Fincher-Burke spin excitations and $\omega/T$ scaling in insulating La$_{1.95}$Sr$_{0.05}$CuO$_4$

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Insulating La$_{1.95}$Sr$_{0.05}$CuO$_4$ shares with superconducting cuprates the same Fincher-Burke spin excitations, which usually are observed in itinerant antiferromagnets. The local spectral function satisfies $\omega/T$ scaling above $\sim 16$ K for this incommensurate insulating cuprate. Together with previous results in commensurate insulating and incommensurate superconducting cuprates, these results further support the general scaling prediction for square-lattice quantum spin $S = 1/2$ systems. The width of incommensurate peaks in La$_{1.95}$Sr$_{0.05}$CuO$_4$ scales to a similar finite value as at optimal doping, strongly suggesting that they are similarly distant from a quantum critical point. They might both be limited to a finite correlation length by partial spin-glass freezing.

It is well known that the $(\pi, \pi)$ peak of the Néel antiferromagnetic order of the parent compounds is replaced by a quartet of incommensurate peaks when cuprates are sufficiently doped to have become superconducting. The very recent excitement is that spin excitations measured in superconducting La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) ($x = 0.10$ and $0.16$), YBa$_2$Cu$_3$O$_{6.6}$ (YBCO)$_2$, as well as in the “stripe-ordered” La$_{1.875}$Ba$_{0.125}$CuO$_4$ with a suppressed $T_C$ share a common spectral feature: broad excitation continua, originating from the incommensurate quartet, disperse towards the $(\pi, \pi)$ point with increasing energy at the rate of the spin-wave velocity of the parent compounds. The spectrum is distinct from the spin-waves in two important ways: 1) the excitations are not resolution-limited; 2) there are no the outward branches. Such a spin excitation spectrum previously has been observed in itinerant spin-density-wave antiferromagnets such as elemental metal Cu and strongly correlated metal V$_{2-x}$O$_x$, and the single-lobed dispersive continuum is referred as the Fincher-Burke mode. A self-consistent theory has been developed by Moriya and others to describe the mode, and quantitative agreement has been achieved for three-dimensional itinerant antiferromagnets. Iterant theories have also been developed to account for the Fincher-Burke-like modes in superconducting cuprates.

Sandwiched between the parent antiferromagnetic insulator and the high-$T_C$ superconductor in the phase diagram of LSCO is a distinct doping regime from $x = 0.02$ to $0.055$. Cuprates in the regime are insulators without the long-range Néel order, and a spin-glass transition occurs at $T_f \lesssim 10$ K. This doping range is often referred to as the spin-glass phase, although the spin-glass phase extends to both lower and higher dopings in the Néel and superconducting states. Magnetic correlations were extensively investigated and regarded as being incommensurate, as in the parent compound. However, with improved single-crystal samples, magnetic correlations show a novel incommensurate doublet, which also differs distinctly from the quartet in the superconductors. In this paper, we report that spin excitations of La$_{1.95}$Sr$_{0.05}$CuO$_4$ in the insulating spin-glass regime are also composed of the Fincher-Burke modes, originating from the incommensurate doublet, with a velocity the same as in superconducting LSCO. Although cuprates in the spin-glass regime are insulators, they are not the usual band insulators, and part of the Fermi surface may have survived. It would be interesting to investigate whether the Fincher-Burke modes reported here can be accounted for by extending theories for similar spin excitations of superconducting cuprates.

