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Magnetic Order and Evolution of the Electronic State around $x = 0.12$ in $La_{2-x}Ba_xCuO_4$ and $La_{2-x}Sr_xCuO_4$

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Muon spin rotation (μ^+SR) measurement provides clear evidence of the antiferromagnetic order of Cu moments below 35 K for $La_{2-x}Ba_xCuO_4$ and below 15 K for $La_{2-x}Sr_xCuO_4$ in the narrow range of x where the high-T_c superconductivity (SC) is suppressed remarkably. The results suggest that the change of the electronic state coupled with the lattice instability is relevant to the local suppression of SC and freezing of spin fluctuations of the Cu moment.

KEY WORDS: High-T_c superconductivity; magnetic order; μSR; $La_{2-x}Sr_xCaO_4$.

1. INTRODUCTION

All of the high-T_c cuprates have similar phase diagrams: namely, an insulating and antiferromagnetically ordered state is quickly destroyed by hole doping and a superconducting and normal, metallic phase appears for further hole doping. However, careful investigations [1,2] demonstrated that the bulk superconductivity is suppressed anomalously in the local range of Ba concentrations around $x=0.12$ of $La_{2-x}Ba_xCu_4$ (LBCO). X-ray studies [3,4] found a structural transition from low-temperature orthorhombic (LTO) to low-temperature tetragonal (LTT) symmetry around the indicated Ba concentration. Since the structural transition to the LTT phase, with a small tilting of the CuO_6 octahedra around T_d= 60 K, turned out to have a pronounced effect on the electronic and magnetic properties [5], the LBCO system has attracted much renewed interest [6].

Some doping effect experiments have indicated that the suppression of T_c is primarily governed by the carrier concentration, not by the Ba atomic ordering with 1/8 number [7,8]. According to heat capacity and thermal expansion experiments [9], the LTT phase exists in a narrower range of x around $x=0.12$ than that reported by Axe *et al.* [3]. The magnitude of Cu moments becomes large in the narrow range around $x=0.12$ in LBCO [10,11], and a magnetic order of Cu moments is revealed by muon spin rotation (μSR) and NMR measurements [12–14]. Since the LTT phase contains more structural disorder than the LTO phase, localization of the electronic states is also expected. Antiferromagnetic (AF) correlations between Cu moments, which is relevant to the unusual magnetic properties in cuprates, seems to evolve with changes in the local lattice symmetry. As the AF spin fluctuations in the CuO_2 plane are considered to be one of the possible candidates for the Cooper pairing interaction, the origin of these anomalies in the La systems is crucial for understanding the high-T_c SC.

Although a slight lowering of T_c near $x=0.12$ was reported in the other La-based system, $La_{2-x}Sr_xCuO_4$ (LSCO) [15], neither the complete disappearance of SC nor the structural phase transition has been observed in LSCO. However, an anomaly of the ultrasonic attenuation [16] suggests the existence of LTO–LTT fluctuations at low temperature in LSCO. The substitution of Cu by small amounts of

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Zn apparently produces the local minimum of T_c around $x = 0.12$ in LSCO [17]. Moreover, in $La_{2-x-y}Sr_xNd_yCuO_4$, the structural LTO–LTT transition and the suppression of SC have been reported [18,19]. Therefore, detailed investigations on the electronic states in carefully prepared $La_{2-x}Sr_xCuO_4$ are required.

In this paper, we report the results of zero field (ZF) μSR measurement in $La_{2-x}Sr_xCuO_4$ as well as in $La_{2-x}Ba_xCuO_4$, which reveals clear evidence of the antiferromagnetic ordering of Cu moments in both systems.

2. EXPERIMENTAL

Samples of $La_{2-x}Ba_xCuO_4$ and $La_{2-x}Sr_xCuO_4$ were prepared by a solid-state reaction method. Details of the sample preparation have been described elsewhere [2,9]. Some samples were annealed at 500°C for 30 days in an O_2 atmosphere to improve homogeneity. The samples were checked by an X-ray diffraction measurement and found to be single phase. Heat capacity and thermal expansion measurements of the present LBCO system indicate that the structural LTO–LTT transition appears in the narrow range of $0.095 < x < 0.13$ [9].

