Novel dynamic scaling regime in hole-doped La$_2$CuO$_4$

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Only 3\% hole doping by Li is sufficient to suppress the long-range 3-dimensional (3D) antiferromagnetic order in La$_2$CuO$_4$. The spin dynamics of such a 2D spin liquid state at $T \ll J$ was investigated with measurements of the dynamic magnetic structure factor $S(\omega, q)$, using cold neutron spectroscopy, for single crystalline La$_{2-\delta}$Cu$_{0.94}$Li$_{0.06}$O$_4$. $S(\omega, q)$ peaks sharply at $(\pi, \pi)$ and crosses over around 50 K from $\omega/T$ scaling to a novel low temperature regime characterized by a constant energy scale. The possible connection to a crossover from the quantum critical to the quantum disordered regime of the 2D antiferromagnetic spin liquid is discussed.

The experimental investigation of the spin dynamics of doped Heisenberg antiferromagnets on a square lattice is essential to understanding cuprate superconductors, as well as to current research on quantum phase transitions\[1\]. For the structurally simplest laminar cuprate, La$_2$CuO$_4$, the 3-dimensional (3D) antiferromagnetic Néel order due to weak interlayer magnetic interactions can be suppressed by 2-3\% hole doping using Sr, Ba or Li, thus allowing experimental investigation of the quantum spin dynamics of a 2-dimensional (2D) spin liquid in a wide temperature range, $0 < T < J/k_B \sim 1000$ K\[2\].

At the critical doping concentration, $y_c$, of La$_2$CuO$_4$ and at $T = 0$, namely at the quantum critical point, spin dynamics is described by classical critical dynamics in 2+1 dimensions\[2\]. At a finite temperature, the extra dimension in imaginary time is reduced to a finite thickness of $\hbar c/k_BT$, where $c$ is the spin wave velocity. As a consequence of this long wavelength cutoff, spin dynamics follows the quantum critical $\hbar \omega/k_BT$ scaling\[3\]. There has been experimentally observed in Ba and Sr doped La$_2$CuO$_4$ in previous inelastic neutron scattering studies\[4, 5\]. At $T = 0$ and above the critical doping concentration $y > y_c$, zero-point quantum fluctuations set another long wavelength cutoff. Therefore, for $y > y_c$, when the thermal cutoff length, $\hbar c/k_BT$, reaches the quantum cutoff during cooling, a crossover in spin dynamics from the quantum critical (QC) regime, where the energy scale is $k_BT$, to the quantum disordered (QD) regime, where the energy scale is fixed at infrared by the quantum cutoff, occurs\[2, 3\]. This crossover, however, has not been explicitly investigated experimentally. Here, we report a cold neutron inelastic scattering study on spin dynamics in Li doped La$_2$CuO$_4$ from 150 K to 1.5 K. A crossover from the $\omega/T$ scaling to a new low temperature regime, which features an infrared energy cutoff, is directly observed.

The critical doping concentration, $y_c$, for Li doped La$_2$CuO$_4$ is 3\%\[5\]. In this study, a $y=6\%$ single crystal, La$_{2-\delta}$Cu$_{0.94}$Li$_{0.06}$O$_4$, weighing 2.1 g, was grown in CuO flux, using isotopically enriched $^7$Li (98.4\%) to reduce neutron absorption of natural Li. The crystal has orthorhombic $C_{mca}$ symmetry with lattice parameters $a = 5.351 \AA$, $b = 13.15 \AA$ and $c = 5.386 \AA$ at 295 K. Using this orthorhombic unit cell to label reciprocal space, the $(\pi, \pi)$ point in the square lattice notation splits to (100) and (001) points. Neutron scattering signal is observed at (100) type but not at (001) type Bragg peaks, as in the stoichiometric antiferromagnet La$_2$CuO$_4$\[7\]. Measurements of seven independent (100) type Bragg peaks, using thermal neutrons to reach 6 $\AA^{-1}$, confirm the magnetic origin of these peaks. This conclusion is consistent with previous thermal neutron scattering studies on Li doped La$_2$CuO$_4$,\[8\], which, however, did not have sufficient energy resolution to resolve the energy spectra at low temperatures. Here, we focus on spin dynamics near $q=(100)$, taking advantage of the enhanced energy resolution of the cold neutron triple-axis spectrometer SPINS at NIST with fixed $E_f = 3.7$ meV or $E_f = 5$ meV. The (002) reflection of pyrolytic graphite was used for both the monochromator and analyzer. A cold Be or BeO filter was put before the analyzer to eliminate higher order neutrons. Horizontal Soller slits of 80’ were used before and after the sample. The temperature of the sample was regulated by a pumped He cryostat.

