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Spin-Stripe Density Varies Linearly With the Hole Content in Single-Layer $Bi_{2+x}Sr_{2-x}CuO_{6+y}$ Cuprate Superconductors

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Spin-stripe density varies linearly with hole content in single-layer Bi2201 cuprate

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We have performed inelastic neutron scattering measurements on the single-layer cuprate $Bi_{2+x}Sr_{2-x}CuO_{6+y}$ (Bi2201) with x=0.2, 0.3, 0.4 and 0.5, a doping range that spans the spin-glass (SG) to superconducting (SC) phase boundary. The doping evolution of low energy spin fluctuations ($\lesssim 11$ meV) was found to be characterized by a change of incommensurate modulation wave vector from the tetragonal [110] to [100]/[010] directions, while maintaining a linear relation between the incommensurability and the hole concentration, $\delta \approx p$. In the SC regime, the spectral weight is strongly suppressed below ~ 4 meV. Similarities and differences in the spin correlations between Bi2201 and the prototypical single-layer system $La_{2-x}Sr_xCuO_4$ are discussed.

The relevance of charge and spin stripes to the phenomenology of hole-doped cuprate superconductors has been gaining currency in recent years. The existence of static stripe order is well established in $La_{2-x}Ba_xCuO_4$ and closely related cuprates [1, 2]. Though stripe order can compete with three-dimensional superconducting phase order, it can coexist with strong superconducting correlations within the CuO_2 planes [3, 4]. Dynamic stripe correlations are inferred in $La_{2-x}Sr_xCuO_4$ based on the quantitative similarities with the magnetic spectra of $La_{2-x}Ba_xCuO_4$ as a function of doping [5]. The stripe picture has been generalized into an electronic liquid crystal analogy [6], and a nematic spin response has been identified in underdoped, superconducting $YBa_2Cu_3O_{6+y}$ (YBCO) [7, 8]. Transport properties of YBCO samples in a strong c-axis magnetic field show clear similarities to those measured in stripe-ordered systems (in zero field) [9]. Evidence for charge-stripe order induced by a strong magnetic field in an underdoped YBCO sample has been reported in a nuclear magnetic resonance study [10], although recent x-ray scattering studies indicate that the charge ordering in YBCO tends to have bidirectional modulations [11, 12].

Real-space imaging of electronic modulations by scanning tunneling microscopy (STM) in $Bi_2Sr_2CaCu_2O_{8+y}$ [13–15] and in $Bi_{2-x}Pb_xSr_{2-z}La_zCuO_{6+y}$ [16] has sometimes been interpreted as evidence of short-range stripe correlations. These unidirectional modulations are found to have a period of approximately $4a$, where $a = 3.8$ Å is the lattice spacing along a Cu-O bond direction. The corresponding wave vector of the modulations, $\mathbf{q}_{\rm STM}$, is $(\frac{1}{4}, 0, 0)$ in reciprocal lattice units $(2\pi/a)$; $q_{\rm STM}$ is observed to decrease with doping, varying in the range of 0.3 to 0.15 [15, 16].

Identifying $\mathbf{q}_{\rm STM}$ with the wave vector $\mathbf{q}_{\rm co}$ [13, 15] associated with charge stripe order in cuprates such as $La_{2-x}Ba_xCuO_4$ [1, 2] leads to a conundrum, as $q_{\rm co}$ *grows* with doping (at least for hole concentrations $p \lesssim 1/8$ [17]), opposite to the behavior of $\mathbf{q}_{\rm STM}$. When spin stripe order also occurs, antiferromagnetic spin correlations are modulated at $\mathbf{q}_{\rm so} = \frac{1}{2}\mathbf{q}_{\rm co}$. It is often possible to observe incommensurate (IC) spin fluctuations split about the antiferromagnetic wave vector $\mathbf{Q}_{\rm AF}$ by $\mathbf{q}_\delta \approx \mathbf{q}_{\rm so}$ even when there is no significant static stripe order, as in $La_{2-x}Sr_xCuO_4$ (LSCO) [18, 19]. Among possible resolutions of the conundrum, it might be that the nature of stripe correlations is not universal among different cuprate families, or that $\mathbf{q}_{\rm STM}$ measures something complementary to $\mathbf{q}_{\rm co}$.

