Magnetoresistance of the
Electron-Underdoped Cuprate
Superconductor \( \text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4 \)

Master Thesis

Ahmed Alshemi

Supervisor
Prof. Dr. Rudolf Gross
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Fakultät für Physik
TECHNISCHE UNIVERSITÄT MÜNCHEN
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Ahmed Alshemi

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List of Abbreviations

HTSC  High-temperature cuprate superconductors
BCS  Bardeen, Cooper, Schrieffer theory
214  Compounds related to RE$_2$CuO$_4$
AFM  Antiferromagnetism
ADMR  Angular dependent magnetoresistance
NIS  Neutron inelastic scattering
$\mu$SR  Muon spin rotation
$h$-doped  hole-doped
$e$-doped  electron-doped
NCCO  Nd$_{2-x}$Ce$_x$CuO$_{4+\delta}$
LCCO  La$_{2-x}$Ce$_x$CuO$_{4+\delta}$
PCCO  Pr$_{2-x}$Ce$_x$CuO$_{4+\delta}$
PCO  Pr$_2$CuO$_4$
PLCCO  Pr$_{1-x}$LaCe$_x$CuO$_{4+\delta}$
$T'$ and $T$  214 crystal structures
RCO  R$_2$CuO$_4$
LSCO  La$_{2-x}$Sr$_x$CuO$_{4+\delta}$
YBCO  YBa$_2$Cu$_3$O$_7$
NCO  Nd$_2$CuO$_4$
a.c.  Alternating current
MR  Magnetoresistance
n-MR  negative magnetoresistance
RE  Rare-earth element
SC  Superconductivity
NSC  Non superconducting
O(1)  Oxygen in in-plane lattice sites
O(2)  Oxygen in out-of-plane lattice sites
O(3)  Oxygen at apical sites
$T_c$  Superconductivity transition temperature
$T_N$  Néel temperature
BZ  Brillouin zone
FS  Fermi surface
Chapter 1

Introduction

The phenomenon of superconductivity, in which the electrical resistance of certain materials completely vanishes at low temperature, is one of the most interesting and sophisticated in condensed matter physics. It was first discovered by the Dutch physicist Heike Kamerlingh Onnes, who was the first to liquify helium (which boils at 4.2 K at standard pressure). In 1911 Kamerlingh Onnes discovered the phenomenon of superconductivity while studying the resistance of metals at low temperature [1].

Since that time it has long been a dream of scientists working in the field of superconductivity to find a material that becomes a superconductor at room temperature. A discovery of this type would revolutionize every aspect of modern day technology such as power transmission and storage, communication, transport and the fast computers. All of these processes would be faster, cheaper and more energy efficient. This has not been achieved to date. However, in 1986 a class of materials was discovered by Bednorz and Müller that led to superconductors that we use today on a bench-top with liquid nitrogen to cool them [2]. The discovery by Bednorz and Müller that superconductivity occurs in the La-Ba-Cu-O system at a temperature as high as 38 K was the beginning of a new era in the field of superconductivity. Soon after their discovery it was clarified that the superconducting phase in the La-Ba-Cu-O system is $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$, a complex non-stoichiometric cuprate which is characterized by the presence of CuO$_2$ planes in the crystal structure. The discovery initiated an intence effort to find other cuprate superconductors with even higher critical temperatures. Within a year YBa$_2$Cu$_3$O$_{7-x}$ ($T_c=93$ K), the first cuprate superconductor with a transition temperature well above the boiling point of nitrogen was found [3].

Despite all that steadfast effort to the study of the cuprates since they where discovered, the pairing mechanism responsible for high $T_c$ superconductivity is still obscure. The electron-phonon interactions that adequately explain pairing for the conventional superconductors within the BCS theory [4, 5] would require phonon frequencies incompatible with the material stability in order to produce materials with high critical temperatures [6].

Up till now the mechanism of superconductivity in cuprates is still not understood. It is believed that the copper-oxygen planes are the main motif for superconductivity, and that
have Mott insulating parent compounds with an antiferromagnetic ground state. Upon hole- or electron doping the antiferromagnetism vanishes and superconductivity appears together with the metallic state.

The electron-doped cuprate Nd$_{2-x}$Ce$_x$CuO$_4$ superconductor with a $T_c$ of 24 K is our main focus in this thesis. It was discovered by J. Akimitsu et al. of Aoyama-Gakuin University in late 1988 [7, 8] and by Y. Tokura et al. of University of Tokyo in early 1989 [9],[10] respectively.

Later, it was known that the antiferromagnetic order is more stable against doping than in hole-doped cuprates [11].

From this perspective and in an attempt to investigate whether the AFM and SC exist as an intrinsic phase separation or in a microscopic coexistence.

In this thesis, the electronic transport measurements were held in order to probe the spin subsystem which is coupled to the charge carriers. This give us an important chance to understand the normal state properties of the high $T_c$ cuprates, where many researchers believe that magnetic interactions, which are observed in all cuprates, may play an important role in the pairing mechanism. For that, $c$-axis magnetotransport studies on Nd$_{2-x}$Ce$_x$CuO$_4$ single crystals with doping levels $x$ close to the border of the superconducting dome on the under doped side of the phase diagram were carried out. Two doping levels were chosen for the present study : a non superconducting composition with $x=0.10$ and the superconducting level with $x=0.12$.

The master thesis is structured as follows:

In the second chapter, the Nd$_{2-x}$Ce$_x$CuO$_4$ superconductor is introduced by presenting the phase diagram, the crystal structure, the spin structure, the electronic band structure and an over view is given on the previous magnetoresistance measurements for the same samples with very lightly doping concentrations and also for similar compounds Nd$_{2-x}$Ce$_x$CuO$_4$, Pr$_{1-x}$LaCe$_x$CuO$_{4±δ}$, etc.

The third chapter, part 1: an overview on crystal growth, crystal preparation for the measurements (i.e sample contacting, fixation, rotation stages) and the experimental techniques.

The fourth chapter the results of the experiments are presented and analysed.

The fifth chapter presents a summary of the main results and a brief outlook is given.
Chapter 2

Electron-doped cuprate superconductors

2.1 Phase diagram (electron-doped VS hole-doped cuprates)

One approach to the understanding of HTSC and its mechanism is to study the similarity and differences between the hole- and electron-doped sides of the cuprate phase diagram. The chemical substitution of heavy ions and/or modification of the oxygen content (either by reduction or oxidation) leads to the doping of the Cu-O planes by holes or electrons.

The phase diagram of cuprates has two regions with distinct properties, one is the antiferromagnetic (AF) region and the other is superconducting (SC). The phase diagram is asymmetric with respect to the type of doping. The AF region extends to a much higher doping level for electron-doped \((e^-\text{doped})\) than for the hole-doped \((h^-\text{doped})\) cuprates while superconductivity is "stronger" for \(h^-\text{doped}\) ones. The number of doping holes \((p)\) or electrons \((n)\) per Cu ion is a fundamental parameter in the physics of cuprates, since the critical temperature for the superconductivity, \(T_c\), is found to be maximum around \(p(n) \approx 0.15\) (the optimal doping) and strongly doping-dependent. Hence, the cuprate samples are called overdoped, underdoped, or optimally doped, when \(p(n)\) is higher, lower or at the optimal concentration (corresponding to the maximum \(T_c\)) respectively, as we can see in (Fig. 2.1) which shows a schematic phase diagram for \(p^-\text{ and } n^-\text{ doping for two crystallographically similar cuprate compounds.}\)

In the \(La_{2-x}Sr_xCuO_4\) (LSCO) system which represents the hole-doped side the trivalent substitution of \(La^{3+}\) by \(Sr^{2+}\) ions dopes the Cu-O planes by holes. Doping \(p\) is equal to \(x\) over a wide range of \(x\) values (up to \(x \approx 0.30\)). For the other side (electron-doped) the \(Nd(Pr)_{2-x}Ce_xCuO_4\) (NCCO, PCCO) system, a replacement of \(Nd^{3+}\) by \(Ce^{4+}\) leads to electron doping of the Cu-O planes.

With no doping the cuprates are poor conductors. They are believed to be an example of the so-called Mott insulator. The Mott insulator is fundamentally different from a conventional band insulator. In the latter the conductivity is blocked by the energy gap
determined by interaction between electrons and the periodic potential of metal ions. By contrast in Mott insulators the conductivity is blocked primarily by the electron-electron Coulomb repulsion in a half filled conduction band. To minimize the potential energy electrons try to be as far as possible from each other and each electron is localized at a minimum of the ionic potential. To generate a current we have to create doubly occupied sites which costs too much energy. So the electrons are "frozen" in their respective positions. There is, however, a virtual hopping between the sites that decreases the kinetic energy without increasing the potential energy too much. Thus there is a magnetic (antiferromagnetic) ordering in the system (two electrons on the neighboring sites must have opposite spins).

With an increase of the doping or temperature, they destroy first all the Mott insulating state, and, thereby AF ordering. However, as we discuss in the case of $n$-doped cuprates the AF ordering seems to be more robust than the Mott state and even exist in the SC region of the phase diagram. After losing the antiferromagnetic ordering (AF insulator phase) due to doping a new fairly-good conducting phase comes to existence which is commonly known as the pseudogap phase. This phase we could consider as a conductor
2.1 Phase diagram (electron-doped VS hole-doped cuprates)

but with properties much different from the properties of the usual conductors (generally Fermi liquids). The importance of this region comes because at this specific region the battle between two different types of order (Mott insulator and SC) takes place. The problem is that the nature of this transition and the new phase that arises is very poorly understood. Most of the theoretical effort in the field has been aimed to understand and describe that new phase. But theory needs well established, reliable experimental data in order to get more information about the properties of this mysterious state. That was a problem in the past, in particular, because, of rather poor crystal quality typical of cuprate crystals. Fortunately, recently high-quality crystals have become available, which triggered further experiments in the field.

At increasing the doping in the pseudogap state is gradually suppressed, whereas superconductivity becomes stronger. $T_c$ becomes higher and higher until it reaches maximum (optimal doping) and then starts decreasing. Like in conventional superconductors the charge carriers are electron pairs (Cooper pairs). According to the conventional BCS theory after forming the condensate of Cooper pairs there is a uniform (hence the name “s-wave”) gap in the $K$-space electron spectrum. In HTSC there is still a gap but it is anisotropic with $d$-wave symmetry. After modifying the BCS theory to incorporate those differences it turns out that it is a pretty good description of the $d$-wave SC. However the microscopic mechanism of the superconductivity is still uncertain. It is clear that the electron-phonon interaction that is responsible for the conventional SC is way to weak to do the same in the cuprates the temperature is too high (this is one of the reasons for the surprise in 1986, nobody was expecting such a high $T_c$).

In electron-doped cuprates we notice that the AFM state exists over a wider doping range, as compared to hole-doped cuprates and the superconducting region is much narrower. Also the pseudogap boundary is rather blurred and its location is quite uncertain (AFM overlap with SC. Furthermore electron-doped cuprates possibly display lower SC $T_c$ unlike what we see in the hole-doped cuprates. So, the intuitive way to visualize the robustness of the AFM order in the electron-doping phase diagram is the spin-dilution picture. While the hole doping introduces carriers to the O $p$-orbitals, the electron doping takes place in the Cu $d$-orbital. The resulting mobile spinless [13].

The asymmetry of some other important differences $p$- and $n$-doped cuprates can be pointed out as the follows:

- In $n$-doped cuprates, for the doping at which $T_c$ is maximum (optimal doping), the resistivity is metallic for both the $ab$-plane and $c$-axis. These particularly distinguish them from many $p$-doped cuprates, which show a non-metallic $c$-axis resistivity.
- The electrical resistivity of the normal state of electron-doped cuprates is known to follow a $T^2$ behavior from $T_c$ up to room temperature, while the hole-doped cuprates show linear resistivity.
- The magnetic field, to suppress superconductivity in $n$-type cuprates, is about 10 T. Therefore the normal state of $n$-doped cuprates is easily accessible in experiments. In
contrary, in optimally \( p \)-doped cuprates, enormously strong magnets are needed to access the normal state.

- In the \( n \)-doped side \( T_c \) is of the order 20 K [10] and therefore significantly smaller than in \( p \)-doped cuprates [2].
- The nature of the superconducting state. While the pairing symmetry of the order parameter in the hole-doped materials is well established to be of the \( d \)-wave symmetry, the situation is far from being settled in the electron-doped materials [14].

### 2.2 Crystal structure

It is well known that in solids, the crystal structure is determined by character of chemical bonding and a number of other related physical properties. Even small changes in structure can considerably change the electronic properties of a solid. In HTSC, the investigation of the crystal structure and its dependence on temperature, pressure and composition plays an important role for understanding such systems and predicting possible ways to synthesize new superconducting compounds.

