

Local magnetic order in La_2CuO_4 seen via μ^+ SR spectroscopy

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Abstract.

The origin of a pseudogap in the underdoped phase of cuprates has become one of the leading issues in understanding the mechanism(s) of high- T_c superconductivity. Several experiments (i.e. polarized neutron diffraction studies) support theoretical models based on the circulating current (CC) picture. These CC models suggest a novel ordered phase formed by orbital magnetic moments caused by spontaneous orbital currents. However to date, μ SR experiments in cuprate superconductors have not revealed any magnetic field higher than 0.2 G associated with orbital moments. Here we present high magnetic field μ SR spectroscopy of a stoichiometric La_2CuO_4 crystal which shows antiferromagnetic splittings and additional 45 G line splittings, consistent with orbital currents.

1. Introduction

The origin of the pseudogap (PG) and its relation to the nature of high- T_c superconductivity has been highly debated for over 20 years. In the underdoped regime of the phase diagram, cuprate superconductors show features of a correlated metal exhibiting a remarkable departure from the standard Fermi liquid behavior. Their transport, magnetic and thermodynamic properties suggest strong reduction of the electronic density of states (DOS) below a temperature T^* , though the DOS does not reach zero at the lowest temperature and the system remains metallic [1]. However, ARPES experiments [2] suggest opening of a q -dependent real gap in a one-particle excitation spectrum, supported by other spectroscopies [3].

Certain theoretical models consider the PG state as an ordered phase with a well defined order parameter and a related broken symmetry [4, 5, 6]. In this scenario, the PG temperature T^* is a phase transition temperature to an ordered state formed by orbital magnetic moments caused by spontaneous orbital currents flowing in specific patterns within or around CuO_2 plaquettes. The fluctuations associated with the broken symmetry are considered to be responsible for both the superconductivity, playing role of a pairing glue, and the non-Fermi-liquid behavior below the T^* phase transition line on the phase diagram.

The key experimental evidence of a novel ordered state comes from spin-polarized neutron scattering experiments which reported commensurate magnetic peaks below T^* in YBCO and H1201 [7, 8], consistent with ARPES data, which point to time-reversal symmetry breaking (TRSB) that nevertheless preserves lattice translation invariance. Sizeable magnetic moments on the order of 0.1-0.2 μ_B are reported in the PG state [7, 8].

However, μ SR experiments which have convincingly demonstrated their sensitivity to TRSB fields in a number of weak magnetic systems, produced no evidence of a magnetic order in the PG state. At first, spontaneous static magnetic fields were reported in YBCO, which were later reinterpreted as being due to spatial charge inhomogeneities [9]. No TRSB is reported in LSCO where μ SR experiments set an upper limit of ~ 0.2 G for magnetic field at the muon site while the expected local field is estimated to amount about 40 G [10].

This discrepancy between neutron and muon experiments has been attributed to screening of the charge density in the metallic-plane unit cells in the vicinity of the muon [11]. On the other hand, orbital currents and associated magnetic moments may be present in the limiting case of the underdoped regime — in the insulating stoichiometric La_2CuO_4 , should CC be an intimate feature of chemical bonding in the CuO_2 plane.

Here we present μ SR spectroscopy of stoichiometric La_2CuO_4 crystal (accompanied by SQUID measurements), in which the magnetic field at the muon should not be affected by charge density screening.

2. Results and discussion

High-quality $\text{La}_2\text{CuO}_{4+x}$ single crystal grown from CuO flux have been used for current studies. Oxygen over its stoichiometric content has been removed by annealing in vacuum. The lattice parameters of the crystal correspond to the low temperature orthorhombic stoichiometric La_2CuO_4 [12]. Néel temperature measured using SQUID amounts to 320 K.

Time-differential μ SR experiments, using 100% spin-polarized positive muons implanted into these samples, were carried out on the M15 surface muon channel using the *HiTime* spectrometer. At low temperature, zero field μ SR spectra consist of two (small and large amplitude) components, well known from numerous previous studies and indicative of two inequivalent muon sites in antiferromagnetic La_2CuO_4 . Figure 1 shows temperature dependences of the corresponding frequencies. Above 150 K, the low-frequency component (signal with a small amplitude) disappears below the background. The Néel temperature $T_N \approx 320$ K and magnetic fields at the muon sites $B = 428.7$ G and $B = 111.8$ G determined from the temperature dependences shown in Figure 1 are consistent with numerous earlier measurements of stoichiometric La_2CuO_4 .