In this Letter, we present the results of inelastic neutron scattering measurements of low-energy spin excitations in the system $Bi_{2+x}Sr_{2-x}CuO_{6+y}$ (Bi2201), demonstrating that $\delta = |\mathbf{q}_\delta| \approx p$ for $0.01 \lesssim p \lesssim 0.12$. This behavior is remarkably similar to that of $La_{2-x}Sr_xCuO_4$, even including the rotation of $\mathbf{q}_{\rm so}$ by 45° for $p \lesssim 0.06$ [18, 19]. These results provide strong circumstantial evidence that $\mathbf{q}_{\rm STM}$ does not correspond to $\mathbf{q}_{\rm co}$; instead, it more likely corresponds to a nesting of antinodal states close to $2k_{\rm F}$, where $k_{\rm F}$ is the nominal Fermi wave vector [16, 20]. This is not incompatible with a stripe origin, but would involve modulations along the charge stripes rather than perpendicular to them.

While several variants of Bi2201 have been studied in the literature, we chose to work with $Bi_{2+x}Sr_{2-x}CuO_{6+y}$ because it is possible both to vary the hole concentration in a controlled fashion and to grow sufficiently large crystals with the floating-zone technique, as previously demonstrated by Luo *et al.* [21]. We prepared single crystals of Bi2201 with x=0.2, 0.3, 0.4, and 0.5. The actual concentrations of Bi and Sr were determined by

TABLE I. Characterizations of the $Bi_{2+x}Sr_{2-x}CuO_{6+y}$ crystals. Elemental concentrations were determined by ICP-AES and hole concentration p was determined from Hall effect measurements.

x	Bi	Sr	Cu	p
0.2	2.173(1)	1.825(1)	0.989(2)	0.12(1)
0.3	2.282(3)	1.717(2)	0.992(4)	0.09(1)
0.4	2.376(1)	1.619(1)	0.992(2)	0.06(1)
0.5	-	-	-	0.01(1)

inductively-coupled plasma (ICP) atomic emission spectroscopy (AES), and the hole densities were determined by measurements of the Hall coefficient at 200 K, following [22, 23]; the results are listed in Table 1. The correspondence between p and x is consistent with the previously reported results based on measurements of the Fermi-surface volume by angle-resolved photoemission spectroscopy [24]. Based on magnetic susceptibility measurements, spin-glass-like behavior was observed below 3 K for $x = 0.4$ [25] and below 4 K for $x = 0.5$; neither magnetic order nor diamagnetism were detected above 2 K in the $x = 0.3$ and 0.2 samples. According to Luo *et al.* [21], the superconducting transition temperature, T_c, is ~ 1 K at $x = 0.2$, rising up to a maximum of 9 K at $x = 0.05$, as shown in Fig. 1(a). The reduced T_c in this system compared to La substitution for Sr is likely associated with structural disorder [26].

Most of the inelastic neutron scattering measurements were performed on thermal triple-axis spectrometer TOPAN installed at reactor JRR-3, Japan Atomic Energy Agency (JAEA). The typical collimator selections were 50′-100′-Sample-60′-180′, and the final energy was fixed at 14.7 meV. To reduce contamination from high-energy neutrons, a sapphire crystal was placed before the sample. A pyrolytic graphite filter was placed after the sample to eliminate higher-order neutrons. Additional measurements below 4 meV were performed at the cold neutron triple-axis spectrometers HER installed in the Guide Hall of JRR-3 and SPINS at the NIST Center for Neutron Research. For each composition, a couple of single crystals with total mass of 10–15 grams were coaligned and positioned so that the scattering plane corresponds to $(h, k, 0)$. Some results for the $x = 0.4$ crystal were reported previously [25].