Many topologically different types of crystal structure of layered copper-oxide superconductors have been studied. These various structures can be divided into several families depending on the type of packing of a small number of structure elements, that is, perovskite-like copper-oxygen \( \text{CuO}_2 \) layers (ex: \( \text{YBa}_2\text{Cu}_3\text{O}_{7-x} \)) and rock salts (ex: \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \)) or fluorite blocks (ex: \( \text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4 \)).

All of the cuprate high temperature superconductors have tetragonal unit cells, or are at least orthorhombic. Some of the simpler cuprate crystal structures are found in the 214 family of cuprate superconductors. The crystal structure varies slightly depending on the choice of rare earth element (RE). For example, a hole-doped 214-compound \( \text{LSCO} \) (\( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \pm \delta \)), crystallizes in the \( T \) phase (Fig. 2.2), whereas an electron-doped 214-compound, \( \text{NCCO} \) (\( \text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4 \pm \delta \)) which is our main focus in this thesis, it crystallizes in the \( T \) phase.

The crystal structure of electron-doped cuprates is illustrated in the left panel of Fig. 2.2. It consists of an alternating stack of \( \text{CuO}_2 \) and rare earth oxide layers. The electronic properties are governed primarily by the \( \text{CuO}_2 \) sheets, whereas the rare-earth oxide layers act as a charge carrier reservoir and spacer for the \( \text{CuO}_2 \) sheets. Hence, the single crystals are characterized by a strong in-plane to out-of-plane electronic anisotropy. The electron-doped 214 compounds crystallize in the body-centered tetragonal (b.c.t) \( T \) structure with the space group \( \text{I}_4/\text{mnm} \). The adjacent \( \text{CuO}_2 \) sheets are shifted with respect to each other along the in-plane diagonal by \( (a/2, a/2) \), and hence, the conventional unit cell is double in the \( c \)-direction. The oxygen atoms occupy two distinct sites: the so-called \( \text{O}(1) \) site, which is the position within the \( \text{CuO}_2 \) sheets, and the \( \text{O}(2) \) site, which denotes the out-of-plane oxygen in the rare-earth sublattice. The rare-earth sublattice \( \text{Ln}_2\text{O}_2^{2+} \) has the fluorite structure with an oxygen coordination number of 8 for the rare earth ion \( \text{Ln} \),
leaving vacant the apex position directly below and above the Cu ions. This is a potential impurity site, which might be occupied partially [15, 16] during crystal growth. Because of steric considerations, the apex occupation leads to a strong, local lattice distortion with considerable influence on the physical properties of the electron-doped compounds.

The arrangement of the O(2) oxygen and hence, the planar coordination of Cu in the electron-doped cuprates is unique, whereas the related hole-doped 214 compound \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) crystallizes in the more common T structure [Fig. 2.2 (right)]. The T structure is characterized by the apex occupation of the O(2) oxygen. In this case the Cu site is sixfold coordinated with oxygen, forming an octahedral environment. The \( \text{La}_2\text{O}_2^{2+} \) layers show rock salt structure with an oxygen coordination number of nine for the La ion. Compared to the T structure, the \( \tilde{T} \) structure is stable at low temperatures. There are no structural phase transitions [66], which influence the electronic properties. The crystallization in the T or \( \tilde{T} \) structure depends on the rare earth ionic radii. 214 crystals with smaller lanthanides favor the \( \tilde{T} \) structure, whereas the compound \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) cannot be crystallized in the \( \tilde{T} \) structure. Concerning single-crystal growth, the system \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) has an additional problem: growth experiments have shown that the dopant Ce is precipitated on the surface and cannot be incorporated into the crystal structure. Thus, although \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) might be the ideal compound for comparative studies with \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \), this system can only be grown in the thin film form.
2.3 Spin structure

Once the positional periodicity of magnetic ions is given by the crystal structure, one of the main features of interest in terms of magnetic correlations is the pattern of spin orientations, often called “magnetic structure”. The magnetic interactions in rare-earth (R) cuprate \( \text{R}_2\text{CuO}_4 \) (RCO) systems have been the subject of extensive studies for various reasons. First and foremost, the R cuprates, which become superconducting under electron doping, have a simpler structure than the hole-doped superconducting cuprates. Second, rare-earth cuprates exhibit novel magnetic properties involving both the Cu and R subsystems. The undoped cuprates are antiferromagnetic insulators where the \( S = \frac{1}{2} \) Cu\(^{+2}\) spins order at high temperatures, typically near or above room temperature. The in-plane Cu\(^{+2}\) exchange interactions are much stronger than along the c-axis, and, thus, the magnetism is two-dimensional in nature. With doping, the materials lose the Cu long-range magnetic order and become high-\( T_c \) superconductors. However, the Cu\(^{+2}\) magnetic moments and thier energetics are still present, and the essential role these quantum spin fluctuations plays in the superconducting state has stated depend on plenty of experimental studies where the electronic properties of the cuprates superconductors have shown that the electron interaction with the spin-fluctuations play an essential role in their anomalous normal state properties [17]. Therefore one could expect that the same interactions may

\[ \text{Figure 2.3: Relative orientations of spins in the chemical unit cell of Nd}_2\text{CuO}_4. \ (a) \text{Noncollinear Phase I } 75 < T < 275 \text{ K and Phase III } T < 30 \text{ K; (b) \text{Phase II } 30 < T < 75 \text{ K; (c) Collinear (Phase I & Phase III) induced by field along [110] direction from Noncollinear (Phase I & Phase III); (d) Collinear (phase II) from noncollinear (phase II). Here the red arrows shows the Cu}^{2+} \text{ ions and the blue ones are Nd}^{3+} \text{ ions.} \]
be responsible for superconductivity in cuprates.

For the Cu$^{2+}$ spins, the central feature that controls many aspects of all the oxide materials is the strong copper-oxygen bonding, which results in a layered Cu-O crystal structure. In the undoped parent materials this strong bonding leads to an electrically insulating antiferromagnetic ground state. The exchange interactions within the layers are much stronger than between the layers, and typically an order of magnitude more energetic than the lattice dynamics, that give rise to both two-dimensional magnetic properties and highly anisotropic superconducting behaviour. The associated spin dynamics and magnetic ordering of the Cu ions are, thus, influenced by this two-dimensional nature. With electronic doping, long-range antiferromagnetic order for the Cu is suppressed and metallic behavior and then superconductivity appears, but strong antiferromagnetic spin correlations still persist in this regime [18].

![Figure 2.4: Magnetic structure of Nd$_2$CuO$_4$:](image)

Figure 2.4: Magnetic structure of Nd$_2$CuO$_4$: (a) in the noncollinear antiferromagnetic phase at zero field and (b),(c) in the collinear phase above the spin-flop transition with a field aligned parallel to the Cu-Cu and Cu-O-Cu direction, respectively, according to Lavrov et al. [19]. Only Cu atoms of two adjacent layers (red solid and red empty circles) are shown.

In the Nd$_{2-x}$Ce$_x$CuO$_4$ structure where the main focus lies on it is this thesis, the magnetic moments of the mother compound Nd$_2$CuO$_4$ are large and interactions between the Rare earth elements ions and localized Cu$^{2+}$ spins give rise to a rich set of properties and signatures of magnetic order.

In Nd$_2$CuO$_4$ has spin reorientation transition takes place due to the competition between various interplanar interactions which arise because of the rapid temperature dependence of the Nd$^{3+}$ moment below about 100 K [20]. The Cu$^{2+}$ spins first order in the noncollinear AF structure phase I below $T_{N1} = 275$ K Fig. 2.3. On further cooling, the Cu$^{2+}$ spins reorder into the noncollinear structure at $T_{N2} = 75$ K phase II. At $T_{N3} = 30$ K the Cu spins experience another reorientation into phase III which has the same noncollinear order as phase I. At $T_{N2}$ all the Cu$^{2+}$ spins rotate by 90° about the c axis, and they rotate back to their original direction at $T_{N3}$. 
Below $T_{N3}$ there is a substantial induced staggered moment on the Nd$^{3+}$ moments, which become ordered at low T (1 K).

The magnetic moments of the Cu-spins lie within the CuO - planes with a relatively strong intralayer coupling pointing along the Cu-O-Cu directions ([100], [010]) \cite{21, 22}. For NCCO the intralayer exchange, $J = 126$ meV is much stronger than $J_{\perp}=\num{5.10^{-3}}$ meV, giving rise to a very anisotropic magnetic behavior \cite{23, 24}. The Cu$^{2+}$ spins order in a noncollinear structure, as it is sketched in Fig. 2.4 (a). The spin orientation is found to alternate between adjacent layers \cite{25, 26}, as indicated by solid and empty red circles for the Cu atoms in Fig. 2.4. For low doping $x \leq 0.03$, a spin-flop transition from the noncollinear into the collinear Cu-spin structure above a critical magnetic field of approximately was observed in magnetization and neutron scattering experiments \cite{23–27}.

### 2.4 Origin of the conducting bands

The parent compounds of all high-$T_c$ cuprates are insulators. In fact, one of the early surprises was that superconductivity could arise in such presumed insulating materials. For example, Nd$_2$CuO$_4$ is insulating and becomes metallic and superconducting only after a sufficient amount of electrons is added via chemical substitution of Ce$^{3+}$ for Nd$^{4+}$.

The undoped compounds are not standard band-theory insulators, but so-called Mott insulators. The Cu$^{2+}$ ions contain nine 3d-electrons out of a maximum of ten, this means that the orbital with the highest energy is half-filled. Due to the tetragonal crystal field this is the 3d$_{x^2-y^2}$ orbital. Naively, one would think that a half-filled electronic band at the Fermi level is the signature of a metal. In a Mott insulator, however, the carriers are highly localized, and there is strong energy cost $U$ for two carriers to be on one site due to the Coulomb repulsion between them. The carriers are thus immobile, and the system is insulating. In the band picture, the half-filled band is split into a filled lower Hubbard band and an empty upper Hubbard band, with a gap energy of $U$. The building

![Figure 2.5: building blocks of cuprate high temperature superconductors (CuO$_2$ planes).](image-url)
blocks of cuprate high temperature superconductors are CuO$_2$ planes like the one shown above. At zero doping, the cuprates are antiferromagnets, meaning the spins on adjacent copper atoms point in opposite directions. They are also insulators, and they are driven to be insulators by strong electron correlations. Judging by the number of electrons per unit cell, these materials are expected to be metals, but because coulomb repulsion between electrons is strong and unscreened, the electrons are instead localized and it is an insulator. You will stand as far apart from each other as possible to avoid smelling each other. As electrons are removed from the CuO$_2$ planes, the remaining electrons have more freedom to move around without bumping into their stinky neighbor, and eventually, the system becomes uncorrelated enough to behave like a Fermi liquid, which is basically a normal metal. Antiferromagnetism is suppressed with a small amount of hole doping. This happens because the dopant holes act magnetic and sit on an oxygen between two copper atoms, and now the two spins that want to point opposite from their neighbor don’t know what to do. Though antiferromagnetism is quickly destroyed, memories of the antiferromagnetic parent compound exist over larger portions of the phase diagrams via short-lived excitations which have a similar periodicity. Some theories explaining superconductivity in the cuprates cite excitations related to antiferromagnetism.

![Figure 2.6: The density of states near the Fermi energy for a cuprate band structure](image)

The situation in the cuprates is slightly more involved, due to the presence of the O$^{2-}$ ions and their valence orbitals. The relevant O$^{2-}$ orbitals are the 2p, i.e., the in-plane orbitals which lie along the Cu-O-Cu directions. The O 2p and Cu 3d$_{x^2-y^2}$ orbitals are shown in Fig. 2.6. The energy level of the oxygen 2p band happens to be in between the upper and lower Hubbard bands of the copper 3d$_{x^2-y^2}$ orbital. In other words, it
is easier to remove an electron from the filled oxygen orbitals than to remove one from the half-filled copper orbital. The Fermi energy lies between the oxygen 2p band and the upper Hubbard band, whose separation is the charge transfer gap $\Delta$, i.e., the energy it takes to transfer an electron from the O$^{2-}$ ion to the Cu$^{2+}$ ion. The undoped cuprates are thus more properly called charge-transfer insulators. The three-band Hubbard model (one copper orbital and two oxygen orbitals) is believed by many to contain all of the relevant low-energy electronic interactions in the CuO$_2$ sheet \[28\]. Because this model is still quite involved, many theorists use the simpler one-band Hubbard model, treating the charge transfer gap $\Delta$ as the effective value for $U$. The one-band Hubbard model is written as

$$H = -t \sum_{\langle j, l \rangle, \sigma} C_{j\sigma}^{\dagger} C_{l\sigma} + U \sum_j n_{j\uparrow} n_{j\downarrow} - \mu \sum_j (n_{j\uparrow} + n_{j\downarrow})$$

(2.1)

where $C_{j\sigma}^{\dagger}, C_{1\sigma}$, are the creation, annihilation and number operators, respectively, for an electron with spin $\sigma$ (up or down) at site $j$. The first term is the kinetic energy: It describes the destruction of an electron of spin $\sigma$ on site $l$ and its creation on site $j$ (or vice-versa). The symbol $\langle j, l \rangle$ emphasizes that hopping is allowed only between two sites which are adjacent. The second term is the interaction energy. It goes through all the sites and adds an energy $U$ if it finds that the site is doubly occupied. The final term is a chemical potential which controls the filling. We refer to the situation where the filling is one electron per site as “half-filling” since the lattice contains half as many electrons as the maximum number (two per site). Studies of the Hubbard model often focus on the half-filled case because it exhibits a lot of interesting phenomena (Mott insulating behavior, anti-ferromagnetic order, etc.)

