

# UC Irvine

## UC Irvine Previously Published Works

### Title

Magnetic fluctuations in La<sub>1.95</sub>Ba<sub>0.05</sub>CuO<sub>4</sub>.

### Permalink

<https://escholarship.org/uc/item/8zm6n6zq>

### Journal

Physical review letters, 66(6)

### ISSN

0031-9007

### Authors

Hayden, SM  
Aeppli, G  
Mook, H  
[et al.](#)

### Publication Date

1991-02-01

### DOI

10.1103/physrevlett.66.821

### Copyright Information

This work is made available under the terms of a Creative Commons Attribution License, available at <https://creativecommons.org/licenses/by/4.0/>

Peer reviewed

### Magnetic Fluctuations in $\text{La}_{1.95}\text{Ba}_{0.05}\text{CuO}_4$

S. M. Hayden,<sup>(1),(a)</sup> G. Aeppli,<sup>(2)</sup> H. Mook,<sup>(3)</sup> D. Rytz,<sup>(4)</sup> M. F. Hundley,<sup>(5)</sup> and Z. Fisk<sup>(5)</sup>

<sup>(1)</sup>*Institut Laue-Langevin, B.P. 156X, 38042 Grenoble, France*

<sup>(2)</sup>*AT&T Bell Laboratories, Murray Hill, New Jersey 07974*

<sup>(3)</sup>*Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831*

<sup>(4)</sup>*Sandoz Optoelectronics, Bau 88, CH-4002 Basel, Switzerland*

<sup>(5)</sup>*Los Alamos National Laboratory, Los Alamos, New Mexico 87545*

(Received 9 November 1990)

Neutron-scattering and resistivity experiments on single crystals of  $\text{La}_{1.95}\text{Ba}_{0.05}\text{CuO}_4$  are described. On warming to as high as 250 K, the correlation length does not change, and the temperature dependence of the generalized susceptibility  $\chi(Q, \omega)$  is due solely to the temperature dependence of the local response. We show that the associated relaxation rate in the metallic regime is  $\hbar\Gamma \cong \frac{1}{2}k_B T$ , in accord with the marginal-Fermi-liquid hypothesis, and relate our results to nuclear spin-lattice relaxation data.

PACS numbers: 75.30.Ds, 71.30.+h, 74.70.Vy, 75.50.Ee

$\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$  is interesting not only because it displays superconductivity at  $x \cong 0.15$ , but also because it undergoes a metal-insulator transition at  $x \cong 0.05$ . In spite of the considerable effort devoted to metal-insulator transitions in this and other systems, relatively little is known about the behavior of the generalized magnetic response function  $\chi(q, \omega)$  near such transitions. We have therefore used cold, thermal, and epithermal neutron-scattering measurements to characterize  $\chi''(q, \omega)$  over a wide range of frequencies,  $0.5 < \hbar\omega < 90$  meV, for a nearly metallic  $\text{La}_{1.95}\text{Ba}_{0.05}\text{CuO}_4$  crystal. The principal conclusion is that for  $3 < T < 250$  K, the temperature dependence of  $\chi''(q, \omega)$  is entirely due to the temperature dependence of the local magnetic response  $\chi''_0(\omega)$  of the Cu ions.  $\chi''_0(\omega)$ , in turn, is characterized by a single energy scale  $\hbar\Gamma$ . For the high-temperature metallic regime,  $\hbar\Gamma \sim \frac{1}{2}k_B T$ , as anticipated from the marginal-Fermi-liquid hypothesis.<sup>1</sup> The framework for understanding the data and the conclusions derived here is probably also applicable to experiments performed over more limited frequency ranges on  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  and  $\text{YBa}_2\text{Cu}_3\text{O}_{6+\delta}$ .<sup>2</sup>

