_2$Cu$_3$O$_{6+x}$ (YBCO) and Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ (BSCCO). The LSCO compound is of high interest because it fills the gap between 3D vortex systems such as YBCO and highly anisotropic 2D systems such as BSCCO. The advantage of LSCO is that the anisotropy factor $\gamma$ depends on the Sr content $x$ and thus allowing a study of the magnetic phase diagram over a wide range of anisotropy ($200 < \gamma^2 < 4000$, see Fig.2.1a) which lies inbetween the values for YBCO ($25 < \gamma^2 < 100$) and BSCCO ($3000 < \gamma^2 < 30000$) [53].

In this chapter, we present a detailed study of the magnetic phase diagram of LSCO over a large doping range (0.075<x<0.20) by means of DC magnetization and AC susceptibility measurements. Four high quality single crystals with different doping have been
Figure 2.1: a) Doping dependence of the anisotropy in LSCO, as determined from resistivity measurements by Sasagawa et al. (ref. [53]) and Kimura et al. (ref. [72]). The line is a guide to the eye. b) Schematic view of the real part of the AC susceptibility in zero field, which is used to determine the SC transition temperature $T_c$, defined by $\chi'(T_c) = \frac{1}{2} \chi'(0 \text{ K})$. The width of the transition $\Delta T_c$ is defined by the 10%-90% criterion.

There are several goals that motivate our macroscopic measurements. First of all we want to obtain a first overview of the magnetic phase diagram of LSCO and its doping dependence, which is not yet completely understood. The knowledge of the magnetic phase diagram is important and crucial for the understanding of SANS (Chapter 3) and INS (Chapter 4) results, since these experiments have also been performed in a magnetic field. Finally, the measurements presented in this chapter are also useful to check the quality of the single crystals.
Table 2.1: List of the sample used for PPMS measurements, with the corresponding doping, mass, the SC transition temperature $T_c$ and its width $\Delta T_c$.

<table>
<thead>
<tr>
<th>Name</th>
<th>Compound</th>
<th>doping $x$</th>
<th>mass ± c</th>
<th>$T_c$</th>
<th>$\Delta T_c$</th>
</tr>
</thead>
<tbody>
<tr>
<td>UD-19K</td>
<td>LSCO</td>
<td>0.075</td>
<td>52 mg</td>
<td>19 K</td>
<td>3.8 K</td>
</tr>
<tr>
<td>UD-29K</td>
<td>LSCO</td>
<td>0.10</td>
<td>37 mg</td>
<td>29.2 K</td>
<td>1.3 K</td>
</tr>
<tr>
<td>OD-36K</td>
<td>LSCO</td>
<td>0.17</td>
<td>293 mg / 84 mg</td>
<td>36.2 K</td>
<td>1.5 K</td>
</tr>
<tr>
<td>OD-31K</td>
<td>LSCO</td>
<td>0.20</td>
<td>51 mg</td>
<td>31.5 K</td>
<td>2.5 K</td>
</tr>
</tbody>
</table>

### 2.2 Experimental setup

DC magnetization and AC susceptibility measurements have been performed using a commercial Quantum Design Physical Properties Measurements System (PPMS) up to magnetic fields of 8 Tesla applied approximately parallel to the c-axis. The angle $\Theta$ between the field direction and the c-axis (determined by x-rays Laue diffraction) was always smaller than 10 degrees. This precision is good enough for the present study, since the critical lines (e.g. melting line $H_m$, upper critical field $H_{c2}$) are known to be only slightly affected by small angles [73], see also Section 2.6.

DC magnetic measurements determine the equilibrium value of the magnetization in the sample. The sample is magnetized by a constant magnetic field and its magnetic moment is measured by induction. Moving the sample through the detection coils induces a voltage and the amplitude of the signal is proportional to the magnetic moment and speed of the sample during extraction.

In AC magnetic measurements, a small AC magnetic field is superimposed on the DC field ($H = H_{DC} + H_{AC} \cdot \cos(\omega_{AC} t)$), causing a time dependent moment in the sample. The field of the time-dependent moment induces a current in the pickup coils, allowing a measurement without sample motion. As long as the frequency $\omega_{AC}$ is small, the measurement is similar to DC magnetic measurements, and the induced AC moment is given by $M_{AC} = dM/dH \cdot H_{AC} \cdot \cos(\omega_{AC} t)$, where $\chi = dM/dH$ is called the susceptibility. At higher frequencies, the magnetization of the sample may lag behind the drive field due to dynamic effects in the sample. Thus two quantities are now measured: the magnitude of the susceptibility and the phase shift between the drive signal and the measured signal. Alternatively one can imagine the susceptibility as having a real (in-phase) and an imaginary (out-of-phase) component: $\chi = \chi' + i\chi''$. The complex AC-susceptibility components are defined as [74]:

$$\chi' = \frac{1}{\pi H_{AC}} \int_0^{2\pi} M(\omega t)\cos(\omega t)d(\omega t)$$  \hspace{1cm} (2.1)

$$\chi'' = \frac{1}{\pi H_{AC}} \int_0^{2\pi} M(\omega t)\sin(\omega t)d(\omega t)$$  \hspace{1cm} (2.2)

where $M(t)$ is the sample magnetization. The physical meanings of $\chi'$ and $\chi''$ are the following: $\chi'$ is proportional to the time average of the magnetic energy stored in the volume occupied by the sample, whereas $\chi''$ is proportional to the energy converted into heat during one cycle of the ac-field (dissipation). The analysis of the susceptibility data requires some care, due to the influence of numerous parameters ($H_{AC}$, $\omega_{AC}$, \ldots). At
high frequency the time available for the flux to penetrate the sample is shortened and the experimental results can therefore be influenced by the time constant involved in the measuring technique. Using low frequency should therefore give better results, since the AC field can see the bulk of the sample. In the present work we used \( H_{AC} = 10 \) Oe and \( \omega_{AC} = 10 \) Hz.

### 2.3 AC susceptibility and DC magnetization measurements

We performed both isothermal and non-isothermal measurements. For the isothermal procedure, the sample is first zero-field cooled (ZFC) starting from temperatures well above \( T_c \) and then stabilized at a desired temperature. This guarantees an almost completely magnetization free initial state of the sample and defines the starting point of the experiment, which consists in a field-scan \( M(H) \) at fixed temperature. In non-isothermal experiments, on the other hand, one measures \( M(T) \) and \( \chi(T) \) as a function of temperature (temperature-scan) while the magnetic field is fixed to the desired value. Such measurements have been performed both in ZFC and field-cooled (FC) mode.

#### 2.3.1 Temperature scans

We start with the complex AC susceptibility \( \chi = \chi' + i\chi'' \). A set of FC temperature scans \( \chi(T) \) for the four LSCO samples at different magnetic fields is shown in Fig. 2.2. The real part and imaginary part of \( \chi(T) \) are directly related: the maximum slope of \( \chi'(T) \) coincides with the peak maximum in \( \chi''(T) \). The sharpness of the transition can be defined by the 10%-90% criterion for \( \chi'(T) \) (see Fig. 2.1) or by the width of the peak in \( \chi''(T) \), and is an indication for the quality of the samples. The transition width \( \Delta T_c \) is very small for OD-36K and UD-29K, whereas is larger in the overdoped (OD-31K) and highly underdoped (UD-19K) samples (see Table 2.1). The magnitude of \( \chi'(0 \text{ K}) \) is found to change from sample to sample, most probably because of the different shape of the crystals (and therefore different demagnetization factors).

In all samples the peak in \( \chi''(T) \) shifts toward low temperatures and sharpens with increasing magnetic field. However this shift is strongly doping dependent: for UD-19K a magnetic field of 6 T is sufficient to shift the peak by \( 0.85 \cdot T_c \), whereas for OD-31K the shift caused by a field of 8 T is only \( 0.45 \cdot T_c \). The detailed field dependence will be discussed later on in this chapter.

In earlier works the peak in \( \chi''(T) \) was interpreted as a signature of the upper critical temperature \( T_{c2} \). This was in disagreement with DC magnetization measurements, and the peak was then attributed to the irreversibility line (IL) [74, 75], which corresponds to the crossover from a phase of pinned vortices (irreversible magnetization, finite critical current) to a phase of unpinned vortices (reversible magnetization, zero critical current). The idea is that the loss peak in the \( \chi''(T) \) occurs when the vortex lines are thermally activated and can exit the pinning centers. Above the irreversibility temperature \( T_{irr} \) reversible magnetization appears and the critical current goes to zero. In resistivity measurements, the irreversibility temperature is defined as the onset of resistance, whereas
2.3 AC susceptibility and DC magnetization measurements

Figure 2.2: Real and imaginary part of the AC susceptibility $\chi(T)$ for UD-19K, UD-29K, OD-36K, and OD-31K measured at different magnetic fields between 0 T and 8 T after field cooling.
Figure 2.3: UD-29K: a) FC and b) ZFC $M(T)$ curves at different magnetic fields. The arrows in b) indicate the values of $T_{FOT}$ as determined by the peak position in $\chi''(T)$, whereas in the inset the same data are shown with an enlarged scale. c) Comparison of $M(T)$ (after subtraction of a linear background taken in the normal state) with $\chi(T)$ at $H_{dc}=3$ T. $T_{FOT}$ is determined by the peak in $\chi''(T)$ which corresponds to the maximum slope in $\chi'(T)$ and to the dip in $M(T)$. Below $T_{irr}$ the FC and ZFC $M(T)$ curves separate. $T_{c2}$ is estimated by extrapolation (see text), whereas $T_{fuct}$ is defined as the point where $M(T)$ deviates from the horizontal normal state line.
2.3 AC susceptibility and DC magnetization measurements

the point where the resistance begins to drop is identified with $T_{c2}$ [53, 76]. Between $T_{irr}$ and $T_{c2}$ the resistivity is due to the motion of vortices (flux flow resistance).

DC magnetization measurements $M(T)$ provide additional information about the vortex behavior. In Fig.2.3a+b and Fig.2.4 ZFC and FC temperature scans at different magnetic fields are shown for the four samples, whereas in Fig.2.3c a representative curve $\chi(T)$ measured at $H_{dc}=3$ T for UD-29K is plotted together with magnetization data $M(T)$. While ZFC and the FC $\chi(T)$ data do not show any difference, FC and ZFC $M(T)$ curves separate below the irreversibility temperature $T_{irr}$ (this is actually the real definition of irreversibility line). To notice is the much larger diamagnetic signal of the ZFC measurements at low temperatures compared to the FC measurements (see inset of Fig.2.3b). Slightly above $T_{irr}$ there is an anomalous dip in the magnetization (see Fig.2.3c), possibly indicating the presence of a first order transition (FOT). Such a FOT line close to the irreversibility line has been previously reported in LSCO [52, 53]. In our samples, $T_{irr}$ obtained by AC susceptibility measurements is slightly larger than the "real" $T_{irr}$, and is concomitant to the dip in $M(T)$ at $T_{FOT}$. We therefore believe that the peak position of $\chi''(T)$ is a signature of $T_{FOT}$ rather than of $T_{irr}$. The small region between the IL and FOT line has been interpreted as a phase where the vortex lattice is unpinned, whereas below IL the vortex lattice is pinned [53]. In the reversible regime above $T_{irr}$ a clear diamagnetic signal is present up to temperatures larger than $T_c$. This region is characterized by fluctuations and there is no well defined upper critical temperature $T_{c2}$. The temperature $T_{\text{fluct}}$, at which diamagnetic (superconducting) fluctuations appear, has been defined as the temperature where the data begin to deviate from the horizontal normal state line (see Fig.2.3c). The simplest way to estimate $T_{c2}$ is to use an extrapolation method based on the linear Abrikosov formula [77] ($\kappa = \frac{\lambda}{\xi}$ is the GL parameter):

$$M(H,T) \sim \frac{H_{c2}(T) - H}{2\kappa^2 - 1}$$ (2.3)

The transition temperature $T_{c2}$ is derived from the intersection of a linear fit with the normal-state horizontal line, as shown in Fig.2.3c. It was shown that this procedure is not totally correct for HTSC, where the Abrikosov linear region is limited to a small temperature range because of the rounding close to $T_{c2}$ [78, 79]. Indeed in the underdoped regime, where fluctuations are larger, using extrapolation we get unphysical values for the upper critical field (positive slope of $H_{c2}(T)$, see later). However, a treatment of the data based on a scaling procedure proposed by Landau and Ott [79], allows one to obtain more reasonable upper critical lines for all the samples (see later).

Such kind of analysis has been performed for each samples. One can notice that the jump in $M(T)$ is more pronounced at high magnetic fields, and in UD-19K only a broad anomaly could be observed (to note is that in this sample the loss peaks in $\chi''(T)$ are very broad, as well). The irreversibility line and the FOT line are found to be close to each other in all samples, and are therefore possibly related to each other. In the following we will consider only the FOT line in the phase diagram, since it can be determined very precisely by $\chi(T)$ measurements in all samples and for each magnetic field.
Figure 2.4: FC (solid lines) and ZFC (dashed lines) $M(T)$ curves for UD-19K, OD-36K and OD-31K at different magnetic fields.
2.3 AC susceptibility and DC magnetization measurements

Figure 2.5: ZFC isothermic magnetisation curves for UD-19K, UD-29K, OD-36K and OD-31K. For OD-36K and OD-31K $H_p$ and $H_{sp}$ have been determined as indicated by the arrows. For UD-19K and UD-29K only $H_{sp}$ could be observed. The insets show some full hysteresis loops with $H_{irr}$.

2.3.2 Magnetic field scans

We also performed isothermic ZFC $M(H)$ measurements at different temperatures (see Fig.2.5). In the OD samples we could observe two minima in the $M(H)$ curves. Starting from zero field we have a first minimum $H_p$, which is known to be related to surface [80] and/or geometrical [81] barriers. Due to these barriers the field doesn’t penetrate the bulk at the lower critical field $H_{cl}$ but only at a higher penetration field $H_p$. By further increasing the field we have a maximum at $H_{on}$, which is then followed by a second (and larger) minimum at $H_{sp}$. $H_{on}$ is called the onset field of the second peak $H_{sp}$, which is related to some flux-pinning mechanism, although its origin is still controversial [82, 83, 37, 84].

In UD samples, on the contrary, only one minimum could be observed. We argue that this
is actually the second peak $H_{sp}$. The penetration field $H_p$ and onset field $H_{on}$ are most probably hidden, due to the low value of $H_{sp}$. This interpretation is supported by the fact that even in the OD samples it is difficult to identify $H_p$ and $H_{on}$ at high temperatures close to $T_c$ (where $H_{sp}$ occurs at low fields). Moreover, very accurate SQUID measurements on UD-29K clearly showed the presence of $H_{on}$ and $H_{sp}$ even in the underdoped regime [45]. We also performed some full hysteresis loops, as shown in the insets of Fig.2.5. The ascending and the descending branches of the hysteresis loops meet at $H_{irr}$, whose values are consistent with those obtained by FC-ZFC $M(T)$ curves.

2.4 Magnetic phase diagram

In order to facilitate the analysis and discussion of the experimental results, the characteristic fields (namely $H_p(T)$, $H_{on}(T)$, $H_{sp}(T)$, $H_{FOT}(T)$, $H_{c2}(T)$ and $H_{fluct}(T)$) of the four samples have been plotted in the $H$ vs $T$ phase diagrams shown in Fig.2.6 (both in linear and logarithmic scale). The magnetic phase diagram of HTSC is usually divided in four main phases [35] (see also Fig.1.6):

1. Above the upper critical field $H_{c2}(T)$ the system is in the non-superconducting state and the magnetic flux is free to enter the crystal homogeneously (normal phase).

2. Between $H_{c2}(T)$ and $H_{FOT}(T)$ (or $H_{irr}(T)$) the magnetic flux is partially expelled from the superconductor. The magnetic field is present in the sample in the form of flux-lines (vortices) which are in a reversible regime. In this region the vortices are thermally activated and highly dynamics in nature (vortex liquid phase).

3. Below $H_{FOT}(T)$ ($H_{irr}(T)$) the vortices are in an irreversible regime, as can be seen by the difference in the FC/ZFC data or in the hysteresis loops. Here the vortices are frozen in a lattice (VL), which can be directly observed in SANS experiments, see Chapter 3. In Fig.1.6 this phase is further divided into a Bragg glass and a vortex glass phase.

4. Below $H_p(T)$ (ideally $H_{c1}(T)$) the system is in the Meissner state and the flux is completely expelled from the bulk of the sample (Meissner phase).

Indeed we can roughly understand our results in LSCO within this description, even though we have some additional lines in the phase diagram. The first observation is that the phase diagrams of OD and UD LSCO are qualitatively similar but quantatively very different. In particular, in the UD samples the reversible region between $H_{FOT}(T)$ and $H_{c2}(T)$ is much larger, whereas the second peak line $H_{sp}(T)$ occurs at much lower fields. Before discussing the possible reasons for this strong doping dependence of the phase diagram we want to look in details at the single lines.

We start from the upper critical line $H_{c2}(T)$, which is not well defined, since fluctuations are very strong near $T_{c2}$. Such SC fluctuations are more pronounced in the underdoped regime, where diamagnetic fluctuations are present even at temperatures $T_{fluct}$ much larger than $T_c$. This anomalous behavior in the underdoped regime has also been observed in Nernst [85, 86, 87, 88, 89] and scanning SQUID microscopy [90] experiments.
This strange effect is present only in hole-doped HTSC and has been interpreted as being due to vortex-like excitations in the pseudogap region. As a consequence $H_{c2}(T)$ as determined by extrapolation shows an unphysical positive slope. In order to get more reasonable upper critical field lines, we used the Landau-Ott scaling procedure [79]. This scaling procedure, based on the Ginzburg-Landau theory, allows to establish the temperature dependence of the upper critical field $H_{c2}(T)$ from measurements of the reversible isothermal magnetization (the absolute value of $H_{c2}(0 \text{ K})$, however, has to be determined experimentally). We applied this procedure to our magnetization data, taking the values of $H_{c2}(0 \text{ K})$ from ref.[76] (see Table 2.2). The resulting $H_{c2}(T)$ lines are plotted in Fig.2.6 (bold lines). Since above $H_{c2}(T)$ a weak diamagnetic signal can still be measured, we can interpret this line as the field where the mixed state disappears and not necessarily as the field where superconductivity is completely suppressed. To note is that in the overdoped regime the upper critical field line determined by extrapolation roughly agrees with that obtained using the Landau-Ott scaling procedure. In the underdoped regime, on the contrary, the two methods give rise to different $H_{c2}$ lines. It is clear that the extrapolation method fails to describe the correct upper critical line when the SC fluctuations are too strong. However, this simple method is able to indicate that SC fluctuations are present above $T_c$, a real effect.

We turn now to the FOT line $H_{FOT}(T)$, which is usually identified with the melting line [46, 47, 48, 34], that is the transition of the vortex solid into a vortex liquid. The temperature dependence of $H_{FOT}(T)$ is predicted by the melting theory to be [34]:

$$ H_{melt}(T) = H_m \cdot \left(1 - \frac{T}{T_c}\right)^m $$

(2.4)

In this case the prefactor is known to depend almost only on the anisotropy of the system. In fact, considering $H_m \sim \gamma^{-2}T_c^{-2}\lambda_{ab}^{-4}$ (where $\lambda_{ab}$ is the in-plane penetration depth)[34] and the fact that $T_c^{-2}\lambda_{ab}^{-4}$ is almost constant, one obtains $H_m \sim \gamma^{-2}$. Fitting our data by this model is not satisfactory, since we obtain a huge doping dependence of the exponent $m$ and the prefactor $H_m$ doesn’t follow the expected $\gamma$ dependence (see Table 2.2). Moreover in all SANS experiments on HTSC [51, 93, 94, 95] the ring-like intensity between $H_{FOT}$ and $H_{c2}$, which is expected in a liquid of straight vortices, has never been observed (see also Chapter 3.3.2., Fig.3.18). A more precise melting theory, still based on the Lindemann criterion [49], predicts a more complicated temperature dependence of the melting line [34]:

$$ H_{melt}(T) = \frac{4c_L^4 H_{c2}(0) \frac{B}{G} \left(\frac{T_c}{T}\right)^2 - 1}{\left(1 + \sqrt{1 + 4c_L^4 B \frac{B}{G} \left(\frac{T_c}{T}\right)^2 - 1}\right)^2} $$

(2.5)

where $G = \frac{1}{2} \left(\frac{\gamma \mu B T_c}{(\pi \mu_0) H_{c2}(0) \lambda_{ab}(0)}\right)^2$ is the Ginzburg number, $B \approx 5.6$ and $c_L$ is the Lindemann number. However, the agreement between the experimental results and the fitted curves is quite poor (see Fig.2.7a for UD-29K). Moreover, $c_L$ is doping dependent and in some cases higher than the expected values ($c_L \sim 0.1-0.2$).
Figure 2.6: Magnetic phase diagram of the four LSCO samples both in linear and double-logarithmic scale. The magnetic field direction is approximately perpendicular to the CuO$_2$ planes. The lines are fit of the different models, see text.
An alternative to the melting transition is given by the sublimation theory [52, 53], based on the strong anisotropy originating from the layered structure intrinsic to all HTSC. In this scenario, the melting is accompanied by the simultaneous decoupling of the vortex lines into 2D pancakes vortices (therefore sublimation from a VL to a vortex gas). The phenomenological scaling law which applies to all HTSC is given by:

\[ H_{\text{subl}}(T)|_{Oc} = 2.85\gamma^{-2}s^{-1}\left(\frac{T_c}{T} - 1\right) \]  

(2.6)

where \( s \) is the intralayer distance (\( s = 6.6 \times 10^{-8} \) cm in LSCO). This formula has been used in order to explain the FOT transition in many HTSC [52, 53] and nicely fits our data (see Fig.2.6 and Fig.2.7a). The good agreement between the extracted and measured values of the anisotropy \( \gamma \) (see Table 2.2) should be noted.

It remains to discuss the second peak line \( H_{\text{sp}}(T) \) which has been explained on the basis of thermal decoupling theory [98, 99, 100], which predicts the suppression of long-range order in the direction of the applied field due to thermal fluctuations. The expected
Table 2.2: Characteristic parameters for LSCO as a function of the Sr concentration $x$. The values of the upper critical field $H_{c2}(0 \text{ K})$ [76], of the penetration depth $\lambda_{ab}$ [96], and of the anisotropy $\gamma$ [52, 53, 72, 97] have been extrapolated from experimental values found in the literature (for $\gamma$, see also Fig.2.1). $H_m$ and $m$ have been obtained by fitting the data using formula (2.4), the Lindemann number $c_L$ using formula (2.5). $\gamma_{\text{dec}}$ and $\gamma_{\text{subl}}$ are the anisotropies obtained by fitting our data using the decoupling, respectively sublimation models. Finally, $n$ is the exponent of the power law (2.8).

<table>
<thead>
<tr>
<th>sample</th>
<th>$x$</th>
<th>$H_{c2}(0) [\text{T}]$</th>
<th>$\lambda_{ab}(0) [\text{Å}]$</th>
<th>$\gamma$</th>
<th>$H_m [\text{T}]$</th>
<th>$m$</th>
<th>$c_L$</th>
<th>$\gamma_{\text{subl}}$</th>
<th>$\gamma_{\text{dec}}$</th>
<th>$n$</th>
</tr>
</thead>
<tbody>
<tr>
<td>UD-19K</td>
<td>0.075</td>
<td>35</td>
<td>~3000</td>
<td>60(5)</td>
<td>15</td>
<td>6.1</td>
<td>0.16</td>
<td>64</td>
<td>85</td>
<td>2.5</td>
</tr>
<tr>
<td>UD-29K</td>
<td>0.10</td>
<td>45</td>
<td>2800</td>
<td>45(5)</td>
<td>15</td>
<td>3.3</td>
<td>0.20</td>
<td>47</td>
<td>40</td>
<td>2.1</td>
</tr>
<tr>
<td>OD-37K</td>
<td>0.17</td>
<td>60</td>
<td>~2400</td>
<td>20(2)</td>
<td>28</td>
<td>1.8</td>
<td>0.29</td>
<td>22</td>
<td>13</td>
<td>2.1</td>
</tr>
<tr>
<td>OD-31K</td>
<td>0.20</td>
<td>45</td>
<td>1970</td>
<td>20(2)</td>
<td>30</td>
<td>1.7</td>
<td>0.28</td>
<td>20</td>
<td>12</td>
<td>2.3</td>
</tr>
</tbody>
</table>

Figure 2.9: Temperature dependence of $H_{sp} \cdot \gamma^3$ plotted in a double logarithmic scale. All the data measured in samples with different doping levels collapse on one line with slope \~2. This indicates that the power law (2.8) has an exponent $n \approx 2$ and $H_0 \propto \gamma^{-3}$. (We have used the values of $\gamma$ obtained by fitting our data using Eq.(2.6) ($\gamma_{\text{subl}}$ in Table 2.2)).

temperature dependence is [100]

$$H_{\text{dec}}(T) = B^* \cdot \left( \frac{T_c}{T} - 1 \right)$$  \hspace{1cm} (2.7)

with $B^* = \Phi_0^3/(16\pi^3 e k_B \mu_0 s \gamma^2 T_c \lambda_{ab}(0)^2)$, where $\Phi_0$ is the flux quantum and $e \approx 2.718$ is the exponential number. This function doesn’t fit well our data, as shown in Fig.2.7b for
OD-36K. Moreover the estimated value for $\gamma$, obtained by substituting the known values of $s$, $T_c$ and $\lambda_{ab}(0)$ in the theoretical expression for $B^*$, are not satisfactory compared to the experimental values (see Table 2.2). Furthermore recent SANS measurements [95] (see Chapter 3) indicate that a well defined vortex lattice persists up to $H_{FOT}(T)$ and therefore discredit the decoupling theory. The origin of the second peak is most probably related to some change of the pinning strength with increasing field. The experimental data are better fitted by a power law [101, 102]

$$H_{sp}(T) = H_0 \cdot \left(1 - \frac{T}{T_c}\right)^n$$

as can be seen in Fig.2.6, Fig.2.7b and Fig.2.9. The value of the exponent is close to $n=2$ in all samples (see Fig.2.9 and Table 2.2). Interestingly the value of $H_0$ seems to be proportional to $\gamma^{-3}$, even though (up to our knowledge) no theory predicts such a $\gamma$ dependence.

The onset of the second peak $H_{on}(T)$ has often been considered to be the relevant field from the physical point of view [103], since there the pinning force density $F_p \sim H \cdot J_c \sim H \cdot \Delta M$ ($J_c$ being the critical current and $\Delta M$ the hysteresis loop width at $H$) has a minimum [72, 104]. This line has been interpreted as the disorder-induced transition from a relatively ordered VL to a highly disordered vortex glass, as a consequence of the competition between the vortex elastic energy and the pinning energy [37, 84, 38, 103]. According to this model, $H_{on}(T)$ has a temperature dependence of the form [103]:

$$H_{dis}(T) = H_d \cdot \left[1 - \left(\frac{T}{T_c}\right)^4\right]^{3/2}$$

This function, however, shows an opposite curvature and can hardly fit our data, which are better described by a simple exponential (see Fig.2.8):

$$H_{exp}(T) = H_1 \cdot \exp\left(-\frac{T}{T_1}\right)$$

Such a temperature dependence has been reported in Tl$_2$Ba$_2$CuO$_6$ [105] and HgBa$_2$CuO$_4$ [106]. We will come back to this on Chapter 3, when macroscopic results will be compared to SANS measurements.

Finally we spend few words about $H_p(T)$, which could be measured only in the OD samples. Its temperature dependent behavior has been predicted by the surface and geometrical barrier models [80, 81]:

$$H_{surf} \sim \exp\left(-\frac{T}{T_0}\right)$$

$$H_{geom} \sim \left(1 - \frac{T}{T_c}\right)$$

As shown in Fig.2.8, the surface barrier model [80] is in good agreement with our data in OD-36K and OD-31K, whereas the geometrical barrier model [81] is not that successful.
2.5 Conclusions from bulk measurements

Our macroscopic measurements of LSCO single crystals in a magnetic field revealed an extremely rich magnetic phase diagram. Besides the three conventional regions present in all Type-II superconductors (Meissner state, mixed state and normal state), we observed more exotic phases (see Fig.2.10):

- Above the upper critical line we observed a region characterized by strong superconducting fluctuations, which are more pronounced in the underdoped regime.
- The mixed phase is split into at least three phases: vortex lattice (phase A and phase B) and vortex gas.

The two vortex lattice phases A and B have different properties, and will be discussed later in Chapter 3 on the basis of our SANS experiments. At high temperatures and fields the vortex lattice undergoes a first order transition to a vortex gas, whereas the conventional melting scenario is not supported by our data. The sublimation transition is accompanied by a jump in the DC magnetization and a peak in the imaginary part of the AC susceptibility. This result is important, since the peak in $\chi''$ has been originally associated with the upper critical line and then with the irreversibility line, whereas our data seem to indicate that it occurs at the first order sublimation transition.

The magnetic phase diagram turned out to be strongly doping dependent, from a quantitative point of view, whereas qualitatively all the samples display the same transitions (second peak, FOT, upper critical line, ...). The quantitative doping dependence of the magnetic phase diagram can mainly be explained by the increasing anisotropy with underdoping (see Fig.2.1):

- The second peak line $H_{sp}(T)$ is proportional to $\gamma^{-3}$
- The sublimation line $H_{FOT}(T)$ is proportional to $\gamma^{-2}$

As a consequence, the vortex gas phase is much more extended in the underdoped regime of LSCO and, more generally, in highly anisotropic systems (e.g. Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$).

The interpretation of the second peak in LSCO is still controversial. We report for the first time a strong anisotropy dependence ($\sim \gamma^{-3}$) which is not predicted by any theory. Even though our macroscopic measurements are insufficient to draw definitive conclusions, our data seem to discredit the decoupling scenario as being responsible for the second peak. We will come back to this problem in Chapter 3 with the support of results from SANS and $\mu$SR measurements.
2.6 Appendix A- Angular dependence of $H_{FOT}(T)$

In this section the angular dependence of the phase diagram, and in particular of $T_{FOT}$, is shortly discussed. Due to the large anisotropy, all critical lines are expected to be strongly dependent on the angle $\Theta$ between the field $H$ and the $c$-axis. Introducing an effective mass tensor [107], one can take into account the effect of the anisotropy on the physical quantities such as the penetration depth or coherence length. Scaling expressions can be found also for other quantities, such as the upper and lower critical fields, depinning and melting lines [73], and have been verified experimentally in YBCO [108, 109].

Following these angular scaling rules for (uniaxial) anisotropic superconductors we expect the FOT line to be:

$$H_{FOT}(T, \Theta) = \frac{1}{\sqrt{\gamma^{-2}\sin^2(\Theta) + \cos^2(\Theta)}} H_{FOT}(T, \Theta = 0)$$  \hspace{1cm} (2.13)

Within the sublimation scenario (see Eq.(2.6)), the theoretical expression for $T_{FOT}(\Theta)$ is then given by [53]:

$$T_{FOT}(\Theta) = \frac{T_c}{\gamma^2 H \sqrt{\gamma^{-2}\sin^2(\Theta) + \cos^2(\Theta)}} + 1$$  \hspace{1cm} (2.14)

Fig.2.11a shows the magnetic phase diagram for UD-29K, as determined by AC susceptibility measurements. The field was applied (approximately) parallel (filled circles) and perpendicular (empty circles) to the c-axis and the transition temperature $T_{FOT}$ has been determined by the peak position of $\chi''(T)$ for fields up to 9 Tesla. The curves have been fitted within the sublimation theory (Eq.(2.6) and Eq.(2.13)). The value of $\gamma=47$ is obtained for $\Theta = 0^\circ$. The data with $H//ab$ have been fitted with $\gamma=47$ fix and $\Theta$ as only free parameter and yields a value of $\Theta = 86^\circ$ which is within the precision of the alignment of the crystal with respect to the external field.
36 Macroscopic measurements

Figure 2.11: a) FOT lines for magnetic fields applied parallel ($H//c$) and perpendicular ($H//ab$) to the CuO$_2$ planes in UD-29K. The lines are fits within the sublimation theory, see Eq.(2.6) and Eq.(2.13). b) Angular dependence of the phase transition line $T_{FOT}$ in UD-29K at different magnetic fields. The curves are the theoretical predictions (see Eq.(2.14)) without free parameters.

