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Evolution of spin excitations in electron-doped $Pr_{0.88}LaCe_{0.12}CuO_{4-\delta}$

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Abstract

We briefly review results of recent neutron scattering experiments designed to probe the evolution of antiferromagnetic (AF) order and spin dynamics in the electron-doped $Pr_{0.88}LaCe_{0.12}CuO_{4-\delta}$ as the system is tuned from its as-grown nonsuperconducting AF state into an optimally doped superconductor ($T_c = 24$ K) without static AF order. For under doped materials, a quasi-two-dimensional spin-density-wave was found to coexist with three-dimensional AF order and superconductivity. In addition, the low energy spin excitations follow Bose statistics. In the case of optimally doped material, we have discovered a magnetic resonance intimately related to superconductivity analogous to the resonance in hole-doped materials. On the other hand, the low energy spin excitations have very weak temperature dependence and do not follow Bose statistics, in sharp contrast to the as-grown nonsuperconducting materials.

Keywords: Electron-doped high-T_c superconductors; Magnetic excitations

1. Introduction

The high-transition-temperature superconductors are fundamentally composed of two-dimensional (2D) copper oxygen planes into which charge carriers, either holes or electrons are doped. Much effort in neutron scattering studies of superconducting copper oxides over the past twenty years has been devoted to hole-doped materials such as $La_{2-x}Sr_xCuO_4$ (LSCO) and $YBa_2Cu_3O_{6+x}$ (YBCO) [1,2]. A unifying picture of the magnetic excitations have now been achieved for these two mostly studied copper oxide materials [3,4]. At low energies, magnetic fluctuations are split away from the wave vector $\mathbf{Q} = (1/2, 1/2)$ in the two-dimensional reciprocal space of the CuO_2 planes, which corresponds to the antiferromagnetic (AF) Bragg position of the undoped compounds (Fig. 1a). With increasing energy, the incommensurate spin fluctuations disperse inward until they reach the commensurate wave vector $\mathbf{Q} = (1/2, 1/2)$, where upon further increases in energy transfer they then disperse back outward rotated from their original orientation. In YBCO, at energies where these incommensurate fluctuations disperse into a commensurate position, a significant enhancement in the spectral weight occurs below T_c, and this is known as the resonance mode excitation. The intensity of the resonance is strongly coupled to superconductivity and its energy scales approximately with $k_B T_c$ [5–8].

Although much is known about magnetic excitations in hole-doped materials, relatively little has been done in electron-doped copper oxides because the difficulty in growing large high-quality single crystals required for inelastic neutron scattering experiments. For prototypical electron-doped $Nd_{2-x}Ce_xCuO_4$ (NCCO) [9], previous neutron scattering experiments show a drastic suppression of the static three-dimensional (3D) AF order when superconductivity is established [10]. However, the static AF order persists even for NCCO with optimal bulk superconductivity ($T_c = 25$ K) [10]. Furthermore, Nd in NCCO has a magnetic ground state, which complicates the interpretation of Cu magnetism from NCCO [11–13]. For these reasons,

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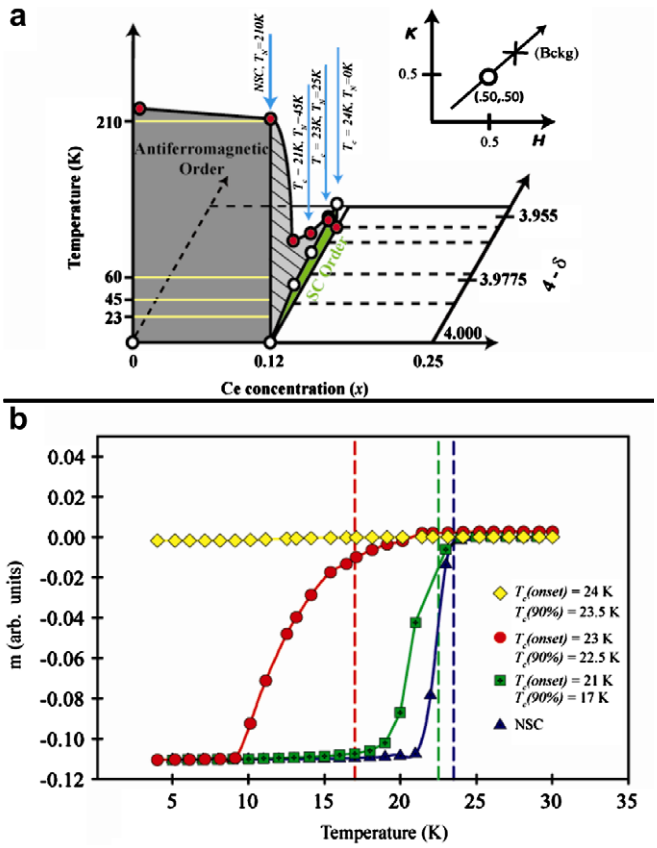

