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Evidence for a generic quantum transition in high-T_c cuprates

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We study the low-energy spin fluctuations and superfluid density of a series of pure and Zn-substituted high-T_c superconductors (HTS) using the muon spin relaxation and ac-susceptibility techniques. At a critical doping state, p_c, we find (i) simultaneous abrupt changes in the magnetic spectrum and in the superconducting ground state and (ii) that the slowing down of spin fluctuations becomes singular at $T=0$. These results provide experimental evidence for a quantum transition that separates the superconducting phase diagram of HTS into two distinct ground states.

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Quantum phase transitions occur at zero temperature at a critical electron density separating distinct ground states. Near a quantum critical point, electrons in metals are highly correlated and the diverging fluctuations may induce unconventional superconductivity.[1–8] For example, in certain heavy fermion compounds a ''bubble'' of superconductivity occurs around the quantum critical point at which itinerant antiferromagnetism is suppressed by applied pressure.[9] The search for an underlying quantum phase transition in high-T_c superconductors (HTS) is motivated by the potential for quantum fluctuations to bind electronic carriers into superconducting Cooper pairs and also to cause the celebrated linear temperature dependence of their electrical resistivity.[1–8,10] HTS exhibit a common generic phase diagram in which the superconducting transition temperature, T_c, rises to a maximum at an optimal doping of approximately 0.16 holes per planar copper atom and then falls to zero on the overdoped side. In addition the underdoped normal state exhibits correlations, which introduce a gap in the density of states that strongly affects all physical properties. There is no phase transition associated with the opening of this gap and so it is called a pseudogap. Analysis of specific heat data, for example, suggests that the pseudogap energy decreases with doping and falls to zero at a critical doping of $p_c \simeq 0.19$, just beyond optimal doping,[10,11] a behavior rather analogous to the quantum-critical heavy-fermion materials.[9]

Many fundamental physical quantities such as the superconducting condensation energy,[10,11] the superfluid density,[12,13] and the quasiparticle weight,[10,14] show abrupt changes as $p \rightarrow p_c$. While compelling in their totality,[10,11] none of the results can be considered as evidence of a quantum transition. In particular there is no evidence for an associated order parameter and slowing down of the relevant fluctuations. With this in mind we examined the evolution with doping of the low-energy spin fluctuation spectrum using muon spin relaxation (μSR) combined with low-field ac-susceptibility measurements of the superfluid density.

The samples studied were: (i) $La_{2-x}Sr_xCu_{1-y}Zn_yO_4$ (LSCO) ($x=0.03$–0.24 and $y=0$, 0.01, and 0.02). Samples were synthesized using solid-state reaction and where necessary followed by quenching and subsequent oxygenation. They were characterized by powder x-ray diffraction as well as extensive transport and thermodynamic measurements, e.g., Refs. 11,12,15 and found to be phase pure. Their T_c values and lattice parameters were also in good agreement with published data, where available.[16,17] (ii) $Bi_{2.1}Sr_{1.9}Ca_{1-x}Y_xCu_2O_{8+y}$ (Bi-2212) ($x=0$, 0.3, 0.5, and appropriate values of y to achieve the desired carrier concentration). Underdoped samples were prepared by deoxygenation and the samples were fully characterized using also thermoelectric power to determine the doping state. (Note that deoxygenation actually removes disorder in this system.) We note that in LSCO $x=p$ and to avoid confusion in the rest of the paper we refer to the carrier concentration as p.

Zero-field (ZF) and transverse-field (TF) μSR studies were performed at the pulsed muon source, ISIS Facility, Rutherford Appleton Laboratory. Spectra were collected down to as low as 40 mK thus allowing the temperature dependence of slow spin fluctuations to be studied to high doping. In a μSR experiment, 100% spin-polarized positive muons implanted into a specimen precess in their local magnetic environment. Random spin fluctuations will depolarize the muons provided they do not fluctuate much faster than the muon precession. The muon decays with a life time 2.2 μs, emitting a positron preferentially in the direction of the muon spin at the time of decay. By accumulating time histograms of such positrons one may deduce the muon depolarization rate as a function of time after implantation. The muon is expected to reside at the most electronegative site of the lattice. In both HTS families studied here it is the O^{2-} nearest to the planes[18] so the results reported here are dominated by the magnetic correlations in the CuO_2 planes. As we show below, this is confirmed by results in samples doped with Zn, which substitutes for Cu in the CuO_2 planes.

