Spin Pseudo Gap in La$_{2-x}$Sr$_x$CuO$_4$ Studied by Neutron Scattering

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Spin excitations of La$_{2-x}$Sr$_x$CuO$_4$ have been studied using inelastic neutron scattering techniques in the energy range of $2 \text{ meV} \leq \omega \leq 12 \text{ meV}$ and the temperature range of $8 \text{ K} \leq T \leq 150 \text{ K}$. We observed a signature of a spin pseudo gap in the excitation spectrum above $T_c$ for the slightly overdoped sample with $x = 0.18$. On heating, the spin pseudo gap gradually collapses between $T = 80 \text{ K}$ and $150 \text{ K}$. For the $x = 0.15$ and $0.20$, although the visibility of gap-like structure at $T \sim T_c$ is lower compared to the $x = 0.18$ sample, the broad bump of $\chi''(\omega)$ appears at $\omega \sim 5 \text{ meV}$, close to the spin-gap energy at base temperature, suggests the existence of the spin pseudo gap in the normal state.

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I. INTRODUCTION

One of the most remarkable phenomena observed in high-$T_c$ cuprates is the opening of a pseudo gap above the superconducting (SC) transition temperature ($T_c$) in excitations of charge as well as spin. To elucidate the basis of this effect, many experimental studies have been carried out using various techniques including photoemission spectroscopy [1-5], NMR [6,7], neutron scattering [8] and others. Nevertheless, the microscopic origin of the pseudo gap remains controversial. Many theoretical models have been proposed based on preformed Cooper pairs or SC pairing fluctuations [9], the RVB state [10,11], a precursor to a spin-density-wave-state [12] and the formation of dynamical charge stripes. To reduce the considerable theoretical and experimental confusion regarding the basis of the spin pseudo gap, further experimental studies are required.

In principle, pseudo gap in magnetic excitations, so-called spin pseudo gap, is observable using NMR as well as neutron scattering. Especially, neutron scattering spectroscopy has the unique benefit of being able to detect directly the energy gap as well as the momentum dependence of spin fluctuation. In fact, neutron scattering measurements on the YBa$_2$Cu$_3$O$_{6+y}$ (YBCO) system observed for the first time a gap-like structure in the energy spectrum of dynamical spin susceptibility $\chi''(\mathbf{q}, \omega)$ near the $(\pi, \pi)$ position in the normal state [8]. For underdoped La$_{2-x}$Sr$_x$CuO$_4$ (LSCO), however, no evidence for the existence of a pseudo gap in the normal state nor for a spin gap in the SC state has been obtained [13, 14]. In contrast, recent neutron scattering studies of optimally or slightly overdoped LSCO have revealed a well-defined energy gap in the incommensurate spin fluctuations below $T_c$ [14-17]. Although results of our previous neutron scattering study indicated the existence of a spin pseudo gap at $T_c$ for slightly overdoped LSCO with $x = 0.18$ [14], no systematic examination of this system has been performed.

In the present work, we report a comprehensive study of the magnetic excitations in the normal state of LSCO with $x = 0.18$ and 0.20. From a comparison of these results with those arising from the previous measurements on LSCO with $x = 0.15$ [14], we conclude that a spin pseudo gap does exist in the LSCO system but the visibility or the stability of the gap-like energy spectrum is sensitively affected by the Sr or hole concentration in these samples.

II. EXPERIMENTAL DETAILS

Single crystals of La$_{2-x}$Sr$_x$CuO$_4$ ($x = 0.15, 0.18, 0.20$) were grown by the traveling solvent floating zone method (TSFZ) using lamp-image furnaces [18, 19]. The as-grown single crystals were annealed under oxygen gas-flow at 900°C for 50 hrs to remove any oxygen defects. Both $x = 0.15$ and $x = 0.18$ crystals had been previously used in neutron scattering measurements with some data also being reproduced in the present paper [14]. Onset temperatures of the SC transition measured by SQUID magnetometers under a magnetic field of 10 Oe are 37.5 K for the $x = 0.15$ sample, 36.5 K for the $x = 0.18$ sample, and 30.0 K for the $x = 0.20$ sample (Fig. 1). $T_c$ values for the $x = 0.18$ and 0.20 samples are lower than that of the $x = 0.15$ sample due to overdoping.