The angular dependence of $T_{FOT}$ at various magnetic fields is plotted in Fig.2.11b together with the theoretical predictions (Eq.(2.14)). To notice is that the curves have been obtained without any free parameter. Even though we have measured $T_{FOT}$ only at two angles, the theory is in good agreement with our experimental data. This result is important for two reasons. On one hand it confirms that the value of the anisotropy obtained in the sublimation scenario is not only consistent to resistivity measurements but also to the angular dependence of $T_{FOT}$. On the other hand it clearly shows that the phase diagram is not strongly affected by small $\Theta$. Therefore, the precision of the alignment of the c-axis with respect to the field direction (in our case $10^\circ$) is sufficient for the investigation of the doping dependence of the phase diagram: our results obtained in Section 2.3-2.5 are intrinsic and not affected by slight misalignments of the samples.

2.7 Appendix B- Specific heat measurements

In this section we present specific heat measurements performed on a piece ($\sim 12$ mg) of the UD-29K used in $M$, $\chi$ and INS experiments. The goal of this experiment was to check the quality of the single crystal and to determine its SC volume fraction. In general, the specific heat of a metal has two contributions:

$$C(T) = C_{el}(T) + C_{ph}(T)$$

(2.15)

where $C_{el}(T) = \gamma_{el} \cdot T$ is the electronic and $C_{ph}(T) = \beta \cdot T^3$ the phononic term.

The low-temperature specific heat of UD-29K, together with measurements on a polycrystalline LSCO (x=0.10) and non-SC Ni-doped polycrystalline La$_{1.9}$Sr$_{0.1}$Cu$_{0.98}$Ni$_{0.02}$O$_4$
2.7 Appendix B- Specific heat measurements

Figure 2.12: a) Low temperature specific heat of $La_{1.9}Sr_{0.1}CuO_4$ single crystal (UD-29K) and polycrystal, and of non-SC $La_{1.9}Sr_{0.1}Cu_{0.98}Ni_{0.02}O_4$, in a $C/T vs T^2$ plot. b) Electronic part of the specific heat in UD-29K, obtained after subtracting the phononic part (see text). The typical anomaly at $T_c$ is clearly observed.

(LSCNO), are shown in Fig2.12a in a $C/T vs T^2$ plot. The data of the single crystal are in agreement with results obtained on polycrystalline samples. To notice is that even in this SC sample, a residual $\gamma^{el}=0.5-1$ mJ/K$^2$mole is present. In the non-superconducting sample (which we consider as a reference for the normal state) we obtain $\gamma^{el}=5.5$ mJ/K$^2$mole. The SC volume fraction can be defined as [110]:

$$SC \text{ volume fraction} = \frac{\gamma_n^{el} - \gamma_r^{el}}{\gamma_n^{el}} \tag{2.16}$$

For UD-29K we obtain a SC volume fraction larger than 80%.

In the superconducting state, the electronic specific heat is modified, and an anomaly at $T_c$ is expected. However, in order to observe it, the phononic contribution has to be removed. This is done by subtracting the specific heat in the normal state, which can be obtained by applying a magnetic field larger than $H_{c2}$. However in LSCO this is not possible, since $H_{c2}(0 \text{ K})\sim50$ T is extremely large [76]. On the other hand we can subtract the phononic term obtained in non-SC LSCNO with the same Sr content as the SC sample, since the partial substitution of Ni has little effect on the phonon term [111]. $C_v(T)/T$ of UD-29K obtained in this way is shown in Fig.2.12b. A sharp anomaly is observed at $T_c$, therefore indicating the good quality of the single crystal.

In future it would be interesting to measure $C_v(H)$ and investigate the field dependence of the specific heat anomaly. In YBCO and BSCCO the anomaly rapidly vanishes at $H \ll H_{c2}$ [112, 113], whereas (up to our knowledge) measurements in LSCO are still missing.
Macroscopic measurements
Chapter 3

Small angle neutron scattering study of the vortex lattice

We report here on the first direct measurements of an ordered vortex lattice in the bulk of LSCO in both underdoped and overdoped regimes. In the overdoped regime we have evidence for a field-induced transition from hexagonal to square coordination of the vortex lattice. This represents the first observation of an intrinsic square vortex lattice in high temperature superconductors. Moreover, it is found that the diffraction signal disappears at temperatures well below $T_c$, due to the sublimation of the vortex lattice. In the underdoped regime we observed a more disordered vortex glass phase by means of $\mu$SR measurements combined with SANS and macroscopic experiments.

3.1 Introduction

Information about the vortex lattice (VL) can be obtained by a number of experimental techniques. As shown in Chapter 2, magnetization measurements are useful to investigate the magnetic phase diagram from a macroscopic point of view. However, in order to investigate the morphology of the vortex system, one needs other experimental methods, such as scanning tunneling microscopy (STM) [114], Bitter decoration [115], magneto optical imaging (MOI) [116, 117], Lorentz microscopy [118], muon spin rotation ($\mu$SR) [67] or small angle neutron scattering (SANS)[119]. Most of these techniques (STM, Bitter decoration, MOI) are limited to imaging the VL at the surface of the superconductor, whereas some of them can be used only at low magnetic fields (Bitter decoration). SANS doesn’t suffer from these limitations and can provide unique information concerning the long-range order of the vortex system in the bulk of the sample up to high magnetic fields (11 Tesla at PSI). $\mu$SR, on the other hand, probes the magnetic field distribution on a local scale, and is complementary to SANS.

The properties of neutrons make the SANS technique unique and useful: due to their neutrality, neutrons can easily penetrate the matter (therefore providing bulk information), and because of their magnetic moment they are sensitive to any varying magnetic field distribution. In particular, from the diffraction pattern the VL symmetry can be directly determined, whereas from the scattered intensity one can obtain (in principle) the SC length scales $\lambda$ and $\xi$. Moreover, the width of the peaks reveals the correlation
Small angle neutron scattering study of the vortex lattice

Figure 3.1: Schematic view of a vortex lattice with a) four nearest neighbors (square VL) and b) six nearest neighbors (hexagonal VL). The d-spacing is given by the condition that there is only one flux quantum $\Phi_0$ per unit cell.

lengths of the VL.

The condition for coherent elastic neutron scattering is the same as the Bragg law for x-rays scattering

$$\lambda_n = 2d \sin(\theta) \quad (3.1)$$

where $\lambda_n$ is the neutron wavelength, $d$ is the spacing of crystal planes, and $\theta$ is the scattering angle. This formula can be applied to a crystal of magnetic vortices (vortex lattice), as well. The distance $d$ is given by the condition that each vortex carries a flux quantum $\Phi_0 = \frac{hc}{2e} \approx 2.0679 \times 10^5$ TÅ$^2$:

$$d = \sqrt{\frac{\sigma}{B}} \Phi_0 \quad (3.2)$$

where $\sigma$ depends on the symmetry of the vortex lattice ($\sigma$ is equal to 1 for square VL, and $\sqrt{3}/2$ for hexagonal VL, see Fig.3.1). $B$ is the average flux density, which for field-cooled measurements is ideally equal to the external magnetic field ($B \approx H$). The most favorable configuration (lowest energy) for an isotropic conventional s-wave Type-II superconductor was calculated to be a hexagonal VL by Kleiner et al. [120], following the pioneering work of Abrikosov [77]. In Fig.3.2 a SANS diffraction pattern in Niobium is shown, which clearly displays a hexagonal symmetry of the VL. On the other hand, in d-wave superconductors, the anisotropic order parameter is able to affect the symmetry and orientation of the VL [121, 122, 123, 124, 125, 126, 127, 128, 129]. As it will be discussed later, these d-wave effects have to be taken into account when analyzing the data at high fields.

Diffraction experiments on VL require a SANS instrument, since $d$ can be very large and consequently $\theta$ becomes very small. An external field of 1 Tesla, for example, gives $d \sim 450$ Å, and using $\lambda_n=10$ Å, one gets a small Bragg angle $2\theta \sim 1.3^\circ$. The intensity $I_{hk}$ of a single $(h,k)$ reflection (integrated over the rocking curve of the VL) can be derived from the neutron scattering magnetic cross section [66], and is given by [119]

$$I(q_{hk}) = 2\pi \phi \left( \frac{g}{4} \right)^2 \left( \frac{V}{\Phi_0^2} \right) \left| F(q_{hk}) \right|^2 \sim \left| F(q_{hk}) \right|^2 \frac{q_{hk}}{q_{hk}} \quad (3.3)$$

---

1W.H. Bragg and W.L. Bragg won the Nobel Prize in Physics in 1915 "for their services in the analysis of crystal structure by means of X-rays"
3.1 Introduction

Figure 3.2: SANS diffraction pattern from Niobium single crystal at $\mu_0 H=0.2$ T, $T=2$ K showing a hexagonal vortex lattice. Higher order spots are clearly seen. (left panel: linear color scale, right panel: logarithmic color scale)

where $\phi$ is the incident neutron flux, $g$ is the neutron magnetic moment, $V$ is the sample volume, and $q_{hk} = \frac{2\pi}{d}$ is the $(h,k)$ reciprocal lattice vector of the VL. $|F(q_{hk})|^2$ is the magnetic form factor, and $F(q_{hk})$ is the Fourier component of the spatial field distribution inside the sample:

$$B(r) = \sum_{h,k} F(q_{hk}) e^{i q_{hk} \cdot r}$$  \hspace{1cm} (3.4)

Within the London theory (high-$\kappa$ approximation valid for fields well below the upper critical field), $F(q_{hk})$ can be calculated. Starting from the London equation

$$B(r) + \lambda^2 [\nabla \times \nabla \times B(r)] = 0$$  \hspace{1cm} (3.5)

and introducing the vortex lines positioned at the sites $r_i$ one gets:

$$B(r) + \lambda^2 (\nabla \times \nabla \times B(r)) = \Phi_0 \sum_i \delta(r-r_i)$$  \hspace{1cm} (3.6)

$\lambda$ is the penetration depth, whereas $\delta(r)$ is a two-dimensional delta function. Combining the Fourier expansion (3.4) and equation (3.6) gives the following result:

$$F(q_{hk}) = \frac{B}{1 + (q_{hk} \lambda)^2} \approx \frac{B}{(q_{hk} \lambda)^2} = \frac{\Phi_0 \sigma}{(2\pi \lambda)^2}$$  \hspace{1cm} (3.7)

The second term in the denominator is usually dominant, and therefore the vortex lattice intensity (given in equation (3.3)) depends on the (temperature and field dependent [67]) penetration depth:

$$I(q_{hk}) \sim \lambda^{-4}$$  \hspace{1cm} (3.8)

In principle, the temperature dependence of the VL intensity is a direct measure of the penetration depth. However, in HTSC this is not necessarily the case, since thermal fluctuations and disorder are likely to play an important role.
Within the London approximation, the form factor is field independent (see Eq. (3.7)), and the field dependence of the VL intensity is given by:

\[ I(q_{hk}) \sim q_{hk}^{-1} \sim H^{-\frac{1}{2}} \] (3.9)

As one increases the field, the vortex lines get closer to each other, and one could expect that the magnetic fields of the single vortices overlap and reduce the spatial variation of the field distribution which gives rise to diffraction. However, the London theory ignores the vortex cores of radius \( \xi \) (coherence length), \( B(r) \) diverges at the center of the vortex line, and this sharp variation is not removed by increasing the density of vortices. These core effects can be included by the introduction of a cutoff factor of the form \( \exp(-2q\xi) \) [130, 131], which may also be written as \( \exp(-\frac{2}{\sigma}\sqrt{\pi}(H/Hc)^{1/2}) \), therefore leading to a quicker fall off of intensity with increasing field:

\[ F(q_{hk}) = \frac{\Phi_0 \sigma}{(2\pi)^2} e^{-\frac{2}{\sigma}\sqrt{\pi}(H/Hc)^{1/2}} \] (3.10)

The field dependence of the VL intensity might also be influenced by the penetration depth. In a d-wave superconductor, \( \lambda \) is expected to increase linearly with field in the Meissner state, due to nonlinear 2 effects [67]:

\[ \frac{\lambda(H)}{\lambda(0)} = 1 + \beta \frac{H}{H_0} \] (3.11)

where \( \beta \) is a temperature dependent coefficient that remains finite at \( T=0 \) K, and \( H_0 \) a characteristic field on the order of the thermodynamic critical field \( H_c \). In the vortex state the situation is complicated by the fact that the distance between vortices decreases with increasing field. \( \mu \)SR measurements in YBCO indicate that \( \lambda(H) \) is linear at low fields (\( H < 2 \) T) with \( \beta \sim 7 \cdot 10^{-2} \) (and \( H_0 \sim H_c = H_{c2}/\sqrt{2\kappa} \sim 1T \)) [133], but deviates from linearity at higher fields [134]. Numerical calculations by Amin et al. [135, 136], which included the influence of nonlinear and nonlocal 3 effects, could reproduce the experimental data very well.

Combining Eq.(3.10) and Eq.(3.11) with Eq.(3.3) we end up with the formula:

\[ I(q_{hk}) \sim H^{-\frac{1}{2}} \left( 1 + \beta \frac{H}{H_0} \right)^{-4} e^{-4\sqrt{2}(H/H_{c2})^{1/2}} \] (3.12)

Due to the large values of \( \lambda \), the VL intensity in HTSC is rather small (see Eq.(3.8)). Successful SANS experiments have been reported in BSCCO [51] and YBCO [137, 138, 139]. On the contrary, only one report about SANS measurements in LSCO is found in the literature [140]. The quality of their data is quite poor, and only a polycrystalline VL could be observed. The lack of information about the long-range order of the vortex

\[ ^2 \text{The supercurrent density } j \text{ is not linear with the velocity of the superfluid } \mathbf{v}_s, \text{ when } \mathbf{v}_s \text{ is larger than a critical velocity, see e.g. [132]} \]

\[ ^3 \text{Nonlocality is caused by the finite size } \xi_0 \text{ of the Cooper pairs. Instead of the local relations between the supercurrent density } j \text{ and the vector potential } \mathbf{A}, \text{ one has to consider that } j(\mathbf{r}) \text{ is determined by } \mathbf{A}(\mathbf{r}) \text{ over a surrounding volume of radius } \xi_0. \text{ Nonlocal effects become important in } d \text{-wave superconductors, since } \xi_0 \text{ diverges at the nodes of the SC energy gap.} \]
lattice in LSCO motivated our SANS investigations. In this chapter we will present SANS measurements on LSCO, which represent the first direct observations of a well ordered VL in this compound. After our first successful experiments in slightly overdoped LSCO ($x=0.17$) at low temperatures ($\leq 5$ K) and low magnetic fields ($\leq 1.2$ T) [141], we extended our investigations to the complete $H - T$ phase diagram (up to 10.5 Tesla) [95] and at different doping levels [142, 45]. In particular, we investigated the field, temperature and doping dependence of the VL.

### 3.2 Experimental setup

Our experiments were performed on the small angle neutron scattering (SANS) instruments of both the PSI (SANS-I and SANS-II), Switzerland, and the Institut Laue Langevin (D22), Grenoble, France. A schematic view of a typical SANS beamline is shown in Fig.3.3. A velocity selector is used to select the neutron wavelength $\lambda_n$. Typical wavelengths used for the study of VL are between 4.5 and 20 Å ($\Delta \lambda_n/\lambda_n=10\%$). Before reaching the sample the incident neutrons are then collimated over a distance which can be varied from 1 to 18 m. The collimator consists of two pinholes, and defines the beam divergence. The samples are glued on aluminum sample holders (masked with Cadmium), mounted in a cryomagnet and placed on a sample table which can be rotated and tilted. For the vortex lattice studies presented here, we used cryomagnets allowing to reach temperatures down to 1.5 K and horizontal magnetic fields up to 11 Tesla. Finally, a 96x96 cm$^2$ position sensitive detector at a variable distance between 4.5 and 18 m was used to detect the scattered neutrons. No analysis of the energy of the neutrons can be performed, and the total number of counted neutrons represents an integration over all neutron energies. Both the collimator and the detector are kept in vacuum in order to reduce air scattering and beam attenuation, and all interfaces between vacuum and air were through single crystal sapphire windows in order to reduce the background.

The VL intensity is usually small compared to small angle defect scattering from the sample (see Fig.1.11). It is therefore necessary to remove this background from the signal.
arising from the VL. This can be done in a clean way by measuring a diffraction pattern above \( T_c \) (with or without field) and subtracting this signal from the data obtained in the superconducting state. The beam center, which defines the origin of the reciprocal space, can be determined by measuring the direct beam (after removing the beam stop and using some attenuator).

The integrated intensity of the VL is determined by measuring the scattered neutrons at a number of discrete angles while rotating the sample. During such a measurement the reciprocal lattice vector is rotated (or rocked) through the Ewalds sphere. Rocking curves can be performed both by rotating (\( \omega \)) or tilting (\( \psi \)) the sample, as shown in Fig.3.4.

The Bragg reflections measured in SANS experiments have a finite width (in three directions: radial, azimuthal and longitudinal) due to VL imperfections and instrumental resolution. After deconvolution, and assuming an exponential decay of the correlation function, the intrinsic width is directly related to the correlation length through the relation \( \xi_{\text{corr}} \approx \frac{2}{\Delta q} \), where \( \Delta q \) is the full width at half maximum of a Lorentzian fit to the data [143].

The resolution in the radial, azimuthal and longitudinal directions is different. In the SANS geometry discussed above, the longitudinal resolution is smaller than the in-plane resolution, therefore allowing the study of the VL order along the direction of the vortices. The instrumental resolution is affected by the velocity selector (\( vs \)), the collimation (\( coll \))
and the detector. Combining the different components one gets [143]:

\[ \Delta q_{\text{radial}} = \sqrt{\left( \frac{2\pi}{\lambda_n} \Delta(2\theta)_{\text{vs}} \right)^2 + \left( \frac{2\pi}{\lambda_n} \Delta(2\theta)_{\text{coll}} \right)^2} \]

\[ \Delta q_{\text{azimut.}} = \frac{2\pi}{\lambda_n} \Delta(2\theta)_{\text{coll}} \]  \hspace{1cm} (3.13)

\[ \Delta q_{\text{longit.}} = \sqrt{\left( q_{hk} \Delta(2\theta)_{\text{vs}} \right)^2 + \left( q_{hk} \Delta(2\theta)_{\text{coll}} \right)^2} \]

with

\[ \Delta(2\theta)_{\text{vs}} = \frac{q_{hk} \lambda_n}{4\pi} \sqrt{\frac{4n2}{3} \Delta \lambda_n} \]  \hspace{1cm} (3.14)

\[ \Delta(2\theta)_{\text{coll}} = 2\sqrt{2n2} \sqrt{\frac{(w_1/l)^2 + (w_2/l)^2}{4}} \]

where \( w_1 \) (\( w_2 \)) is the diameter of the pinhole at the beginning (end) of the collimation and \( l \) is the distance between them.

## 3.3 Overdoped regime of La\( _{2-x}\)Sr\( _x\)CuO\( _4\)

We investigated LSCO samples in the overdoped regime at two different doping levels: \( x=0.17 \) (\( T_c=37 \) K) and \( x=0.20 \) (\( T_c=31 \) K). All single crystals have been grown by the TSFZ method to a cylindrical shape, with the \( c \)-axis perpendicular to the axis of the cylinder. At both doping levels we obtained similar results, therefore suggesting that these are generic features of the overdoped regime of LSCO. We will mainly concentrate on LSCO \( (x=0.17) \), since it is on these samples that we performed most of the experiments.

### 3.3.1 Field-induced hexagonal-to-square transition

#### Experimental results in LSCO \( (x=0.17), \; 0 \; T < \mu_0 H < 1.2 \; T \)

We start with the results obtained on slightly overdoped LSCO \( (x=0.17) \) at low temperatures \( (T=1.5 - 5 \) K). In Fig.3.5 are shown two SANS diffraction patterns measured at \( 0.1 \) T and \( 1 \) T at base temperature \( (T=1.5 \) K) cooled in a field (FC), with the \( c \)-axis oriented along the field direction. At \( \mu_0 H=0.1 \) T a large number of spots are distributed around a ring, whereas at \( \mu_0 H=1 \) T a completely different pattern emerges, with all of the magnetic scattering concentrated in four intense spots appearing along the Cu-O bonds directions, forming a perfect square. To notice is that the rocking curves were found to be extremely broad (see Fig.3.6), indicating that the VL is deformed. We could therefore measure the intensity at a fixed angle.

Such 2-dimensional patterns can be analyzed in different ways\(^4\). For example, one can produce radially averaged \( (I \; vs \; q) \) or azimuthal averaged \( (I \; vs \; \varphi) \) one dimensional plots. In Fig.3.7 two examples of such analysis are shown for data taken at \( \mu_0 H=1 \) T. From

\(^4\)For the analysis of the data we used the Graphical Reduction and Analysis SANS Program for Matlab (GRASP) by Charles Dewhurst (dewhurst@ill.fr), www.ill.fr/lss/grasp/
Figure 3.5: LSCO (x=0.17): SANS diffraction pattern obtained after field cooling in a) 0.1 T, and b) 1 T after subtracting a background measured above \( T_c \). The Cu-O bond directions (\{1,1,0\} in orthorhombic notation) is indicated by the arrows.

Figure 3.6: a) Rocking curve of the four spots and b) sum of all rocking curves. Within the available \( \omega \)-range a maximum is not visible, therefore indicating extremely broad (flat) rocking curves. The line in b) is a guide to the eye.

\( I \) vs \( \varphi \) plots, the square symmetry is clearly established (the four spots are 90 degrees apart from each other), whereas \( I \) vs \( q \) curves can be fitted by a Gaussian and are used to determine the position of the Bragg peaks in reciprocal space. The value of \( q_{\text{peak}} \) is very important, since it is directly related to the magnetic induction \( B \) through a symmetry dependent quantity \( \sigma \) (see Eq.(3.2)):

\[
\sigma = \left( \frac{2\pi}{q_{\text{peak}}} \right)^2 \frac{B}{\Phi_0}
\]  

(3.15)
Figure 3.7: LSCO (x=0.17): Examples of two types of analysis performed on SANS patterns (in this case at $\mu_0H=1$ T): radially averaged ($I$ vs $q$) and azimuthal averaged ($I$ vs $\varphi$) 1D plots can be obtained from the 2D data.

Some $I$ vs $q$ curves measured at different magnetic fields are shown in Fig.3.8a. As expected from Eq. (3.15), $q_{\text{peak}}$ gets larger with increasing field, therefore establishing the VL origin of the neutron signal.

The value of $\sigma$ obtained at different fields is plotted in Fig.3.8b: for magnetic fields larger than $B^* \sim 0.4$ T, the experimental values of $\sigma$ are as expected for a square VL ($\sigma=1$), while at low fields ($\lesssim 0.1$ T) they are consistent with that of a hexagonal VL ($\sigma=0.866$). An alternative analysis of the data consists in monitoring the intensity ratio of sectors ($\pm 15^\circ$) containing the $\{1,0\}$ and $\{1,1\}$-type directions. As shown in Fig.3.8b this ratio decreases steadily from 1 (isotropic ring-like intensity distribution) at $\mu_0H=0.1$ T to about 0.2 at $\mu_0H=0.5$ T (intensity concentrated in four spots oriented along the $\{1,1,0\}$ directions) and then remains constant. These results confirm that at low fields the VL has a hexagonal symmetry, whereas at higher fields the symmetry of the VL becomes square.

---

5 The intensities have been rescaled to allow a common vertical axis. The field dependence of the VL intensity will be discussed later.

6 In principle, the intensity ratio should go to zero when the VL is square. In our case the intensity ratio saturates to a positive value ($\sim 0.2$), indicating that some intensity is still present in the $\{1,0\}$ sectors. This intensity is mainly due to the slightly disordered vortex lattice which leads to flat rocking curves and broad spots in reciprocal space. Moreover, there might be some contribution from the (broad...
Figure 3.8: LSCO (x=0.17): a) $I$ vs $q$ curves for $\mu_0H=0.2$ T (circles), 0.5 T (squares) and 0.8 T (triangles) at $T=5$ K. The intensities have been rescaled to allow a common vertical axis. The shift in $q$ illustrates the expected dependence of the vortex lattice spacing on increasing magnetic field. b) The black circles show the field dependence of $\sigma = (2\pi/q_{\text{peak}})^2 B/\Phi_0$ at $T=1.5$ K. The horizontal lines represent the expected values of $\sigma$ for hexagonal and square VL’s. The open circles show the intensity ratio of sectors containing the $\{1,0\}$ and $\{1,1\}$ directions, see text.

The ring-like pattern observed at low fields most likely consists of a superposition of diffraction from various domain orientations of hexagonal coordination. In order to confirm this hexagonal coordination, it is necessary to reduce the high degree of degeneracy apparent in Fig.3.5a (the same figure is shown as a contour plot in Fig.3.9a). This was achieved by rotating the c-axis (about the vertical axis) away from the field direction by a sufficiently large angle $\Theta$. The result of this procedure is shown in Fig.3.9b where a hexagonal pattern is indeed observed, and is very reminiscent of what has been seen in untwinned YBCO crystals \[144\]. In that system, a small amount of residual twin planes appears to control the orientation of the VL, giving rise to a high level of degeneracy. As the c-axis is rotated away from the field direction, the hexagonal symmetry becomes evident. We have therefore evidence for a field-induced transition from a hexagonal VL at low fields to a square VL at higher fields in LSCO.

Twin planes might also affect the symmetry and orientation of the VL at higher fields and must be taken into account in order to establish the significance of the fourfold diffraction pattern observed above the crossover field $B^*$. In YBCO, at intermediate fields ($\mu_0H \lesssim 5$ T), the apparent square alignment of the VL was attributed to the influence of (and weak) higher order spots which lie along the $\{1,0\}$ direction. In Section 3.3.3 we’ll show that the order of the VL can be improved by an appropriate oscillation of the field, which leads to sharper rocking curves and well-defined spots in reciprocal space, see Fig.3.23. In that case the intensity ratio at 1 Tesla is well below 5%. 


3.3 Overdoped regime of La$_{2-x}$Sr$_x$CuO$_4$

Figure 3.9: LSCO (x=0.17): SANS diffraction patterns taken at $T=1.5$ K, after field cooling from 40 K in $\mu_0 H=0.1$ T. In a) the c-axis lies along the field direction. In b) the c-axis has been rotated 10° (about the vertical axis) away from the field direction.

Figure 3.10: LSCO (x=0.17): SANS diffraction patterns obtained for a field $\mu_0 H=0.8$ T. One of the \{1,1\} in-plane axes is aligned at 32 degrees to the horizontal axis in the plane perpendicular to the field. The c-axis of the sample is either a) parallel to the field or b) rotated 30 degrees away from it. The result of the two-dimensional fits of the diffraction patterns are shown in a) by the white square and in b) by the white rhomboid which illustrates the distortion induced by the rotation of the c-axis away from the direction of the applied magnetic field.
anisotropic vortex cores [138], whereas more recent measurements on untwinned YBCO crystals [144] have confirmed earlier suggestions [145, 146, 147] that the results in twinned crystals are actually explained in terms of strong alignment of the hexagonal VL mainly due to twin-plane pinning. Thus in YBCO, for magnetic fields roughly perpendicular to the \(CuO_2\) planes, an intrinsic hexagonal coordination may give rise to a predominantly fourfold diffraction pattern. This conclusion is also inferred in twinned YBCO by rotating the field away from the c-axis so that the influence of the twins is severely reduced, resulting in the recovery of a hexagonal coordination [144]. In YBCO for small tilt angles \(\Theta < \Theta_c \approx 5^\circ\) a fraction of vortices remains pinned to the c-axis, since the vortices bend in order to lie for part of their length within the planar defects [34, 145]. For larger angles this is no longer energetically favourable and the vortices lie along the direction of the applied field [148]. Estimates of this critical angle \(\Theta_c\) for LSCO would not suggest a significantly different behaviour from YBCO, given the slightly increased values for \(\lambda\) and \(\gamma\), although the precise value is difficult to predict [34] and falls with increasing field. Our measurements at low field (see Fig.3.9) indicate that for our LSCO sample \(\Theta_c\) is lower than 10°. Hence, for data taken at high field, we can be sure that \(\Theta = 30^\circ\) exceeds the critical angle. One can see in Fig.3.10 that a four-spot pattern is retained at this angle. It is thus extremely unlikely that this pattern arises in LSCO from pinning distortions due to the presence of twin boundaries, and these data represent the first such observations of an intrinsically square VL in a cuprate HTSC. Recently, an intrinsically square VL has been observed in YBCO at high magnetic fields (\(\mu_0H \gtrsim 10T\)) [139] and in electron-doped NCCO at low magnetic fields (\(\mu_0H \gtrsim 0.05 T\)) (see Appendix). Later on in this chapter we will discuss the similarities and differences of square VL’s obtained in different families of HTSC.

For completeness we note that for \(\Theta = 30^\circ\) there exists a slight distortion of the square pattern which is in accord with expectations for fields tilted away from the c-axis. Due to the approximately uniaxial superconducting anisotropy, the tilting causes an increase of the penetration depth of the vortex currents along one direction, distorting the vortex shape and hence the vortex lattice. In the London approach for anisotropic uniaxial superconductors [107] a rhomboid is expected. The expected value for the ratio \(\epsilon = a/b\) between the sides \((a\ and \ b)\) of the rhomboid is \(\epsilon=0.88\), and for the internal angle is \(\beta = 86.4^\circ\). A two dimensional fit of the neutron data yields \(\epsilon = 0.90(3)\) and \(\beta = 87.3^\circ(1.7^\circ)\), which agree well with the theoretical values.

**Experimental results in LSCO (x=0.17), 2 T< \(\mu_0H\) <9.5 T**

Until now we presented data up to magnetic fields slightly larger than 1 Tesla. Measurements at higher fields have also been performed [95] and are shown in Fig.3.11. As it can be seen, we didn’t observe any further change in the symmetry and/or orientation of the VL: the square VL is oriented along the Cu-O bond direction up to the highest field available (\(\sim 10\) Tesla). Such measurements at high fields are extremely difficult, since the VL intensity rapidly decreases with increasing field. Counting times up to 12 hours were needed for taking SANS diffraction patterns with sufficient statistic.
### 3.3 Overdoped regime of La$_{2-x}$Sr$_x$CuO$_4$

Figure 3.11: LSCO (x=0.17): SANS diffraction patterns from the vortex lattice measured at T=5 K at an applied magnetic field of $\mu_0 H = 2$ T, 5 T and 9.5 T.

![Figure 3.11: SANS diffraction patterns from the vortex lattice measured at T=5 K at an applied magnetic field.](image)

Figure 3.12: LSCO (x=0.17): Field dependence of the VL intensity measured at T=1.5 K: a) linear scale and b) double-logarithmic scale. The fitted lines are explained in the text.

![Figure 3.12: Field dependence of the VL intensity measured at T=1.5 K.](image)

**Field dependence of the vortex lattice intensity in LSCO (x=0.17)**

Since the rocking curves were found to be very broad (see Fig.3.6), we couldn’t measure them as a function of field. Nevertheless, assuming that the rocking curve width remained constant, we measured the intensity at a fixed angle, which then would be proportional to the integrated intensity. The VL intensity is obtained by the integrated intensity of Gaussians fitting the $I$ vs $q$ curves.

In Fig.3.12 the field dependence of the VL intensity for fields smaller than 1.2 Tesla is shown in both linear and logarithmic scales. The data have been fitted within the London approximation (see Eq.(3.9)), including core effects (see Eq.(3.10)), the field dependence of $\lambda$ (see Eq.(3.11)), or both (see Eq.(3.12)). In LSCO $\mu_0 H_c \approx 0.4$ T, whereas $\beta \approx 0.12$ for LSCO (x=0.15) (as determined by $\mu$SR [149]). For the fitted curves we therefore used $\mu_0 H_c = 0.4$ T, $\beta = 0.12$, $\mu_0 H_{c2} = 60$ T [76] while for simplicity $\sigma$ was kept fix to 1. It is clear from Fig.3.12 that the simplest London approximation is not good, whereas including...
core effects and $\lambda(H)$ gives better agreement with our data. However, the best fit is given by a simple power law with exponent -1 (note that the London approximation predicts a power law with exponent $-\frac{1}{2}$).