We grew  $\text{La}_{1.95}\text{Ba}_{0.05}\text{CuO}_4$  crystals with volumes of order  $1 \text{ cm}^3$  by top seeding a solution of 0.85 CuO and 0.15  $\text{La}_{1.2}\text{Ba}_{0.2}\text{CuO}_4$ . After growth, the crystals were subjected to 45-h anneals at 600°C and 114 bars of  $\text{O}_2$ . When prepared in this fashion, the crystals displayed no antiferromagnetic (AFM) Bragg peaks (see discussion below), and the orthorhombic  $T=5$  K lattice constants were  $a=5.361(3)$  Å,  $b=13.18(2)$  Å, and  $c=5.411(3)$  Å. Recall that the parameters  $a$  and  $c$  characterize the basal planes such that in the tetragonal limit, the nearest-neighbor Cu-Cu separation would be  $a/\sqrt{2} = c/\sqrt{2}$ .

Figure 1(a) shows the temperature-dependent basal-plane resistivity  $\rho_{ab}$ . On cooling from 300 K, there is a substantial decrease in  $\rho_{ab}$  (with a linear asymptote as  $T \rightarrow 300$  K) until  $T \cong 50$  K, where localization effects set in and  $\rho_{ab}$  begins to rise. Finally, at the lowest  $T$ , there is a slight decrease in  $\rho_{ab}$ , due to small supercon-

ducting regions in the conducting path. In a qualitative sense, these results are very similar to others<sup>3</sup> obtained for the same nominal composition. However, the absolute magnitude of  $\rho_{ab}$  is lower than reported elsewhere, most probably because our samples are single crystals.

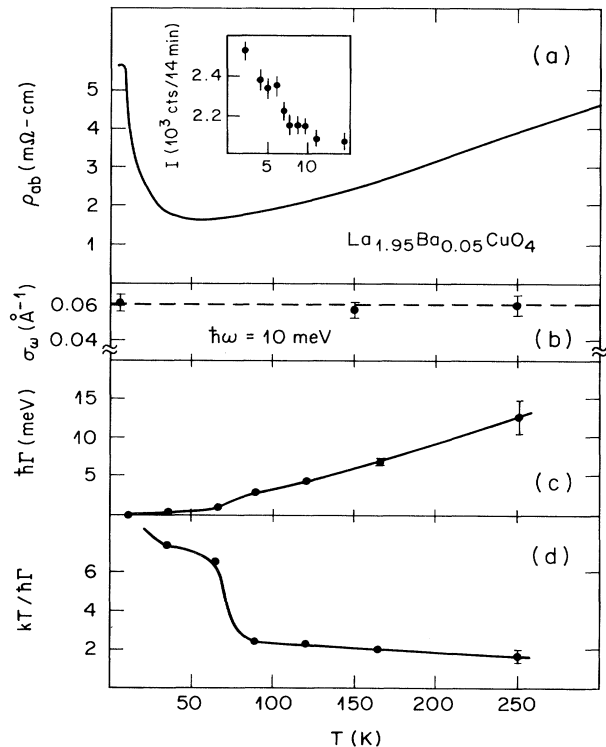


FIG. 1. Temperature dependence of (a) basal-plane resistivity (inset: elastic signal proportional to the spin-glass order parameter), (b)  $\sigma_\omega$  as measured at  $\hbar\omega = 10$  meV, (c) magnetic relaxation rate  $\hbar\Gamma$ , and (d) calculated ratio  $k_B T / \hbar\Gamma$ , proportional to nuclear spin-lattice relaxation rate.

The minimum value (1.8 m $\Omega$  cm) is close to  $(h/e^2)b/2 = 1.7$  m $\Omega$  cm, where  $h/e^2 = 25813 \Omega$  and  $b/2$  is the interlayer spacing in our sample.