Since $T_c$ is relatively insensitive to Sr concentration near the optimally doped region, we investigated the structural phase transition temperature ($T'_c$) between the high temperature tetragonal (HTT) and low temperature orthorhombic (LTO) phases. The (1,1,0) (in I4/mmm notation) fundamental Bragg peak intensity was monitored as a function of temperature for both $x = 0.18$ and 0.20
samples. We note that the intensity changes like an order parameter upon entering the LTO phase due to the suppression of extinction effects on the neutron beam caused by formation of twinned domain. For the 0.15 sample, intensity of the (0,1,4) (in Bmab notation) superlattice reflection was monitored as a function of temperature. We fitted the observed temperature dependence of the peak intensity using a phenomenological function of \((1-T/T_s)^2\), including a Gaussian distribution of \(T_s\) (the half width at half maximum of the Gaussian is defined as \(\Delta T_s\)) to evaluate \(T_s\) quantitatively. The index of \(\beta\) was fixed at a value of 0.35. As a result, \(T_s\) and \(\Delta T_s\) were respectively determined to be 191 K and 10 K for the x = 0.15 sample, 111 K and 13 K for the x = 0.18 sample, and 92 K and 22 K for the x = 0.20 sample. Larger \(\Delta T_s\) for the overdoped sample is due to larger Sr dependence of \(T_s\). Details of sample preparation and characterization have been reported elsewhere [14, 18-20].

Inelastic neutron scattering measurements were performed using the Tohoku University triple-axis spectrometer TOPAN in JRR-3M of JAERI at Tokai. The incident (final) neutron energy was fixed at \(E_i\) (\(E_f\)) = 14.75 meV or 13.75 meV using the (002) reflection of a pyrolytic graphite monochromator and an analyzer. The typical horizontal collimator sequence was 40'-100'-S-60'-80' or 40'-60'-S-60'-80' where S denotes the sample position. A pyrolytic graphite filter and a sapphire crystal were inserted to reduce neutron beam flux from the higher order reflection and high-energy neutrons, respectively. In order to increase the sample volume, two or three single crystalline rods were assembled and mounted in an Al container filled with He thermal exchange gas. A closed cycle \(^4\)He refrigerator was used to cool samples down to 8 K with temperatures monitored by a Si diode.

III. ANALYSIS OF NEUTRON SCATTERING EXPERIMENTS

The energy dependence of incommensurate peak intensity is depicted (Fig. 2) after making following correction on raw data arisen from instrument. Background-subtracted inelastic scattering intensities, \(I\), taken with \(E_i\)-fixed mode are corrected into \(I'\) using the following equation,

\[
I' = I \cdot \frac{\tan \theta_A}{k_f^2} \tag{1}
\]

where \(k_f\) and \(\theta_A\) denote wavenumber of the scattered neutrons and the analyzer angle, respectively [21]. On the other hand, for experiments with \(E_f\)-fixed mode, the counting time was corrected. This was necessary as the count rate of a fission monitor for the incident beam flux depends on the incident neutron energy due to the energy dependence of intensity of the higher order reflected beam.

