High-energy spin excitations in heavily electron-doped Pr$_{1-x}$LaCe$_x$CuO$_4$

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Abstract. We performed neutron scattering measurements on the overdoped sample of electron-doped superconductor Pr$_{0.83}$LaCe$_{0.17}$CuO$_4$ ($T_c$~16 K) to clarify an overall spin excitation spectrum. Clear commensurate low-energy spin fluctuations were observed at the $(\pi, \pi)$ reciprocal position below $\sim$10 meV. With increasing energy transfer, the width of commensurate peak drastically broadens and the intensity rapidly decreases. However, magnetic scattering intensities at high-energy region around 100 meV were confirmed to exist. These results are qualitatively same with those obtained for optimally-doped Pr$_{0.80}$LaCe$_{0.21}$CuO$_4$, while the broadening of peak-width against energy transfer is more marked and the intensities at high-energy region are weaker in the present overdoped sample. Detailed comparison of excitation spectra with the optimally-doped system is given.

1. Introduction

High-$T_c$ superconductivity is induced by either electron- or hole-doping into Mott insulators. Although Néel ordered phase disappears by carrier doping, spin fluctuations remains even in the superconducting (SC) phase[1, 2]. Therefore, spin correlations are widely believed to play a crucial role for the emergence of high-$T_c$ superconductivity. Extensive neutron-scattering experiment indeed clarified the intimate relation between the spin correlations and the superconductivity in the hole-doped ($p$-type) system[3]. In the $p$-type SC La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) and YBa$_2$Cu$_3$O$_{6+y}$, the spin excitation spectrum is known to persist up to high-energy region of $\sim$300 meV, which is comparable to the zone boundary energy of spin-wave excitation in non-doped mother compounds, and the spectral weight in the wide energy region drastically changes upon doping[4, 5, 6, 7]. Furthermore, the incommensurate spin fluctuations are observed below a characteristic energy of 40-50 meV and the incommensurability linearly increases with increasing $T_c$ in the underdoped region. These experimental results suggest a potential role of magnetism in the high-$T_c$ mechanism. However, the role of the spin dynamics for the pairing mechanism is not fully understood. Moreover, compare to the systematic study for $p$-type cuprate, there are quite few investigations of spin dynamics for the electron-doped ($n$-type) system, although the comparative study for two system is important for the unified understanding of superconductivity mediated by spin fluctuations.

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In this letter, we present results of recent high-energy neutron-scattering measurement performed on the overdoped n-type Pr$_{0.83}$LaCe$_{0.17}$CuO$_4$ (PLCCO, $T_c=16$ K), which shows commensurate low-energy spin fluctuations. We confirmed that the magnetic excitations extends up to at least high-energy region of $\sim$100 meV. With increasing energy transfer, the width of commensurate peak drastically broadens and the intensity rapidly decreases. These results are quantitatively same with those obtained for optimally-doped Pr$_{0.89}$LaCe$_{0.11}$CuO$_4$[8], while the broadening of peak-width against energy transfer is more marked and the intensities at high-energy region are much weaker in the present overdoped sample.

2. Sample preparation and neutron-scattering experiments

For the experiment, we have grown single crystals of PLCCO by a traveling-solvent floating-zone method. Obtained crystals were subsequently annealed under Ar gas flow at 940-960 °C for 10-12 hours, to induce the superconductivity. High energy ($\omega \lesssim$180 meV) neutron-scattering measurements were carried out on the time-of-flight spectrometers MAPS at the ISIS pulsed spallation neutron source, Rutherford Appleton Laboratory (RAL) in the U.K. Twelve crystals in the shape of columnar, 30-35 mm in length and 6 mm in diameter, were assembled so that the c-axis is parallel to the incident neutron beam and AF wavevector of $(\pi,\pi,0)$ is in the scattering plane. We selected several incident neutron energies $E_i$ between 50 and 200 meV. In addition, to measure the spin fluctuations below 12 meV more precisely, we have done further experiments using the thermal triple-axis spectrometers TOPAN installed in the reactor of JRR-3 at Japan Atomic Energy Agency, Japan.

![Figure 1. Constant energy spectra with the $\omega$ = (a) 6±2.5 meV, (b) 105±5 meV and (c) 150±10 for Pr$_{0.83}$LaCe$_{0.17}$CuO$_4$ measured using chopper spectrometer MAPS.](image1)

![Figure 2. $\omega$-dependence of resolution corrected peak-width $\kappa$ for Pr$_{0.83}$LaCe$_{0.17}$CuO$_4$ measured using MAPS (closed circles) and TOPAN (open circles) spectrometers. The results for Pr$_{0.89}$LaCe$_{0.11}$CuO$_4$ is drawn by dashed lines as a reference. Solid lines are guides to the eye.](image2)
3. Results and discussion

