Scaling Behavior of the Generalized Susceptibility in $La_{2-x}Sr_xCuO_{4+y}$

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We have measured the static and dynamic spin fluctuations in a crystal of $La_{2-x}Sr_xCuO_{4+y}$ in the spin-glass doping regime. The static spin correlations are well described by a simple model in which the inverse correlation length $\kappa(x,T)=\kappa(x,0)+\kappa(0,T)$. The dynamic spin fluctuations exhibit a simple scaling behavior in ω/T for temperatures 10 K $\leq T \leq$ 500 K for the measured energies $4.5 \leq \omega \leq 12$ meV.

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The nature of the magnetism in the lamellar copper oxides continues to be a subject of extensive study both because of its intrinsic interest and because of its possible role in high-temperature superconductivity. Spin fluctuations in antiferromagnetic, spin-glass, and superconducting samples have been probed using neutron-scattering [1], nuclear-magnetic-resonance [2], and muon-spin-resonance techniques [3]. The former two, in particular, have shown the importance of magnetic fluctuations in superconducting samples. Of special importance for the studies reported here is the observation in the $La_{2-x}Sr_xCuO_{4+y}$ series that the spin fluctuations at low energies (say $\omega \simeq 6$ meV, using $\hbar = k_B = 1$ as we will throughout) have comparable strengths in antiferromagnetic, spin-glass, and superconducting samples [4].

In this paper we report on a detailed study of the instantaneous and dynamic spin fluctuations for a crystal of $La_{1.96}Sr_{0.04}CuO_4$ which is in the intermediate spin-glass regime. These measurements are part of a more global study of the nature of the crossover from antiferromagnetic to spin-glass behavior which will be reported at a later date [5]. However, our results for the dynamics are sufficiently novel and, in our view, important that we wish to report those results separately here. We find that the instantaneous correlations are well described by a simple model in which $\kappa(x,T)=\kappa(x,0)+\kappa(0,T)$ [5,6]. The spin-spin correlation length, $\xi=\kappa^{-1}(x,0)$, is $\sim$40 Å and $\kappa(x,T)$ is temperature independent below 300 K. Studies of the dynamics in the CuO_2 plane reveal that the fluctuations are centered about the (π,π) position (square lattice, unit lattice constant notation) with negligible weight at other high-symmetry positions such as $(\pi,0)$ and $(2\pi,0)$. Our essential new result is that we find that the integrated response around (π,π) as a function of temperature T exhibits a simple scaling behavior in ω/T for temperatures 10 K $\lesssim T \leq$ 500 K and energies 4.5 meV $\leq \omega \leq$ 12 meV. This scaling leads to a natural explanation of a variety of normal-state properties of the copper oxides.

Before discussing the experiments, we note that the transport in our $La_{2-x}Sr_xCuO_{4+y}$ single crystals has been extensively investigated by Preyer *et al.* [7]. In the doping regime $0.02 < x \lesssim 0.06$, the resistance within the CuO_2 planes is metallic and linear in T over a wide range of temperatures above $\sim$80 K with an amplitude, normalized to carrier concentration, comparable to that in $YBa_2Cu_3O_7$. Below $\sim$80 K logarithmic corrections to the resistivity appear, possibly the result of weak-localization effects, thence producing an apparent metal-to-semiconductor crossover. As we shall show, these weak logarithmic corrections to the conductance have no important consequences for the static and dynamic spin fluctuations. This, in turn, suggests that our results should apply generally for metallic lamellar CuO_2 samples.