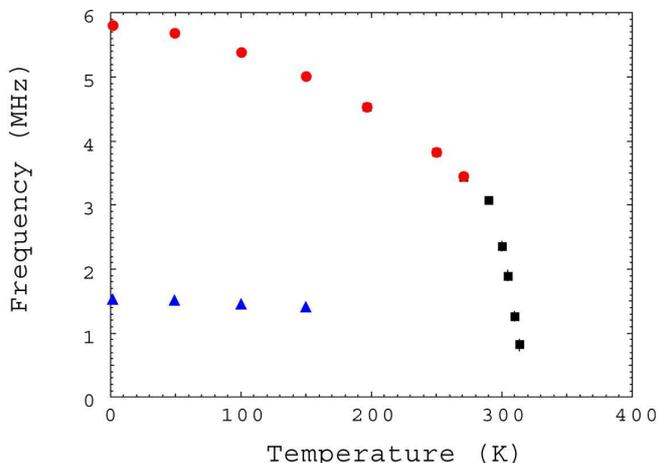


Figure 1. Temperature dependences of the muon spin precession frequencies in stoichiometric La_2CuO_4 in zero applied magnetic field. Red circles and blue triangles: frequencies obtained via a 2-component fit of μ SR spectra at low temperature. Black squares: frequencies obtained via a 1-component fit of μ SR spectra at higher temperature.

In high magnetic field B transverse to the initial muon spin polarization and parallel to the c -axis of La_2CuO_4 crystal, muon spin precession spectra exhibit at least 7-component pattern. The characteristic Fourier transform is shown in Figure 2. Central line at about 135.6 MHz coincides with the single line detected in CaCO_3 , a non-magnetic reference sample. We attribute this line to those muons (small portion of the muon flux) which missed the sample and stopped in a non-magnetic environment. Two small peaks positioned symmetrically around the central line correspond to the AFM splitting of the low-frequency (low-amplitude) signal observed in zero magnetic field. Our main interest is focused on large-amplitude signals again positioned symmetrically around the central line. We attribute these signals to high-frequency (large-amplitude) signal in Figure 1.

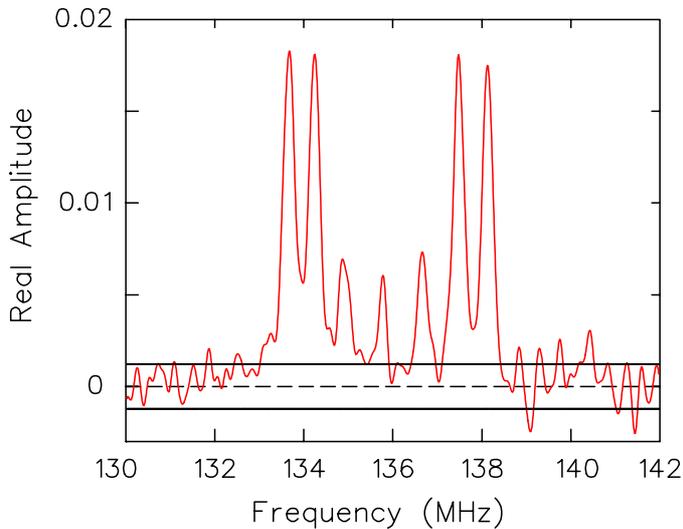


Figure 2. Frequency spectrum of muon spin precession in La_2CuO_4 in a transverse magnetic field of $H = 1$ T at $T = 100$ K. Central line frequency coincides with that of a single peak detected in the reference non-magnetic sample (CaCO_3).

In order to uncover the origin(s) of the magnetic field at the muon site, which may come from different sources, we must exactly determine the muon positions in the La_2CuO_4 lattice.

Stoichiometric La_2CuO_4 is a collinear antiferromagnetic material with four sublattices. Magnetic moment on Cu atoms is about $0.5 \mu_B$ directed along the diagonal of the CuO_2 plaquette [13]. A peculiar feature of magnetic ordering in La_2CuO_4 is the presence of a weak ($\sim 0.002 \mu_B$) ferromagnetic coupling of spins within CuO_2 layers [13, 14] which is believed to originate from Dzyaloshinskii-Moriya exchange interaction. These moments are orthogonal to CuO_2 layers and have opposite directions in the neighbouring planes. They are coupled to in-plane magnetic moments on Cu via tilting of CuO_6 octahedra. Therefore, CuO_2 is an antiferromagnet with hidden, weak ferromagnetism. A spin-flop transition to this hidden phase (weak ferromagnetism) takes place in high magnetic field applied along the c -axis [13, 14]. Within this phase, the direction of ferromagnetic moments in every CuO_2 plane becomes the same. Our SQUID measurements (Figure 3) at 250 K show that the spin-flop transition is observed at magnetic field of about 3.5-4 T. The magnetization jump $\sim 0.002 \mu_B$ is consistent with literature data [13, 14].