**Experimental results in LSCO ($x=0.20$)**

Ideally it would be nice to have a tetragonal crystal, where the complication due to the presence of twin boundaries is absent. Indeed the LSCO compound is tetragonal down to low-temperatures for $x \gtrsim 0.22$. However, it is quite difficult to grow sizable single crystals with doping larger than $x=0.20$.

Alternatively, one can try to detwin the crystal by applying an uniaxial pressure. In this case the crystal would still be orthorhombic, but without twin boundaries. We managed to partially detwin a LSCO ($x=0.17$) sample by applying a pressure of about 60 bar along the cylinder axis (starting from a population ratio of $\frac{9}{10}$ at $p=0$ bar we ended with a population ratio of about $\frac{1}{2}$ at $p \approx 60$ bar). However, no effect on the vortex lattice could be observed. This further supports the fact that the square VL oriented along the Cu-O bond direction is an intrinsic feature of the overdoped regime of LSCO.

We present here the results obtained in a $x=0.20$ single crystal [142], where the HTT-LTO structural transition occurs at lower temperatures ($T_0 \sim 70$ K, see Fig.1.10). The orthorhombicity is therefore strongly reduced (compared to the $x=0.17$ sample where $T_0 \sim 130$ K) and the related effect of twin planes should be less relevant. In Fig.3.13 are shown SANS diffraction patterns at different fields. As for the $x=0.17$ sample, at very low fields ($B^* < 0.05$ T) we observed a ring-like intensity, whereas at higher fields ($B^* > 0.4$ T) a square VL oriented along the Cu-O bond direction is seen. By further increasing the magnetic field, the square VL remains oriented along the $\{1,1,0\}$ directions (see Fig.2.2). At 9.5 Tesla some additional intensity is found along the $\{1,0,0\}$ directions (between the four bright spots). Since at high magnetic fields the VL signal is very weak, this additional intensity most probably arises from a bad background subtraction. However, we can not rule out that this intensity is intrinsic and indicates that at higher magnetic fields there is a reorientation of the square VL (rotation by $45^\circ$), as predicted by some theories [150], or a crossover to a more disordered (vortex glass?) phase (see later).

The field-induced hexagonal to square transition has been analyzed following the procedure used for the $x=0.17$ sample. As shown in Fig.3.14 no big difference is found between the $x=0.17$ and $x=0.20$ samples, even though in the latter the transition might occur at slightly lower fields. These results are consistent with the data obtained on the slightly overdoped sample and are therefore intrinsic features of the overdoped regime of LSCO.

**Discussion of the square vortex lattice (theory)**

We now discuss the possible origin of the fourfold coordination of the VL that we observed in the overdoped regime of LSCO for applied magnetic fields larger than $\sim 0.5$ T. Abrikosov predicted the vortex state from a periodic solution of the GL equations close to $H_c2$ [77]. Since the interaction between vortices is repulsive, they will position themselves in a regular arrangement. The equilibrium structure of the VL can be obtained by minimizing the free energy of the system, and the most favourable configuration for an isotropic s-wave superconductor was calculated to be hexagonal [120]. A number of
Figure 3.13: LSCO (x=0.20): SANS diffraction patterns from the vortex lattice in overdoped, measured at T=1.5 K at an applied magnetic field of $\mu_0 H=0.05$ T, 0.4 T, 5T and 9.5 T.

Figure 3.14: LSCO (x=0.20): Quantitative analysis of the field-induced hexagonal to square transition. The full circles indicate the experimental value of $\sigma$ as a function of field, while the empty circles represent the intensity ratio between the {1,0,0} and {1,1,0} directions.
Figure 3.15: a) Current distribution around a vortex in a $d$-wave superconductor: the amplitude of the current $J(r)$ is fourfold symmetric. The breaking of the cylindrical symmetry is stronger in the core region, while toward the outer region the cylindrical symmetry is gradually recovered. b) The associated magnetic field distribution $B(r)$ ($\mathbf{J}(r) = \nabla \times B(r)$), reflecting the current distribution, is also fourfold symmetric. (from Ichioka et al. [124])

experiments confirmed this prediction [151, 114].

In general, the symmetry and orientation of the VL relative to the crystallographic axes is determined by the microscopic electronic properties (e.g. Fermi surface and SC gap symmetry). If an anisotropy is present, the supercurrents surrounding the vortex cores may break the isotropic cylindrical symmetry and influence the symmetry of the VL [124]. Since the difference in energy between the hexagonal and square configurations is very small (about 2% for isotropic systems), the presence of a weak anisotropy is often sufficient to render the square VL energetically more favorable.

Vortex lattices of square coordination have indeed been observed in many superconductors and may occur for a number of reasons. In low-$\kappa$ materials, non-local electrodynamic effects are expected to be significant due to stronger interactions between the cores of neighboring vortices. This may give rise to a VL of square coordination, although at low magnetic fields (large vortex separation) or close to $T_c$ (reduced influence of non-local effects) the VL tends to a hexagonal coordination. In combination with the anisotropy of the Fermi velocity, which reflects the anisotropy of the Fermi surface, this explains the observations of hexagonal to square transitions in borocarbide superconductors [152, 153, 154, 131].

A square VL can also arise in extended Ginzburg-Landau theories that allow for the existence of more than one order parameter, which may account for the robustness of the square VL in the candidate $p$-wave system $\text{Sr}_2\text{RuO}_4$ [155, 156].

The presence of an anisotropic superconducting gap can also give rise to square vortex lattices. Numerous theoretical papers, stimulated by the observations of Keimer et al. in YBCO [138], have tried to develop a model for the VL in a $d$-wave superconductor (such as HTSC) [121, 122, 123, 124, 125, 126, 127, 128, 129]. Studies of an isolated vortex line
3.3 Overdoped regime of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$

in a $d$-wave superconductor within different theoretical frameworks (Ginzburg-Landau or quasiclassical Eilenberger) [121, 122, 124] indicate that the supercurrent and magnetic field distributions around the vortex core have a fourfold symmetry (see Fig.3.15). At low fields, the distance between vortices is too large and the anisotropic vortex cores do not affect the VL. With increasing field the core regions occupy a larger area, and the $d$-wave anisotropy gradually influences the whole structure of the VL [125, 123]. A square VL has been calculated to have a lower free energy at fields larger than $B^* \approx 0.15 \cdot H_{c2}$ (Eilenberger theory) [125] or $B^* \approx H_{c2}/\kappa$ (extended GL theory) [123]. Not only the symmetry, but also the orientation of the VL is affected. The square VL with the nearest neighbor vortices oriented along the gap node directions is found to be energetically more favorable [124, 125, 123, 128]. A square VL with this orientation of the nearest neighbors has indeed been observed in the heavy-fermion $d$-wave superconductor CeCoIn$_5$ [157].

In HTSC, both the Fermi surface and SC gap anisotropies may affect the symmetry and orientation of the VL. If the position of the SC gap nodes and Fermi velocity minima are rotated 45° to each other (as it is the case at least for BSCCO [158] and LSCO [159]), these anisotropies have a competitive effect and try to orient the square VL to a different direction (gap nodes and Cu-O bond direction, respectively; see also Fig.3.16) [150]. While the $d$-wave nature of the order parameter is well established in all HTSC

Figure 3.16: Schematic view of the superconducting gap and Fermi velocity anisotropies in LSCO and corresponding vortex lattice (see text).
Table 3.1: Comparison of the experimental values of the crossover field $B^*$ from a hexagonal to a square VL in various HTSC with the theoretical predictions for $d$-wave SC.

<table>
<thead>
<tr>
<th>Compound</th>
<th>orientation</th>
<th>$B^*$ (exp.)</th>
<th>$B^*$ (Ref.[125])</th>
<th>$B^*$ (Ref.[123])</th>
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<tr>
<td>LSCO</td>
<td>Cu-O bonds</td>
<td>$\sim$0.5 T</td>
<td>$\sim$9 T</td>
<td>$\sim$0.6 T</td>
</tr>
<tr>
<td>YBCO</td>
<td>gap-nodes</td>
<td>$\sim$10 T</td>
<td>$\sim$15 T</td>
<td>$\sim$1 T</td>
</tr>
<tr>
<td>NCCO</td>
<td>gap-nodes</td>
<td>$&lt;0.05$ T</td>
<td>$\sim$1.2 T</td>
<td>$\sim$0.4 T</td>
</tr>
</tbody>
</table>

The orientation of the intrinsic square VL (with nearest neighbors along the nodal direction) recently observed in YBCO [139] at high magnetic fields ($B^* \approx 10$ T), on the other hand, is consistent with $d$-wave theories. To notice is that in YBCO the Fermi velocity is quite isotropic, while the Fermi surface tends to a squarish shape oriented so as to favor the observed square VL orientation [163]. In BSCCO, a hexagonal VL could be observed only at extremely low fields [51]: due to the high anisotropy the VL decouples into pancakes vortices already at 0.08 T, and therefore makes impossible the observation of a field-induced hexagonal to square transition. Recent measurements in the electron-doped NCCO also indicate the presence of a square VL [164] (see Appendix). As in YBCO, the nearest neighbors are along the gap nodes, but $B^*$ is found to be extremely low (<0.05 T).

It remains a challenge to understand on a quantitative way the interplay/competition between the various anisotropies present in the CuO$_2$ planes of different members of the HTSC family. While the different orientations of the square VL’s might be related to changes in the Fermi surface anisotropies, the discrepancy in the values of $B^*$ is more difficult to understand. The prediction of Ref.[125] agrees with the results obtained in YBCO, the one of Ref.[123] is consistent to our experiments in LSCO, while the NCCO data cannot be explained in a quantitative way within existing theories (see also Table 3.1). In order to elucidate the physical origin of the observed square VL, more investigations from both the experimental and theoretical point of views are required. In particular,
detailed ARPES measurements of the anisotropies in the CuO$_2$ planes of HTSC combined with SANS experiments in other HTSC compounds and at different doping levels are needed.

3.3.2 Temperature dependence

The measurements described in the previous section have been all performed at low temperatures. In this section we will present temperature-scans performed at different fields. The motivation for such measurements is given by the prediction [165, 48, 47] and experimental observations [166, 167, 168] of the so-called melting of the vortex lattice in HTSC (see Chapter 1). From SANS experiment one obtains a microscopic measurement of the global long-range order of the vortex lines, and the SANS intensity represents a phenomenological order parameter of the VL.

In Fig.3.17 we show the temperature dependence of the VL intensity for LSCO ($x=0.17$) at $\mu_0H=0.2$ T and 5 T after FC. As a function of temperature, the intensity rapidly decreases and finally vanishes at some temperature $T_m$. In the high field data, the intensity clearly vanishes below the upper critical temperature $T_{c2}$ (as determined by magnetization measurements, see Chapter 2), and $T_m$ is usually interpreted as the melting temperature of the VL [93, 94].

The temperature dependence of the VL signal arises in principle from two causes:

- from the variation of the penetration depth $\lambda(T)$ (since $I \sim \lambda(T)^{-4}$, see Eq.(3.8))
- from the variation of the VL order due to thermal fluctuations, which increases at high temperatures.

$\lambda^{-2}$ is proportional to the superfluid density $n_s$ (density of Cooper pairs) [92] and decreases with increasing temperature, when quasiparticles (QP) are thermally excited. In a conventional s-wave superconductor, this temperature dependence is typically weak at low-$T$ because the isotropic energy gap exponentially cuts off the QP excitations as $T \to 0$ K. In d-wave SC, on the contrary, a linear dependence of the penetration depth down to $T = 0$ K is expected, since the nodes (where the SC energy gap vanishes) allow extreme low-energy QP excitations. Such a behavior has indeed been observed both in the Meissner and vortex state of LSCO and YBCO [67, 134, 149]. However, at higher fields, $\lambda^{-2}$ was found to be roughly constant at low temperatures [134]. While such a behavior could be explained by a decrease in superfluid density $n_s$ or by a field-induced transition to a fully gapped state [169, 170], it was shown later that this could be as well due to nonlinear and nonlocal effects [136]. Such effects are more important at high fields, because of the greater overlap of the regions in the vicinity of the vortex cores.

The shape of the two curves shown in Fig.3.17 is consistent with the results obtained in YBCO by $\mu$SR [134]. At $\mu_0H=0.2$ T (low fields) the VL intensity is roughly linear all over the temperature range, and doesn’t flatten at low temperatures. At 5 T (high fields), on the other hand, the VL signal below about 15 K deviates from the linear behavior. Later in this chapter we will show more detailed temperature scan at intermediate fields ($\mu_0H=2$ T), which indicate that at low temperatures the VL signal (and therefore $\lambda$) is roughly temperature independent (see Fig.3.28).
Figure 3.17: LSCO (x=0.17): Temperature dependence of the VL intensity a) at low fields (0.2 Tesla) and b) at high fields (5 Tesla). The intensity of the VL signal has been normalized to its value at 5 K. Lines are guides to the eye.

Figure 3.18: LSCO (x=0.17): SANS diffraction pattern at 5 Tesla, measured a) at low temperatures and b) just above $T_m$. c) $I$ vs $q$ plots at 5 T, $T=1.8$ K and $T=28$ K.
3.3 Overdoped regime of La$_{2-x}$Sr$_x$CuO$_4$

Figure 3.19: Theoretical calculation of the structure factor of a hexagonal VL a) before and b) after the melting transition, from Ref.[171]

Figure 3.20: Magnetic phase diagram of LSCO (x=0.17). The melting line $H_m$ (filled circles) has been determined by SANS measurements. $H_{FOT}$ (dotted line) and $H_{c2}$ (dashed line) have been obtained by macroscopic measurements, see Chapter 2. The hexagonal ($\Delta$) to square (□) transition line is schematically shown by the thick gray horizontal line.
From the above discussion it is clear that a quantitative analysis of the temperature dependence of the VL intensity requires a good model for $\lambda(T)$. The simple Ginzburg-Landau model gives $\lambda(T) = \lambda(0)(1 - \frac{T}{T_c})^{-\frac{1}{2}}$, but is valid only close to $T_c$, while the two fluid model $\lambda(T) = \lambda(0)(1 - (\frac{T}{T_c})^4)^{-\frac{1}{2}}$ has been sometimes used [137]. However, independently from the model, $\lambda(T)$ always diverges at $T_c$ and cannot therefore explain the observed vanishing intensity of the VL at $T < T_{c2}$. Thermal fluctuations must therefore be responsible for it! Interestingly, the scattered intensity is negligible above $T_m$, as shown in Fig.3.18. Similar results have been observed in BSCCO [51] and YBCO [93]. This result is surprising, since in a liquid of straight vortices a ring of intensity is expected [171] (see Fig.3.21), and might indicate that the vortices are decoupled into a gas of pancake vortices above $T_m$.

At this point it is worth to compare the SANS results to macroscopic measurements (see Chapter 2). In Fig.3.20 the magnetic phase diagram of LSCO ($x=0.17$) is shown. The values of $T_m$ obtained by SANS are in good agreement to the FOT line obtained by magnetization and AC-susceptibility. Such an agreement was also found in YBCO [93]. In Chapter 2 we identified the FOT with the sublimation transition from a VL to a gas of vortices. The absence of intensity above $T_m$ in SANS measurements seems to confirm this interpretation. Within this scenario, the melting is therefore accompanied by a simultaneous decoupling of the vortices into a system of 2D pancakes (the structure factor of a gas of pancakes vortices is probably too small to be measured in SANS experiments).

Finally we would like to address some words about the temperature dependence of the field-induced hexagonal-to-square transition. At low temperatures we found that this smooth transition occurs around $B^* \approx 0.4$ T. In order to study the evolution of $B^*$ with increasing temperature, we can follow the temperature dependence of the symmetry dependent parameter $\sigma$ (see Eq.(3.15)). As shown in Fig.3.21 the value of $\sigma$ is roughly constant both at $\mu_0 H=0.1$ T (hexagonal VL) and $\mu_0 H=0.6$ T (square VL). The field-induced...
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transition to a square VL is therefore roughly temperature independent and occurs at 0.1 T $< B^* < 0.6$ T. This horizontal transition line is included in the magnetic phase diagram shown in Fig.3.20

3.3.3 Non field-cooled measurements

A VL can be grown within the superconducting crystal by various combinations of field and temperature histories as the sample enters the superconducting state from above the upper critical line $H_{c2}$. The data presented in the previous sections have all been obtained after field cooling (FC) through $T_c$ in the chosen magnetic field. In principle, this FC procedure should generate a magnetic-field distribution close to the equilibrium magnetization and VL arrangement. On the other hand, vortex pinning often disorders and introduces mosaicity in the FC VL structure due to competition between Meissner expulsion of flux and pinning as the sample is cooled below $T_c$. As a consequence, broad rocking curves might be observed when measuring after FC.

In order to depin the vortices and end up with a VL state closer to its equilibrium, we used a special oscillating field procedure. Measurements after ZFC have also been performed. Since results obtained on three LSCO ($x=0.17$) samples are consistent to each other, we believe that the following results are intrinsic features and not sample-dependent.

Oscillating field procedure

In an attempt to improve the spatial order of the vortex lines and achieve a VL arrangement closer to equilibrium, we have used an oscillating field procedure (OFP) to anneal the VL. Such a technique has been employed successfully in SANS experiments [172, 173], and is analogous to the shaking of the VL which has been applied in other experiments [174, 175, 176].

The OFP technique replaces the standard FC procedure and consists of the following steps (see also Fig.3.22):

1. Starting from above $T_c$, the sample is field cooled in the desired field $H_{FC}$ down to a temperature $T_{osc}$

2. When the temperature is stable, the field is oscillated (shaked) around its average value $H_{FC}$ in the following way: $H_{FC} \rightarrow H_{FC} - \Delta H \rightarrow H_{FC} + \Delta H \rightarrow H_{FC} - \frac{\Delta H}{2} \rightarrow H_{FC} + \frac{\Delta H}{2} \rightarrow H_{FC}$

3. The temperature is then decreased to a temperature $T_{meas}$ at which the VL is measured

Fig.3.23 shows the result obtained for $\mu_0 H_{FC}=1$ T, $T_{osc}=13$ K and $\Delta(\mu_0 H)=0.2$ T, after integration over rocking curves (see later). It is clear from these Figures that this OFP strongly affects the vortex lattice. The symmetry and orientation of the VL are not changed, but the spots are now much stronger (note the different scale for Fig.3.23a and Fig.3.23b). At 1 Tesla the maximal VL intensity is increased by a factor of 12. This

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7 The rate of the field variation was limited to 0.5 T per minute by the specifications of the magnet. It would be also interesting to investigate the OFP with a faster variation of the field.
enhancement of the vortex lattice signal is due to the sharpening of the rocking curve. While after FC VL the rocking curves are very broad (see Fig.3.6), the OFP produces much sharper rocking curves (see Fig.3.25). This indicates that the VL has now a much better longitudinal correlation (with a correlation length larger then 6400 Å, that is 14 times the distance between vortices at 1 T).

One can also notice in Fig.3.23b and Fig.3.24 that after OFP one observes two strong and two weak spots. The two strong (resp. weak) spots are approximately aligned (resp.
3.3 Overdoped regime of La$_{2-x}$Sr$_x$CuO$_4$

Figure 3.24: LSCO (x=0.17): a) I vs q and b) I vs $\phi$ plot of the data shown in Fig.3.23

Figure 3.25: LSCO (x=0.17): Rocking curves for a) tilt angle $\psi$ and b) rotational angle $\omega$.

perpendicular) to the axis of the cylindrical sample. It is natural to explain this effect in terms of sample geometry. By oscillating the field, one creates an anisotropic field gradient inside the sample, due to the cylindrical shape of the crystal. As a result, the square VL consists of well aligned Bragg planes approx. perpendicular to the axis of the cylinder (contributing to two strong spots) and of less correlated vortices in the direction approx. aligned with the cylindrical axis (giving rise to weaker spots with broader rocking curves).

In order to test this possibility we performed the same experiment on a smaller sample with aspect ratio 1 (a short cylindrical sample with diameter= length $\approx$ 5 mm). Indeed in this sample we obtain four strong spots of similar intensity (see Fig.3.26). It is also worth noticing that weak higher order spots can be seen in Fig.3.26b, therefore excluding the possibility that what is observed is arising from a one-dimensional vortex arrangement.
Figure 3.26: SANS diffraction pattern in a LSCO (x=0.17) sample of aspect ratio 1, measured at $\mu_0 H=1$ T, $T=1.5$ K a) after FC, b) after oscillating the field at $T_{osc}=13$ K with $\Delta(\mu_0 H)=0.2$ T.

within twin boundaries. We will see later that such a vortex system can be observed in underdoped LSCO. The oscillation of the field simply shakes the VL to promote ordering by allowing vortices to move away from randomly distributed pinning sites. The energy given by the oscillating field is used by the vortices to overcome the pinning barriers and to reach a configuration closer to the equilibrium state (in the absence of pinning). Apart from the practical advantage (reduced counting time), such oscillation procedure provides a new, unique and direct method to study the (bulk) pinning strength, which is usually temperature and field dependent [177].

The vortices need an activation energy, in order to overcome the pinning barrier. This energy must be given at the beginning of the oscillating procedure. Indeed we found that the initial decrease of the external field is sufficient to affect the VL. We performed several measurements using this modified oscillating procedure which simply consists in dropping the field from $H_{FC}$ to $H_{FC} - \Delta H$ and then in going back to $H_{FC}$. In Fig.3.27a results obtained at $\mu_0 H_{FC}=1$ T are shown. At $T=12$ K, for $\Delta(\mu_0 H) > 0.15$ T, one observes a strong increase of VL intensity, whereas at base temperature ($T=1.8$ K) this procedure doesn’t work even for $\Delta(\mu_0 H)=0.5$ T. There is therefore a (temperature dependent) threshold value for $\Delta(\mu_0 H)$ which has to be reached in order to have an effect on the vortex lattice. To notice is that after the dropping procedure, the field inside the sample $B \approx 0.95$ T (obtained from the position of the Bragg spots) is smaller than the external magnetic field $\mu_0 H_{FC}=1$ T, indicating some flux expulsion. For a full oscillation of the field, on the other hand, $B \approx H_{FC}$ and the enhancement of the VL intensity is larger. Therefore the vortices are activated already when the field is decreased for the first time, but the full oscillation procedure leads to a "better" VL. Similar measurements at different fields $H_{FC}$ indicate that the threshold value $\Delta H$ increases at higher fields (see Table 3.2), and for $\mu_0 H > 5$ T the dropping/oscillation procedure is completely unsuccessful.

The next step was to investigate the detailed temperature dependence of the pinning strength. As shown in Fig.3.27b, there is a well-defined temperature range $T_1 < T < T_2$
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where the dropping/oscillating procedure works, and both $T_1$ and $T_2$ are field dependent. While at 1 Tesla the VL intensity is not enhanced at low temperatures (<5 K), at larger fields (e.g. at 3 Tesla) the VL intensity is enhanced by the OFP down to the lowest temperatures. To notice is that the ratio between the maximal intensity after OFP and the VL intensity after FC, namely $I_{\text{osc}}^{\text{max}}/I_{\text{FC}}$, is also field-dependent. As shown in Table 3.2 this ratio is maximal at $\mu_0 H=1$ Tesla ($I_{\text{osc}}^{\text{max}}/I_{\text{FC}}=12$) and decreases at higher fields. These different temperature regimes result from the interplay/competition between the vortex elastic energy $E_{el}$, the pinning energy $E_{\text{pin}}$ and the thermal energy $E_{th}$. As a result, while the density of pinning centers (and the related absolute pinning profile) is independent of temperature and field, the vortices might experience a different effective pinning profile as a function of increasing field, since the density of vortices is larger, or temperature, when thermal fluctuations become important.

Table 3.2: Results from the dropping field experiments on LSCO ($x=0.17$), see text

<table>
<thead>
<tr>
<th>$H_{\text{FC}}$ [T]</th>
<th>$B$ [T]</th>
<th>$\Delta H_{\text{thresh}}$ [T]</th>
<th>$T_1$ [K]</th>
<th>$T_2$ [K]</th>
<th>$I_{\text{osc}}^{\text{max}}/I_{\text{FC}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td>0.45</td>
<td>$\sim 0.15$</td>
<td>$\sim 10$</td>
<td>$\sim 25$</td>
<td>$\sim 3$</td>
</tr>
<tr>
<td>1</td>
<td>0.95</td>
<td>$\sim 0.15$</td>
<td>$\sim 5$</td>
<td>$\sim 20$</td>
<td>$\sim 12$</td>
</tr>
<tr>
<td>2</td>
<td>1.67</td>
<td>$\sim 0.2$</td>
<td>$\sim 2$</td>
<td>$\sim 20$</td>
<td>$\sim 9$</td>
</tr>
<tr>
<td>3</td>
<td>2.49</td>
<td>$\sim 0.6$</td>
<td>$\sim 0$</td>
<td>$\sim 15$</td>
<td>$\sim 7$</td>
</tr>
<tr>
<td>4</td>
<td>3.44</td>
<td>$\sim 0.8$</td>
<td>$\sim 0$</td>
<td>$\sim 10$</td>
<td>$\sim 4$</td>
</tr>
<tr>
<td>5</td>
<td>-</td>
<td>$&gt;1$</td>
<td>-</td>
<td>-</td>
<td>$\sim 1$</td>
</tr>
</tbody>
</table>

Furthermore we measured the temperature dependence of the annealed VL (after OFP),
and compared it to the results obtained after FC. As shown in Fig.3.28 for $\mu_0 H=2$ T, the intensity is quite constant at low temperatures ($T < 10$ K), and then decreases roughly linearly with increasing temperature. Above $20$ K most of the intensity has disappeared, but a tail can be observed up to about $30$ K. To notice is that $T=10$ K is the temperature where we had the maximum gain in intensity after oscillating the field, $T=20$ K is the temperature where we observed the second peak in magnetization measurements, and $30$ K is close to the irreversibility line. Without oscillating the field, on the other hand, we observe a weak signal which vanishes around $30$ K, and which could be responsible for the tail observed in the OFP data. These results indicate that the OFP gives rise to an additional intensity which vanishes at the second peak line, whereas the FC VL intensity disappears at the FOT line (see also previous section). A more detailed comparison between SANS and macroscopic results will be given later in Section 3.3.4.

Finally we used the OFP in order to better understand the field-induced hexagonal to square transition. In Fig.3.29 are shown 2D pattern obtained at $T=10$ K after oscillating the field at $T_{osc}=15$ K ($\Delta(\mu_0 H)=0.1$ T), for fields between $0.15$ T and $0.5$ T. The OFP enhances two diagonal spots along the $\{1,1,0\}$ direction, but also indicate a mainly hexagonal (square) symmetry at low fields (high fields). Moreover, the hexagonal pattern oriented along the two bright spots seems to be favored. At intermediate fields, on can understand the complex SANS pattern as a superposition of hexagonal and square domains, as shown schematically in Fig.3.29f. The $I$ vs $q$ curves can be therefore fitted by two Gaussians, as shown in Fig.3.30. The centers of the two Gaussian is fixed to the values expected for the square and hexagonal VL, whereas their widths are forced to be the same. The results of such an analysis confirms the smooth field-induced hexagonal-to-square transition presented in Fig.3.14. At low fields the VL is completely hexagonal, while as a function of increasing field the contribution from a square VL becomes more and more important. Finally, above $B^*$ the VL is totally square. At intermediate fields the VL consists of a superposition of both hexagonal and square VL’s. This can also
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Figure 3.29: LSCO ($x=0.17$): 2D pattern after OFP. At intermediate fields the SANS pattern consists of the superposition of square and hexagonal VL’s.

explain that at intermediate fields, the value of $\sigma$ was found to lie in between the values expected for square and hexagonal VL’s (see Fig.3.8b).

The coexistence of hexagonal and square domains might indicate that the transition is of first order, as in the borocarbides [173] and in CeCoIn$_5$ [157]. However, the field-range of coexistence is quite large. This could be due to weak disordering (pinning) of the VL [173], whereas strong disorder can transform the first order into a second order phase transition [178]. To notice is that in YBCO the hexagonal VL is smoothly distorted to a square VL with increasing field [139], therefore suggesting a continuous (second order) phase transition.

Zero-field cooled measurements

We also performed ZFC measurements, and more precisely field-scans at different temperatures, which are the analogue of the ZFC magnetization data presented in Chapter 2. In Fig.3.31 we show the results obtained at $T=10$ K. Similarly to the OFP, we observe a strong increase of the VL intensity and sharpening of rocking curves. To notice is that, after ZFC, the internal field $B$ inside the sample (as determined by the $q$-value of the spots of the square VL) is lower than the external magnetic field. From SANS data it is clear that at low magnetic fields the vortices cannot penetrate the sample (surface and geometrical barriers). As soon as a threshold field has been reached, the VL intensity increases very fast, whereas at higher fields the VL intensity decreases. Finally, after removing the external field, trapped flux inside the sample can still be observed. Interestingly, the observed SANS pattern still displays a square VL. To note is however that
Figure 3.30: LSCO (x=0.17): Analysis of the field-induced hexagonal-to-square transition after OFP. For clarity the intensities at different fields have been rescaled and have a vertical offset. Horizontal segments indicate the instrumental resolution. $I$ vs $q$ curves have been fitted by two Gaussians with the position fixed to the $q$-value expected for square and hexagonal VL’s respectively, while the widths of the two Gaussians was forced to be the same. In a) the total fit is shown, whereas in b) the two Gaussians are plotted separately: the dotted (solid) curve indicate the contribution from a square (hexagonal) VL. The square contribution becomes more and more important with increasing field. In c) the population rate of the square VL $I_{\text{square}}/(I_{\text{square}} + I_{\text{hex}})$ is plotted as a function of field.
the mean value of the trapped internal field is still larger than $B^*$, and the observation of a square VL is therefore not too surprising.

A direct comparison to macroscopic measurements can be done: the VL signal appears at the penetration field, and a peak is observed at the onset field. This observation is confirmed by measurements at $T=5$ K and 15 K (not shown), since the peak in the VL intensity shifts to higher magnetic field with decreasing temperature, as expected. These results indicate that around the onset field one can increase the VL intensity. During the OFP, the field is decreased close to the onset region, where the vortices can move and exit from the pinning centers. As a consequence the correlation length of the VL is increased, the rocking curves sharpen and the VL intensity is enhanced. The onset field region can be therefore seen as a regime of weak pinning. This interpretation could also explain the fact that at high fields we need larger amplitude of the field-oscillation in order to see an effect (see Table 3.2). This is due to the fact that the onset field is far away from the applied field $H_{FC}$, and in order to go close to $H_{on}$ one has to increase the wiggling amplitude, as shown schematically in Fig.3.32.

### 3.3.4 Comparison to macroscopic measurements

In Chapter 2 we presented macroscopic measurements ($M$, $\chi$), which represent the total response of the sample to an external magnetic field. Such measurements do not give direct information about the behavior of the vortices, but are extremely helpful to have a general idea of the magnetic phase diagram and to identify several transition/crossover lines. On the other hand, the SANS experiments described in this section, are sensitive to the long range order of the VL.

Macroscopic and SANS measurements are therefore complementary, and in order to understand the magnetic phase diagram, one has to compare their results. The question is: how do the vortices behave during a macroscopic measurement (e.g. hysteresis loop)? One can model and speculate, but SANS is able to directly answer this question!

Here I summarize the main combined results of Chapter 2 and 3 for overdoped LSCO ($x=0.17$):

#### Penetration field

At low magnetic fields, the ZFC $M(H)$ curve decreases linearly and in ZFC SANS experiments no VL signal is observed. This indicates that the sample is in the Meissner phase, which is actually extended to fields larger than $H_{c1}$ due to surface and geometrical barriers. The magnetic flux can therefore enter the sample only at a penetration field $H_p > H_{c1}$.