The neutron-scattering experiments were carried out at the National Institute of Standards and Technology Research Reactor. For consistency with previous work [1,4] for orthorhombic $La_{2-x}Sr_xCuO_4$, we use the crystallographic notation pertaining to space group *Cmca*; reflections in the CuO_2 planes then have Miller indices $(h0l)$ while $[0k0]$ is the direction perpendicular to the CuO_2 planes. Because of twinning, h and l cannot be distinguished. In this notation, (100) or (001) corresponds to the (π,π) position in reciprocal space for a square lattice with unit lattice constant. The static measurements were carried out on a two-axis spectrometer (BT4) with incident neutron energy $E_i=30.5$ meV and collimator configuration 40′-20′-S-20′. The sample was oriented in the $(hk0)$ plane and scans were carried out across $(h\,0.38\,0)$. As discussed previously [1,4], in this geometry one automatically integrates over the 2D spin fluctuations which have energies $\sim -T < \omega < 30.5$ meV. The inelastic measurements were carried out on a triple-axis spectrometer (BT9) with fixed incoming neutron energy $E_i=30.5$ meV and collimator configuration 40′-40′-S-40′-80′. The sample was oriented with [010] vertical so that the entire 2D in-plane reciprocal lattice could be studied. Pyrolytic graphite (PG) (002) was used as both monochromator and analyzer. For both experiments a PG filter was placed before the sample to remove higher-order neutrons. For measurements between 10 and 350 K the sample was placed in a Displex cryostat; for higher temperatures we used a standard oven.

Before discussing the experimental results we comment briefly on the crystal growth and characterization. A series of single crystals of $La_{2-x}Sr_xCuO_{4+y}$ were grown using the CuO flux technique [8]. The specific crystal studied here, of volume 0.9 cm^3, was determined to have $x=0.04\pm0.01$ by electron microprobe analysis; it was used as grown. The exact oxygen concentration y is unknown although as-grown crystals tend to have a slight excess of oxygen. For all of our crystals, the homogeneity has been characterized with unusual thoroughness. For samples in the spin-glass regime, the sharpness of the tetragonal-to-orthorhombic phase transition provides the best measure of the homogeneity since it is very sensitive to macroscopic concentration gradients and microscopic astatistical distributions of the Sr and/or O atoms. For our sample we find a sharp structural phase transition at $T_0=435\pm10$ K. This is consistent with previous studies [4] for $x\simeq0.04$. The width of the transition implies that the variation in the Sr and O contents is less than ±0.01.

We now discuss the results of the neutron-scattering studies. For a Heisenberg system, the neutron magnetic cross section is proportional to [9]

$$S(\mathbf{Q},\omega)=\frac{1}{2\pi}\sum_{\mathbf{r}}\int_{-\infty}^{\infty}dt\,e^{i\mathbf{Q}\cdot\mathbf{r}-\omega t}\langle\mathbf{S}_0(0)\cdot\mathbf{S}_{\mathbf{r}}(t)\rangle \sim\frac{1}{1-e^{-\omega/T}}\mathrm{Im}\chi(\mathbf{Q},\omega)\,. \quad (1)$$

A full integration of Eq. (1) over energy yields the instantaneous spin-spin correlation function $S(\mathbf{Q})=(1/2\pi)\sum_{\mathbf{r}}e^{i\mathbf{Q}\cdot\mathbf{r}}\langle\mathbf{S}_0\cdot\mathbf{S}_{\mathbf{r}}\rangle$.

Our experimental technique and method of analysis for the energy integrating measurements are indentical to those discussed in Ref. [4]. We find that the static structure factor is peaked at (100), indicating that the local magnetic order is commensurate with the lattice. Fits of the measured structure factor to a two-dimensional Lorentzian $S(\mathbf{Q})=A/[\kappa^2+(\mathbf{Q}-\mathbf{G}_{100})^2]$ yield the values for κ shown in Fig. 1. We show, in addition, in Fig. 1, results from a pure crystal of La_2CuO_4 with $T_N=320$ K [5]. Clearly, in the doped sample the inverse correlation length κ is nearly independent of temperature from 10 to 300 K and then increases gradually with increasing temperature above room temperature. The solid line in Fig. 1 for the pure sample, $\kappa(0,T)$, is the prediction of Chakravarty, Halperin, and Nelson [6] with the spin-wave velocity fixed at the measured value of $v=850$ meV Å [10]. For the doped sample, the solid line corresponds to $\kappa(x,T)=\kappa(x,0)+\kappa(0,T)$, where $\kappa(x,0)$ is determined by the lowest-temperature data and, as above, $\kappa(0,T)$ is the pure system behavior. The implications of these results for this and other samples are discussed in detail in Ref. [5]. For our purposes here it is sufficient to note that the spin correlation length below room temperature is about 40 Å and there are no evident effects on the spin correlations from the logarithmic conductance effects which typically set in below ~80 K.