The spin-flop transition discussed above is clearly seen in μSR measurements at 250 K (Figure 4), where half of the lines in the spectra vanish at a magnetic field of about 4 T. A remarkable consequence of the spin-flop transition is that the magnetic unit cell becomes equivalent to the crystallographic unit cell; the number of magnetic sublattices decrease from 4 to 2. From symmetry considerations this means that tilting of the CuO_6 octahedra forces the muon to occupy only half of the otherwise crystallographically equivalent positions. However, in order to go beyond general considerations, possible muon positions should be clearly established.

The muon stopping site(s) in La_2CuO_4 have been discussed in several papers [15]. Information obtained from zero-field μSR spectra is insufficient and additional data are typically sought from

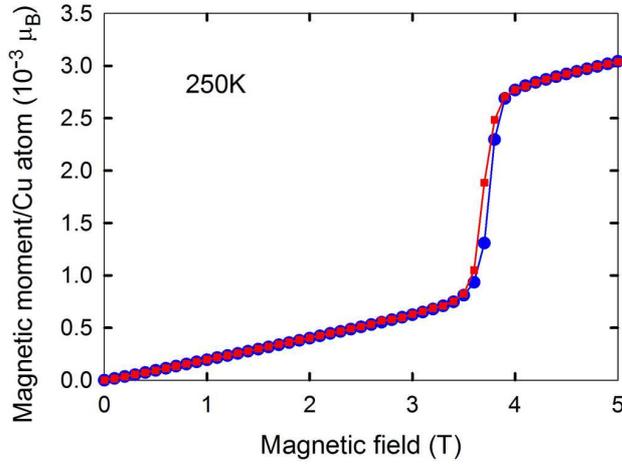


Figure 3. Magnetic field dependence of magnetic moment per Cu atom along the c -axis. Blue color: measurements in increasing magnetic field; red color: measurements in decreasing magnetic field.

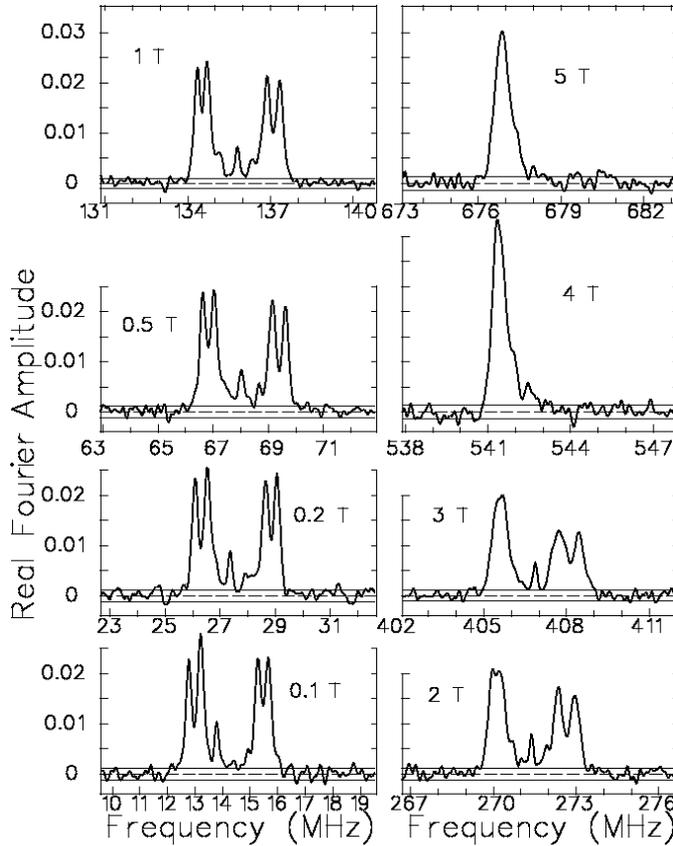


Figure 4. Fourier transforms of the muon spin precession signal in La_2CuO_4 at $T = 250$ K in different transverse external magnetic fields.

electronic structure calculations. It is common to predict the muon position to be in the vicinity of the apical oxygen atom but the exact positions are quite different in a number of publications leading to much confusion. In contrast, high-field μSR measurements with different setups provide data on projections of the magnetic field on the muon which correspond perfectly to the frequencies determined from the zero-field experiments.

