Magnetic Field Effect on the Static Antiferromagnetism of the Electron-Doped Superconductor $Pr_{1-x}LaCe_xCuO_4$ ($x = 0.11$ and 0.15)

M. Fujita,[1,*] M. Matsuda,[2] S. Katano,[2] and K. Yamada[1]

[1]*Institute for Materials Research, Tohoku University, Katahira, Sendai 980-8577, Japan*
[2]*Advanced Science Research Center, Japan Atomic Energy Research Institute, Tokai, Ibaraki 319-1195, Japan*

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Effects of magnetic fields (applied along the c axis) on static spin correlation were studied for the electron-doped superconductors $Pr_{1-x}LaCe_xCuO_4$ with $x = 0.11$ ($T_c = 25$ K) and $x = 0.15$ ($T_c = 16$ K) by neutron-scattering measurements. In the $x = 0.11$ sample, which is located near the antiferromagnetic (AF) and superconducting phase boundary, a commensurate magnetic order develops below around T_c at zero field. Upon applying a magnetic field up to 9 T both the magnetic intensity and the onset temperature of the order increase with the maximum field effect at ~5 T. In contrast, in the overdoped $x = 0.15$ sample any static AF order is neither observed at zero field nor induced by the field up to 8.5 T. Difference and similarity of the field effect between the hole- and electron-doped high-T_c cuprates are discussed.

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Magnetism in lamellar copper oxides is widely believed to play an important role in the mechanism of high-T_c superconductivity [1]. Extensive neutron-scattering measurements have indeed shown an intimate relation between incommensurate (IC) low-energy spin fluctuations in the hole-doped (p-type) $La_{2-x}Sr_xCuO_4$ (LSCO) and their superconductivity [2]. Recently, neutron-scattering study on the superconducting (SC) LSCO with $x \sim 1/8$ and excess-oxygen doped La_2CuO_{4+y} revealed an enhancement of the long-ranged IC magnetic order [3–6]. Lake *et al.*, furthermore, found field-induced slow spin fluctuations below a spin-gap energy, which shows a tendency toward the magnetic order, in the optimally doped LSCO [7]. These authors discussed that the antiferromagnetic (AF) insulator with IC correlations is a possible ground state after vanishing the superconductivity, as supported by theoretical studies [8–10]. Thus, in order to clarify the universal nature of magnetism hidden behind the superconductivity, it is necessary to investigate whether the AF order is commonly observed by suppressing the superconductivity.

Important challenges have been made on the prototypical electron-doped (n-type) system of $Nd_{2-x}Ce_xCuO_4$ (NCCO) which shows commensurate spin fluctuations at the tetragonal (1/2 1/2 0) reciprocal-lattice position in both AF and SC phases [11]. In $Nd_{1.86}Ce_{0.14}CuO_4$, at least down to 154 K, Matsuda *et al.* observed no field effect on the AF order which coexists or phase separates with the bulk superconductivity [12], whereas Kang *et al.* subsequently reported a field-enhanced huge magnetic intensity for $Nd_{1.85}Ce_{0.15}CuO_4$ and asserted the AF order as a competing ground state with the superconductivity, irrespective of carrier type [13]. This discrepancy could be originated from drastic (first-order-like) doping dependence of the static magnetism near the boundary between AF and SC phases [14,15]. However, since the bulk superconductivity with identical T_c of 26 K as well as the AF order appear in both samples, field effects on the AF order and the superconductivity are still controversial. Furthermore, an effect on Nd^{3+} spins is expected to be significant in NCCO, and therefore the results may conceal the inherent field dependence of Cu^{2+} spin correlations. Thus, a more comprehensive study on the n-type system is required using a sample with small rare-earth magnetic moments.