The neutron-scattering measurements were performed using various three-axis spectrometers [IN1, IN8, and IN12 at the Institut Laue-Langevin (ILL), and TAS6 at Risø National Laboratory] on hot, thermal, and cold sources at the ILL high-flux and Risø medium-flux reactors. Figure 2 shows a series of constant- $\hbar\omega$  scans as a function of in-plane momentum transfer  $q$  relative to the nearest two-dimensional AFM zone center of pure  $\text{La}_2\text{CuO}_4$ . For  $\hbar\omega = 0$  and  $T = 3$  K (solid circles in scan labeled (d)), we observe a peak at  $q = 0$  with a half width at half maximum (HWHM) of  $\xi^{-1} = 0.055 \pm 0.003 \text{ \AA}^{-1}$ , indistinguishable from the inverse correlation length of  $0.055 \pm 0.015 \text{ \AA}^{-1}$  reported<sup>3</sup> for a sample of  $\text{La}_{1.94}\text{Sr}_{0.06}\text{CuO}_4$ . The width and amplitude of the peak do not change when the momentum transfer perpendicular to the basal planes is changed from  $0.4b^*$  (as in the figure) to 0. In particular, no magnetic Bragg scattering is found in the latter case. Thus, at  $T = 3$  K, the sample is divided into two-dimensional AFM microdomains, of typical side length 20  $\text{\AA}$ , which are frozen on time scales  $\tau \sim 6 \times 10^{-10}$  sec, corresponding to the instrumental energy resolution (HWHM) of 65  $\mu\text{eV} \equiv 16$  GHz. On warming to  $\sim 15$  K, magnetic scattering is no longer ap-

parent in the  $\hbar\omega = 0$  scan, as indicated by the open circles in Fig. 2(d). Thus the lifetime of the AFM clusters has become significantly shorter than  $10^{-10}$  sec. The inset of Fig. 1(a), which shows the temperature dependence of the magnetic intensity, illustrates the freezing process in our sample: On the time scale of the present measurements, the freezing temperature is  $T_g = 8$  K. For more detailed descriptions of spin-glass phenomena in  $\text{La}_{2-x}(\text{Sr},\text{Ba})_x\text{CuO}_4$ , the reader should consult Refs. 4.

Of greater interest here are higher-energy fluctuations. In Fig. 2, the  $\hbar\omega = 15$  meV scan (c) is somewhat broader than that at  $\hbar\omega = 0$ . At larger  $\hbar\omega$ , the scans in Fig. 2 become substantially broader than at  $\hbar\omega = 0$ , and the fluctuations must therefore also have components with wavelengths  $< \xi$ . In other words, excitations *within* the AFM clusters must now be taken into account. To provide a framework for understanding the data, it is convenient to write the dynamical structure function as

$$S(q, \omega) = \chi_0''(\omega) [1 - \exp(-\beta \hbar \omega)]^{-1} F_\omega(q), \quad (1)$$

where  $\int F_\omega(q) d^2q = 1$ . Because of the normalization condition satisfied by  $F_\omega(q)$ ,  $\chi_0''(\omega)$  represents the imaginary part of the local ( $\text{Cu}^{2+}$ ) response function, and  $F_\omega(q)$  the form factor for excitations with energy  $\hbar\omega$ . For classical 2D AFM's such as  $\text{La}_2\text{CuO}_4$  (Ref. 5) and at energies small compared to the zone-boundary spin-wave energies, (a)  $\chi_0''(\omega)$  is  $\omega$  independent and (b)  $F_\omega(q) = (2\pi q_\omega) \delta(|q| - q_\omega)$  is the characteristic function for circles of radius  $q_\omega = \omega/c$ , where  $c$  is the spin-wave velocity. We find that our data for  $\text{La}_{1.95}\text{Ba}_{0.05}\text{CuO}_4$  cannot be described in terms of this simple form for  $F_\omega(q)$ , a result hardly surprising in view of the large width of our constant- $(\hbar\omega = 0)$  scans. However, there are many other forms which can account for the data. Arguably the simplest is the Gaussian,

$$F_\omega(q) = (2\pi\sigma_\omega^2)^{-1} \exp - \frac{1}{2} (|q|^2/\sigma_\omega^2),$$