For consistency, we will continue to index the scattering in terms of a tetragonal unit cell with $a_t = b_t \approx 3.81$ Å, although the symmetry is actually orthorhombic, with in-plane basis vectors along $[1\bar{1}0]$ and [110] corresponding to a_o and b_o, respectively. Although we cannot resolve the very small orthorhombic strain, we can distinguish the b_o^* direction by the presence of superlattice peaks (at $\sim 0.2b_o^*$) corresponding to the modulation of the BiO layers. We find that b_o^* runs in a unique direction in each crystal (*i.e.*, there is little, if any, twinning),

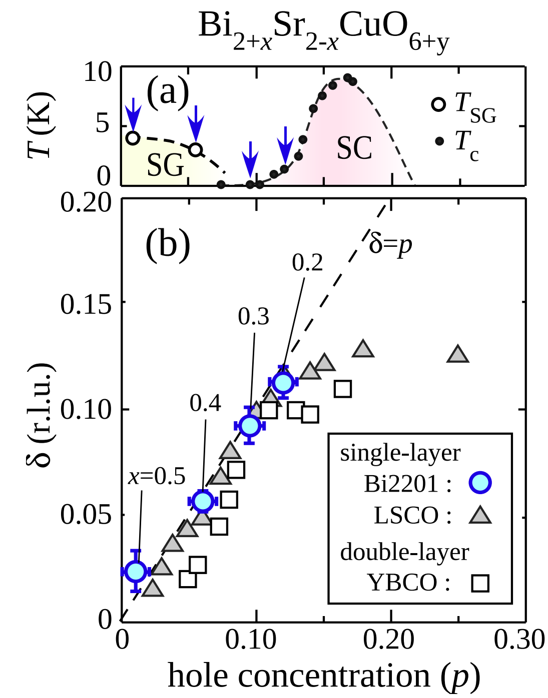

FIG. 1. (Color online) (a) Electronic phase diagram of $Bi_{2+x}Sr_{2-x}CuO_{6+y}$. The spin-glass transition temperature is plotted by open circles. T_c data (filled circles) are from [27]. The dashed lines are guides to the eye. (b) Hole concentration dependence of the incommensurability δ of low-energy spin fluctuations in $Bi_{2+x}Sr_{2-x}CuO_{6+y}$ (blue circles) compared with results for LSCO (gray triangles) [28–30] and YBCO (open squares) [7, 31] (with p estimated from T_c via [32]). The dashed line represents $\delta = p$.

and we will see that this results in a unique orientation of the IC spin fluctuations in the more underdoped crystals.

Inelastic neutron-scattering spectra for the $x = 0.2$ sample ($p = 0.12$) obtained for an excitation energy of $\hbar\omega = 11$ meV and a temperature of $T = 70$ K are shown in Fig. 2(a)-(c). Scans A and B exhibit IC peaks split about $\mathbf{Q}_{\mathrm{AF}}$ in the [100] and [010] directions, while the transverse scan C shows no structure. The pattern is identical to that observed in the superconducting phase of LSCO [28] and twinned YBCO [7, 31]. The intensity at this and lower energies is weak compared to that from LSCO for the same p and mass, measured under identical experimental setups, but, of course, the Cu mass fraction in Bi2201 is smaller by a factor of two.