Fig. 1. Phase diagram for single layer electron- and hole-doped copper oxides PLCCO and magnetic susceptibility for PLCCO samples studies. (a) The inset shows the direction of **Q**-scans with the circle denoting (1/2, 1/2, 0) and the cross indicating where the offset background was measured. (b) Bulk susceptibility measurements showing the superconducting transitions for the PLCCO samples studies.

we have focused on another electron-doped material $Pr_{0.88}LaCe_{0.12}CuO_4$ (PLCCO), where Pr^{3+} has a singlet nonmagnetic ground state [14] and the optimally doped concentration is phase pure without static AF order to coexist with superconductivity [15–17]. In the following, we will discuss how spin excitations evolve as PLCCO is annealed from a nonsuperconducting (NSC) antiferromagnet to an optimally doped superconductor with $T_c = 24$ K.

2. Neutron scattering results on PLCCO

Compared to hole-doped materials, one of the unique features of electron-doped copper oxides is that as-grown, these materials are not superconducting. To make superconductivity in electron-doped materials requires annealing of the samples in a reduced oxygen atmosphere, which removes a tiny amount oxygen. Although the precise microscopic role of annealing process to the resulting superconductivity is still under debate [17], it is clear that the phase diagram of electron-doped materials is 3D where T_c can be controlled by either traversing the Ce concentration and/or tuning the oxygen content in the sample. As a first step, we have carried out neutron scattering experiments to establish the phase diagram of PLCCO as a function of annealing process. In our previous publications [16,17], we have shown that an elastic spin-density-wave (SDW) exists in the under doped PLCCO. We now discuss how low energy spin excitations evolve with increasing superconducting transition temperature through annealing. Fig. 1 shows the susceptibility of the four PLCCO samples used in our experiments. For the as-grown sample, we find that spin excitations are completely controlled by Bose statistics as expected for classical spin-wave excitations. Fig. 2 shows **Q**-scans for energy transfers $E = 0.5$, 1.5 and 4.0 meV at different temperatures. At $T = 1.4$ K, spin-wave excitations are gapped at $E = 0.5$ meV and the intensity of the scattering increases with increasing energy. The **Q**-widths of the scattering are entirely resolution-limited and can be well-fitted by Gaussians on linear backgrounds. This is consistent with the very large magnetic exchange ($J > 100$ meV) coupling in the as-grown materials [18]. To test whether the temperature dependence of the spin-wave excitations follow Bose statistics, we plot in Fig. 3 the **Q**-scan data as a function of temperature. For energies above the spin-gap, the spin-wave excitations increase in intensity

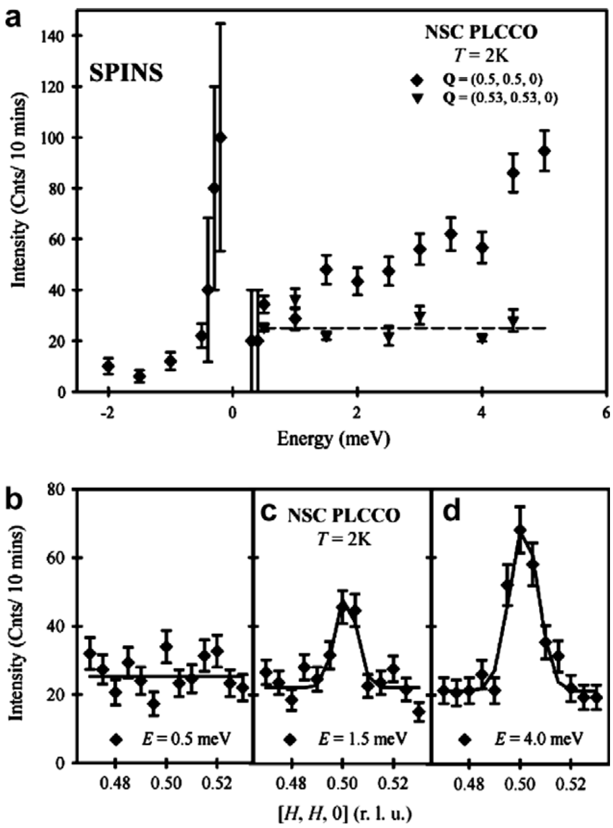

Fig. 2. Low energy excitations in NSC PLCCO. (a) Constant-**Q** scans at signal [$\mathbf{Q} = (0.5, 0.5, 0)$] and background [$\mathbf{Q} = (0.53, 0.53, 0)$] positions at 2 K. The dashed line is a linear fit to the energy dependence of the nonmagnetic background. (b)–(d) Constant energy scans along the $[H, H, 0]$ direction at $E = 0.5$, 1.5, and 4.0 meV. Solid lines are Gaussian fits on linear background described in the text.

