Static magnetism in superconducting stage-4
$\text{La}_2\text{CuO}_4+y(y = 0.12)$

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Abstract

Zero-field (ZF) and transverse-field (TF) $\mu$SR measurements have been performed on a superconducting $\text{La}_2\text{CuO}_4+y$ crystal with $y = 0.12$ where excess oxygen is intercalated to form a stage-4 structure. Measurements are made on the same single-crystal sample studied in the neutron scattering measurements of Lee et al. [Phys. Rev. B 60 (1999) 3643] who find that both superconductivity and static incommensurate magnetic modulation develop below $T_c \sim T_N \sim 42$ K. The amplitude of the muon precession signal observed in ZF-$\mu$SR increases progressively below $T_N$ and saturates at $T \rightarrow 0$ to the value which indicates that a static magnetic field from Cu moments exists in only a part of muon sites corresponding to a magnetic site fraction of $V_M = 41\%$. The observed frequency $\sim 3.5$ MHz of the precession signal suggests the development of a large (at least 0.4 $\mu_B$) static Cu moment near the “magnetic” muon sites. Comparing the TF-$\mu$SR results with computer simulations, we obtained $V_M(T \rightarrow 0) = 42\%$. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: Superconductivity; Magnetic site fraction; $\mu$SR; Stripes

1. Introduction

Despite a large number of experimental and theoretical studies in the past 10 years on copper oxide superconductors, details of the interplay between superconductivity and magnetism are yet to be clarified. Undoped $\text{La}_2\text{CuO}_4$ is an antiferromagnetic insulator, but doping can change the hole density on the CuO$_2$ planes. One can substitute La$^{3+}$ ions with other metallic ions (Sr$^{2+}$, Ba$^{2+}$, etc.) or one can include excess oxygen. Dynamic incommensurate magnetic correlations have been observed by inelastic neutron scattering studies [1–5] in the superconducting state of La214 and Y123 with a wide range of hole concentrations. Recently, static correlations have been seen using neutron scattering [6,7], $\mu$SR [8–10] and other techniques in systems with the hole concentration near 1/8 per Cu. These incommensurate spin correlations are often discussed in terms of the formation of charge and spin stripes [6,11].

Recently Lee et al. [7] have performed neutron scattering studies on a single crystal of $\text{La}_2\text{CuO}_4+y$ with $y = 0.12$. Electrochemical oxidation leads, in
this case, to an intercalation compound with stage-4 structure [4]. Incommensurate Bragg peaks were found to develop below \( T_N \approx 42 \text{K} \), which coincides with the superconducting transition temperature \( T_c \) of the crystal. Here we report zero-field (ZF) and transverse-field (TF) \( \mu \)SR measurements performed using this same single crystal. We find that a static internal magnetic field from Cu moments exists at only about a half of the muon sites even at \( T \to 0 \). We will compare our results with those from an earlier \( \mu \)SR study of \( \text{La}_2\text{CuO}_4+y \) by Pomjakushin et al. [12], on samples with \( y = 0.02 \) and 0.04 which are in the miscibility gap between the lightly doped antiferromagnetic phase and the stage-6 superconductor.

2. ZF-\( \mu \)SR results

A single-crystal sample of \( \text{La}_2\text{CuO}_4+y \) (\( y = 0.12 \)) was mounted in an He gas flow cryostat, which can attain temperatures down to 2 K. The crystal was twinned with equal volumes in each domain, as confirmed by neutron scattering. \( \mu \)SR measurements were performed for two different orientations of the crystal: with the muon beam (and muon polarization) parallel in configuration I (C-I), and perpendicular (C-II) to the \( c \)-axis. Fig. 1 shows the time spectra of the muon spin polarization in zero external field in C-I. A slow relaxation is observed above 42 K: this is due to nuclear dipolar fields. The spectra below 42 K exhibit a damped oscillation, corresponding to development of static internal fields at the muon site from nearby Cu moments. The amplitude of this oscillation increases gradually with decreasing temperature below \( T_N \), but saturates towards a value of about 25-35\% of the full muon asymmetry.