also used successfully to describe the paramagnetic scattering from the itinerant AFM  $\text{Cr}^6$ . Figure 3(a) shows the frequency dependence of the width parameter  $\sigma_\omega$  obtained from fits to constant- $\hbar\omega$  scans such as those reproduced in Fig. 2. The important result is that between  $\hbar\omega = 0$  and 40 meV,  $\sigma_\omega$  rises with  $\omega$  more rapidly than  $c_0^{-1}$ , where  $\hbar c_0 = 850 \text{ meV \AA}$  is the spin-wave velocity of pure  $\text{La}_2\text{CuO}_4$ .<sup>4</sup> Indeed, the solid line in the figure has a slope of  $1.9 \times 10^{-3} (\text{\AA meV})^{-1} \equiv (0.6c_0)^{-1}$ . Thus, "long"-wavelength (i.e., with  $q \lesssim 0.12 \text{ \AA}^{-1}$ ) disturbances propagate considerably more slowly in  $\text{La}_{1.95}\text{Ba}_{0.05}\text{CuO}_4$  than in  $\text{La}_2\text{CuO}_4$ . For  $\hbar\omega \gtrsim 50$  meV, the errors in  $\sigma_\omega$  are sufficiently large that  $\sigma_\omega$  may depend more weakly on  $\omega$  than indicated by the solid line. In other words, a crossover to the behavior of the pure compound may begin at  $\sim 50$  meV.

To characterize the magnetic dynamics of a material, it is important to specify not only  $F_\omega(q)$ , but also the

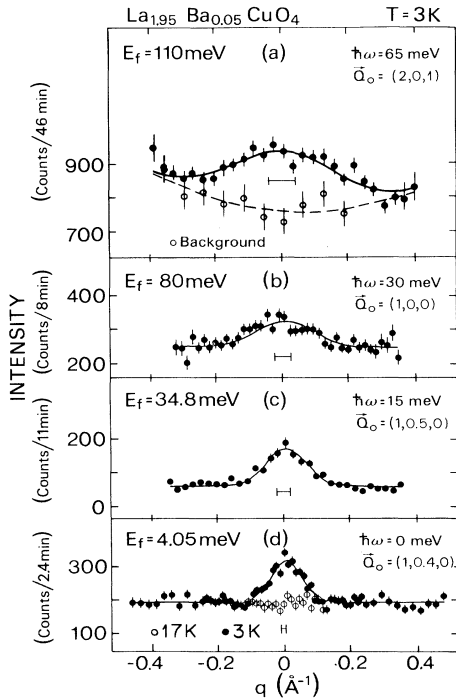


FIG. 2. Constant- $\hbar\omega$  scans through AFM zone centers (100) and (201). Horizontal bars represent the response (FWHM) of the instrument for spin waves of infinite velocity and lifetime.

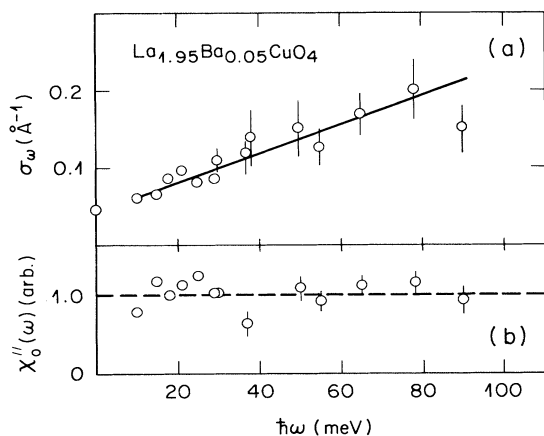


FIG. 3. Dependence of width parameter  $\sigma_\omega$  and amplitude  $\chi''_0(\omega)$  [see Eq. (1)] on  $\hbar\omega$  for  $T=3$  K.

amplitude  $\chi''_0(\omega)$  in Eq. (1). We estimate  $\chi''_0(\omega)$  by fitting the constant- $\hbar\omega$  scans assuming (to reduce scatter) that the straight line in Fig. 3(a) describes  $\sigma_\omega$ . Figure 3(b) shows the outcome, namely, that  $\chi''_0(\omega) \sim \text{const}$ , as in pure  $\text{La}_2\text{CuO}_4$ . Thus, although the spatial extent, characterized by  $F_\omega(q)$ , of the excited-state wave functions is quite different in  $\text{La}_2\text{CuO}_4$  and  $\text{La}_{1.95}\text{Ba}_{0.05}\text{CuO}_4$ , the low-temperature single-site dynamics for  $\hbar\omega \geq 10$  meV, reflected in  $\chi''_0(\omega)$ , of the two compounds are indistinguishable.