#### Onset field

By further increasing the magnetic field during ZFC field scan, $M(H)$ displays a maximum (onset field) followed by a minimum (second peak). Around the onset field, the critical current is lower, therefore indicating that the vortices can easily escape from the pinning barriers. This is indeed seen in ZFC SANS experiments. Moreover the OFP works only
Figure 3.31: LSCO (x=0.17): a) ZFC field scan at \( T=10 \) K together with magnetisation curves. The VL signal appears at the penetration field, whereas the peak in SANS data is coincident to the onset of the second peak. The point at zero field has been measured after measuring at 2 Tesla, and is indicative of the trapped flux inside the sample (consistent to magnetization data). b) \( I \) vs \( q \) plots at selected magnetic fields.

Figure 3.32: LSCO (x=0.17): Magnetization data at \( T=5 \) K and \( T=10 \) K. The length of the arrows indicate the amplitude of the field oscillation, whereas their thickness indicate the effectiveness of the OFP (dotted: no effect, solid line: small effect, thick line: strong effect).
when the oscillation is able to reach the onset field region. We can therefore describe the onset field as a weak-pinning region.

Second peak
Around the second peak something happens, the density of vortices is larger, and they experience a different (stronger) pinning profile. The critical current is maximal, and the VL gets more disordered. This can be seen in SANS experiments, where the intensity gained after the OFP is lost at the second peak temperature (see Fig.3.28). Above the second peak we have therefore a sort of vortex glass, with a small correlation length of the VL. However, a weak square VL can be observed above the second peak line up to the FOT line, therefore it is not a depinning line.

Sublimation
In Chapter 2 we interpreted the FOT as being the concomitant melting and decoupling of the VL into a gas of pancakes vortices (therefore sublimation). This was inferred from the doping dependence of the FOT line. SANS experiments are consistent with this interpretation, since the VL intensity completely vanishes at the FOT line.

Magnetic Phase Diagram
Finally, in Fig.3.33, a schematic magnetic phase diagram is plotted within the above interpretation. As in conventional Typ-II SC we have the Meissner state, the mixed state and the normal state. However, the mixed state is further split into three main phases. The well-ordered VL at low temperatures/fields undergoes a transition to a more disordered vortex glass, followed by a sublimation to a vortex gas. Finally, above $T_c$, strong (vortex-like) SC fluctuations survive.
3.4 Underdoped regime of La$_{2-x}$Sr$_x$CuO$_4$

As already discussed in the introduction (see Section 1.4.1), the underdoped regime of LSCO represents a unique system from the vortex matter point of view, because of the combination of intermediate values of the anisotropy $\gamma$ between YBCO (and overdoped LSCO) and BSCCO, and long penetration depth $\lambda$. The significance of these parameters can be understood by considering the Josephson length $\lambda_J = \gamma \cdot s$, where $s$ is the spacing of the copper oxide planes. While higher values of $\lambda_{ab}$ and $\gamma$ increase the susceptibility of the vortices to disorder, the ratio $\lambda_J/\lambda_{ab}$ determines the effectiveness of the Josephson currents, that tunnel between the conduction planes, to maintain the stiffness of a vortex line. In BSCCO $\lambda_J/\lambda_{ab} \gtrsim 1$, and consequently the vortex lines are very flexible, while in YBCO $\lambda_J/\lambda_{ab} \ll 1$, and the vortices resemble the rigid rods of flux found in conventional isotropic superconductors. In contrast, the material parameters of underdoped LSCO ($\lambda_J \approx 250 \, \text{Å}$) give rise to a system of fairly rigid vortex lines which are nonetheless highly susceptible to transverse fluctuations. This system is therefore a good candidate for investigating experimentally the transition (with increasing field) from a vortex lattice to a vortex glass, due to the changing scale of disorder as the separation of the vortices decreases.

SANS investigations in underdoped LSCO are complicated by the large penetration depth. Since the intensity of the Bragg reflection is proportional to $\lambda^{-4}$, see Eq.(3.3), the VL signal is extremely weak. Moreover, due to the large anisotropy, we expected that an ordered vortex lattice is present only at very low fields. This requires the use of neutrons with long wavelength, and therefore weaker neutron flux. Despite all these experimental difficulties, we report here on the first direct observation of a vortex lattice in a crystal of underdoped LSCO ($x=0.10$).

3.4.1 Experimental results

We performed two experiments on the same underdoped LSCO sample ($T_c=29$ K) on D22 at ILL, the first one in 2003 and the second one in 2004. Low-field SANS diffraction pattern are shown in Fig.3.34: during the first experiment we observed a weak ring-like pattern whereas in the second experiment only two strong spots could be observed. We will discuss the possible reason for this surprising difference between the two measurements later in this section. For the moment we just remark that the intensity shown in Fig.3.34a (counting time of about 10 hours) is about 10 times weaker than the one in Fig.3.34b (counting time of about 1 hour). To notice is also that the two strong spots (Fig.3.34b) have extremely sharp rocking curves (see Fig.3.35) whereas rocking curves of the ring-like intensity (Fig.3.34a) were found to be very broad (not shown).

In Fig.3.36a an $I$ vs $q$ plot of the two strong spots measured at $\mu_0 H=1000 \, \text{Oe}$ and $T=5$ K is shown together with fitted Lorentzian and Gaussian curves. The temperature dependence of the two spots measured at $\mu_0 H=800 \, \text{Oe}$ (see Fig.3.36b) clearly indicates

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*Unfortunately a exact comparison of the intensities is not possible, because a new detector with different efficiency was used during the second experiment.*

---

8LSCO $(x=0.10)$: $\gamma \approx 45$, $\lambda_{ab} \approx 3000 \, \text{Å}$ (see Table 2.2); LSCO $(x=0.2)$: $\gamma \approx 20$, $\lambda_{ab} \approx 2000 \, \text{Å}$ (see Table 2.2); YBCO (opt. doped): $\gamma \approx 4$ [179], $\lambda_{ab} \approx 1300 \, \text{Å}$ [133]; BSCCO: $\gamma \gtrsim 150$ [180], $\lambda_{ab} \approx 1400$-$2000 \, \text{Å}$ [34]
3.4 Underdoped regime of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$

Figure 3.34: Low-field ($\mu_0H=13\text{ mT}$ and $\mu_0H=30\text{ mT}$) SANS diffraction pattern taken at $T\approx5\text{ K}$ after subtraction of a background taken at $T=35\text{ K}$. a) During the first experiment (in 2003), we observed a ring-like pattern. b) In a second experiment (in 2004) we observed two strong spots, see text. The integrated intensity in a) is about 10 times lower than the one in b) (the color scales in a) and b) are arbitrarily different).

![Image of diffraction patterns](image)

Figure 3.35: Rocking curves of the two strong spots measured in 2004: a) tilt angle $\psi$ and b) rotational angle $\omega$. To notice is the sharp rocking curve in $\psi$.

![Image of rocking curves](image)

Figure 3.36: a) $I$ vs $q$ curves measured in 2004 at $\mu_0H=100\text{ mT}$, $T=5\text{ K}$, fitted by a Gaussian and a Lorentzian. b) Temperature dependence of the signal measured at $\mu_0H=80\text{ mT}$.

![Image of I vs q curves and Temperature dependence](image)
Figure 3.37: Field dependence of the peak position \( q_{\text{peak}} \) as determined by Gaussian fits of \( I \) vs \( q \) curves: a) ring-like pattern (2003) and b) two spots (2004). The lines indicate the expected behavior for hexagonal (dashed line) and square (solid line) vortex lattices.

Figure 3.38: Field dependence of the integrated intensity: a) ring-like pattern (2003) and b) two spots (2004). The fitted curves are described in the text.

Figure 3.39: Field dependence of the radial width for the data measured in 2004. In a) the raw width is shown together with the instrumental resolution. In b) the corrected intrinsic width is shown. The line is a guide to the eye.
3.4 Underdoped regime of La$_{2-x}$Sr$_x$CuO$_4$

that the signal is real and appears in the superconducting state. From $I$ vs $q$ curves we can obtain, as usual, the peak position $q_{\text{peak}}$ and the integrated VL intensity. The field-dependence of $q_{\text{peak}}$ is shown in Fig.3.37: $q_{\text{peak}}$ increases with field, as expected due to the increased vortex density inside the sample, for both the ring-like pattern measured in 2003 and the two strong spots measured in 2004. In Fig.3.38 the integrated intensity is plotted as a function of field: the intensity of both the ring-like pattern and the two strong spots decreases with increasing field faster than in the London model. While the field dependence of the ring-like intensity can be roughly explained by Eq.(3.12), the decrease of the intensity of the two strong spots is much faster and is well fitted by a power law with exponent -2. Interestingly the loss of intensity is accompanied by an increase in the radial width, as shown in Fig.3.39. Both results indicate that disorder increases with increasing field.

Finally we discuss the apparent discrepancy between the results obtained in 2003 and 2004. Most probably the signal that has been measured during the two experiments arises from different parts of the crystal. While the ring-like intensity arises from the bulk of the crystal, the two strong spots possibly result from the vortices lying in the twin boundaries. These are capable of pinning the VL because they represent planar regions with a suppressed order parameter [181, 145]. Indeed the two strong spots are aligned to the direction of the twin boundaries approximately aligned to the axis of the cylindrical crystal (see Fig.3.34b). Due to the sharp rocking curve, expected for vortices lying in well-defined planar defects, it is possible that during the first experiment we missed this strong signal, due to a slight misalignment of the sample and measured the weak signal from vortices outside the twin boundaries. Such an arrangement of vortices pinned by correlated disorder is also of high interest, and has been described in terms of Bose glass from the theoretical point of view [182].

It remains to understand why only two spots are seen: twin boundaries are present also in the direction perpendicular to the one shown in Fig.3.34b and one would therefore expect four spots. The cylindrical shape of the crystal might be the reason for it, as it was the case in overdoped LSCO after the oscillation of the field. However, measurements in underdoped LSCO have been performed after FC, and it is therefore more complicated to identify the mechanism leading to well aligned vortices in the twin-planes approximately perpendicular to the cylinder axis, and badly aligned vortices in the orthogonal family of twin planes. Experiment in samples with aspect ratio 1 should be performed in order to clarify this aspect.

3.4.2 $\mu$SR experiments

The SANS results described in the previous section are insufficient to completely understand the vortex matter in underdoped LSCO. In particular the vortex phase cannot be investigated at large magnetic fields, because of the weak SANS signal. In order to clarify the situation, we performed also muon spin rotation ($\mu$SR) experiments on the same crystal using the GPS spectrometer at the Paul Scherrer Institute [45]. $\mu$SR is complementary to SANS because it allows the study of the local order of the vortices, and is therefore

\textsuperscript{10}The name Bose glass is used because the physics of vortices localized by correlated pinning is analogous to the problem of localization of quantum mechanical bosons in two dimensions [182, 183].

Figure 3.40: a) Temperature-dependent probability distributions of the internal field, $p(B)$, due to the presence of the vortices at applied fields of a) 80 Oe and b) 6 kOe. The curves are normalized to allow easy comparison of the shapes.

able to image the vortex system even in the more disordered vortex phase at high fields.

Polarized muons (with their spin perpendicular to the applied field) were randomly implanted in the sample. Each muon precesses at a rate proportional to the local field $B(r)$. The muon spin precession is monitored via the positrons which are emitted preferentially along the muon spin direction during the decay of the muon. The precession signal, averaged over $\sim 10^7$ muons, reveals the probability distribution $p(B)$ of the spatial variation $B(r)$.

In a $\mu$SR experiment the presence of a vortex lattice is revealed by a $p(B)$ which is characterized by a peak at $B_{pk}$ followed by a tail extending towards fields higher than the mean field, reflecting the small volume of the core region of the vortices. In Fig.3.40a are shown probability distributions measured in underdoped LSCO at low field, and those taken at the lowest temperatures correspond most closely to the lineshape expected from an ideal VL [67]. Since the muon probe measures the local field, which is determined by contributions from vortices only within $\lambda_{ab}$ of the muon, the general form of $p(B)$ is remarkably robust to changes in long range order. For this reason, $\mu$SR is fairly insensitive to the subtle difference between a VL, possessing modest local positional fluctuations while maintaining long range correlations, and a Bragg glass (BG). In contrast, the technique is highly sensitive to changes in the local environment due to thermally-induced or pinning-induced distortions, as has been demonstrated in a number of systems [184, 185]. It is this property which allows us to observe the transition from the ordered state to the more disordered vortex glass phase in underdoped LSCO.

The main results can be appreciated immediately by comparing the normalised experimental lineshapes of Fig.3.40a and Fig.3.40b, measured at 80 Oe and 6 kOe respectively. Referring firstly to the lowest temperature lineshape at each field, it is clear that they are
Figure 3.41: The variation with field of the line width at selected temperatures. The initial reduction with increasing field corresponds to a motional narrowing due to the field dependent influence of thermal fluctuations on the field distribution. Above about 1 kOe, the width dramatically rises due to the onset of static disorder. Note that the high-field dependence has almost disappeared by a temperature of 23 K, and disappears completely at the irreversibility line.

very different. While the 80 Oe distribution has the characteristics of an ideal vortex lattice described above, the 6 kOe signal is highly symmetric, indicating a strong departure from an ideal vortex or Bragg glass arrangement. Monte Carlo simulations indicate that the vortex system at large fields is disordered having short-ranged translational correlations of order of a few inter-vortex spacings. In these simulations, the loss of long range order is due to a transition from a Bragg glass phase to a multi-domain glass comprising a size distribution of domains within which the vortices are locally ordered [186]. Increasing the field leads to a rapid fall in the average domain size just above the Bragg glass phase. Further conclusive evidence for the transition from a BG to a VG phase, which is independent of any particular model, comes from considerations of the width of the lineshapes $\Gamma = \sqrt{\langle (\Delta B)^2 \rangle}$, given by the square root of the second moment of $p(B)$ \footnote{$\langle (\Delta B)^2 \rangle = \langle (B - \langle B \rangle)^2 \rangle = \langle B^2 \rangle - \langle B \rangle^2 \propto \frac{\Phi_0}{A}$ where $\langle B \rangle$ is the average magnetic field.}. This quantity is plotted in Fig.3.41 as a function of applied field, for several temperatures. At low temperatures, the signal measured at 6 kOe is considerably broader than that at 80 Oe. Such a broadening of the signal from the vortex lattice can only arise from static disorder in the positions of vortex lines within a plane perpendicular to the field [187, 188], because local density variations, due to positional disorder of the vortices, give rise to regions with field values both higher and lower than in the well-ordered lattice [187, 188]. Conversely, in a system composed of two-dimensional pancakes, transverse fluctuations having short
wavelength along the field direction would always lead to a narrowing of the field distribution \( p(B) \) [187]. This is indeed the situation found experimentally in the very anisotropic BSCCO [184]. Thus it is clear that in our case we are dealing with a highly disordered vortex line arrangement, which we identify with the vortex glass phase. We now turn our attention to the temperature dependence of the lineshapes of Fig.3.40a, where it can be seen that with increasing temperature there is a truncation of the high-field tails. This reflects the increasing amplitude of thermally-induced fluctuations \( \sqrt{\langle u^2 \rangle} \) of the vortex positions on a timescale faster than the characteristic muon sampling time [189]. The muons thus experience a time-averaged field distribution in which the high-field values arising from close to the vortex cores are smeared out. At a given temperature this will also lead to increased narrowing as the field is increased, because of the increasing ratio \( \sqrt{\langle u^2 \rangle}/d \), \( d \) being the distance between vortices. This dynamical reduction in width with increasing field can be seen up to fields of around 1 kOe, and occurs at all temperatures. Thus the minimum in \( \Gamma \) at each temperature should be viewed as arising from a competition between motional narrowing and broadening due to the formation of the vortex glass phase. It should be pointed out that narrowing of the lineshapes could also arise in a quasi-two-dimensional pancake-vortex system, from static or dynamical transverse fluctuations [187], as was demonstrated experimentally for BSCCO [184]. However, this would be extremely unlikely given the increased broadening at high fields, which may only be explained by increasing in-plane disorder of vortex lines \(^{12}\).

### 3.4.3 Vortex glass transition

These \( \mu \)SR results can be now compared to our SANS results. The rapid loss of intensity in SANS experiment together with the broadening of the radial width as a function of increasing field may result from the increasing influence of strong disorder combined with the onset of the loss of transverse long range order over this field range. That is, the reduction in transverse correlation length which broadens the \( \mu \)SR linewidth at higher field will also reduce the SANS intensity.

In the literature, evidence for the existence of the BG-VG transition is frequently taken from signatures in the bulk magnetisation, since the transition to the glassy state is accompanied by an increased effectiveness of point pinning above the onset field of the second peak \( H_{on} \) [103]. While our macroscopic measurements in underdoped LSCO didn’t reveal the presence of \( H_{on} \), more detailed SQUID measurements on a piece of the sample used in \( \mu \)SR experiments clearly indicate that an onset field can be identified even in LSCO (\( x=0.10 \)) [45]. The temperature dependence of \( H_{on}(T) \) is plotted in the magnetic phase diagram of Fig.3.42. The occurrence of a field-induced disorder transition has been discussed theoretically by many authors including Giamarchi and Le Doussal [190]. The Bragg glass is expected to be unstable to the proliferation of topological defects (dislocations) at large fields. These two phases, one without topological defects in equilibrium

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\(^{12}\)The onset of magnetic order can also lead to a broadening of the lineshapes. This effect is indeed observed for this sample below 3 K, at which temperature both the longitudinal and transverse \( \mu \)SR signals indicate the onset of magnetic order (see later in Chapter 4). However, the highly broaden transverse lineshapes observed below 3K in the magnetically ordered regime retain the general characteristics of an ordered vortex line arrangement at low field, and indicate the transition to a disordered phase at high field.
Figure 3.42: LSCO ($x=0.10$): the magnetic phase diagram is derived from the changes observed in the $\mu$SR field distribution. $B_{cr}(T)$ indicates the onset of the broadening at high fields, and should be considered as an upper limit of the BG-VG phase transition. This uncertainty in the exact position of the transition is represented by the shading below the line $B_{cr}$. The irreversibility line $B_{irr}$ and the onset field $B_{on}$ as determined by SQUID magnetisation measurements are also plotted.

and the other in which there are defects in equilibrium (VG), cannot be smoothly connected, thus implying the existence of an equilibrium phase transition between them. It is worth noting in this context that the muon is sensitive only to the local magnetic field, so in general this transition will be manifested as a crossover of behaviour which reflects the underlying sharp transition. This feature should therefore be viewed as reflecting the existence of the vortex glass phase rather than being directly identified with the BG-VG transition, and indeed it provides an upper limit for the latter. Nonetheless at high fields the broadened signal clearly indicates that the vortex lines are in a highly disordered state. The signature in the magnetisation also reflects the underlying disorder transition, but measures very different properties under different conditions to the $\mu$SR experiment. The muons measure the field probability distribution in a field-cooled state and the signature reflects two competing field-dependent processes. The magnetisation measurements determine the macroscopic properties of a disturbed system possessing strong macroscopic flux gradients, and reflect the changing dynamic response to the pinning landscape. The SANS measurements are sensitive both to thermal fluctuations and to the loss of long range coherence in the vortex structure, but in practice are unlikely to provide information deep into the vortex glass phase. Further comparison between data derived from all of these techniques with detailed Monte Carlo simulations should further resolve some of
these complex issues. The existence of an ordered BG was recently suggested by SANS measurements in the isotropic superconductor (K,Ba)BiO₃ [191, 44]. In those experiments the characteristic length scale for the decay of vortex transverse correlations was found to give a reduction in neutron intensity with increasing field due to finite instrumental resolution, having the appearance of a crossover. Moreover also in electron-doped NCCO we have possibly evidence of a transition to a disordered phase from SANS [164], see Appendix. The present case in LSCO (x=0.10) is distinguished, however, by the fact that it provides unambiguous evidence for a crossover with increasing field from a nominally BG phase to a more disordered VG state.

3.5 Conclusions

In conclusion, we have used SANS to make the first clear observations of the vortex lattice in all doping regimes of La₂₋ₓSrₓCuO₄.

In the overdoped regime:

- We discovered a field-induced transition from a hexagonal to a square vortex lattice. Intrinsically square VL had never been observed before in HTSC, and are indicative of the coupling of the VL to some anisotropy in the CuO₂ planes. Subsequent to our discovery, square VL’s have been observed in other members of the HTSC family, namely in YBa₂Cu₄O₆₊ₓ and Nd₂₋ₓCeₓCuO₄ (by ourselves, see Appendix).

- We were able to directly compare results from SANS and magnetization measurements. Our results strongly support the existence of a sublimation transition (concomitant melting and decoupling) of the vortex lattice at temperatures well below Tc.

- Moreover we were able to investigate the different pinning regimes by means of SANS. Such experiments turned out to be helpful for a better understanding of the complex magnetic phase diagram of LSCO and, more generally, of HTSC.

In the underdoped regime:

- SANS experiments have been performed successfully at low magnetic fields, and the VL intensity was found to rapidly decrease with increasing field. The effect of twin boundaries on the VL could also be investigated.

- Complementary μSR experiments of the the vortex state clearly indicate a change with increasing field from a vortex lattice (or Bragg glass) to a more disordered vortex glass phase. This new result is consistent to SANS and magnetization measurements.
Chapter 4

Inelastic neutron scattering study of the spin excitations

We present a low-energy inelastic neutron scattering study of the spin dynamics in the mixed phase of LSCO. In the overdoped regime ($x=0.17$) the application of a magnetic field induces a spectral weight redistribution of the low-temperature susceptibility centered at the zero-field spin gap energy ($\Delta_{SG} \approx 6.5$ meV). Moreover, the opening of the spin gap seems to be related to the temperature at which the vortex lattice sublimates rather than to the upper critical temperature $T_c^2$, indicating an unusual interplay between the mesoscopic and microscopic properties in LSCO. In the underdoped regime ($x=0.10$) our zero-field data indicate an anomalous increase of the imaginary part of the susceptibility $\chi''$ at low energies ($\hbar \omega < 2$ meV) and low temperatures ($T < 15$ K), which is possibly related to the spin glass phase observed in $\mu$SR experiments. These results, together with the magnetic field dependence of $\chi''$, are consistent with a strongly reduced spin gap ($\Delta_{SG} \approx 2.5$ meV) which vanishes in the vortex gas phase. Furthermore, we have investigated the elastic incommensurate magnetic peaks in underdoped samples ($x=0.10$ and $x=0.12$) close to $x=1/8$. In both samples we observe a field-induced signal in the superconducting phase, which is discussed in relation to the spin glass phase and to the possibility that static magnetism resides in the vortex cores.

4.1 Introduction

4.1.1 Neutron scattering

Compared to other experimental techniques, neutron scattering is a valuable and sometimes unique tool for the investigation of condensed matter. Due to their charge neutrality, the interaction of neutrons with matter is limited to short-ranged nuclear and magnetic interactions. The weakness of the interaction has the advantage that neutrons easily penetrate the samples, therefore probing their bulk properties. On the other hand, this requires a large amount of sample (at least several grams for inelastic neutron scattering). Since the wavelength of the neutrons can be chosen to be comparable to the mean

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1 B.N. Brockhouse and C.G. Shull won the Nobel Prize in Physics in 1994 "for pioneering contributions to the development of neutron scattering techniques for studies of condensed matter"
interatomic separation in solids (some Å), neutrons are ideal for the study of atomic structures (elastic neutron scattering, diffraction experiments). Moreover, their kinetic energy is comparable to that of elementary excitations in solids and liquids (phonons, magnons, etc.), thus allowing the study of their dynamic properties (inelastic neutron scattering).

**Cross section**

The basic expression for the partial differential scattering cross, representing the number of neutrons scattered into a small solid angle dΩ with final energy between \( h\omega \) and \( h\omega + dh\omega \), is given by [66]:

\[
\frac{d^2\sigma}{d\Omega d\omega} = \left( \frac{m}{2\pi \hbar^2} \right)^2 \frac{k'}{k} \sum_{\lambda} p_\lambda \sum_{\lambda'} |\langle k', \lambda' | \hat{U} | k, \lambda \rangle|^2 \delta(h\omega + E_\lambda - E_{\lambda'})
\]  

(4.1)

\( m \) is the mass of the neutron, \( k \) and \( k' \) are the initial and final wavevectors, |\( \lambda \)\rangle and |\( \lambda' \)\rangle denote the initial and final states of the sample with energy \( E_\lambda \) and \( E_{\lambda'} \), respectively. The averaging over the initial states is done on the basis of |\( \lambda \)\rangle being occupied with the probability \( p_\lambda \), whereas the summation over the final states is done by summing over the index |\( \lambda' \)\rangle. \( \hat{U} \) designates the interaction operator between sample and neutrons.

**Nuclear scattering**

In the case of nuclear scattering, \( \hat{U} \) can be described by the Fermi pseudopotential:

\[
\hat{U} = \frac{2\pi \hbar^2}{m} \sum_j b_j \delta(\mathbf{r} - \mathbf{r}_j)
\]  

(4.2)

where \( b_j \) is the scattering length of the atom \( j \), while \( \mathbf{r} \) and \( \mathbf{r}_j \) are the position of the neutron and of the nucleus \( j \), respectively. The scattering length corresponds to the amplitude of the wavefunction of the scattered neutron, and its magnitude and sign is different for each atom (and isotope). Inserting plane wave functions for the neutrons, |\( k \rangle = \exp(ik \cdot r) \), one obtains the following cross section [66]:

\[
\frac{d^2\sigma}{d\Omega d\omega} = \frac{k'}{k} \sum_{\lambda} p_\lambda \sum_{\lambda'} |\langle \lambda' | \hat{U} | \lambda \rangle|^2 \delta(h\omega + E_\lambda - E_{\lambda'})
\]  

(4.3)

\( \mathbf{Q} = \mathbf{k} - \mathbf{k}' \) is called the scattering vector. The evaluation of such a cross section is usually complicated. For example, assuming harmonic interatomic forces in the crystal, the coherent one-phonon cross section for a Bravais crystal is given by:

\[
\frac{d^2\sigma}{d\Omega d\omega} = \frac{\sigma_{coh} k'}{k} \frac{(2\pi)^3}{K^3} \frac{1}{2M} \frac{c}{\omega_s} \sum_s \sum_{\tau} \frac{(\mathbf{Q} \cdot \mathbf{e}_s)^2}{\omega_s^2}
\]

\[
\cdot \left( \langle n_s + 1 \rangle \delta(\mathbf{Q} - \mathbf{q} - \tau)\delta(\omega - \omega_s) + \langle n_s \rangle \delta(\mathbf{Q} + \mathbf{q} - \tau)\delta(\omega + \omega_s) \right)
\]

(4.4)

\( \sigma_{coh} = 4\pi(b)^2 \) is the coherent scattering factor, where \( b \) is the averaged scattering length, \( M \) is the averaged mass of the atoms, \( v_0 \) is the volume of the unit cell, \( \tau \) is a reciprocal lattice
4.1 Introduction

vector and \( e^{x(-2W)} \) is the Debye-Waller factor. \( \mathbf{e}_s \) are the eigenvectors of the phonon belonging to the branch \( s \) defining the movement of the atoms around their equilibrium position. The \( \delta \)-function at \( \omega + \omega_s \) gives rise to Stokes lines (phonon absorption) and the \( \delta \)-function at \( \omega - \omega_s \) to anti-Stokes lines (phonon emission). \( \mathbf{q} \) is the wavevector of the phonon of energy \( \hbar \omega_s \). The \( \delta \)-functions are responsible for the conservation of energy and momentum and define the dispersion of the excitations: \( \omega = \omega_s(q) \). Due to the polarisation factor \( \mathbf{Q} \cdot \mathbf{e}_s \), only lattice vibrations along \( \mathbf{Q} \) contribute to the cross section, which allows an experimental discrimination between different eigenmodes (for example longitudinal and transversal). Important are also the Bose-Einstein population factors

\[
\langle n_s \rangle = \frac{1}{e^{\hbar \omega_s / kT} - 1},
\]

and

\[
\langle n_s + 1 \rangle = \frac{e^{\hbar \omega_s / kT}}{e^{\hbar \omega_s / kT} - 1}
\]

that give rise to the asymmetry between Stokes and anti-Stokes peak intensities (which can be corrected by means of the detailed balance factor). As \( T \to 0 \), \( \langle n_s + 1 \rangle \to 1 \) and \( \langle n_s \rangle \to 0 \), which means that the cross section for phonon absorption tends to zero. This is consistent with the fact that at zero temperature all the modes are in their ground states and there are no phonons to be absorbed.

Magnetic scattering

Due to their spins, neutrons also interact with the magnetic field arising from unpaired electrons in the scattering material. For magnetic scattering, the interaction operator is given by:

\[
\hat{U} = \frac{2\pi \hbar^2}{m} \sum_j p_j F_j(Q) \delta(\mathbf{r} - \mathbf{r}_j)
\]

where \( p_j \) is the magnetic scattering length and \( F_j(Q) \) is the magnetic form factor of atom \( j \) at the position \( \mathbf{r}_j \). The magnetic form factor is the Fourier transform of the normalized spin density of the unpaired electrons \( s(\mathbf{r}) \):

\[
F_j(Q) = \int s_j(\mathbf{r}) e^{iQ \cdot \mathbf{r}} d\mathbf{r}
\]

For unpolarised neutrons and identical magnetic ions with localized electrons one obtains the following magnetic cross section [66]:

\[
\frac{d^2\sigma}{dQd\omega} = (\gamma \hbar_0)^2 \frac{\hbar^2}{k} F^2(Q) e^{-2W} \sum_{\alpha,\beta} \left( \delta_{\alpha\beta} - \frac{Q_{\alpha}Q_{\beta}}{Q^2} \right) S^{\alpha\beta}(Q,\omega)
\]

where \( S^{\alpha\beta}(Q,\omega) \) is the magnetic scattering function:

\[
S^{\alpha\beta}(Q,\omega) = \sum_{ij} \exp(iQ \cdot (\mathbf{r}_i - \mathbf{r}_j)) \sum_{\lambda,\lambda'} \int p_\lambda(\lambda | \hat{S}_i^\alpha | \lambda') \langle \lambda' | \hat{S}_j^\beta | \lambda \rangle \delta(\hbar \omega + E_\lambda - E_{\lambda'})
\]
\( \gamma = -1.913 \) is the gyromagnetic ratio, \( r_0 = 0.282 \cdot 10^{-12} \text{ cm} \) is the classical radius of electron and \( S_\alpha^\beta(\alpha = x, y, z) \) is the spin operator of \( i \)th ion at the position \( \mathbf{r}_i \).

Two factors govern the cross section for magnetic neutron scattering in a characteristic way: the magnetic form factor \( F(Q) \), which usually falls off with increasing modulus of the scattering vector \( Q \), and the polarization factor \( (\delta_{\alpha\beta} - \frac{Q_\alpha Q_\beta}{Q^2}) \), which allows only the coupling of neutrons to magnetic moments (or spin fluctuations) perpendicular to \( Q \).

The essential factor in Eq.(4.9) is the magnetic scattering function \( S^{\alpha\beta}(Q, \omega) \), which is related to the imaginary part of the generalized magnetic susceptibility \( \chi''(Q, \omega) \) through the fluctuation-dissipation theorem:

\[
S^{\alpha\beta}(Q, \omega) = \frac{\hbar}{\pi} \frac{1}{1 - e^{-\hbar \omega / k_B T}} \chi''(Q, \omega)
\]  

(4.11)

This implies that the magnetic moment of the neutron acts as a wavevector and frequency dependent magnetic field that probes the dynamic magnetic response of the sample.

**Triple-axis spectrometer**

The determination of the scattering function \( S(Q, \omega) \) by neutron scattering techniques requires a controlled access to the variables \( Q \) and \( \omega \). An efficient experimental method is the triple-axis spectrometer (TAS), which allows the measurement of the scattered intensity as a function of momentum transfer \( \hbar Q \) and energy transfer \( \hbar \omega \) to the sample. A schematic view of a TAS instrument is shown in Fig.4.1. The Bragg scattering from the monochromator crystal (first axis) defines the wavevector \( k \) of the incoming neutrons,
which are scattered from the sample (second axis) along a direction defined by the scattering angle $\theta$. The scattered neutrons are Bragg reflected from the analyzer crystal (third axis) defining their wavevector $k_f$. The controlled access to the variables $k_i$, $k_f$ and $\theta$ completely determines the scattering process. The advantage of this spectrometer is that the data can be taken at a pre-determined point in reciprocal space (constant-$Q$ scan) as a function of energy transfer or for a fixed energy transfer (constant-$E$ scan) along a particular line in reciprocal space, so that measurements of dispersion relations in single crystals can be performed in a controlled manner. Of course, general scans in $Q$ and $\omega$ are also possible.