### 2.5 Review of previous MR experiments of the electron-underdoped cuprates

For both sides of the phase diagram, hole- and electron-doped, the main target of experimental activities has been to investigate the evolution of the electronic state from an AFM Mott insulator to a high temperature superconductor. To investigate the electronic structure of cuprates, during the last five years several experiments in high magnetic fields on hole-doped compounds have found evidence for the existence of a well established Fermi surface (FS) in the normal state at different doping levels \[29\].

In the frame work of Toni Helm’s, master thesis and PhD thesis \[30, 31\] at Walter-Meissner Institute, the angle- dependent magnetoresistance oscillations and Shubnikov-de Haas oscillations were observed for electron-doped cuprates (at different dopings) for the first time. The observed oscillation in the physical quantities such as resistivity and magnetization, have proven a powerful tool for investigating the fermiology of such compounds \[29, 32\]. Since these effects can be related directly to the Fermi surface (FS)
topology in the bulk of the crystals, those observations reveal the existence of a well defined FS in NCCO and provide quantitative information on the FS properties [33, 34]. The inter-layer magnetotransport studies on a set of Nd$_{2-x}$Ce$_x$CuO$_4$, \([x = 0.13; 0.15; 0.16; 0.17]\) single crystals, covering the superconducting part of the phase diagram of electron-doped cuprates, have been performed in high magnetic fields.

The striking difference between the SdH frequencies for the compositions \(x = 0.15\) and 0.16 on one hand, and for \(x = 0.17\) on the other implies that the superstructure gap is either strongly suppressed or absent at \(x = 0.17\). The complete suppression of the gap and corresponding transformation of the FS would be consistent with suggestions of quantum phase transition at a doping level between 0.16 and 0.17, see, e.g., [35–40].

The obtained data [41–45] shows a high-frequency quantum oscillations revealing a reconstructed Fermi surface for various doping levels in the superconducting overdoped regime, \(x \geq 0.15\) of NCCO with a demonstration of the orbital effects. At the same time for underdoped cuprates the small Fermi surface sections are revealed with a suppression of SdH oscillations and the orbital effects. This behaviour in underdoped regime is possibly related to spin ordering effects like those observed recently in magnetotransport of electron-underdoped cuprates [46]. Understanding how the electrons in the antiferromagnetic underdoped regime of NCCO are coupled to the spin system is a very important.

For strongly underdoped NCCO samples with \(x=0.033\) and 0.025 which was published by Wu et al [47]. The anisotropic MR and magnetization have been studied by rotating the magnetic field within the CuO$_2$ plane. A giant anisotropy was observed in a form of spin-flop transition by applying the field along [100] and [110] directions. Additionally, a sharp feature was found in high-field angular sweeps which was suggested to be an evidence to support the spin-flop transition of Nd$^{+3}$.

Similar data was obtained for lightly electron-doped Pr$_{1.3-x}$La$_{0.7}$Ce$_x$CuO$_4$ for \(x=0.01\) [19]. Where both the in-plane and out-of-plane resistivity are surprisingly sensitive to spin reorientation. The results also show coupling between the charge carries and magnetism at such strongly underdoped cuprates.

MR of strongly underdoped, \(x = 0.10\) and 0.05, were performed by Toni Helm [31], for NCCO samples in magnetic field oriented parallel to the CuO$_2$-layers. A step-like feature was recorded for both samples, for \(B \parallel [100]\) and \(B \parallel [110]\), respectively as can be seen in Fig. 2.8. These observed features represent a spin-flop transition induces by a certain magnetic field.

Also, MR oscillations with a fourfold symmetry was recorded for NCCO 0.05 see Fig. 2.9. by rotating the magnetic field within the CuO$_2$-layers. A step-like feature with a hysteretic behavior was observed at \(T = 1.4\) K and 4.2 K, respectively. The observed features were attributed to a spin-flop transition in the Cu$^{2+}$ spin-structure, where at a certain critical field, \(B_{SF}\), the Cu$^{2+}$ spin-lattice undergoes a spin-flop transition from the noncollinear to collinear antiferromagnetic ordering which is already well known for the undoped NCO.
Figure 2.7: (a) In-plane and out-of-plane resistivity of PLCCO ($x=0.01$) single crystals. The MR in $\rho_c$ (b) and $\rho_{ab}$ (c) measured for the in-plane magnetic field $B \parallel [110]$ [19].

Figure 2.8: Interlayer MR for the field oriented parallel to the conducting layers, (red) $B \parallel [110]$ and (black) $B \parallel [100]$, for $x=0.10$ (a) and 0.05 (b) at 1.4 K.
Figure 2.9: Angle-dependent interlayer MR of an $x = 0.05$ sample for field rotations in the plane parallel to the conducting layers at $T = 1.4$ K (top) and $T = 4.2$ K (bottom).
Comparing with other samples which have quite similar crystal structure as NCCO, a similar set of measurements were performed on Pr\textsubscript{2-x}Ce\textsubscript{x}CuO\textsubscript{4} thin films. The transport measurements were used in order to detect the antiferromagnetic phase in PCCO [48–50]. The measurements results in that AFM phase persists up to \( x = 0.16 \) [19]; indicates significant coexistence between antiferromagnetism and superconductivity. The angular magnetoresistance oscillations in Pr\textsubscript{2-x}Ce\textsubscript{x}CuO\textsubscript{4} are observed to decrease with temperature and doping. This behaviour of magnetoresistance anisotropy was attributed to the long-range order antiferromagnetism.

From the previous measurements, it is clear that the Magnetoresistance provides new insight into the coupling between the charge carriers and the background magnetism in all underdoped cuprates.

In this thesis, we are going to measure the interlayer magnetoresistance as a function of the strength and orientation of the applied field in order to probe the spin subsystem which is coupled to the charge carriers. This is expected to give us an important chance to understand the normal state properties of the high \( T_c \) cuprates, where many researchers believe that magnetic interactions, which are observed in all cuprates, may play an important role in the pairing mechanism. For that, \( c \)-axis magnetotransport studies on Nd\textsubscript{2-x}Ce\textsubscript{x}CuO\textsubscript{4} single crystals with doping levels \( x \) close to the border of the superconducting dome on the under doped side of the phase diagram were carried out. Two doping levels were chosen for the present study: a non superconducting composition with \( x = 0.10 \) and the superconducting level with \( x = 0.12 \). In this thesis we are going to investigate whether superconductivity and AF is coexist in the electron under-doped Nd\textsubscript{2-x}Ce\textsubscript{x}CuO\textsubscript{4} for \( x = 0.10 \) and 0.12.
Chapter 3

Sample preparation and experimental techniques

In this part an overview is given on the preparation and preliminary characterization of the single-crystalline samples that were used in the studies carried out in this thesis. Thereafter, details on different measurement techniques and setups that were applied for investigating the high-field properties of these samples will be presented.

3.1 Crystal growth

Single crystals of NCCO, characterized by the world’s best quality, were grown by using the traveling solvent floating zone technique (TSFZ) see [51–54] in our WMI crystals lab. Single crystals of NCCO with $x=0.10$ and 0.12 have been provided by Alma Dorantes and Andreas Erb.

3.1.1 Advantages of the TSFZ method

The preference for the TSFZ arises from the advantage to grow crystals from materials which undergo an incongruent melting. Thus, only the growth by the TSFZ technique enables the control of the correct stoichiometry in our NCCO crystals. Moreover, no crucibles are required and contamination and reaction with crucible materials are avoided. Using the TSFZ technique, two rods of the material (seed and feed) are melted via an optical setup of mirrors. In order to ensure homogeneity of the melt, both rods are rotated against each other. Via a vertical movement of the rods, the melting zone travels through the rod. This leads to a directional solidification and crystallisation. Impurities usually stay within the melt or stay at the surface of the crystal and can therefore easily be removed. Control and optimization of the crystal growth is done by supervising and adapting the growth parameters such as the type of gas and pressure of the gas, speed of rotation, speed of pulling, composition of the rods and temperature of the melt. The most obvious advantages of travelling solvent floating zone technique can be summarized as follows [55]:
• No crucible is necessary.
• Both, congruently and incongruently melting materials can be grown.
  • The relatively high thermal gradient on the crystallization front decreases the chance for constitutional supercooling and allows for a more rapid growth of incongruently melting ones.
  • Oxides melting at temperatures as high as $2500^\circ C$ can be grown.
  • The growth can be conducted at high pressure (up to 10 atm) and in specific atmospheres.
  • Solid solutions with controlled chemical composition can be prepared.
  • Finally, in contrast to a crucible method, a steady state can be achieved. This is beneficial for crystal growth of doped materials (with a distribution coefficient different than 1) and for incongruent crystallization [53, 56, 57].

3.1.2 Preparation of the feed rods

The first step to crystal growth is the preparation of a polycrystalline feed rod of the desired material. High quality feed rods are characterized by their homogeneity and uniformity in density and shape. Furthermore, phase purity and homogeneous distribution of the dopant are important as otherwise the small solvent zone changes continuously its composition during the growth process along the vertical feed rod, thereby affecting the stability of the floating zone and the crystallization. Rods of high density avoid the penetration of a larger quantity of liquid flux into the feed rod and hence, lead to a well defined upper solid-liquid interface. The sequence of the actual growth of a 214 phase material is shown in Fig. 3.1.

At first, the 214 phase is prepared by a solid state reaction. For this purpose the corresponding rare earth oxide and CuO powders with a purity of 99,99% are mixed together according to the desired stoichiometric composition. The phase is generated via a fivefold pre-reaction of the mixture at temperatures of $900^\circ C, 920^\circ C, 950^\circ C, 980^\circ C$ (twice) for 10 h in air. After each cycle the powder is homogenized using a ball mill. The multiple calcination steps improve the homogeneity. After the calcination the phase formation is checked by X-ray powder diffraction.

After the pre-reaction the powders are ready to be packed in a rubber tube which has the required diameter and length. This is firstly done by hand, which requires extra care from the experimentalist. For a better compact state, the rod is pressed in a hydrostatic press at 2,000 kg/cm². Then it is prepared for the next stage, sintering.

The purpose of sintering is to eliminate any remaining porosity from the powders. This is done at temperatures near the melting point. If any porosity is found in the feed rod, there is high probability of bubble formation in the melt zone or penetration of the melt into the feed rod. Bubbles in the rod can join together and then collapse, which puts the stability of the molten zone in high danger. Another side effect can occur when the
Figure 3.1: Illustration of the single crystal growth of 214 high temperature superconductors. The growth process starts with the generation of the floating zone of an appropriate composition by melting a flux pellet(a). The growth velocity usually amounts to 0.5 mm/h. After a few days stable conditions are obtained. In (b) a snapshot after 7 days of successful growth is provided, illustrating the 6 mm thick polycrystalline feed rod with a small region of flux penetration, the stable floating zone of 4.5 mm in length with a slightly concave crystallization line and the grown single crystal rod with its shiny surface. (c) The thick polycrystalline feed rod with a neck, indicating the starting point of the growth process, the grown crystal rod with its shiny surface and the eutectically solidified residual flux on the top, Taken from[51].

bubbles stay in place and form defects in the crystal [55].

The sintering process is performed in a rotational lifter in O\textsubscript{2} at temperatures of 1050\textdegree C, 1100\textdegree C and 1200\textdegree C for 5 hours each. The bar is rotated inside the alumina tube to obtain the straight and uniform density rod. It is also lifted up and down continuously for temperature regularity.

Finally the flux material is also prepared from a combination of powders, further pre-reacted and annealed at 1010\textdegree C for 10 hours in air. The correct calculation of the composition is vital to grow a single and uniform crystal. Size and volume are also important matters which plays a role in the stability of the molten zone and the interface.

