NEUTRON SCATTERING STUDIES OF $\rm La_{2-x}Ba_xCuO_4$

NEUTRON SCATTERING STUDIES OF La_{2-x}Ba_xCuO₄

By

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Abstract

Elastic neutron scattering studies have been performed on high quality single crystal samples of lightly doped La_{2-x}Ba_xCuO₄ with concentrations of x=0, 0.006, 0.0125 and 0.025. These samples display x dependent magnetic structures with transition temperatures which closely follow the usual phase diagram for this material. A low temperature, 2D antiferromagnetic transition has been observed in samples with hole concentration near the 'spin-glass' transition at x~0.02. In data collected on the N5 triple axis spectrometer at Chalk River, the sample with x=0.0125 demonstrates the onset of 3D antiferromagnetism with $T_N \sim 151$ K, followed by attenuation of the scattering intensity of the (1/2,1/2,0) magnetic Bragg peak below T=21K, indicating the onset of 2D magnetic ordering. This result is similar to that obtained by Matsuda et al. in lightly doped La_{2-x}Sr_xCuO₄.[1] This result is confirmed by direct observations of 2D 'rods' of magnetic scattering in time-of-flight measurements performed on the Disk Chopper Spectrometer at NIST.

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Chapter 1

Introduction

The discovery[2] of high temperature superconducting materials in the latter part of the twentieth century has stimulated a great deal of activity in the field of condensed matter physics. In particular, the cuprate family of superconductors has been a highly productive area of study and includes materials such as YBa₂Cu₃O₇ and Bi₂Sr₂CaCu₂O₈ with their astoundingly high transition temperatures of T_C~100K.[3][4] The first of the layered cuprate materials, La_{2-x}Ba_xCuO₄ (LBCO), discovered in 1986 by Bednorz and Müller, has been studied relatively little.[5][6] This is owing to the rather difficult process of crystal growth that made high quality single crystal samples rare, if not non-existent during the first decade after their discovery. With the advent of the floating-zone image furnace technique, the growth of large single crystal LBCO samples has been made possible.[2]

For many years, experiments have clearly demonstrated a relationship between the structural and magnetic properties of these materials and the superconducting phase.[7][8] Both LBCO and its well studied sister material, $La_{2-x}Sr_xCuO_4$ (LSCO), exhibit the usual characteristics of the cuprate superconductors including the 1/8 anomaly in which the superconducting transition temperature is suppressed at a Ba or Sr concentration of x=1/8.[9] This and other behaviours including the high T_c phenomenon itself, have remained unexplained for over two decades. However, recent progress has been made in exploring the interplay of various properties of these materials through the use of neutron scattering.[10][1][11] Changes in the phase of the material are directly tied to the static magnetic structure. Recent studies have demonstrated changes in the incommensurate magnetic structure as the material is doped into the superconducting regime.[12] The cause underlying this and other phenomena is highly controversial, and ultimately this study seeks to contribute to the determination of the mechanism of high temperature superconductivity in the cuprates. By studying the evolution of magnetic scattering in lightly doped, non-superconducting LBCO, this study will help elucidate the unusual behaviour of this interesting and poorly understood family of novel materials.

Due to the highly penetrating nature of the neutron, neutron scattering experiments, unlike x-ray diffraction, require relatively large samples in order to produce a sufficiently large coherent signal. Since the late 1980s, crystals of LSCO of appropriate size have been grown for such a purpose using a variety of different methods. This has made LSCO a well studied material in the field of neutron scattering. Subtle differences in the phase diagrams of LBCO and LSCO, however, demonstrate that these studies do not provide a sufficient amount of data on the doped La-214 materials to explain their behaviour in general. While LSCO and LBCO have similar electronic and chemical structures, the choice of dopant has a significant effect on the behaviour of the material.

Single crystal samples of underdoped $La_{2-x}Ba_xCuO_4$ have been prepared using the floating-zone image furnace technique including pure La_2CuO_4 (x=0) and x = 0.006 as well as electron doped $La_{2-x}Ce_xCuO_4$ with concentrations of x = 0.0125 and x = 0.025. Each of these samples has been characterized using neutron scatter-

ing techniques as well as SQUID magnetometry. The magnetic structures of these materials have been probed as a function of temperature using neutron scattering. In particular, elastic magnetic scattering has been observed utilizing the N5 triple axis spectrometer at Chalk River National Laboratories in Ontario as well as the DCS time-of-flight spectrometer at the National Institute of Standards and Technology in Maryland. This document will describe the growth of several samples, and the subsequent neutron studies that have been conducted.

Chapter 2

$La_{2-x}R_xCuO_4$

2.1 Introduction

In this chapter, background material pertaining to chemical structure, magnetism and preparation of high quality single crystals of $La_{2-x}Ba_xCuO_4$ and $La_{2-x}Ce_xCuO_4$ will presented. For this study, low dopings of LBCO and LCCO have been grown for use in neutron scattering experiments. The organization of this chapter will include a section detailing the structure of the parent material and the effects of doping. This will be followed by a discussion of the magnetic properties of these materials. Finally, the process of material preparation and crystal growth will be outlined.

2.2 Chemical Structure

Lanthanum cuprate has a simple, layered chemical structure which is unlike most other materials in the cuprate family. Figure 2.1 a) shows the crystal structure of the parent material La_2CuO_4 . The crystal is composed of alternating layers of copper and lanthanum oxide. This creates an orthorhombic unit cell with spacegroup (*Bmab*) with lattice parameters a = 5.354Å, b = 5.401Å, c = 13.153Åwhere c is oriented along the stacking direction.[5] During the synthesis of this material, the lanthanum atoms become oxidised, donating valence electrons to the bonding of the CuO₂. When a dopant is introduced to produce a La_{2-x}R_xCuO₄ type material, the new ions (R) are substituted directly onto the lanthanum sites in the lattice. The substitution of a Ba²⁺ ion for a La³⁺ in the lattice effectively removes an electron, or adds a nonmagnetic hole into the system. This would seem to indicate a priori that the crystal structure would remain unchanged. However, this is not always the case as will be discussed later in this section. Typically, this type of doping is achieved using Sr²⁺ as the dopant. This is due to the fact that it is simply easier to produce single crystals of LSCO. Although single crystals of LBCO are more difficult to acquire, there is a need to study them in order to produce a complete picture of superconductivity in this family of cuprates. Figure 2.1 b) shows the general phase diagram for LSCO with the antiferromagnetic and superconducting domes labeled as well as the orthorhombic and tetragonal phases.

Doping the lattice with barium produces two additional structural phases on either side of the orthorhombic phase which has become known as the middle temperature orthorhombic (MTO) phase. A high temperature tetragonal (HTT) phase (I_4/mmm) occurs for which the transition temperature is highly doping dependent. For low dopings, this transition occurs well above room temperature but is suppressed to T_c=305K for x=0.08.[7] Upon cooling, the sample undergoes a HTT to MTO transition which is a continuous, second order transition. The low temperature tetragonal (LTT) phase (P_{4_2}/ncm) appears at the same doping as the onset of superconductivity although the transition temperatures are not the same. Unlike the HTT to MTO, the MTO to LTT transition is a sharp, first order transition.[7] The structural as well as superconducting transitions are labeled in figure 2.2 a), which shows the phase



Figure 2.1a: Diagram of the unit cell of $La_{2-x}Cu)_4$ showing the simple layered structure of this material. The diagram shows the unit cell in tetragonal notation with the orthorhombic lattice parameters shown as running diagonally from Cu to Cu.[5]



Figure 2.1b: Phase diagram for LSCO which shows all of the relevant phases applicable to the hole doped lanthanum cuprates. [13]

diagram specific to LBCO. The interrelation between the LTT and superconducting phases remains unclear; however, it does demonstrate a clear connection between the structural and magnetic properties of these materials.

Single crystal samples of lightly doped $La_{2-x}Ce_xCuO_4$ have also been grown with cerium concentrations of x=0.0125 and x=0.025. In these crystals, Ce⁴⁺ is substituted onto the La³⁺ sites, effectively introducing an additional electron into the system. Electron doping of La₂CuO₄ is a less prominently studied area of doped lanthanum cuprates. In these materials, the antiferromagnetic region persists over a much larger range of dopings and the superconducting dome is much smaller than in the hole doped case. The antiferromagnetic region is shown on the left side of the phase diagram in figure 2.2 b). A structural determination has been conducted using x-rays which confirms that these crystals do indeed belong to the same MTO phase as the parent material at room temperature. Further studies are required to determine the effect of doping with Ce⁴⁺ ions on the chemical structure of this material.