In this Letter, we report the result of neutron-scattering measurements performed under magnetic fields on the n-type $Pr_{1-x}LaCe_xCuO_4$ (PLCCO) with $x = 0.11$ ($T_c =$ 26 K) and 0.15 ($T_c = 16$ K), in which the effect of rare-earth moments is negligibly small compared with that in NCCO [16,17]. Samples with $x = 0.11$ and 0.15 are located in the vicinity of the phase boundary between AF and SC phases and in the overdoped SC phase, respectively [16,18]. At zero field, the former sample shows a short-ranged commensurate AF order, while no evidence of a magnetic order was observed in the latter sample. The intrinsic field effect on the AF order is, hence, expected to be clarified by the comparative study of these samples.

Single crystals of PLCCO with $x = 0.11$ and 0.15 were grown using a traveling-solvent floating-zone method. We annealed the as-grown single crystalline rods with ~30 mm in length and 6 mm in diameter under argon gas flow. The edge of the annealed crystals was cut into a plate to characterize the superconductivity by magnetic susceptibility measurements. As shown in Fig. 1, both $x = 0.11$ and 0.15 samples exhibit bulk superconductivity with T_c (onset) of 26 and 16 K, respectively.

Using the main part of the crystals, elastic neutron-scattering measurements were performed on the triple-axis spectrometer TAS-2 installed at the thermal neutron guide of JRR-3M of the Japan Atomic Energy Research Institute. We selected the incident neutron energy E_i of

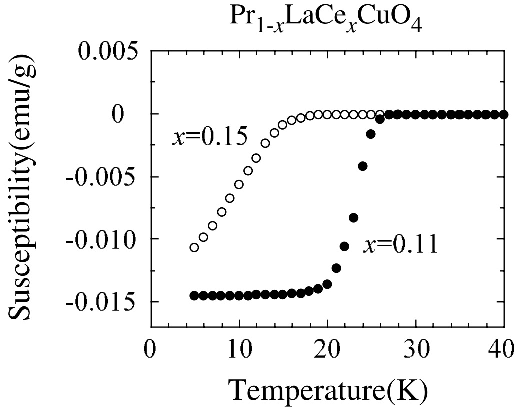

FIG. 1. Magnetic susceptibility measured for single crystals of $Pr_{0.89}LaCe_{0.11}CuO_4$ and $Pr_{0.85}LaCe_{0.15}CuO_4$ after the zero-field-cooling process.

14.7 meV with the collimation sequence of 17′-20′-sample-20′-80′ (or 17′-80′-sample-40′-80′) for the investigation of the peak profile with a high resolution (or the peak intensity against the temperature and the magnetic field). We mounted each single crystal with the CuO_2 sheets in the horizontal scattering plane and applied magnetic fields vertically to this scattering plane using a split-type cryocooled superconducting magnet. Crystallographic indexes are denoted as $(h\ k\ 0)$ in the tetragonal notation with the reciprocal-lattice unit (r.l.u.) at 3 K of 1.579 $Å^{-1}$, corresponding to the lattice constant of 3.979 Å along the Cu-O bonding. All measurements were done in the field cooling process after the field was set at above $T_c(B = 0)$ and $T_m(B)$.

The spatial static spin correlations were studied by scans along the $[1 - 1\ 0]$ direction through the AF zone center of (1.5 0.5 0) in the as-grown samples [19]. Figure 2 shows the scan profiles at 3 K after subtracting the background at high temperatures. Intensities for both samples are normalized by their volumes. At low temperatures in zero field, a weak intensity due to the magnetic order appears in the $x = 0.11$ sample at a commensurate (1.5 0.5 0) position. Assuming the same spin structure as that for Pr_2CuO_4, a magnetic moment of Cu averaged over the whole sample m_{Cu} was evaluated to be $(0.01–0.02)\mu_B$. We note that the magnetic intensity is superposed on the temperature-independent nonmagnetic superlattice peak, whose origin was discussed from the viewpoint of lattice distortion [20] and/or an impurity phase caused by heat treatments [21–23]. Measurements with the tighter collimation revealed that the Gaussian linewidth is broader than the resolution limited value indicated with the horizontal bar in Fig. 2(a). The intrinsic half width was evaluated to be 0.0049 ± 0.0008 $Å^{-1}$, corresponding to the inverse of the size of ordered region of 205 ± 32 Å in the CuO_2 planes. The peak width shows no remarkable field dependence, in contrast to the result for LSCO in which the long-ranged AF order is stabilized under the field [3–6].