The μSR was measured at UT-MSL/KEK by a ZF longitudinal relaxation method using a pulsed 4 MeV μ^+ beam. The stopped muon decays with a lifetime of 2.2 μsec and emits positrons preferentially along the spin direction. We detected these positrons using counters placed at forward and backward directions along the muon spin. The asymmetry of the muon spin polarization is defined as $[F(t) - B(t)]/(F(t) + B(t)]$, where $F(t)$ and $B(t)$ are the number of positrons detected by the forward and backward counters at time t.

3. RESULTS AND DISCUSSION

Figure 1 shows the composition dependence of T_c of the well-annealed LSCO samples. T_c is obtained by ac susceptibility measurements. The bulk superconductivity for $x = 0.115$ is almost completely suppressed. The volume fraction of diamagnetism is less than 1% for $x = 0.115$. Though the structural LTT–LTO transition on a macroscopic scale has not been observed in LSCO so far, some structural phase fluctuations are expected [16]. As a result, the electronic states in the narrow range of x are modified by the phonon instability even in LSCO as well as in LBCO.

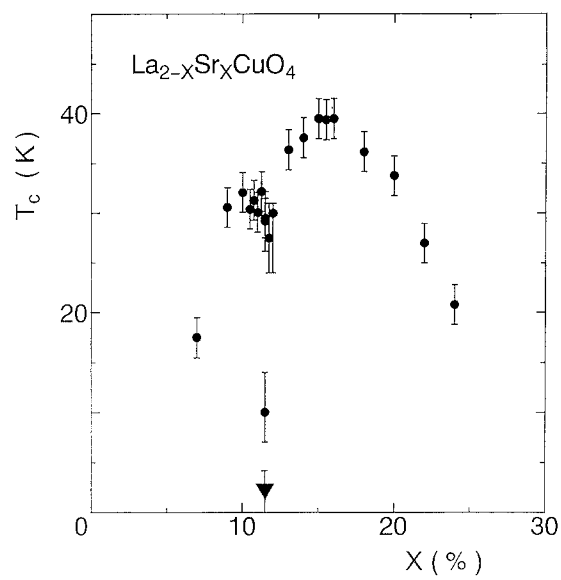

Fig. 1. x-dependence of superconducting transition temperature, T_c, of $La_{2-x}Sr_xCuO_4$. ▼ for $x = 0.115$ shows disappearance of the bulk SC down to the lowest temperature.

Thus, we believe that the anomalies of T_c in both systems is due to the same origin.

Figure 2 shows the time evolution of the asymmetry parameter of the long-annealed sample of $La_{1.875}Ba_{0.125}CuO_4$ at 7 K and $La_{1.885}Sr_{0.115}CuO_4$ at 2.5 K. Similar precession patterns are observed in both LBCO and LSCO samples near $x = 0.12$, although the precession amplitude shows a gradual decrease with time [12–14]. The observed precession pattern indicates the existence of a rather uniform magnetic field at the muon sites. Since the μSR is

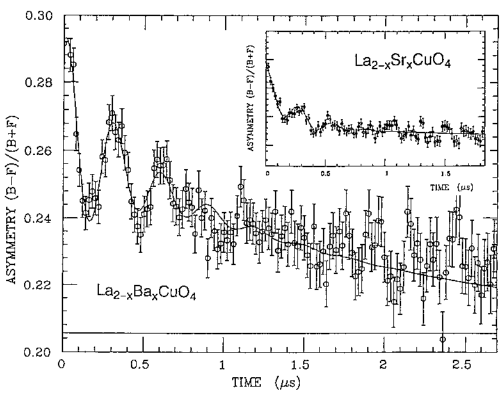

Fig. 2. Time evolution of the muon spin polarization under zero field for the long-annealed samples $La_{1.875}Ba_{0.125}CuO_4$ and $La_{1.855}Sr_{0.115}CuO_4$. The coherent precession indicates that an AF-like ordered state appears at low temperature.