2.3 Magnetic Structure

The undoped parent material La₂CuO₄ is a classic example of a Mott insulator. The antiferromagnetism within the system is due to the S=1/2 moments on each of the Cu²⁺ ions within the CuO₂ plane after bonding. The organization of these in-plane spins within the basal plane of the tetragonal lattice is an excellent example of the 2D square lattice quantum Heisenberg antiferromagnet. This has motivated many groups to thoroughly study the pure material in order to understand the Hamiltonian for the exchange interaction. [5] While a great deal of interesting behaviour is associated with the in-plane interaction, there exists another significant magnetic interaction within this material. This second interaction is a relatively weak antiferromagnetic



Figure 2.2a: Phase diagram for LBCO. Both the structural transitions (T_{d1} and T_{d2}) and the superconducting transition (T_C) are shown in this diagram. [7][14]



Figure 2.2b: Phase diagrams for electron doping and oxygen doping in the cuprates. The antiferromagnetic region in the electron doped materials persists over a wide range of concentrations. The oxygen doped case displays the same antiferromagnetic dome as hole doping with Sr or Ba. [5]

exchange that occurs between corresponding copper ions in adjacent CuO layers. In the undoped material, both of these interactions lead to a 3D antiferromagnetic structure below $T_N=325$ K.[5]

The antiferromagnetic phase is readily disrupted by the introduction of Ba into the lattice which induces holes in the CuO_2 plane. The Neel temperature drops sharply with x and the system enters a 'spin-glass' phase for x > 0.02. The results reported in Chapter 4 and 5 support the existence of a 2D antiferromagnetic phase for Ba x > 0.02as an alternative to the notion of a spin-glass phase in this material. At low temperatures in this region of the phase diagram, the antiferromagnetic ordering becomes restricted to the basal plane, leaving the system in a layered, 2D antiferromagnetic state. Magnetic neutron scattering studies have show evidence of such a transition.[1] As the material enters this phase, the magnetic Bragg peaks that develop during the onset of 3D AFM order experience a decline below 25K. This can be interpreted as the 3D antiferromagnetic Bragg peaks becoming 2D. Since the 2D AFM order is left intact in the CuO_2 plane, this 'incoherent'scattering is confined to rods running parallel to c^{*}. While the evidence suggests this is the case, no direct measurements of rod-like scattering have been reported in the literature. In Chapter 5, the direct observation of rod-like scattering is reported in samples with Ba concentrations of x=0.0125 and x=0.035.

Of course, hole doping also influences the magnetic ordering within the CuO_2 plane itself. Magnetic Bragg peaks display static incommensurate order in the doped samples which increases with hole concentration.[12] The non-magnetic holes have the effect of breaking the long range AFM order. The specific mechanism behind this disruption has not been determined and has been the subject of debate for several years.

There are two predominant models used to describe the effect of hole doping on the

antiferromagnetism in this material. The first is the stripe picture in which the holes align into quasi-1D stripes in the CuO₂ plane. These stripes serve as boundaries between antiferromagnetic domains which induces a π phase shift in the magnetic ordering along the domain edges.[15][16] This creates magnetic ordering which is incommensurate with respect to the crystal lattice. Neutron scattering experiments are effective for observing this incommensuration around the (1/2,1/2,0) Bragg positions. [10][17] The incommensuration increases dramatically with hole concentration until finally the peaks become colinear, rather than diagonal with respect to the tetragonal axes, at higher doping levels.

The competing model that accounts for the incommensurate structure is known as the itinerant picture. In this picture, fermi surface nesting accounts for the incommensurability in the magnetic structure where flat pieces of the Fermi surface can be connected by neutron scattering at an appropriate wavevector \vec{Q} .

While the cuprates are, in general, a well studied family of high T_c superconductors, the details of the superconducting mechanism still remain unexplained. Studying the magnetic structure of these materials may provide the clues necessary to construct a holistic picture of superconductivity and exotic magnetism in the cuprates.

2.4 Sample Preparation & Crystal Growth

High quality single crystal samples of LBCO and LCCO have been grown using the floating-zone image furnace technique. Crystals are grown from ceramic rods prepared from stoichiometric quantities of the reactants. Samples of pure La_2CuO_4 , $La_{1.994}Ba_{0.006}CuO_4$, $La_{1.9875}Ce_{0.0125}CuO_4$ and $La_{1.975}Ce_{0.025}CuO_4$ have been grown using this technique. For the pure material, a mixture of powdered La_2O_3 and CuO_2 in the proportions 1:1 was prepared and mixed for several hours in a ball mixer. Since La_2O_3 is extremely hygroscopic, a large quantity of it was dried in an oven at 1200°C for 12 hours before each mixture was prepared. Although it is much less hygroscopic than La_2O_3 , a large amount of CuO was also dried at 800°C to ensure the powder contained as little H₂O as possible.

A pressed powder rod was then made by filling a rubber balloon with 35g of the mixture and applying ~ 1.2 MPa of pressure using a hydrostatic press for 20 minutes. The rods are typically 12cm in length which provides ample material for a crystal growth. The rod was then transferred to a furnace and annealed at 1150°C for 12 hours in order to sinter the powder into a ceramic. Rods of the LCCO materials were prepared using a similar method and the addition of CeO_2 as a starting material. Preparations with Ce dopings of x=0.0125 and x=0.025 were produced using La_2O_3 : CeO₂: CuO as starting materials in the ratios of 1.9875: 0.0125:1 and 1.975:0.025:1 respectively. Although the mixtures are prepared as accurately as possible, the precise doping level of the final product cannot be easily determined at this stage. The orthorhombicity of the final material can be used as a more accurate test of the final concentration due to the high sensitivity of the lattice parameters to the quantity of dopant. An additional 1.5% CuO is added and mixed for all individual rod preparations to compensate for evaporation during crystal growth. To balance the reaction, additional O_2 is supplied to the material from the atmosphere during annealing and in the sample environment during crystal growth.

Once a rod has been annealed, it is immediately ready for growth. The floatingzone image furnace is an optical furnace which uses two high powered halogen lamps to heat the material. The energy from the lamps is focused onto a small region of the ceramic rod using two parabolic mirrors. The growth environment in enclosed by a transparent quartz tube that can be pressurized with a gas that is appropriate for a particular material. For all samples of LBCO and LCCO the chamber is pressurized to ~180kPa with dry $O_{2(gas)}$. The ceramic rod is fed into the growth chamber from the top of the quartz tube using a steel feed rod from which the growth rod is hung using platinum wire. A seed, which is composed of a small piece of sintered rod or previously grown crystal with similar composition, serves as a base on which to grow the crystal. It is mounted on a second steel feed rod at the bottom of the chamber which holds the seed in place, directly beneath the growth rod within the sample environment. A small quantity of flux (~3g) is applied to the top of the seed to serve as a solvent to facilitate more rapid melting of the growth rod. The flux is a powdered mixture composed of La₂O₃ and CuO in the ratio 1:3.

By focusing the energy from the lamps onto a small area, the growth can be conducted in a controlled manner. The materials are melted by gradually increasing the power output of the lamps until a small, liquid zone is established between the seed and the ceramic rod. Once the zone is established, and an appropriate rate of melting is achieved, the lamps are moved slowly up the growth rod, causing the liquid zone to solidify into a single crystal at the bottom, and liquify more of the growth rod. In this manner, the entire growth rod is melted, bit by bit, and crystalized as the zone travels up the ceramic rod. The rate of motion of the lamps generally varies from material to material. For these materials in particular, a rate of 1mm/hour or less is required to ensure good crystalization of the final product. The maximum displacement of the lamps is 100mm, meaning that the average growth takes roughly four days.

The size of the crystals is restricted by the limits of the vertical translation of the lamps in the furnace. Under optimal conditions, a growth can be as long as 10cm and in general has a width of 4-6mm. However, as mentioned above, La_2O_3 is extremely hygroscopic and if any is left unbonded within the crystal, it will readily absorb water from the atmosphere. As a result, an apparent single crystal can break into segments and partially deteriorate back into a powder. With these materials, it is inevitable that some portion of a crystaline sample will break down in this way. Once this has occurred, the remaining single crystal is generally stable and can be stored in a sealed vessel containing a desiccant. Typically, a crystal will decay into one or more fragments which can be anywhere from 2-6cm in length. The quality of these crystals is unknown once the growth is complete and the pieces have stabilized. The crystals must then be characterized using SQUID magnetometry and x-ray or neutron diffraction to more precisely determine their phase and mosaicity.

2.5 Sample Characterization

The preparation of these materials requires as much precision as possible during synthesis. Due to the limitations of the process, however, there still exists some element of chance concerning the final composition and quality of the crystals. Once a single crystal has been grown and has stabilized, there are several methods that can shed light on some of the uncertainties in the final product. As mentioned in the previous section, it is not possible to precisely control the exact doping level of the material through stoichiometric preparation alone. Evaporation of CuO and the uniformity of the powdered mixture can have a significant effect on the doping in the final product. Measuring the orthorhombicity and determining the magnetic phase of the material can be highly effective ways of establishing accurate values of x.

The highly concentration dependent phase diagram of LBCO can be used in conjunction with SQUID magnetometry and structural determination in order to establish the phase of a particular crystaline sample. Crystals have previously been prepared and characterized using these techniques with concentrations of x=0.035, x=0.025, x=0.0125.[12][7] New crystals have been grown with concentrations of Ce x=0.0125, x=0.025, Ba x=0.006 as well as two samples of pure La₂CuO₄.

A known issue with crystals of this type is that they are highly sensitive to oxygen doping. Since all of these crystals have been grown in pressurized oxygen, there is some likelyhood that they contain some quantity of excess oxygen. Since this type of doping has the same effect as hole doping with Ba, the effect of excess oxygen may be to negate the contribution of electrons from the Ce atoms. The evidence from the susceptibility measurements does not suggest that this is the case; however, a more in depth study may be required. Since all crystals were grown in similar environments, it is likely that the oxygen doping is at least consistent accross all Ba doped samples and thus has a minimal effect on this study.

For all relevant experiments, crystals were aligned using the E3 triple axis spectrometer at the Canadian Neutron Beam Centre at the Chalk River National Labs. During these alignments, several mesh scans were performed in order to determine the quality and mosaicity of the crystals. Due to their relatively low resolution, in general, these scans give limited information. They allow the quality of each crystal to be determined to a certain extent and reveal the number of grains present. While most crystals were oriented without issue, one of the two pure La_2CuO_4 samples was found to contain several, randomly oriented grains. While pure sample #1 was found to have a relatively small mosaic spread, pure sample #2 was found to be inappropriate for neutron scattering studies.

The samples are mounted on aluminum stands which have moveable joints to allow for minor adjustments of the alignment of the crystal (See figure 2.3). The samples are held to the stands using thin aluminum straps. The stands are held onto aluminum base plates with aluminum screws. The base plates are designed specifically for the mounting systems at either Chalk River or NIST. For some samples, it was necessary to use an additional aluminum adaptor plate which allows a Chalk River base plate to be mounted in a DCS sample space and vice versa.



