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Enhancement of static antiferromagnetic correlations by magnetic field in a superconductor $La_{2-x}Sr_xCuO_4$ with $x=0.12$

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Elastic neutron-scattering experiments have been performed to study effects of high magnetic field on static antiferromagnetic correlations coexisting with the superconductivity in $La_{1.88}Sr_{0.12}CuO_4$ ($T_c=12$ K). Under the field applied perpendicular to the CuO_2 plane its superconductivity is severely suppressed at 10 T. The intensity of incommensurate elastic magnetic peaks around the (π,π) point is, on the other hand, significantly increased at 10 T by as much as 50% of that of 0 T.

Neutron-scattering experiments on oxide high-T_c superconductors, in particular, those on the $La_{2-x}Sr_xCuO_4$ (LSCO) system[1,2] have revealed the existence of dynamical incommensurate spin fluctuations coexisting with the superconductivity. At present it is considered that such spin fluctuations would play an important role for the high-T_c superconductivity. Recent neutron-scattering studies on $La_{2-x}Sr_xCuO_4$, however, showed that *static* (*or quasistatic*) long-range antiferromagnetic correlations, which also coexist with the superconductivity, appear in the compounds with x of around 0.12.[3,4] These magnetic correlations are of a long period with the incommensurability similar to that observed for the dynamical spin fluctuations. The experiments further indicate that the antiferromagnetic correlations of these compounds show a long-ranged character in the CuO_2 plane but only a short-ranged one between the planes. The form of the coexistence of the incommensurate magnetic ordering and the superconductivity is, however, not clear at present.

The static antiferromagnetic correlations in the oxide superconductors were first observed in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ ($x=0.12$) in the studies of the so-called $\frac{1}{8}$ problem.[5] Around this hole concentration the superconductivity is dramatically suppressed, and such suppression is enhanced by a tetragonal structure induced at low temperatures. In the Nd-doped compound, a charge ordering is also clearly observed with this incommensurate magnetic order. These results were hence interpreted as ''the stripe ordering,'' in which antiferromagnetically ordered regions are separated by charged domain walls that act as magnetic antiphase boundaries.

The static antiferromagnetic correlations found in the superconducting $La_{2-x}Sr_xCuO_4$ system also may be directly related with this stripe ordering. For the samples with $x\sim 0.12$, the superconducting transition temperature T_c (onset) is ordinarily observed at around 32 K, showing a small dip in the curve which represents T_c vs Sr concentration x. In these compounds the antiferromagnetic correlations are observed below T_m (the magnetic onset temperature) of around 32 K—almost the same temperature as their T_c.[4] Quite recently, a compound with the same concentration x was grown very carefully under the air. This sample was then annealed at the oxygen atmosphere as usual; however, it shows a superconducting transition at T_c of 12 K (onset). The temperature T_c observed is considerably lower than that of the compounds described above. This suggests that the superconductivity of this sample is strongly suppressed corresponding with the $\frac{1}{8}$ problem in the related systems. The static antiferromagnetic correlations of this sample were investigated by neutron scattering in connection with its characteristic superconductivity. The results showed that the magnetic onset temperature T_m is about 25 K and is a little lower than that of the ordinary sample. Small shifts from the conventional incommensurate positions, $(\frac{1}{2}-\varepsilon\ \frac{1}{2}\ 0)$, etc., were also observed as in excess-oxygen doped La_2CuO_{4+y} (Ref. 6) and in $La_{2-x}Sr_xCuO_4$ grown at the oxygen atmosphere.[7] The details of the magnetic ordering of this sample will be shown elsewhere.[8] In the present work, effects of magnetic field on the superconductivity and static antiferromagnetic correlations of this compound have been studied up to 10 T. Under this high field the superconductivity and the static antiferromagnetic correlations are strongly affected. The results obtained here show aspects of the antiferromagnetic correlations of these superconducting compounds.

A single crystal with $x=0.12$ was grown with great care by the traveling-solvent-floating-zone (TSFZ) method under the air—the mixture gas of oxygen 20% and nitrogen 80% with the flow rate of about 100 cc/min. The speed of the crystal growth was 0.4 mm/h, which was two or three times slower than the usual cases. The size obtained was about 7 mm in diameter and 30 mm in length. The sample was then annealed at 900 °C for 4 days fully under the oxygen atmosphere. The superconducting transition temperature T_c of this sample was determined by a magnetization measurement with a superconducting quantum interference device (SQUID) magnetometer. From an additional increase of the elastic constant in the mixed state under magnetic fields, the volume fraction of the superconductivity was estimated to be more than 80%. As mentioned before, T_c of 12 K determined is rather low compared with that of the sample grown in the oxygen atmosphere. This low T_c is not due to simple impurity effects since any other impurity elements were not detected in the ICP (inductive coupled plasma) spectroscopy.