We show in Fig. 2 a series of inelastic scans at 6 meV

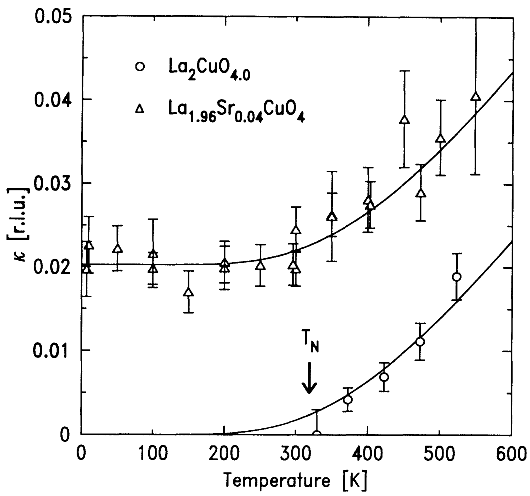

FIG. 1. Inverse magnetic correlation length of pure ($T_N=320$ K) and Sr-doped La_2CuO_4. The solid line through the undoped crystal data is the prediction of Chakravarty, Halperin, and Nelson for the spin-$\frac{1}{2}$ Heisenberg antiferromagnet [6], while for the doped system the solid line is $\kappa(x,T)=\kappa(x,0)+\kappa(0,T)$ with $\kappa(x,0)=0.02$ r.l.u. (r.l.u. denotes radiation length unit).

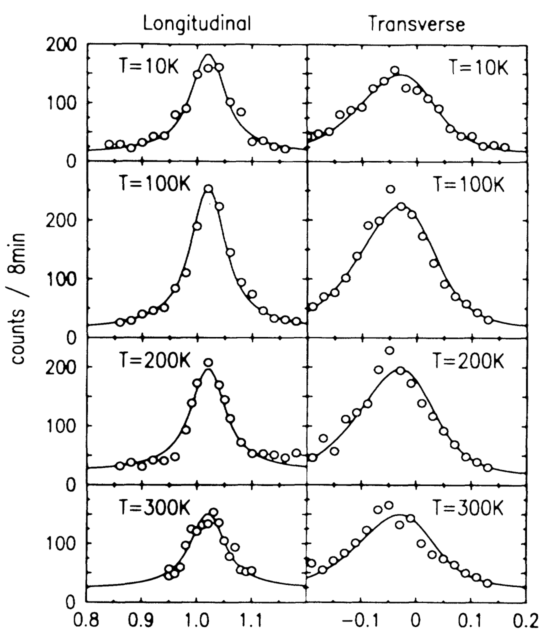

FIG. 2. Longitudinal (left half) and transverse (right half) scattering profiles through (1.02 0 −0.03) at 6-meV energy transfer. The solid lines are convolutions of an isotropic 2D Lorentzian of width 0.02 r.l.u. with the instrumental resolution function. The slight incommensurability of the peaks is due to a subtle monochromator misalignment.

at temperatures between 10 and 300 K. We note that the peaks are sharp and symmetric and that the background is relatively flat and featureless. Thus we are able to extract integrated intensities quite reliably. The solid lines in the figure are the results of fits by an isotropic 2D Lorentzian whose width in **Q** is fixed at the measured inverse correlation length $\kappa(x,T)$, convoluted with the instrumental resolution function. This simple functional form describes our data adequately for all temperatures and energies. We note that the apparent anisotropy of the (100) peak width is entirely due to the instrumental resolution. We carried out scans in both the longitudinal and transverse directions through (100) for temperatures between 10 and 500 K at energies of 4.5, 6.0, 9.0, and 12.0 meV. We also surveyed reciprocal space extensively at 10 and 300 K; we found no significant scattering above background at any other place in reciprocal space including especially the $(\frac{1}{2}0\frac{1}{2})$ and (101) [or in square-lattice notation, the $(\pi,0)$ and $(2\pi,0)$] positions.