Related scans for the $x = 0.4$ sample ($p = 0.06$) are shown in Fig. 2(d)-(f); these were measured at $\hbar\omega = 4$ meV and $T = 40$ K using the SPINS spectrometer with $E_f = 5$ meV. Here we see that IC peaks are only present in scan A′, which is along b_o^*, with no IC peaks along scan C′, in the direction of a_o^*. Similar scans at $\hbar\omega = 1$ meV are reported in [25], where the intensity is shown to fall off with temperature in a fashion consistent with magnetic correlations. An earlier study demonstrated that

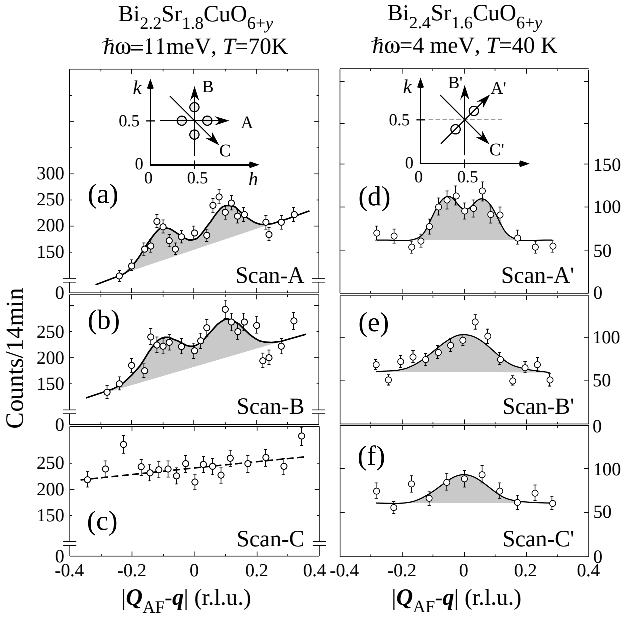

FIG. 2. Inelastic neutron scattering spectra in $Bi_{2+x}Sr_{2-x}CuO_{6+y}$ $x = 0.2$ at 11 meV, 70 K (a)-(c) and $x = 0.4$ at 4 meV, 40 K (d)-(f); solid lines are fits to the data. Insets show the IC peak geometry and the scan trajectories. IC peaks are seenn in (a) [100] and (b) [010] direction for x=0.2, and (d) [110] direction for x=0.4 sample, respectively.

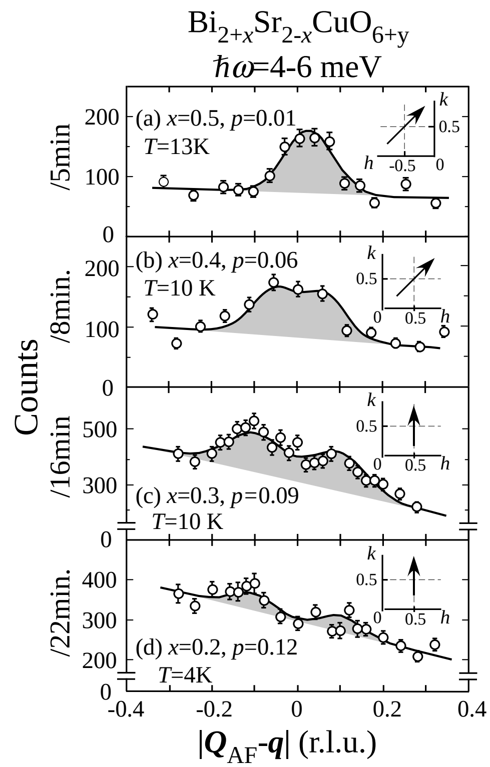

FIG. 3. Inelastic neutron scattering spectra at 4-6 meV and low temperatures in $Bi_{2+x}Sr_{2-x}CuO_{6+y}$ with (a) $x = 0.5$, (b) 0.4, (c) 0.3 and (d) 0.2; solid lines are fits to the data. Spectra were measured along [110] and [010] directions for (a,b) and (c,d), respectively, as indicated in insets.

the signal falls off in higher Brillouin zones, as expected for a magnetic form factor [33]. The observation of a longitudinal IC splitting along a unique orthorhombic axis corresponds perfectly with the behavior previously found in the spin-glass phase of LSCO [19, 29]. From the unique orientation we infer that static order is likely, and it is strongly indicated by bulk susceptibility measurements to occur below 3 K [25]; however, we were not able to detect IC peaks in elastic scattering for any of the samples. Of course, there is a substantial background in the elastic channel from nuclear diffuse scattering resulting from structural disorder, and that limits the sensitivity.