To examine effects of magnetic field on the superconductivity of the sample, electrical resistivity was measured under the fields up to 8 T generated by a superconducting magnet. Measurements were carried out in the CuO_2 plane by the conventional four-probe dc method. Magnetic fields were applied vertically to this CuO_2 plane. After field cooling the resistivity was measured under magnetic field with increasing temperature.

Neutron-scattering experiments were performed on the triple-axis spectrometer TAS-2 installed at the thermal neutron guide of JRR-3M of Japan Atomic Energy Research Institute (JAERI). The sample placed in an Al can filled with He gas was mounted in a conventional cryostat, and was cooled down to 4.2 K. The scattering measurements were done with the initial energy E_i of 14.7 meV under the diffraction arrangement. The monochromator was a set of pyrolytic graphite (PG) crystals, and the collimation was blank ~17′-(monochromator)-40′-(sample)-40′- (analyzer) - blank. A PG filter was placed before the sample to eliminate the higher-order harmonics. The scattering plane was set in the CuO_2 plane, which was denoted as the $(h\,k\,0)$ plane in the tetragonal notation. In this setting the lattice constants determined were $a\ (=b)=3.772$ Å in the high-temperature tetragonal (HTT) phase at room temperature. Upon cooling the system undergoes a structural transition to the low-temperature orthorhombic (LTO) phase. In the present work the tetragonal notation was conventionally used as in Ref. 7, even though the system is orthorhombic at low temperatures.

The transition temperature observed for this structural transition was the same as that in the previous report.[7] This implies that the Sr concentration x is very close to the nominal one. As will be shown later, furthermore, the incommensurability of the static magnetic correlations also shows that the Sr content is consistent with 0.12. The crystal mosaic is 0.4° full width at half maximum, and two twin domains populate almost equally. The crystallographic characteristics of the sample therefore indicate that the present compound has the same quality as the crystals studied before.[7] The inhomogeneity of this single crystal has not been observed up to now in the careful crystallographic studies. Here it should be noted that a T_c of around 10 K has not been reported before for crystals of LSCO with around $x=0.12$; however, in the literature such a low T_c was found for powder samples.[9] Since the powder samples are generally less prone to inhomogeneities or residual stresses, the low T_c of the present crystal may not be due to these extrinsic effects. However, further characterization of this unique crystal would be necessary.

Neutron-scattering experiments under magnetic fields were performed up to 10 T using a new type of split-pair superconducting magnet cooled by cryocoolers.[10] The field was applied vertically to the scattering plane. The experiment under magnetic field was carried out after field cooling. In these experiments the magnetic field was not changed in the superconducting state; that is, the field was always changed after the temperature was warmed up to 35 K, well above its superconducting transition temperature. Temperatures under magnetic fields were measured by a Cernox thermometer of Lake Shore Cryotronics, Inc.

The resistivity in the CuO_2 plane ρ_{ab} measured under magnetic fields is shown in Fig. 1 as a function of tempera-

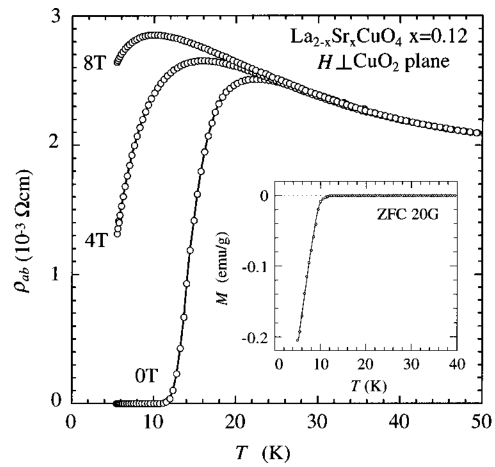

FIG. 1. Temperature dependence of the resistivity in the CuO_2 plane of $La_{2-x}Sr_xCuO_4$ with $x=0.12$. Magnetic fields up to 8 T were applied vertically to this plane. The inset shows the magnetization measured at 20 G after cooling in zero field.

ture. Here fields were applied up to 8 T in the direction perpendicular to the CuO_2 plane. The inset of this figure indicates the magnetization measured at 20 G, showing that the superconducting transition temperature T_c (onset) is 12 K. The resistivity observed indicates an upturn with decreasing temperature. This feature can be related with the strong suppression of the superconductivity due to the localization of holes. The zero-field resistivity at 50 K is about 2×10^{-3} Ω cm, which is rather high—about five times larger than that of the nominally identical crystals.[11,12] This resistivity is, however, still six times higher than that of Nd codoped LSCO sample that shows a similar severe suppression of the superconductivity near $x=\frac{1}{8}$;[13] thus the high resistivity may not be explained simply by the $\frac{1}{8}$ problem. The difference in the resistivity in these compounds should be clarified further.