To find muon positions based on this extended set of experimental data, we performed calculations based on the dipole-field approximation [16] assuming the periodic antiferromagnetic structure with the known magnetic moments on Cu atoms. There are only two types of inequivalent muon positions for the high-frequency high-amplitude signal in Figure 1: one is

close to the apical oxygen and the other is very close to La atom. From chemical bonding considerations the first type is far more stable. Similar analysis is possible for the low-frequency signal and only one of the two positions is chemically substantiated. Figure 5 shows possible muon positions for both low- and high-frequency signals. The tilting of the CuO_6 octahedra produces two types of chemically inequivalent positions differing by the distance between the muon and the apical oxygen atom. Only one type of these positions is occupied by muons. Our calculations confirm that it is consistent with vanishing half of the spectral lines at the spin-flop transition (accompanied by 10% decrease of the line shift from the free muon signal).

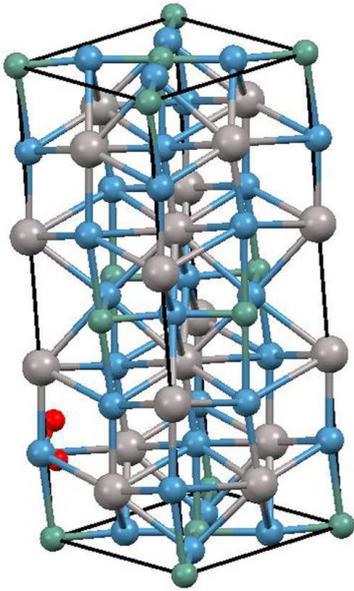


Figure 5. Unit cell for the low temperature orthorhombic phase of La_2CuO_4 : green - Cu atoms, grey - La atoms, blue - O atoms, red - muon positions (high- and low-frequency signals come from the muon residing near Cu and La, respectively); atomic positions are shown for pristine La_2CuO_4 , the perturbation caused by the muon is not taken into account.

One should expect 4 distinct signals in the high-field μSR spectra from two independent muon positions known from zero-field μSR experiments and the antiferromagnetic nature of the sample. Instead, at least 6 peaks have been observed in high magnetic fields of different directions. That means that apart from the AFM field, there is *an additional source of magnetic field on the muon* where some muons experience magnetic field larger than that coming from AFM while the other feel smaller magnetic field which causes the characteristic splitting of the large-amplitude signals (see Figure 4). The apparent absence of splittings in low-frequency signals can be explained by the increased distance from the CuO_2 planes leading to much smaller splittings which are not well resolved in the experiments.

There are two types of models explaining intra-unit-cell magnetic order in cuprates which may reveal the nature of the additional source of magnetic field on the muon. One relies upon spins being ascribed to oxygen atoms while the other is based on circulating orbital currents. Polarized neutron experiments have not been able to differentiate the existing models beyond establishing TRSB [7, 8]. It seems to be possible using high-field μSR experiments. While the strength of additional magnetic fields on the muon (which causes the observed splitting in the first place) is not known *a priori*, the ratio of signal splittings for different directions of the external magnetic field can be used to rule out certain models; although, a lot can be inferred from simple symmetry considerations.

The first class of models is based on the antiferromagnetic ordering of spins on in-plane oxygen atoms. When the spins are directed along the *b*- or *c*-axis of the crystal [17] the spectra are not described even qualitatively due to absence of splitting along the *c*-axis. The case of oxygen spins directed along the *a*-axis [7] is less obvious as the symmetry is correct but the ratio

of the splittings along the b - and c -axes is about 1.6, while our experiments determine it to be close to 1.

Magnetic orders based on CC provide a happy alternative. The model dubbed $CC-\Theta_I$ and formed by 4 orbital current loops for each CuO_2 plaquette [4] is not consistent with polarized neutron experiments as well as with our μSR data complemented by symmetry considerations. The most widely used model is $CC-\Theta_{II}$ with 2 opposite current loops O-Cu-O per CuO_2 plaquette [5]. This model has correct symmetry properties and predicts the ratio of the splittings along the b - and c -axes to be about 1.1, which is close to the observed value. The orbital magnetic moment extracted from our data amounts about $0.04 \mu_B$, which is comparable with the theoretical value on the order $0.1 \mu_B$ [4]. An alternative staggered orbital current phase with the same symmetry as $CC-\Theta_{II}$ but Cu-Cu-Cu currents [18] is also unlikely because the ratio of the splittings along the b - and c -axes is too large.

In summary, high magnetic field μSR studies in stoichiometric La_2CuO_4 reveal additional source of magnetic field on the muon consistent with Varma model of circulating currents [5].

Acknowledgments

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