3.1.3 Annealing treatment

Electron-doped crystal in their as-grown state are not superconducting even at optimal doping. They are antiferromagnetic insulators with a Néel temperature \((T_N)\), between 125-160 K. The superconducting transition appears only after an appropriate temperature
treatment. Since the crystals grown by the TSFZ technique do not show SC in their as-grown state, all crystals, which were used in the experiments reported in this thesis, were annealed under the same conditions to reduce the apical oxygen content. These crystals received a standard reduction treatment in an argon gas flow at $900 - 950^\circ C$, close to the decomposition temperature [51], for 20h followed by moderate cooling (50-100 K/h) to room temperature to achieve sharp superconducting transitions in the zero-field temperature curves.

3.2 Sample contacts, fixation and measurement geometry

3.2.1 Sample contacts

3.2.1.1 Silver Paste (EpoTek) contacts

Transport measurements all rely on making good electrical contacts to the material. The contacts for NCCO crystals are generally made by hand under an optical microscope. Annealed platinum wires of 20 $\mu$m diameter were attached to the sample surface manually by using silver paste (for the electrical contacts the two-component silver paste EpoTek H20E conducting epoxy was used), see Fig. 3.2 (b). The contact resistances achieved by simply drying under ambient conditions are in the range of several hundred ohms up to kiloohms. Therefore, the contacted crystals, including the wires, were cured by a heat treatment in three stages, first by annealing the samples at 140$^\circ$ for ~40 min which is needed for solidifying EpoTek where it does not solidify at room temperature. In a second stage, we anneal it at a much higher temperature 500$^\circ C$ for at least 1h in air, after that

Figure 3.2: (a) Mounted and contacted two NCCO samples (0.3×0.3×1) mm$^3$ for the interlayer transport measurements under the optical microscope. (b) Platinum wires of 20 $\mu$m diameter attached to the sample two sides by silver paste then the sample is fixed by Stycast (blue) to a sapphire substrate.
3.2 Sample contacts, fixation and measurement geometry

the contacts are reinforced with a little bit of silver paste and annealed again at 140°C for \( \sim 40 \) min. This whole thing leads to low-ohmic contact resistances of \( \leq 5 \Omega \) which is crucial for us to get sufficiently low-noise signals. It has to be noted that this short heat treatment does not affect notably the oxygen content of the samples, since the oxygen mobility at these temperatures is very small in \( n \)-doped cuprates see [58, 59].

It turned out that the samples felt a strong torque mainly induced by the neodymium moments in a magnetic field. Therefore, Stycast 2850 FT, prepared with Catalyst 24 LV, was used as a glue to fix the samples on a sapphire plate. Sapphire is chosen because of its perfect electrical insulating and good thermal conducting properties. Stycast 2850 FT is characterized by a high thermal conductivity, small thermal expansion and a low viscous consistency, before it hardens.

It should be noted that before attached the platinum wires the sample two side surfaces was polished by grinding them mechanically. To avoid stress, induced by the fixation onto the sapphire, upon cooling to liquid \( ^4 \)He temperatures, the samples were embedded in pillows made from blue Stycast 2850FT that kept the bar slightly above the sapphire surface. To guaranty a homogeneous current distribution, the silver contacts were attached so that the full sides of the crystal bar were fully covered.

3.2.1.2 Gold contacts

A second technique was tested in order to get low-ohmic contact resistances which is crucial to get sufficiently low-noise signals as we discussed before. Samples were prepared with gold contacts on the surface. For that, UHV electron Beam Evaporation System was used. The UHV metal system allows for the growth of high quality metallic thin layers by electron beam evaporation Fig. 3.5 (b),(c). A gold layer with a thickness of 200 nm was...
obtained with a growth rate $= 1 \, \text{Å/sec}$ see Fig. 3.5 (a). Again under an optical microscope platinum wires of 20µm diameter were attached to the gold pads on the sample surface manually by using Dupont 4529. After that, the contacted samples, including the wires, were cured by a heat treatment at different temperatures. At $T=500^\circ C$ and for one hour and half, the samples were annealed. That leads to contact resistances of 120 - 140 $\Omega$. In order to decrease the contact resistances the samples were annealed again at $T=580^\circ C$. Contact resistances of 10-12 $\Omega$ could be reached by this method.

Since this values are comparable or even slightly higher than those obtained by using EpoTek silver epoxy, the latter method was left for further experiments.

### 3.3 Experimental setups and techniques

#### 3.3.1 Magnet system

In this thesis steady-field experiments in fields of up to 14 T were performed in a liquid $^4$He cooled superconducting magnet system available at the Walther-Meissner Institut (WMI). The system is operated with a maximum current of 111.08 A, to apply a steady magnetic field of 14 T. Two coils of different superconducting materials ($\text{Nb}_3\text{Sn}$ for the inner and $\text{NbTi}$ for the outer coil) are mounted co-axially on a common base and coupled in series. Cooling is realized by a bath of liquid $^4$He surrounding the coils completely. For applying magnetic fields the magnet coils are connected to an external power supply, for that an "Oxford IPS 120-10" was used in our lab, which enable us to apply currents up to 120 A. For experiments at a constant field the coils can be brought in the persistent mode. For that reason, the coil system is equipped with a superconducting shunt. During the charging of the coil this shunt has to be heated to become normal conducting, i.e. resistive. When the desired field is reached the shunt heating can be stopped and the external power supply disconnected. Thus very stable fields are achieved and the noise level is small, since the power supply is decoupled. The limiting factors for superconducting magnets are the finite critical currents and fields of the coil materials.

#### 3.3.2 Temperature control

Within this experimental work, the measurements were performed at temperatures between 1.4 K and 300 K. In order to allow a continuous control of the temperature in this range a variable temperature insert (VTI) was used. The VTI consists of two coaxial tubes with a space in between which can be either filled with an exchange gas or evacuated. This is to make sure that the sample space i.e. inner tube is thermally decoupled from the environment (i.e. the $^4$He bath). As can be seen in Fig. 3.4, where the bottom part of the VTI is shown, a capillary with a rather high gas flow impedance provides a connection between the sample space and the main bath, when the VTI is submerged into the helium.
Then, as the sample space of the VTI is being pumped, a constant helium flow enters the VTI. Resistance with 60Ω and a temperature cernox, placed next to the sample, is used to adjust a certain temperature by applying a heating power. Here temperature sweeps with a ramp speed of $\sim(0.3-3)$ K/min can be performed. For temperatures above 4.2 K, it controlled by the heater power in presence of constant helium gas flow. That

![Diagram of VTI with impedance](image)

**Figure 3.4:** Principle of the VTI with the impedance [30].
way the temperature can be controlled and stabilized between 1.4 K and 80 K. To reach 300 K the VTI must be taken out of the helium bath to stop the helium liquid flow. Without using heater and only by regulating the pressure, temperatures between 1.4 K and 4.2 K can be stabilized due to the pressure dependent boiling temperature of $^4$He. For measuring temperature Cernox and RuO$_x$ resistive thermometers were used. The RuO$_x$ thermometer was used for measurements of temperatures between 1.4 K and 4 K and a calibrated Cernox was used for temperatures above 4 K, with a precision of a few mK. The temperature was read out by a Lake Shore 340 temperature controller. When heating was necessary, the heater was also controlled by the Lake Shore device. Taking into consideration that the Cernox resistor has a weak magnetoresistance, therefore the temperatures below 4.2 K were determined according to the $^4$He pressure in the sample space.

### 3.3.3 Resistance measurements (a.c. 4-probe technique)

Resistivity measurements are carried out by the a.c. (alternating current) four-probe method in order to get rid of the contact resistance effects from the measurements and measure the sample resistance only. As shown in Fig. 3.5, four contacts are attached to the sample, two on each side. The four-wire resistance measurement circuit includes two current leads (I$_+$) and (I$_-$) and two voltage leads (V$_+$) and (V$_-$) electrically connected to the sample. A current is maintained between I$_+$ and I$_-$ and voltage is measured between V$_+$ and V$_-$. Samples are cut and polished into suitable shapes, and the current contacts are carefully placed to cover the sample’s side faces. We cut the sample at with respect to the orthorhombic a and b axes (i.e., along the Cu-Cu direction). Due to the layered crystal structure, NCCO shows a large anisotropy in the resistivity for currents within or

![Figure 3.5](image.png)

**Figure 3.5:** (a) and (b), Illustration of the interlayer transport configurations, i.e. current applied perpendicular to the CuO$_2$-layers, for two different sample geometries characterized by a short or large length in the c- direction, respectively.
perpendicular to the conducting CuO$_2$-layers [41, 60, 61]. The anisotropy ratio is:

$$\rho_c/\rho_{ab} \approx 10^3,$$

(3.1)

with the interlayer resistivity:

$$\rho_c = (U/I).(w/t)$$

(3.2)

The resistance, $R$, is an extrinsic property and depends upon the size and shape of the sample: the length ($L$); width ($w$); and thickness ($t$). Therefore, $\rho$, the resistivity, which does not depend upon sample geometry but is rather an intrinsic property of the material, is more useful. The resistance of the samples was measured in the direction perpendicular to the conducting CuO$_2$ layers, for two reasons:

![Figure 3.6](image)

**Figure 3.6:** Block-diagram of the measuring setup with a variable reference resistor $R_1 = 10\,\Omega$ to 100 $\Omega$ and a load resistor $R_2 = 1\,k\Omega$ to 100 $k\Omega$. The sample voltage ($V$) is measured by using a lock-in amplifier.

Firstly, for the angle-dependent magnetoresistance studies, the ADMR phenomenon is an inherent property of the interlayer magnetoresistance and should be much more pronounced in this configuration [41, 60]. Secondly, due to the high resistivity anisotropy $\sim 10^3$, the interlayer resistance value is usually much higher than the in-plane resistance and, hence, easier to measure. For that, it is necessary to know the geometry of the sample, and it is frequently useful to modify the sample geometry for measurement convenience.
For the samples, which were used in this thesis, the sample dimensions were about $0.3 \times 0.3 \times 1 \text{mm}^3$, with the largest dimension along $c$-axis. The current can be regarded as uniformly distributed over the whole bulk. The sample dimensions were chosen quite small in order to avoid the crystal inhomogeneity. Then, we could obtain a sample resistance of 100-400 $\Omega$ at room temperature [41].

A sketch of our measuring circuit is shown in Fig. 3.6. To measure the resistance an a.c. current of 10 to 100 $\mu$A with a frequency of the order of 300 Hz or 10-18 Hz in case of very small signals is applied and the voltage is amplified and detected by a highly sensitive lock-in amplifier (Stanford system, model 830 or Princeton Applied Research, model 5210). The low current value serves to prevent overheating of the sample at low temperatures and has to be adjusted to the given experimental conditions, e.g. contact resistances and temperature range. To keep the current amplitude constant and stable during the measurement a high resistance $R_2$ (typically 100 k$\Omega$ - 1 M$\Omega$) is placed in series.

For the adjustment of the current and the phase a reference resistance $R_1$ (of 10-100 $\Omega$) is placed into the circuit in series with the sample. By measuring the a.c. voltage across this resistance a desired current value can be set. The absolute sample resistance at low temperatures (with or without a magnetic field) was thus checked to be detected to an accuracy of at least 5%. The signal to-noise ratio during our measurements was typically $\geq 10^4$. Because of the large resistance value of $R_2$ compared to the sample resistances, the change to zero resistance in the superconducting state affects the current by less than 1%, and therefore guarantees a stable current during the whole experiment.

### 3.3.4 Definition of the angles for the magnetic field orientation

The definition of the angles describing how the magnetic field is oriented relative to the sample is given in Fig. 3.7.

The principles of the rotating sample stages used in steady fields are presented as the following: Field rotations in a plane parallel to the crystallographic $c$-axis are described by the polar angle $\theta$, where $\theta = 0^\circ$ corresponds to $B \parallel [001]$. The azimuthal orientation, i.e. the direction of the field component parallel to the CuO$_2$-layers, will be described by $\varphi$, with $\varphi = 0^\circ$ and $\varphi = 45^\circ$ corresponding to $B \parallel [100]$ and $B \parallel [110]$, respectively.

### 3.3.5 Two-axes rotational

A two-axes rotator designed for the 14T superconducting resistive magnet systems was used in our experiments. This insert fits to the superconducting magnet available at the WMI. It was constructed in the framework of the Ph.D. thesis of D. Andres [62].

In Fig 3.8 the principle of rotation is illustrated.

The rotation is provided by two worm gear units. The $\theta$-rotation is driven by a long rod coupled to a piezo-electric motor on top of the whole insert outside the cryostat. The azimuthal $\varphi$-orientation can be controlled by a screwdriver only when the rotation
3.3 Experimental setups and techniques

**Figure 3.7:** (a) Interlayer transport configurations, i.e. current applied perpendicular to the CuO$_2$-layers, with a largest dimension along the $c$-axis. (b) Definition of the angles $\theta$ and $\varphi$ with respect to the crystal axes.