To illustrate the variation of the spin correlations for all four of our samples, Fig. 3 shows representative scans through $\mathbf{Q}_{\rm AF}$ obtained with thermal neutrons at excitation energies in the range of 4–6 meV. For $x = 0.3$, the orientation of the IC peaks is the same as for $x = 0.2$, but the splitting δ is slightly smaller. For both $x = 0.4$ and 0.5, the instrumental resolution is too broad to resolve IC peaks clearly, but the narrower width for $x = 0.5$ suggests a smaller splitting.

For quantitative analysis, we model the scattered intensity $I(\mathbf{Q}, \omega) \sim \chi''(\mathbf{Q}, \omega)(1 - e^{-\hbar\omega/k_{\rm B}T})^{-1}$ with the formula,

$$\chi''(\mathbf{Q}, \omega) = \chi''(\omega) \exp\left[-\ln(2)(\mathbf{Q} - \mathbf{Q}_{\rm AF} \pm \mathbf{q}_\delta)^2/\kappa^2\right].$$

$\chi''(\omega)$ and κ correspond to the local spin susceptibility and the peak-width (half-width at half-maximum), respectively. In fitting the data, the model intensity was convolved with the instrumental resolution function and a linear background was included. For the $x = 0.2$ and 0.3 samples, we defined $\mathbf{q}_\delta$ to include $(\delta, 0, 0)$ and $(0, \delta, 0)$. For $x = 0.4$ and 0.5, we set $\mathbf{q}_\delta = (\delta/\sqrt{2}, \delta/\sqrt{2}, 0)$; fitting the high-resolution data of Fig. 2(d) yielded $\delta = 0.057(5)$. Since the fitted values of κ for $x = 0.4$, 0.3, and 0.2 in Fig. 2(d) and Fig. 3(c,d) were comparable (~ 0.04 r.l.u.), we fitted the $x = 0.5$ data with κ fixed at 0.04 and with the assumption of IC peaks oriented as for $x = 0.4$.

The values of δ obtained for Bi2201 from the fitting are plotted as a function of p in Fig. 1(b), where they are compared with results for LSCO [28–30] and YBCO [7, 31]. We find that $\delta \approx p$ in Bi2201, which appears to be quantitatively identical to LSCO and qualitatively similar to YBCO. Comparing with the spin-glass and nominal superconducting transitions indicated in Fig. 1(a), the rotation of $\mathbf{q}_\delta$ occurs between the spin-glass and superconducting phases, just as in LSCO [19].

The degree of similarity between Bi2201 and LSCO is a bit surprising, given that photoemission studies have indicated significant differences [34]. In particular, the chemical potential in LSCO remains rather constant with

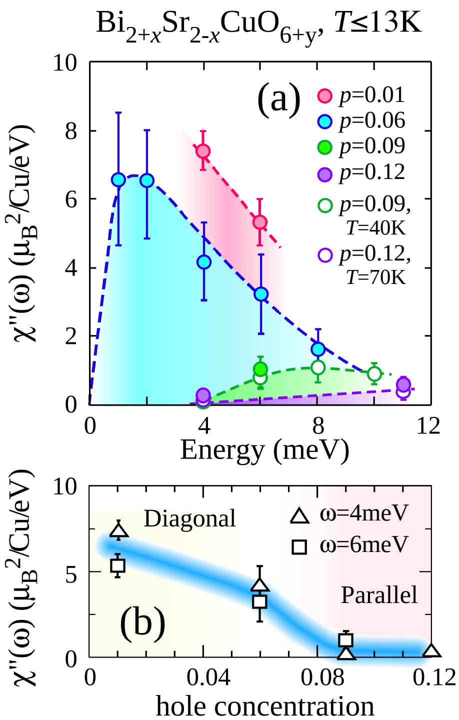

FIG. 4. (Color online) (a) Energy dependence of the local spin susceptibility $\chi''(\omega)$ for $Bi_{2+x}Sr_{2-x}CuO_{6+y}$ with x=0.2, 0.3, 0.4 and 0.5. Results at low temperatures below 13 K and high temperatures of 70 K for x=0.2 and 40 K for x=0.3 are plotted by closed and open circles, respectively. (b) Hole concentration dependence of $\chi''(\omega)$ at 4 meV (triangles) and 6 meV (squares). Dashed lines in (a) and broad line in (b) are guides to the eye.