Figure 2.3: Single crystal LBCO x=0.025 mounted on a Chalk River base plate. The white markings indicate the c direction. This sample has been aligned for scattering in the (HHL) plane.

Chapter 3

Principles of Neutron Scattering

3.1 Introduction

In this chapter, all relevant information pertaining to the theoretical aspects of neutron scattering will be discussed. In the first section, the properties and production of neutrons will be discussed. This will be followed by a section describing the theory of triple axis spectrometry. Finally, time-of-flight neutron scattering will be discussed.

3.2 The Neutron

Discovered by Chadwick in 1935, the neutron has had an immense impact on the world of science over the past several decades. In the field of condensed matter physics, the neutron has been invaluable and has facilitated a great deal of progress and discovery.

Neutron scattering experiments are made possible by the wave-particle duality inherent in all massive particles. The wavelength of such a particle is defined by the De Broglie equation which depends on both the mass and the energy of the particle in question. It is particularly advantageous that the wavelengths of thermal neutrons tend to be on the same length scale as the interatomic spacing of the lattice in most solids. The relationship between the energy, temperature, mass and wavelength of the neutron is given by

$$E = \frac{h^2}{2m\lambda^2}.$$
(3.1)

The second main advantage of the neutron is that it is charge neutral. Since it does not experience any Coulombic interaction with either the electron cloud or the nucleus, the neutron is also an extremely penetrating form of radiation. This means that a neutron can not only penetrate deeply into a solid mass, such as a large crystal, but that the neutron can interact directly with the nucleus. The intrisic S=1/2 spin of the particle also makes it an excellent probe of magnetic structure through the electron-neutron, dipole-dipole interaction.

Generally, there are two methods of producing neutrons. A linear accelerator can be used to propel protons at a heavy metal target. When the protons collide with the target, neutrons are produced through the process of spallation. More commonly, neutrons are produced through the use of a fission reactor. In a typical reactor core, U^{235} undergoes fission and splits into fission products which include a high energy neutron. Such a neutron is not immediately useful for scattering as it is far too high in energy, and thus low in wavelength, to be useful for the study of materials. A moderator is used as a way to control the energy of the neutrons produced by the reaction. The moderator is simply a medium through which the neutrons pass that is kept at a desired temperature. As the neutrons pass through the moderator, they thermally equilibriate with the surrounding medium, lowering their energy to a useful level. Another advantage that neutrons have over x-rays is contrast in scattering from material to material. X-ray scattering depends directly on the density of the electron cloud of an atom. Thus atoms with high atomic numbers, and therefore denser electron clouds, readily scatter x-rays whereas a light atom like hydrogen will be a very poor x-ray scatterer. Since the neutron does not directly interact with the electron cloud, the strength of the interaction of a neutron with a particular element depends on the interaction with either the nucleus via short range interactions or via magnetic scattering. The strength of the scattering is determined by the neutron scattering cross-section, which will be discussed in the next section. It is important to note that the scattering length of an element does not vary uniformly across the periodic table. For example, hydrogen is an excellent scatterer of neutrons as compared to x-rays.

While the neutron shares a number of properties with the x-ray, they provide a great deal of complimentary information pertaining to a material. While x-ray diffraction may give detailed structural information, neutron scattering can provide details about the magnetic structure and dynamics of the lattice using similar scattering techniques.

3.3 Elastic Neutron Scattering

The derivation of scattering cross-sections and Bragg's law in this chapter follows G.L. Squire's work *Introduction to the Theory of Thermal Neutron Scattering*.[18] For a given flux of neutrons in a neutron beam, a sample will scatter some number of these neutrons per second in all directions. This value is known as the total scattering cross-section and is denoted by σ_{tot} . Rarely, if ever, does a detector collect all neutrons scattered in all directions for all values of energy. Thus a partial differential cross-section is defined as the number of neutrons scattered into an element of solid angle

 $d\Omega$ with some small range of energy dE. This is written as

$$\frac{\delta\sigma^2}{\delta\Omega\delta E}.$$
(3.2)

By integrating over energy, the total number of neutrons per second in a given solid angle can be recovered using the following form

$$\frac{d\sigma}{d\Omega} = \int_0^\infty \frac{\delta\sigma^2}{\delta\Omega\delta E} dE.$$
(3.3)

Thus the total scattering cross-section can also be found by integrating over all directions

$$\sigma_{tot} = \int \frac{d\sigma}{d\Omega}.$$
(3.4)

A single neutron moving through space can be represented by the wave equation

$$\Psi_i = exp(i\vec{k}\vec{r}) = exp(ikz) \tag{3.5}$$

since the direction of propagation can be taken to be z for simplicity. When the neutron comes into contact with a nucleus fixed at the origin, it is scattered with spherical symmetry due to the relatively large wavelength of the typical neutron which is $\sim \text{Å}$. Thus the wavefunction for the scattered neutron can be written as

$$\Psi_s = \frac{-b}{r} exp(i\vec{k}\vec{r}). \tag{3.6}$$

In order for the wavefunction to remain dimensionless, b must have the units of length. This value is known as the scattering length and is a unique constant for each isotope and its individual spin state. Using the wavefunctions for the incident and scattered neutrons, the scattering cross-section can be calculated for a single nucleus. The flux of scattered neutrons can be written as the product of the probability distribution, velocity and area through which the neutrons pass. Thus $v|\Psi_s|^2 dA = vb^2 d\Omega$ and the flux of the incident neutrons is given simply by v. This coupled with the definition of the differential cross-section gives the fundamental result that

$$\sigma_{tot} = 4\pi b^2. \tag{3.7}$$

In order to derive the partial differential cross-section for the general case, Fermi's Golden Rule must be used. In this case, we treat the system as a collection of neutrons which are scattered from an initial state defined by λ_i , \vec{k}_i to a final state with λ_f , \vec{k}_f . Using this set of conditions and summing over the set of all neutrons that are scattered in this way, the differential cross-section becomes

$$\frac{d\sigma}{d\Omega} = \frac{1}{\Phi d\Omega} \sum_{\vec{k}} W_{\lambda_i, \vec{k}_i \to \lambda_f, \vec{k}_f}.$$
(3.8)

By applying Fermi's Golden Rule, the differential cross-section becomes

$$\frac{d\sigma}{d\Omega} = \frac{k_f}{k_i} (\frac{m}{2\pi\hbar^2})^2 |\langle \vec{k}_f \lambda_f | V | \vec{k}_i \lambda_i \rangle|^2.$$
(3.9)

In order to produce the general partial differential cross-section that is used to describe general scattering, a term must also be introduced to represent the range in energy across which the neutrons are distributed in a given cross-section.

$$\frac{\partial^2 \sigma}{\partial \Omega \partial E} = \frac{k_f}{k_i} (\frac{m}{2\pi\hbar^2})^2 |\langle \vec{k}_f \lambda_f | V | \vec{k}_i \lambda_i \rangle|^2 \delta(E_{ni} - E_{nf} + E_{si} - E_{sf}). \tag{3.10}$$

The delta function in equation 3.10 represents the conservation of energy for the neutron and scattering system. The introduction of this partial derivative is valid due to the following property of the delta function;

$$\int_{-\infty}^{\infty} \delta(E_{ni} - E_{nf} + E_{si} - E_{sf}) dE = 1.$$
 (3.11)

It is more common for the partial differential cross section to be written in terms of the scattering vector \vec{Q} and the time dependent Heisenberg operator $\vec{R}(t)$, where \vec{R} is the displacement vector between the neutron and the scattering body. The potential can be written in terms of $\vec{Q} = \vec{k}_i - \vec{k}_f$ by rewriting the interaction such that

$$\langle \vec{k}_f \lambda_f | V(\vec{r}) | \vec{k}_i \lambda_i \rangle = \langle \lambda_f | V(\vec{Q}) exp(-i\vec{Q}\vec{x}) | \lambda_i \rangle.$$
(3.12)

The final step involves representing the delta function for energy conservation as a function of time. This is done by using the integral of a plane wave to represent the delta function.

$$\delta(E_{ni} - E_{nf} + E_{si} - E_{sf})dE$$

$$= \delta(E_{ni} - E_{nf} + \hbar\omega)dE$$

$$= \frac{1}{2\pi\hbar} \int_{-\infty}^{\infty} exp((E_{ni} - E_{nf})t/\hbar)exp(-i\omega t)dt.$$
(3.13)

Using eq. 3.10, 3.12 and 3.13, the partial differential cross section can be written as

$$\frac{\partial^2 \sigma}{\partial \Omega \partial E} = \frac{k_f}{k_i} \frac{1}{2\pi\hbar} \sum_{ij} b_i b_j \int_{-\infty}^{\infty} \langle exp \left\{ -i\vec{Q} \cdot \vec{R}_i(0) \right\} exp \left\{ -i\vec{Q} \cdot \vec{R}_j(t) \right\} \rangle \\ \times exp \left\{ -i\omega t \right\} dt$$
(3.14)

Where the sum is taken over all scatterers in the system. Due to the variation of spin states and isotopes of a particular element, each b_i will vary from nucleus to nucleus, even in a crystal composed of a single element. In a typical scattering system, the number of nuclei is extremely large. This means that, for simplicity, the scattering cross-section can be written in terms of the the average scattering length \bar{b} . This allows eq 3.14 to be written as

$$\frac{\partial^{2}\sigma}{\partial\Omega\partial E} = \left\{ \left[\frac{k_{f}}{k_{i}} \frac{(4\pi(\bar{b})^{2})}{4\pi} \frac{1}{2\pi\hbar} \right] \sum_{i\neq j} \int_{-\infty}^{\infty} \langle exp\left\{ -i\vec{Q}\cdot\vec{R}_{i}(0) \right\} exp\left\{ -i\vec{Q}\cdot\vec{R}_{j}(t) \right\} exp\left\{ -i\omega t \right\} dt \right\} + \left\{ \left[\frac{k_{f}}{k_{i}} \frac{(4\pi[(\bar{b}^{2}) - (\bar{b})^{2}])}{4\pi} \frac{1}{2\pi\hbar} \right] \sum_{ij} \int_{-\infty}^{\infty} \langle exp\left\{ -i\vec{Q}\cdot\vec{R}_{i}(0) \right\} exp\left\{ -i\vec{Q}\cdot\vec{R}_{j}(t) \right\} exp\left\{ -i\omega t \right\} dt \right\} \right\}$$

$$(3.15)$$

The first term in this equation is the coherent scattering cross-section. In a real neutron scattering experiment, this term gives the interference effects which are necessary to produce Bragg scattering. For a perfect neutron scattering system, $b = \bar{b}$ for all nuclei, and every neutron would contribute to the coherent scattering. The second term represents the incoherent scattering which is the scattering contribution from nuclei which deviate from the definition of a 'perfect'scattering system; that is nuclei with $b \neq \bar{b}$.

