Resonance in Optimally Electron-Doped Superconductor Nd$_{1.85}$Ce$_{0.15}$CuO$_{4-\delta}$

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We use inelastic neutron scattering to probe magnetic excitations of an optimally electron-doped superconductor Nd$_{1.85}$Ce$_{0.15}$CuO$_{4-\delta}$ above and below its superconducting transition temperature $T_c = 25$ K. In addition to gradually opening a spin pseudo gap at the antiferromagnetic ordering wavevector $Q = (1/2,1/2,0)$, the effect of superconductivity is to form a resonance centered also at $Q = (1/2,1/2,0)$ but at energies above the spin pseudo gap. The intensity of the resonance develops like a superconducting order parameter, similar to those for hole-doped superconductors and electron-doped Pr$_{0.88}$LaCe$_{0.12}$CuO$_4$. The resonance is therefore a general phenomenon of cuprate superconductors, and must be fundamental to the mechanism of high-$T_c$ superconductivity.

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In conventional Bardeen-Cooper-Schrieffer (BCS) superconductors, the superconducting phase forms when electrons are bound into pairs with long-range phase coherence through interactions mediated by lattice vibrations (phonons). Since high-transition-temperature (high-$T_c$) superconductivity arises in copper oxides when sufficient holes or electrons are doped into the CuO$_2$ planes of their insulating antiferromagnetic (AF) parent compounds, it is important to determine if spin fluctuations play a fundamental role in the mechanism of high-$T_c$ superconductivity. For hole-doped superconductors, it is now well documented that the spin fluctuations spectrum forms an ‘hourglass’ dispersion with the most prominent feature, a collective excitation known as the resonance mode, centered at the AF ordering wavevector $Q = (1/2,1/2)$ in YBa$_2$Cu$_3$O$_{6+x}$ (YBCO). The intensity of the mode tracks $T_c$ and its intensity behaves like an order parameter below $T_c$ for materials such as YBa$_2$Cu$_3$O$_{6+x}$ (YBCO) and La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) displays negligible changes across $T_c$. Instead, the effect of superconductivity in optimally hole-doped LSCO is to open a spin gap and pile density of states along incommensurate wavevectors at energies above the spin gap, and thus appears to be different from YBCO.

If the resonance is fundamental to the mechanism of superconductivity, it should be ubiquitous to all high-$T_c$ superconductors. Although the superconductivity-induced enhancement at incommensurate wavevectors in LSCO has been argued to be comparable to the commensurate resonance in YBCO, the intensity gain of the resonance below $T_c$ may not always be compensated by opening of a spin gap and spectral weight loss at lower energies. For example, the resonance intensity gain in the electron-doped Pr$_{0.88}$LaCe$_{0.12}$CuO$_4$ (PLCCO, $T_c = 24$ K) below $T_c$ is not compensated by spectral weight loss at lower energies. On the other hand, while neutron scattering measurements found a low-temperature spin gap (about 4 meV) in the electron-doped superconductor Nd$_{1.85}$Ce$_{0.15}$CuO$_4$ (NCCO), there have been no report of the resonance or spectral weight gain at energies above the spin gap below $T_c$. Therefore, the relationship between the superconducting spin gap and the resonance is still an open question.

In this Letter, we report the results of inelastic neutron scattering studies of temperature dependence of the spin fluctuations in an optimally electron-doped NCCO ($T_c = 25$ K) below $T_c$. We confirm the presence of a low-temperature spin (pseudo) gap and show that the effect of superconductivity also induces a resonance at energies similar to electron-doped PLCCO. Our results thus demonstrate that the resonance is an ubiquitous feature of optimally electron-doped superconductors. Its intensity gain below $T_c$ in NCCO is due in part to the opening of a spin pseudo gap and spectral weight loss at low energies. This is remarkably similar to the optimally hole-doped LSCO, and thus suggesting that the enhancement at incommensurate wavevectors below $T_c$ in LSCO has the same microscopic origin as the commensurate resonance in other high-$T_c$ superconductors.

