

# Ce-substitution effects on the spin excitation spectra in $\text{Pr}_{1.4-x}\text{La}_{0.6}\text{Ce}_x\text{CuO}_{4+\delta}$

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**Abstract.** We performed inelastic neutron scattering measurements on as-grown single crystals of electron-doped  $\text{Pr}_{1.4-x}\text{La}_{0.6}\text{Ce}_x\text{CuO}_{4+\delta}$  with  $x = 0, 0.08$  and  $0.18$ . We succeeded in observing the spin excitation spectra up to  $300\text{meV}$  in the all samples. In both parent and electron-doped samples, a magnetic signal was observed at the magnetic zone center  $(0.5, 0.5)$  in the low-energy region below  $40\text{ meV}$ . In the high-energy region above  $100\text{ meV}$ , a double-peak structure reflecting the spin-wave excitation was detected in the constant-energy spectrum for the parent sample, while a single broad peak centered at  $(0.5, 0.5)$  was observed for the Ce-doped samples. These results suggest that the dispersion of spin excitation spectra become steeper upon doping. Furthermore, we newly found a drastic reduction of the magnetic intensity with Ce-substitution.

## 1. Introduction

The high-transition-temperature superconductivity in cuprate oxide emerges by the carrier-doping in antiferromagnetically ordered Mott insulators. To understand the mechanisms of the superconductivity, clarifying the electron-hole symmetry in physical properties is quite important. In the hole-doped system, the effect of carrier-doping on spin excitation has been extensively studied by neutron scattering measurements. An hourglass-shaped excitation, which consists of an inwardly dispersive low energy excitation and a spin wave like high energy excitation, is commonly observed in the superconducting phase of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  and  $\text{YBa}_2\text{Cu}_3\text{O}_{6+\delta}$  [1, 2]. Appearance of the hourglass excitation in the superconductivity phase suggests the connection between characteristic spin correlations and the superconductivity. In contrast to the study for hole-doped system, systematic experiments of spin excitation in electron-doped systems is limited due to the difficulties in preparing the high quality crystal. Although high-energy inelastic neutron scattering measurements have revealed a steeply standing magnetic dispersion along the energy direction and a weak dynamic spin susceptibility for the superconducting  $\text{Pr}_{1-x}\text{LaCe}_x\text{CuO}_4$  [3, 4], the origin of such characteristic feature is still unclear. It has been widely accepted that both the Ce substitution and the reduction annealing are essential for the emergence of superconductivity in the electron-doped cuprate. Therefore, the individual effect on the magnetism should be clarified. Our previous neutron and X-ray studies reported that the Ce-substitution effect is characterized by an elongation of spin excitation spectrum along the energy direction, which is quite different from the negligible doping effect on the high-energy spin excitation in the hole-doped system [5]. However, the data analysis was not done for the neutron scattering measurements. In this paper, we present the results of analysis, which shows the reduction of magnetic intensity as well as the elongation of magnetic excitation by Ce-substitution.



## 2. Experimental Details

The single crystals of  $\text{Pr}_{1.4-x}\text{La}_{0.6}\text{Ce}_x\text{CuO}_{4+\delta}$  ( $x = 0, 0.08$  and  $0.18$ ) were grown by a traveling-solvent floating-zone method. The set of single crystals are co-aligned with the mass of 41.3g ( $x = 0$ ), 42.6g ( $x = 0.08$ ) and 28.1g ( $x = 0.18$ ), respectively. Inelastic neutron scattering measurements were performed with Fermi chopper spectrometer 4SEASONS at Materials and Life Science experimental facility in J-PARC. The incident neutron energies of 357 meV, 88 meV and 39 meV were selected with the multi- $E_i$  method [6]. These experimental conditions are identical to those for previously reported measurements [7]. The scattering intensity was converted into the absolute value of  $S(\mathbf{Q}, \hbar\omega)$  using the vanadium standard. The differential cross section for the inelastic neutron scattering can be written as

$$\frac{d^2\sigma}{d\Omega dE} = \frac{2(\gamma r_e)^2 k_f}{\pi g^2 \mu_B^2 k_i} S(\mathbf{Q}, \hbar\omega) |F(\mathbf{Q})|^2, \quad (1)$$

where  $F(\mathbf{Q})$  is the magnetic form factor for Cu  $3d_{x^2-y^2}$  orbital.  $(\gamma r_e)^2 = 0.2905$  barn,  $g = 2$ , and  $\mu_B$  is Bohr magneton.  $k_i$  and  $k_f$  are incident and scattered neutron wave vectors, respectively.  $S(\mathbf{Q}, \hbar\omega)$  is a Fourier transform of the spin-spin correlation function, and is connected with the dynamic spin susceptibility  $\chi''(\mathbf{Q}, \hbar\omega)$  via the fluctuation-dissipation theorem.

$$S(\mathbf{Q}, \hbar\omega) = \chi''(\mathbf{Q}, \hbar\omega) \frac{1}{1 - e^{-\frac{\hbar\omega}{k_B T}}} \quad (2)$$

In this paper, we label the momentum transfer in the tetragonal notation.