Figure 1 shows the representative constant-$\omega$ spectra of spin fluctuations measured at MAPS spectrometer. The magnetic spectra were obtained in the energy range between $\omega-\Delta\omega$ and $\omega+\Delta\omega$ after subtracting the constant background. In the low-energy region of 6±2.5 meV, the clear commensurate peak was observed at $h=1$, which corresponds to $(\pi, \pi)$ reciprocal position. This is consistent with the result obtained at the triple-axis spectrometer TOPAN with higher experimental accuracy. As seen in Fig. 1 (b), even at $\omega=105±5$ meV, the spin fluctuations centered at the commensurate position are confirmed, although the width of commensurate peak is much broader than that observed in the low-energy region. However, the scattering intensity at $\omega=150±10$ meV is nearly zero, meaning that the magnetic signal is indistinguishable from the background level. No well-defined magnetic signal at high-energy of $\omega\sim150$ meV is contrastive to the existence of clear commensurate signal in the optimally-doped Pr$\text{La}_{0.89}\text{Ce}_{0.11}\text{CuO}_4$, which is confirmed by the measurement performed under the similar experimental conditions. Therefore, the magnetic signal at high-energy region becomes weak by electron-doping.

In order to evaluate the $\omega$-dependence of peak-width and integrated intensity around $(\pi, \pi)$, the observed scattering intensity $S(\vec{Q}, \omega)$ was fitted to a following single Gaussian function,

$$S(\vec{Q}, \omega) = \frac{1}{1 - \exp(-\omega/k_B T)} \chi''(\vec{Q}, \omega)$$

$$\chi''(\vec{Q}, \omega) = \chi''(\omega) \sqrt{\frac{\ln(2)}{\pi \kappa^2}} \exp \left( -\ln(2) \frac{(\vec{Q} - \vec{Q}_\text{AF})^2}{\kappa^2} \right)$$

where $k_B$ is the Boltzmann constant, $\chi''(\omega)$ is the imaginary part of the dynamic susceptibility and $\kappa$ is the half width at half maximum of the observed peak. In Fig. 2, the evaluated $\omega$-dependence of resolution corrected peak-width $\kappa$ is plotted. The results obtained at triple-axis spectrometer TOPAN are also plotted by the open circles in the figure. We should note that the magnetic signal from the Cu spins could not evaluated at $\omega\sim90$ meV, due to the extrinsic background from the crystal field excitation of Pr ions. Furthermore, the huge intensity of phonon branches at $\omega\sim30$ meV prevents the precise determination of magnetic signal. We therefore excluded the data around these energies from the result of analysis. Although the data in insufficient, it safely can be said that the $\omega$-dependence of $\kappa$ shows drastic increases with increasing $\omega$, while it shows weak $\omega$-dependence in the high-energy region above $\sim100$ meV. The $\kappa$ above $\sim100$ meV is consistent with the value at $\omega\sim50$ meV. The $\chi''(\omega)$ as a function of $\omega$ is shown in Fig. 3. In the low-energy region below 12 meV, $\chi''(\omega)$ gradually decreases as $\omega$ increases. Although the spin excitation was confirmed to persist up to $\omega\sim140$ meV, the scattering intensity at high-energy region is quite weak and less that 20% of maximum value at $\omega\sim2$ meV.

We now compare the results for the overdoped sample and those previously obtained for the optimally-doped sample. In the present study, we observed the characteristic $\omega$-dependence of $\kappa$ showing different behavior separated by $\omega_c(\sim50$ meV). As shown by a dashed lines in Fig. 2, substantial peak broadening in the low-energy region and weak $\omega$-dependence of $\kappa$ in the high-energy are consistently seen in the optimally-doped sample. Therefore, existence of characteristic energy $\omega_c$, where the $\omega$-dependence of $\kappa$ changes, would be common feature in the spin excitation spectrum in the electron-doped superconductor. Compared with the $\omega$-dependence of $\kappa$ in the low-energy region for the optimally-doped sample, increase of $\kappa$ with $\omega$ is marked in the present overdoped sample, while the value at high-energy region in the two sample is approximately same, resulting into the reduction of $\omega_c$ in the heavily doped sample. On the other hand, scattering intensity in the all measured region is weaker in the overdoped sample. This means the reduction of magnetic intensity and the large reconstruction of intensity-distribution in $\omega$ with electron-doping. In contrast, in the hole-doped LSCO, the high-energy spin excitations
Figure 3. $\omega$-dependence of local spin susceptibility $\chi$ for Pr$_{0.83}$LaCe$_{0.17}$CuO$_4$ measured using MAPS (closed circles) and TOPAN (open circles) spectrometers. The results for Pr$_{0.88}$LaCe$_{0.11}$CuO$_4$ is drawn by dashed lines as a reference. The hatched area represents the region where the intensity from the Cu spin was not evaluated owing to contamination from the crystal field excitation of the Pr ions at $\sim$90 meV. Solid lines are guides to the eye.

persistently exist with sizable scattering intensity even in the heavily overdoped samples[9, 10]. Although the low-energy component of spin fluctuations are diminished in the overdoped region of LSCO with the disappearance of superconductivity, the high-energy spin fluctuations are robust against hole-doping. Therefore, the doping-dependence of spin fluctuations at high-energy region in the two systems is different and such a distinct doping evolutions suggest the existence of electron-hole asymmetry in the spin correlation is the wide energy scale. Further study is required to understand the magnetism in the doped Mott insulator and clarify its relevance to high-$T_c$ superconductivity.

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