Fig. 1 shows some constant-energy ($\hbar \omega$) scans (a) in the basal plane and (b) perpendicular to the basal plane around $q=(100)$ at various energies and temperatures. The sharp peak at $(\pi, \pi)$ in Fig. 1(a) does not show appreciable change in its width over a wide energy and temperature range, suggesting a resolution-limited in-plane peak. Flat scans in Fig. 1(b) reaffirm the 2-dimensional character of the spin dynamics in our sample\[11\]. An extensive search along the (100) and (101) directions in the basal plane yielded no incommensurate peaks, such as those found in Sr doped La$_2$CuO$_4$\[12, 13\]. This means that the spin liquid in La$_{2-\delta}$Cu$_{0.94}$Li$_{0.06}$O$_4$ is composed of simple chess-board type dynamic antiferromagnetic spin clusters in the CuO$_2$ plane, which have grown to substantial size below 100 K with the correlation length, $\xi \gg 42$ A, the inverse of the half-width-at-half-maximum of the in-plane peak. The correlation length is much
The fact that there is little change in measured $q$ by the fluctuation-dissipation theorem [22], related to the dynamic magnetic structure factor $S(q,\omega)$. Background in (b), triangles, was measured at 1.5 K either at $-0.5$ meV, or at $(1.2,k,0)$ with $\omega=0.3$ and 1.2 meV.

longer than the mean distance between Li dopants, 15 Å.

The commensurate spatial magnetic structure in Li doped $\text{La}_2\text{CuO}_4$, which is simpler than incommensurate structures observed in Sr doped $\text{La}_2\text{CuO}_4$ [12, 14], may be understood in light of the mobility of doped holes [8, 15]. Vortices associated with the holes, which are a long-range and effective topological disturbance to the 3D Néel order, preserve the commensurate ($\pi,\pi$) magnetic correlations [16, 17].

The incommensurate magnetic structures, on the other hand, through either the “stripe” [18] or the nesting Fermi surface [19] mechanism, require more mobile holes such as those in Sr doped $\text{La}_2\text{CuO}_4$ [17]. The effect of hole mobility on spatial magnetic correlations has also been demonstrated in numerical simulation [20].

After delimiting the spatial part of spin dynamics in $\text{La}_2\text{CuO}_4\text{Li}_{0.06}\text{O}_4$, we now turn to the temporal dependence of the 2D ($\pi,\pi$)-correlated spin liquid. Fig. 2 shows energy scans at $q=(100)=(\pi,\pi)$ at various temperatures. Since magnetic intensity is sharply confined in $q$-space in a rod passing through (100), as shown in Fig. 1, the energy scan at (1.39,0,0) which is far away from the rod (triangles in Fig. 2), offers a good measure of background. In addition to the flat background, neutron scattering intensity in Fig. 2 consists of a convolution of the instrument resolution function with the dynamic magnetic structure factor $S(\omega, q)$ plus the elastic and incoherent peak at $\omega=0$ [21]. The imaginary part of the generalized magnetic susceptibility $\chi''(\omega, q)$ is related to the dynamic magnetic structure factor $S(\omega, q)$ by the fluctuation-dissipation theorem [22].

$$\chi''(\omega, q) = \pi \left(1 - e^{-\hbar\omega/k_BT}\right) S(\omega, q).$$

The fact that there is little change in measured $q$ widths in our energy and temperature ranges (Fig. 1 indicates that magnetic intensity in Fig. 2 is proportional to the local dynamic magnetic structure factor, $I(\omega)$, to Eqs. 2-6). Triangles represent a flat background measured at (1.39,0,0).

FIG. 1: Const.-$q=(100)$ scans with various energy transfers and at various temperatures (a) in the basal CuO$_2$ plane and (b) perpendicular to the basal plane. The $q=(100)$ here in the orthorhombic notation corresponds to the ($\pi,\pi$) point of the CuO$_2$ square plane. Background in (b), triangles, was measured at 1.5 K either at $-0.5$ meV, or at $(1.2,k,0)$ with $\omega=0.3$ and 1.2 meV.

FIG. 2: Const.-$q=(100)$ scans from 1.5 to 150 K. The shaded area represents least-squares fit of inelastic magnetic intensity, $I(\omega)$, to Eqs. 2-5. Triangles represent a flat background measured at (1.39,0,0).
In this regime, from Eqs. (2), (5) and (6) and noting the constant magnetic behavior. Below the crossover temperature, La,

the spin liquid. Fig. 3(c) shows the normalized scaling scheme describes the low temperature regime of

with the energy scale $\Gamma_0 \approx 1$ meV. The solid line is the scaling function, Eq. (6).

thermal energy $k_B T$ is similar to that in the insulating phase of Ba doped La$_2$CuO$_4$.[3]

Below 50 K, however, the $\omega/T$ scaling breaks down in La$_2$Cu$_{0.94}$Li$_{0.06}$O$_4$, as clearly shown in Fig. 3(b). Data taken at 1.5, 6 and 21 K no longer fall on the solid $\omega/T$ scaling curve, Eq. (4), valid above 50 K. Instead, a new scaling scheme describes the low temperature regime of the spin liquid. Fig. 3(c) shows the normalized $\chi''(\omega)$ as a function of energy for data taken at low temperatures. All data are described by

$$\frac{\chi''(\omega)}{\chi'_\pi} = g(h\omega/\Gamma_0)$$

with the energy scale $\Gamma_0 \approx 1$ meV and the scaling function

$$g(x) = \frac{x}{1 + x^2}.$$  

(6)

Notice also that $\chi_\pi$ is basically constant below 50 K (Fig. 4), therefore, $\chi''(\omega)$ is essentially independent of temperature in the new low temperature regime.