**Figure 3.8:** (a) Photo of the two-axes rotator with introduced rotation angles: $\varphi$ is controlled by the screwdriver, which can be decoupled from the rotator platform, and $\theta$ is controlled by a driving axis coupled via a worm gear in the upper wall. (b) Sample holder with two samples mounted with the CuO$_2$-layers parallel to the rotator platform.

Platform is in its initial position parallel to the screwdriver, as it is shown in Fig. 3.8(a). The screwdriver can be manually controlled from outside. Thus, during an experiment the $\varphi$-position has to be set manually by first sliding the screwdriver in, turning it and finally sliding it out and out. After decoupling the screwdriver manually from the platform, a continuous $\theta$-rotation can be performed fully automatically. Both angles can be set to
an accuracy of $\leq 0.05^\circ$. The sweeping rate of the sample rotation can be continuously changed in a range of 0.003-10$^\circ$/s via a mechanical gear placed outside between the motor and the driving rod. Two samples can be placed, as depicted in Fig. 3.8(b), usually with their crystallographic $c$-axis oriented perpendicular to the rotation platform. Thus, any angular orientation with respect to the magnetic field can be set with this setup.
Chapter 4

Results and discussion

It has long been known that the cuprate superconductors have parent compounds with an antiferromagnetic (AFM) insulating ground state which is suppressed with doping and for that, in this state the magnetic moments are localized on the copper atoms. The mechanism by which the magnetism is suppressed is not symmetric with doping: in hole-doped materials the magnetism is suppressed by spin frustration, whereas in electron-doped materials magnetism is suppressed by spin dilution. One of the consequences of these differing mechanisms is an asymmetry in the phase diagram. Unlike in hole doped systems, where the AFM state is rapidly suppressed well before superconductivity appears, electron doped cuprates exhibit a much more gradual suppression of AFM, leading to questions of competition and/or coexistence (being either macroscopic or microscopic) in both underdoped superconducting and non-superconducting samples.

As known the pairing necessary for superconductivity in cuprates involves the interplay between the doped charges and AFM spin correlations. The study of lightly doped, insulating AFM state is important because the density of the carriers can be sufficiently low such that the interaction between them is small relative to their interaction with the Cu$^{2+}$ spins for that it gives us a chance to study the coupling between charge and Cu$^{2+}$ spins and this is due to:

(1) In contrast to the contorted CuO$_2$ in hole-doped cuprates, the CuO$_2$ plane in electron-doped cuprates is flat, so the spin ordering is pure antiferromagnetic without a ferromagnetic component along the c-axis. In hole-doped cuprates; such a ferromagnetic component along the c-axis makes the study of the coupling between charge and Cu$^{2+}$ spin more complicated.

(2) The spin structure can be tuned by an external magnetic field [63].

Understanding the anomalous features of the out-of-plane normal state transport, particular magnetotransport, in layered cuprates remains a challenge because we deal with the general problem of the transport properties of a single electron in a strongly correlated antiferromagnetically ordered quasi-2D cuprate which continues to be the topic of much debate, both theoretically and experimentally [64]. Previous experiments [65–67] have demonstrated that out-of-plane resistivity is sensitive to the interlayer magnetic order of the spins. Magnetoresistance MR provides a new insight into the coupling between the
charges and the background magnetism with a electron-doped cuprates.

Here we systematically studied out-of-plane MR because it is more sensitive to the spin structure than the in-plane MR and its angular dependence for under-doped Nd$_{2-x}$Ce$_x$CuO$_4$ with $x=0.10$ and 0.12, respectively.

The interlayer MR was measured as a function of the magnetic field strength at different orientations as well as a function of the polar and azimuthal field orientations at different strengths of the field.

### 4.1 Magnetoresistance measurements on NCCO$_{10}$ non-superconducting sample #1

#### 4.1.1 Cooling curves:

In high-$T_c$ cuprates both normal and superconducting states depend on the carrier concentration in the CuO$_2$ planes (doping). In hole doped ($p$-doped) cuprates, the overdoped region is believed to be metallic (Fermi liquid-like), whereas in the underdoped region, at low temperatures the resistivity increases with decreasing temperatures. A similar behavior with decreasing doping is found in electron-doped ($n$-doped) cuprates for underdoped and optimally doped samples, as the temperature falls down, the resistivity decreases till it reaches minimum at $T_{min}$ and then starts to increase [68].

For our NCCO$_{10}$ non-superconducting samples the $c$-axis resistance ($\rho_c$) grows quadratically with high temperature $\sim T^2$ which could be an indication of electron-electron scattering [69, 70], whereas it shows an insulating "upturn" ($d\rho/dT < 0$) at low temperature, which has a log $T$ dependence. As we see in Fig. 4.1, the zero-field resistance as a function of temperature shows a minimum at $T_{min} \approx 63$ K for sample #1 and at $T_{min} \approx 36$ K for sample #2. It is clear that sample #2 appears more metallic than sample #1. This is maybe due to a higher doping concentrations since $T_{min}$ increases with decreasing doping [71].

The question that comes to mind is: what is the origin of this anomalous upturn in resistivity at low temperature?. Actually, the reason for the upturn in the resistivity vs. temperature curves for $T \to 0$, and related to it for some unidentified localization of itinerant charge carriers is up to now not fully understood. However, three suggestions as to where the resistivity upturn comes from were published recently. The first one comes from Fournier et al.[68], where they interpreted that the upturn behavior in resistivity, as well as the negative magnetoresistance (n-MR), are a result of two-dimensional (2D) weak localization by disorder. The second scenario was proposed by Sekitani et al.[21], and they claimed that the upturn behavior and the n-MR could be due to the scattering off Cu$^{2+}$ Kondo impurities associated with the residual apical oxygen. The third suggestion comes from Greene et al.[71], where they concluded that the spin dependent MR exists in the same temperature range as the upturn.
4.1 Magnetoresistance measurements on NCCO_10 non-superconducting sample #1

![Figure 4.1](image)

(a) The zero field resistance as a function of temperature for NCCO_010 non-superconducting sample #1 showing a minimum at \( T_{\text{min}} \approx 63\)K. (b) The same for sample #2 where it shows \( T_{\text{min}} \approx 36\)K.

4.1.2 Interlayer MR for magnetic field parallel to the conducting layers:

In all undoped layered cuprate structures it is known that the spins of the Cu\(^{+2}\) ions have AFM ordering in the CuO\(_2\) plane due to in-plane exchange interaction. At zero-field the spins orientation in the adjacent planes is noncollinear ruled by weak pseudo-dipolar interaction between the planes, since the exchange potential on each copper ion which is created by neighboring planes is canceled due to the body-centered tetragonal crystal symmetry [72], the evidence of this magnetic ordering has been studied using neutron inelastic scattering (NIS) and muon spin rotation (\(\mu\)SR) techniques [13, 39, 73–75].

The zero-field spin structure of the electron doped cuprates is noncollinear antiferromagnetism. The spins are aligned antiferromagnetically, alternating along crystallographic directions [100] and [110], respectively in adjacent CuO\(_2\) layers. At sufficiently high magnetic fields applied in the plane a transition from AFM noncollinear structure to a collinear structure is observed. In this state, the spin alignments in adjacent planes are no longer perpendicular to each other; it has become parallel. In this case Antiferromagnetism can be detected due to a slight angular dependence of magnetoresistance [47].

From this perspective, the electronic in-plane magnetotransport measurements were performed to trace the AFM ordering in the under-doped samples. We have started our interlayer magnetoresistance measurements by applying the magnetic field along the Cu-O-Cu (hard axis) and also along the Cu-Cu (easy axis). In our experiment the Cu-O-Cu axis corresponds to a certain azimuthal angle \(\phi=0^\circ\), whereas for the Cu-Cu axis \(\phi=45^\circ\). The MR measurements were performed by sweeping the magnetic field up and
down between 0T and 14T at a fixed temperature 1.4 K.

![Graph showing interlayer magnetoresistance](image)

**Figure 4.2:** Interlayer magnetoresistance (MR) for the field oriented parallel to the conducting layers, $B \parallel [110]$ (red curve) and $B \parallel [100]$ (black curve), for $x=0.10$ at 1.4 K.

In the noncollinear AFM state at $B=0$, the spins do prefer to align along the crystal axes, i.e. along the [100] and [010] directions, respectively. By applying a magnetic field in a direction parallel to the sublattice magnetization, at small magnetic fields the magnetic moments do not rotate. Then, as the field grows further and at a certain critical field the system suddenly snaps into a different configuration this is called spin-flop transition. Step-like features so called kinks observed at certain critical magnetic fields $B_{SF}=3.5\,\text{T}$ and $B_{SF}=1.1\,\text{T}$ as the field is applied along [100] and [110], respectively as shown in Fig. 4.2.

Those two observed features represent a spin flop induced by a certain magnetic field as mentioned. Upon applying the field a long [100] the Cu spins in the sublattice [100] flop by $90^\circ$, which causes a first order transition from noncollinear into collinear phase in which all of the ordered moments are approximately perpendicular to the direction of the applied field. Here the spins in the noncollinear configuration do require high energy in order to snap into the new collinear configuration; i.e (perpendicular to the magnetic field direction). On the other hand, as the field is applied along Cu-Cu the spins do rotate in-plane by about $45^\circ$ to the same collinear structure but in this case the system undergoes
a second-order spin orientation phase transition [27] so called (Cross over transition). In this case the spins do rotate easily to the new collinear configuration. This explains why the critical field which is required to cause spin-flop along [100] axis which was observed at $B_{SF} = 3.5 \text{T}$ is much higher than what observed when the field is applied along [110] axis to reorient the spin structure where $B_{SF} = 1.1 \text{T}$.

As we see in Fig. 4.2, the anisotropy became opposite, where it shows a noteworthy change in the magnetoresistance sign from positive to negative MR above the critical magnetic field $B_{SF}$ at which the spin flop observed. Also around 4.5 T - 8 T a change in the MR slop is observed but the MR keeps linear decrease with a difference between the two extremal orientations about $\sim 2\%$. The behavior of MR for $B$ along the Cu-Cu direction is almost the same as that for $B$ along the Cu-O-Cu direction in the collinear structure (above $B_{SF}$).

Similar data for strongly under-doped NCCO for $x = 0.033$ and 0.025, recently published by Wu et al.[47], are shown in Fig. 4.3. Step-like features are observed at almost the same critical fields $B_{SF}$ as we recorded along [100] and [110], respectively.

The recorded data for these strongly underdoped samples shows that, above $B_{SF}$, the behavior of MR for $B$ along the Cu-Cu direction is totally different from that for $B$ along the Cu-O-Cu direction in the collinear structure. The MR with $B$ along the Cu-Cu direction slightly changes above $B_{SF}$, while the MR monotonically increases with increasing $B$ for $B$ along the Cu-O-Cu direction. A giant anisotropic MR between the fields $B$ along
the Cu-Cu and Cu-O-Cu directions is observed which comes in contrary with what we found for our 10% samples. For the $x = 0.025$ crystal, the MR at 12 T is as high as 235% with $B$ along the Cu-O-Cu direction, while it is only 17% with $B$ along the Cu-Cu direction [47].

In addition to that, a similar MR behavior for anti-ferromagnetic Pr$_{1.3-x}$La$_{0.7}$Ce$_x$CuO$_4$ has been observed by Lavrov et al.[19] with $x=0.01$. But in this case, the magnitude of the MR and the MR anisotropy are much larger than what we observed for our 10% samples and even than for $x=0.025$ and 0.033 [47].

Comparing between the above mentioned MR measurements [19, 47] and our measurements, it seems that the magnitude of the MR and the MR anisotropy increases as the doping decreases for all doped curates in the electron-underdoped regime. The reasonable argue for that, for very lightly doped samples where $x=0.01$-0.05 the samples normally shows an insulating behavior causes that unambiguous increase of the resistance in the spin flop phase and it starts to decrease due to the influence of doping as we see in our NCCO$_{10}$ sample which is quite high doped as compared to the others [19, 47]. Also it is clear that the MR behavior is surprisingly sensitive to the doping concentration, giving a definite evidence for the itinerant electrons directly coupled to the localized spins even at such very lightly doped samples.

4.1.3 Intermediate field orientations: a second (step-like) feature:

According to what was found for the undoped mother compounds [63], a magnetic field exactly aligned along the [100] direction causes a first-order spin-flop transition. For intermediate orientations, it first induces a collinear ordered spin structure with the staggered moment ordered along [110]. As the field grows further, it gradually rotates to a configuration perpendicular to the field. This consistently explains the lower $B_{SF}$ for the [110]-direction, where the step in the field-dependent MR indicates the spin-flop.

But, interestingly, a second sharp feature was observed in MR at some intermediate angles $\varphi$, when the field was first swept up to 14 T along the [100]-direction and then down at the angle $\varphi$. Examples of such measurements are shown in Fig. 4.4. Here, every time the magnetic field the magnetic field was first applied parallel to [100]. Then, it was turned by an angle $\varphi$ with respect to [100] and swept down.