## 3. Results and Discussion

Figure 1 represents the constant energy spectra of the spin excitations at  $\omega =$  (a) 40 meV, (b) 160 meV, (c) 200 meV and (d) 240 meV for  $x = 0, 0.08$  and  $0.18$ . The excitations were sliced along [100] direction through the antiferromagnetic zone center. In the figure, the spectra were shifted along the vertical direction for the clear visualization. To evaluate peak-position, peak-width and the

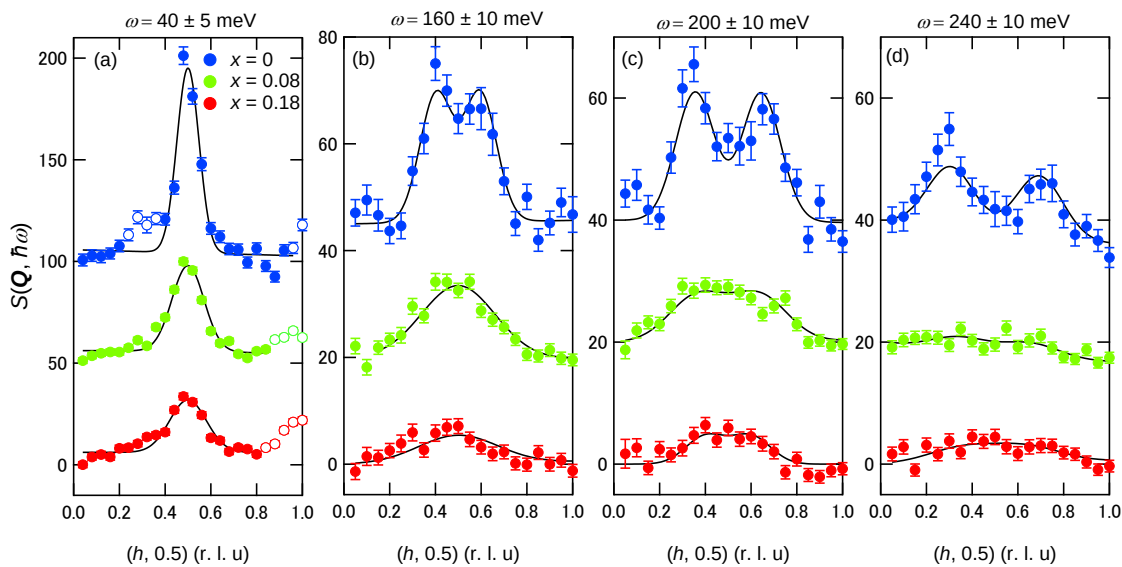


Fig. 1. Constant energy spectra at  $\omega =$  (a) 40 meV, (b) 160 meV, (c) 200 meV and (d) 240 meV for the parent and electron doped samples. Solid lines are results of fitting by Gaussian function with slope background. The open circles correspond to phonons and those data were removed from the fitting analysis.

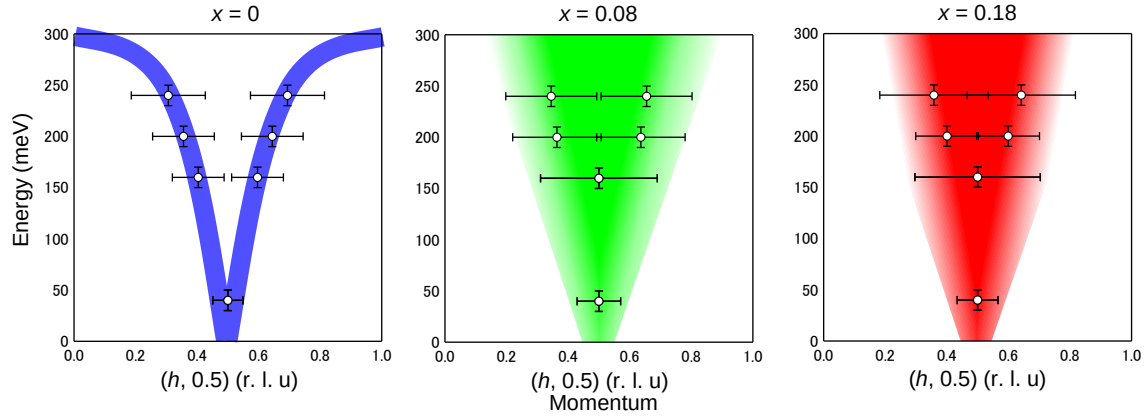


Fig. 2. Schematic pictures for the doping evolution of spin excitation spectra in as-grown  $\text{Pr}_{1.6-x}\text{La}_{0.6}\text{Ce}_x\text{CuO}_{4+\delta}$ . Open circles are the peak-positions evaluated from the fitting analysis. Vertical and horizontal bars represent energy width for the intensity-integration and the peak-width in half-width at half-minimum.

intensity, we carried out fitting analysis. With assuming the slop background, the spectra above 160 meV in the  $x = 0$  sample and 200 meV in  $x = 0.08$  and 0.18 sample were fitted by two Gaussian function and other spectra were fitted by using single Gaussian function. At  $\omega = 40$  meV, as seen in Fig. 1(a), a magnetic peak was observed at the antiferromagnetic zone center (0.5, 0.5) in all samples. Although the peak broadens with Ce-doping, their integrated intensity in the momentum space is comparable (see Fig. 3).

