Long-range magnetic ordering in the spin ladder compound $LaCuO_{2.5}$ probed by muon-spin relaxation

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(Received 16 May 1996)

A clear sign of long-range magnetic ordering has been observed by muon-spin rotation/relaxation in the two-leg spin-ladder compound $LaCuO_{0.25}$ below 125 K with a relatively large hyperfine field (~0.1 T). This result demonstrates that the ground state of the undoped $LaCuO_{2.5}$ is a magnetically ordered state, rather than the predicted quantum spin-liquid state.

Spin-ladder compounds such as $Sr_{n-1}Cu_{n+1}O_{2n}$ ($n=3,5,7,\ldots$), consisting of one-dimensional (1D) Cu-O atomic chains connected by Cu-O-Cu atomic rungs, have been drawing much interest due to the unique magnetic property of their ground states compared with other one- or two-dimensional systems. Theoretical calculation predicted[1–6] that ladders with an even number of legs have a ground state of correlated Cu^{2+} spin-singlet pairs (i.e., ''quantum spin-liquid'' state) and the first excited state is separated by a ''spin-gap'' energy determined by intraladder antiferromagnetic coupling J. On the other hand, the ladders consisting of an odd number of legs may be in a magnetically ordered state. Since the successful synthesis of single-phase materials under high pressure,[7] various experimental studies including susceptibility,[8] NMR,[8] and muon-spin relaxation[9] (μSR) measurements have been performed in the two-leg ($SrCu_2O_3$) and three-leg ($Sr_2Cu_3O_5$) compounds and confirmed much of the predicted features of their ground-state properties.

Meanwhile, the more interesting aspect of these compounds is the possible superconductivity in ladders with even number legs. It is well known that most of the recently discovered high-T_c superconductors have a common lammellar structure consisting of two-dimensional (2D) CuO_2 planes. In these materials the superconductivity emerges as the 2D antiferromagnetic ground state of the parent compounds is destroyed by carrier doping to the CuO planes, suggesting that the 2D antiferromagnetic spin correlation between spin-1/2 Cu^{2+} ions may play a crucial role in the mechanism of superconductivity. Since light doping of holes into two-leg ladders may also lead to a superconductivity due to the remaining spin gap,[1,6] the detailed study of such systems must be of great help for the understanding of high-T_c superconductivity.

Recently Hiroi and Takano found a ladder compound $LaCuO_{2.5}$ which exhibits the character of a two-leg spin-ladder system with a spin-gap energy on the order of 400 K estimated from the temperature dependence of susceptibility.[10] Unlike the previously found SrCuO compounds, it can be hole doped by replacing La with Sr, thereby opening a possibility for the systematic study of various spin/charge dynamics in this material. However, the ground-state property of undoped $LaCuO_{2.5}$ is still unclear: the sharp increase of the spin-lattice relaxation rate and subsequent disappearance of the ^{63}Cu NMR signal with decreasing temperature suggest some magnetic anomaly below about 150 K,[11] whereas no such anomaly is expected in an ideal two-leg ladder system because of the spin-singlet nature of the ground state. Thus, to shed light on the new magnetic property of this presumed ''spin-gap'' ground state, we investigated the undoped $LaCuO_{2.5}$ by muon-spin rotation/relaxation technique (μSR). The μSR technique has a unique feature complementary to both NMR and neutron scattering that it can be used to detect the local magnetic ordering with a sensitive time range of fluctuation from 10^{-4} to 10^{-9} s.

The μSR experiment was conducted at the RIKEN-RAL Muon Facility in the Rutherford Appleton Laboratory which provides a pulsed (70 ns width and 50 Hz repetition) beam of nearly 100% spin-polarized muons with a moment of 29 MeV/c. Polycrystalline samples prepared under the same procedure as reported elsewhere[10] (net weight ~2 g) were coarse ground and mounted on a sample holder made of 99.99% silver which was placed in a cryostat. Time-differential μSR measurements were performed over a temperature range of 8–270 K under both zero (ZF) and transverse (TF, $\simeq$0.02 T) magnetic fields. The magnitude of local magnetic (hyperfine) field at the muon site was determined by the longitudinal field (LF) dependence of the residual muon polarization (see below).

In the paramagnetic phase the relaxation is predominantly from randomly oriented Cu *nuclear* moments and we have a

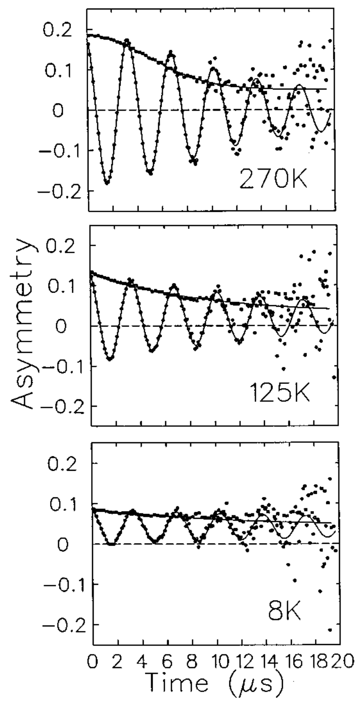

FIG. 1. Time differential μ-e decay asymmetry $A_0P\rho(t)$ ($A_0 \simeq 0.2$) under zero ($\rho=z$) and transverse ($\rho=x$) external fields in $LaCuO_{2.5}$ at some typical temperatures. Solid curves are fit results by simple models for $P_\rho(t)$ described in the text.