On warming, the spin-glass order disappears and the sample acquires metallic transport properties. However, the widths of the constant- $\hbar\omega$  scans do not change discernibly on heating to  $T$  as high as 250 K. For example, the Gaussian widths  $\sigma_\omega$  measured at  $\hbar\omega = 10$  meV and shown in Fig. 1(b) are temperature independent.

While heating does not change  $F_\omega(q)$ , it has a large effect on  $\chi''_0(\omega)$ . Figure 4(a) shows low- $\omega$  spectra obtained at 36 K for fixed momentum transfers at (solid circles) and away from (open circles) the 2D antiferromagnetic zone center. Because of the good energy and momentum resolution of the cold neutron spectrometer used here, and in view of the negligible variation of  $F_\omega(q)$  over the energy range explored, a measure of  $\chi''_0(\omega)$  can be obtained directly from the data by subtracting the background from the signal and then dividing by the statistical factor  $n(\omega)+1 = [1 - \exp(-\beta\hbar\omega)]^{-1}$ . Figures 4(b)–4(d) show the results of this procedure at several temperatures. As required,  $\chi''_0(\omega)$  is an odd function of  $\omega$ . Also, as expected given the results [see Fig. 3(b)] of the higher-energy experiments,  $\chi''_0(\omega)$  at low  $T$  (10 K) is virtually independent of  $\omega$  for  $\omega \gtrsim 0$ . Raising  $T$  affects only the low- $\omega$  portion of  $\chi''_0(\omega)$ : The high- $\omega$  amplitude of  $\chi''_0(\omega)$  appears to be unchanged. Furthermore, there is no obvious contribution of the type associated with a single-pole response function,  $\chi(\omega) = \chi_0\Gamma/(\Gamma - i\omega)$ , where  $\chi''_0(\omega) = \chi_0\Gamma\omega/(\Gamma^2$

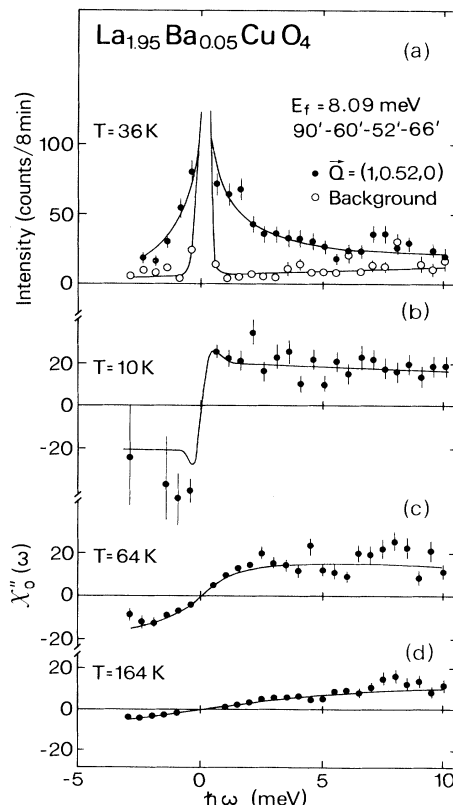


FIG. 4. Constant  $Q=(1,0.52,0)$  scans at various temperatures. Incoherent energy resolution is 0.5 meV (FWHM). (a) Constant- $Q$  scan at (solid circles) and away from (open circles) the 2D antiferromagnetic zone center. (b)–(d) The measured  $\chi''_0(\omega)$  at various temperatures obtained from data as in (a) by the procedure described in the text.

$+\omega^2$ ). The data themselves suggest a better form for the single-site response function,

$$\chi''_0(\omega) = A \tan^{-1} \left[ \frac{\omega}{\Gamma} \right] = A\omega \int_{\Gamma}^{\infty} \frac{d\gamma}{\gamma^2 + \omega^2}. \quad (2)$$

Note that  $\chi''_0(\omega)$  now crosses smoothly from a linear form  $A\omega/\Gamma$  to the constant  $A\pi/2$  at high  $\hbar\omega$ .