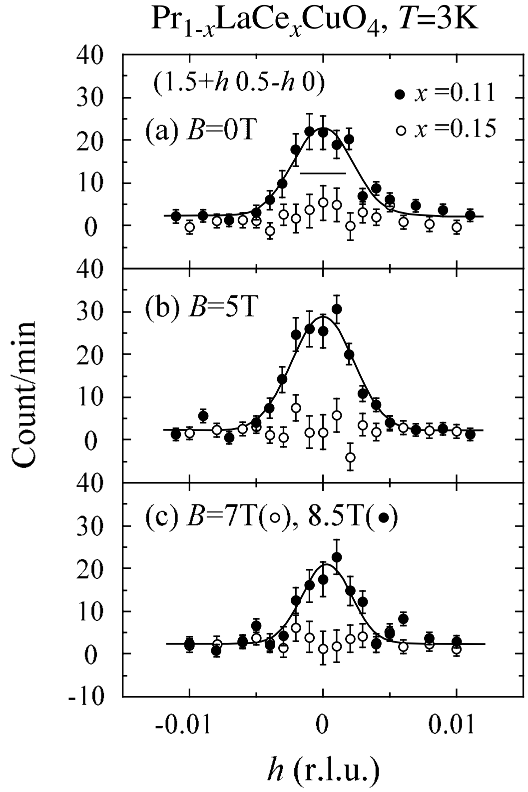

FIG. 2. Background-subtracted peak profiles for $Pr_{1-x}LaCe_xCuO_4$ with $x = 0.11$ (solid circles) and $x = 0.15$ (open circles) at 3 K under different magnetic fields at (a) 0 T, (b) 6 T, and (c) 8.5 T (7 T for $x = 0.15$). The horizontal bars indicate the instrumental Q resolution. The solid lines are results fitted with a single Gaussian function by convoluting the resolution.

The field-enhanced intensity shows a maximum at ~5 T, which is about 25% of the intensity at 0 T [see Fig. 5(a)]. With increasing field beyond 5 T, the intensity starts to decrease and at ~9 T becomes comparable with that at zero field. We note that Kang and co-workers observed similar field dependence of the magnetic intensity in $Nd_{1.85}Ce_{0.15}CuO_4$ [13]. However, the intensity is much stronger than that of PLCCO; therefore, a strikingly large effect of Nd spins should be considered in the field dependence of its magnetic intensity at (π, π). Furthermore, a field-induced complicated spin structure appears for $Nd_{1.85}Ce_{0.15}CuO_4$, while no evidence of such a structure was seen in $Pr_{0.89}LaCe_{0.11}CuO_4$. In the $x = 0.15$ sample, on the other hand, magnetic signal was not induced even under magnetic fields. The fact that field effect on the static spin correlations is absent in the overdoped sample provides important clues for understanding the field effect in n-type cuprates, as we discuss later.

We next investigated the temperature and field dependences of the peak intensity in the $x = 0.11$ sample. In

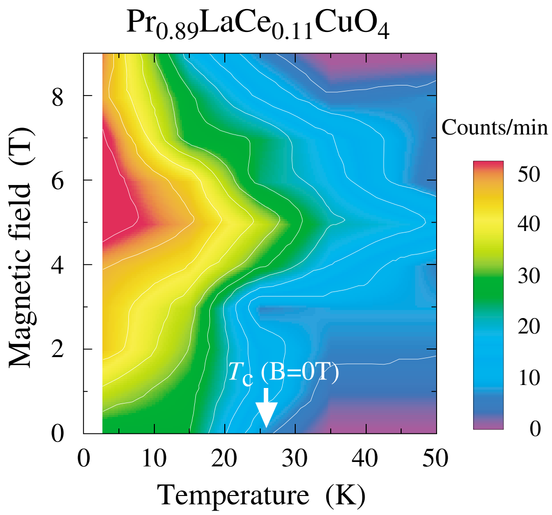

FIG. 3 (color). Magnetic peak intensity as a function of temperature and magnetic field in $Pr_{0.89}LaCe_{0.11}CuO_4$.