As shown in Fig. 4.4, intermediate orientations for fields parallel to the conducting layers was held. This were performed by sweeping the field up along [100] and sweep it down at different angles inclines from [100] direction.

For a field applied directly along [100] direction, a spin-flop transition is observed at $B_{SF}=3.5$ T. This step-like feature (as we discussed in section 4.2) occurs as the field induces the spins to be reoriented from the noncollinear to collinear structure.

Then, by rotating the azimuthal angle $\varphi$ at different angles between $\varphi=1.5^\circ$ to $\varphi=23^\circ$ and sweep the field down. Surprisingly a step-like feature was recorded at high fields.
4.1 Magnetoresistance measurements on NCCO$_{10}$ non-superconducting sample #135

Figure 4.4: Interlayer MR for intermediate orientations of the applied field parallel to the conducting layers, $B \parallel [100]$ sweep up (black curves) and $B \parallel$ (different $- \phi$) inclined from $[100]$ (red curves), where the graphs from (a) to (g) show measurements at 1.4 K and (h) is taken at 4.2 K. Note: Curves are vertically shifted for clarity.
The field at which this feature occurs depends on how far the angle was from the [100] direction. It clearly increases gradually as the azimuthal angle $\varphi$ increases. Also, the observed feature becomes much more pronounced (sharper) as the azimuthal angle $\varphi$ increases, as clearly seen in Fig. 4.4. At $\varphi=1.5^\circ$ it is recorded at $B_{SF2}=5.3$ T Fig. 4.4 (c), whereas at $\varphi=23^\circ$ (the maximum angle at which the second feature was observed in our experiment) it is shifted to a higher field $B_{SF2}=13.8$ T Fig. 4.4 (g). It should be noted that the magnetic field was swept up at $\varphi \leq 0^\circ$ before the strong field is aligned at an angle $\varphi>0^\circ$.

Also, it was possible to obtain the second step-like feature by first sweeping the field up at $\varphi_0<0^\circ$, then turning it to $\varphi_1>0^\circ$ and sweep the field down. But if the field was applied at $\varphi_0>0^\circ$, then no feature is observed at $\varphi_1>0^\circ$ during the down sweep. (Note: we are talking about $\varphi$ variation in the range $-45^\circ \leq \varphi \leq 45^\circ$.

Fig. 4.5, shows how the second feature critical field increases as the azimuthal angle $\varphi$ increased.

![Figure 4.5](image)

**Figure 4.5:** Magnetic fields at which the second step-like feature observed at $T=1.4$ K vs the azimuthal angles $\varphi$.

What we observed could be explained as follows: As we discussed in section 2.3, at zero field the spin magnetic moments are ordered antiferromagnetically within the layers forming a noncollinear (crosslike) magnetic structure which has the lowest energy state.
From this scene, let us consider two vector moments \( L_1 \) and \( L_2 \) where \( L_1 \) corresponds to the lattice where the spins are staggered along \([100]\) i.e the magnetization moment \( \vec{M} \parallel [100] \) and \( L_2 \) corresponds to the sublattice where the spins are aligned along \([010]\) i.e \( \vec{M} \perp [010] \). Then, as the applied field direction coincides with the spin orientation along \([100]\), a first order transition in a form of spin flops appears. This corresponds to the critical field \( B_{SF1} \) which is recorded in our measurements at \( B_{SF1} = 3.5 \) T. This transition occurs due to the flops of the sublattice spins to the direction perpendicular to the field i.e the sublattice spins rotates by \( 90^\circ \), while the initial positions of the spins which oriented a long \([010]\) is almost unchanged. Here, at \( B > B_{SF1} \) i.e in the collinear configuration, the spins in both subsystems are staggered perpendicular to the field direction as shown in Fig. 4.4 (black curves).

For the second step-like feature which we have observed at different angles tilted away from the \([100]\) orientation by sweeping the field down from 14 T directly after a field sweep up along \([100]\) direction can be discussed as follows: At high fields our spin structure already is in the collinear configuration. The spins here are aligned perpendicular to the applied field direction. Then, upon rotation of the external field the magnetic moments do not rotate but keep "frozen" in the same collinear configuration. Hence, as the field swept down the spins shows a hysteretic behavior at critical field \( B_{SF2} \) which arises due to the minimization of zeeman energy. So that, the spins can easily overcome the energy barrier to reach absolute minimum energy. Then, as the field decreases, the spins undergo another phase transition from the collinear to noncollinear structure at a particular field at which the spins in one of the two sub-lattices flop to be parallel to the applied field orientation. At this particular field the spins undergoes a second order phase transition where the angle between the two subsystems is the order parameter as shown in Fig. 4.4 (red curves). By sweeping the field up to 14 T at the same angles, the second-step like feature disappeared giving a clear evidence that it is appearance not due a spin flop transition, this we can see clearly in Fig. 4.4 (e).

Another scenario : by sweeping the field up along \([100]\), at high field our spin structure in already collinear, the spins here are aligned perpendicular the applied field direction. Upon rotation of the azimuthal angle \( \varphi \). The spins are no longer perpendicular to the field direction but they are slightly inclined from their original orientation by a small angle. Let’s call it \( \alpha \) (the angle between the two subsystems). Hence, when we start to sweep the field down the spins then do rotate again to its preferred easy axis at which the spin again oriented perpendicular to the field direction where the angle \( \alpha \) between the two subsystems starts to decrease gradually till it reaches zero at a critical field \( B_{SF2} \). The spins again aligned to be perpendicular to the field which is energetically favorable for the spin collinear configuration. Then, as the applied field decreases down to zero the spins experiences another phase transition from collinear to non collinear structure \( B_{SF1} \) at which the spins in one of the two sub-lattices flop to be parallel to the applied field orientation. This transition is a first order phase transition similar to what we have
recorded when the field was applied along [100] Cu-O-Cu axis.

The same set of measurements was performed at $T=4.2$ K. At $\varphi=10^\circ$ a second step-like feature was observed at a critical field $B_{SF2}=11.3$ T with a small hysteretic in comparison to what we observed at $T=1.4$ K at the same condition which is a normal behavior of hysteresis as the temperature increases.

Also, such observed features at such temperature $T=4.2$ K give us a clear evidence that this features related to the Cu-Cu interaction which means Nd-Cu interaction is irrelevant to long range order antiferromagnetism where as discussed before the Nd moments becomes ordered at temperature lower than 1 K.

### 4.1.4 Out-of-plane field rotations :

#### 4.1.4.1 ADMR $R(\theta)$ at $\varphi=0^\circ$ (Cu-O-Cu):

![Figure 4.6: ADMR curves ($\theta$-rotations) at different fields for an $x=0.10$ sample at $\varphi=0^\circ$ along Cu-O-Cu and $T=1.4$ K.](image)

The ADMR of a strongly underdoped, $x=0.10$, sample recorded at $\theta$-rotations where $\theta$ is the angle between the applied field and the $c$-axis. The measurements were performed at temperature of 1.4 K as shown Fig. 4.6.
4.1 Magnetoresistance measurements on NCCO_10 non-superconducting sample

Starting from low applied fields 1\,T up to 3\,T no features were observed in our $\theta$ dependent measurements. Once we increase the applied field up to $B = 3.8\,T$, and exactly at $\theta = 66^\circ$ a sharp step-like feature is observed. The feature position shifts towards smaller $\theta$ and becomes weaker at increasing field.

At $B = 6\,T$, the step-like feature around $\theta = 23^\circ$ and $-23^\circ$ is observed. Then as the field grows further above 6\,T the step-like feature starts to come close to $\theta = 0^\circ$ where $B \parallel C$-axis. Obviously, the in-plane component becomes weaker and weaker as the field increases which means that the in-plane field component is no longer strong to stabilize the collinear phase within the CuO$_2$ layers.

![Figure 4.7: Field sweeps at different $\theta$ for an $x = 0.10$ sample at $\phi = 0^\circ$, starting from $\theta = 0^\circ$ where $B \parallel (Cu-O-Cu)$ (red curve) to $\theta = 90^\circ$ where $B \parallel C$-axis (black curve) at $T = 1.4\,K$.](image)

It is striking that what we have observed from this set of measurements comes with conformity with what we have seen from the interlayer (MR) for field parallel to the Cu-O-Cu measurement see Fig. 4.2, at which the spin structure experience a phase transition from noncollinear to collinear at critical field $B_{SF} = 3.5\,T$.

In order to see how this step-like feature position changes toward $B \parallel [001]$ as we go with the field higher than $B = 3.5\,T$, field dependence measurements at different $\theta$ were performed with $\theta$ changing from $\theta = 0^\circ$ to $\theta = 90^\circ$, as we see in Fig. 4.7. The usual spin flop at $B = 3.5\,T$ is observed at $\theta = 90^\circ$. As the angle $\theta$ decreases gradually, the feature shifts...
towards high fields and at the same time starts to lose its sharpness. Around $\theta=45^\circ$ the feature appears to be flattened, and it seems to be totally disappeared at $\theta=0^\circ$.

The positions of the step in the $R(\theta)$ curves at $4T$ and $5T$, in Fig. 4.6, correspond to the in-plane field component $B_\parallel = 3.8 - 4T$. However, at $B \geq 6T$ the observed step does not scale with the in-plane field component $B_\parallel = B \sin \theta$.

### 4.1.4.2 ADMR $R(\theta)$ at $\varphi = 45^\circ$ (Cu-Cu):

The same set of measurements was performed and a similar step-like feature in the ADMR was observed for $\theta$-rotations in the plane at $\varphi = 0^\circ$ along Cu-Cu direction. The feature also shifts towards the $B \parallel c$-direction upon increasing field. As shown in Fig. 4.8, at very low applied fields 1 T-1.1 T no features was observed. Then, at $B = 2T$ a clear step-like feature observed around $\theta=33^\circ$. Its tempting to associate the step-like feature with the spin-flop transition observed at $B_{SF} = 1.1T$ in the field sweeps for $B \parallel [110]$ cause spin reorientation transition from collinear configuration to non collinear configuration. Then, as the field increases the feature is still there and again it shifts towards $B \parallel [001]$. The positions of the step-like feature in the angular sweeps $R(\theta)$ at $B = 2T$ up to $B = 4T$ scale with the in-plane field component $B_\parallel = B \sin \theta$. However, at $B \geq 6T$ the the recorded

**Figure 4.8:** ADMR curves ($\theta$-rotations) at different fields for an $x = 0.10$ sample at $\varphi = 45^\circ$ along Cu-Cu and $T = 1.4K$. 
4.1 Magnetoresistance measurements on NCCO-10 non-superconducting sample

Figure 4.9: Field sweeps at different $\theta$ for an $x = 0.10$ sample at $\varphi = 0^\circ$. The curves were recorded at different fixed $\theta$ starting from $\theta = 90^\circ$ where $B \parallel (Cu-Cu)$ (red curve) to $\theta = 0^\circ$ where $B \parallel c$-axis (black curve); and $T = 1.4$ K.

step-like feature does not scale with the in-plane field component. This could be due to that the in-plane component at high fields is much weaker than the out-of-plane component which is reasonable as the observed step-like features positions in our measurements at $B \geq 6$ T is already shifted towards $B \parallel c$-axis.

In order to check the origin of the shifted features in the ADMR, field sweeps at different angles $\theta$, $R(B)_\theta$ were held by changing $\theta$ from $\theta = 90^\circ$ where $B \parallel (Cu-Cu)$ to $\theta = 0^\circ$ where $B \parallel c$-axis.

Interestingly, the kink like-feature is observed for all the angles ranging from $\theta = 90^\circ$ up to $\theta = 10^\circ$ within $[R(B)]_\theta$ measurements. As shown in Fig. 4.9, the kinks position are shifted towards the high fields as $\theta$ is tilted towards $c$-axis. Here the in-plane field component gets weaker than the out-of-plane component as long as $\theta$ varying gradually away from $B \parallel (Cu-Cu)$. This explains the behaviour of the observed features at high fields in the angular sweeps measurements as shown in Fig. 4.8, where it associates with the transition of the Cu$^{2+}$ spin lattice back to the noncollinear configuration as the in-plane field-component weakens.

No step-like feature is observed at $\theta = 0^\circ$ where $B \parallel c-axis$ cause the in-plane
components vanishes. Also, a prominent central hump in Fig. 4.8 is observed in the ADMR curves for $-30^\circ \leq \theta \leq 30^\circ$ at $B=2T$. This could be associated to the spins reorientation from collinear to noncollinear configuration at low fields and it gets smaller or less pronounced due to the weakness of the in-plane component as the field increases.