We grew a high quality (mosaicity $< 1^\circ$, 3.5 grams) NCCO single crystal using a mirror image furnace. Figure 1a plots the magnetic susceptibility measurements showing an onset $T_c$ of 25 K with a transition width of 3 K. Our neutron scattering experiments were performed on the IN-8 thermal triple-axis spectrometer at the Institute Laue Langevin, Grenoble, France. We define the wave vector $Q$ at $(q_x,q_y,q_z)$ as $(h,k,l) = (q_xa/2\pi,q_ya/2\pi,q_za/2\pi)$ reciprocal lattice units (r.l.u) in the tetragonal unit cell of NCCO (space group $I4/mmm$, $a = 3.95$, and $c = 12.07$ Å). For the experiment, the...
NCCO sample is mounted in the $[h,k,0]$ zone inside a cryostat. We chose a focusing Si(111) as monochromator and PG(002) as analyzer without collimation. The final neutron energy was fixed at $E_f = 14.7$ meV with a pyrolytic graphite (PG) filter in front of the analyzer. This setup resulted an energy resolution of about 1 meV in full-width-half-maximum (FWHM) at $Q = (-0.5,0.5,0)$. To understand the effect of superconductivity on the Cu$^{2+}$ spin fluctuations, we must first determine the temperature dependence of the magnetic excitations from Nd$^{3+}$ crystal electric field (CEF) levels in NCCO. For Nd ions in the tetragonal NCCO crystal structure, the three lowest energy CEF magnetic excitations are at $h\omega = 12.2 \pm 0.3$ meV, $20.3 \pm 0.1$ meV, and $26.5 \pm 0.3$ meV. Our energy scans at $Q = (-0.5,1.5,0)$ confirm these results and show that the intensities of these CEF levels have small temperature dependence between 2 K and 30 K (Figure 1c).

Figure 2 summarizes the transverse and longitudinal $Q$-scans around $(-0.5,0.5,0)$ at different energy transfers and temperatures. Consistent with earlier results on NCCO and PLCCO, the scattering is commensurate and centered at $Q = (-0.5,0.5,0)$ for all energies probed. Figures 2a-d show the raw data (with scan directions marked) below and above $T_c$ at $h\omega = 2.5$, 8 meV. At $T = 30$ K ($T_c + 5$ K), the magnetic scattering above the linear backgrounds decreases slightly with increasing energy from 2.5 meV to 8 meV (Figs. 2e and 2f). On cooling to below $T_c$, the peak intensity is drastically suppressed for $h\omega = 2.5$ meV (Figs. 2a and 2b), and it increases for $h\omega = 8$ meV (Figs. 2c and 2d). Figures 2e-g show background subtracted transverse scans at various energies. It is immediately clear that cooling below $T_c$ suppresses the Q = (-0.5,0.5,0) peak at $h\omega = 2.5$ meV but enhances scattering at $h\omega = 8$ and 10 meV. On the other hand, magnetic scattering at $h\omega = 36$ meV changes negligibly from 2 K to 50 K (Fig. 2h).

Figures 3a and 3b show energy scans at the signal $Q = (-0.5,0.5,0)$ and background $Q = (-0.34,0.66,0)$ positions above and below $T_c$. Although the large Nd$^{3+}$ CEF level dominated the magnetic scattering at $h\omega = 12$ meV, one can still see clear Cu$^{2+}$ spin fluctuations centered at $(-0.5,0.5,0)$ for energies between 2 and 10 meV. In the normal state, the magnetic scattering decreases with increasing energy, consistent with $Q$-scans.
FIG. 3: a) The temperature dependence of the scattering at the peak \([Q = (-0.5, 0.5, 0)]\) and background \([Q = (-0.34, 0.66, 0)]\) positions below and above \(T_c\). Note the intensity is plotted in log-scale to display the large intensity difference between the \(\text{Nd}^{3+}\) CEF level at \(\bar{\hbar}\omega = 12\) meV and \(\text{Cu}^{2+}\) spin fluctuations centered at \(Q = (-0.5, 0.5, 0)\) for energies between 2 and 10 meV. b) Background subtracted magnetic scattering at \(Q = (-0.5, 0.5, 0)\) below and above \(T_c\). The data are cross checked by constant-energy scans in Fig. 2. c) The temperature difference plot showing the resonance at \(E_r = 9.5\pm2\) meV. The large error is due to the uncertainty in obtaining \(\text{Cu}^{2+}\) magnetic signal above 10 meV.

at \(\hbar\omega = 2.5, 8,\) and 10 meV (Figs. 2c-g). In the superconducting state, the low-energy spin fluctuations at \(Q = (-0.5, 0.5, 0)\) are suppressed for \(\hbar\omega \leq 4\) meV and there is a clear scattering intensity gain for \(6 \leq \hbar\omega \leq 10\) meV. The contrast between the normal and superconducting states becomes more obvious when changes in background scattering are taken into account (Fig. 3b). The large \(\text{Nd}^{3+}\) CEF scattering between 10 < \(\hbar\omega < 33\) meV (Fig. 1c) overwhelmed \(\text{Cu}^{2+}\) magnetism. The background corrected difference plot between the superconducting and normal states shows a resonance at \(\hbar\omega = 9.5\pm2\) meV, similar to that for PLCCO [16].