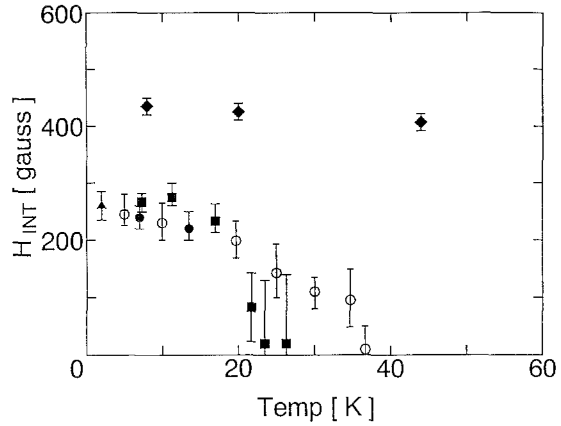

Fig. 3. Temperature dependence of the internal field at the muon sites. ●: $La_{1.875}Ba_{0.125}CuO_4$; ■: $La_{1.775}Sr_{0.125}Nd_{0.01}CuO_4$; ▲: $La_{1.885}Sr_{0.115}CuO_4$; ◆: La_2CuO_4 [20]; ○: $La_{1.875}Ba_{0.125}CuO_4$ [14].

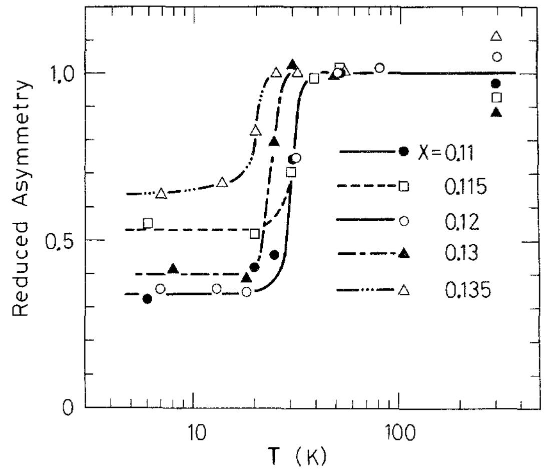

Fig. 4. Temperature dependence of the initial asymmetry of the muon spin polarization for $0.11>x>0.135$ in $La_{2-x}Ba_xCuO_4$. Each value is normalized by the value at 50 K.

measured under zero external field, the internal magnetic field at the muon site is attributed to a magnetic ordering of Cu moments. The rather homogeneous internal field shows the appearance of a coherent order of Cu moments, as opposed to a spin glasslike order. Thus, evidence for the AF-like ordering at low temperature is demonstrated in LSCO as well as in LBCO reported previously [12–14].

The temperature dependence of the internal field, H_{int}, at the muon site is shown in Fig. 3. H_{int} for LBCO and LSCO around $x=0.12$ is about 240 Oe which is about a half of that (420 Oe) in La_2CuO_4 [20]. Assuming the internal field arises from the dipole fields of Cu moments, the magnitude of the magnetic Cu moments is estimated to be about 0.3 μ_B, which is in good agreement with the value of 0.26 μ_B obtained from the low-temperature heat capacity measurement of LBCO [10,11]. The fact that there is no difference of the internal field between LBCO and LSCO indicates that the magnetic and electronic state are basically similar in both systems.

Recently, other evidence of magnetic order has been obtained by zero-field NQR investigations [6,21]. The broadened Cu spectra over 20–80 MHz are observed for $x<0.14$ in LBCO. The width of the Cu–NQR spectra does not change so much through both the HTT–LTO and the LTO–LTT phase transition [21]. The extremely broadened Cu spectra at low temperature are attributed to the Zeeman interaction through magnetically ordered Cu moments. A large enhancement of $1/T_1$ near 35 K (not at $T_d=60$ K) has been observed [21]. Recent La-NMR in $La_{2-x}Sr_xCuO_4$ and $La_{2-x}Ba_xCuO_4$ reveals the broadening of the La spectra at the 90 edge [22], which suggests that the internal field has a well-defined direction perpendicular to the c-axis and that the magnetic order is not of a spin glass-like but of the coherent AF-like order, consistent with the conclusion obtained from the μSR study.