We show in Fig. 3 the resolution-corrected 2D integrated intensities as a function of temperature. In each case, the intensity peaks at a temperature of $T \simeq 2\omega$. This suggests that one should replot the data in a scaled form in terms of ω/T. Since most theories and many derivative quantities involve $\text{Im}\chi(\mathbf{Q},\omega)$, we have used Eq. (1) to convert the measured $\int_{(\pi,\pi)} d\mathbf{Q}\, S(\mathbf{Q},\omega)$ to $\int_{(\pi,\pi)} d\mathbf{Q}\, \text{Im}\chi(\mathbf{Q},\omega)$. We have normalized the data at the maximum intensity in Fig. 3 and we have chosen the overall scale such that the integrated susceptibility for $\omega/T > 1$ is ~ 1. We will discuss the significance of the normalization constant below. The results are plotted in Fig. 4.

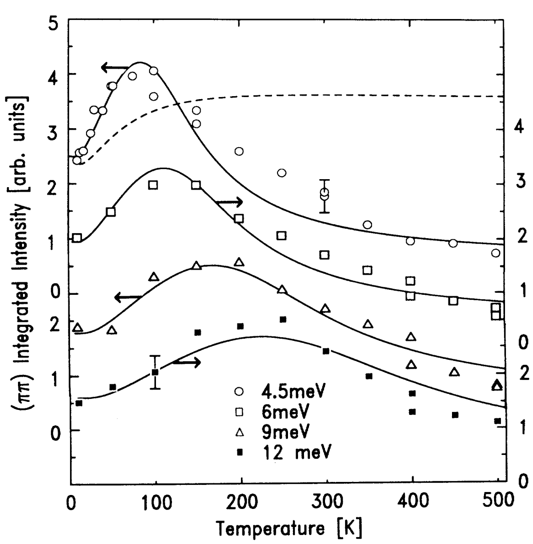

FIG. 3. Intrinsic 2D integrated intensities of the scattering profiles at various temperatures and energy transfers. The solid lines are of the form $(1-e^{-\omega/T})^{-1}\tan^{-1}[a_1(\omega/T)+a_3(\omega/T)^3]$ with $a_1=0.43$, $a_3=10.5$. The dashed line associated with the 4.5-meV data is the simplest marginal-Fermi-liquid (Ref. [12]) prediction with $a_1=2.2$ and $a_2=0$.

Figure 4 contains the salient result of this paper. We see that all of the data for temperatures between 10 and 500 K and energies between 4.5 and 12 meV fall on a single universal curve when plotted in this fashion. This scaling occurs in spite of the fact that the correlation length is nearly independent of temperature over the entire temperature interval. We also note that there is no difference for data at temperatures above and below 80 K. Thus $\text{Im}\chi(\mathbf{Q},\omega)$, along with the static correlations, is insensitive to localization effects as one might have expected on physical grounds since such localization typically involves the behavior of the carrier wave functions at large distances.

In the absence of any theory for these results, we use the following phenomenological model to parametrize our data. In mean-field theory for an antiferromagnetically correlated system $\chi(\mathbf{q},\omega)\sim\xi^2(1+q^2\xi^2-if)^{-1}$, where $\mathbf{q}=\mathbf{Q}-\mathbf{G}_{100}$. For relaxational dynamics one typically writes $f=\omega/\Gamma$, with $\Gamma\sim\xi^{-2}$. More generally, however, $f=f(q\xi,qa,\omega/\Gamma,\omega/T)$. The data in Fig. 4 suggest that the ω/T dependence dominates. We therefore ignore all but the ω/T dependence of f and write f