The crossover from the high temperature QC regime to the new low temperature regime also manifests itself in low-energy magnetic neutron scattering intensity. At high temperatures, from Eqs. 2, 3 and 4,

$$I(\omega \to 0) \propto \chi_{\pi} \sim T^{-1}.$$  

(7)

Therefore, magnetic intensity at the low energy limit increases during cooling as the local magnetic susceptibility, $\chi'(0) = \pi \chi_\pi/2$, does, consistent with common paramagnetic behavior. Below the crossover temperature, from Eqs. 2, 3 and 4 and noting the constant $\chi_{\pi}$ in this regime,

$$I(\omega \to 0) \propto T,$$  

(8)

which is markedly different from that in the QC regime, Eq. 4. Circles in Fig. 4 show $I(\omega)$ measured at $q=100\bar{h}$ and $h\omega = 0.2$ meV, $I(\omega \sim 0)$. The diamonds and squares were measured during cooling and warming, respectively.

FIG. 3: (a) $\omega/T$ scaling is valid for La$_2$Cu$_{0.94}$Li$_{0.06}$O$_4$ in the high temperature QC regime. The solid line is the scaling function, Eq. 4. (b) $\omega/T$ scaling becomes invalid in the low temperature regime. (c) A new scaling for the low temperature regime, with a constant energy scale $\Gamma_0 \approx 1$ meV. The solid line is the scaling function, Eq. (6).

FIG. 4: (left scale) Temperature dependence of inverse local magnetic susceptibility $\chi_{\pi}^{-1}$ (circles) for La$_2$Cu$_{0.94}$Li$_{0.06}$O$_4$, see Eq. 8 and 9. (right scale) Temperature dependence of magnetic intensity (open symbols) measured at $h\omega = 0.2$ meV, $I(\omega \sim 0)$. The diamonds and squares were measured during cooling and warming, respectively.

In Sr or Li doped La$_2$CuO$_4$, a spin glass transition occurs in a wide doping range at a similar temperature, $T_{sf} \sim 10 K$.[23, 24, 25]. In La$_2$Cu$_{0.94}$Li$_{0.06}$O$_4$, $T_{sf} = 8 K$.[25]. This raises the possibility that the departure from the $\omega/T$ scaling at low temperature might be caused by the spin glass transition. However, the key spectral signature of a spin glass transition is a zero energy scale as the time scale becomes infinite.[24]. Instead of approaching zero, the energy scale of La$_2$Cu$_{0.94}$Li$_{0.06}$O$_4$ saturates at a finite energy, $\Gamma \approx 1$ meV, at the crossover. Therefore, the crossover around 50 K is not related to the spin glass transition.

A second possibility is to identify the low temperature regime with the quantum disordered regime of the 2D Heisenberg antiferromagnet, which is characterized by a constant energy scale. As discussed in the introduction, the finite energy scale is determined by the quantum
long wavelength cutoff, which depends on the distance from the quantum critical point, \( y - y_c \). Physically, the distance can be tuned by microscopic mechanisms, such as frustrating magnetic interactions, vortex-like topological magnetic defects associated with holes, or other mechanisms, such as frustrating magnetic interactions.

In real materials, there are additional low temperature phase transitions, which are caused by effects ignored in theoretical models of the 2D quantum antiferromagnet. These transitions include (1) a finite temperature Néel transition due to weak interlayer magnetic interaction, (2) a spin glass transition due to disorder unavoidable with doping, (3) “stripe” formation due to the interaction between antiferromagnetic order and mobile holes, and (4) superconducting transition. Nevertheless, 2D quantum antiferromagnetism can be accessed in the temperature window between \( J/k_B \) and the undoped low temperature phase transitions. The QC regime, as well as the renormalized classical regime, of the 2D antiferromagnetic spin liquid have been successfully investigated experimentally in this window. Here, a possible QC to QD crossover around 50 K falls also within the \( J \sim 1000 \) K and \( T_{sf} \).

In effective low temperature theories based on the 2D quantum non-linear \( \sigma \) model, which simulates the Néel order suppressing effect of hole doping with frustration in magnetic interactions, the scaling function \( g(x) \) is usually gapped. When interaction between spin excitations and doped fermions is explicitly considered, however, \( g(x) \) becomes gapless. The gapless \( g(x) \) observed in La\(_2\)CuO\(_4\), refer to B. J. Sternlieb et al., Phys. Rev. B 30, 1691 (2002).

In summary, the \((\pi, \pi)\)-correlated dynamic spin clusters in hole-doped La\(_2\)CuO\(_4\) have developed to substantial size below 150 K. The dynamics of such spin clusters cross over around 50 K from the quantum critical \( \omega/T \) scaling to a new low temperature regime with a saturated energy scale at \( T_0 \approx 1 \) meV. The observed crossover possibly corresponds to the theoretically expected quantum critical to quantum disordered crossover for 2D antiferromagnet.

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