For structural studies (thus elastic neutron scattering) there is no need to perform an energy analysis by the third spectrometer axis. In this case the scattering law $S(Q,\omega)$ is integrated in $\omega$-space to give the structure factor $S(Q)$.

### 4.1.2 Spin excitations in HTSC

**Experimental facts**

As described in Chapter 1, the undoped parent compound of all HTSC is an insulator with the copper spins ordered antiferromagnetically. This antiferromagnetic state is characterized by an elastic magnetic peak at $(\pi, \pi)$ (see also Fig.4.2a), due to the double size of the magnetic unit cell compared to the nuclear unit cell, which can be observed in neutron scattering experiments [192]. Spin waves measured in La$_2$CuO$_4$ [193] are consistent with the conventional theory of the 2D $S=1/2$ Heisenberg Hamiltonian with $J \approx 135$ meV (see also Fig.4.3a). A small amount of doped holes is sufficient to destroy the AF order, which is replaced by an insulating spin glass phase [27, 28, 65]. By further increasing the doping level, in the superconducting state, the spin excitation spectrum of YBa$_2$Cu$_3$O$_{6+x}$ is dominated by a so-called magnetic resonance at an energy $E_{\text{res}} \approx 40$ meV (at optimal doping) located at the commensurate AF zone center $(\pi, \pi)$ [194]. Polarized neutron scattering experiments have confirmed the magnetic origin of the signal [195]. The resonance is present in the superconducting state below $T_c$ [196] but seems to survive up to the pseudogap temperature $T^*$ [197] and its energy decreases in the underdoped regime following the relation $E_{\text{res}} \approx 5k_B T_c$ [198, 199, 200]. Similar resonance peaks have been measured in Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ [201, 202, 203] and in the single-layer system Tl$_2$Ba$_2$CuO$_{6+x}$ [204], but has never been observed in La$_{2-x}$Sr$_x$CuO$_4$. This lead to the conclusion that LSCO has, for some unclear reason (e.g. different Fermi surface [205, 206], small values of $T_c$ and of the superconducting gap [207], disorder/impurity effects [208],...), a different spectra of the spin excitations and has to be regarded as a special case. Indeed in the LSCO family the spectral weight of the magnetic susceptibility is concentrated in four incommensurate magnetic peaks. For $0.02 < x < 0.05$, in the non-superconducting spin glass phase, the system is characterized by one-dimensional static spin modulations, which give rise to

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2Spin waves are described by the Hamiltonian $H = \sum_{ij} J \cdot S_i \cdot S_j$, where $J$ is the exchange operator and $S_i$, $S_j$ are the spin operators. Considering only nearest neighbor interaction, one obtains the excitation spectrum $\hbar \omega = 4S J \sqrt{1 - \gamma^2(Q)}$, with $\gamma(Q) = \frac{1}{2} [\cos(Q_x) + \cos(Q_y)]$.

3In the overdoped regime $T^*$ coincides with $T_c$. 

Inelastic neutron scattering study of the spin excitations

a) 0.00 < x < 0.02

\[(\pi, \pi)\]

\[\frac{1}{2} \quad \frac{1}{2}\]

\[(jt, jt)\]

b) 0.02 < x < 0.05

\[2\delta\]

\[\frac{1}{2} \quad \frac{1}{2}\]

c) 0.05 < x < 0.25

d) \(\delta \) (r.l.u)

Figure 4.2: Evolution of the magnetic response of LSCO as a function of doping (see text). a) and b) are cut in reciprocal space at zero energy transfer, whereas the cut in c) is at finite energy.

\[\textbf{diagonal\ incommensurate magnetic peaks [209, 210, 211]} \] \(^4\). Due to the presence of twin domains, four peaks at \(Q_\delta = (\frac{1}{2} \pm \frac{\delta}{\sqrt{2}}, \frac{1}{2} \pm \frac{\delta}{\sqrt{2}})\) are usually observed [209], as shown schematically in Fig.4.2b. By further increasing the hole concentration (0.05 < x < 0.25), LSCO becomes superconducting and inelastic parallel incommensurate peaks at \(Q_\delta = (\frac{1}{2} \pm \frac{\delta}{\sqrt{2}})\) and \((\frac{1}{2} \pm \frac{\delta}{\sqrt{2}})\) are observed [212, 29, 30], as shown in Fig.4.2c \(^5\). The incommensurability \(\delta\) was found to be strongly doping dependent (it increases linearly with doping and saturates at \(\delta = 0.13\) for x > 0.13 [29], see Fig.4.2d) but roughly independent from the energy transfer (at low energies). Static magnetic modulations with the same incommensurability have been observed in the very underdoped regime of LSCO (x < 0.13) as well as in La_{1.875}Ba_{0.125-x}Sr_xCuO_4 [213] and in La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4 [214, 215].

In LSCO, at higher doping (x > 0.13) where no elastic peaks are observed, a spin gap (\(\Delta_{SC}\)) opens below \(T_c\), e.g. spectral weight disappears below \(\Delta_{SC}\) (\(\approx 6.5\) meV at optimal doping) [216, 217, 218]. A similar spin gap has been observed in YBCO [219, 220, 221, 222].

\(^4\) The term diagonal arises from the fact the the spin-modulation vectors are at an angle of 45 degrees respect to the Cu-O bonds.

\(^5\) The term parallel arises from the fact the the spin-modulation vectors are parallel with respect to the Cu-O bonds.
In both LSCO and YBCO, $\Delta_{SG}$ is doping dependent, and decreases (vanishes) in the underdoped regime [218, 223, 222]. Interestingly, the spin gap seems to be absent also in the overdoped regime ($x > 0.20$) [218, 30]. While in the underdoped regime the absence of a spin gap might be related to the spin glass phase coexisting with superconductivity, in the overdoped regime it has been suggested that the spin gap cannot be observed due to the reduction of the coherence length of the spin correlation [218].

Recently, parallel incommensurate peaks (similar to those observed in LSCO) at low energies $\Delta_{SG} < \hbar \omega < E_{res}$ [224, 225, 226, 227, 222, 220, 221] and diagonal incommensurate peaks above $E_{res}$ [228] have been observed in YBCO. In the underdoped regime of YBCO, it has been suggested that the magnetic excitations are one-dimensional [229, 230], as for lightly doped LSCO. However, very recent data in untwinned YBCO crystals indicate a two-dimensional nature of the magnetic fluctuations [231]. Moreover, it was found that even for LSCO the incommensurability is not constant but decreases with increasing energy [232] as in YBCO. The horizontal incommensurate peaks most likely join at the commensurate wave vector $(\pi, \pi)$ [233, 234, 232] and disperse into diagonal peaks at higher energies, as it was found in La$_{1.875}$Ba$_{0.125}$CuO$_4$ [235].

These new results seem to indicate that all HTSC are characterized by a universal dispersion of the spin excitations, which is schematically drawn in Fig.4.3b. Since the incommensurate and commensurate magnetic peaks appear to be smoothly connected, it is therefore reasonable to assume that they have a common origin. A valuable theory for the spin excitations in HTSC should be able to account for such a downward (upward) dispersion below (above) $E_{res}$.

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In this case the incommensurate peaks are parallel at low energies, and diagonal at high energies, as in YBCO [228].
Inelastic neutron scattering study of the spin excitations

Theories

The presence of strong spin excitations in the cuprates has been interpreted within different competing theoretical models. For simplicity we will limit here our discussion to some of them.

In a first scenario, static incommensurate peaks are the consequence of the formation of stripes [33], e.g. a phase separation into hole-rich (metallic) and hole-poor (AF insulating) one-dimensional regions (see Fig.4.4). In this picture, the one-dimensional hole-rich stripes represent an anti-phase domain wall between the AF domains, giving rise to magnetic incommensurate peaks. The incommensurability $\delta$ is a measure of the distance between the stripes, and is expected to be proportional to the hole concentration, e.g. the doping $x$, as it was experimentally observed in LSCO [29]. Moreover, incommensurate charge peaks are also expected, due to the modulation of the charge distribution. The periodicity of these charge modulation is only half of that of the spins, and the incommensurability (in reciprocal space) is therefore expected to be $\epsilon = 2\delta$ [214]. Charge and spin incommensurate elastic peaks have been indeed observed in Neodymium and Barium doped LSCO [214, 236, 213, 237], where superconductivity is strongly suppressed. To notice is that the introduction of Nd/Ba distorts the usual low-temperature orthorhombic (LTO) structure into the low-temperature tetragonal (LTT) structure and suppresses the superconducting transition temperature. It was shown that (parallel) stripes are pinned in the LTT phase [214], but not in the LTO phase. Therefore one expects static charge and magnetic peaks in the LTT phase, whereas only dynamical stripe correlations should survive in the LTO phase [237]. For instance, the incommensurate magnetic peaks observed in LSCO and YBCO have been interpreted as dynamical stripes 7. Due to the suppressed $T_c$ in the presence of static stripes, one would expect that stripes are in competition to superconductivity [238]. However, it has been also suggested that dynamic stripes might be beneficial for superconductivity [239, 240]: pairs of holes from the stripes may obtain the pairing correlation by a virtual hopping into the magnetic domains. Obtaining commensurate peaks from the stripe scenario seems to be hard, but recently the downward dispersion has been interpreted as originating from the spin wave dispersion in an incommensurate antiferromagnet [241, 242, 243, 244, 245]. When the two incommensurate spin waves branches intersect at $(\pi, \pi)$, a sort of resonance is observed. Such a dispersion of (incommensurate) spin waves has been recently measured in the ideal stripe-ordered system $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$ [246, 247], but is quite different from the dispersion observed in the cuprates. Alternatevely, it has been recently suggested that the spectrum in HTSC can be understood in terms of weakly coupled two-leg spin ladders [235].

Within the SO(5) theory [32, 248], a unified theory of antiferromagnetism (AF) and superconductivity (SC) which describes the three AF degrees of freedom and the two SC degrees of freedom (real and imaginary part of the SC gap) in a unified Hamiltonian, the resonance peak at $(\pi, \pi)$ is interpreted as a collective mode describing a rotation from the SC state to the commensurate AF state [249, 250]. While this bound state in the spin-triplet particle-particle channel naturally results from the theory, recovering incommensurate peaks in this scenario is more challenging. However, the downward dispersion could be reproduced even within the SO(5) scenario [251].

7In both LSCO and YBCO, no charge peaks could be measured, yet.
Figure 4.4: Stripe pattern in a CuO$_2$ plane of LSCO (x=1/8). Arrows represent the copper spins, dark circles are doped holes. The magnetic modulation has a period of 8·a, whereas the charge modulation has period 4·a, a being the distance between copper atoms.

An alternative to the stripe and SO(5) approaches is based on a Fermi liquid like picture, where the spin excitations originate from Fermi surface pseudo-nesting [205, 252, 253, 254, 255, 256, 257]. In such a fermiology approach, quasiparticles close to the Fermi surface interact via a residual nearest neighbor AF interaction or a Hubbard-type repulsion. This situation is intermediate between spin-waves in Mott insulators, where all electrons are localized due to strong Coulomb interaction, and a non-interacting gas of electrons (Fermi liquid). In the latter case, a continuum of low-energy excitations are created by flipping the spin of an electron close to the Fermi surface (k, ↑) and moving it to a different place in reciprocal space (k + q, ↓). In the superconducting state, the energy of the (BCS) quasiparticle is given by $E_k = \sqrt{\varepsilon_k^2 + \Delta_k^2}$, where $\varepsilon_k$ is the band dispersion $^8$ and $\Delta_k$ is the momentum dependent superconducting gap (which can be measured in ARPES experiments). A good starting point for computing the imaginary part of the dynamical susceptibility is given by the non-interacting magnetic susceptibility [253]:

\[
\chi_0(\omega, q) = \sum_k \left[ \frac{1}{2} \left( 1 + \frac{\varepsilon_k \varepsilon_{k+q} + \Delta_k \Delta_{k+q}}{E_k E_{k+q}} \right) \frac{f(E_{k+q}) - f(E_k)}{\omega - (E_{k+q} - E_k) + i\Gamma} \\
+ \frac{1}{4} \left( 1 - \frac{\varepsilon_k \varepsilon_{k+q} + \Delta_k \Delta_{k+q}}{E_k E_{k+q}} \right) \frac{1 - f(E_{k+q}) - f(E_k)}{\omega + (E_{k+q} + E_k) + i\Gamma} \\
+ \frac{1}{4} \left( 1 - \frac{\varepsilon_k \varepsilon_{k+q} + \Delta_k \Delta_{k+q}}{E_k E_{k+q}} \right) \frac{f(E_{k+q}) + f(E_k) - 1}{\omega - (E_{k+q} + E_k) + i\Gamma} \right]. 
\]

where $f(E) = [exp\left(\frac{E}{k_B T}\right) + 1]^{-1}$ is the Fermi distribution function and $\Gamma$ encodes phenomenologically the broadening of the spin excitations (due to disorder) and is inversely proportional to their lifetime. The first term describes thermally excited quasiparticles in the presence of a superconducting gap, whereas the second (third) term describes the

$^8$The Fermi surface is defined by the condition $\varepsilon_k = 0$. 
annihilation (creation) of a spin-triplet from a Cooper pair (spin-singlet). At low temperatures the first two processes can be neglected and only the spin-triplet creation process is relevant:

\[
\chi_0(\omega, q) \approx \sum_k \frac{1}{4} \left( 1 - \frac{\varepsilon_k \varepsilon_{k+q} + \Delta_k \Delta_{k+q}}{E_k E_{k+q}} \right) \frac{f(E_{k+q}) + f(E_k) - 1}{\omega - (E_{k+q} + E_k) + i\Gamma} \tag{4.13}
\]

The first factor is called the coherence factor, whereas the second factor is a sort of two-particle density of states, which is used to describe the magnetic spectrum of simple metals. For low-energy excitations (energies much smaller than the Fermi energy) one can set

\[
\varepsilon_k = \varepsilon_{k+q} = 0 \tag{4.14}
\]

and the coherence factor simplifies to

\[
1 - \text{sign}(\Delta_k) \cdot \text{sign}(\Delta_{k+q}) \tag{4.15}
\]

For an isotropic (s-wave) gap, the coherence factor is therefore zero and the pair creation process is forbidden. On the other hand, for an anisotropic d-wave gap, the coherence factor is maximal when the nesting vector \(Q_n\) connects parts on the Fermi surface with a different sign of the gap (see Fig.4.5a). Moreover the two-particle density of states is maximal at the energy

\[
\omega_{\text{max}}(q, k) = |\Delta_k| + |\Delta_{k+q}| \tag{4.16}
\]

Usually, the interaction between quasiparticles is taken into account within the random phase approximation (RPA) and is given by:

\[
\chi_{\text{RPA}}(\omega, q) = \frac{\chi_0(\omega, q)}{1 - U \cdot \chi_0(\omega, q)} \tag{4.17}
\]
where $U$ is a Hubbard-type coupling constant. The imaginary part of (4.17) is the quantity measured in neutron scattering experiments (see Eq.(4.11)) and can be written as:

$$\chi''_{RPA}(\omega, \mathbf{q}) = \frac{\chi''_0(\omega, \mathbf{q})}{[1 - U\chi_0(\omega, \mathbf{q})]^2 + |U\chi''_0(\omega, \mathbf{q})|^2} \quad (4.18)$$

Due to the Kramers-Kronig relation, any step-like discontinuity in $\chi''_0(\omega, \mathbf{q})$ at some frequency $\omega_n(\mathbf{q})$ results in a logarithmic divergence of $\chi''_{RPA}(\omega, \mathbf{q})$ at $\omega_n(\mathbf{q})$ which guarantees that the condition $1 - U\chi_0(\omega, \mathbf{q}) = 0$ is met at an energy $\omega_n(\mathbf{q}) < \omega_0(\mathbf{q})$, therefore enhancing $\chi''_{RPA}(\omega, \mathbf{q})$. Numerical calculations [257] for optimally doped BSCCO are shown in Fig.4.5b. The main experimental observations can be reproduced: a strong magnetic resonance at $(\pi, \pi)$ is observed together with a downward dispersion into parallel incommensurate peaks and the opening of a spin gap $\Delta_{SG}$. Moreover, above the resonance energy, diagonal incommensurate peaks were also predicted [254, 257]. However, within this fermiology approach, commensurate and incommensurate peaks are obtained only in the superconducting phase below $T_c$, whereas numerous experiments (in particular in LSCO) have shown strong spin excitations even in the normal state ($T > T_c$). A practical drawback of RPA calculations is that they require precise measurements of the band dispersion (and the related Fermi surface) and of the SC gap function. Such information can be obtained by means of ARPES [160, 262, 263], but detailed measurements are nowadays available only for BSCCO. However, due to the small size of BSCCO samples, only few neutron scattering measurements have been reported so far.

By solving Eq.(4.14) for $\mathbf{q}=\mathbf{Q}_\delta$, one obtains two solutions $\mathbf{k}_1$ and $\mathbf{k}_2$ [257, 264]. In a first approximation, $\omega_{\max}(\mathbf{Q}_\delta, \mathbf{k}_1)$ determines the threshold to the 2-particle continuum, e.g. the spin gap value $\Delta_{SG}$. As it can be seen in Fig.4.5a, $\omega_{\max}(\mathbf{Q}_\delta, \mathbf{k}_1)$ is associated with excitations whose nesting vectors connects parts of the Fermi surface close to the nodes of the SC gap. We will discuss later how this observation might be important for the understanding of the doping dependence of the spin gap [257].

**Effect of magnetic fields on the spin excitations**

None of the theoretical models discussed in the previous section is able to completely explain the experimental observations. Each model has advantages but also drawbacks. Since these theories differ in the way the AF state of the undoped compound is related to the superconducting state, the application of a magnetic field provides a possibility to differentiate between them [265]. While conventional vortices with metallic cores emerge from the Fermi-liquid like picture, the SO(5) model predicts unusual vortices with bound AF-states [266, 267]. Finally, within the stripe scenario, vortices locally pin the charged stripes and lower the energy of the dynamic spin excitations, possibly leading to a field-induced quantum phase transition from a superconducting phase to a mixture of superconducting and spin density wave phase [268, 269].

Experiments in YBCO revealed that the resonance is significantly suppressed by a modest magnetic field (less than 10 Tesla) applied perpendicular to the CuO$_2$ planes [270].

---

9 The parameters for the dispersion $\varepsilon_\mathbf{k}$ and for the $d$-wave gap are taken from ARPES experiments [258, 259, 260, 261].

10 Due to its large anisotropy, this material is easy to cleave. Cleavage is needed to have a perfect surface, which is required in surface-sensitive techniques such as ARPES.
Inelastic neutron scattering study of the spin excitations

whereas for fields applied parallel to the CuO$_2$ planes the resonance remains unaffected [271, 270]. Within the fermiology approach, this result is interpreted as due to the suppression of the resonance in the vortex cores, due to a suppression of either the gap or the coherence factor [272]. It was also proposed that the resonance can account for the total condensation energy [250], which can be measured in specific heat experiments [112]. This connection between magnetic resonance and specific heat anomaly is corroborated by the fact that both features are strongly decreased under the influence of a moderate magnetic field [273]. However, there is no consensus about what the resonance can do [274] and cannot do [275, 276] for superconductivity and its relation to electronic excitations [277].

In LSCO, many experiments have also investigated the field dependence of the (incommensurate) spin excitations [278, 279, 280]. In optimally doped LSCO ($x=0.163$) sub-gap excitations induced by a magnetic field of 7.5 Tesla applied parallel to the c-axis have been observed at low-temperatures ($T<10$ K). Moreover, the spin gap was found to close at the irreversibility temperature rather than at $T_{c2}$ [278]. In underdoped LSCO ($x=0.10$) elastic neutron scattering experiments revealed a field-induced static signal below $T_c$ [279]. Similar results have been obtained in LSCO ($x=0.12$) [281] and in oxygen doped La$_{2-x}$CuO$_{4+y}$ [282, 283]. However, in non-superconducting lightly-doped LSCO ($x \approx 0.02$) [284] and in superconducting La$_{1.15}$Nd$_{0.4}$Sr$_{0.15}$CuO$_4$ ($T_c=15$ K) [285] an opposite result is observed, e.g. a decrease of intensity in a magnetic field. These results in superconducting LSCO seem to indicate a coexistence of antiferromagnetism and superconductivity, where the interplay between the two kinds of order can be continuously tuned by the external magnetic field. A possible interpretation of neutron scattering experiments in underdoped LSCO is that at large enough fields ($H > H_{c2}$) the normal state becomes an incommensurate AF insulator. This scenario is supported by resistivity [71, 286] and thermal conductivity [287] measurements. At lower fields ($H < H_{c2}$), it has been suggested that the field-induced magnetic signal arises from the AF vortex cores [266]. Enhanced AF spin correlations in the vortex core region have been indeed observed in NMR experiments [288, 289]. However, both the correlation length and the size of the magnetic signal are too large to arise just from the vortex cores. A possible solution to this problem is that the vortices nucleate magnetic regions which extend beyond the cores into the surrounding superconducting regions [267, 290]. In a simple model which assumes that the field-effect is originating from the vortices, a linear increase of the magnetic signal is expected, since the number of vortices is proportional to $H$ [13]. Considering the AF and SC order to be in competition, three different phases might exist [290]: a pure SC phase, a phase in which the stripe (or spin density wave (SDW)) order coexists and couples to superconductivity, and a pure SDW phase. The transition between these phases occurs by changing the doping (in zero field) or, alternatively, by applying a magnetic field [265]. In the latter case, the SDW

---

11 We have some evidence that the resonance in BSCCO is also unaffected by a magnetic field of 3.5 Tesla [59], whereas no result is available for fields perpendicular to the planes.

12 About this dispute, it is interesting to notice that Ref. [275] is entitled "What the resonance cannot do" in cond-mat/0110478, whereas Ref. [274] is entitled "What the resonance peak does" in cond-mat/0209435. The titles in the published versions in Phys. Rev. Lett. have been modified to a more conventional form (see Bibliography).

13 Due to the absence of vortices in non-SC samples, such an approach would be also consistent to the experimental results obtained in lightly doped LSCO [284].
order originates from the suppression of the order parameter in the vortex-core and in the surrounding region. The expected field dependence of the elastic incommensurate peak intensity at $T=0$ K is then given by \[ I(H) = I(0) + C \cdot \frac{H}{H_{c2}} \cdot \ln \left( \frac{H_{c2}}{H} \right) \] (4.19)

where $C$ is a constant. This prediction is in quite good agreement with the experimental results [279, 282, 283]. Recently, a further possible explanation supported by thermal conductivity experiments has been suggested, where the dynamical stripes are pinned by the vortex cores [269], thus giving rise to enhanced static signal.

### 4.1.3 Motivation and goals of our studies

It remains a challenge to understand the details of the doping dependence of the field-effect on the spin excitations in HTSC. In an attempt to better understand the open questions presented in the previous section, we have performed both inelastic and elastic neutron scattering measurements of the incommensurate peaks on slightly overdoped ($x=0.17$) and underdoped ($x=0.10, x=0.12$) LSCO single crystals in magnetic fields up to 14.5 Tesla. In Section 4.3 and Section 4.4 we will present INS results on overdoped ($x=0.17$) and underdoped ($x=0.10$) samples, respectively. Elastic measurements on underdoped LSCO ($x=0.10, x=0.12$) are described in Section 4.5.

### 4.2 Experimental setup

We investigated one slightly overdoped ($x=0.17, T_c=37$ K, $\Delta T_c=1.5$ K, $m=2.73$ g) and two underdoped ($x=0.10, T_c=29$ K, $\Delta T_c=1.5$ K, $m=3.86$ g and $x=0.12, T_c=27$ K, $\Delta T_c=1.5$ K, $m=3.21$ g) LSCO single crystals. The two underdoped samples are close to the $x=1/8$ anomaly (see Fig. 1.9).

The overdoped sample is the same crystal measured in macroscopic and SANS experiments (see Chapter 2 and Chapter 3). The inelastic neutron scattering data were taken on the IN22 spectrometer at the high-flux reactor of the Institute Laue Langevin (ILL) in Grenoble, France. We used a graphite vertically curved monochromator and graphite horizontally curved analyser with a fixed final energy of 13.8 meV with an energy resolution of about 1 meV. In order to avoid contamination by higher-order reflections a PG-filter in $k_f$ was installed. The sample was mounted in a cryostat with the c-axis oriented perpendicular to the scattering plane. A magnetic field up to 5 Tesla could be applied parallel to the c-axis. The Q-scans were performed by rotating the sample around its c-axis. Within such a scan, one can measure two different incommensurate peaks without changing the analyser and detector positions (see Fig.4.6a).

Inelastic neutron scattering measurements on underdoped LSCO ($x=0.10$) have been performed on IN22 and IN14 at ILL. On IN22 we used a similar experimental setup as for overdoped LSCO. On IN14 we used a graphite vertically curved monochromator, and a graphite horizontally curved analyser with fixed final energy ranging from 3.5 to 5 meV with an energy resolution of about 0.2 meV. A Berillium-filter was installed in $k_f$ to remove higher-order reflections. In a first experiment (in 2003) the sample was measured...
Inelastic neutron scattering study of the spin excitations

Figure 4.6: a) Reciprocal space of the CuO$_2$ superconducting planes. The four spots mark the incommensurate locations of the spin excitations around the ($\frac{1}{2},\frac{1}{2}$) point. The solid arc shows the wavevectors spanned in a constant-energy scan (Section 4.3 and 4.4). The solid vertical line indicate a Q-scan through one incommensurate peak (Section 4.5). The magnetic field $H$ was applied perpendicular to these planes. In b) and c) we show a photograph of the LSCO ($x=0.10$) sample measured on IN14 in 2003 and 2004, respectively, mounted on an Aluminum sample holder, and oriented with the c-axis vertical. For the experiment in a magnetic field, the crystal was cut in two pieces in order to fit the magnet.

In zero-field, whereas in 2004 we repeated the experiment in vertical magnetic fields up to 14.9 Tesla. In order to fit into the magnet, the sample has been cut in two pieces, as shown in Fig.4.6b and c. On a piece of this crystal we also performed the macroscopic measurements presented in Chapter 2.

Moreover we performed elastic neutron scattering experiments in fields up to 14.5 Tesla on the V2/FLEX spectrometer at the Hahn-Meitner Institute (HMI Berlin) on the same $x=0.10$ sample used at ILL. The energy was fixed at 7.5 meV using a graphite vertically curved monochromator and flat graphite analyser, and a tunable PG filter was used to eliminate second-order scattering. A similar elastic neutron scattering experiment in fields up to 8.5 Tesla has been performed on Rita-II at PSI on the $x=0.12$ underdoped sample. The energy of the neutrons was 5.57 meV (vertically curved monochromator, flat analyser) and a PG-filter was installed before the sample. During our elastic measurements we concentrated on one incommensurate peak and we performed vertical Q-scan as shown in Fig.4.6a.

All the measurements have been performed after cooling in a field (FC) applied parallel to the c-axis.
4.3 Overdoped La$_{2-x}$Sr$_x$CuO$_4$($x=0.17$)

4.3.1 Measurements in zero field

We start with the description of the data obtained at zero-field. Fig.4.7a shows Q-scans through two incommensurate peaks at an energy transfer $\hbar\omega=4$ meV for temperatures above and below $T_c$. Fig.4.7b shows a similar scan at $\hbar\omega=11$ meV and $T=5$ K. The trajectory of the scan is shown in Fig.4.6a. Above $T_c$ we observe clear peaks at the incommensurate positions $Q_\delta=(\frac{1}{2}, \frac{1}{2}+\delta)$ and $(\frac{1}{2}+\delta, \frac{1}{2})$ with $\delta=0.13(1)$. The incommensurability $\delta$ is consistent to the values found in the literature for similar doping levels of LSCO [29, 217]. Below $T_c$ the magnetic intensity at low energy disappears, which is a clear sign for the opening of a spin gap in the superconducting state. The presence of the gap is further confirmed by the energy scans performed at $Q_\delta$, see Fig.4.8a. For $T=5$ K ($< T_c$), $\chi''(Q_\delta, \omega)$ drops sharply below about 6.5 meV, which we identify with the spin gap energy $\Delta_{SG}$, whereas for $T=40$ K ($> T_c$), $\chi''(Q_\delta, \omega)$ doesn’t vary much as a function of energy $^{14}$ The energy dependence of the susceptibility at the incommensurate wave vector $Q_\delta$ can be analyzed using the phenomenological function introduced by Lee et al. [218]:

$$\chi''(Q_\delta, \omega) \propto \left( \frac{\Gamma}{\Gamma^2 + (\hbar\omega)^2} \right) \cdot \text{Re} \left( \frac{\hbar\omega - i\tilde{\gamma}}{\sqrt{\hbar\omega - i\tilde{\gamma})^2 - \Delta_{SG}^2}} \right)$$

(4.20)

This corresponds to Lorentzian spin fluctuations in the normal state multiplied by a gap-function in the superconducting state, where $\Delta_{SG}$ is the spin gap energy, $\tilde{\gamma}$ a broadening term and $\Gamma$ the inverse lifetime of the spin fluctuations. At low temperature and zero-field, we obtain a spin gap value of $\Delta_{SG} = 6.5 \pm 0.4$ meV consistent with earlier results [218, 217].

In Fig.4.8b we have plotted the difference of the high- and low-temperature susceptibility $\chi''(Q_\delta, \omega)$ in zero-field. A weight transfer from the high-energy to the low-energy region is observed, most likely indicating a conservation of spin as reported earlier [292] and also expected in more recent theoretical studies [264].

Finally, we present the temperature dependence of the spin fluctuations. Fig.4.9 shows the neutron counts (without background subtraction) as a function of temperature at $Q_\delta$ and at an energy transfer of 2.5 meV. The background was found to be dependent on the energy transfer, but independent on temperature (5 K $< T <$40 K) and magnetic field ($H \lesssim 5$ T). The sharp decrease of the intensity with decreasing temperature indicates the opening of the spin gap. At zero field the intensity drops at $T_{SG} \approx T_c$, thus demonstrating that the spin gap is related to the SC gap.

4.3.2 Measurements in a magnetic field

We turn now to the magnetic field dependence of the spin excitations. In Fig.4.10a are shown Q-scans similar to those shown in Fig.4.7a but in a magnetic field of 5 Tesla applied parallel to the c-axis. On can notice that the incommensurate peaks are not strongly affected by the application of an external magnetic field at $\hbar\omega=4$ meV. In particular,

$^{14}$It has been proposed that the peaked feature at $\hbar\omega \approx \Delta_{SG}$ is due to the presence of a pseudogap [291].
Figure 4.7: Constant energy-scans through two incommensurate peaks at zero field: a) for an energy transfer $\hbar \omega = 4$ meV, $T=5$ K and 40 K, and b) for $\hbar \omega = 11$ meV, $T=5$ K.