Meanwhile, for quasi-two-dimensional (2D) spin $S=1/2$ cuprates, the temperature range where spin fluctuations are investigated is within $T \ll J/k_B \sim 1000$ K, where classical statistical mechanics has to be replaced by quantum statistical mechanics. The general scaling argument of 2D quantum statistical systems leads to a prediction for samples which are not exactly at a quantum critical point (QCP) that for $T \ll J/k_B$ but above a low-temperature limit $T_X$, the energy scale for 2D spin fluctuations at long wavelengths is $k_B T_X^{15,16}$. Hence, for $T_X < T < J/k_B$, the spin excitation spectrum follows the $\omega/T$ scaling, which is not a robust feature of the Moriya theory. For $T < T_X$, a constant energy gap is predicted. We test these predictions against the distinct incommensurate spin excitations from the doublet in La$_{1.95}$Sr$_{0.05}$CuO$_4$. We also compare our results with previous investigations of incommensurate spin fluctuations from the quartet in superconducting cuprate and commensurate ones at the $(\pi, \pi)$ point in insulating cuprates. Our data support the scaling above $T_X$ but no gap is observed below $T_X$. Surprisingly, scaling analysis of the $q$ and $\omega$ dependent spin excitation spectra indicates that La$_{1.95}$Sr$_{0.05}$CuO$_4$ and optimally doped LSCO are similarly distant from the QCP.
A single piece of La$_{1.95}$Sr$_{0.05}$CuO$_4$ crystal of 5.2 g was used in this work. It was grown using a traveling-solvent floating-zone method as described previously. We use the orthorhombic Cmca unit cell ($a = 5.338\,\text{Å}$, $b = 13.16\,\text{Å}$, $c = 5.404\,\text{Å}$ at 1.5 K) to describe the $q$-space for measurements at NIST using the cold neutron triple-axis spectrometer SPINS. The sample temperature was controlled by a pumped He cryostat. The horizontal collimations before and after the sample were both 80', and a Be filter cooled by liquid nitrogen was used after the sample to reduce higher order neutrons passing through the pyrolytic graphite (002) used for both monochromator and analyzer. The intensity of magnetic neutron scattering was counted against a flux monitor placed before the sample in a fixed $E_F = 5\,\text{meV}$ configuration and normalized to yield $S(q, \omega)$ in absolute units. Such a cold neutron spectrometer readily resolves the incommensurate doublet near (100), i.e., the $(\pi, \pi)$ point (see Fig. 1), while it is difficult to resolve the doublet using a thermal neutron triple-axis spectrometer due to its coarser resolution. Hence, we will present only the cold neutron scattering results here.

We first present the nominal elastic signal at various temperatures. Fig. 1(a) shows constant-energy $\hbar \omega = 0$ scans through the incommensurate doublet from 1.2 to 80 K. The sharp peak at (100) is due to higher-order diffraction of (200). Its width indicates the instrument resolution. The magnetic doublets at $q_{\perp} = (1.0, \pm 0.058(2))$ are obviously broader than the resolution. The deconvoluted peak width yields the in-plane correlation length for the nominally elastic spin correlations, $\xi_\perp = 34(2)\,\text{Å}$ at 1.2 K, about 9 nearest-neighbor Cu spacings. With increasing temperature, the doublet monotonically decreases in intensity without appreciable change in either the peak width or position. Above $\sim 20\,\text{K}$, the doublet disappears, consistent with previous studies.

At finite energies, however, the temperature dependence of the doublet is entirely different from that at $\hbar \omega = 0$. Fig. 1(b) shows constant-energy $\hbar \omega = 0.5\,\text{meV}$ scans measured in the same temperature range. Instead of monotonically decreasing, the intensity first increases, reaches a maximum between 15 and 20 K, and then decreases with further rising temperature. The intensity at the peak shoulder, e.g., at $q = (1.0, 0.2)$, in Fig. 1(a)-(b) measures a temperature-independent background, which has been subtracted in Fig. 1(c). Note that at 1.2 K, $S(q, \omega)$ at 0.5 meV is more than one order of magnitude weaker than at $\hbar \omega = 0$. This reflects the fact that the energy spectrum of $S(q, \omega)$ shows a prominent sharp “central peak” at $\omega = 0$ at low temperatures, see Fig. 1(c). The “central peak” is energy-resolution-limited at the SPINS spectrometer, with the full-width-at-half-maximum (FWHM) of 0.3 meV. However, the nominal elastic signal from La$_{1.95}$Sr$_{0.05}$CuO$_4$ is not truly static at $T > T_g \approx 5\,\text{K}$ as determined by our $\mu$SR measurements, which has an energy resolution of $\sim 10^{-6}\,\text{meV}$. Details of the $\mu$SR study will be published elsewhere. Similar “central peak” phenomenon has been reported for Li-doped La$_2$CuO$_4$ and YBa$_2$Cu$_3$O$_{6+x}$, and the very slow spin dynamics is attributed to a partial spin-glass transition.

What is the energy dependence of the doublet? Fig. 2(a) shows scans at various energies at 20 K. Below 3 meV, the two incommensurate peaks are clearly distinguishable. As energy increases, the doublet merges into a flat-top peak. The scans in Fig. 2(a) can be fitted using two gaussians of the same width. The peak positions are shown as the black circles in Fig. 2(b). The dispersion is consistent with the inner branches of spin-waves (solid lines). The same dispersion rate has been reported for superconducting La$_{2-x}$Sr$_x$CuO$_4$ ($x = 0.10$ and 0.16). The shaded area in Fig. 2(b) covers the FWHM, which grows slowly with energy from 0.089(3) Å$^{-1}$ at 0.5 meV.