3.3.1 Bragg Scattering

Bragg scattering is produced by the specular reflection of some form of radiation from a series of parallel planes of atoms in a crystal lattice. The incoming radiation reflects off of successive planes, causing a variation in the total path length travelled by the radiation. For radiation of wavelength λ and set of planes with spacing d, there will be some reflecting angle θ for which the difference in path length between reflections produces a phase shift of 2π causing constructive interference of the reflected waves. This is the nature of Bragg's Law and is written as

$$2dsin\theta = n\lambda \tag{3.16}$$

The condition for Bragg scattering can be derived from the coherent scattering term in eq. 3.15. In a crystal lattice, a lattice translation vector is given by

$$\vec{T} = a\vec{x} + b\vec{y} + c\vec{z} \tag{3.17}$$

where a, b and c are integers. All nuclei vibrate around their equilibrium position with some thermal motion. Thus the actual diplacement vector for a given nucleus will be

$$\vec{R} = \vec{T} + \vec{u} \tag{3.18}$$

where \vec{u} is the thermal displacement from the equilibrium location. Due to the translational symmetry of any Bravais crystal lattice, the sum over all scatterers given in eq. 3.14 depends only on $\vec{T_i} - \vec{T_j}$. So, for all nuclei with $\vec{T_j}$ the sum over $\vec{T_i}$ will be identical. Therefore the coherent scattering term can be written as

$$\frac{d\sigma}{d\Omega_{coh}} = \left[\frac{k_f}{k_i} \frac{(4\pi(\bar{b})^2)}{4\pi} \frac{1}{2\pi\hbar}\right] \times N \int_{-\infty}^{\infty} \langle exp\left\{-i\vec{Q}\cdot\vec{u}(0)\right\} exp\left\{-i\vec{Q}\cdot\vec{u}(t)\right\} \rangle exp\left\{-i\omega t\right\} dt$$
(3.19)

The condition for Bragg scattering can be obtained by writing eq. 3.19 in terms

of the temperature factor and by integrating over energy to yield.

$$\frac{d\sigma}{d\Omega_{coh}} = \frac{\sigma_{coh}}{4\pi} N \frac{(2\pi)^3}{V} exp\left\{ \langle [\vec{Q} \cdot \vec{u}(0)]^2 \rangle \right\} \sum_{\vec{Q}'} \delta(\vec{Q} - \vec{G})$$
(3.20)

where \vec{G} is a reciprocal lattice vector with magnitude $G = n(2\pi)/d$. The delta function gives the condition for Bragg scattering since it implies that $\vec{Q} = \vec{k}_i - \vec{k}_f = \vec{G}$. For an elastic scattering process, $k_i = k_f$ which gives the result

$$2\vec{k}\cdot\vec{G} = G^2 \tag{3.21}$$

which can be written as

$$2dsin\theta = n\lambda \tag{3.22}$$

Hence, both formulations lead to Bragg's Law. [19]

3.4 Magnetic Scattering

The neutron possesses a magnetic dipole moment given by

$$\vec{\mu}_n = -\gamma \mu_N \vec{\sigma} \tag{3.23}$$

where $\gamma = 1.913$, $\mu_N = e\hbar/2m_p$ and $\vec{\sigma}$ is the Pauli spin operator. This yields a magnetic potential V_m which causes neutrons to scatter from magnetic ions. Similarly for the electron

$$\vec{\mu}_e = -2\mu_B \vec{s} \tag{3.24}$$

with \vec{s} as the spin operator for the electron and $\mu_B = e\hbar/2m_e$. An electron with momentum \vec{p} produces a magnetic field via the Biot-Savart law. The potential of the neutron in the fields produced by both the momentum and dipole moment of the electron is given by

$$V_m = -\frac{\mu_0}{4\pi} \gamma \mu_N 2\mu_B \vec{\sigma} \cdot \left\{ \nabla \times \left(\frac{\vec{s} \times \hat{R}}{R^2} \right) + \frac{1}{\hbar} \left(\frac{\vec{p} \times \hat{R}}{R^2} \right) \right\}$$
(3.25)

Substituting this potential into eq. 3.10 yields

$$\frac{\partial^2 \sigma}{\partial \Omega \partial E} = \left(\frac{\gamma \mu_0 e^2}{4\pi m_e}\right)^2 \frac{k_f}{k} |\langle \sigma_f \lambda_f | \vec{\sigma} \cdot \vec{Q}_\perp | \sigma_i \lambda_i \rangle|^2 \times \delta(E_{\lambda_i} - E_{\lambda_f} + \hbar\omega)$$
(3.26)

where the operator \vec{Q}_{\perp} contains terms representing both the dipole-dipole and dipole-current interactions.

$$\vec{Q}_{\perp} = \sum_{i} exp\left\{i\vec{Q}\cdot\vec{r}_{i}\right\} \left[\hat{Q}\times(\vec{s}_{i}\times\hat{Q}) + \frac{i}{\hbar Q}(\vec{p}_{i}\times\hat{Q})\right].$$
(3.27)

The cross products in the operator indicate that a magnetic scattering interaction will only occur if the magnetization of the ion involved is perpendicular to the scattering vector \vec{Q} .

3.5 Triple Axis Spectrometry

The triple axis spectrometer was developed by Bertram Brockhouse in the 1950s for which he won the Nobel Prize in 1994. Figure 3.1 shows a schematic diagram for a triple axis spectrometer. This type of spectrometry utilizes an intense, continuous beam of neutrons produced by a reactor. Three rotating stages are used to control the scattering angles of the monochromator, sample, and the analyser crystal, as shown in the figure. Thermal neutrons exit the reactor with a Maxwellian distribution of energies. The monochromator, which is usually made of pyrolytic graphite, makes use of the Bragg scattering condition in order to select a beam of neutrons with a specific λ . Monochromators can also be made from Si, Ge or Cu. Neutrons with wavelengths of λ/n for n = 1, 2, 3... also meet the condition for Bragg reflection given in eq. 3.16 and so a nitrogen cooled Be filter can be placed in the beam after the monochromator to eliminate any higher order scattering that has made it passed the monochromator. After passing through a collimator, the beam is then scattered by the sample and proceeds to the analyser crystal.

The analyser crystal is a second monochromator which allows scattered neutrons of a particular λ to pass through to the detector. During an experiment, the analyser and the monochromator are usually set to fixed positions. This effectively fixes the values of k_i and k_f . The scattering vector \vec{Q} can then be controlled by changing the relative directions of \vec{k}_i and \vec{k}_f . The final and incident energies can also be changed while \vec{Q} remains constant. This method allows the energy and \vec{Q} resolution of an experiment to be finely tuned.

3.6 Time-of-Flight Spectrometery

Time-of-Flight spectrometers measure the time it takes for a neutron to pass through a sample and into the detector in order to determine the final energy of the neutron. This type of spectrometer makes use of a large bank of neutron detectors in order to capture as much of the scattered signal as possible. By measuring the flight time of the neutron, the energy transferred to or from a crystal lattice can be determined. Also, by measuring the scattering angle of a neutron, the momentum transfer can be



Figure 3.1: Schematic depiction of a triple axis spectrometer with each 'axis 'labeled determined. In this way, a ToF spectrometer is capable of detecting the elastic and inelastic scattering simultaneously.

Figure 3.2 show a schematic diagram of a ToF spectrometer. Neutrons emerge from the source as a white beam which travels down the beam guide to the choppers. The DCS at NIST operates with a cold neutron source. The choppers are used to filter out all neutrons but those with a specific value of λ , pulse the neutron beam and remove higher order neutrons. Pulsing the beam effectively defines t=0 from which the instrument determines the total flight time of the neutron. The Disk Chopper Spectrometer (DCS) at NIST uses a set of seven disk choppers to



Figure 3.2: Schematic diagram of a time-of-flight spectrometer with choppers.

Detectors

control the wavelength of neutrons used in an experiment. Each chopper is a disk with a rectangular slit cut into it to allow neutrons to pass through. Several of these choppers are placed in succession and are rotated with relative phases. Only a neutron with a particular velocity is be capable of passing through the slits in each disk and proceeding to the sample. These choppers replace the monochromator used in the triple axis spectrometer and have the added advantage of eliminating higher order neutrons at the same time.
Chapter 4

Triple Axis Neutron Scattering Study of Lightly Doped La_{2-x}Ba_xCuO₄

4.1 Introduction

Neutron scattering experiments have been performed using the N5 Triple Axis spectrometer at Chalk River Nuclear Laboratory. The purpose of this study was to acquire high resolution scattering data concerning the shape and intensity of the magnetic Bragg features at the (1/2,1/2,0) position in reciprocal space for several samples of LBCO. Four single crystals of LBCO and one crystal of LCCO were studied in order to examine the temperature dependent evolution of the magnetic structure across the 3D antiferromagnetic to 'spin-glass' regions of the phase diagram.