The superfluid density, λ_{ab}^{-2}, results shown here are for pure $La_{2-x}Sr_xCuO_4$ ($y=0$) and were determined from measurements of the in-plane magnetic penetration depth λ_{ab} using the low-field ac-susceptibility technique at an ac field of 1 G rms (parallel to the c axis) and a frequency 333 Hz for

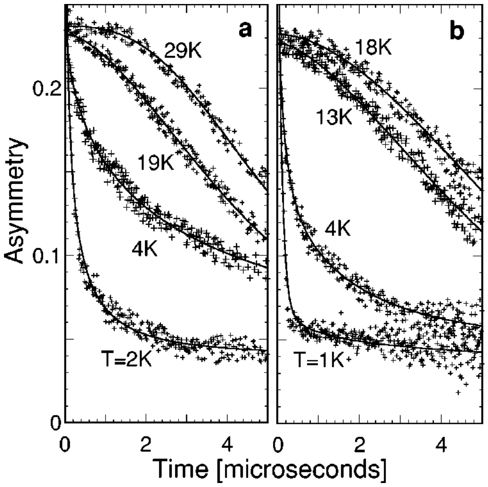

FIG. 1. Typical zero-field μSR spectra as a function of temperature of (a) pure $La_{2-x}Sr_xCuO_4$ with $x=p=0.08$ and (b) $La_{2-x}Sr_xCu_{1-y}Zn_yO_4$ with $x=p=0.15$ and $y=0.01$. The solid lines are the fits discussed in the text.

grain-aligned powders.[12,15,19] In total, 16 samples were investigated for each doping content. We note that grain agglomerates can be a cause of poor alignment, and to eliminate these, powders were ball milled in ethanol and dried after adding a deflocculant. Scanning electron microscopy confirmed the absence of grain boundaries and showed that the grains were approximately spherical. The powders were mixed with a 5-min curing epoxy and aligned in a static field of 12 T at room temperature. Debye-Scherrer x-ray scans showed that approximately 90% of the grains had their CuO_2 planes aligned to within approximately 2°. The values of $\lambda_{ab}(0)^{-2}$ for samples with $p\geqslant 0.15$ were also confirmed by standard TF μSR experiments performed on unaligned powders at 400 G.[12,13]

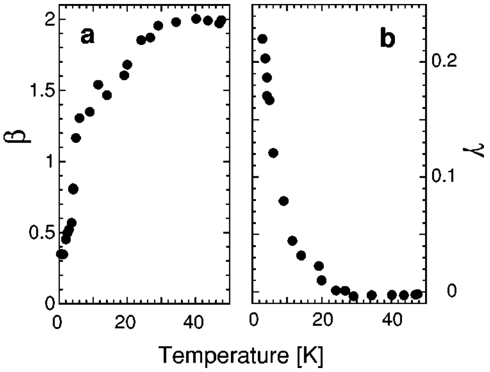

FIG. 2. The temperature dependence of (a) the (stretched-exponential) exponent β and (b) the relaxation rate, γ, within the Kubo–Toyabe function obtained by fitting muon depolarization data for $La_{2-x}Sr_xCuO_4$ ($Sr=0.08$).

Figure 1 shows the typical time dependence at several temperatures of the ZF muon asymmetry[20–23] for (a) pure $La_{2-x}Sr_xCuO_4$ with $x=p=0.08$ and (b) $La_{2-x}Sr_xCu_{1-y}Zn_yO_4$ with $x=p=0.15$ and $y=0.01$. In all samples the high-temperature form of the depolarization is Gaussian and temperature independent, consistent with dipolar interactions between the muons and their near-neighbor nuclear moments. This was verified by applying a 50-G longitudinal field, which completely suppressed the depolarisation. Here the electronic spins in the CuO_2 planes fluctuate so fast that they do not affect the muon polarization. At low enough temperatures, typical of other spin glass systems,[18,20–25] there is a fast relaxation due to a static distribution of random local fields, followed by a long-time tail with a slower relaxation resulting from remnant dynamical processes within the spin glass. By decoupling experiments in a longitudinal field we also confirmed the static nature of the magnetic ground state and at very low temperatures oscillations in the asymmetry were observed for $p\leqslant 0.08$. For $p>0.08$ oscillations were not observed and, as discussed below, the data were better represented by an exponential relaxation indicating either a very strongly disordered static field distribution or rapid fluctuations. Between the high and low temperature limits the spin correlations slow down through the experimental μSR time window and modify the depolarization process in a distinctive fashion.[22–25]