Figure 4.8: a) Energy dependence of $\chi''(Q_\delta, \omega)$ in zero-field at $T=5$ K and 40 K. A background measured away from $Q_\delta$ has been subtracted from the raw neutron data. While at high temperature the intensity is roughly constant, at low temperature a gap is present. The data at low temperature have been fitted by Eq.(4.20). b) Difference of the high- and low-temperature susceptibility $\chi''(Q_\delta, \omega)$ (shown in Fig.4.8a) measured at $\mu_0 H=0$ T.
4.3 Overdoped La$_{2-x}$Sr$_x$CuO$_4$ ($x=0.17$)

Figure 4.9: Temperature dependence of the neutron intensity for an energy transfer $\hbar \omega=2.5$ meV taken in zero-field, indicating the opening of the the spin gap below $T_c$.

our slightly overdoped sample, there is no clear indication of field-induced sub-gap excitations at low temperatures, as shown also in Fig.4.10b for $h\omega=3$ meV. However, one can notice that in the $Q$-scan performed at $\mu_0 H=5$ T ($T=5$ K, Fig.2.4a) there is slightly more intensity than in the $Q$-scan performed at $\mu_0 H=0$ T ($T=5$ K, Fig.2.2a). By taking the zero-field data at $T=5$ K as background and fitting two Gaussians with fixed positions ($H=\pm \delta$) and widths, we estimate that the value of $\chi''(\mu_0 H=5$ T, $T=5$ K) is about $30\%\pm15\%$ of $\chi''(\mu_0 H=0$ T, $T=40$ K). This value is in agreement with $\chi''(Q_0, \omega)$ obtained from the energy scan shown in Fig.4.11a. A fit of the energy scan with Eq.(4.20) indicate that the spin gap $\Delta_{SG}$ decreases by about $25\%$ ($\pm20\%$). However, due to the large error on the determination of the spin gap value, no definitive conclusion can be reached as for the field-dependence of the spin gap.

It is very instructive to compare the difference between the 5 Tesla and zero-field susceptibility $\chi''(Q_0, \omega)$ at $T=5$ K (see Fig.4.11b) to the difference of the high- and low-temperature susceptibility $\chi''(Q_0, \omega)$ in zero-field (shown in Fig.4.8b). In both cases we observe a weight transfer from the high-energy to the low-energy region centered at $\Delta_{SG}(0$ T). The low-energy field-induced weight transfer at 5 Tesla is about $35\%$ ($\pm15\%$) of the temperature-induced weight transfer measured between $T=5$ K and $T=40$ K through $T_c$. This result is very surprising if one considers that the applied field represents only $10\%$ of $H_{c2}(5$K)$\sim50$ Tesla (as determined from resistivity data [76]).

Finally, we present the temperature dependence of the spin fluctuations as a function of magnetic field. Fig.4.12 shows the neutron counts (without background subtraction) as a function of temperature at $Q_0$ and at energy transfers of $\hbar \omega=2.5$, 4 and 11 meV. For energies lower than the zero-field spin gap ($\hbar \omega < \Delta_{SG}(0$ T)), the spin gap opens at about $T_{SG}=22$ K, which is about 15 K lower than $T_c$. For $\hbar \omega=11$ meV $> \Delta_{SG}(0$ T), on the other hand, the intensity is found to decrease around $T_{SG}$. This second observation is consistent with the spectral weight redistribution observed before: when the spin gap
Figure 4.10: a) Constant energy-scans in a field of \( \mu_0 H = 5 \) T for \( h\omega = 4 \) meV, \( T = 5 \) K and 40 K. b) Similar scan at \( h\omega = 3 \) meV, \( T = 1.55 \) K.

Figure 4.11: a) Energy dependence of \( \chi''(Q_0, \omega) \) at \( T = 5 \) K and \( \mu_0 H = 5 \) T. The filled circle indicates the value of \( \chi'' \) at 4 meV obtained independently from the Q-scan measured at \( \mu_0 H = 5 \) T, see text. The curve is a fit of the data using Eq.4.20. b) Difference of the \( \mu_0 H = 5 \) T and zero-field susceptibility \( \chi''(Q_0, \omega) \) (shown in Fig.2.4b and Fig.2.3a) measured at \( T = 5 \) K. Similarly to the temperature effect, the application of a magnetic field induces a transfer of weight from high- to low-energies. The field-induced increase of \( \chi'' \) at 4 meV, obtained independently from the Q-scan (see text), has also been included (filled circle).

opens the intensity which is lost at low energies is transferred to higher energies. What is surprising is that the application of a modest field suppresses \( T_{SG} \) by about 40\%, whereas \( T_c \) is only slightly affected by a field of 5 T, see Chapter 2. The application of a magnetic field has therefore an unusually strong influence on the temperature dependence of the
Figure 4.12: Temperature dependence of the neutron intensity a) for energy transfers below the zero-field spin gap $\Delta_{SG}$ ($\hbar\omega=2.5$ meV and 4 meV), and b) for $\hbar\omega=11$ meV (> $\Delta_{SG}$). Solid lines are guide to the eye. The dotted line represent the temperature dependence in zero-field (see Fig.4.9). Measurements have been performed in a field of $\mu_0 H=5$ T heating up after field-cooling. Vertical dashes lines indicate the value of $T_{FOT}$ at 5 T as determined by macroscopic measurements, see Fig.4.13.

spin excitations.

4.3.3 Discussion

In order to understand the field effect on $T_{SG}$, we can compare the INS results to the macroscopic data presented in Chapter 2 and performed on a piece of the crystal used for neutron scattering experiments. Data taken at 5 Tesla are reproduced in Fig.4.13. One can notice that between zero and 5 Tesla, $T_c$ changes by only 4 K. Therefore, one would expect that the application of such a magnetic field would only marginally affect the temperature at which the spin gap opens. As can be seen in Fig.4.12, this is clearly not the case since in a field of 5 Tesla the point at which the gap starts to fill in is shifted toward a much lower temperature than the measured $T_c$. Interestingly, $T_{SG}$ seems to agree well with the measured $T_{FOT}$, which is also the temperature at which we lose the vortex lattice intensity in SANS experiments (see Chapter 3).

The suppression of the spin gap in the vortex gas phase above $T_{FOT}$ can be understood qualitatively by assuming that in this region of the phase diagram, the time scale characterizing the dynamics of the vortices becomes comparable to that of the Cu-spin excitations [278]. Such a coupling between the electronic and vortex-fluid degrees of freedom has been inferred earlier, on one hand from Hall measurements where a sharp change of the conductivity is observed at the melting transition [294], and on the other hand from the observation of an unusually large increase of the specific heat when passing from the solid- to the fluid-vortex state [168]. The combination of our INS, SANS and macroscopic measurements could be also consistent with the scenario, suggested by Cooper et al. [295],
where the FOT line corresponds to the $H_{c2}$ line which has been suppressed by thermodynamic fluctuations. The vortex fluid phase is then interpreted as a state with large (critical) superconducting fluctuations. Another recent theoretical model suggests that in magnetic fields close to $H_{c2}$ the conventional vortex structure is replaced by the formation of superconducting filaments embedded in the matrix of the normal metal [296]. With decreasing magnetic field, the configuration of superconducting filaments then undergoes a first-order phase transition into the conventional vortex lattice state. All these theoretical models and experiments, including our own, point toward a more complex nature of the vortex fluid state than believed so far [34, 35]. It remains a theoretical challenge to understand, at a quantitative level, the connection existing between these anomalous observations.

A second important result of our investigation in slightly overdoped LSCO ($x=0.17$) is the absence of clear magnetic field induced sub-gap excitations. This result is in contrast to the data of Lake et al. in optimally doped LSCO ($x=0.163$). It may appear astonishing that such a small change in Sr-content strongly affects the spin dynamics in a magnetic field. This drastic change of the field-effect on the spin excitations is possibly due to the proximity of a quantum critical point at $x=0.19$, which separates the overdoped regime, where a Fermi-liquid like description is adequate, from the underdoped regime, characterized by unusual (non Fermi-liquid) normal state properties and by the formation of a pseudogap below $T^*$ [297, 298, 22]. On the other hand, we do observe an unusually large spectral weight redistribution centered at the spin gap energy when a magnetic field of
4.4 Underdoped La$_{2-x}$Sr$_x$CuO$_4$($x=0.10$)

In a first experiment on IN22, we performed a similar experiment on underdoped LSCO ($x=0.10$) as the one presented in the previous section 4.3 on overdoped LSCO ($x=0.17$) [299, 223]. The results of such measurements are summarized in Fig.4.15. In contrast to the slightly overdoped sample, the spin excitations at $Q_s$, with $\delta \approx 0.10$, are present both above and below $T_c$, and are not affected by the application of a magnetic field of 5.5 T parallel to the $c$-axis for $h\omega > 2$ meV. From the energy dependence of $\chi''(Q_s,\omega)$ there is no evidence of a spin gap down to energies as low as 2 meV. Moreover, $\chi''(Q_s,\omega)$ is almost temperature independent and no anomaly at $T_c$ could be observed (not shown). However, the presence of a gap smaller than 2 meV couldn’t be ruled out. In order to clarify whether a small spin gap is present or not in the underdoped regime, we performed INS experiment on IN14 with increased energy- and $Q$-resolution on the same single crystal. We investigated the low-energy excitations for energy transfers in the range $0.3$ meV$\leq h\omega \leq 3$ meV.
4.4.1 Measurements in zero field

We start with the results obtained in zero field. Constant energy scans through two incommensurate peaks are shown in Fig. 4.16. Measurements have been performed at base temperature ($T=1.5$ K) and above $T_c$ ($T=33.5$ K). The data have been fitted by a double Gaussian with correlated peak positions $x = \pm \delta$ and width $w$ (similar fits with all free parameters lead to similar results):

$$f(x) = A \cdot x + B + C_1 \cdot e^{-(x-\delta)^2/w} + C_2 \cdot e^{-(x+\delta)^2/w}$$ (4.21)

The background $A \cdot x + B$ was found to be slightly energy dependent. Apart from the data taken at $\hbar \omega=0.3$ meV, a flat background was chosen ($A=0$). Moreover the high- and low-temperature background were forced to be identical.

One can notice that spin excitations are clearly observed at low-temperatures down to $\hbar \omega=0.3$ meV, therefore signifying the absence of a spin gap in this underdoped sample. Moreover, the calculated susceptibility $\chi''$ is found to be larger at $T=1.5$ K than at $T=33.5$ K. This is exemplified in Fig. 4.16a’ where the neutron counts have been multiplied, after subtraction of a linear background, by the Bose factor $(1 - \exp(-\frac{\hbar \omega}{k_B T}))$ (see Eq.(4.11)).

In Fig. 4.17 the energy dependence of the incommensurability $\delta$ and width $w$ are shown. The values obtained from the fits by Eq.(4.21) didn’t show any temperature dependence (the different value of $\delta$ at 0.3 meV is probably not intrinsic) while a slight energy dependence was found both in $\delta$ and $w$. In particular the width of the peaks seems to broaden at 3 meV. However, this might be also a resolution effect.

The other quantity that can be extracted from the fits of Q-scans, the intensity $(C_1 + C_2)$ of the incommensurate peaks, is shown as a function of energy in Fig. 4.18. Data from energy scans at $Q_6$ (after subtraction of a background measured at one point in reciprocal space away from $Q_6$) have been also included. One could argue that the correct quantity to investigate is the integrated intensity and not the maximal intensity. However,
4.4 Underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4 (x=0.10)$

Figure 4.16: a)-e) Zero-field constant energy scans through two incommensurate peaks in LSCO ($x=0.10$) at various energy transfers ($0.3 \text{ meV} < \hbar \omega < 3 \text{ meV}$) measured above (empty circles) and below (full circles) $T_c=29 \text{ K}$. Data have been fitted with double-Gaussians and linear background, see Eq.(4.21). In a’) the raw data shown in a) have been transformed into the susceptibility $\chi''$.

since the width of the peaks is temperature and energy independent (at least for $\hbar \omega \leq 2 \text{ meV}$, see Fig.4.17b) we can equally consider the maximal intensity, which allows us to directly compare data from Q- and energy-scans. As shown in Fig.4.18 the intensity at $T=33.5 \text{ K}$ is roughly energy independent, whereas at $T=1.5 \text{ K}$ the intensity is peaked at $\hbar \omega=1.5 \text{ meV}$ and decreases at lower energies. After multiplication with the Bose factor the energy spectrum is strongly modified. Above $T_c$, $\chi''$ steadily decreases with decreas-
Figure 4.17: Energy dependence of a) the incommensurability $\delta$ and b) the width $w$ of the peaks.

Figure 4.18: a) Energy dependence of the maximal intensity of the spin excitations obtained from fits of the $Q$-scans (squares) and by energy scans performed at $Q_\delta$ (circles) at $T=1.5$ K (full symbols) and $T=33.5$ K (empty symbols). b) Energy dependence of the imaginary part of the susceptibility $\chi''(Q_\delta, \omega)$ at $T=1.5$ K and $T=33.5$ K.

The imaginary susceptibility $\chi''$ deviates from the high energy susceptibility below about 2 meV and extrapolates to a finite value at zero energy transfer. The anomalous increase of $\chi''$ at low temperatures and low energies is surprising, since one expects a decrease if a spin gap is formed (as it is experimentally observed in the overdoped LSCO sample, see Fig.4.8a). To notice is that the raw intensity actually decreases below 1.5 meV compared to the normal
state intensity, mimicking the opening of a spin gap. The anomalous enhancement of $\chi''$ observed in Fig.4.18b is a consequence of the Bose-factor which is very sensible to the value of the energy at low temperatures, as shown in Fig.4.19. As a consequence, one should be careful about the interpretation of the raw data.

We also investigated the temperature dependence of the spin excitations. Temperature scans at $h\omega=0.3$, 1 and 3 meV are shown in Fig.4.20. We measured only at the peak position $Q_\delta$ (for the foreground) and a background point, therefore assuming that the width is temperature independent (this seems to be the case at least up to $\sim$35 K). For all energies, the maximal intensity of the incommensurate peaks increases steadily with decreasing temperature down to $T_c$. This increase is stopped by the onset of superconductivity and the intensity slightly decreases below $T_c$. If one plots the imaginary part of the susceptibility instead of the intensity, one observes that $\chi''$ increases monotonously with decreasing temperature. This increase is weaker below $T_c$ but a further strong enhancement is observed at low energy transfers ($h\omega=0.3$ meV and 1 meV) below about 15 K ($=T'_{sg}$). Therefore, the anomalous increase of $\chi''$ at low energies ($h\omega < 2$ meV) shown in Fig.4.18b occurs at low temperatures ($T < T'_{sg}$) and is not directly related to $T_c$. By plotting $\chi''(Q_\delta, \omega)/\omega$ measured at different energies in a single graph (see Fig.4.21) one can notice that above $T_c$ all curves follow a power law with exponent $-2$:

$$\frac{\chi''(Q_\delta, \omega)}{\omega} \propto T^{-2} \quad (4.22)$$

A similar temperature dependence in the normal state has been observed in slightly underdoped LSCO ($x=0.14$) and was attributed to the proximity to a quantum critical point (QCP) [300]. Below $T_c$ the data taken at different energies do not coincide any more: for $h\omega=3$ meV $\chi''$ remains constant down to the lowest temperatures, whereas for $h\omega=0.3$ meV and 1 meV, $\chi''$ sharply increases below $T'_{sg}$. 

Figure 4.19: a) Bose factor (BF) $(1-e^{-\frac{h\omega}{k_BT}})$ as a function of energy calculated for $T=1.5$ K and $T=33.5$ K. b) Ratio of the Bose factors at $T=1.5$ K and $T=33.5$ K shown in a). To notice is the sharp increase of the ratio below 1 meV.
Figure 4.20: a)-c) Temperature dependence of the maximal intensity of the incommensurate peak at \( h\omega = 0.3, 1 \) and 3 meV. d)-f) Calculated susceptibility \( \chi''(Q,\omega) \) as a function of temperature.

### 4.4.2 Zero-field muon spin relaxation measurements

In order to better understand the anomalous increase of the susceptibility at low energies and low temperatures, we performed zero-field muon spin relaxation (\( \mu \)SR) experiments on the same crystal used in INS experiments. In Fig. 4.22a zero-field \( \mu \)SR time spectra at different temperatures are shown. Such spectra represent the time evolution of the \( \mu \)SR decay process.

\footnote{These measurements have been performed by C. Niedermayer at the Paul Scherrer Institute, Switzerland.}
4.4 Underdoped La_{2-x}Sr_xCuO_4 (x=0.10)

Figure 4.21: a) Temperature dependence of \( \chi''(Q_\delta, \omega)/\omega \). Inset: low-temperature region. \( T_{sg}^n \) is defined as the temperature where the susceptibility begins to increase with decreasing temperature. b) Double logarithmic plot of the data shown in a). Above \( T_c \) the data at different energies collapse on one a common straight line with slope = -2.

Asymmetry parameter \( A(t) \), defined as

\[
A(t) = \frac{F(t) - B(t)}{F(t) + B(t)}
\]

where \( F(t) \) and \( B(t) \) are the total muon events of the forward and backward counters, which were aligned with respect to the beam direction, respectively. Data has been fitted with a two component fit. The first component is a Gaussian Kubo-Toyabe function [301] fitted at high temperature \( (T=30 \text{ K}) \) and kept constant at all other temperatures. The second component is an exponential relaxation \( A_0 \cdot \exp(-\lambda t) \), where the relaxation rate \( \lambda \) is the main fitting parameter. The baseline and initial asymmetry were fitted at low and high temperatures respectively, and kept constant for the whole temperature range. The fitted values of the relaxation rate \( \lambda \) are shown in Fig.4.22 as a function of temperature. Below 10 K \( \lambda \) increases due to the freezing of the Cu spin. Such a behavior is known to be a signature of the spin glass phase coexisting with superconductivity in the underdoped regime of LSCO [27]. Interestingly the value of \( T_{sg}^n \) obtained by \( \mu \)SR roughly agrees with \( T_{sg}^a \) obtained in neutron scattering experiments (see inset of Fig 4.21a) [16]. This suggests that the spin glass phase is responsible for the magnetic signal observed in our INS investigations (and vice versa). As a consequence, a spin gap could be present, but not observable due to the superposition of the signal occurring from the spin glass. In other words, the additional excitations in the spin glass phase could obscure the possible (and expected) existence of a spin gap.

\[16\text{More precisely, } T_{sg}^n \text{ is slightly higher than } T_{sg}^a, \text{ due to the different time resolutions of the two techniques.}\]
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Figure 4.22: a) $\mu$SR time spectra measured at different temperatures on the same under-doped LSCO ($x=0.10$) sample used for the INS investigations. b) Relaxation rate as a function of temperature.

Figure 4.23: Q-scan measured at 10 Tesla after cooling in a field down to 2 K. a) $\hbar\omega=0.5$ meV and b) $\hbar\omega=2$ meV.

4.4.3 Measurements in a magnetic field

Our INS experiment in zero field combined with $\mu$SR experiments indicate that the magnetic signal at low temperatures possibly arises from a spin glass phase coexisting with superconductivity. However, it remains to demonstrate whether the additional and conventional "gapped" spectra belong to two separated objects or are part of a unique response. In order to clarify this aspect we performed INS experiments as a function of magnetic field.

In Fig.4.23 constant energy scan in a magnetic field of 10 Tesla measured at 2 K after field-cooling for $\hbar\omega=0.5$ meV and 2 meV are shown. Compared to the measurements in zero field we didn’t notice large differences, neither in the intensity, nor in the incom-
m) Underdoped La_{2-x}Sr_xCuO_4 (x = 0.10)

Figure 4.24: Temperature scan at the peak position \( Q_{\delta} \) in zero field and at 10 Tesla (FC) for \( h\omega = 1 \) meV. a) Neutron counts, b) \( \chi''(Q_{\delta}, 1 \) meV).

Figure 4.25: Temperature scan at the peak position \( Q_{\delta} \) in zero field and at 12.5 Tesla (FC) for \( h\omega = 0.3 \) meV. a) Neutron counts, b) \( \chi''(Q_{\delta}, 0.3 \) meV). Lines are guides to the eye.

density \( \delta \) and width \( \omega \). Hence, we conclude that there is no strong field effect on the incommensurate peaks at base temperatures. The temperature dependence of the intensity at the peak position \( Q_{\delta} \) is shown in Fig.4.24 and Fig.4.25 for \( h\omega = 1 \) meV and 0.3 meV, respectively. The background was found to be independent of the temperature and magnetic field and is indicated by the grey area. In zero field, we could reproduce the data of the first experiment presented in Section 4.4.1. In particular we have now more temperatures and better statistics for \( h\omega = 1 \) meV. The intensity is peaked at \( T_c \), indicating a strong influence of superconductivity on the spin excitations, and \( \chi'' \) is enhanced below \( T^a_{sg} \approx 15 \) K. The application of a magnetic field of 10 Tesla enhances the intensity in a finite temperature range \( 10 \text{ K} \lesssim T \lesssim 30 \text{ K} \), whereas above \( T_c \) and at base temperatures the spin excitations are not much affected. Similar results have been obtained at
Figure 4.26: \( \mathbf{Q} \)-scan at \( h\omega=0.3 \) meV, \( T=22.3 \) K in zero field and in a field of 12.5 Tesla. Data have been fitted by a Gaussian, see Table 4.1

Table 4.1: Fitted parameters of the \( \mathbf{Q} \)-scans performed at \( h\omega=0.3 \) meV and \( T=22.3 \) K shown in Fig.4.26.

<table>
<thead>
<tr>
<th>Field</th>
<th>Max. Intensity (counts/12 min)</th>
<th>Width (r.l.u.)</th>
<th>Int. Intensity (a.u.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 Tesla</td>
<td>44.5 ± 3.2</td>
<td>0.030 ± 0.002</td>
<td>1.33 ± 0.13</td>
</tr>
<tr>
<td>12.5 Tesla</td>
<td>88.7 ± 5.3</td>
<td>0.024 ± 0.003</td>
<td>2.23 ± 0.29</td>
</tr>
</tbody>
</table>

\( h\omega=0.3 \) meV, even though these data have been taken with half-statistics and less points with respect to the data at 1 meV. At 0.3 meV the field-induced enhancement is even stronger, but still limited to a finite temperature range. The absence of an effect of the magnetic field on the spin excitations at base temperature is in contrast to the elastic measurements of Lake et al. [279], which indicate a strong field-induced enhancement of the elastic incommensurate peaks down to 2 K (see also Section 4.5).

In Fig.4.26 we show the field dependence of one incommensurate peak measured at \( T=22.3 \) K. One can notice that the peak at 12.5 Tesla is not only stronger but also sharper than the peak in zero field. The fitted values are summarized in Table 4.1. Remember that at base temperature we didn’t observe any enhancement nor sharpening of the incommensurate peaks in a magnetic field.

Energy scans have been performed at the peak position \( \mathbf{Q}_s \) at different fields and temperatures. In Fig.4.27 are shown the results obtained at \( T=22.3 \) K, \( \mu_0 H=0 \) T and 10 T. A field-induced signal is clearly observed below about 1.5 meV. Fig.4.27b is reminiscent of Fig.4.18b, where a similar low-energy enhancement of \( \chi'' \) was observed upon cooling below 15 K. The application of a magnetic field seems therefore to increase the temperature...
4.4 Underdoped La$_{2-x}$Sr$_x$CuO$_4$($x=0.10$)

Figure 4.27: Energy dependence of a) the maximal intensity of the incommensurate peaks and b) $\chi''(Q_s, \omega)$ measured at $T=22.3$ K for $\mu_0H=0$ T and 10 T. A clear field-induced signal is observed below $\hbar\omega \approx 1.5$ meV. Lines are guides to the eye.

range where $\chi''(Q_\delta, \hbar\omega < 1.5$ meV) is enhanced. It is therefore possible that the spin glass phase is extended up to higher temperatures upon the application of a magnetic field. Field-scan at $Q_\delta$ have been performed at $T=22.3$ K for $\hbar\omega=0.3$, 0.5, 1 and 2 meV (see Fig.4.28). Measurements at different fields have been always performed after field-cooling from above $T_c$. For $\hbar\omega \leq 1$ meV we observed a clear field-induced enhancement of the incommensurate peaks, whereas at $\hbar\omega=2$ meV the peak intensity is basically unaffected by the application of a field up to 14.9 Tesla. These results are consistent to the temperature-, $Q$- and energy-scans shown in Fig.4.24, Fig.4.25, Fig.4.26 and Fig.4.27, respectively. The data can be well fitted by Eq.(4.19) using the value $H_{c2} = H_{c2}(0 \text{ K}) = 45$ T [76, 279], even though this theoretical prediction is in principle valid only for $T=0$ K. Such a field dependence has been previously reported for the elastic incommensurate signal in LSCO ($x=0.10$) [279]. However, in that experiment, the field-scan was measured at base temperature, whereas in our case we performed the measurements at finite temperature. We should therefore use $H_{c2}(T=22.3$ K)$\approx 20$ T (see Fig.4.29), which leads to a less satisfying agreement between experiment and theory.

In general, our experimental results indicate a non-linear field-induced signal at low energies and finite temperatures, and we have some evidence for a saturation of the signal at large fields. However, since we are measuring at the peak position $Q_\delta$, a field-dependence of the width of the incommensurate peaks might have a strong influence on the field dependence of the integrated susceptibility $\chi''$.

4.4.4 Discussion

Before starting the discussion of our measurements in underdoped LSCO, it is worth to summarize the experimental results. Measurements in zero-field revealed an unusual energy and temperature dependence of the incommensurate spin excitations. We couldn’t observe the expected opening of a spin
gap below $T_c$. On the contrary, the low-energy susceptibility ($\hbar \omega < 2 \text{ meV}$) is enhanced at low temperatures ($T < T_{sg}^m \approx 15 \text{ K}$). Muon-spin relaxation experiments on the same sample revealed a sudden increase of the relaxation rate below $T < T_{sg}^m \approx 10 \text{ K}$, which is usually associated with the freezing of the copper spins in a spin glass phase. It is therefore attempting to interpret the low-energy spin excitations observed in INS at low temperatures as being caused by the spin glass phase coexisting with superconductivity. As a consequence, it is possible that a spin gap is present, but cannot be seen due to this additional contribution.

The application of an external magnetic field perpendicular to the CuO$_2$ planes affects the spin excitations only at low energies ($\hbar \omega \lesssim 1.5 \text{ meV}$) and in a finite temperature range.

Figure 4.28: Field dependence of the intensity at the incommensurate peak position $Q_\delta$ at different energies. The solid and dashed lines are fits to the data using Eq.(4.19) with $H_{c2} = H_{c2}(0 \text{ K}) \approx 45 \text{ T}$ and $H_{c2} = H_{c2}(22.3 \text{ K}) \approx 20 \text{ T}$, respectively. The dotted lines are linear fits.

---

**Equation (4.19)**

$$ H_{c2} = H_{c2}(0 \text{ K}) \approx 45 \text{ T} \quad \text{Eq. (4.19) with } H_{c2} = H_{c2}(22.3 \text{ K}) \approx 20 \text{ T} $$

---

**Linear fit**
Figure 4.29: Magnetic phase diagram of LSCO ($x=0.10$). Full circles are the measured values for the first order transition (FOT) line between the vortex lattice and vortex gas phases (see Chapter 2). The solid line is a fit to the data using Eq.(2.6) (sublimation), whereas the dashed line is an estimated curve for the upper critical field, using a parabolic temperature dependence $H_{c2}(T) = H_{c2}(T=0\,\text{K})\cdot(1-(\frac{T}{T_c})^2)$ with $H_{c2}(T=0\,\text{K})=45\,\text{T}$. The blue (red) arrows indicate the temperature (field) scans performed during the experiment.

$\text{(10 K} \lesssim T \lesssim 30\,\text{K)}$. The absence of an enhancement at base temperature is surprising, since it differs from the results of Lake et al. which indicates a field-induced enhancement of the elastic signal down to the lowest temperatures in a LSCO sample with the same (nominal) doping of $x=0.10$.

It is interesting to note that the vortex gas phase is very extended in underdoped LSCO, as discussed in Chapter 2. In Fig.4.29 is shown the magnetic phase diagram of LSCO ($x=0.10$) as determined from our macroscopic measurements. Red and blue arrows indicate the field- and temperature-scans presented in Section 4.4.3. It can be appreciated that our INS measurements in a magnetic field have been performed mainly in the vortex gas phase above $H_{\text{FOT}}(T)$. A possible consequence for our INS results is that the low-energy field-induced signal occurs only in the vortex gas phase (between $H_{\text{FOT}}(T)$ and $H_{c2}(T)$). This is what we observe in the temperature scans at 0.3 meV in a field of 12.5 Tesla, where a field-induced signal is observed for $T_{\text{FOT}}(12.5\,\text{T})\approx 5\,\text{K} \lesssim T \lesssim T_{c2}(12.5\,\text{T})\approx 25\,\text{K}$ (see Fig.4.25b). At 1 meV the field induced signal is smaller, but the trend is similar. The field dependence measured at $T=22.5\,\text{K}$ (see Fig.4.28) is also consistent with this interpretation. All measurements in a magnetic field have been taken in the vortex gas phase and indeed we observe a first sharp increase at low fields followed by a
Inelastic neutron scattering study of the spin excitations

Figure 4.30: Schematic representation of the swiss cheese model. a) Impurities (black circles) nucleate antiferromagnetic regions (orange) which extend in the superconducting region (blue). If percolating, these AF regions might display long range order. An increase of the fraction covered by AF regions can be achieved by b) having larger AF nucleation regions or c) having more impurities (or larger fields in the case of vortices).

Swiss cheese model

How can we understand the absence of a spin gap and the field-induced signal in under-doped LSCO? Interestingly, enhanced susceptibility below the spin gap energy $\Delta_{SC}$ and at low temperatures has been recently reported in La$_{1.85}$Sr$_{0.15}$Cu$_{1-y}$Zn$_{y}$O$_4$ upon doping with Zn$^{2+}$ ions (3d$^{10}$, $S = 0$) [302, 303] and in optimally doped LSCO ($x=0.163$) upon application of a magnetic field [278].

In the first case, non-magnetic Zn impurities induce additional sub-gap spin excitations, which are enhanced and shift to lower energies with increasing doping and eventually become static [302]. This is consistent with the swiss cheese model, which has been originally suggested in order to explain $\mu$SR experiments [304, 305]. In this model, charge carriers around Zn impurities are localized, and the remaining superconducting region resembles a slice of swiss cheese (Emmental), see also Fig.4.30.

In the second case, sub-gap spin excitations are induced by an external magnetic field. Since the superconducting order parameter vanishes in the center of each vortex (see Fig.1.4), one can consider that vortices act as impurities in the superconducting background. The area where the order parameter is strongly depressed is equal to $\pi \xi^2$, where $\xi$ is the superconducting coherence length, and is proportional to $\frac{H}{H_{c2}}$. However, this is insufficient to explain the experimentally observed field-induced sub-gap excitations, since their strength in a field much lower than $H_{c2}$ ($H/H_{c2} \approx 12$%) is similar to the strength measured in the normal state in zero field [278]. This might actually indicate that antiferromagnetic regions are nucleated around each vortex core and are not only present...
inside the vortex core. By increasing the field, more and more vortices are present in the sample and one would expect the sub-gap excitations to lower their energy and eventually become static at large enough magnetic fields. In the underdoped regime of LSCO, on the other hand, a strong field enhancement of the elastic incommensurate peaks has also been observed [279]. It has been suggested that, compared to optimally doped LSCO, in the underdoped regime the nucleated magnetic regions are larger and lead to long-range magnetic order, as schematically shown in Fig.4.30. Within this approach, one would also expect that enhanced susceptibility is observed in the vortex-fluid phase. Due to the thermal motion of the vortices, several Cu sites are visited by vortices and keep a memory of the induced magnetization. As a consequence, the effective fraction of regions covered by vortices is larger than that observed at a given instant. This might explain the fact that, in LSCO (x=0.17), we observe a spin gap only in the vortex lattice phase, where vortices are pinned and are sticked to the same sites, and not in the vortex gas phase.