### 4.1.5 Interlayer MR for field parallel to the conducting layers at different temperatures $R(B)_T$:

![Graphs](image)

**Figure 4.10:** Interlayer MR for the field oriented parallel to the Cu-O-Cu axis ($\varphi=0^\circ$), at different temperatures. (a) Shows MR measurements at temperatures ranging from 1.4 K up to 10 K and the curves are shifted vertically for clarity. (b),(c),(d) Shows MR measurements at $T = 15$ K, $T = 20$ K and $T = 30$ K, respectively.

Interlayer MR for fields parallel to the conducting layers at different temperatures $R(B)_T$ was measured for $B||[100]$ and $B||[110]$, respectively as shown in Fig. 4.10 and Fig. 4.11. Interestingly, the observed step-like features as result of a spin flop transition as we discussed before survived up to 30 K accompanied by the n-MR. The amplitude of n-MR exhibits a variation, showing a tendency to decrease as the temperature increases.
4.1 Magnetoresistance measurements on NCCO$_{10}$ non-superconducting sample #1

Figure 4.11: Interlayer MR for the field oriented parallel to the Cu-Cu axis ($\phi = 45^\circ$), at different temperatures. (a) Shows MR measurements at temperatures ranging from 1.4 K up to 10 K and the curves are shifted vertically for clarity. (b),(c),(d) Shows MR measurements at $T = 15$ K, $T = 20$ K and $T = 30$ K, respectively.

Also, the position of the kink feature seems to be shifted towards a lower field as the temperature increases. However, since the AFM ordering of Nd$^{3+}$ spins in formed below 1.4 K [47], the observed step-like features at such relatively high temperatures give us an evidence of the main role of Cu-Cu magnetic interactions in the presence of the AFM long range order in such non-superconducting samples.
4.2 Magnetoresistance measurement on NCCO\textsubscript{10} non-superconducting sample \#2

4.2.1 Interlayer MR for field parallel to the conducting layers

For the second sample the same set of measurements was performed for the field applied parallel to both Cu-O-Cu (Hard axis) and the Cu-Cu (Easy axis), respectively. As we discussed for the first NCCO\textsubscript{10} sample, as the magnetic field applied in the ab-plane will force the copper magnetic moments to switch to a collinear AFM state in the direction perpendicular to the applied field. A weak kink feature was observed at $B_{SF}=0.8$ T along [100] direction and a hardly discernible kink feature at $B_{SF}=3.4$ T along [110] axis as shown in Fig. 4.12 (black curve). The MR changes sign and the anisotropy is opposite, with a difference between the two extremal orientations of $\approx 2\%$, it is rather small. Around 5 - 8 T the MR changes its slope but decreases further almost linearly.

It’s clear that the critical fields corresponding to both observed step-like features (kinks) have quite different values comparing with the previous sample. The reason for that could be due to the higher doping concentration or stronger annealing treatment which results

![Figure 4.12: Interlayer magnetoresistance (MR) for the field oriented parallel to the conducting layers, $B \parallel [110]$ (red curve) and $B \parallel [100]$ (black curve), for $x=0.10$ at 1.4 K.](image)
4.2 Magnetoresistance measurement on NCCO\(_{10}\) non-superconducting sample \#2

in a small amount of remnant interstitial oxygen for this particular sample giving that metallic behavior with decreasing temperature as we discussed before.

### 4.2.2 In-plane angular sweeps:

The anisotropic MR measurements were performed by rotating the magnetic field \(B\) within the CuO\(_2\) plane on NCCO\(_{10}\) non-superconducting sample\#2. The samples were mounted on a rotator stage that allowed 0-220° of rotation with the axis of rotation parallel to the \(c\)-axis of the crystal structure. As the magnetic field was rotated in the CuO\(_2\) plane, the copper spins were alternately aligned along the easy and hard axes, [110] and [100] respectively.

![Figure 4.13: Angle-dependent interlayer MR for \(x = 0.10\) for fields oriented parallel to the conducting layers.](image)

As Fig. 4.13 shows, the \(\phi\)-dependence measurements are recorded at different fields in the range between \(B = 1\) T and \(B = 14\) T. One can see that the resistance decreases as the field increases. This is of course consistent with the n-MR data presented in section 4.1.2 in the form of field sweeps for \(B \parallel [100] \& [010]\). The resistance alternating in 45° shows minimum MR for \(B \parallel [100]\) and maximum for \(B \parallel [110]\). This resulting in fourfold oscillation of the angle dependent magnetoresistance (ADMR), where MR diagram rotates by 90°. These oscillations in MR are due to an underlying magnetically ordered state and therefore their observation is an indication of the magnetic structure of the crystal lattice.
which appears due to the tetragonal symmetry of our crystal. Similar anisotropy with four-fold symmetry has been observed in Pr$_{1.3-x}$La$_{0.7}$Ce$_x$CuO$_4$ with $x=0.01$ crystal [19]. Such behaviour has been explained by V. P. Plakhty et al [63]. These authors proposed that the relative orientation of spins with respect to the crystal axes comes from the fact that the spin structure always stays collinear at high fields because the total energy does not change due to the interplane pseudo-dipolar interactions when the spin sublattices of the adjacent CuO$_2$ planes rotate in opposite directions [76, 77]. So, the continuous spin rotation is induced by rotation of the applied field because the spins gradually rotate toward a configuration perpendicular to the field orientation at high fields.

As we see in Fig. 4.13, the amplitude of the MR oscillations decreases with decreasing the applied field. Thus, the anisotropy in the MR changes its sign. Here, it is clear that the negative MR increases gradually within the collinear phase as the field grows up.

At $B \geq 6$ T a clear step-like feature is observed as the field is tilted away from the [100]-direction with a hysteretic behaviour depend on the direction of the angular sweep. The feature becomes more pronounced as the field grows up. Again the spins undergo transitions due to the energy competition from the collinear configuration with local minimum energy to a collinear configuration with absolute minimum which shows a hysteretic effect due to the energy barrier.

For lower field, at $B \leq 3.5 - 4$ T, a step-like features was observed in the vicinity of [100] and [010] directions, at which the spin configuration snaps into noncollinear structure from the collinear one.

In recapitulation of the NCCO$_{10}$ it has been observed experimentally that the magnetoresistance takes on measurably different values, depending upon whether the field is aligned along the Cu-Cu direction, [110], or along the Cu-O-Cu direction, [100]. This hysteretic behavior which observed is a manifestation for the itinerant electrons coupled to the localized spins.

### 4.2.3 Out-of-plane field rotations:

The ADMR of the second $x = 0.10$ (sample #2) was recorded for $\theta$-rotations, where $\theta$ is the angle between the applied field and the c- axis. The measurements were performed a constant temperature of 1.4 K as shown in Fig. 4.14.

At low applied fields between 2 T and 6 T a clear features in the vicinity of $\theta = 0^\circ$ and $\theta = 180^\circ$ were observed in our $\theta$- dependent measurements. The positions of these step-like features do scale with the in-plane field component $B \parallel = B \sin \theta$.

Again the observation of these features seems to be related to the critical spin-flop field $B_{SF}$, associated with the transition of the Cu$^{2+}$ spin lattice back to the noncollinear configuration as the in-plane field component weakens.

For fields between 10 T and 14 T and for orientations close to perpendicular, $\theta=0^\circ$, no step-like feature was observed.
4.2 Magnetoresistance measurement on NCCO_10 non-superconducting sample #2

Figure 4.14: ADMR curves ($\theta$-rotations) at different fields for an $x = 0.10$ sample at $\varphi = 0^\circ$ along Cu-O-Cu and $T = 1.4$ K.

Obviously, the in-plane component of the staggered magnetization becomes weaker and weaker as the field increases which means that the in-plane field component is not strong enough any longer to stabilize the collinear phase within the CuO$_2$ layers which could be the reason of step-like feature disappearance at $B \geq 6$ T.

4.2.4 Interlayer MR for field parallel to the conducting layers at different temperatures $R(B)_T$:

Interlayer MR measurements were performed on the NCCO 10% non-superconducting (second sample) for field parallel to the conducting layers at different temperatures $R(B)_T$.

The results are shown in Fig. 4.15 and Fig. 4.16, a shift in the critical field position to lower fields is observed as the temperature increases. The step-like feature which arises due to the spin reorientation into the collinear configuration as we discussed before becomes less pronounced as the temperature increases and it has the same behaviour in both cases i.e the field applied along Cu-O-Cu axis and along Cu-Cu axis. Such behavior at relatively high temperature give us an evidence of the Cu-Cu magnetic interactions is the main driving force in our spin reorientation mechanism and its influence on whether
we have long range order antiferromagnetism cause at such high temperature the Nd-Cu interaction becomes insignificant and can be considered as a perturbation, also the Nd-Nd has nothing to do with our spin configurations cause at high temperature $T > 50$ K, the rare-earth lattice is para magnetic [78], even at low temperature where the Nd-Nd interaction begin dominate at $T < 1$ K the influence of the rare-earth ions can be easily ignored as long as our step-like feature survives at $T \geq 1.4$ K.

The obtained results shows that the amplitude of the n-MR decreases as the temperature increases and it surprisingly vanishes approximately at the same temperature at which the upturn disappears i.e. at $T_{min}=36$ K for this particular sample as shown in Fig. 4.1(b). The MR sigh starts to be positive as the temperature further increases. Hence, one can conclude that there is a direct relation between the n-MR for $B \parallel a - b$ plane and the upturn behaviour in resistivity.

A correlation between the upturn in the zero - field $R(T)$ dependence and the isotropic spin related MR was noticed by Dagan et al.[71]. According to these authors the upturn behaviour is a spin scattering process.
4.2 Magnetoresistance measurement on NCCO_10 non-superconducting sample #2

Figure 4.15: (a) Interlayer magnetoresistance (MR) for the field oriented parallel along (Cu-O-Cu) axis where $\varphi = 0^\circ$, at different temperature where $1.4K \leq T \leq 35K$, the measurements were performed for the NCCO 10% non-superconducting (sample 2). (b) The rest of measurements at $T \geq 35K$.

Figure 4.16: (a) Interlayer magnetoresistance (MR) for the field oriented parallel along (Cu-Cu) axis where $\varphi = 45^\circ$, at different temperature where $1.4K \leq T \leq 35K$, the measurements were performed for the NCCO 10% non-superconducting (sample 2). (b) The rest of measurements at $T \geq 35K$. 
4.3 Magnetoresistance measurements on NCCO_012 (SC) samples

4.3.1 Cooling Curve:

For this SC sample similar interlayer MR measurements are carried out at various field orientations. This particular Nd$_{1.88}$Ce$_{0.12}$CuO$_4$ sample with Ce concentration $x=0.12$ had been tested by the magnetic measurements showing a SC signal equivalent to 16.5 % of the ideal diamagnetic shielding. The out-of-plane resistance of this sample at room temperature was about $\approx 320 \Omega$.

![Figure 4.17](image)

**Figure 4.17:** The resistance as a function of temperature for Nd$_{1.88}$Ce$_{0.12}$CuO$_4$ Superconducting sample shows $T_c = 25$K at zero field.

In contrast to hole-doped cuprates, the critical fields for this electron-doped sample is low. This give us an easy access to the normal state, since superconductivity can easily be suppressed by applying a magnetic field $B \sim 6$-8 T (perpendicular to CuO$_2$ layers). This is true for any doping level even at the lowest temperatures.

Fig. 4.17 shows the $T$ dependent zero field out-of-plane resistance curve. The critical temperature $T_c = 18$K and a little step in the cooling curve $R(T)$ has been detected at
4.3 Magnetoresistance measurements on NCCO_012 (SC) samples

$T = 25\, K$ (not resolved in the scale of Fig. 4.17). This step reveals a minor fraction of a SC phase with $T_c = 25\, K$. Above $T_c$ the resistance shows a monotonic behavior with a temperature dependence close to $T$-linear dependence.

**4.3.2 Interlayer MR for field parallel to the conducting layers**

The set of measurements presented in this section similar to that performed on the NCCO_10 samples in field parallel to the conducting layers. Here all measurements have been done at $T = 1.4\, K$. We started the interlayer MR measurements by applying the field along the [100] axis where $\varphi = 0^\circ$, no features were observed by sweeping the field up to $=14\, T$ and down to $0\, T$ as can be seen in Fig. 4.18 (black curve). Here one could expect that it is reasonable that no feature was observed because we were already measuring in the superconducting state at $1.4\, K$. To make sure of that measurements, intermediate

![Figure 4.18: Interlayer MR for intermediate orientations of the applied field parallel to the conducting layers, Field sweep up $B \parallel [100]$ (black curve) and Field sweeps down to 0 T $B \parallel$ (different angles $\varphi$) inclined from [100] starting from $\varphi=9^\circ$ (red curve) to $\varphi=41^\circ$ (green curve), at 1.4 K. Note: the curves are shifted vertically for clarity, the sample resistance is zero at $B = 0\, T$. Arrows point to the observed second step-like features.](image)
orientations of the applied field were undertaken in order to search for the second hysteresis feature which we have already observed for NCCO\textsubscript{10} non-superconducting samples. That was by sweeping the field down at different angles tilted from \[100\] direction directly after a field sweep up along \[100\] direction, see section 4.1.3.