With increasing magnetic field, as was observed in the resistivity of other samples with $x=0.12$,[12] the temperature where the resistivity begins to fall decreases significantly and the width at this transition does not show any appreciable broadening. These results sharply contrast with the field dependence of the resistivity of the optimum doped samples, in which the temperature where the resistivity starts to fall does not change but the width at the transition broadens remarkably. Since these features are attributed to strong superconducting fluctuations under magnetic field, the results obtained suggest that these fluctuations are fairly suppressed at this hole concentration of 0.12. Such a suppression of the superconducting fluctuations may be related to an appearance of the static antiferromagnetic correlations observing at $x\sim0.12$. As shown in the electrical resistivity at 0 T, the temperature where the resistivity falls to the zero value is 12 K, which corresponds well with the onset superconducting transition temperature T_c. Under high fields, this temperature where the resistivity reaches zero, that is, the onset T_c becomes fairly low. Under the field of 10 T the superconductivity is very severely suppressed.

The antiferromagnetic peaks were observed at four reciprocal lattice points of $(\frac{1}{2}\mp\varepsilon\ \frac{1}{2}\pm\delta\,0)$ and $(\frac{1}{2}\mp\delta \times \frac{1}{2}\pm\varepsilon\ 0)$—the incommensurate positions around the (π,π)

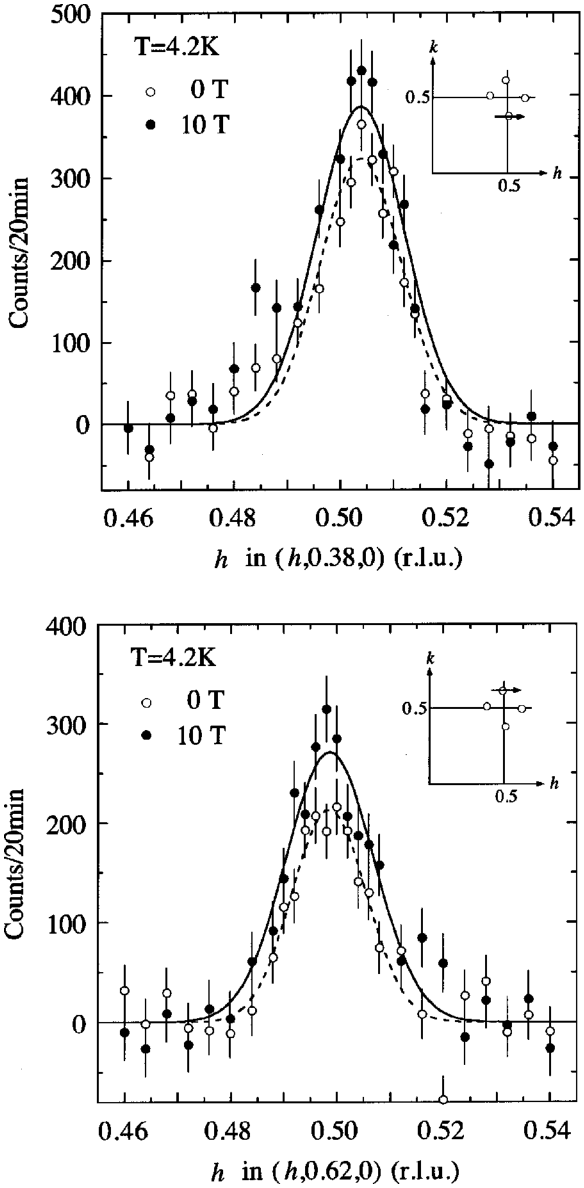

FIG. 2. Intensities of antiferromagnetic peaks of $La_{2-x}Sr_xCuO_4$ with $x=0.12$ measured at 4.2 K under magnetic field of 0 and 10 T at two different positions. Each scan was performed along the lines in the reciprocal space indicated in the inset. The background intensities measured at 35 K were subtracted.

point, as was shown in the previous studies.[6,7] These positions are indicated in the insets of Fig. 2. Here, the incommensurate wave vector ε was determined to be 0.120 (± 0.002). The wave vector δ shows a shift from the conventional incommensurate position, and was obtained to be 0.004 (± 0.001) at 4 K. The incommensurability ε is consistent with that of $La_{2-x}Sr_xCuO_4$ with $x=0.12$ and of $T_c=32$ K, which was grown and annealed under the oxygen atmosphere. However, the value of δ is a little smaller than that of the sample grown under the oxygen atmosphere. This shift is fairly small, and at present the physical meaning of this shift is not clear.

Figure 2 shows the intensities of these antiferromagnetic peaks measured at 4.2 K under magnetic fields of 0 and 10 T. These intensities were measured at around $(\frac{1}{2}\mp\delta\ \frac{1}{2}\pm\varepsilon\ 0)$ along the lines in the reciprocal space indicated in the inset.