![FIG. 1: (color online) The generalized spin correlation function $S(q, \omega)$ as a function of $q$ along the c-axis measured at (a) $\hbar \omega = 0$ and (b) 0.5 meV, respectively, at various temperatures. (c) $S(q, \omega)$ as a function of energy at $q = (1.0, 0.05)$ and at 1.2 and 20 K, respectively. The “central peak” at 1.2 K is also shown on a 1/3 scale with open circles. Background has been subtracted in (c).](image-url)
mensurate spin correlations with the very different dynamic magnetic susceptibility relates to of our improved sample. The imaginary part of the local liquid theory. The arctan function is stipulated by the marginal Fermi liquid theory, and \( \Gamma \) the spin relaxation constant. In previous studies, the local staggered static magnetic susceptibility, \( \chi_S(S,T) \) using cuprate samples showing commensurate spin correlations. It is re-examined with the very different incommensurate spin correlations of our improved sample. The imaginary part of the local dynamic magnetic susceptibility relates to \( S(\omega) \) by the fluctuation-dissipation theorem

\[
\chi''(\omega) = \pi (1 - e^{-\hbar \omega/k_B T}) S(\omega).
\]  

(1)

Fig. 2 shows \( \chi''(\omega) \) from 1.2 to 80 K, which is well described by the Debye relaxor model

\[
\chi''(\omega) = \frac{\chi_0(\hbar \omega/T)}{1 + (\hbar \omega/T)^2}.
\]  

(2)

where \( \chi_0 \) is the local staggered static magnetic susceptibility, and \( \Gamma \) the spin relaxation constant. In previous studies, the local \( \chi''(\omega) \) was modeled by

\[
\chi''(\omega) = I(\omega) \arctan[a_1(\hbar \omega/k_B T) + a_2(\hbar \omega/k_B T)^2].
\]  

(3)

The arctan function is stipulated by the marginal Fermi liquid theory, but \( I(\omega) \) has no determined analytic form. Hence, we opt for the well-known Debye relaxor model, Eq. (2), to fit our data. The Debye relaxor model has also successfully described measured \( \chi''(\omega) \) of insulating \( \text{La}_{2} \text{CuO}_{4} \), which has commensurate magnetic correlations.

Now we turn to examination of scaling behavior of spin excitations. Historically, it was done in the spin-glass regime through the local spin correlation function \( S(\omega) = \int dqS(q,\omega) \) using cuprate samples showing commensurate spin correlations. It is re-examined with the very different incommensurate spin correlations of our improved sample. The imaginary part of the local dynamic magnetic susceptibility relates to \( S(\omega) \) by the fluctuation-dissipation theorem

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On the base plane of Fig. 3, the spin relaxation constant \( \Gamma \) obtained from the least-squares fit is shown as a function of temperature. The good instrument resolution, 0.3 meV (FWHM), has a negligible effect during fitting. One interesting result is that \( \Gamma = 0.73(2) \) for temperatures above \( T_X \approx 16 \) K. Hence, when \( \chi''(\omega) \), normalized by its maximum \( \chi_0/2 \) at \( \hbar \omega = \Gamma \), is plotted as a function of \( \hbar \omega/k_B T \), Eq. (2) dictates that all data collected above \( T_X \) collapse onto a single universal curve

\[
y = 2/[1 + (x/0.73)^2]
\]  

and Fig. 2(a) bears this out. The result is commonly referred to as the \( \omega/T \) scaling, and \( \Gamma/k_B T = O(1) \) is a hallmark of quantum magnetic theory. For \( T < T_X \), Fig. 3 shows that \( \Gamma \) departs from the proportionality to temperature. Consequently, the low temperature data would not follow the scaling curve, as demonstrated by Fig. 3(b). Note that the spectral function Eq. (2) does not become gapped below \( T_X \), contrary to non-random quantum theory, but can be explained by dopant scattering.