Surprisingly, a step-like feature was found by sweeping the field down at different angles away from \[100\] directly after a field sweep up along \[100\] direction. As can be seen in Fig. 4.18, a step-like feature was detected as the field oriented a way from \[100\]. At $\varphi = 9^\circ$ (red curve), while the field swept down, the step-like feature was observed at 4.3 T. Then, by further the field direction from \[100\], it is obvious that the step-like feature position shift smoothly towards the higher field. At $\varphi = 41^\circ$ the critical field of the observed feature is 5 T.

From that one can easily see that the observed step-like features for this NCCO\textsubscript{12} (SC) sample behaves in a similar way to what we observed for the previous NCCO\textsubscript{10} non-superconducting samples. This is an evidence of spin reorientation from collinear configuration to non-collinear one below the critical fields as the field is swept down directly after a field sweep up along the \[100\] direction. Hence, one can estimate that there is a hardly discernible step-like feature for $B \parallel [100]$ in order to change the spin configuration to a stable collinear phase at high fields which is required for the observed down sweeps features.

It should be noted that the explanation of the second feature proposed in section 4.1.3 crucially relies on the presence of the long-range collinear AF ordering with the staggered magnetization aligned exactly perpendicular to the strong field applied along \[100\]. The observation of this hysteretic feature on the NCCO\textsubscript{12} is a strong evidence of the existence of the long-range AF order in this sample. It also suggests there is a spin flop transition at $B \parallel [100]$. The black curve corresponding to this field direction which shown in Fig. 4.18, has a weak hump at $B \approx 2$ T which may be a manifestation of the spin flop. We note that the strength of the resistive anomaly associated with the spin flop is sample dependent. For example the NCCO\textsubscript{10} sample #2 showed almost no feature at this field orientation, see Fig. 4.12.

Interestingly, the whole observed step-like features was recorded between 4 T to 5 T, which is below the known superconductivity critical field $B_{c2}=6$-8 T for field applied \perp to the conducting layers, at which the superconductivity is completely suppressed. Form that, one could estimate that the observed feature appear as a result of the long range order AFM without any influence of the SC properties. The arguments that the "second step-like feature" is not due to due to SC properties (vortex melting, irreversibility field, etc) can be discuss as follow:

Firstly, the second step-like feature is a consequence of the hysteresis in resistive behavior where the hysteresis in the resistance of the superconductors in the mixed state is highly unusual by contrast to the hysteresis in SC magnetization. On the other hand, the procedure which is required for obtaining this feature is equal to that we had for
NCCO_10. That implies an AF magnetism is the origin of this feature. Secondly, the observed feature is unlikely because of a fraction of a lower-doped phase where the feature characteristic fields at different $\varphi$ from [100] direction is quantitatively different from what we observed for NCCO_10 non-superconducting samples. Thirdly, the second feature critical field increases as the temperature increase which in turn violates the behavior of all characteristic field of SC state as we are going to discuss in the next section.

4.3.3 Observation of the second step-like feature for intermediate orientations of the applied field at different temperatures:

In order to see how the observed step-like feature behaves at temperatures above 1.4 K, the same set of interlayer MR measurements were held at several temperatures between 1.4 K and 4 K. In this experiment the field was swept down at $\varphi=10^\circ$ directly after a field sweep up along [100] direction at $\varphi=0^\circ$.

As can be seen in Fig. 4.19, a clear step-like feature is recorded at $T=2.3$ K and 3 K, respectively. At 1.4 K, the feature was observed at 4.3 T and by further increasing the

![Figure 4.19: Field sweeps down at $\varphi=10^\circ$ at different temperatures between 1.4 K and 4 K. Before each down sweep, the field was swept up at $\varphi=0^\circ$; i.e $B || [100]$. Arrows point to the observed second step-like feature for each temperature.](image-url)
temperature the feature recorded at 4.5 T and 5 T at temperatures 2.3 and 3 K, respectively. Results in a smooth shift in the critical field position towards the high fields is observed as the temperature increases.

The same trend has been obtained for the NCCO_10 non-superconducting samples, at which the observed features show a spin reorientation transition from the collinear configuration to the noncollinear one by sweeping the field down directly after a sweep up along [100]. This give us a strong support of the AF origin of this feature in the present sample. By contrast, if this feature had a SC origin associated with some transformation of the vortex system in the mixed state, one would expect a shift to lower field at higher temperatures. Moreover, a hysteresis in the resistance would be very unusual.

Thus, we conclude that the long-range AF order and the superconductivity coexist in the present NCCO_12 sample.

4.3.4 Azimuthal field orientation variation at $T = 27 K$:

![NCCO_0.12 (SC) T=27 K](image)

**Figure 4.20:** Interlayer MR for $x = 0.12$ for the field oriented parallel to [100], at $T = 27 K > T_c$.

According to what we observed from the $\phi$-dependence measurements for the previous NCCO_10 non-superconducting samples, it seems that the relationship between angular
Magnetoresistance and antiferromagnetism is largely empirical. For that and in order to trace the antiferromagnetic features in this NCCO$_{12}$ (SC) sample, the angle dependence of the interlayer MR for fields oriented parallel to the conducting layers were held.

The $\varphi$-dependent measurements were performed at temperature above $T_c$ at which the superconductivity is completely suppressed. For that the temperature was stabilized at $T = 27\,K$.

Before starting the $\varphi$-dependence measurements, field sweep measurements up and down have been done at $B \parallel [100]$. As shown in Fig. 4.20, by applying the field along $[100]$ the MR is flat up to $\approx 6\,T$. Then, as the field grows further, a very weak n-MR is recorded between 6-8T. As the field increases above 8T an obvious sharp step up in the MR observed, then it increases further monotonically. Returning to our $\varphi$-dependence measurements, as shown in Fig. 4.21 the measurements were performed by rotating the magnetic field $B$ within the CuO$_2$ plane. The effect of the azimuthal field orientation at different fields for this sample seems to be the similar to that for NCCO$_{10}$ non-superconducting samples.

Step-like features close to [100]and [010] with a hysteretic behavior are clearly seen.

![Figure 4.21: Angle-dependent interlayer MR for fields oriented parallel to the conducting layers for $x=0.12$ at $T = 27\,K$.](image)
upon rotating the azimuthal angle $\varphi$ up and down at fixed fields. The curves show a minimum MR for $B \parallel [100]$ and a maximum MR for $B \parallel [110]$.

The amplitude of the step-like feature develops starting from relatively low fields $B \sim 4 - 6$ T, indicating spin reorientation transition and it becomes much more pronounced as the field increases to 14 T; i.e (simultaneously a hysteresis appears and significantly grows at increasing the field). As we discussed before, the observed MR oscillations arise from a change of the relative orientation of the spins with respect to the crystal axes because the spin structure always stays in the collinear arrangement and the spins are gradually rotate towards a configuration at which they lie perpendicular to the applied field direction where the hard and easy axis spins are tuned by the field [79]. Below $B = 4$ T this step-like feature disappeared where we have a stable non-collinear configuration.

Here the observed features is related to a field induced reorientation of the ordered Cu spins where the Cu spins alternately aligned along the easy axis [110] and the hard axis [100].

An interesting thing is that the MR diagram for this sample at such high temperature shows two-fold symmetry which is arising due to the collinear phase (I) symmetry see section (2.3). At this temperature range i.e $1.4$ K $<$ $T$ $<$ $30$ K, at zero field the spin configuration appears in a noncollinear phase (I) then above the critical field the spins ordered in a collinear phase (I) and in this collinear configuration at high fields the spins are rotates by $90^\circ$ depend at which direction the field is oriented whether along [100] or [110]. That explains why with further cooling the four-fold symmetry developed and this comes in consistent with what we have seen for the azimuthal field orientation measurements for the NCCO\(_{10}\) non-superconducting samples at $T=1.4$ K, where the MR oscillations appears to be symmetric for the noncollinear structure phase (I) and (III). This observed features shows a clear evidence of the long range antiferromagnetism and the reason for the surprisingly breakdown of the four-fold symmetry is still to be understood.
4.3.5 Azimuthal field orientation variation for $T > 27$ K at $B = 14$ T:

At temperatures higher than 27 K, the same set of measurements was performed by rotating the magnetic field $B$ within the CuO$_2$ plane. As shown in Fig. 4.22, the measurements were performed at different temperatures between 35 K and 90 K. At $T = 35$ K, a step-like features close to [100] and [010] with a hysteretic behavior are clearly seen upon changing the azimuthal angle $\varphi$ up and down at fixed field; the curves alternating shows minimum MR for $B \parallel [100]$ and a maximum MR for $B \parallel [110]$.

At temperatures between 35 K and 80 K As shown in Fig. 4.22, as the temperature increases, the amplitude of the observed features decreases, the step-like feature disappeared totally. Moreover, the paramagnetic state is not normal even at these high temperatures. As one can see a very strong hysteresis between the up two downwards angular sweep is conserved with a clear shift between the two extreme points at which the field is parallel to [110]. The reason for that at such high temperatures could be due to the randomness of the magnetic moments ordering which is appears as so called spin glass state.

Further studies are necessary in order to reveal the evolution of this glassy state and it is detailed characteristics at such high temperatures.
Figure 4.22: Angular-dependent interlayer MR for $x=12$ for field oriented parallel to the conducting layers for $35\,\text{K}<T<90\,\text{K}$. 
Chapter 5

Conclusion and outlook

In this thesis, the out-of-plane magnetoresistance measurements were done for underdoped Nd$_{2-x}$Ce$_x$CuO$_4$ with $x=0.10$ and 0.12.

These measurements were used as a tool to provide information on the interaction between charge carriers and magnetic moments. The focus was laid on manifestations of spin-dependent transport characteristic of a magnetically ordered state.

The measurements were carried out for doping levels $x$ close to the border of the SC doping on the underdoped side of the phase diagram in order to inquire the relation between the AF state and SC state of the electron-doped cuprate superconductors and try to observe if there is a coexistence of these two states.

From this perspective, the interlayer MR was measured as a function of the magnetic field strength at different orientations as well as a function of the polar and azimuthal field orientations at different strengths of the field.

The main results of this work are summarized in the following:

Firstly, for NCCO 10% sample: The interlayer magnetoresistance measurements show a spin subsystem related feature associated with a spin-flop transition by applying the field along the two crystal axes $a$ and $b$. A hysteretic second step-like feature was observed for intermediate in-plane field orientations. This feature shows an evidence of spin reorientation from a meta stable spin configuration to a stable one as the field is swept down directly after a sweep up along the [100] direction. From that, it was clear for us that these features are attributed directly to the Cu$^{2+}$ spins subsystem and not due to the Nd$^{3+}$ ions. Oscillations in the magnetoresistance with four-fold symmetry were observed during the in-plane angular sweeps. These observed oscillations were accompanied by clear sharp features with a hysteretic behaviour. These features came in consistent with the second step-like feature in the field sweeps, at which the spin configuration is a meta stable collinear one at high fields and it snaps into a stable structure at lower fields. Also, the out-of-plane field rotations in a fixed magnetic field up to 14 T have shown a significant effect of the magnetic subsystem reorientation.

Secondly, for NCCO 12% superconducting sample: A second step-like feature was observed at $T$=1.4 K by sweeping the field down at different angles inclined away from the [100] direction directly after a field sweep up along the [100] direction. The observed fea-
tures have a similar behavior to what we observed for the NCCO 10% non-superconducting samples. Again these features show an evidence of a spin subsystem reorientation and a presence of an long range order AF. From that, we concluded that the magnetoresistance measurements show a coexistence of the long-range AF order and the superconductivity. At $T \geq 27$ K at which the superconductivity is totally suppressed, the in-plane angular sweeps were done by rotating the field within CuO$_2$ planes. A sharp feature with a hysteretic behavior has been observed, where the amplitude of the step-like feature was developed as the field increases. Also the observed magnetoresistance oscillation was attributed to the change of the relative orientations of the spins as mentioned before.

The obtained experimental results brought an insight into the normal state properties of the electron-doped cuprate Nd$_{2-x}$Ce$_x$CuO$_4$. They also expected to have a significant impact on the understanding of superconductivity in this type of materials, in particular, concerning the interplay of superconductivity and magnetism.

Much more measurements in the doping range $12.5 \geq x \geq 14.5$ is needed to be done with the same technique which we used in our measurements in order to see the evolution of the long range AF order near the optimal doping level. Such experiments could put us on the path of understanding the pairing mechanism in superconductivity where it may have magnetic origin.
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Declaration

I declare that I prepared and wrote this thesis work independently and with no other means than those referenced in the text.

Ahmed Alshemi