The samples were aligned using the E3 triple axis alignment spectrometer at the CNBC. All samples were aligned with the (HK0) plane coincident with the horizontal scattering plane in tetragonal notation. Samples were mounted and cooled using

a displex closed-cycle refrigerator with He as an exchange gas. Thermal neutrons with a fixed final energy of $E_f=1.23$ THz ($\lambda = 4$ Å) were used in combination with tight collimation in order to achieve high resolution. The magnetic scattering in this system is weak due to the small moment (S=1/2) on each Cu²⁺ ion. A liquid nitrogen cooled Be filter was used to eliminate contamination due to higher order scattering ($\lambda/2$) which can be problematic considering the relatively high intensity of the (1,1,0) Bragg peak. Several samples were run over the course of five weeks in November and December 2008, and March and April 2009.

Highly detailed mesh scans were compiled with counting times of 1min/point. These maps of the magnetic Bragg peaks were used to analyse the peak shape and track any incommensurability as a function of doping. The (1/2,1/2,0) magnetic peaks were profiled in this way for several temperatures in each sample. Once the temperature dependence of the peak profile had been roughly determined, several locations in and around the magnetic Bragg scattering were tracked as a function of temperature. The results demonstrate the onset of antiferromagnetism in the materials and also indicate the presence of a magnetic phase transition below T=25K in the sample with x=0.0125. This result is consistent with that observed by Matsuda et al. in LSCO.[1] As discussed in Chapter 2, this would also be consistent with the rod-like scattering that is assumed to exist perpendicular to the (HK0) plane.

4.2 Experimental Results

4.2.1 Undoped La₂CuO₄

Undoped La_2CuO_4 is an excellent example of a Mott insulating material. While the pure material has been well studied, it serves as a benchmark with which we may



Figure 4.1a: Mesh scan at 3K for undoped La_2CuO_4 with cold Be filter.



Figure 4.1b: Mesh scan repeated without filter.

compare the rest of the data taken in such a study. Figure 4.1 shows the mesh scans taken for the pure crystal of La₂CuO₄ at base temperature with and without a Be filter placed in the incident beam. Each of these images represents the scattering around a small portion of reciprocal space surrounding the (1/2,1/2,0) magnetic Bragg position. These images demonstrate the high resolution with which these scans were performed. For this case, the collimation used was [Open, 0.2°, 0.2°, 1.2°]. Each of these values represents the angular confinement of the neutron beam after passing through a collimator. The beam is open (no collimation) before the monochromator, has a collimation of 0.2° in the beam between the monochromator and sample, 0.2° between the sample and analyser and 1.2° between the analyser and the detector. The scan showing the double peak is representative of the higher order wavelength contamination that results from $\lambda/2$ scattering from (1,1,0). From this figure, it is easy to see that this particular sample is twinned and that one orientation in the a-b plane is more heavily populated. It also demonstrates that this crystal is of generally high quality.

In an orthorhombic unit cell, the nuclear scattering in the (HK0) plane should produce four individual Bragg peaks. As the lattice parameters approach the tetragonal limit a = b, the features would appear merge into one. In this case, however, the small orthorhombicity in the lattice produces four peaks near each other which can be resolved in these scans. This crystal displays four peaks clearly with some mosaic spread between each pair.

The image shown in figure 4.1a) shows a mesh scan performed with a cold Be filter in the incident beam. The higher order intensity is removed and only the magnetic Bragg feature remains. The fact that this feature coincides with the structural feature at higher \vec{Q} indicates that the magnetic moments are parallel to b* within the CuO₂ plane.[20] Due to b > a, in reciprocal space a > b > b. Since the magnetic feature is



Figure 4.2a: Magnetic peak labeled with locations at which temperature dependence was measured.



Figure 4.2b: Magnetic scattering vs. temperature for all points measured in the above diagram.

located at the higher \vec{Q} position, the magnetic scattering can be associated with the a axis. Also, from Chapter 3, if the moment direction is parallel to the scattering vector, then no magnetic scattering will be detected. Since there is no scattering in the low \vec{Q} or b^{*} position, the magnetic moments must be parallel to the b axis.

Here, the magnetic scattering shows strongly as a mostly singular Bragg peak at the high \vec{Q} position. In this sample, T_N is expected to be approximately 325K.[5] This does not appear to be the case as is shown in figure 4.2. The temperature dependent scattering is shown along with a map detailing the positions of both points sampled on the peak. While only one position was measured from base temperature up to 280K, all temperature dependencies clearly show a pure Neel type transition that begins almost precisely at 250K. The various sample points near the region of highest intensity all drop off sharply to background levels at just over 250K. This transition is marked by the solid black line. This suppression of T_N is most likely due to the inclusion of excess oxygen from the crystal growth environment during synthesis. While the suppression of the Neel temperature shows that this, and thus other samples may all be contaminated by oxygen doping, the sharpness of the Neel transition shows that this doping may have a minimal effect on the nature of the magnetic structure. Studies of oxygen doped La_2CuO_4 by Birgeneau et al. have shown that the introduction of intercalant O^{-2} ions creates a rather complex phase diagram. [21] The suppression of $T_{\rm N}$ to 250K in this material would be consistent with an increased oxygen content of ~ 0.005 which can be determined using the phase diagram for the oxygen doped material in figure 2.2b). [5] While the transition temperature appears to be consistent for all sampled points, the regions farther from the peak intensity reach their maxima faster than the higher intensity region. This behaviour is quite similar to what was seen in the x=0.0125 sample which may suggest some disruption of long range AFM order by the excess oxygen although the effect is minimal. Unlike the x=0.0125 sample, however, this sample shows no sign of any further magnetic transition at low temperatures. Near base temperature, there is no down-turn in intensity. This is, of course, expected for the pure material.



Figure 4.3: Average of T dependent scattering for peaks measured from room temperature to 150K in La₂CuO₄.

Figure 4.3 shows the average trend in the temperature dependencies taken from room temperature to 150K. The more gradual increase in intensity shown by the most central point is dominated by the sharp Neel type transitions of the other points. While the average does not extend to base temperature, it does demonstrate that the transition for this material, although supressed, is sharp. From this figure, the transition temperature is confirmed to be 250K.

4.2.2 La_{1.994}Ba_{0.006}CuO₄

This sample, like the previous, is also shown to be of relatively high quality having one highly populated twinning domain, and another minimally populated domain. In the structural profile of the (1,1,0) scattering, it can be seen that for this crystal, the Bragg scattering is contained mainly in the low \vec{Q} region (see figure 4.4b). Unfortunately, this means that since the magnetic scattering is localized in the high \vec{Q} region, the majority of the sample is oriented such that the magnetic moments make no contribution to the magnetic scattering visible at this position.

In this case, the magnetic peak is confined to a small region in Q space due to the small mosaic in the system. This made the temperature dependence of this material easy to measure. Figure 4.5 shows the temperature dependence of the magnetic scattering for this lightly doped sample at several points representing the region of highest intensity as well as several points probing the regions of weaker scattering. Even at this small level of doping, the sharpness of the Neel transition is lost. In addition to this, there is another issue that is apparent from the temperature dependence of several points. In particular, the intensity measured at (0.5125, 0.4945, 0) begins to set in rapidly at 220K and suddenly drops off below 145K. This unusual behaviour is due to structural changes occuring in the crystal during cooling which appear in the magnetic scattering as a change in mosaicity. As this peak drops off, some of the other regions which began weakly, begin to increase as the peak moves from one region to another. While this occurs in the regions of weaker scattering, the intense regions of the AFM peak steadily climb to a maximum. In order to determine the transtion temperature for this sample, a region has been marked out using dashed lines on the figure. Since the only reliable temperature dependence can be obtained from the intense region which remains stationary throughout the scan, the transition



Figure 4.4a: Mesh scan at 3K with cold Be filter for LBCO x=0.006.



Figure 4.4b: Repeated scan without filter.

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must heavily depend on these two sets of data. The initial increase in intensity at the (0.5125, 0.4945, 0) position provides some evidence that, even in the weaker scattering regions, the long range order sets in at approximately the same temperature across the entire peak.

The change in the longitudinal line shape of the peak introduce a large error in the determination of T_N at this doping. Coupled with the complex trends seen in figure 4.5b), it is difficult to pin point an exact transition temperature in this material. The upper bound can be placed on the transition by noting that, at 200K, three of the major sampling points have begun to increase in intensity and diverge from one another. The lower point can be set by noticing that above 220K, the scattering intensity approaches background levels for all points. Using these two markers as a guide and weighting the decision heavily on the onset of magnetic scattering at the two prominant scattering positions, the Neel temperature for this material can be estimated at $T_N=218$ K.

 T_N has been marked on this diagram by a solid, vertical line. This temperature dependence was measured in several steps, stopping at various intervals in order to profile the magnetic peak. The first set of measurements were performed at intervals of 1K as the temperature was increased from base to 125K. From 125K to 280K the temperature was incremented in 3K steps which was more than sufficient T resolution to make the transition apparent.

4.2.3 La_{1.9875}Ba_{0.0125}CuO₄

Figure 4.6 shows two typical mesh scans for LBCO x=0.0125. On the top and bottom of the figure are images showing scans taken at base temperature, with and without a Be filter respectively. In the absence of the filter, the scan clearly shows two separate



Figure 4.5a: Mesh scan labeled with locations at which temperature dependent scattering was measured.