Two-components model

The previously discussed swiss cheese model can account qualitatively for the results obtained by Lake et al. [278, 279]. However, in our slightly overdoped LSCO sample we didn’t observe the same strong sub-gap excitations as in optimally doped LSCO. On the other hand, magnetic vortices are present and well ordered in our sample, as indicated by our SANS investigations. If arising from AF regions around the vortex cores, this effect should be seen even in the overdoped regime. Moreover, the swiss cheese model cannot explain our results in the underdoped regime. First of all, we observed enhanced susceptibility (and no spin gap) even in the absence of vortices (zero-field) or any other impurities apart from the intrinsic disorder of the Sr ions. We observed no field-induced enhancement of the spin excitations at low temperatures, where the vortices are present in a glassy state (see Chapter 3), but only at finite temperatures (in the vortex gas phase). Alternatively, we can assume that two components contribute to the spin susceptibility in zero field. The first component is the usual susceptibility arising from interacting quasiparticles, with a spin-gap developing in the superconducting state. The second component is associated with the spin-glass phase observed only in the underdoped regime and which is present at low-energies and low-temperatures. Within this scenario, both contributions must have the same wavevector \( \mathbf{Q}_s \). It might be surprising that the susceptibility of the spin-glass is incommensurate in nature. However, such an incommensurability has been observed even in the spin-glass phase of lightly doped (non-superconducting) LSCO [211, 63], and in our experiments we didn’t observe any intensity at the commensurate \((\pi, \pi)\) wavevector. For the total susceptibility one can therefore write:

\[
\chi''_{\text{tot}}(\mathbf{Q}_s, \omega) = \chi''_{\text{sc}}(\mathbf{Q}_s, \omega) + \chi''_{\text{sg}}(\mathbf{Q}_s, \omega)
\]  

(4.24)

Within this approach, a spin gap is present even in the underdoped regime of LSCO, but cannot be observed due to the overlapping spin-glass component. \( \chi''_{\text{sc}}(\mathbf{Q}_s, \omega) \) is defined as in Eq.(4.20) with a temperature dependent spin gap

\[
\Delta_{SG}(T) = \Delta_{SG}(0) \cdot \left(1 - \left(\frac{T}{T_c}\right)^2\right)
\]  

(4.25)
Inelastic neutron scattering study of the spin excitations

Figure 4.31: Calculated dynamical spin susceptibility for underdoped LSCO in zero field. a) $\chi''_{sc}(Q_\delta, \omega, T)$ using $\Delta_{SG}(0)=2.5$ meV, $T_c=29$ K, $\Gamma=10$ meV and $\bar{\gamma}=0.8$ meV. b) $\chi''_{sg}(Q_\delta, \omega, T)$ with $E_0 \rightarrow 0$ meV and $\bar{\Gamma}=1.3$ meV. c) Total susceptibility $\chi''_{tot}(Q_\delta, \omega, T) = \chi''_{sc}(Q_\delta, \omega, T) + \chi''_{sg}(Q_\delta, \omega, T)$.

and the temperature dependence in the normal state given by Eq.(4.22). For the spin glass contribution $S_{sg}(Q_\delta, \omega)$ we have (arbitrarily) chosen a damped harmonic oscillator (DHO):

$$S_{sg}(Q_\delta, \omega) \propto \frac{\bar{\Gamma} \hbar \omega}{\left((\hbar \omega)^2 - \Omega^2\right)^2 + 4\bar{\Gamma}^2(\hbar \omega)^2}$$ (4.26)

where $\Omega^2 = E_0^2 + \bar{\Gamma}^2$. The susceptibility $\chi''_{sg}(Q_\delta, \omega)$ is then obtained by multiplying $S_{sg}(Q_\delta, \omega)$ with the Bose-factor.

In Fig.4.31 $\chi''_{sc}$, $\chi''_{sg}$ and $\chi''_{tot}$ are plotted as a function of energy and temperature assuming a spin gap of $\Delta_{SG}=2.5$ meV, $E_0 \rightarrow 0$ meV and $\bar{\Gamma}=1.3$ meV. The spin gap present in Fig.4.31a is masked by the spin glass contribution shown in Fig.4.31b. As a consequence, the total susceptibility (Fig.4.31c) doesn’t display a spin gap at low energies and low temperature. On the contrary, an enhancement of susceptibility is observed below 2 meV and 10 K. This is in qualitative agreement with our measurements in zero field. In Fig.4.32 is shown a simulation of the energy scans shown in Fig.4.18b. By comparing the two figures, one can notice that our simple model can reproduce the observed enhancement of suscep-
4.4 Underdoped La_{2-x}Sr_xCuO_4 (x=0.10)

Figure 4.32: Simulation of the experimental energy-scans shown in Fig.4.18b by the two-components model. The blue curve represents the expected gapped spectra at 2 K, the green curve is the spin-glass signal. The superposition of both contributions (red curve) leads to an enhancement below 2 meV compared to the normal state susceptibility (black curve). The same input parameters as those given in the caption of Fig.4.31 have been used.

Figure 4.33: Simulated temperature dependence of \( \chi''(T) \) for a) \( h\omega=0.3 \) meV and b) \( h\omega=3 \) meV. At \( h\omega=0.3 \) meV, the superposition of the spin glass signal (green curve) to the gapped spectra (blue curve) leads to enhanced susceptibility below about 10 K. This is in qualitative agreement with the experimental data shown in Fig.4.20d. At \( h\omega=3 \) meV, the calculate susceptibility is also consistent with the experimental results shown in Fig.4.20f.

tibility below 2 meV as being due to the superposition of a gapped spectra and a DHO spin glass component. Moreover, the simulated temperature dependence at \( h\omega=0.3 \) and 3 meV is also qualitatively consistent with the experimental data, as shown in Fig.4.33. While the zero-field data can be well described within the two-component model, it remains to show that this model can account for the observed field-dependence of the susceptibility. On the basis of our experimental data in overdoped LSCO (see Section
4.3), we assume that the spin gap vanishes in the vortex gas phase above the FOT line. For \( \mu_0H \approx 10 \) T our macroscopic measurements indicate that the FOT transition occurs around \( T_{\text{FOT}}(10 \text{ T}) \approx 5 \) K (see Fig.4.30). We therefore replaced \( T_c \) by \( T_{\text{FOT}}(10 \text{ T}) \) in Eq.(4.25) and reduced the value of \( \Delta_{SC}(0) \) from 2.5 meV to 2 meV. The temperature dependence of the susceptibility in the fluid phase between \( T_{\text{FOT}}(10 \text{ T}) \) and \( T_{c2}(10 \text{ T}) \approx 27 \) K is unknown. For simplicity, we will keep \( \chi'' \) constant in this temperature range, whereas above \( T_{c2} \) we expect the same temperature dependence as for the zero-field simulations. A further assumptions is that the spin glass signal is unaffected by the application of an external magnetic field. The idea behind all these assumptions is that the observed field-induced signal is just a consequence of the suppressed spin gap in the vortex gas phase.

\[ \Delta_{SG}(H) = \Delta_{SG}(0) \cdot (1 - H/H_{c2}) \]

---

Figure 4.34: Calculated dynamical spin susceptibility for underdoped LSCO in a magnetic field of 10 Tesla. a) \( \chi''(Q, \omega, T) \) using \( \Delta_{SC}(0) = 2 \) meV, \( T_{\text{FOT}} = 5 \) K, \( T_{c2} = 27 \) K, \( \Gamma = 10 \) meV, \( \gamma = 0.8 \) meV, \( E_0 \rightarrow 0 \) meV and \( \Gamma = 1.3 \) meV. b) Difference between the susceptibility in a field of 10 Tesla (shown in a)) and in zero field (shown in Fig.4.31c).

Figure 4.35: a) Simulated energy scans at \( T = 20 \) K in zero field and in a field of 10 Tesla (to be compared with Fig.4.27b). b) Calculated temperature-scan at \( h\omega = 0.3 \) meV for \( \mu_0H = 0 \) and 10 T (to be compared with Fig.4.25b). c) Calculated field-dependence of \( \chi'' \) at \( h\omega = 0.3 \) meV for \( T = 20 \) K (to be compared with Fig.4.28).
4.4 Underdoped La$_{2-x}$Sr$_x$CuO$_4$ ($x=0.10$)

In Fig. 4.34a is shown the calculated total susceptibility in 10 Tesla whereas in Fig. 4.34b is plotted the difference between the susceptibility in a field and in zero-field $\chi''_{tot}(10 \text{ T}) - \chi''_{tot}(0 \text{ T})$. One can notice that, despite the simplicity of our model, we can reproduce the experimental observation of enhanced susceptibility at finite temperatures and low-energies, whereas the low-energy excitations at base temperature remain unaffected by the application of a magnetic field. A more detailed comparison of simulation and experimental data is presented in Fig. 4.35. Both temperature- and energy-scans shown in Fig. 4.25b and Fig. 4.27b can be qualitatively explained within our model. Even though we cannot reproduce the exact temperature and energy dependence of the neutron intensity, the model is able to reproduce qualitatively the observed dependence upon the application of a magnetic field. In particular, both our experimental results and the calculated susceptibility indicate a field-induced enhancement below about 1.5 meV and in a finite temperature range. Furthermore, we have calculated the field-dependence of $\chi''$ at 20 K and 0.3 meV (see Fig. 4.35c). Our model predicts a sharp increases at low fields up to $H_{FOT}$ followed by a slower linear increase. Such a behavior is qualitatively consistent to the experimental results shown in Fig. 4.41.

Summarizing, the simulations presented in this section possibly indicate that our experimental data in underdoped LSCO are not much different from the results obtained in overdoped LSCO. In both cases there is a spin gap in the superconducting phase, which vanishes in the vortex gas phase. However, in the underdoped regime the spin gap is hidden by the the low-energy/temperature spin glass susceptibility which is absent in the overdoped regime.

Influence of higher $d$-wave gap harmonics on the spin gap value

The two-component simulations are consistent with the presence of a small spin gap in the underdoped regime of LSCO. However, its value ($\Delta_{SG} \approx 2.5 \text{ meV}$) is much lower than the values obtained in the optimally/overdoped regime ($\Delta_{SG} \approx 6.5 \text{ meV}$). This result is surprising, since in a fermiology approach the spin gap should roughly scale with the superconducting gap, which increases with underdoping [17]. Moreover, even the decrease of the superconducting transition temperature with underdoping ($T_c(x=0.10)/T_c(x=0.17) \approx 0.8$) cannot account for the observed doping dependence of the spin gap ($\Delta_{SG}(x=0.10)/\Delta_{SG}(x=0.17) \approx 0.4$). One might be attempted to interpret this result in terms of stripes, which have been observed when superconductivity is depressed in LSCO. The strong decrease of the spin gap in the underdoped regime could be then regarded as some kind of precursor of static stripes. However, we would like to discuss here an alternative scenario based on a fermiology approach [257]. Several ARPES measurements of the superconducting gap in underdoped BSCCO [14, 261, 306] indicate that there are deviations from a pure $d$-wave order parameter. As a result, the gap function is smoother in the vicinity of its nodes and can be described by including the second order term to the $d$-wave gap expansion:

$$ \Delta_k = \Delta_1 [\cos(k_x) - \cos(k_y)] + \Delta_2 [\cos(2k_x) - \cos(2k_y)] $$

(4.27)

Representing the wavevector dependence of the SC gap on the Fermi surface using polar coordinates, one can write

$$ \Delta(\phi) = \Delta_{\text{max}} [B \cos(2\phi) + (1 - B) \cos(6\phi)] $$

(4.28)
Figure 4.36: a) Schematic view of the Fermi surface as measured by ARPES. The incommensurate nesting vector $Q_\delta$ is indicated by the arrow. The angle $\phi$ measures the position along the Fermi surface. b) Normalized superconducting gap as a function of angle $\phi$. The solid line represents a pure $d$-wave gap function, whereas the dashed line includes the second order term ($B=0.8$, see Eq.(4.28)). The red arrows describe the energy transfers which corresponds to the lowest-lying incommensurate spin excitations.

where $\phi$ is the Fermi surface angle (see Fig.4.36a) and $B$ is a parameter which decreases with underdoping, as indicated by experiments in BSCCO. The $\cos(2\phi)$ and $\cos(6\phi)$ terms correspond to nearest-neighbors and next-nearest-neighbors interactions, respectively, indicating that the pairing interaction becomes more "long-range" with underdoping [261]. In order to visualize what happens when the gap changes shape, we have drawn in Fig.4.36b a cartoon describing the connection between the spin gap, the Fermi surface and the superconducting gap. The solid line corresponds to a pure $d$-wave gap ($B=1$) of the optimally doped region, while the dashed line corresponds to the gap in the underdoped region with higher order term ($B=0.8$). The red arrows indicate the energy transfers of the lowest-lying incommensurate spin excitations. The corresponding Fermi nesting wavevector $Q_\delta$ is shown in Fig.4.36a. One can notice that:

- In the superconducting state, no low-lying spin excitations can be created at $Q_\delta$ up to an energy corresponding to the spin gap $\Delta_{SG} \approx |\Delta(\phi_1)| + |\Delta(\phi_2)|$.

- The spin gap value at $Q_\delta$ is determined by the local SC gap close to the nodes ($\phi=45^\circ$) and not by the overall SC gap value $\Delta_{\text{max}}$.

- When the second order $d$-wave term is included ($B < 1$), the spin gap is strongly renormalized.

Beyond these qualitative considerations, numerical calculations have been performed for BSCCO [257]. In this compound the band dispersion, the Fermi surface and the SC gap have been measured in detail as a function of doping by means of ARPES, and can therefore be used as input parameters for the RPA calculations. Unfortunately, only few ARPES experiments have been reported in LSCO so far, and precise input parameters...
4.4 Underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4(x=0.10)$

Figure 4.37: $\chi''_{\text{RPA}}$ calculated numerically for a) optimally doped and b) underdoped BSCCO [257]. The strong decrease of the spin gap at the incommensurate position is due to the effect of higher order $d$-wave harmonics in the underdoped regime.

for the calculations are still missing. However, we believe that the results in BSCCO might be generic features of all HTSC. Details about the RPA treatment and numerical calculations have been shortly described in the introduction of this Chapter, and can be found in Ref.[257], as well. Here we will just present the main result of these calculations, which indicate that a strong decrease of the spin gap in the underdoped regime might be caused by the doping dependence of the $d$-wave higher harmonics. In Fig.4.37 are shown the intensity of $\chi''(\omega, q_x, \pi)$ in the $(\omega, q_x)$ plane at fixed $q_y = \pi$ for optimally doped ($T_c=87 \text{ K}, \Delta_{\text{max}}=35 \text{ meV}, B=0.96$) and underdoped ($T_c=68 \text{ K}, \Delta_{\text{max}}=41 \text{ meV}, B=0.84$) BSCCO. As a guide to the eye, contour lines are drawn at an intensity of 0.5% of the maximum intensity of the dynamical susceptibility, and can be associated to the momentum-dependent spin gap. One observes that, despite the fact that $T_c$ is reduced by only 20% with underdoping, the decrease of the spin gap $\Delta_{SG}$ at the incommensurate position $Q_5$ is much stronger (about 50%). This result could explain our results in LSCO, where a decrease of $T_c$ of about 22% is accompanied by a decrease of $\Delta_{SG}$ of about 60%.
4.5 Elastic measurements in underdoped La$_{2-x}$Sr$_x$CuO$_4$ ($x=0.10$, $x=0.12$)

As already discussed in this Chapter, it has been shown that static antiferromagnetism can be induced by a magnetic field down to the lowest temperatures in underdoped LSCO ($x=0.10$, $T_c=29$ K) [279]. In Fig.4.38 are summarized the results obtained by Lake et al. [279]:

- In zero field, incommensurate elastic peaks are present in the superconducting state and their intensity steadily increases with decreasing temperature.

- The application of a magnetic field perpendicular to the $CuO_2$ planes strongly enhances the elastic signal. The temperature dependence of the signal in a field is different from that in zero-field: it increases more rapidly just below $T_c$ and saturates below about 10 K, where the zero field signal is still evolving.

- Both zero-field and field-induced peaks are resolution limited with an in-plane correlation length $\zeta$ larger than 400 Å.

- The field-induced signal at base temperature ($T=1.9$ K) increases rapidly with small fields, and more slowly at higher fields.

The observation that the onset temperature of the field-induced signal coincides with $T_c$ lead to the idea that this signal arises from AF order confined in the vortex cores. The large value of $\zeta$, however, indicate that the AF ordered regions are much larger than the vortex core size $\xi \sim 20$ Å.

On the other hand, our INS measurements on a LSCO sample of identical nominal doping $x=0.10$ indicate that the low-energy spin excitations as well are strongly affected by the application of a magnetic field, but only in an intermediate temperature range ($10$ K $< T < 30$ K). In order to clarify the relation between the field induced spin excitations and elastic signal, we performed elastic neutron scattering measurements as a function of temperature and magnetic fields on the same $x=0.10$ sample used for our INS experiment, as well as in a $x=0.12$ sample. Experiments on LSCO $x=0.10$ and $x=0.12$ have been performed on V2/FLEX at HMI and on Rita-II at PSI, respectively.

In Fig.4.39 are shown Q-scans through one incommensurate peak at different fields and temperatures. The background was found to be temperature- and field-independent. In both samples we observed elastic incommensurate peaks in zero field at low temperature, which are enhanced by the application of a magnetic field perpendicular to the $CuO_2$ planes. However, there are several differences between the $x=0.10$ and $x=0.12$ samples. Apart from the expected difference in incommensurability ($\delta=0.108(3)$ for $x=0.10$ and $\delta=0.117(2)$ for $x=0.12$), the elastic peak in $x=0.12$ was found to be much stronger and sharper than in $x=0.10$. The peak is (almost) resolution limited for $x=0.12$, therefore implying a correlation length $\zeta$ larger than 200 Å, whereas for $x=0.10$ we estimated from the width of the peak that the correlation length is about $\zeta=100$ Å. From the comparison of the magnetic intensity to a transverse acoustic phonon measured at 190 K, it results that the integrated intensity in zero field in the $x=0.10$ sample at 2 K is roughly three times weaker than that in the $x=0.12$ sample. This result is also consistent to muon spin
4.5 Elastic measurements in underdoped La$_{2-x}$Sr$_x$CuO$_4$ ($x=0.10$, $x=0.12$)

relaxation experiments, which also indicate a ratio of about three between the magnetic signal in the two samples. A further difference between the two samples is found in the onset temperature of the elastic peak (see Fig.4.40). Due to the weak signal, for the $x=0.10$ sample we measured the temperature dependence of the neutron counts at the peak position, whereas for the $x=0.12$ sample we could measure full Q-scans and extract the integrated intensity as a function of temperature. As shown in Fig.4.40, both the zero-field and field-induced signal appear below about 20 K $^{18}$ (a temperature much lower than $T_c=29$ K) in LSCO $x=0.10$, and below 25-30 K ($\approx T_c=27$ K) in LSCO $x=0.12$. It is therefore probable that the onset temperature of the elastic peak is doping/sample dependent and not directly related to the onset of superconductivity. This result is important, since it complicates the interpretation of the field induced signal as being due to AF vortex cores. Moreover, the comparison of Fig.4.40 with Fig.4.38 seems to indicate that the results of Lake et al. in LSCO ($x=0.10$) are more similar to our results in LSCO ($x=0.12$) rather than to LSCO ($x=0.10$). This observation, together with the values of the incommensurability $\delta$ [279] and of the HTT-LTO transition temperature [307], indicates

$^{18}$To notice is that this temperature is similar to $T_{sy} \approx 15$ K obtained from our INS experiments (see Fig.4.21)
that the sample measured by Lake et al. is closer to $x=1/8$ than our $x=0.10$ sample. Fig.4.41 shows the field-dependence of the peak intensity. For the $x=0.10$ sample, a linear behavior is observed at $T=10$ K, whereas at 2 K the field-induced signal saturates above about 10 T and is well described by Eq.(4.19). The observed field dependence of the elastic signal is similar to that observed in our INS experiment (see Fig.4.28), with the difference that on IN14 we measured it at $T=22.3$ K. In that case, the low-energy field-induced signal was present only in a finite temperature range $10$ K $< T < 30$ K (see Fig.4.25) and not at base temperature. The reason for this discrepancy and the relation between the elastic and inelastic measurements is still unclear. The experiments on the $x=0.12$ sample were limited to fields lower than 9 Tesla, but the trend is similar to the $x=0.10$ sample. At 2 K the data are well fitted by Eq.(4.19), whereas the width of the peak is roughly field-independent (and almost resolution limited) apart from an anomalous minimum at 7.5 Tesla. At this point it is natural to wonder if the observed features are generic of the underdoped regime or just confined to a small doping range around $x=1/8$. It is interesting to notice that in both $x=0.10$ and $x=0.12$ samples the zero-field signal at $T=2$ K is doubled by the application of a magnetic field of about 7.5 Tesla. This observations (together with the fact that the onset temperature of the zero-field and field-induced signal is the same) is important because it indicates that the field-induced signal is coupled to the initial zero-field signal. In other words, if the zero-field elastic peak is weak (resp. strong), then the field-induced peak will also be weak (resp. strong). This is exemplified in Fig.4.42, where the field-induced signal at 2 K shown in Fig.4.41 is presented in two alternative ways. In Fig.4.42a we plotted the field-induced signal ($\text{Intensity}(H) - \text{Intensity}(0T)$) in units
Figure 4.40: Temperature dependence of the incommensurate peak intensity measured at different fields for a) LSCO ($x=0.10$) and b) LSCO ($x=0.12$). In the lower panels is shown the difference between the data with and without field of the upper panels. Lines are guides to the eye.

of the zero-field intensity in the $x=0.10$ sample. It is clear from this plot that the field-induced signal in the $x=0.12$ sample is much stronger than in the $x=0.10$ sample. This seems to be in contradiction with the interpretation of the field-induced signal as being due to AF vortex cores, since within this scenario one would expect a doping independent moment induced by a magnetic field. However, the larger moment induced in $x=0.12$ might be just due to the fact that larger AF regions are nucleated around the vortex cores, as indicated by the larger correlation length $\zeta$. The field-induced signal at 8.5 Tesla in $x=0.12$ is roughly four times larger than in $x=0.10$, therefore implying a correlation length two times larger, which is consistent to our estimations ($\zeta \approx 200 \AA$ for $x=0.12$ and $\zeta \approx 100 \AA$ for $x=0.10$). It remains to explain why this correlation length is maximal at $x=1/8$. It has been suggested that the enhanced tendency towards magnetic ordering at this special doping is due to the formation of stripes, as illustrated in Fig.4.4.

In Fig.4.42b is shown the ratio between the intensity in a field and in zero-field ($\text{Intensity}(H)/\text{Intensity}(0T)$). We also included the data of Lake et al. [279].

$^{19}$We have assumed here that the zero-field intensity in $x=0.10$ is three times smaller than in $x=0.12$, as indicated from the normalization of the magnetic intensity to a transverse acoustic phonon. In principle, this calibration is able to yield the ordered spin moment per Cu$^{2+}$ ion [279].
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Figure 4.41: Field dependence of the incommensurate peak intensity measured for a) LSCO ($x=0.10$) at $T=2, 10$ K and b) LSCO ($x=0.12$) at $T=2$ K. For the $x=0.12$ sample, the field-dependence of the width of the peak is also shown. Solid lines are fits to the data using formula (4.19) with $H_{c2}=45$ T, whereas dashed lines are linear fits. The data have been always measured after cooling in a field from above $T_c$.

Figure 4.42: Field-induced signal in LSCO ($x=0.10$) and LSCO ($x=0.12$) at $T=2$ K. a) Difference between the intensity in a field and in zero-field. b) Ratio between the intensity in a field and in zero-field. The data of Lake et al. [279] have been included in this plot. Lines are guides to the eye.
Interestingly, the data of three different sample with different doping roughly collapse on one curve. This possibly indicates that the field-induced signal is directly connected to the zero-field signal. Assuming that the zero-field signal arises from frozen spins in the spin glass phase, the field-induced signal would also be related to such a spin glass. However, very little is known about the field-dependence of this spin glass. A possibility is that in zero field the spin glass consists of disordered clusters of AF ordered frozen spins, and the application of a magnetic field is able to better order such clusters, therefore leading to a field-induced signal. It is interesting to note that at 2 K the field-induced signal saturates at high fields, whereas at 10 K the signal increases linearly up to 15 Tesla (see Fig.4.41a for $x=0.10$). This might be an indication that at 10 K the copper spins are still dynamic and need a larger magnetic field to order, whereas at 2 K they are already frozen and a smaller field is needed to order them.
4.6 Conclusions

In conclusion, we presented inelastic and elastic neutron scattering experiments in LSCO as a function of magnetic field at three different doping levels ($x=0.10$, $0.12$, $0.17$) and compared our results to other experimental works and to various theories.

**Overdoped LSCO ($x=0.17$)**

- Our INS investigations in zero-field revealed incommensurate spin excitations, which are gapped below $\Delta_{SG} \approx 6.5$ meV and $T_c=37$ K.
- The application of a modest magnetic field (5 Tesla) induces an unusually large spectral weight redistribution centered at the zero-field spin gap $\Delta_{SG}$.
- Moreover, we have some evidence that the spin gap vanishes in the vortex gas phase, indicating that the Cu-spin dynamics is surprisingly related to the dynamics of magnetic vortices.

**Underdoped LSCO ($x=0.10$, $0.12$)**

1. *Inelastic* neutron scattering study of LSCO ($x=0.10$)

- We observed incommensurate low-energy excitations at low-temperatures which are possibly related to the so-called spin glass phase observed in $\mu$SR experiments.
- Our data are consistent with the presence of a small spin gap of about $\Delta_{SG} \approx 2.5$ meV, which is hidden by the spin glass signal. The strong decrease of the spin gap as a function of underdoping can be understood quantitatively by including higher-order $d$-wave harmonics.
- The application of a magnetic field influences the low-energy ($\hbar\omega <1.5$ meV) excitations in a finite temperature range, but not at base temperature. This behavior is in qualitative agreement with a vanishing spin gap in the vortex gas phase, as observed in the overdoped sample.

2. *Elastic* neutron scattering study of LSCO ($x=0.10$, $x=0.12$)

- Elastic incommensurate peaks in zero-field have been observed in both $x=0.10$ and $x=0.12$ samples below a sample/doping dependent temperature which is *not* related to $T_c$. The peak in the $x=0.12$ sample is stronger and sharper than in the $x=0.10$ sample.
- The zero-field signal is strongly enhanced by the application of a magnetic field in both samples. The field-induced signal could arise from AF ordered regions nucleated by the vortex cores or be related to the spin glass phase present in zero-field.
It remains a challenge to understand the relation between the results of elastic and inelastic neutron scattering experiment obtained on underdoped LSCO ($x=0.10$). While the elastic incommensurate peak is increased by the application of a magnetic field down to base temperature, the incommensurate spin excitations are enhanced by a field only in a finite temperature range. In order to elucidate this point, further INS investigation in LSCO ($x=0.12$) are needed.
Chapter 5

Conclusions and outlook

This work has been dedicated to the study of the hole-doped \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) (LSCO) and electron-doped \( \text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4 \) (NCCO) high-\( T_c \) superconductors (HTSC) in high magnetic fields. We have employed different experimental techniques, mainly neutron scattering, in order to better understand both the generic and magnetic phase diagrams of HTSC. We hope and believe that the presented novel results will contribute significantly to clarify the understanding of the static and dynamic magnetic correlations present in the superconducting state and their relation/interplay to the magnetic vortices induced by an external magnetic field.

A strong doping dependence of the magnetic phase diagram (vortex matter) has been inferred by the combination of macroscopic, SANS and \( \mu \)SR experiments in LSCO single crystals (0.075 < \( x \) < 0.2). For the first time, we were able to observe an ordered vortex lattice (or Bragg glass) in both overdoped and underdoped LSCO.

In the overdoped regime (\( x=0.17 \) and \( x=0.20 \)) we discovered a field-induced transition from a hexagonal to a square vortex lattice (VL) oriented along the Cu-O bonds. This represents the first observation of an intrinsically square VL in HTSC, and is indicative of the coupling of the VL to a source of in-plane anisotropy, such as those provided by the \( d \)-wave superconducting gap or Fermi velocity anisotropies. Subsequent to our discovery, square VL’s have been observed in YBa\(_2\)Cu\(_3\)O\(_{6+x}\) and NCCO (by ourselves), indicating that these results are generic features of all HTSC. However, the physical reason for this field-induced change in symmetry from hexagonal to square has not yet been clarified, and will need further experimental and theoretical efforts. Moreover, we have strong evidence that the vortex lattice undergoes a first order sublimation transition (concomitant melting and decoupling) to a gas of vortices at temperatures well below \( T_c \).

In the underdoped regime (\( x=0.10 \)) an ordered VL could be observed only at very low fields, whereas at higher fields the VL undergoes a transition to a more disordered vortex glass state. This is mainly due to the larger out-of-plane anisotropy that makes the vortex system more susceptible to disorder and fluctuations. The increasing anisotropy with underdoping strongly influences both the second peak (related to the vortex glass transition) observed in magnetization measurements, and the sublimation transition line (leading to a much more extended vortex gas phase in the underdoped regime). Our SANS investigations of NCCO might indicate that a similar vortex glass phase is present even in electron-doped compounds.

The magnetic correlations present in the superconducting regime of LSCO were also
Conclusions and outlook

found to be strongly dependent on the doping level. Inelastic neutron scattering experiments revealed that for $x \geq 0.14$ the spin excitations are gapped in the superconducting state, whereas for $x \leq 0.13$ static incommensurate peaks are present and no spin gap could be observed so far. In overdoped LSCO ($x=0.17$) the application of a magnetic field induces a large spectral weight redistribution of the susceptibility centered at the zero-field spin gap. Moreover, the spin gap vanishes in the vortex gas phase, therefore indicating an unexpected connection between the copper-spin and vortex dynamics. In underdoped LSCO ($x=0.10$), low-energy excitations have been observed at low-temperatures, and are associated with the spin glass phase coexisting with superconductivity in the underdoped regime of HTSC. These excitations are strongly affected by a magnetic field in a finite temperature range, but not at base temperature. These results are consistent with the presence of a strongly reduced spin gap even in underdoped LSCO, which is however hidden from the signal arising from the coexisting spin glass phase. The strong decrease of the spin gap with underdoping can be understood by including higher-order $d$-wave harmonics to the superconducting gap function (this higher-order effect has been experimentally observed in Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ by means of ARPES). Moreover, we investigated the field-dependence of the elastic incommensurate peaks in the vicinity of the special doping concentration $x=1/8$, where a decrease of $T_c$ is accompanied by an enhancement of the spin glass phase. The static magnetic correlations are strongly increased by an external magnetic field in both $x=0.10$ and $x=0.12$ LSCO samples. It has been suggested that the field-induced signal arises from antiferromagnetic regions nucleated by the vortices, but it might also be that this effect is related to the spin glass phase present in zero-field. Further neutron scattering experiments are needed to elucidate this aspect.

It is difficult to draw a final unified conclusion from our experimental results, since it does not exist a detailed understanding of the many new phenomena observed, yet. In general, our findings have created several new unresolved questions. Regarding the vortex matter, it is now well established that a square vortex lattice is observed in HTSC at high enough magnetic fields, but its origin is still unclear. In order to shed some light on this aspect, one should try to investigate the vortex lattice in other HTSC compounds. In particular, experiments in detwinned or untwinned crystals at different doping levels should clarify which electronic anisotropy is driving the hexagonal-to-square transition. Experiments in both tetragonal LSCO crystals ($x > 0.22$), de-twinned LSCO and YBCO crystals and in electron-doped Pr$_{1-x}$LaCe$_x$CuO$_4$ are planned in collaboration with Prof. Keimer’s group (Max Planck Institute, Stuttgart) and Prof. Forgan’s group (University of Birmingham, UK). $\mu$SR and SANS experiments, together with theoretical calculations and Monte Carlo simulations, are still needed in order to gain more information within the vortex glass phase, where three-body correlations are likely to play an important role. This will be done in collaboration with Prof. G.I. Menon (Institute of Mathematical Sciences, Chennai, India) and Prof. S.L. Lee (University of St. Andrews, UK).

Despite our experimental effort, it remains a challenge to understand both the origin and the role played by magnetism for the occurrence of superconductivity in the cuprates. In particular, the doping dependence of the field-induced magnetic signal in LSCO should be further investigated, as well as the unusual interplay between the vortex- and Cu-spin-dynamics. Moreover, it is so far unclear to what degree the proximity of the $x=1/8$
instability influences the behavior of both vortex lattice and Cu-spin dynamics. Since the 1/8 anomaly can be further enhanced by a partial substitution of La by Nd, neutron scattering experiments on La$_{2-x}$Nd$_x$Sr$_x$CuO$_4$ ($x=1/8$) are foreseen. Such measurements, in comparison with those presented in this thesis, will allow us to better understand to what degree the proximity of the 1/8 instability influences the magnetic properties in the underdoped regime.
Appendix A

Vortex lattice in electron-doped \( \text{Nd}_2-x\text{Ce}_x\text{CuO}_4 \)

In this Appendix we present macroscopic, \( \mu \)SR and SANS measurements in electron-doped \( \text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4 \). For the first time, a vortex lattice could be directly observed in the bulk of an electron-doped HTSC. A square vortex lattice with the nearest neighbors oriented at 45° from the Cu-O bond direction has been measured, which is consistent with theories based on the d-wave superconducting gap. However, the square symmetry persists down to unusually low magnetic fields. Moreover, the diffracted intensity from the vortex lattice is found to decrease rapidly with increasing magnetic field, possibly due to the field-induced crossover to a more disordered vortex-glass phase.