For quantitative analysis, we fitted the observed magnetic intensity, which is proportional to the dynamical structure factor, \(S(q, \omega)\), and dynamical magnetic susceptibility, \(\chi''(q, \omega)\), using the following equations convoluted with an instrumental resolution function,

\[
S(q, \omega) = \frac{1}{1 - \exp(-\frac{\omega}{\xi_f^2})} \cdot \chi''(q, \omega) \tag{2}
\]
FIG. 3: Energy dependence of the q-spectrum of the incommensurate magnetic signals for x = 0.18 obtained at T = Tc by a constant energy-scan around (π, π). The scan trajectory and the instrumental resolution ellipsoid are schematically shown in the inset of (c). The incident neutron energy was fixed at Ei = 14.75 meV. Solid lines are the results of fits convoluted with the instrumental resolution using the background shown by the dashed line (see text for detail). The energy dependence of back ground is depicted in the inset of (a). Horizontal bars depict the instrumental q-resolution. The intensity at around (π, π), k = 0.5 in the figure, reflects the effect of incommensurate peaks located outside of the scan trajectory (see inset of (c)).

\[ \chi''(q, \omega) = A_\omega \sum_{\delta=1,4} \left\{ \frac{\kappa_\omega}{|q - q_{\delta, \omega}|^2 + \kappa_\omega^2} \right\} \quad (3) \]

where \( k_B \), \( q_{\delta, \omega} \), \( \kappa_\omega \), and \( A_\omega \) denote the Boltzmann constant, the four-fold positions of incommensurate peaks around (π, π), the q-width and a scaling factor, respectively. The absolute values of q-integrated dynamical magnetic susceptibilities, \( \chi''(\omega) \), are determined using phonon intensities as described previously [14].

IV. RESULTS

Figure 2 shows energy dependence of incommensurate peak intensity for x = 0.15 and 0.18. At T \( \ll T_c \), both samples have a clear gap spectrum as reported in ref. [14, 15]. Upon heating to \( T = T_c \), on the other hand, the gap structure is disappeared for x = 0.15. Whereas, for x = 0.18, although the intensity does not drop into 0, reduction of peak intensity at low energy region still occurs, which suggests that a spin pseudo gap is open. Note that the intensity below \( \omega = 6 \) meV increases with increasing temperature while it decreases above \( \omega = 6 \) meV, which can be owing to sum rule.

Typical raw q-spectra of magnetic peaks for the x = 0.18 sample taken by the \( E_i \)-fixed mode at \( T \sim T_c \) are shown in Fig. 3. Trajectory of the scan is illustrated in the inset of Fig. 3(c). Solid lines depict the results of fits using the scattering function described in equation (2) and (3) convoluted with the resolution function. Long dashed lines depict background. As shown, the solid lines reproduce the observed q-spectra quite well. Larger magnetic intensity near the (π, π) position at \( \omega = 6 \) meV than the left side tail dominantly arises from incommensurate peaks outside of the scan trajectory collected by the finite instrumental resolution (see inset of Fig. 3(c)). Energy dependence of back ground at incommensurate peak position (k = 0.37 in Fig. 3) is depicted in inset of
The energy spectrum at $x = 0.18$ sample. As presented in Fig. 4(a) and in our previous paper [14], the energy spectrum at $T = 8$ K due to the opening of energy gap, the line-width is not defined below the gap energy. Solid lines provide guides to the eyes.

Fig. 3(a). Background at $T_c$ is nearly independent of temperature, while the bump around $\omega = 6$ meV remains, suggesting that the bump near the energy gap disappears with the closing of the spin pseudo gap. Possibly, the bump is an effect of the spin pseudo gap as discussed in the previous paper [14].

Figure 4 shows energy dependence of line-width at $x = 0.18$ sample. As presented in Fig. 4(a) and in our previous paper [14], the energy spectrum at $T = 8$ K exhibits a clear cut-off near $\omega = 6$ meV ($E_{gap}$) with a broad bump, while $\chi''(\omega)$ completely vanishes into the background below $\omega \sim 4.5$ meV. Upon heating the sample to $T_c$, magnetic intensities are found to appear below $\omega = 4.5$ meV (Figs. 2(b) and 3(a)). Nevertheless, steep decrease in $\chi''(\omega)$ below $\omega = 6$ meV remains, suggesting a presence of spin pseudo gap. The bump around $\omega = 6$ meV is also survived. Dashed line in Fig 4(b) depicts a fit assuming a gapless state using Eq. (4), which details are described in section 5. Upon heating the sample to $T = 80$ K, the gap-like structure becomes broad, while at $T = 150$ K, $\chi''(\omega)$ is nearly independent of temperature, consistent with a collapse of the spin pseudo gap.