Figure 4.5b: Temperature dependence of magnetic scattering at all locations labeled in the above diagram.

peaks of relatively high intensity. The appearance of two features is representative of the orthorhombic twinning that occurs in this material in the MTO phase. This contamination is due to the higher order Bragg scattering from the (1,1,0) structural position.

Once the cold filter has been placed in the beam, the higher order contamination is eliminated as before. While some incommensurability is expected in this material at low dopings, this sample does not seem to display any signs of an incommensurate structure. The feature is, however, long and thin along the diagonal. While it is difficult to observe in this example, each elongated feature is produced by the mosiac spread of two Bragg peaks which overlap in the centre.

Figure 4.7 shows similar scans for three separate temperatures. From the three different temperatures in this figure, a clear evolution of both the structure and the magnetism can been seen. On the left hand side are the scans performed in the absence of the filter. These maps represent the structural scattering from the (1,1,0) peak caused by $\lambda/2$. From high to low temperature, the peak separation increases which indicates that the orthorhombicity of the crystal is increasing as the temperature decreases.

More significantly, an evolution of the magnetism can be seen in the scans which utilise the Be filter. The absence of any AFM structure is evident at 280K as would be expected in the paramagnetic phase above T_N . Upon cooling, the material enters the antiferromagnetic phase and the (1/2,1/2,0) magnetic Bragg feature appears. As expected, this phase persists down to base temperature but changes slightly in intensity. While it may be predicted that an AFM Bragg feature increases in intensity as the temperature is lowered to base temperature beyond T_N , this does not appear to be the case accross the entire range of temperatures. From these three mesh scans, there appears to be a slight loss in intensity as the sample is cooled to base from 80K.



Figure 4.6a: Mesh scan at 3K for LBCO x=0.0125 with a cold Be filter in the incident beam.



Figure 4.6b: Mesh scan at 3K with no filter.

Temperature dependencies of several different positions on the magnetic peak were taken all of which are displayed in figure 4.8. Figure 4.8 a) shows a base temperature map on which the locations of each scan have been labeled. These locations were chosen because they represent areas of interest and capture many different regions of the magnetic peak. By performing these more detailed scans, a great deal of information is gained regarding the evolution of the feature from regions of Bragg and diffuse scattering. All of these regions collectively provide the details necessary to compile a complete picture of the antiferromagnetism in the material. From this diagram, the onset of AFM order can be seen along with some unusual low temperature behaviour. A well defined Neel transition is not readily apparent from the temperature dependent behaviour displayed in the figure. While the scattering begins to develop at temperatures as high as 180K, many of the regions do not begin to show signs of magnetic ordering down to 160K. Due to the diversity of this behaviour, T_{N} must be determined from the onset of magnetic order in all regions measured on the peak. Using this as a guide, the region in which all points appear to intensify is marked with the vertical dashed lines. On the low T side, 125K, the least intense of the features, those at the edges of the peak rapidly increase and begin to level off. For the more intense regions near the centre of the peak, the scattering sets in at higher temperatures where the steepest increase begins to occur around 150K. Using these markers as upper and lower bounds for the transition temperature, a mid range value of 151K can be used as T_N . This value is indicated by the solid vertical line. For the regions of higher intensity, the trend continues rapidly upwards until the intensity approaches a maximum at roughly 35K. The more diffuse scattering at the edges of the peak level off more rapidly and maintains its intensity down to base temperature. For the central regions, however, a second transition occurs at 21K, which is also marked by a solid line, in which the peak intensity drops sharply and continues to decrease as the sample is cooled to 3K.

Figure 4.9 shows two T dependent trends which represent the average scattering for both the intense and diffuse regions of scattering on the peak. From this figure, a clearer picture of the magnetic evolution is visible, which allows for a refinement of T_N based on the average behaviour. Both regions begin to increase in intensity as the temperature is decreased, however, at $T_N=130$ K, the peripheral regions settle into a regime of relatively low intensity scattering, diverging from the high intensity regions which continue to climb as long range AFM order sets in. The separation of these two trends signals an excellent location to mark T_N as it occurs while the scattering is becoming well developed, but occurs in conjunction with the full onset of magnetic scattering on the periphery of the peak.

This figure also shows the prominence of the second magnetic transition at the central region of the peak. Averaging like regions displays the sharpness of the transition and demonstrates that the second transition temperature of 20K is an accurate evaluation of the transition temperature for this phenomenon. As mentioned in Chapter 2, this low temperature transition has been shown to occur in samples of LSCO with similar hole concentrations.[1] The transition has been interpreted as being the onset of the diagonal stripe phase for samples with dopings of x>0.2. While this sample is still well within the AFM dome for this material, the hole doping clearly influences the 3D ordering. This transition may be consistent with loss of AFM order along the c axis, resulting in a 2D AFM structure localized in the CuO₂ plane. This type of transition would produce rod-like scattering along (00L) and this redistribution of scattering intensity would account for the loss of intensity at the magnetic position. While this result does not directly demonstrate the existence of 2D AFM order, it does suggest it and at least confirms the result seen by Matsuda et al in LSCO.

4.2.4 La_{1.975}Ba_{0.025}CuO₄

Unlike the other samples in this experiment the temperature dependent magnetic scattering for this sample was difficult to measure. This is due to a lack of intensity in the observed magnetic scattering. Figure 4.10 shows the mesh scans taken a base temperature with the filtered and unfiltered beams. The structural contamination for this sample is unusual compared to the other samples in this study. This sample appears to show three peaks, two at high \vec{Q} and one at low \vec{Q} which is different from the structure observed for all other samples. When the Be filter is placed in the incident beam, little magnetic structure is observed at all. In order to compensate for the extremely low intensity at this high resolution, these mesh scans were collected with the collimation reduced to [open, 0.6°, 0.799°, 1.2°]. Here the magnetic intensity appears at three points in reciprocal space which correspond to the most intense regions in the structural scattering. This may indicate that regardless of the Be filter, some structural contamination may still be passing through. There is some indication, however, that there does exist some magnetic signal in this material. Surrounding the main high intensity features is a diffuse, but noticeably elevated 'background'.

This diffuse scattering is more readily visible when compared with another mesh scan at a higher temperature as in figure 4.11. In this figure, the 3K data is plotted along side data collected with the same collimation at 50K. This comparison shows clearly the existence of diffuse, magnetic scattering at base temperature which does not exist at higher temperatures. In these figures, it can also be seen that the peak intensities in both plots follow the same pattern, although the diffuse scattering is gone. This may further indicate that the peak intensities are due to the $\lambda/2$ contamination from the nuclear Bragg scattering. The fact that the majority of this magnetic scattering intensity is localized on the low \vec{Q} region is also somewhat unusual. This could indicate that the Cu^{2+} magnetic moments no longer lie along the b axis but also along the shorter a-axis at this doping.

Figure 4.12a shows the same mesh scan which has been binned in square blocks of four pixels (2x2 binning). The binning eliminates the peaks as prominant features. In particular, the magnetic peaks at higher \vec{Q} blend into the background showing the diffuse nature of the magnetic scattering. In this image, the scattering can be seen more readily as a general increase of the background levels surrounding the peak. Figure 4.12b shows diagonal cuts through the magnetic peaks as labeled on the mesh scan in figure 4.11a. This figure shows the double peak along the diagonal (H, 1-H, 0) direction in the region of the magnetic peak. From this figure, it can be seen that the magnetic scattering has begun to display incommensuration as is expected for samples at this doping. The data is fit to a two gaussian form which is an excellent fit to the peak with an $R^2=0.94$. This incommensurate structure is in stark contrast to the lack of scattering seen in the diagonal scan at 50K. This scan shows a weak, single sharp peak in the centre of the scan. It is clear from this comparison that the magnetic scattering does not set in until well below 50K and that a significant magnetic structure develops at low temperature. The incommensuration can be measured using the distance from peak centre to peak centre which is $\delta = 0.021 \pm 0.008$. Results from previous time-of-flight measurements on this material show that the incommensuration is $\delta = 0.017 \pm 0.001$.[10] Although these results appear to be consistent, the large value of error on the value obtained from the Chalk River data makes this estimate of the incommensuration somewhat unreliable. This value is, however more consistent with the results seen in LSCO by Matsuda et al where $\delta \approx x$. [22]

Naturally, the temperature dependence of this diffuse scattering was also measured at a single point which represents a region which is uncontaminated by higher order scattering. This temperature dependence can be seen in figure 4.13a. While this temperature scan does not cover the entire range of temperatures, there is a clear transition which occurs at about 25K. Above 25K, a seemingly constant count rate is observed. Below 25K, the diffuse scattering sharply increases. In this sample, the usual 3D AFM state does not occur and only a 2D AFM structure is observed. The onset of this two dimensionally ordered state at low temperature is also consistent with the incommensurate spin structure observed in this and other samples with similar doping.[12][10] Similar triple axis work has been done on LBCO with x=0.035 at HFIR at Oak Ridge National Laboratory. The temperature depedence of the magnetic scattering for this material is shown in figure 4.13b. Although the transition temperature is lower for this sample, it still displays a similar trend in which the low temperature magnetic scattering sets in amongst a diffuse background.

Studies of this particular sample have been undertaken at both NIST and Chalk River which report the appearance of AFM Bragg scattering at low temperatures which has been assumed to be rod-like along L. [12] Incommensurate rod-like scattering is consistent with data collected on DCS at NIST. It is clear that a full temperature dependence of the magnetic structure in this material would benefit this study and may be necessary in the future.

4.2.5 La_{1.9875}Ce_{0.025}CuO₄

While it is not a part of the same class of materials, one Ce doped sample was included in this study to ascertain the quality and explore the magnetic structure of this newly synthesized material. It is expected that a material with such a low electron concentration is to be well within the AFM dome on the electron doped phase diagram (see figure 2.2b). Since this material has never before been studied in single crystal form with neutrons, this study was an excellent opportunity to collect data concerning its magnetic structure.