doping for $0 \leq p \lesssim 0.12$, while it shifts downward linearly with doping in Bi2201 [34]. There have been variety of models proposed to explain the doping dependence of δ in cuprates [35, 36], not all of which involve stripes; nevertheless, if stripes are involved, then it is reasonable to expect that $\mathbf{q}_{\rm co} \approx 2\mathbf{q}_\delta$, and hence $|\mathbf{q}_{\rm co}| \approx 2p$ for Bi2201 with $p \lesssim 0.12$ based on the present results.

The experimental frequency dependence of the **Q**-integrated magnetic response $\chi''(\omega)$ is plotted in Fig. 4(a). The relative signal strength was normalized by measuring the same transverse acoustic phonon for each sample on the TOPAN spectrometer. To determine the absolute scale, we have compared with time-of-flight neutron scattering results obtained for the Bi2201 $p = 0.06$ sample, which were calibrated to measurements of elastic incoherent scattering of a vanadium standard, yielding $\chi''(6\text{ meV}) = 3.1\ \mu_{\rm B}^2/\text{eV/Cu}$ [37]. The resulting $\chi''(\omega)$ of 1–7 $\mu_{\rm B}^2$/eV/Cu for $\hbar\omega < 10$ meV in Bi2201 $p = 0.06$ is comparable to the magnitude of 3–9 $\mu_{\rm B}^2$/eV/Cu in the same energy range reported for lightly-doped LSCO $x = 0.05$ from time-of-flight measurements [5]. For the two samples near the spin-glass regime ($p = 0.01$ and 0.06), the low-temperature $\chi''(\omega)$ is large at low energy, consistent with proximity to an ordered state. For the more highly-doped samples ($p = 0.09$ and 0.12), $\chi''(\omega)$ is strongly reduced at low energy, and there is not much change when the temperature is raised (open symbols). The doping trend for the low-energy weight is illustrated in Fig. 4(b). From Fig. 4(a), the magnitude of the spectral weight at $\sim$ 10 meV is comparable to that in superconducting LSCO [5], but the apparent gapping of low-frequency spin fluctuations at these modest dopings is different from the case of LSCO with $p < 0.13$ [38]. In particular, the results indicate that the system is farther from static spin order of the type associated with the 1/8 anomaly in LSCO.

Our result $\delta \approx p$ in Bi2201 provides further evidence of universal behavior of spin correlations in the cuprates. The implication for possible coexisting charge modulations is in conflict with a common interpretation of STM studies [13–15]. Specifically, the doping dependence of $\mathbf{q}_{\rm STM}$ is not consistent with the simplest stripe interpretation. The static electronic modulations are more likely due to $2k_{\rm F}$-like oscillations associated with the large antinodal density of states [16, 20], as they are a screening response to disorder. We are aware that $\mathbf{q}_{\rm STM}$ does not precisely match $2k_{\rm F}$ measured by photoemssion [39]; however, this is not a problem as the STM modulations have maximum amplitude at bias voltages comparable to the pseudogap energy, so that $\mathbf{q}_{\rm STM}$ need not be determined by states precisely at the Fermi level. It may be relevant that the antinodal pseudogap energy is also the scale on which antiferromagnetic spin fluctuations become strongly damped [40]. Hence, the low-energy spin fluctuations and the STM modulations appear to detect different electronic features with distinct energy scales.

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