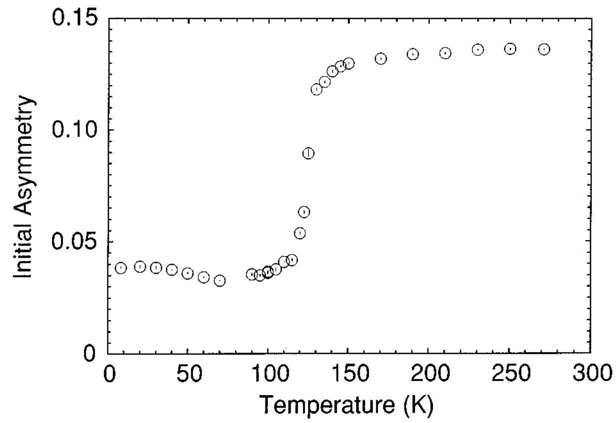

FIG. 2. Temperature dependence of the initial μ-e decay asymmetry $A_0P_z(0)$. (The component from the sample holder is subtracted, i.e., $A_0 \simeq 0.136$ corresponds to 100% muon polarization *in the sample*.) The sharp decrease around 125 K implies the onset of a long-range magnetic ordering.

simple Gaussian decay form $P_z(t) \simeq \exp(-\sigma_z^2 t^2)$ for the longitudinal and $P_x(t) \simeq \cos\omega_\perp t \exp(-\sigma_x^2 t^2)$ for the transverse relaxation where $\omega_\perp = \gamma_\mu H_\perp$ ($\gamma_\mu = 2\pi \times 135.54$ MHz/T) is the muon gyromagnetic ratio and $H_\perp$ is the external field perpendicular to the initial muon polarization) and σ_ρ ($\rho = z,x$) is the decay constant determined by nuclear dipolar moments. Figure 1 shows the observed ZF- and TF-μSR time spectra at some typical temperatures, where the observed time-dependent asymmetry is given by $A_0P_\rho(t)$ with $A_0(\simeq 0.2)$ being the μ-e decay asymmetry. The spectrum at 270 K is well fitted by assuming these functions with one component from the sample ($\sigma_z \simeq \sqrt{2}\sigma_x = 0.95 \times 10^5$ s^{-1}) and another component from the silver backing ($\sigma_z = \sigma_x = 0$). As temperature decreases the relaxation rate gradually increases below about 200 K and steep decrease of the muon polarization sets in below about $T_N \simeq 150$ K as shown in Fig. 2. This is a clear indication that the sample is in a magnetically ordered state.

In the ordered phase of a sample with randomly oriented powder grains (assuming that the muons occupy a unique crystallographic site), the longitudinal muon-spin polarization $P_z(t)$ has the form

$$P_z(t) = \tfrac{1}{3}\exp(-t/T_1) + \tfrac{2}{3}\exp(-t/T_2)\cos\omega t, \qquad (1)$$

where T_1 and T_2 are the respective longitudinal and transverse spin relaxation times and $\omega = \gamma_\mu H_{loc}$ is the muon angular frequency determined by the local magnetic-field magnitude H_{loc}. The transverse depolarziation is caused by a combination of temporal fluctuations in the direction of $\mathbf{H}_{loc}$, static spatial inhomogeneity of its magnitude, and the ambiguity of the time origin distributed over the range of beam pulse width δ. When the local field is large enough to satisfy $\omega\delta \gg 1$, the polarization for the second component is lost completely (effective $T_2 \ll \delta$) and only the first term is observed. The asymmetry from the $LaCuO_{2.5}$ sample is reduced from 0.136 to 0.038, which is in line with the interpretation that the residual asymmetry corresponds to this $\frac{1}{3}$ component. [The effect of the low transverse field is negligible in the ordered phase (i.e., $H_\perp \ll H_{loc}$) and the precession component at 8 K seen in Fig. 1 gives the background signal from sample holder made of silver.] The $\frac{1}{3}$ component comprises the muons stopped in grains where a component of $\mathbf{H}_{loc}$ is parallel with the initial muon polarization and therefore the relaxation rate $1/T_1$ is a function only of the fluctuation of $\mathbf{H}_{loc}$ in time. We found that $1/T_1$ is less than 10^4 s^{-1} at 8 K, indicating that the ordered magnetic moments are static.