This material behaves in a similar manner to the hole doped materials. The mesh scans in figure 4.14 show higher order contamination of the magnetic Bragg position by a twinned orthorhombic structural peak. When the contamination is eliminated, the magnetic scattering is localized in the high \vec{Q} region of the structural feature as before. This sample also appears to have a small mosaic spread and gives a clean antiferromagnetic signal.

Figure 4.15 shows the temperature dependence taken at several points on the peak as indicated by the 3K mesh scan. As with the Ba x=0.006 sample, the temperature scans of this material show a wide spread of T dependent scattering. While none of the peaks shift during cooling, a number of the different peak positions display the onset of magnetic ordering at slightly different temperatures. This is captured by the placement of the dashed vertical lines on the plot. From these initial boundaries, the AFM order can be said to appear by 230K and increase down to base. Although the transition temperature is near that of the undoped sample, the forms of the temperature dependence for these materials varies a great deal. The loss of the sharpness at T_N which was displayed by the pure sample shows conclusively that the Ce is indeed being incorporated into the lattice.

This material does not seem to show any sign of the onset of 2D AFM order at low temperature. Both the high T_N and the persistance of 3D AFM ordering are signs that this material is well within the wide antiferromagnetic dome that is typical for the electron doped cuprates. While this does not necessarily provide conclusive evidence of the antiferromagnetic structure in this material, it serves as a preliminary study with which to orient our future endeavours. Further studies are required in order to understand the behaviour of this material.



Figure 4.7: Typical mesh scans for LBCO x=0.0125 as the sample is cooled down to base temperature. The onset of antiferromagnetic order can be seen, as well as a decrease in peak intensity from 80K to base temperature.



Figure 4.8a: Mesh scan for LBCO x=0.0125 at 3K with locations of temperature scans labeled on the peak.



Figure 4.8b: Plot of scattering intensity vs. temperature for all points shown in the above diagram.



Figure 4.9: Average scattering intensity vs. temperature for regions with relatively high intensity, displaying down turn near base temperature. Diffuse magnetic scattering on the periphery (low intensity regions) also sets in and remains constant down to 3K, showing only a very small decrease in intensity at base T.



Figure 4.10a: Mesh scan of LBCO x=0.025 showing three structural peaks and the diffuse scattering which occurs around these features.



Figure 4.10b: Mesh scan at 3K without filter.



Figure 4.11a: Diffuse scattering around the magnetic Bragg position in LBCO x=0.025.



Figure 4.11b: Mesh scan repeated at 50K. Note the decrease in both diffuse scattering and peak intensity.



Figure 4.12a: Diffuse scattering around magnetic Bragg position with 2x2 binning.



Figure 4.12b: Cuts along diagonal between peaks as shown in figure 4.11a.



Figure 4.13a: Temperature dependence of diffuse scattering taken from base to 50K in LBCO x=0.025.



Figure 4.13b: Temperature dependence of magnetic scattering in LBCO x=0.035 taken from base to 35K at HFIR.



Figure 4.14a: Mesh scan with filter for Ce doped crystal at 3K.



Figure 4.14b: Repeated scan without filter.



Figure 4.15a: Mesh scan of LCCO x=0.025 at 3K, indicating locations at which temperature dependence was measured.



Figure 4.15b: Temperature dependent scattering in LCCO x = 0.025.



Figure 4.16: Comparison of I vs. T trends for all hole doped materials in this study with the addition of LBCO x=0.035 measured at HFIR. The development of dopant dependent magnetic structure can been seen in this figure.

4.3 Conclusions

This high resolution neutron scattering experiment has confirmed and uncovered a great deal of interesting behaviour in extremely lightly doped LBCO. Collectively, the temperature dependence of the magnetic scattering for these materials create a new way of viewing the magnetic phases which exist in this material as a function of doping. Transition temperatures that have been determined in the previous sections can be compiled to show the evolution of magnetic structure in this material.

Figure 4.16 shows the progression of T_N shown by the data collected at Chalk River. It is clear that T_N follows the expected pattern for the AFM dome in the lightly doped materials, apart from the potential influence of excess oxygen which has lowered T_N for all the materials in this study. As x increases, T_N begins to drop off, and for x=0.0125, a second magnetic transition is observed at 20K. While this phase diagram does not explicitly demonstrate the existence of the 2D AFM phase, it does indeed show some form of a reentrant transition that is consistent with the expected 'spin-glass' phase of hole doped La-214 as reported by Matsuda et al.[1] As the doping is increased into the spin-glass regime, the 3D antiferromagnetic transition is lost and the 2D AFM order remains. This 2D transition occurs at low temperature in both the x=0.025 and the x=0.035 samples in which it sets on quickly.

Using this data and the transition temperatures determined from the previous analysis, a phase diagram of the antiferromagnetic order can be constructed for hole doped La₂CuO₄ as is shown in figure 4.17. Coupled with data found in the superconducting regime[12], an antiferromagnetic phase diagram emerges which follows the usual phases of this material. In the pure and lightly doped materials, the usual 3D antiferromagnetism can be seen, as was demonstrated above. The transition temperatures for these materials follow the expected trend of the antiferromagnetic dome for doped La-214. For x=0.0125, a 2D AFM transition occurs at 20K (which is confirmed by the results in Chapter 5). This shows the beginnings of some spin-glass like behaviour near the edge of the AFM dome at x=0.02. As the doping increases further, there is a noticable lack of 3D antiferromagnetism, but the onset of incommensurate 2D AFM ordering can be seen at low temperature. This 2D antiferromagnetism continues in through the 'spin-glass' regime up to the edge of the superconducting dome at x=0.05. From here up to higher concentrations, the incommensurate structure continues to develop as reported by S. Dunsiger et al.[10] The correlation of the mag-



Figure 4.17: Diagram of antiferromagnetic phases in LBCO. T_N in the SC dome have been taken from Y. Zhao et al.[7]

netic behaviour to the superconducting and other phases in this material may provide more clues about the mechanism of superconductivity, and provide a complete picture of the complex behaviour displayed by this material.

Chapter 5

Time of Flight Neutron Scattering Study of Lightly Doped $La_{2-x}Ba_xCuO_4$

5.1 Introduction

Time-of-flight neutron scattering experiments have been performed using the Disk Chopper Spectrometer (DCS) at NIST. The purpose of this experiment was to directly observe the 2D AFM order which has been discussed in previous sections. In order to bring the 'rod' of scattering into the scattering plane for this experiment, several samples of LBCO were aligned in the (HHL) scattering plane. The realignment was done on the Chalk River E3 alignment spectrometer.

The sample environment used for this study was an ILL orange cryostat which cooled the crystals using liquid He, allowing the samples to be cooled to a base temperature of 1.5K. For the x=0.025 and x=0.035 samples, an aluminum adapter base plate was required in order to make the sample mounts compatible with the ILL Orange Cryostats used at NIST. As an instrument operating with a cold neutron source, an $E_i = 0.79$ THz ($\lambda = 5$ Å) was chosen. This incident energy had the advantage of eliminating aluminum powder lines from the regions of interest in reciprocal space for these materials as well as providing high energy and \vec{Q} resolution. In this configuration, detailed maps of the (HHL) scattering plane were made at various temperatures in an effort to discern the 2D antiferromagnetism which is indicated in the previous analysis.

5.2 $La_{1.9875}Ba_{0.0125}CuO_4$

In the previous section, it was shown that the sample with the most complex antiferromagnetic behaviour was the LBCO x=0.0125 crystal. Naturally, this material was the most thoroughly examined material in this study. Figure 5.1 shows two maps of reciprocal space made on DCS at 1.5K (base temperature) and 35K. These maps were produced by rotating the single crystal sample in 1° steps spanning a 90° rotation with a collection time of 5min/point. From this diagram, the behaviour observed in the previous section is immediately confirmed. It is clear that the 2D antiferromagnetic phase is present at low temperature and dies off at some T<35K.

The rod of scattering occurs at (H,H,L) for H=0.5. Of course, coexisting with the rod-like scattering are the antiferromagnetic peaks which persist at this doping. While there are peaks at all integer values of L along the rod, only the peaks at odd integers represent purely magnetic scattering. For even L, the peaks are much more intense and persist at all temperatures measured in this study, up to the maximum 300K. This would appear to indicated that these features are a result of nuclear, rather than magnetic structure. Further investigation is required in order to determine the nature of these unusual features.



Figure 5.1: Reciprocal space maps for LBCO x=0.0125 at 1.5K and 35K. The 2D antiferromagnetism is clearly visible at base temperature and vanishes at higher T.

Another unusual feature that appears in each of these maps is the shadow which cuts through a wide region near L=0. The cause of this shadow is unknown but is likely due to sheilding by the crystal. With the long cylindrical shape of these samples and the wide angular range covered by these scans, it is possible that the crystal itself may have attenuated the scattering simply by changing its orientation in the beam. Regardless of the source, it was clearly a requirement that this region be avoided during analysis so as to avoid inconsistencies in the data.

While the presence of the 2D AFM feature is clear at base temperature, it is also clear that it is absent at 35K. The analysis of the Chalk River data lead to the conclusion that a transition into the 2D AFM phase occurred at a temperature of 21K. From this preliminary example, it appears that this transition temperature is roughly correct. From this figure, it is difficult to determine if the rod of scattering has been completely eliminated by 35K as there appears to be some high background around the magnetic peak position. In order to determine the nature of this transition, more detailed temperature scans were compiled.