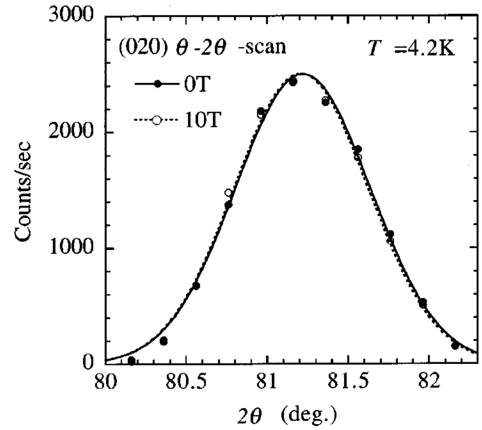

FIG. 3. Intensities around the (0 2 0) nuclear peak of $La_{2-x}Sr_xCuO_4$ with $x=0.12$ under magnetic field of 0 and 10 T.

Here the background intensities measured at 35 K were subtracted for each scan. In the profile of the scattering intensity small shoulders around the main antiferromagnetic peaks are observed. The origin of the shoulders is, however, unclear. In the present experimental conditions the resolution is estimated to be at about 0.012 Å^{-1}. When the magnetic peaks are fitted to a Gaussian function, these peaks are found to be only a little broader than the resolution limit. The μSR experiments, on the other hand, indicated that the magnetic transition temperature T_m observed is lower than that measured in neutron scattering,[14] and therefore the static antiferromagnetic correlations still fluctuate in time. In neutron-scattering experiments the time constant is rather fast and the resolutions have widths for space and energy. Thus the antiferromagnetic correlations observed in neutron scattering should be considered as a quasistatic ordering which fluctuates in space and time in some extent.

As shown clearly in the lower panel of this figure, the intensity under 10 T at $k=0.620$ is considerably increased compared with that under 0 T. The width of the diffraction peak also increases a little under magnetic field. Thus, the increase in the integrated intensity is rather large and estimated to be about 50(± 12)% of the intensity at 0 T. The enhancement for the intensity at $k=0.380$ under 10 T shown in the upper panel is also apparent; however, the increase in the intensity is a little small. In this case the intensity is increased at the rate of about 30(± 9)%. The experiments at the other two incommensurate positions $(\frac{1}{2}\mp\varepsilon\ \frac{1}{2}\pm\delta\, 0)$ also showed the similar enhancement of the intensity. Within the experimental accuracy the positions where the magnetic peaks appear were not changed by magnetic field.

Figure 3 shows the scans around the nuclear peak (0 2 0) under 0 and 10 T. The peak position and the intensity were not changed under these fields within the experimental accuracy. This result indicates that there are no appreciable changes in the crystal lattices and also in the conditions of the sample. It is noted that the sample was not moved under the field. Thus the changes in the magnetic intensity shown above are intrinsic in the system.

These experimental results indicate that under the magnetic field of 10 T the superconductivity is severely suppressed and, on the other hand, the static antiferromagnetic correlations are strongly enhanced. Such a large enhancement of the static magnetic correlations could be explained by the suppression of spin fluctuations along the direction of the magnetic field. Since the field of 10 T corresponds to about 0.6 meV in energy, the weight of the spin fluctuations with the low energies (∼below around 1 meV) would be effectively increased. The suppression of these spin fluctuations can be observed as a static (or quasistatic) magnetic ordering in the neutron-scattering experiment with the finite energy resolution. The increase of the low-energy spin fluctuations, on the other hand, may break the superconducting Cooper pairing and lead to the suppression of the superconductivity of the system. The remarkable effects observed are due to the matching of the energy scale among the *high* magnetic field (10 T∼0.6 meV), the *low* superconducting transition temperature (12 K∼1 meV), and the energy width of the elastic neutron scattering (∼0.8 meV). It is pointed out here that vortexes also can induce a magnetic ordering. In the vortexes the normal state is produced over the correlation length. There the energy of the spin fluctuations would become low. The spin fluctuations with these low energies can be suppressed by magnetic field as was mentioned above, and the static antiferromagnetic correlations will be enhanced. Such an antiferromagnetic order induced in the vortex cores was recently discussed in theoretical consideration on the *d*-wave superconductivity.[15]

To conclude, neutron-scattering experiments on newly grown $La_{2-x}Sr_xCuO_4$, $x=0.12$, with the low T_c of 12 K showed that the static antiferromagnetic correlations are significantly enhanced under magnetic field of 10 T. At this field the superconductivity of the system is severely suppressed. Since T_c is fairly low in this system, the weight of the spin fluctuations with low energies would be effectively increased by the field of 10 T. The enhancement of the static antiferromagnetic correlations can be explained by the suppression of these spin fluctuations under magnetic field.

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