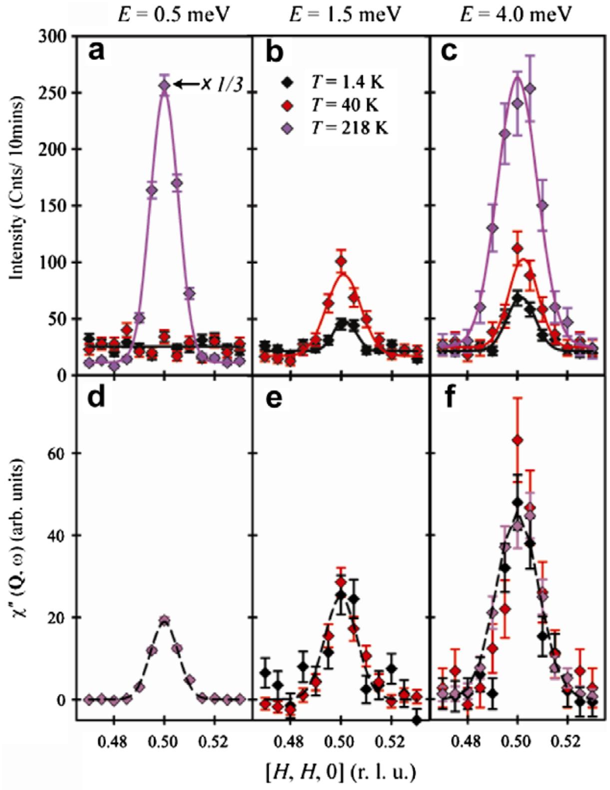

Fig. 3. Temperature dependence of the low energy excitations in NSC PLCCO. (a)–(c) Raw **Q**-scans around (0.5,0.5,0) at different temperatures at $E = 0.5$, 1.5, and 4.0 meV. A clear un-isotropy spin gap at 0.5 meV persists above 40 K until the 3D order breaks down above T_N. (d)–(f) Measured dynamic susceptibility at different temperatures for these three energy transfers. Besides the data at 0.5 meV, it is clear that susceptibility follows the standard Bose statistics.

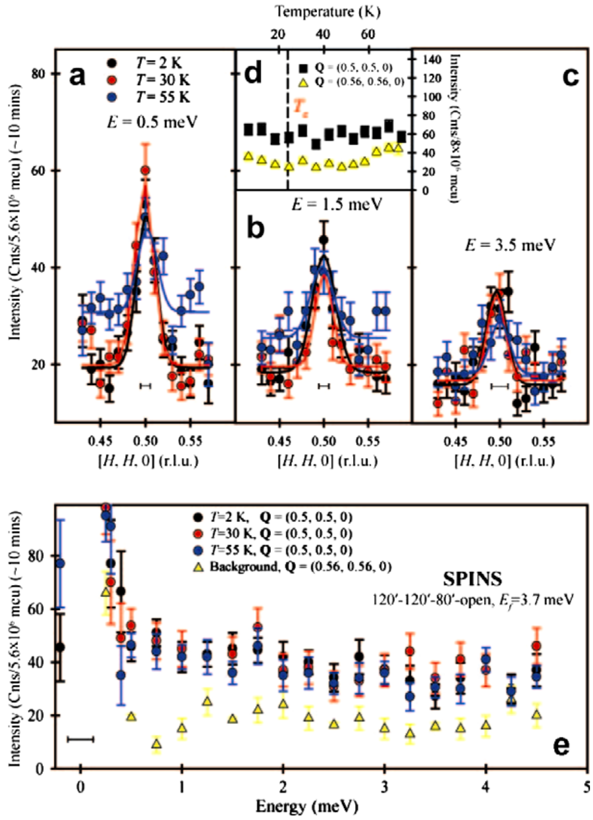

Fig. 4. Temperature dependence of the low energy excitations in superconducting PLCCO using the same spectrometer as Figs. 2 and 3. (a)–(c) Raw **Q**-scans around (0.5,0.5,0) at different temperatures at $E = 0.5$, 1.5, and 3.5 meV. (d) Temperature-dependent scattering at $Q = (1/2, 1/2, 0)$ and (0.56,0.56,0) for $E = 1.5$ meV. The background is independent of temperature below ~50 K and the magnetic signal becomes weaker at 80 K. (e) Constant-Q scans obtained on SPINS spectrometer with collimations as shown, at the ridge of magnetic scattering at 2, 30 and 55 K, compared with the temperature independent background scattering for temperatures between 2 K and 30 K.

with increasing temperature following the Bose population factor. Removing the nonmagnetic background contributions to the scattering, we find that the imaginary part of the dynamic susceptibility, $\chi''(\mathbf{Q},\omega)$, becomes temperature independent (Fig. 3d–f). This is a direct indication that spin-wave excitations in as-grown PLCCO follow Bose statistics.