In order to avoid the shadowy intrusion discussed above, a more restricted set of angular scans was performed spanning 27° with step sizes of 0.5-1°. These scans captured all of the regions of reciprocal space left untouched by the shadow region. These scans were performed at 10K, 15K, 20K, 22.5K, 25K, 30K and 35K in order to observe the evolution of the magnetic structure. It is clear from this figure that the rod gradually decreases in intensity until 20K at which point the rod-like scattering drops sharply. After this, the gradual decrease in intensity continues until the rod has virtually been eliminated by 35K, and only the Bragg scattering (magnetic and otherwise) remains.

Figure 5.3 shows cuts made along the length of the rod of scattering (i.e. along the L direction) at three temperatures. At all three temperature, it is clear that



LBCO x=0.0125, 1.5K, 5A Elastic LBCO x=0.0125, 10K, 5A Elastic LBCO x=0.0125, 15K, 5A Elastic LBCO x=0.0125, 20K, 5A Elastic 20 0 [....] 20 0 1111111111111111 20 0 [.....]

Figure 5.2: Small reciprocal space maps for LBCO x=0.0125 at various temperatures. These scans show a more detailed picture of the evolution of the rod of scattering with temperature.

the magnetic Bragg features are prominent and show little temperature dependence. The difference is the intensity along the rod. At base temperature, the scattering intensity is high relative to that at 22.5K. Between these two temperatures there is a significant decrease in intensity across the entire length of the rod. There is a similar but much smaller decrease in intensity between 22.5K and 35K. This demonstrates that the entire rod of scattering changes consistently with temperature and does not vary in intensity at different values of L. It also shows that the majority of the tran-


Figure 5.3: Cuts along the rod of scattering at various temperatures in LBCO x=0.0125.

sition occurs below 22.5K. From this perspective, however, it is difficult to pinpoint an exact temperature at which a transition between 3D and 2D antiferromagnetism occurs. One important fact is that the onset of the rod-like scattering is consistent for all values of L and does not develop outward from the Bragg features. Although the scattering is weak away from the Bragg peaks, these scans collected enough information to create cuts through the rod at various temperatures. In order to do this, data sets at each temperature were compiled and binned along the L direction using DCS Mslice, an analysis package included in DAVE. The satellite peaks near the main Bragg features posed a problem and restricted the area which could be binned over to a small region of $\Delta L=0.3\text{\AA}^{-1}$ between each set of Bragg peaks. Once the regions between Bragg peaks were binned together along L, the data sets were added together, resulting in a reliable, average cut through the rod of scattering visible in each map.



Figure 5.4a: Colour contour map showing the evolution of the cross-section of rod-like scattering as a function of temperature in LBCO x=0.0125.



Figure 5.4b: Temperature dependence of rod intensity from peak values on colour contour map above.

The result of this process is the colour contour map shown in figure 5.4. While the previous figure gave some idea as to the shape of the rod along L with respect to temperature, the colour contour map is able to give a more accurate picture of the temperature dependence of the cross-section of the feature. It is interesting to note that the cuts perpendicular to the rod do not appear to change in shape as the scattering diminishes. Rather, the entire rod simply decreases in intensity until 25K, above which it appears to fade to background levels. This behaviour is readily seen in the I vs. T plot corresponding to the data in the colour contour map.

In the Chalk River data presented in Chapter 4, the decrease in 2D scattering would correspond to an increase in intensity of the antiferromagnetic Bragg peaks. While the reciprocal space maps shown above are ideal for observing the weak rod of scattering, the Bragg features are too intense to show any temperature dependence on this scale. Following the evolution of the AFM Bragg peaks required a technique that could distinguish the Bragg scattering from the rod which is superimposed upon it. In order to do this, a cut had to be made parallel to the length of the rod which passed through the Bragg feature. Then, using the PAN (Peak ANalysis) package in DAVE, the features at (0.5,0.5,-1) and (0.5,0.5,-3) were fitted to a gaussian form. With this method, at each temperature, however intense the rod-like scattering, it would always be included in the fit as background surrounding the Bragg peak. The background could then be subtracted from the fit and the peak intensity of the fit would represent the pure Bragg intensity of the AFM feature.

The result of such fitting can be seen in figure 5.5a) and 5.5b) which show a fit for a typical Bragg feature and the temperature dependence of the maximum intensity for each Bragg peak. Figure 5.5b) does not provide nearly the same resolution on the temperature axis as the triple-axis study, however a similar trend can be readily observed. For (0.5, 0.5, -3), a high temperature, the magnetic scattering is weak and



Figure 5.5a: Example cut parallel to rod showing magnetic Bragg peak in LBCO x=0.0125.



Figure 5.5b: Temperature dependence of magnetic Bragg scattering for two peaks in LBCO x=0.0125.

begins at background levels for T=300. At T=35K, the intensity approaches its peak intensity and, at 30K reaches its maximum value. Below 30K, the intensity follows a downward trend similar to that measure at Chalk River. In this case however, the transition temperature appears to be higher by roughly 10K. For the Bragg feature of less intensity, (0.5,0.5,-1), the maximum value appears to dip only slightly at base temperature. Both of these trends are consistent with what has been seen in this material in the previous study.

5.3 $La_{1.965}Ba_{0.035}CuO_4$

The transition observed in the previous material has been confirmed using data from Chalk River and DCS. The x=0.035 material, however, has not been studied in a similar way, and thus serves as an excellent opportunity to obtain information regarding the rod-like scattering in the 'spin-glass' regime. Scans similar to those run on the x=0.0125 material were performed in order to gain a picture of the 2D AFM order in this material.

Figure 5.6 shows a comparison of reciprocal space maps at base temperature (1.5K) and 35K. From this figure, a relatively faint rod of scattering can be seen at (1/2,1/2,L). In this case, the rod appears to be much weaker and also broader then in the previous sample. The broadening of the rod of scattering is consistent with the incommensurate picture of the antiferromagnetism in this material. From the antiferromagnetic to spin-glass phases the antiferromagnetism moves from being commensurate to incommensurate. As the doping is increased, the incommensuration increases as well and is visible in the profile of the AFM Bragg feature. In this material, however, there exists little 3D antiferromagnetism if any at all. This is indicated by the absence of the (0.5, 0.5, -1) and (0.5, 0.5, -3) peaks which were very



Figure 5.6: At base temperature in LBCO x=0.035, a weak rod-like feature is observed which is completely absent in the 35K scan.





prominent in the previous sample.

Figure 5.7 shows several maps around the magnetic position at various temperatures for this sample. It can clearly be seen that the scattering declines rapidly and completely vanishes by 15K. This is quite consistent with the analysis from the previous section where the transition temperature for this material was determined to be roughly 17K. As the temperature continues to increase, some diffuse features appear in the region of the rod but are not consistent with the form observed at low temperature.

Figure 5.8 shows three scans along the rod of scattering at 1.5K, 15K and 35K.



Figure 5.8: Cuts along rod at various temperatures.

It is quite evident from these cuts that the magnetic Bragg peaks which appeared in the x=0.0125 sample are no longer present except for the peak at (1/2,1/2,2). Again, as the temperature is decreased, the intensity of the rod decreases consistently along its length. In this case, however, the intensity appears to reach a minimum value by 15K and remains roughly constant up to 35K.

Figure 5.9, constructed in a similar way as for the x=0.0125 sample, provides a more conclusive look at the temperature dependent behaviour in this material. Binning the scattering along the rod creates a prominent feature at both 1.5K and 10K. This feature then obviously drops off to background levels and is maintained up to 35K. The corresponding plot of the temperature dependence of the rod intensity shows more complex behaviour. After the rod-like scattering appears to vanish, some other diffuse intensity seems to reappear at higher temperatures. This is not easily seen in from the colour map itself. Although the maps show the appearance of a weak feature on the (0.5,0.5,1) position, these data were binned such that no scattering in the vicinity of the Bragg positions would be included. This must indicate that, for these higher temperatures, there is a small increase in the overall intensity of the diffuse scattering along the rod. Further investigation into this phenomenon is required in order to confirm this observation.



Figure 5.9a: Colour map showing the evolution of the cross-section of rod-like scattering as a function of temperature in LBCO x=0.035.



Figure 5.9b: Temperature dependence of rod-like scattering in LBCO x=0.035.

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5.4 Conclusion

While providing complementary information to the results obtained in the tripleaxis experiment, this study has shown a great deal of interesting behaviour in hole doped La-214. Although the scattering in LBCO x>0.02 has always been assumed to be rod-like, it has not, until now, been directly observed. This new observation of 2D antiferromagnetic order, as well as the confirmation of the temperature-dopant dependent AFM phenomena in this material both provide new ways of viewing the magnetism that is inherent in LBCO.

Chapter 6

Conclusion

The neutron scattering studies described in chapters 4 and 5 have demonstrated a rather complex and interesting set of doping dependent magnetic phenomena in lightly doped $La_{2-x}Ba_xCuO_4$. The evolution of the magnetic scattering from pure 3D antiferromagnet to 2D antiferromagnet has been observed as a function of doping. Using magnetic transition temperatures alone, the form of the general phase diagram for LBCO has been reproduced. This result clearly demonstrates the interconnection between magnetism, structure and superconductivity in this family of cuprates. Also, 3D antiferromagnetism has been observed in the electron doped material with the same orthorhombic phase as the hole doped material which is an unusual result.

Direct measurements of rod-like scattering at low temperatures near the 'spinglass' phase demonstrate the existence of 2D antiferromagnetic scattering. In the sample with x=0.0125, this rod of scattering is observed to coexist with the expected 3D antiferromagnetism. The transition temperature of the 2D AFM phase determined in the triple axis experiment is confirmed in this study. The sample with x=0.035 displays rod-like scattering in the absence of 3D antiferromagnetism, but with a significantly lower transition temperature. These experiments have elucidated some very interesting phenomena which demonstrate the need to continue studies of the cuprates. Ideally, these results will contribute to the general understanding of superconductivity in this family of the cuprates and will motivate future work in this field.

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