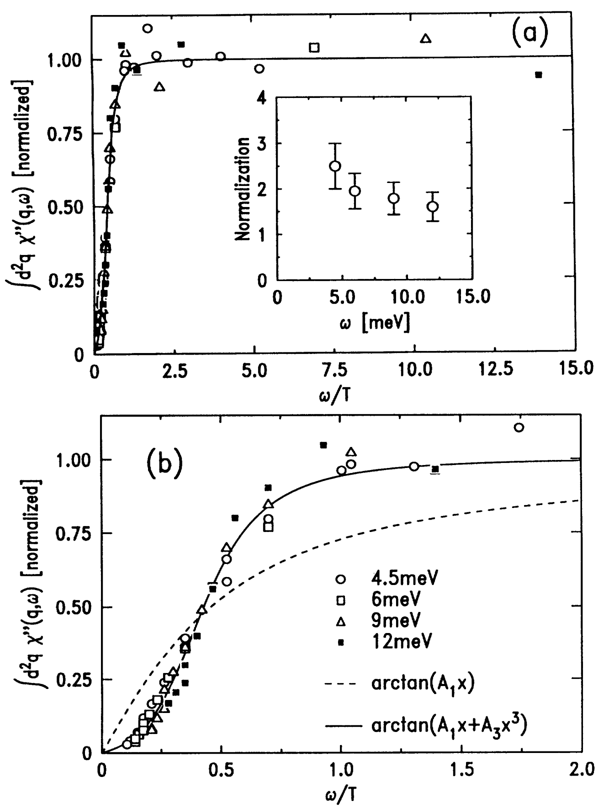

FIG. 4. Normalized, integrated spin susceptibility around (π,π) as a function of the scaling variable ω/T. Inset in (a): The normalization constant. (b) A closeup of panel (a). The lines are the function $(2/\pi)\tan^{-1}[a_1(\omega/T)+a_3(\omega/T)^3]$ with $a_1=0.43$, $a_3=10.5$ (solid line), and $a_1=2.2$, $a_3=0$ (dashed line).

$=\sum_{n=1,3,\ldots} a_n(\omega/T)^n$ with n odd as required by time-reversal symmetry. Some simple algebra [11] then gives

$$\int_{(\pi,\pi)} d\mathbf{q}\,\mathrm{Im}\chi(\mathbf{q},\omega) = C(\omega)\tan^{-1}\left[a_1\left(\frac{\omega}{T}\right)+a_3\left(\frac{\omega}{T}\right)^3+\cdots\right], \quad (2)$$

where $C(\omega)$ allows for a weak ω dependence in the overall amplitude. The solid line in Fig. 4 represents a fit by this form normalized to unity at large ω/T with $a_1=0.43$ and $a_3=10.5$. Inclusion of higher-order terms does not improve the fit. Clearly this simple phenomenological model works very well.

We note that Eq. (2) is consistent with the marginal-Fermi-liquid model of Varma *et al.* [12] which hypothesizes, as part of a phenomenological picture of the fluctuations in the optimal superconducting concentration regime, $\mathrm{Im}\chi\sim\omega/T$ for $|\omega|\ll T$, ~ 1 for $|\omega|\gg T$. The physics, however, appears to be more general than that assumed in Ref. [12] since, as we shall discuss in future publications [13], Eq. (2) works well for $La_{2-x}Sr_xCuO_{4+y}$ and $YBa_2Cu_3O_{6+\delta}$ samples with antiferromagnetic correlation lengths varying between 150 and 10 Å. It should be noted that the scaling form evident in Fig. 4 must break down at large ω since we expect a cutoff at high energies $\omega\sim 2J$ where J is the Heisenberg exchange coupling.

Many physical quantities depend directly on $\mathrm{Im}\chi(\mathbf{Q},\omega)$. For example, Moriya, Takahashi, and Ueda [14] have argued that the electrical resistivity due to antiferromagnetic fluctuations should scale like

$$R\sim T\int_{-\infty}^{\infty}\left(\frac{\omega}{T}\right)d\left(\frac{\omega}{T}\right)\frac{e^{\omega/T}}{(e^{\omega/T}-1)^2}\int d\mathbf{Q}\,\mathrm{Im}\chi(\mathbf{Q},\omega)\,. \quad (3)$$