Fig. 3 the intensity contour is shown. At zero field, the magnetic intensity appears below $\sim T_c$ and lineally increases with decreasing temperature, which is similar to the case of the magnetic order in random systems [24,25] (see also Fig. 4). Interestingly, T_m increases up to ~60 K at 5 T and starts to decrease upon further increase of the field. More interestingly, as shown in Fig. 5, the field dependences of T_m and the peak intensity at 3 and ~35 K are quite similar. We note that the linear relation between the Néel temperature and the staggered moment and their large field dependence is often seen in the weak itinerant antiferromagnets and theoretically interpreted

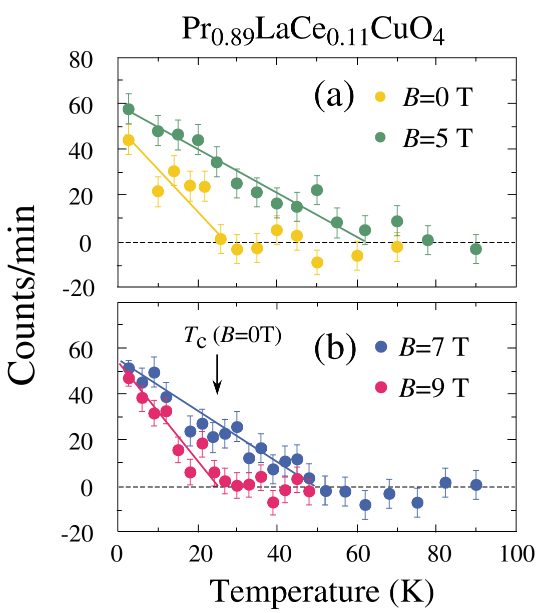

FIG. 4 (color). Temperature dependence of peak intensity at (1.5 0.5 0) reciprocal-lattice position measured at (a) $B = 0$ T and 5 T and (b) 7 T and 9 T for $Pr_{0.89}LaCe_{0.11}CuO_4$.

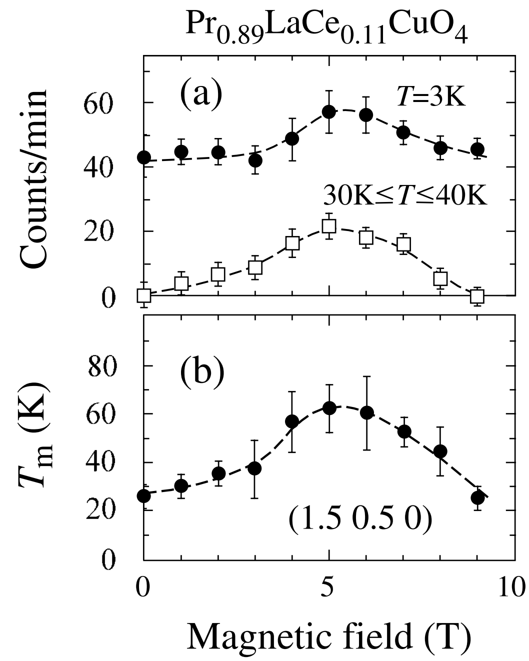

FIG. 5. Field dependence of (a) the peak intensity measured at (1.5 0.5 0) at 3 K (solid circles) and averaged intensity measured between 30 and 40 K (open squares), and (b) the magnetic ordering temperature for $Pr_{0.89}LaCe_{0.11}CuO_4$. Solid lines are guides to the eyes.

within the Fermi-liquid framework [26]. A recent nuclear magnetic resonance study [27] for optimally doped PLCCO also predicted the Fermi-liquid ground state. Therefore, the magnetism in the n-type cuprate near the AF-SC phase boundary can be regarded as the weak itinerant antiferromagnet. The shift of T_m by magnetic field may correspond to the that of quantum critical concentration x_c for the magnetic order which is located between $x = 0.11$ and 0.15 at zero field. If this shift occurs, the field-induced magnetic order is observable in the ground state at a specific concentration x'; $x' > x_c$ at zero field and $x' < x_c$ under field.