The magnitude of H_{loc} is determined by measuring the recovery of the residual muon polarization under a longitudinal field, which is given by

$$P_z(t \to \infty) \simeq (\tfrac{1}{3}\ H_{loc}^2 + H_\parallel^2)/(H_{loc}^2 + H_\parallel^2), \qquad (2)$$

where $H_\parallel$ is the external field parallel with the initial muon polarization. Figure 3 shows such ''repolarization'' results at 120 and 8 K, where the field dependence is well reproduced by Eq. (2), indicating that the internal field $\mathbf{H}_{loc}$ takes a well-defined unique value rather than those distributed over a certain range in a spin-glass-like system. This result, together with the sharp onset of the reduction of asymmetry near 125 K strongly evidences the presence of a long-range antiferromagnetic ordering below 125 K. The magnetic moment develops with decreasing temperature and the corresponding H_{loc} becomes larger at lower temperatures. The fit result at 8 K yields $H_{loc} = 0.089(5)$ T [i.e., $\omega = 2\pi \times 1.21(6) \times 10^7$ s^{-1}], which is consistent with the previous assumption in Eq. (1) that $1 \ll \omega\delta$ ($\simeq 5$).

We lack the knowledge of the hyperfine coupling constant and the location of the muon site in the crystal which are necessary for the accurate determination of the magnitude of Cu moments. However, we can give a crude estimation of the value assuming that the hyperfine coupling is mainly due

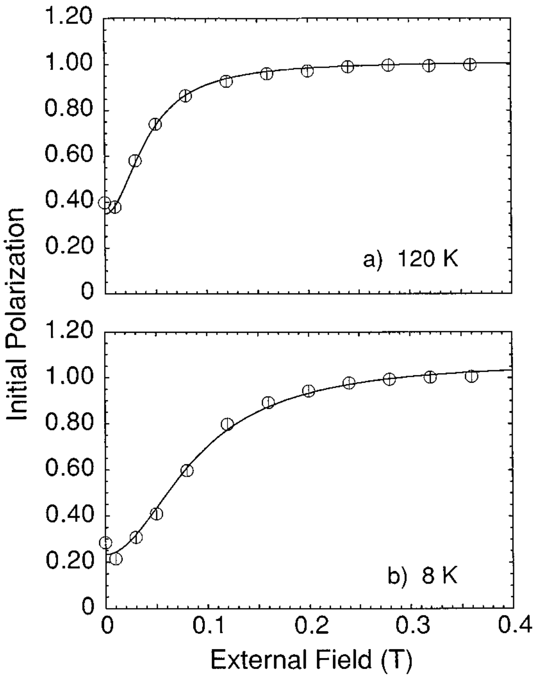

FIG. 3. LF quenching of the internal magnetic field at muon sites at (a) 120 K and (b) 8 K. Solid curves are fit results with a model described in the text. The magnitude of the local field at the muon site is deduced to be 0.043(2) T at 120 K and 0.089(5) T at 8 K from the fitting.

to the dipolar interaction (which is the case for the other magnetic oxides, e.g., MnO) and that the muon occupies a high-symmetry site between ladders. For example, if the muon site is the center of four oxygen atoms between two legs and rungs and the Cu moments are along with the ladder leg or rung, the observed local field $H_{loc}\simeq0.089$ T may correspond to the dipolar field from 0.3 to 0.7μ_B Cu moments with the alternating antiferromagnetic spin structure along the ladder. (The actual value of the moment depends on the direction of the ordered moments relative to the ladder.) A similar value is expected for the interladder sites surrounded by four oxygen atoms.

It is evident that the estimated magnitude of Cu moment is far too large to be reconciled with the ordinary three-dimensional antiferromagnetic spin structure: the development of such a large moment should be observed as a kink of the susceptibility near 125 K, which is absent in the previous data.[10] However, there remains a possibility that the susceptibility measurement was not sensitive enough to detect the antiferromagnetic ordering due to imperfect sample stoichiometry. Then the primary possibility is that the magnetic ordering is due to the ferromagnetic interladder coupling, which is relatively strong compared with that in SrCuO compounds where the interladder coupling is effectively weakened due to the frustration between neighboring sites.

Another possibility is the effect of the nonstoichiometric oxygen component which is often substantial in this compound. In a recent discussion on the possible coexistence of a spin-singlet ground state and antiferromagnetism in the Si-doped spin-Peierls system $CuGe_{1-x}Si_xO_3$ ($x\leq0.01$), it was concluded by Fukuyama *et al.*[12] that the spin-Peierls and/or ladder systems can easily be destroyed by a small amount of impurities. The appearance of antiferromagnetism observed in the current experiment might suggest the destruction of the singlet ground state due to the oxygen deficiency. Further investigation of the magnetic structure by neutron diffraction measurement would be of great help for the understanding of this peculiar magnetic property.

We thank the RIKEN staff at the RIKEN-RAL Muon Facility for technical support. This work was partially supported by a Grant-in-Aid for Scientific Research from the Ministry of Education, Science and Culture, Japan.

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