To study the doping dependence of this slowing down we determine two characteristic temperatures. (i) The temperature, T_f, where the spin correlations first enter the μSR time window, i.e., where the muon asymmetry first deviates from Gaussian behavior and (ii) the temperature, T_g, at which these correlations freeze into a glassy state thus causing an initial rapid decay in the asymmetry. μSR is sensitive to spin fluctuations within a time window of 10^{-9}–10^{-6}s (Ref. 22) and we may therefore associate T_f and T_g, respectively, with these lower and upper thresholds. In general the relaxation data (Fig. 1) may be fitted to the form $G_z(t)=A_1\exp(-\gamma_1 t)+A_2\exp(-(\gamma_2 t)^{\beta})+A_3$ where the first term is the fast relaxation in the glassy state (i.e., at higher temperatures $A_1=0$), the second stretched-exponential term is the slower dynamical term, and A_3 accounts for a small time-independent background arising from muons stopping in the silver backing plate. As in some other spin glass systems, in the high-temperature Gaussian limit $\beta=2.0$.[18,22,23] Consequently, any departure below $\beta=2.0\pm 0.06$ is taken as the onset temperature, T_f, at which spin fluctuations slow down sufficiently to enter the time scale of the muon probe (10^{-9} s). A typical temperature dependence for β for $Sr=0.08$ is shown in Fig. 2(a).[23] [As a further check on the assignment of T_f we fitted the high-temperature data to the full Kubo–Toyabe function $G_z(t)=A_1\exp(-\alpha t^2)\exp(-\gamma t)+A_2$ and values of the relaxation rate γ are plotted in Fig. 2(b). As expected γ is found to rise from zero at the same temperature at which the exponent β departs from 2, indicating the entrance of the spin correlations into the experimental time window.] At low temperatures the exponent β falls rapidly toward the value 0.5. We identify T_g as the temperature at which $\beta=0.5\pm 0.06$.[18,22,23] This "root exponential" form for the relaxation function is a common feature of spin glasses, and in the present samples the temperature T_g coincided with a maximum in the longitudinal relaxation rate γ [see Fig. 2(b)] and the appearance of the fast relaxation.[21–24] We have also tested other methods of analyzing the data as well as a different choice of criteria, for example choosing $\beta=0.3$ instead of 0.5, to identify T_g.[24] Different approaches were