In the present study, the field effect was observed in the $x = 0.11$ sample analogous to the case of underdoped LSCO. The enhanced m_{Cu} at 3 K and 5 T, however, is quite small ($\sim 10^{-4}\mu_B$), compared with that of $\sim 10^{-1}\mu_B$ in LSCO with $x = 0.10$ measured at a comparable temperature as well as at a magnetic field. On the other hand, the critical field for the full suppression of superconductivity (B_{c2}) in PLCCO is less than 10 T [27], while B_{c2} in LSCO is ~45 T. Thus, the competitive coupling between AF order and superconductivity, which is theoretically discussed in terms of AF order induced around the vortex cores [8–10], is much weaker or absent in the n-type system. Furthermore, in contrast to the case of PLCCO, T_m in LSCO with $x = 0.10$ is independent of the field. These contrasts between the two systems, therefore, suggest different magnetic ground states in the optimally doped n-type cuprate and underdoped p-type La-214 in which the evidence of stripe correlations are observed [28,29].

Recently, Mang *et al.* proposed a model to describe the field effect on the magnetic order observed for NCCO [23]. According to their model, the field-induced intensities originate from the secondary phase of $(Nd, Ce)_2O_3$, which is epitaxially intercalated between the adjacent CuO_2 layers induced by heat treatment. The cancellation effect on the magnetic structure factor from the two Nd sites with distinct magnetic moments reproduces the field dependence of magnetic intensities. Here we discuss whether Mang's model is applicable to the field effect of PLCCO. As shown in Figs. 3 and 5(a), similar to the case of NCCO the magnetic intensity exhibits a peak as a function of field strength. However, the magnetic field at the maximum intensity is independent of temperature, in contrast to the peak position scaled by (H/T) in NCCO. Furthermore, in PLCCO, the evidence of secondary phase of $(Pr, Ce)_2O_3$ is seen in both the $x = 0.11$ and 0.15 samples. Nevertheless, the field-induced intensity is observed only in the $x = 0.11$ sample. Therefore, the field effect seen in PLCCO is difficult to be explained only by Mang's model.

Finally we discuss the possible scenario for understanding the field effects on the spin correlation in PLCCO. Systematic muon spin rotation measurements on the PLCCO systems revealed a decrease in the volume fraction of the magnetic order, staggered moment, and T_m upon electron doping near the AF-SC phase boundary [16]. This result suggests an existence of AF ordered islands segregated from the SC background in the $x = 0.11$ sample. The nonmagnetic $3d^{10}$ Cu sites introduced by doped electrons in Cu $3d$ orbitals may play some role in the formation of the AF islands [11,27]. In this situation, magnetic field affects both SC and AF ordered regions. In the present case, since the maximum value of T_m under magnetic field exceeds $T_c(B = 0\ \mathrm{T})$, we speculate that the observed field-enhanced magnetic order does not correspond to the vortex-induced one. The absence of field-induced AF order in the SC region may relate to an energy gap in magnetic excitation as seen in NCCO [11].

In conclusion, we have investigated the effect of magnetic field on the static spin correlations in the electron-doped superconductors $Pr_{1-x}LaCe_xCuO_4$ by neutron-scattering measurements. The field dependences of the magnetic intensity at low temperatures and the onset temperature of the commensurate AF order are similar in the $x = 0.11$ sample, showing a peak at $\sim$5 T. In contrast, any static AF order was not induced in the $x = 0.15$ sample under the fields up to 8.5 T. The present results suggest that the competitive coupling between the AF ordering and superconductivity is much weaker or absent in the n-type system.

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*Electronic address: fujita@imr.tohoku.ac.jp

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