The functional form of this oscillation is close to the Bessel function \( j_0(\omega t) \), which is expected in incommensurate spin-density-wave (SDW) systems [13] and was observed by ZF-\( \mu \)SR in the “1/8” materials [8,9]. The observed spectra in our crystal exhibit a damping faster than that of the Bessel function. Therefore, we use the fitting function

\[
P_\mu(t) = A_1 j_0(\omega t) \exp(-\lambda t) + A_2 G_{KT}(A, t),
\]

where \( G_{KT}(A, t) \) is the Kubo–Toyabe function which represents muons stopped at sites without static magnetic fields. The value of the total amplitude \( A_{tot} \equiv A_1 + A_2 \) is obtained in the measurements with a weak transverse field, and is fixed in the analyses of ZF results.

In Fig. 2(a) we show the precessing amplitude \( A_p \), defined as \( A_1/A_{tot} \), as a function of temperature. The amplitude increases gradually below \( T_N \) and saturates around the value of \( A_p \approx 35\% \) for C-I and 25\% for C-II at low temperatures. In crystal specimens with a unique orientation of magnetic field, \( A_p \) depends on the angle \( \gamma \) between the internal field and the muon spin direction, as \( A_p = \sin^2(\gamma) \). We assume the internal fields \( H_1 \) and \( H_2 \), as illustrated in the inset of Fig. 1, for the two twin domains.

When only a part of the muon sites, with site fraction \( V_M \), has a static internal magnetic field from the ordered Cu moments, the actual value of \( A_p \) becomes \( V_M \sin^2(\gamma) \). In the case of fully ordered powder samples, \( A_p \) should be 66\%, the average value of \( \sin^2(\gamma) \). As an example, we also include \( A_p \) obtained by ZF-\( \mu \)SR in a ceramic specimen of \( \text{La}_{1.475}\text{Sr}_{0.125}\text{Nd}_{0.4}\text{CuO}_4 \), which shows \( V_M \) approaching 100\% at \( T \to 0 \) [9].
In the case of twinned crystals, $A_p$ is the sum of the contributions of the two magnetic domains: $A_p = A_{p1} + A_{p2}$. For C-I, $\gamma_1 = \gamma_2 = \gamma$, as shown in Fig. 1. The angle between the projections of the two magnetic fields in the $ab$ plane is 90°. For C-II with the muon polarization perpendicular to the $c$-axis, $\cos^2(\gamma_1) = \sin^2(\gamma)\cos^2(\phi)$ and $\cos^2(\gamma_2) = \sin^2(\gamma)\sin^2(\phi)$. Therefore, $A_p = V_M \sin^2(\gamma)$ for C-I and $A_p = V_M(1 - 0.5\sin^2(\gamma))$ for C-II. As shown in Fig. 2, we have $A_p = 34\%$ for C-I and $A_p = 24\%$ for C-II. Thus we obtain $V_M = 34/2 + 24 = 41\%$.

Fig. 2(b) shows that the frequency of the oscillating signal increases very sharply below $T_N$ and rapidly attains a constant value of 3.6 MHz. This frequency is very close to that observed by $\mu$SR in $\text{(La}_{1.475}\text{Sr}_{0.125}\text{Nd}_{0.4})\text{CuO}_4$ and many other “1/8” systems [9], indicating that these systems and the “magnetic” sites of $\text{La}_2\text{CuO}_4$ share a common local spin structure. The observed frequency corresponds to a static Cu moment of about 0.4 $\mu_B$ around the muon site, as scaled from the 5 MHz $\mu$SR frequency in undoped $\text{La}_2\text{CuO}_4$, which has 0.6 $\mu_B$ per Cu.

3. TF-$\mu$SR results

We have also performed transverse-field $\mu$SR measurements in the $\text{La}_2\text{CuO}_{4.12}$ crystal with an external field of 1 kG applied parallel to the $c$-axis. The relaxation rate increases with decreasing temperature below $T \approx 42$ K. The observed spectra fit reasonably well with a simple phenomenological form

$$P_{\mu}(t) = A\exp\left(-\frac{t}{\tau_{TF}}\right)\cos(\omega t).$$

At low temperatures, the amplitude $A$ from this simple fit decreases to about 80% of the full amplitude at high temperatures, as shown in Fig. 3.