Because neither the high- $\omega$  amplitude nor the structure factor  $F_\omega(q)$  are temperature dependent, the problem of the temperature dependence of  $S(q,\omega)$  in  $\text{La}_{1.95}\text{Ba}_{0.05}\text{CuO}_4$  reduces to finding the temperature dependence of a *single* parameter,  $\hbar\Gamma$ . The solid lines in Fig. 4 correspond to fits of the resolution-corrected form (2) to the data, while Fig. 1(c) shows the values  $\hbar\Gamma$  obtained from fits at various  $T$ . For  $T \geq 80$  K,  $\hbar\Gamma$  rises in roughly linear fashion with  $T$ . The constant of proportionality is unusually large, namely,  $\hbar\Gamma \cong \frac{1}{2} k_B T$ . This result is also apparent in Fig. 1(d), which shows the temperature dependence of the dimensionless quantity  $k_B T/\hbar\Gamma$ . A somewhat astonishing finding is that  $k_B T/\hbar\Gamma$  increases

threefold between 88 and 64 K, which is far from  $T_g$  but close to where  $\rho_{ab}$  achieves its minimum. Thus, the loss of metallic conductivity is associated with a large increase in the lifetime of magnetic fluctuations.

Our neutron-scattering data shed considerable light on Cu nuclear spin-lattice relaxation experiments. In the latter, one measures  $T_1^{-1} \sim [n(\omega_0) + 1]\chi_0''(\omega_0)$ , where  $\omega_0$  is an appropriate nuclear resonance frequency, typically of order  $10^7$  Hz. Because both  $\hbar\Gamma$  and  $k_B T \gg \hbar\omega_0$ ,

$$T_1^{-1} \sim \int S(Q, \omega_0) d^2Q \cong \chi_0''(\omega_0) [n(\omega_0) + 1] = A \frac{k_B T}{\hbar\Gamma}. \quad (3)$$

The last quantity in (3) is precisely what is shown in Fig. 1(d). Note that because  $\hbar\Gamma \sim k_B$  for  $T \gtrsim 100$  K, we reproduce the well-known result<sup>7</sup> that  $T_1^{-1}$  is temperature independent for 2-1-4 based materials in this temperature range. Our neutron-scattering experiments, which yield the full  $\omega$  dependence of  $\chi_0''(\omega)$ , thus demonstrate unambiguously that the behavior of the planar Cu  $T_1^{-1}$  (for  $T \gtrsim 100$  K) is due primarily to the linear relation  $\Gamma \sim T$ .<sup>1</sup> The same experiments [Fig. 1(b)] also show that the linear relation is not due to an evolution of  $\xi$ , as in proposals<sup>8</sup> where the scaling hypothesis  $\Gamma \sim \chi \sim \xi^2$  and the Curie-Weiss ansatz  $\chi = (T + \theta)^{-1}$  are used to account for the magnetic-resonance data on layered cuprates.

It is possible to understand the low- $T$  behavior of  $\chi''(q, \omega)$  at small  $\omega$  by thinking of  $\text{La}(\text{Sr}, \text{Ba})_2\text{CuO}_4$  as a system where ferromagnetic (FM) bonds, due to localized holes, are randomly inserted into the otherwise antiferromagnetic Cu-O planes.<sup>9</sup> Therefore, the compound could be considered a two-dimensional  $S = \frac{1}{2}$  Heisenberg spin glass. Unfortunately, little is known about spin glasses of this kind,<sup>10</sup> and we can only hope that the present measurements will motivate calculations of the temperature-dependent parameters appearing in Eq. (2).