To determine if the low temperature spin fluctuations’ suppression below 4 meV and enhancement between 6 to 10 meV are indeed associated with the opening of a superconducting gap below \(T_c\) as in the tunneling experiments [22], we carefully measured the temperature dependent scattering at the peak \([Q = (-0.5, 0.5, 0)]\) and...
background positions for $h\omega = 2.5$ and 8 meV. From previous low-energy inelastic neutron scattering work on NCCO [18], we know that the spin gap in NCCO opens gradually with decreasing temperature until it reaches to about 4 meV at 2 K. While peak intensity in the Q-scans at $h\omega = 2.5$ meV show a clear low temperature suppression, there is still a peak present at $Q = (-0.5, 0.5, 0)$ even at 2 K. Therefore, optimally electron-doped NCCO does not have a clean spin gap as in the case of the optimally hole-doped LSCO [10]. The temperature dependence of the scattering at the peak and background positions (Figs. 4a and 4b) reveals that the intensity suppression at $h\omega = 2.5$ meV does not happen at $T_c$ but at 9 K ($T_c = 16$ K). While this result confirms the earlier report [18], it also suggests that the gradual opening of the (pseudo) spin gap is not directly related the temperature dependence of the superconducting gap which is BCS-like [22] and becomes essentially fully opened with $2\Delta \approx 7$ meV below 12 K (50% of $T_c$).

On the other hand, the temperature dependence of the scattering at $h\omega = 8$ meV is clearly coupled to the occurrence of superconductivity. With increasing temperature, the scattering at $Q = (-0.5, 0.5, 0)$ first decreases like an order parameter, showing a kink at $T_c$, and then increases again above 30 K. It turns out that the large intensity rise above 30 K at $h\omega = 8$ meV is due to the CEF transition from 12 meV to 20 meV as the 12 meV state is being populated with increasing temperature (Fig. 4c) [20]. As the CEF levels are weakly Q-dependent, the large intensity increase above 30 K is also seen in the background (Fig. 4c). The difference between signal and background shows a clear order-parameter-like temperature dependence of the resonance, remarkably similar to the temperature dependence of the resonance in PLCCO [16] and hole-doped superconductors [4, 13, 14, 16, 18, 19].

The discovery of the resonance in another class of electron-doped superconductors suggests that the mode is a general phenomenon of electron-doped superconductors independent of their differences in rare-earth substitutions [13]. For hole-doped LSCO [10, 11, 12, 13, 14], the intensity enhancement in spin susceptibility above the spin-gap energy has been characterized as the magnetic coherence effect [11]. The variation of the susceptibility enhancement at energies ($6 \leq h\omega \leq 13$ meV) just above the spin pseudo gap energy of 4 meV in NCCO is consistent with this picture, although the temperature dependence of the spin pseudo gap in NCCO behaves rather differently from those in LSCO [10, 18]. In our search for the excitations responsible for electron pairing and high-$T_c$ superconductivity, one of the arguments against the relevance of the resonance has been the inability to observe superconductivity-induced commensurate resonance in LSCO [11, 12, 13, 14]. If the resonance is a phenomenon associated with the opening of a superconducting gap and the subsequent local susceptibility enhancement, it is natural to regard the susceptibility gain in both NCCO and LSCO as the resonance. Adding these two points to the universal $E_c = 5.8k_BT_c$ plot in Fig. 1b suggests that while the resonance energy itself is intimately related to $T_c$, other details such as the spin gap, commensurability, and hourglass dispersion found in different materials may not be fundamental to the superconductivity.

For hole-doped superconductors, the hourglass dispersion has been interpreted either as the signature of “stripes” where doped holes are phase separated from the Mott-like AF background [23, 24, 25], or as a bound state (spin exciton) within the gap formed in the non-interacting particle-hole continuum of a Fermi-liquid [26, 27]. Although the resonance in PLCCO has been interpreted as an over damped spin exciton [28], it remains a challenge to understand how the resonance can arise both from NCCO which has a spin pseudo gap and from the gapless PLCCO [29].

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