A.1 Introduction

It is a matter for debate whether hole-doped and electron-doped high-\( T_c \) cuprate superconductors (HTSC) can be described within a unified physical picture [308, 309, 310]. Indeed, electron-doped HTSC have markedly different properties from hole-doped HTSC. For example electron-doped materials have comparatively low values of the superconducting transition temperature, \( T_c \), and much lower values of upper critical field, \( H_{c2} \). Furthermore, their normal-state resistivity varies as \( T^2 \) as expected for a Fermi-liquid [311, 312], and the presence of a pseudogap is still under discussion [23]. Electron-doped HTSC also appear much closer to long-range antiferromagnetic order, which may in fact co-exist with superconductivity [313, 314, 315, 316, 317, 318]. In hole-doped HTSC the \( d \)-wave nature of the order parameter is well-established. However, the evidence for the symmetry of the superconducting gap in electron doped materials (which has important implications for the pairing mechanism [310]) is somewhat contradictory. Earlier measurements of the penetration depth [319, 320] and tunneling experiments [321, 322] indicated an \( s \)-wave symmetry, whereas more recently a \( d \)-wave superconducting order parameter has been observed in phase-sensitive [6, 323] and ARPES experiments [324, 325]. The electron-doped superconductors are of particular interest in this respect, since they have a tetragonal structure (rather than orthorhombic) and therefore should show pure \( d \)-wave behaviour, unaffected by twin planes or admixture of an \( s \)-wave component associated with the orthorhombicity [6]. Recently, there has been considerable interest in the na-
ture of the vortex lattice (VL) in unconventional superconductors. It has been shown theoretically that the symmetry of the superconducting order parameter in \( d \)-wave superconductors has a profound impact on the structure of the vortex core, which exhibits four-fold symmetry \([121, 127, 126, 123, 125]\). This anisotropy is manifest in vortex interactions, particularly at high magnetic fields where the vortices are closely spaced, resulting in a square VL in an orientation with the nearest neighbour directions aligned with directions of the nodes of the superconducting order parameter (see also Section 3.3.1).

If electron-doped HTSC, e.g. \( \text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4 \) (NCCO), are indeed bulk \( d \)-wave superconductors, it is expected that their VL will undergo a similar transition as observed in YBCO \([139]\) and LSCO \([141]\). Moreover the tetragonal structure of the electron-doped compounds avoids the complicating effects (present in orthorhombic systems such as YBCO and LSCO) of twin planes, which are able to pin the VL \([181, 145, 326]\). Finally, the low values of the upper critical field \( (H_{c2} \sim 10 \text{ T} \text{ compared to } H_{c2} \sim 100 \text{ T} \text{ in hole-doped HTSC}) \) allow the investigation of the whole magnetic phase diagram by means of neutron scattering.

Electron-doped HTSC were discovered shortly after the hole-doped ones \([327, 328]\), but only recently large crystals could be produced, and to our knowledge, a SANS investigation of their VL has not been published so far. In Section A.4 we will present the first successful SANS experiments in electron-doped \( \text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4 \) single crystals, which were first characterized by means of macroscopic (Section A.2) and \( \mu \)SR measurements (Section A.3).

### A.2 Macroscopic measurements

We performed DC magnetization and AC susceptibility measurements on a piece \((m=40 \text{ mg})\) of the NCCO \((x=0.15)\) crystal used in SANS experiments (see section A.3). The experimental setup is the same as the one described in Chapter 2, and magnetic fields up to 2 Tesla were applied approximately parallel to the c-axis.

In Fig.A.1 FC temperature scans of the AC susceptibility \( \chi = \chi' + i\chi'' \) are shown. The onset of the superconducting transition temperature in zero field is \( T_{c}^{\text{onset}} = 25 \text{ K} \), whereas the main transition occurs at \( T_c = 17.4 \text{ K} \) \((\Delta T_c = 3.2 \text{ K})\). As for LSCO, the peak in \( \chi''(T) \) rapidly shifts toward lower temperatures with increasing magnetic field, and is interpreted as the signature of the sublimation transition from a vortex lattice to a vortex gas.

FC and ZFC temperature scans of the magnetization \( M(T) \) are shown in Fig.A.2. At low fields \((\mu_0 H < 0.1 \text{ T})\) the magnetization curves are conventional, with the diamagnetic signal appearing in the superconducting state. At larger fields, however, we observe an anomalous temperature dependence of \( M(T)/H \). Starting from high temperatures, the FC magnetization gradually increases with decreasing temperature. At intermediate fields \((0.1 \text{ T} < \mu_0 H < 0.4 \text{ T})\) the FC magnetization decreases in the superconducting states, but then increases again at lower temperatures. At high fields \((\mu_0 H > 0.5 \text{ T})\), on the contrary, \( M(T) \) is monotonically increasing with decreasing temperature. At \( \mu_0 H = 2 \text{ T} \) the data can be well fitted by the Curie-law:

\[
\chi = \frac{M}{H} = \frac{C}{(T + \Theta_c)} \quad (A.1)
\]
A.2 Macroscopic measurements

Figure A.1: a) Real and b) imaginary part of the AC susceptibility $\chi(T)$ measured in NCCO at different magnetic fields between 0 T and 2 T after field cooling. In c) a zoom of a) is shown for $\mu_0 H=0$ T.

Figure A.2: a) FC and b) ZFC $M(T)/H$ curves in NCCO. The red dotted line in a) is a fit to the Curie law of the FC data measured at 2 T.
with $\Theta_c = 3$ K. This typical paramagnetic behavior is most probably due to the contribution of the Nd ions to the total magnetization, and already indicates a much stronger magnetic response compared to the LSCO compound. ZFC magnetization curves are also anomalous at large magnetic fields ($\mu_0 H \geq 0.1$ T), as shown in Fig.A.2b. To notice is also the large difference in magnitude between the FC and ZFC diamagnetic signals at low temperatures and fields. For example, at $\mu_0 H = 10$ mT, the ratio between the FC and ZFC diamagnetic signal at 3 K is less than 2%. Assuming that the ZFC signal represents the maximal diamagnetic signal of the Meissner phase and that the diamagnetic signal vanishes in the normal state, we can estimate that only a small fraction of the applied field $H$ is expelled after FC.

We also performed isothermic ZFC $M(H)$ measurements at different temperatures (see Fig.A.3). Only one peak is seen in the magnetization loops, as in underdoped LSCO (see Fig.2.5) \(^1\). Above the irreversibility field $H_{irr}$, defined as the meeting point between the increasing and decreasing branches of the hysteresis loop, one can notice that the magnetization continues to increase linearly with field. This effect is also due to the paramagnetic signal of the Nd ions, which was visible in the $M(T)$ data shown in Fig.A.2a (see also Eq.(A.1)).

A.3 $\mu$SR measurements

The magnetic properties of the NCCO crystal used in SANS experiment have been also investigated by means of zero-field muon spin relaxation measurements \(^2\). In Fig.A.5a zero-field $\mu$SR time spectra at different temperatures are shown. Such spectra represent the time evolution of the asymmetry parameter $A(t)$, defined by Eq.(4.23). The data can

\(^1\)However, several groups could observe a second peak in the magnetization loops of optimally doped $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$ ($x=0.15$) \([329, 103]\).

\(^2\)These measurements have been performed by A. J. Drew, University of St. Andrews, at the Paul Scherrer Institute, Switzerland \([330]\).
A.3 μSR measurements

Figure A.4: a) Zero-field μSR time spectra of the asymmetry $A(t)$ at three temperatures ($T=1.7\, K < T_{N2}, T_{N2} < 25\, K < T_{N1}$ and $T=75\, K > T_{N1}$). The green lines are exponential fits to the data. b) Longitudinal field μSR time spectra measured at $T=1.6\, K$ at different fields.

Figure A.5: Zero-field relaxation rate $\Lambda$ as a function of temperature in NCCO. $T_{N1}$ and $T_{N2}$ represent the temperatures where the Cu and Nd spins are slowed down. The line is a guide to the eye. In the inset is shown the low-temperature region.

be fitted by a single exponential function $A_0 exp(-\Lambda \cdot t)$ where $A_0$ is the initial asymmetry and $\Lambda$ is the relaxation (depolarization) rate. If the nuclear spins are randomly oriented, a Gaussian-like depolarization is expected. As shown in Fig.A.5a, muon-spin precession is observed already at high temperatures. With decreasing temperature, $\Lambda$ increases steadily and saturates below $T_{N1} \approx 45\, K$. This temperature is associated with the ordering of the
Cu spins, as observed in neutron scattering experiments [331]. At lower temperatures, below \( T_{N2} \approx 6 \) K, the depolarization rate further increases because of the slowing down of the Nd spins. These results are consistent to Ref.[317, 318]. By applying a longitudinal field, one observes that the relaxation gets smaller with increasing field. This is consistent with a glassy phase with static moments, since for fluctuating moments one would not expect any field-dependence of the relaxation.

### A.4 SANS measurements

We report here the first direct observation of a VL in NCCO (\( x=0.15 \)) [164]. Our SANS experiments were performed on the instrument D22 at the Institut Laue Langevin, France, using neutrons with a wavelength \( \lambda_n = 6 \) Å - 20 Å. Crystals of NCCO were grown in a mirror furnace and annealed as described in ref. [332]. Samples with two shapes were investigated in applied magnetic fields up to 0.4 T. In high fields, a cylinder of 5 mm diameter, consisting of two nearly aligned crystals, was mounted in a cryostat with the magnetic field direction bisecting the two \( c \)-directions and at \( 7^\circ \) to each of them. Single crystal plates of \( \sim 1.5 \) mm thickness were used at low fields: this required a long neutron wavelength and the smaller thickness reduced the effects of neutron absorption in the sample. They were mounted so that the \( c \)-direction was within \( \sim 2^\circ \) of the applied field, which was approximately parallel to the incident beam. There were no significant differences in the VL diffraction patterns obtained from different samples in measurements at the same field. The samples were oriented so that the \{100\} crystal directions were vertical/horizontal.

In Fig.A.6 we show the VL diffraction patterns obtained at different magnetic fields after subtraction of a background measured above \( T_c \). For magnetic fields larger than 50 mT, the VL clearly has square coordination. The nearest-neighbor directions are parallel to the \{110\} crystal directions, which correspond to the nodes of the superconducting order parameter, in agreement with predictions for \( \delta \)-wave HTSC [121, 127, 126, 123, 125]. Further evidence for a square VL is given by the position of the Bragg spots in reciprocal space. The relationship between the magnetic field \( H \) and the magnitude \( q \) of the wave vector is given by Eq.(3.15). As usual, \( q \) is obtained by fitting the tangential average of the neutron signal with a Gaussian (Fig.A.7a). As expected, the position of the peak shifts to higher \( q \) with increasing magnetic field (confirming the VL origin of the neutron signal), and the extracted values of \( \sigma \) are consistent with a square VL at all field measured (see Fig.A.7b). As discussed in the previous section, the comparison of FC and ZFC magnetization measurements indicates that the value of the trapped flux at low fields is within \( \sim 1\% \) of the applied field (\( B \approx H \)). Hence, flux expulsion has an insignificant effect on the value of the \( q \)-vector.

As shown in Fig.A.6, the VL orientation is not perfect at any field. However, the orientation becomes rather more disordered as the field is decreased below 50 mT and at

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\(^3\)There is an optimal sample thickness \( t \) in SANS experiments since the scattered intensity from the VL is proportional to \( t \), whereas the absorption depends exponentially on \( t \). The scattered intensity is then proportional to \( t \cdot \exp(-\alpha t) \) which has a maximum at \( t_m = 1/\alpha \). Due to the large absorption cross section of Nd, in NCCO (\( x=0.15 \)) \( \alpha = 0.058 \cdot \lambda_n \) mm\(^{-1} \) (\( \lambda_n \) in Å) which gives an optimal thickness \( t_m \approx 1 \) mm at \( \lambda_n \approx 18 \) Å.
Figure A.6: SANS diffraction patterns measured at \( T \approx 2.5 \) K, after field cooling from 30 K in \( \mu_0 H = 20 \) mT, \( \mu_0 H = 50 \) mT, \( \mu_0 H = 100 \) mT and \( \mu_0 H = 200 \) mT. A background taken at \( T = 30 \) K has been subtracted. The \{100\} directions correspond to the Cu-O bond directions in the CuO\(_2\) planes.

At 20 mT, the intensity distribution becomes ring-like. A similar distribution of intensity at low magnetic fields has been observed in LSCO (see Fig.3.5a), and was attributed to the superposition of diffraction patterns from various domain orientations of hexagonal coordination, since the value of \( \sigma \) at low fields was consistent with that of a hexagonal VL (see Fig.3.8b). Moreover, by rotating the c-axis 10° away from the field direction, the degeneracy of the VL system could be reduced, and the hexagonal coordination of the VL in LSCO was confirmed (see Fig.3.9b). In NCCO, on the contrary, the values of \( \sigma \) at low magnetic fields are still consistent with that of a square VL (see Fig.A.7b), and measurements at 20 mT with the c-axis rotated 10° away from the field direction showed a similarly disordered ring-like pattern. The square VL is robust even upon the rotation of the c-axis away from the field direction. In Fig.A.8 are shown SANS pattern measured at 50 mT, which represent the lowest field where a clear square VL is observed, as a function of angle \( \Theta \). For \( \Theta = 20^\circ \) the pattern resembles very much the one observed at \( \Theta = 0^\circ \), whereas at \( \Theta = 30^\circ \) it becomes more disordered and the scattered intensity lies on an ellipse as expected for the uniaxial anisotropy of the crystal. To notice is that the value of \( \sigma \) is consistent to a square VL for all \( \Theta \). It is surprising that the vortex lattice maintains square coordination without being aligned to the crystal lattice.

In addition to the \( d \)-wave scenario for square VL coordination, one should also consider
Figure A.7: a) Tangential average of the neutron signal for $\mu_0 H = 50$ mT and $\mu_0 H = 100$ mT at $T \approx 2.5$ K. b) Field dependence of $\sigma$: the horizontal lines indicate the expected values for hexagonal and square VL.

Figure A.8: SANS diffraction patterns measured at $T=2$ K, after field cooling from 30 K in $\mu_0 H = 50$ mT. In the left panel the c-axis is oriented along the field-direction, whereas in central and right panels the c-axis was rotated away from the field direction by an angle of 20 and 30 degrees, respectively. The growth direction and long axis of the sample lay 15° counterclockwise to the vertical and may be associated with the stronger intensity in these quadrants. The ellipses drawn in the right panel have axial ratio of $\cos(30^\circ)$.

the effects of Fermi surface anisotropy. An appropriate theory for large-$\kappa$ materials well below $B_{c2}$ is London theory with non-local corrections [131], which has been extensively used to account for VL phase transitions in the borocarbides [154, 173]. If both these effects are present [150], we would expect the strong angular variation of the $d$-wave gap to dominate over the usually smaller variation of the Fermi velocity. ARPES experiments together with band structure calculations [160] indicate that NCCO and YBCO both have nearly isotropic hole-like Fermi surfaces with a slight four-fold distortion oriented so as to favor the observed square VL orientation. In overdoped LSCO, on the other hand, the Fermi surface is electron-like [161, 159] with a square shape oriented at 45° to that of the
Figure A.9: a) VL intensity as a function of magnetic field. The dotted line is the expected field dependence taking into account of core effects and the (possible) field-dependence of the penetration depth (see text). b) Temperature dependence of the VL intensity at 50 mT.

other two compounds. Moreover LSCO exhibits a pronounced anisotropy in the Fermi velocity. Both the shape of the Fermi surface and the Fermi velocity anisotropy would indicate (via nonlocal effects) the VL orientation actually observed in LSCO [141]. However, both d-wave and nonlocal effects should only be important at fields which are substantial fraction of $B_{c2}$, when the inter-vortex spacing is comparable to the coherence length. For instance, d-wave calculations indicate that the square symmetry has lower free energy than the hexagonal one at applied fields greater than $0.15 B_{c2} \approx 1.2 \text{T}$ [125], or than $B_{c2}/\kappa \approx 0.4 \text{T}$ [123] (taking the values of $B_{c2} \approx 8 \text{T}$ [333], penetration depth $\lambda_L \approx 1250 \text{Å}$ [334] and coherence length $\xi \approx 60 \text{Å}$ [333], therefore $\kappa = \lambda_L/\xi \approx 20$). Although the characteristic fields in electron-doped HTSC are generally lower than in the hole-doped materials, these estimates are too large to explain our results. Hence, our observation of a square VL in NCCO down to very low magnetic fields is rather surprising, unless another source of anisotropy in the CuO$_2$ planes is present. One candidate for this is the Cu antiferromagnetic correlations, whose characteristic wavevector [314] coincides in direction with the VL reciprocal lattice wavevector. We have confirmed by µSR experiments (see Fig.A.5) that enhanced AF correlations do exist in our sample below $T_{N1} \approx 45 \text{K}$ in agreement with Ref. [317, 318].

In addition to the VL structure and orientation, a matter of great interest is the variation of the diffracted intensity with field. In our NCCO samples, the rocking curves of the VL diffracted intensity were found to be so broad that we could not measure them. This result is consistent with the values of $\lambda_L$ quoted in µSR measurements [334], which imply very broad rocking curves to give the integrated intensity implied by Eqns. (3.3) and (3.7). We therefore measured the intensity as a function of field at a fixed sample angle. Assuming that the rocking curve width remains constant with field, the measured intensity is proportional to the integrated intensity. In contrast with the London prediction, see Eq. (3.9)), we observe a strong field-dependence of the scattered intensity (see Fig.A.9a). The intensity decreases rapidly with field and becomes unmeasurable above $H \approx 0.4 \text{T}$,
which is well below $H_{c2}$. If represented by a power law, this variation has an exponent of about -2. (If the rocking curve becomes narrower with increasing field, then the field dependence of the VL intensity is even more extreme than this. If it becomes broader, then this may form part of the explanation of the field dependence).

We consider two possible intrinsic reasons for the fast decrease of the VL intensity with field, as discussed in Chapter 3. The first effect is due to the finite size of the vortex core, which leads to deviations from the London predictions at large values of the wavevector $q$. The second one, which is expected to be a specifically $d$-wave effect, is the variation of the penetration depth with field, which affects the intensity via $I_{hh} \propto \lambda_{c}^{-1}$ (see Eqs.(3.8)). Including both effects, we expect approximately the following variation of the VL intensity with field (see also Eq.(3.12)):

$$I_{hh} \sim H^{-\frac{1}{2}} \left(1 + \beta \frac{H}{H_0}\right)^{-4} \exp(-4\sqrt{\pi}(H/H_{c2})^{\frac{1}{2}})$$

(A.2)

For NCCO we used $\beta = 7 \cdot 10^{-2}$ (as for YBCO [134]) and $(H_0 \sim H_{c2}/\sqrt{2\kappa} = 0.28$ T).

As can be seen in Fig.A.7c, we cannot explain the strong field-dependence of our experimental data by these intrinsic effects. This rapid decrease of the VL intensity is more likely to be due to a transition to a more disordered vortex system (e.g. short distance vortex displacements). Indeed such a transition from a Bragg glass (or quasilattice) to a vortex glass (or entangled solid) has been predicted theoretically [43, 84, 38], and was associated with the onset of a second peak in magnetisation measurements [103]. These authors reported that in NCCO the onset field at low temperatures is about 30 mT. SANS experiments revealed a rapid loss of diffracted intensity with increasing field even in the isotropic (K,Ba)BiO$_3$ system [191]. The VL intensity is completely lost at the onset of the second peak in magnetisation, suggesting that the peak effect is indeed related to a change in the vortex structure. In hole-doped BSCCO a similar strong decrease of the VL intensity with increasing field was attributed to a crossover from a 3D to a 2D vortex system [51]. In NCCO, however, such a dimensional crossover is expected to occur at much higher magnetic field $H_{c2D} = \Phi_0/(\gamma s)^2 \sim 13$ T (which is unphysical, since it is far above $H_{c2}$), where $s \sim 6$ Å is the distance between CuO$_2$ planes and $\gamma \sim 21$ [311] is the anisotropy. In this respect, NCCO seems to be similar to underdoped LSCO [45], in which $\mu$SR measurements have given clear evidence of a field-induced crossover to a more disordered, but still three-dimensional VL (see section 3.4.3). In addition to this effect, another possible cause of VL disorder at high fields is the field-induced antiferromagnetic order which has been observed in electron-doped materials [313, 335, 316] and which may also cause distortions of the VL. However, it has also been suggested that in Nd$_{2-x}$Ce$_x$CuO$_4$ the field induced AF moment is due to spurious effects associated with the Nd moments [336, 337].

Finally, in Fig.A.9b, the temperature dependence of the VL intensity at 50 mT is plotted. The intensity linearly decreases with increasing temperature and vanishes around $T_c$. The linearity at low temperatures might be a further indication for $d$-wave superconductivity in the bulk of electron-doped HTSC. Moreover, we couldn’t observe any sizable change in the value of $\sigma$ at high temperatures, indicating the absence of a square-hexagonal transition as a function of temperature.
A.5 Conclusions

In conclusion, we have made the first SANS observation of the VL in electron-doped NCCO, which is the first tetragonal HTSC to be investigated by SANS. Our main results are:

- The observation of a square vortex lattice down to very small fractions of $H_{c2}$, which is contrary to theoretical expectations.
- The observation of an unusually fast decrease of the VL intensity with increasing magnetic field, which is probably due to a crossover to a more disordered vortex state.

Both effects are possibly a fascinating consequence of the coexistence of antiferromagnetism and superconductivity in electron-doped materials. Our macroscopic and $\mu$SR measurements confirmed that indeed magnetism is much stronger in electron-doped than in hole-doped HTSC, and that it has a strong impact on the magnetic phase diagram of NCCO. Further studies in other electron-doped compounds and at different doping levels are needed to clarify the unresolved questions of our experimental investigations.
Bibliography

[1] H. K. Onnes, Communication from the Physical Laboratory of the University of Leiden 120b (1911).


[14] J. M. Harris et al., Anomalous superconducting state gap size versus $T_c$ behavior in underdoped $\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{Dy}_x\text{Cu}_2\text{O}_{8+\delta}$, Physical Review B 54, 15665 (1996).


[19] D. Rubio-Temprano et al., Oxygen and copper isotope effects on the pseudogap in the high-temperature superconductor $\text{La}_{1.84}\text{Ho}_{0.04}\text{Sr}_{0.15}\text{CuO}_4$ studied by neutron crystal-field spectroscopy, Physical Review B 66, 184506 (2002).


[21] A. G. Loeser et al., Excitation gap in the normal state of underdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_3\text{O}_{8+\delta}$, Science 273, 325 (1996).


[26] S. Nakamae et al., The ground state of heavily-overdoped non-superconducting $\text{La}_{2.2}\text{Sr}_x\text{CuO}_4$, Physical Review B 68, 100502(R) (2003).

[27] C. Niedermayer et al., Common phase diagram for antiferromagnetism in $\text{La}_{2.2}\text{Sr}_x\text{CuO}_4$ and $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_3\text{O}_6$ as seen by muon spin rotation, Physical Review Letters 80, 3843 (1998).


[51] R. Cubitt et al., Direct observation of magnetic flux lattice melting and decomposition in the high-$T_c$ superconductor $Bi_2Sr_2CaCu_2O_{8+x}$, Nature 365, 407 (1993).


[59] R. Gilardi, Neutron scattering study of the high-temperature superconductors $La_{2-x}Sr_xCuO_4$ and $Bi_{2}Sr_{2}CaCu_{2}O_{8+x}$, Diploma thesis, ETH, 2001.
[60] M. Braden et al., Coupling between superconductivity and structural deformation in $La_{2-x}Sr_xCuO_4$, Physical Review B 47, 12288 (1993).

[61] R. Yoshizaki et al., Superconductivity of $(La_{1-x}Sr_x)_2CuO_4$ ($0 \leq x \leq 0.03$), Physica C 156, 297 (1988).


[68] Q. Li, M. Suenaga, T. Kimura, and K. Kishio, Reversible magnetic properties of $La_{2-x}Sr_xCuO_4$ single crystals with $0.05 < x < 0.10$, Physical Review B 47, 11384 (1993).


[95] R. Gilardi et al., *A small angle neutron scattering study of the vortex matter in La\(_{2-x}\)Sr\(_x\)CuO\(_4\) (x=0.17)*, International Journal of Modern Physics B 17, 3411 (2003).


[100] I. M. Sutjahja, A. A. Nugroho, M. O. Tjia, A. A. Menovsky, and J. J. M. Franse, *Peak effects and the solid vortex phase of a T*-phase SmLa\(_{0.3}\)Sr\(_{0.2}\)CuO\(_4-\delta\) single crystal*, Physical Review B 64, 134502 (2001).

[101] M. Okuya et al., *The second peak effect observed in Sr-overdoped (La\(_{1-x}\)Sr\(_x\))\(_2\)CuO\(_4-\delta\) single crystals with various oxygen deficiencies delta*, Physica C 271, 265 (1996).


[141] R. Gilardi et al., Direct evidence for an intrinsic square vortex lattice in the overdoped high-$T_c$ superconductor La$_{1.83}$Sr$_{0.17}$CuO$_4$, Physical Review Letters 88, 217003 (2002).


[184] S. L. Lee et al., *Evidence for flux-lattice melting and a dimensional crossover in single-crystal Bi$_{2}$Sr$_{1.85}$CaCu$_{2}$O$_{8+\delta}$ from muon spin rotation studies*, Physical Review Letters 71, 3862 (1993).

[185] S. L. Lee et al., *Evidence for two-dimensional thermal fluctuations of the vortex structure in Bi$_{2}$Sr$_{1.85}$CaCu$_{2}$O$_{8+\delta}$ from muon spin rotation experiments*, Physical Review Letters 75, 922 (1995).


[201] H. F. Fong et al., Neutron scattering from magnetic excitations in $Bi_2Sr_2CaCu_2O_{8+x}$, Nature 398, 588 (1999).


[208] B. Keimer et al., Resonant spin excitations in $YBa_2Cu_3O_{6+x}$ and $Bi_2Sr_2CaCu_2O_{8+x}$, Journal of Physics and Chemistry of Solids 60, 1007 (1999).

[209] S. Wakimoto et al., Observation of incommensurate magnetic correlations at the lower critical concentration for superconductivity in $La_{2-x}Sr_xCuO_4$ ($x=0.05$), Physical Review B 60, R769 (1999).

[210] S. Wakimoto et al., Direct observation of a one-dimensional static spin modulation in insulating $La_{1.95}Sr_{0.05}CuO_4$, Physical Review B 61, 3699 (2000).


[218] C. H. Lee et al., *Energy spectrum of spin fluctuations in superconducting La$_{2-x}$Sr$_x$CuO$_4$ (0.1<x<0.25)*, Journal of the Physical Society of Japan 69, 1170 (2000).


[223] R. Gilardi et al., *Spin dynamics in the mixed phase of La$_{2-x}$Sr$_x$CuO$_4$ (x=0.10, x=0.17), Physica B 350, 72 (2004).


[258] H. Ding et al., Evolution of the Fermi surface with carrier concentration in Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$, Physical Review Letters 78, 2628 (1997).

[259] A. A. Kordyuk et al., Doping dependence of the Fermi surface in (Bi,Pb)$_2$Sr$_2$CaCu$_2$O$_{8+δ}$, Physical Review B 66, 014502 (2002).


[269] K. Kudo et al., Field-induced magnetic order in La$_{2-x}$Sr$_x$CuO$_4$ ($x = 0.10, 0.115, 0.13$) studied by the in-plane thermal conductivity measurements, Physical Review B 70, 014503 (2004).


[299] R. Gilardi et al., *Spin dynamics in the mixed phase of La$_{2-x}$Sr$_x$CuO$_4$ (x=0.10, x=0.17)*, proceeding SCNS Workshop (2002), http://whisky.ill.fr/Events/ONSITE/SCNS/.


[305] T. Adachi et al., Muon-spin-relaxation and magnetic-susceptibility studies of the effects of nonmagnetic impurities on the Cu-spin dynamics and superconductivity in La$_{2-x}$Sr$_x$Cu$_{1-y}$Zn$_y$O$_4$ around $x = 0.115$, Physical Review B 69, 184507 (2004).


[308] N. C. Yeh, Recent advances in high-temperature superconductivity, Bulletin of Associations of Asia Pacific Physical Societies (AAPPS) 12, 2 (2002), cond-mat/0210656.


[313] H. J. Kang et al., Antiferromagnetic order as the competing ground state in electron-doped Nd$_{1.85}$Ce$_{0.15}$CuO$_4$, Nature 423, 522 (2003).


[316] M. Fujita, M. Matsuda, S. Katano, and K. Yamada, Magnetic-field-effect on static antiferromagnetism in electron-doped superconductor, Pr$_{1-x}$LaCe$_x$CuO$_4$ ($x=0.11$ and 0.15), cond-mat/0311269 (2003).
[317] T. Uefuji et al., Coexistence of antiferromagnetic ordering and high-$T_c$ superconductivity in electron-doped superconductor $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$, Physica C 357-360, 208 (2001).

[318] I. Watanabe et al., Muon spin relaxation study on magnetic properties of $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$ around a boundary between the magnetically ordered and superconducting states, Physica C 357-360, 212 (2001).


[320] A. Andreone et al., Temperature dependence of the penetration depth in $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ superconducting thin films, Physical Review B 49, 6392 (1994).

[321] Q. Huang et al., Tunneling evidence for predominantly electron phonon coupling in superconducting $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$ and $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$, Nature 347, 369 (1990).


[329] A. A. Nugroho et al., Vortex state in a $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ single crystal, Physical Review B 60, 15379 (1999).


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   *Square vortex lattice at unusually low magnetic fields in electron-doped Nd_{1.85}Ce_{0.15}CuO_4*
   http://xxx.lanl.gov/abs/cond-mat/0410250

   *Unusual interplay between copper-spin and vortex dynamics in slightly overdoped La_{1.85}Sr_{0.17}CuO_4*

   *Spin dynamics in the mixed phase of La_{2-x}Sr_xCuO_4 (x=0.10, x=0.17)*

   *Field-induced hexagonal to square transition of the vortex lattice in overdoped La_{1.8}Sr_{0.2}CuO_4*

   *Direct observation of the flux-line vortex glass phase in a type-II superconductor*

6. A. Schnyder, A. Bill, C. Mudry, **R. Gilardi**, H.M. Rønnow and J. Mesot
   *Influence of higher d-wave gap harmonics on the dynamical magnetic susceptibility of high temperature superconductors*
   http://xxx.lanl.gov/abs/cond-mat/0405607

2003

   *A small angle neutron scattering study of the vortex matter in La_{2-x}Sr_xCuO_4 (x=0.17)*
   Field-induced transition from hexagonal to square vortex lattice in $La_{1.83}Sr_{0.17}CuO_4$
   ILL Annual Report 2002 - Scientific Highlights (April 2003) p.52
   http://www.ill.fr/AR-02/

2002

   Direct Evidence for an intrinsic square vortex lattice in the overdoped high-$T_c$ superconductor $La_{1.83}Sr_{0.17}CuO_4$

    Spin dynamics in the mixed phase of $La_{2.3-x}Sr_xCuO_4$ ($x=0.10$, $x=0.17$)
    http://whisky.ill.fr/Events/ONSITE/SCNS/

    Doping dependence of the tetragonal-orthorhombic phase transition in the superconducting compound $La_{2-x}Sr_xCuO_4$