For  $T > 50$  K, where the resistance minimum occurs, strong coupling of charge and spin fluctuations is manifested by a dramatic increase of the magnetic fluctuation rate. Further evidence for such coupling is the striking similarity of inelastic light<sup>11</sup> and neutron<sup>2</sup> scattering spectra in the metallic cuprates. Thus, descriptions of the magnetic fluctuations must take the mobility of the holes into account, and the idea that static FM bonds determine the spin dynamics must be abandoned. It is more appropriate to compare our results to theories of the metallic state of the cuprates. Of particular interest is the notion of a marginal Fermi liquid<sup>1</sup> where  $\chi_0''(\omega)$  is

constant for  $\hbar\omega > \hbar\Gamma \sim k_B T$  and linear in  $\hbar\omega$  for  $\hbar\omega < \hbar\Gamma$ . This is, of course, precisely what we find in our experiment.

We thank Risø National Laboratory and the Aspen Center for Physics for their kind hospitality during the time when this work was being completed. Our gratitude is also extended to K. Clausen and M. Nielsen for valuable assistance during the TAS6 experiment, and J. Als-Nielsen and J. Kjems for encouragement and support. Finally, it is a pleasure to thank A. Balatsky, P. Littlewood, A. Millis, I. Morgenstern, D. Pines, and C. Slichter for stimulating discussions, and E. Abrahams and C. M. Varma for useful comments on the manuscript. The U.S. Department of Energy funded work at the Oak Ridge (Contract No. DE-AC05-84OR21400 with Martin Marietta Energy Systems) and Los Alamos National Laboratories.

<sup>(a)</sup>Present address: H. H. Wills Physics Laboratory, Royal Fort, Tyndall Ave., Bristol, BS8 1TL, United Kingdom.

<sup>1</sup>C. M. Varma, P. B. Littlewood, S. Schmitt-Rink, E. Abrahams, and A. E. Ruckenstein, *Phys. Rev. Lett.* **63**, 1996 (1989).

<sup>2</sup>T. R. Thurston *et al.*, *Phys. Rev. B* **40**, 4585 (1989); G. Shirane *et al.*, *Phys. Rev. B* **41**, 6547 (1990); J. Rossat-Mignod *et al.*, in *Proceedings of the Nineteenth International Conference on Low Temperature Physics*, Brighton, 1990 [*Physica (Amsterdam) B* (to be published)].

<sup>3</sup>See, e.g., Y. Maeno *et al.*, in *Proceedings of the Nineteenth International Conference on Low Temperature Physics*, Brighton, 1990 (Ref. 2); B. Ellman *et al.*, *Phys. Rev. B* **39**, 9012 (1989).

<sup>4</sup>D. Harshman *et al.*, *Phys. Rev. B* **38**, 852 (1988); J. Budnick *et al.*, *Europhys. Lett.* **5**, 651 (1988); H. Mook *et al.*, in "Dynamics of Magnetic Fluctuations in High  $T_c$  Materials," edited by G. Reiter, P. Horsh, and G. Psaltakis (Plenum, New York, to be published); B. J. Sternlieb *et al.*, *Phys. Rev. B* **41**, 8866 (1990).

<sup>5</sup>G. Aeppli *et al.*, *Phys. Rev. Lett.* **62**, 2052 (1989); S. M. Hayden *et al.*, *Phys. Rev. B* **42**, 10220 (1990).

<sup>6</sup>B. H. Grier, G. Shirane, and S. A. Werner, *Phys. Rev. B* **31**, 2892 (1985).

<sup>7</sup>K. Ishida, Y. Kitaoka, and K. Asayama, *J. Phys. Soc. Jpn.* **58**, 26 (1989); Y. Kitaoka *et al.*, report, 1990 (to be published).

<sup>8</sup>A. J. Millis, H. Monien, and D. Pines, *Phys. Rev. B* **42**, 167 (1990); B. S. Shastry, *Phys. Rev. Lett.* **63**, 1288 (1989).

<sup>9</sup>A. Aharony *et al.*, *Phys. Rev. Lett.* **60**, 1330 (1988).

<sup>10</sup>I. Morgenstern, *Z. Phys. B* (to be published).

<sup>11</sup>F. Slakey *et al.*, Illinois University, Urbana-Champaign, report, 1990 (to be published).