Fig. 5 shows the energy dependence of line-width. At $T = 8$ K (Fig. 5(a)), the line-width increases with decreasing the energy down to $\omega = 6$ meV. Below the energy gap, the line-width cannot be defined due to the absence of magnetic intensity. At $T = 36$ K, the enhancement around $\omega = 6$ meV still remains as a small bump (Fig. 5(b)). On the other hand, upon heating to $T = 80$ K, the width becomes nearly independent of energy. It seems that the bump near the energy gap disappears with the closing of the spin pseudo gap. Possibly, the bump is an effect of the spin pseudo gap as discussed in the previous paper [14].

Figure 6 shows the raw q-spectrum of magnetic peaks for the $x = 0.20$ sample at $T \sim T_c$, taken at $\omega = 3, 4$ and 6 meV under the $E_f$-fixed mode. Well-defined incommensurate peaks with the peak-width similar to that of the $x = 0.18$ are observed. We find that the peak intensity has a weaker energy dependence compared to the $x = 0.18$ sample. Note that a clear energy gap exists for $T \ll T_c$ (see Fig. 7(a)) with the gap energy of $5 \sim 6$ meV, which is slightly smaller than that of the $x = 0.18$ sample. At $T \sim T_c$, as shown in the Fig. 7(d), however, no clear gap-like structure is observed, whereas a weak bump is seen at $\omega \sim E_{gap}$.

Fig. 7 summarizes the energy dependences of $\chi''(\omega)$ for the $x = 0.15$ (data from ref. [14]), 0.18 and 0.20 samples. At $T = 8$ K, all three samples show a well-defined energy gap with a broad maximum near $\omega = 6$ meV. Upon
heating to \( T = T_c \), a gap-like structure of \( \chi''(\omega) \) remains only for the \( x = 0.18 \) sample. For the \( x = 0.15 \) and 0.20 samples, \( \chi''(\omega) \) decreases linearly with decreasing energy below \( \omega \sim 5 \) meV, approaching zero only as \( \omega \to 0 \). The broad peak in \( \chi''(\omega) \), on the other hand, is still observed in all three samples.

V. DISCUSSION

A signature of spin pseudo gap in the energy spectrum of incommensurate spin fluctuations was first obtained for the 2-1-4 type hole-doped cuprates. The present neutron scattering experiment shows that a gap-like structure at \( T \sim T_c \) is observable only in a narrow Sr concentration range near \( x = 0.18 \). For the \( x = 0.15 \) and 0.20 samples, although a clear gapped spectrum was observed at \( T \ll T_c \), the spin pseudo gap was poorly defined at \( T \sim T_c \).

We first discuss the broad peak in \( \chi''(\omega) \) with \( \omega \sim 6 \) meV in view of the spin pseudo gap. In general, for the correlated spin systems without magnetic long range order, \( \chi''(\omega) \) exhibits a broad peak even in the normal state.

For example, \( \chi''(\omega) \) can be described in a Lorentzian form

\[
\chi''(\omega) = C \frac{\omega^\gamma}{\gamma^2 + \omega^2},
\]