At $T \leq T_c$, i.e., $T \leq T_N$, there are two sources of relaxation for TF-$\mu$SR in the present case: (a) static
internal fields from Cu moments at the muon sites involved in the magnetic site fraction $V_M$; and (b) the field distribution due to the superconducting screening current which affects muons in the non-magnetic site fraction $(1 - V_M)$ and also possibly those muons in $V_M$ as well. The reduction of the amplitude $A$ is presumably due to the muons residing at sites with static internal magnetic fields large enough to depolarize the muon spin before the initial time range of the fit.

For a twinned crystal, when the transverse field is applied along the $c$-axis, the two orientations of the field give the same signal. We have calculated the expected field distribution at muon sites for a given value of the angle $\gamma$. For the muons within $V_M$, we assume a sinusoidal spatial modulation of the static internal field with the maximum amplitude corresponding to the 3.6 MHz muon frequency and obtained the local field as a vector sum of the internal and external fields. For the muons at the non-magnetic sites, we assume some broadening for a flux vortex structure to be consistent with the observed relaxation rate. With the resulting total field distribution, we simulate the TF-$\mu$SR signal, fitted with Eq. (2), and obtain the amplitude $A$ for different $\gamma$ as shown in the inset of Fig. 3. By comparing the simulation and experiments, we obtain $\gamma = 65^\circ$, which implies the magnetic site fraction $V_M$ of 42%. This is in a very good agreement with the ZF results.

4. Discussion

Our main results are: (1) the development of static magnetic order below $T_N$, which coincides with the superconducting $T_c$; (2) the site fraction $V_M$ increases progressively below $T_N$ towards $\sim 40\%$ at $T \to 0$; (3) the precession frequency $\nu$ below $T_N$ is almost independent of temperature; (4) $\nu \sim 3.5$ MHz at $T \to 0$; and (5) the line shape of the ZF relaxation is consistent with an incommensurate/stripe spin structure.

Pomjakushin et al. [12] performed $\mu$SR measurements on single crystals of La$_2$CuO$_{4.02}$ (A) and La$_2$CuO$_{4.04}$ (B). In (A), they observed the features (1), (2) and (3) with $T_N \sim T_c \sim 15$ K and $V_M(T \to 0) \geq 50\%$. In (B), they observed an onset of antiferromagnetism at $T_{N1} \sim 230$ K, with $V_M \sim 10\%$, followed by the features (1), (2) and (3) below $T_{N2} \sim T_c \sim 25$ K with $V_M$ increasing gradually below $T_{N2}$ to $V_M \sim 40\%$ at $T \to 0$. These samples are both in the miscibility gap between the lightly doped antiferromagnetic phase and the stage-6 superconductor which in clean samples has $T_c = 32$ K [4]. We infer therefore that in sample (A) impurities suppress both the phase separation and $T_c$. In (B), phase separation occurs and $T_c$ is only slightly reduced. Neutron scattering shows that the superconducting portion has at best a small ordered moment at low temperatures [14]. Presumably, therefore, the increased moment which Pomjakushin et al. [12] observed below 25 K in (B) arises from re-entrant spin glass behaviour in the antiferromagnetic portion of the sample. We note that, nevertheless, the ordered moments in (A) and (B) correspond to $\nu(T \to 0) \sim 5$ MHz as expected from La$_2$CuO$_4$ in the Néel state.

The present results (4) and (5) indicate that the spin configuration around the “magnetic site” of La$_2$CuO$_{4.12}$ is identical to those of the “1/8” systems, with incommensurate/stripe spin modulation. Features (2) and (3), together with neutron results [7], indicate that such a spin correlation exists in a dynamic/inelastic channel above $T_N$, and gradually comes into the static time window below $T_N$. Further measurements are required to determine whether superconductivity occurs in a location associated with “non-magnetic” muon sites or “magnetic” muon sites or both; and to determine the length scale associated with a possible separation between “magnetic” and “non-magnetic” regions.

Acknowledgements

This work has been supported by NSF-DMR-98-02000 (at Columbia) and NSF-DMR-97-04532 and DMR-98-08941 (at MIT).

References