Clearly, any form for $\int d\mathbf{Q}\,\mathrm{Im}\chi(\mathbf{Q},\omega)$ which is homogeneous in ω/T will lead to a resistance linear in T due to spin fluctuations. Our data indeed manifest such homogeneity provided that $C(\omega)\sim$const. As shown in the inset in Fig. 4(a), $C(\omega)$ is indeed $\sim$const, except for the point at 4.5 meV. Heuristically, for $\omega\gtrsim v\kappa\sim 20$ meV, one expects a crossover to spin-wave behavior; $C(\omega)\sim$const. Thus, overall, we expect $C(\omega)$ to depend weakly on ω over most of the relevant energy range. This in turn implies $R\sim T$. Clearly, additional neutron experiments at high energies are required to measure the relevant behavior of $\int d\mathbf{Q}\,\mathrm{Im}\chi$ directly [15]. It is also apparent from Eq. (3) that one would expect an isotropic, negative magnetoresistance due to the diminution in $\mathrm{Im}\chi$ by an applied field. This agrees with the recent results of Preyer *et al.* [7]. However, a detailed theoretical treatment is required to explain their observed H/T scaling [7] although our measured ω/T scaling is certainly suggestive. The above arguments also seem capable of explaining the optical conductivity [14]. Finally, Eq. (2) appears to be consistent with Cu^{2+} NMR relaxation data in both $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+\delta}$ [2].

As noted above, the essential result of this work is shown in Fig. 4. Prediction of this form for $\int_{(\pi,\pi)} d\mathbf{Q}\times\mathrm{Im}\chi(\mathbf{q},\omega)$ represents a challenge for any model of the CuO_2 superconductors.

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[1] See, for example, R. J. Birgeneau and G. Shirane, in *Physical Properties of High Temperature Superconductors I,* edited by D. M. Ginsberg (World Scientific, Singapore, 1989), pp. 151–212; R. J. Birgeneau, Am. J. Phys. **58**, 28 (1990).

[2] See, for example, C. H. Pennington and C. P. Slichter, in *Physical Properties of High Temperature Superconductors II,* edited by D. M. Ginsberg (World Scientific, Singapore, 1990), pp. 269–368; K. Ishida *et al.*, J. Phys. Soc. Jpn. **58**, 26 (1989).

[3] See, for example, Y. J. Uemura *et al.*, J. Phys. (Paris), Colloq. **49**, C8-2087 (1989).

[4] R. J. Birgeneau *et al.*, Phys. Rev. B **38**, 6614 (1988); T. R. Thurston *et al.*, Phys. Rev. B **40**, 4585 (1989).

[5] B. Keimer *et al.* (unpublished).

[6] S. Chakravarty, B. I. Halperin, and D. R. Nelson, Phys. Rev. B **39**, 2344 (1989).

[7] N. W. Preyer *et al.* (unpublished); N. W. Preyer *et al.*, Physica (Amsterdam) **162-164C**, 1003 (1989).

[8] Y. Hidaka *et al*, J. Cryst. Growth **85**, 581 (1987).

[9] W. Marshall and R. D. Lowde, Rep. Prog. Phys. **31**, 705 (1968).

[10] G. Aeppli *et al.*, Phys. Rev. Lett. **62**, 2736 (1989).

[11] We thank V. Emery and J. Tranquada for first pointing this out to us.

[12] C. M. Varma *et al.*, Phys. Rev. Lett. **63**, 1996 (1989).

[13] Y. Endoh *et al.* (unpublished); P. Gehring *et al.* (unpublished).

[14] T. Moriya, Y. Takahashi, and K. Ueda, J. Phys. Soc. Jpn. **59**, 2905 (1990); (unpublished).

[15] After submission of this paper we received a preprint of an article by S. M. Hayden *et al.* [now published in Phys. Rev. Lett. **66**, 821 (1991)] which indeed confirms our assumption $C(\omega)\sim$const at large ω. This paper reports results and analysis of neutron experiments in $La_{1.95}Ba_{0.05}CuO_4$ which generally are consistent with work reported here although there are apparent discrepancies for $T<80$ K which require further investigation.