where the peak energy nearly corresponds to \( \gamma \), a characteristic energy-scale of spin fluctuations of the system. Thus, the broad peak in \( \chi''(\omega) \) does not simply correspond to a spin gap or spin pseudo gap. For the optimally doped LSCO, however, \( \chi''(\omega) \) has already been studied over a wide energy range by pulse neutron scattering and a broad peak was observed at \( \omega = 22 \sim 40 \) meV [22-24]. Then, combining both results from the present low energy experiments and pulse neutron scattering at high energy, there should exist two peaks in \( \chi''(\omega) \). A poorly fit using Eq. (4) to \( \chi''(\omega) \) of \( x = 0.18 \) at \( T = 36 \) K where \( \gamma \approx 9 \) meV is obtained (Fig. 4(b)) suggests that the \( \chi''(\omega) \) at low energy region can not be explained by the simple Lorentzian form. Possibly, the peak at lower energy is the result of forming an energy gap below \( T_c \) and even a spin pseudo gap at \( T \sim T_c \). Fig. 8 depicts a conceptual drawing of \( \chi''(\omega) \). In normal state, \( \chi''(\omega) \) depends linearly on \( \omega \) at low energies near \( \omega = 0 \). If spin gap or spin pseudo gap opens, the \( \chi''(\omega) \) near 0 deviates downward from the linear dependence and a bump appears near the gap-energy.

According to the fact that the broad peak in \( \chi''(\omega) \) with \( \omega \sim 6 \) meV is also observed in \( x = 0.15 \) and 0.20, we speculate that the spin pseudo gap remains at \( T \sim T_c \) in these samples although steep decrease of \( \chi''(\omega) \) is less defined. We note that the visibility or the stability of the pseudo gap highly depends on the gap-edge structure of the energy spectrum at \( T \ll T_c \), which is broader at \( x = 0.15 \) and 0.20 than at \( x = 0.18 \). If gap-edge is broader at base temperature, gap-like structure or spin gap - energy.

![FIG. 7](image-url)

![FIG. 8](image-url)
pseudo gap is more easily smeared out by heating. Then the spin pseudo gap is difficult to detect even if it exists. One possible reason for the gap-edge broadening in the ground state is the spatial distribution of the size of energy gap due to the inhomogeneous carrier distribution and/or chemical potential-randomness. The effect of the randomness becomes dominant below $x \sim 0.15$ due to the substantial decrease in the effective carrier concentration [25]. For the $x = 0.20$ sample, on the other hand, the spin pseudo gap is possibly degraded by overdoping as reported by many other indirect experiments. At present, however, the direct relation between the instability of spin pseudo gap and the gap-edge broadening is not clear.

Many studies on pseudo gap have been predicting the existence of two characteristic crossover temperatures, $T_o$ and $T^*$ ($T_o > T^*$). For example, at both temperatures of $T_o$ and $T^*$, suppression of magnetic susceptibility and downward deviation of inplane resistivities from linear temperature dependence were observed [26-28]. Furthermore, at $T^*$, the electronic specific-heat coefficient is slightly suppressed [29]. The origin of such $T_o$ is interpreted by the onset of antiferromagnetic correlation. Below $T^*$, on the other hand, the pseudo gap is expected to open. According to NMR measurements, the value of $T^*$ for $x = 0.20$ is about $T = 200$ K [7]. Taking into account the difficulty of determining $T^*$ correctly, the thermal scale of the spin pseudo gap observed in the present study appears to be consistent with the results of NMR.

The relation between the pseudo gap and superconducting gap is also of great interest. According to angle-resolved photoemission measurements in LSCO, large and small charge pseudo gaps exist with magnitudes of about 100 meV and 30 meV, respectively [4, 5]. The present results, however, suggest that a single spin gap structure in the $\chi''$ spectrum at base temperatures gradually transforms into the spin pseudo gap upon heating.

VI. CONCLUSION

We observed a signature of a spin pseudo gap in the energy spectrum of $\chi''(\omega)$ for the slightly overdoped $x = 0.18$ samples of LSCO by neutron scattering experiments. Upon heating, the spin pseudo gap gradually collapses between $T = 80$ K and 150 K. For the $x = 0.15$ and $x = 0.20$ samples, on the other hand, the gap-like structure is poorly defined at $T = T_c$. However, the broad bump in $\chi''(\omega)$ at $T \sim T_c$ suggests the remaining of the spin pseudo gap.

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