The \( \omega/T \) scaling and its departure below \( T_X \) shown in Fig. 3 for \( \text{La}_{1.95} \text{Sr}_{0.05} \text{CuO}_{4} \) bears a striking similarity to what reported for \( \text{La}_{2} \text{CuO}_{4} \). The two cuprates have similar hole concentration and develop spin-glass at similar \( T_g \). However, they differ in several important ways: i) The dopants are out of the \( \text{CuO}_2 \) plane in the Sr compound, but directly replace \( \text{Cu}^{2+} \) in the Li compound. ii) The former becomes a superconductor with additional 0.5% more holes, but the latter always remains an insulator. iii) Magnetic correlations are incommensurate in the former, but commensurate in the latter. iv) The in-plane correlation length \( \xi_0 \approx 34(2) \) Å for the glassy spin component in the former, but \( \xi_0 \approx 274 \) Å in the latter, and the \( \kappa(\omega,T) \) shown in Fig. 2(c) is more than double that in the latter.
\[ \frac{\Gamma}{k_B T} = 0.73 \text{ for the former, and 0.18 for the latter.} \]

In spite of these differences, \( \Gamma \) saturates at \( \Gamma_0 \approx 1 \text{ meV} \) and \( \chi''(\omega) \) becomes essentially \( T \)-independent (see Fig. 3) for both cuprates below \( T_X \). As a consequence, Eq. (1) requires a reduced \( S(\omega) \) at low energies when the temperature decreases below \( T_X \), as observed in Fig. 1(b) and (c), in sharp contrast to a magnet at the QCP.

Scaling of spin excitations has also been examined near the optimal doping for \( \text{La}_{1.86}\text{Sr}_{0.14}\text{CuO}_4 \). The material is concluded to be near a QCP, namely, \( T_X = 0 \), with some caveats. The \( \chi''(\omega) \) in the \( T \rightarrow 0 \) limit, equaling to \( \chi_0 / \Gamma \) of Eq. (2), would saturate below \( T_X \), but \( T_C = 35 \text{ K} \) sets the upper limit for measurable \( T_X \) in \( \text{La}_{1.86}\text{Sr}_{0.14}\text{CuO}_4 \). Hence it cannot be determined whether \( \text{La}_{1.86}\text{Sr}_{0.14}\text{CuO}_4 \) or \( \text{La}_{1.95}\text{Sr}_{0.05}\text{CuO}_4 \) is closer to a QCP with a lower \( T_X \).

Another method to assess the distance from the QCP is to examine the width of constant-\( \hbar \omega \) scans, see Fig. 2(d). Adapting the ansatz in [22], the \( \kappa(\omega, T) \) plotted as a function of a reduced temperature better collapses our data in Fig. 2(d), and the solid line is

\[ \kappa(\omega, T) = \kappa_0^2 + (k_B T/c)^2(1 + (\hbar \omega/0.73 k_B T)^2), \]

where \( \kappa_0 = 0.044(1) \text{ A}^{-1} \) and \( c = 4(1) \times 10^2 \text{ meV}\AA \). At the QCP, \( \kappa_0 \) is expected to be zero, and its value for \( \text{La}_{1.95}\text{Sr}_{0.05}\text{CuO}_4 \) is comparable to \( \kappa_0 \) in the superconducting state, but narrower than \( \kappa_0 \) at 40 K in the normal state for \( \text{La}_{1.84}\text{Sr}_{0.16}\text{CuO}_4 \). Hence, \( \text{La}_{1.95}\text{Sr}_{0.05}\text{CuO}_4 \) and the optimally doped LSCO with a short \( 1/\kappa_0 \), about 6 Cu-Cu spacings, seem equally distant from the QCP.

In conclusion, the Fincher-Burke modes, the broad and dispersive spin excitations of itinerant antiferromagnets, are observed in the spin-glass regime of \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \). Theoretical understanding of similar excitation modes in superconducting cuprates now has an added task in the insulators. Befitting to the generality of its theoretical argument, the \( \omega/T \) scaling is shown to be valid for a new type of cuprates above \( T_X \). Spin excitations below \( T_X \) remain gapless contrary to the prediction of non-random quantum theory. The spin-glass transition at finite doping introduces an extra component of slow spin fluctuations. It would be interesting to explore whether the glassy state, limiting the correlation length of the rest of spins, is responsible for the equal distance from the QCP for \( \text{La}_{1.95}\text{Sr}_{0.05}\text{CuO}_4 \) and \( \text{La}_{1.84}\text{Sr}_{0.16}\text{CuO}_4 \).

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