Figure 4 shows the temperature dependence of the initial asymmetry parameter of μSR for $La_{2-x}Ba_xCuO_4$. When all the muons experience a static magnetic field, the initial asymmetry decreases to one-third of its magnitude in the paramagnetic state. Actually, the initial asymmetry at the lowest temperature was found to be almost one-third of that at high temperature in $La_{1.875}Ba_{0.12}CuO_4$, implying that the static magnetic order appears in the whole volume of the sample. The rapid decrease of the initial asymmetry below 35 K indicates the appearance of the internal static field at the muon sites at temperatures lower than $T_d=60$ K, and suggests that the magnetic ordering does not occur at the LTO–LTT transition. In addition, muon spin relaxation for $x=0.12$ has an exponential form as a function of T below 50 K and is largely enhanced near 35 K, suggesting that the magnetic transition is accompanied by a critical slowing down of the Cu moments and is rather long range in nature. The failure to observe a clear precession pattern only around $x=0.12$ is ascribed to an unavoidable inhomogeneity of the internal field owing to the random distribution of La and Ba atoms.

Similar decays of asymmetry parameter are also obtained for $0.105<x<0.12$ in LSCO. We define the magnetic transition temperature, T_N, as the temperature at which the initial asymmetry starts to decrease with decreasing temperature. The magnetic phase

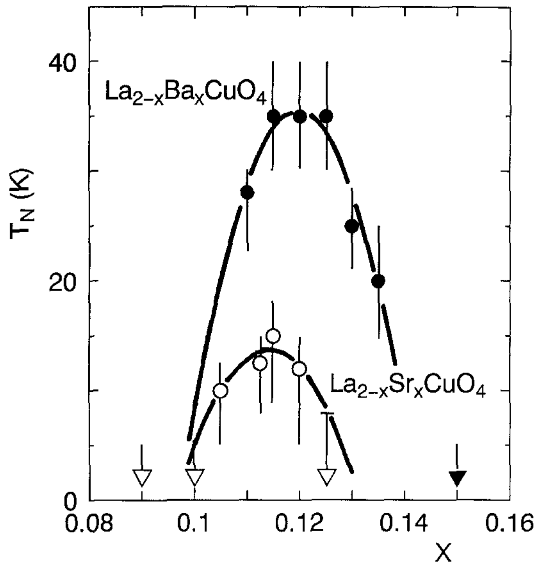

Fig. 5. Magnetic phase diagram for $La_{2-x}Ba_xCuO_4$ and $La_{2-x}Sr_xCuO_4$. The symbols ▼, ▽ indicate that no signs of magnetic order are observed by μSR.

boundaries of $La_{2-x}Ba_xCuO_4$ and $La_{2-x}Sr_xCuO_4$ are shown in Fig. 5. T_N shows its maximum value around $x=0.12$ for LBCO, but the maximum of T_N is at $x=0.115$ for LSCO. We would like to stress that any signs of magnetic ordering in the temperature dependence of the initial asymmetry and in the muon spin relaxation rate are not observed either for $x=0.08$ and 0.15 in LBCO, or for $x=0.9$, 0.10, and 0.125 in LSCO, indicating that the magnetism appears only in the narrow range of x near $x=0.12$. Clearly, the magnetically ordered state of Cu moments competes exclusively with the SC state.

The rotation of the CuO_6 octahedron below T_d lowers the local crystal symmetry of in-plane oxygen sites and introduces disorder in the system. The non-equivalence in the oxygen potential in the CuO_2 plane stabilizes the LTO–LTT transition [23] and may also cause charge transfer between in-plane oxygen and Cu sites, which causes localization of the electronic state and hence the restoration of the magnetic Cu moments near $x=0.12$ in the La systems. A recent theoretical study [24] of the role of spin–orbit interaction, which is responsible for the scattering of electrons in the CuO_2 plane from oxygen displacements, suggests that for the d-wave gap function, the spin–orbit interaction can be a strong pair breaker. The spin–orbit interaction is enhanced in a structural disordered phase [24]. Therefore, under the anharmonicity in the dynamical phonon mode as expected in LSCO or the disorderness on a microscopic scale as realized in LBCO, pair breaking of magnetic origin through spin–orbit interactions will be one of the possible causes of the strong suppression of T_c.

4. SUMMARY

We have investigated the ZF-μ^+SR for $La_{2-x}Ba_xCuO_4$ and $La_{2-x}Sr_xCuO_4$ around $x=0.12$ at which SC is remarkably suppressed. The observation of the precession pattern of the asymmetry of the muon spin polarization indicates the appearance of antiferromagnetic-like order of Cu moments in a narrow range of x. The present study suggests strongly that the static magnetic ordering of Cu moments, and hence, the freezing of the spin fluctuations of Cu moments, relates to the suppression of high-T_c superconductivity.

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