In the high-energy region at 160 meV, 200 meV and 240 meV, a double-peak structure was observed in the  $x = 0$  sample, and the distance between the two peaks becomes larger at higher energy transfer, reflecting the outwardly dispersive spin wave excitation as seen in the  $\text{La}_2\text{CuO}_4$ . In contrast, a broad single peak centered at (0.5, 0.5) was observed for the Ce-substituted samples at 160 meV. Even at higher-energy of 240 meV, no well-defined double-peak structure was detected. The intensity is rather gathered at the zone center, indicating that the excitation spectrum becomes steeper by Ce-substitution. Schematic picture for the evolution of spectra induced by Ce-substitution is shown in Fig. 2. This doping evolution is quite different from that in the hole-doped  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ , which shows weak doping dependence of high energy dispersion with remaining the considerable intensity [5]. Therefore, present results combined with those for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  indicates the doping asymmetry of spin excitation spectrum against the type of carrier. The elongation of the spectra toward the higher energy region was more clearly confirmed in the previous our resonant inelastic X-ray scattering study. [7] We newly found the drastic reduction of magnetic intensity in the high-energy region.

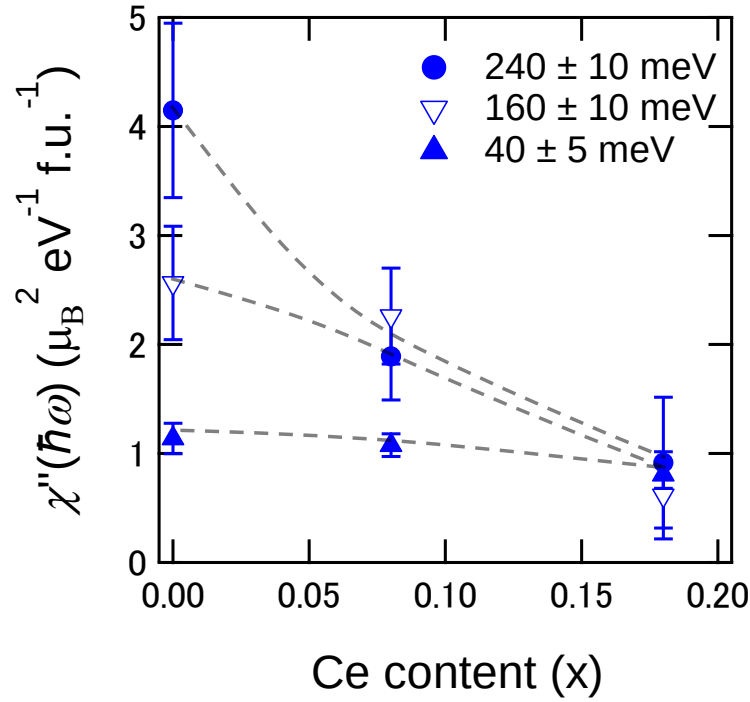


Fig. 3. Ce-substitution dependence of local spin susceptibility  $\chi''(\hbar\omega)$ .

In Fig. 3, Ce-substitution dependences of the local spin susceptibility  $\chi''(\hbar\omega)$  obtained from the integration of intensity in the antiferromagnetic Brillouin zone is summarized. The  $\chi''(\hbar\omega)$  at 40 meV exhibits the weak Ce-concentration dependence, while that at 240 meV firmly decreases upon Ce-substitution and the value for  $x = 0.18$  is less than half of that for  $x = 0$ . Therefore, the redistribution of intensity by Ce-substitution shows the energy-dependence. This behaviour is similar to the suppression of high energy inelastic neutron intensity reported for the superconducting  $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$ . [8] Stock and co-workers claimed that the intensity reduction in  $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$  is originated from the opening of pseudogap. As we mentioned above, the doping evolution of high-energy dispersion is different between the hole-doped and electron-doped systems. The boundary energy for PLCCO with  $x = 0.18$  exceeds 600 meV, which is approximately double of that for  $x = 0$ . Therefore, the intensity reduction in PLCCO is mainly attributed to the elongation of excitation spectrum, and the origin of intensity reduction between PLCCO and YBCO would be different.

#### 4. Summary

To study the Ce-substitution effects on the spin excitation, we performed inelastic neutron scattering measurements on the as-grown single crystals of  $\text{Pr}_{1.4-x}\text{La}_{0.6}\text{Ce}_x\text{CuO}_{4+\delta}$  ( $x = 0, 0.08$  and  $0.18$ ). We confirmed that the change of spectrum by Ce-substitution is characterized by the intensity reduction remarkably seen in the high energy region and the elongation of spectrum along the energy direction. These results are different from those in the hole-doped  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ , indicating the doping asymmetry of spin excitation spectrum against the type of carrier.

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