Now turning to superconducting PLCCO, one can tune T_c by simply annealing the samples at different temperatures. Because of the space limitation, we will only discuss magnetic excitation results for optimally doped PLCCO ($T_c = 24$ K). Fig. 4 shows similar scans as that of Fig. 2 for this material [19]. In remarkable contrast to Fig. 2, the scattering at $E = 0.5$, 1.5 and 4.0 meV is essentially temperature independent for temperature from 2 K to 30 K but decreases slightly at 55 K owing to the increased background scattering. Compared to the resolution-limited **Q**-widths in as-grown PLCCO (Fig. 2), the scattering centered around $\mathbf{Q} = (1/2, 1/2)$ is considerably broader. Fourier transforms of the Gaussian peaks give dynamic spin correlation lengths of $\xi \sim 96$, 80, and 94 Å. The temperature dependence of the scattering at $\mathbf{Q} = (1/2, 1/2)$ confirms the results of the **Q**-scans. To directly compare the results of the superconducting PLCCO with that of the as-grown material, we plot in Fig. 5 the temperature dependence of the scattering together with the extracted $\chi''(\mathbf{Q},\omega)$. It is immediately clear that $\chi''(\mathbf{Q},\omega)$ behaves quite differently from that of the as-grown sample.

Recently, we have discovered that optimally doped PLCCO has a resonance, analogous to that of the hole-doped materials [19]. Similar to hole-doped materials, the resonance in electron-doped PLCCO increases in intensity below T_c and its energy scales with $5.8k_BT_c$ forming a universal plot for all superconducting copper oxides irrespective of electron or hole doping [19]. However, in contrast to hole-doped materials, magnetic scattering below the resonance in electron-doped PLCCO form commensurate and gapless scattering (Figs. 4 and 5). Therefore the hour-glass shaped dispersion in the magnetic scattering of hole-doped superconducting materials may not be a universal feature of all copper oxides. Instead, the resonance itself appears to be a fundamental property of the superconducting copper oxides.

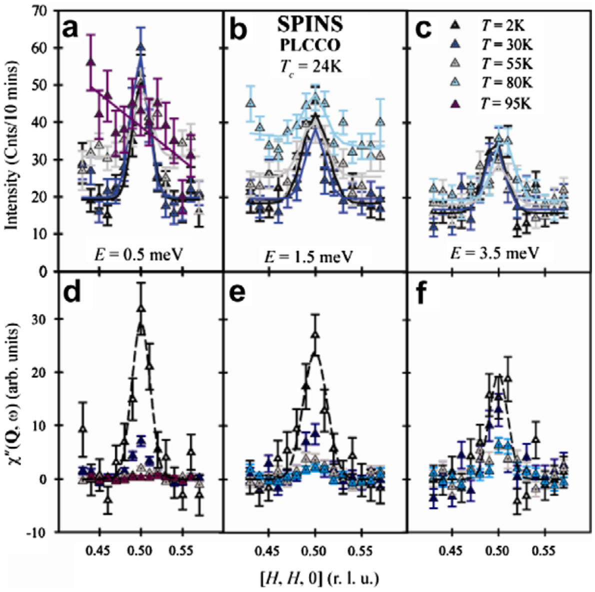

Fig. 5. Temperature dependence of the low energy excitations in superconducting PLCCO. (a)–(c) Raw **Q**-scans around (0.5, 0.5, 0) at different temperatures at $E = 0.5$, 1.5, and 3.5 meV. (d)–(f) Temperature dependence of the dynamic susceptibility at these energies are clearly different from those in Fig. 3 for NSC PLCCO.

3. Conclusions

The discovery of the resonance in electron-doped PLCCO suggests that resonance may play an essential role in the mechanism of high-T_c superconductivity. While the intensity of the resonance in hole-doped materials appears to arise from opening of a spin-gap in low energy part of the spin excitations spectrum [3–8], the gapless and weakly temperature-dependent low energy magnetic scattering in optimally electron-doped PLCCO is puzzling. At present, it is not clear where the resonance draws its spectral weight below the superconducting transition temperature. On the other hand, the weakly temperature dependent behavior observed in the low-energy spin excitations of PLCCO remarkably resembles the quantum critical scattering seen in heavy fermion compounds [20–22]. To determine the microscopic origin of the resonance and its correlation with high-T_c superconductivity, one must now systematically study the doping, magnetic field, and energy dependence of the mode for different electron-doped materials.

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