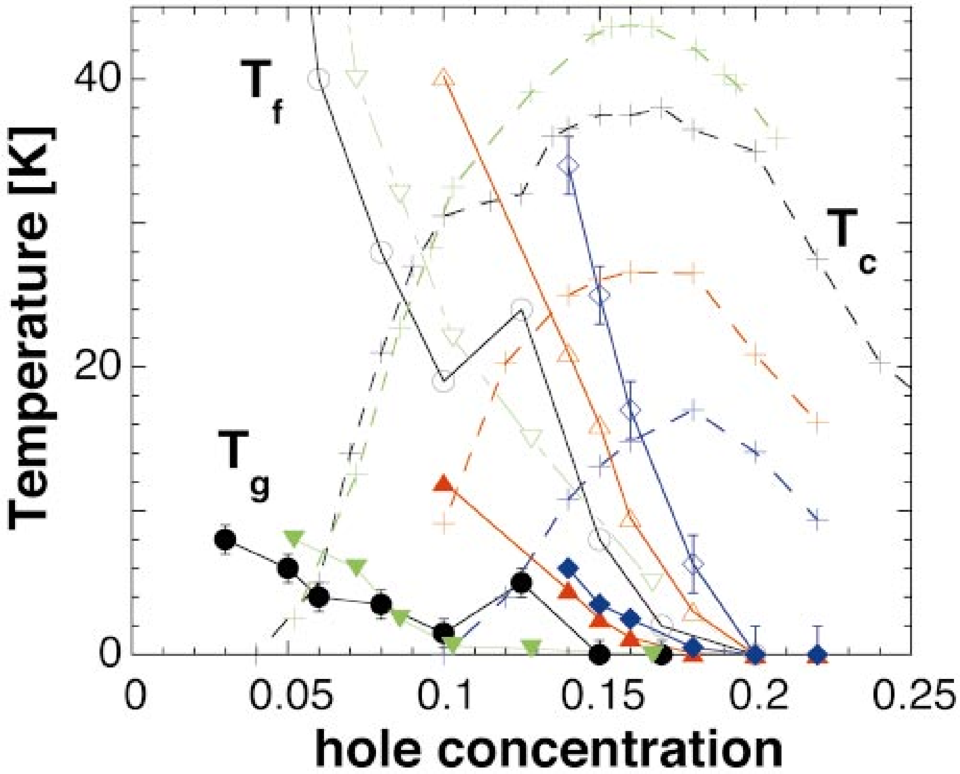

FIG. 3. (Color) The doping dependence of the crossover temperatures T_f (open symbols) where the spin fluctuations enter the μSR time window and T_g (closed symbols) where the spin fluctuations leave the μSR time window. Below T_g the fluctuations freeze out into a spin glass. Black, red, and blue data are for $La_{2-x}Sr_xCu_{1-y}Zn_yO_4$ with $y=0$, $y=0.01$, and $y=0.02$, respectively. The green symbols are T_f (open) and T_g (closed) values, determined in the same way, for $Bi_{2.1}Sr_{1.9}Ca_{1-x}Y_xCu_2O_{8+y}$. Typical error bars for T_f and T_g are shown for two of the sample series. The T_c values for all samples are shown as crosses in the respective colors. T_c's for Bi-2212 have been divided by 2 for clarity. Solid lines are drawn as a guide to the eye.

found to affect slightly the magnitude of T_g and T_f but not the trends with doping. We note that our values for T_g agree with published data obtained from different techniques, where available.[17,21,26–30]

We first discuss the data for pure LSCO (i.e., $y=0$ in Fig. 3). Values of T_g and T_f summarized in Fig. 3 indicate that the spin-glass phase persists beyond $p=0.125$. In fact the onset of the spin glass phase for $p=0.125$ occurs at a higher temperature than that for $p=0.10$. This may be due to the formation of strongly correlated antiferromagnetic stripe domains in this range of doping.[17,31–33] For $p=0.15$ and 0.17, T_g becomes very small (less than 45 mK) and T_f is approximately 8 and 2 K, respectively. For $p \geqslant 0.20$, there are no changes in the depolarization function to the lowest temperature measured (40 mK).

Figure 3 clearly shows that although the freezing of spins occurs at very low temperatures, low-frequency spin correlations enter the experimental time window at significantly higher temperatures. Both T_g and T_f are found to decrease with increasing doping and tend to zero at $p \simeq 0.19$. Their behavior resembles that of the pseudogap[10,11] which vanishes at the same doping. The LSCO results are reproduced in the Bi-2212 system (Fig. 3), which shows precisely the same trend with T_g and $T_f \rightarrow 0$ as $p \rightarrow 0.19$. We note that this similarity does not rule out possible effects of striped phases in our LSCO data. We also know from a couple of data points[34] that the $YBa_2Cu_3O_{7-\delta}$ system shows similar trends as LSCO and Bi-2212. Similar slowing down of spin fluctuations has also been observed in the most ordered of all underdoped HTS namely the $YBa_2Cu_4O_8$.[34] Furthermore, the doping dependence of T_g seen here has been found in $Y_{1-y}Ca_yBa_2Cu_3O_{6.02}$ up to 0.09 holes per planar copper atom.[21] These observations indicate that the behavior shown in Fig. 3 is common to all HTS. They also indicate that the observed trends are not a consequence of a structural transition or inhomogeneity peculiar to a specific HTS family.

Earlier spectroscopic studies have shown that substitution with Zn slows down the spin correlations.[17,35,36] This, as depicted in Fig. 3, enhances the muon depolarization rate at low temperatures and causes an increase in both T_g and T_f. The striking result which Fig. 3 summarizes is the apparent convergence of both $T_g(p)$ and $T_f(p)$ to zero, for all Zn concentrations, at the critical doping $p_c \simeq 0.19$. Although we do not have data for exactly $p=0.19$, the presence of finite values of T_g and T_f for $p=0.18$ but their absence for 0.20, indicates that the two magnetic scales go to zero somewhere in between and in particular very near 0.19. In fact this was confirmed by fitting (not shown) the data in Fig. 3 to the function $T(1\text{-}p/0.19)^n$. Therefore, the data are consistent with a scenario in which T_g and T_f decay away to zero at $p=p_c=0.19$. While this effect is not so obvious for the pure LSCO samples it is very clear in the two Zn-substituted series. The fact that $T_f(p) \rightarrow 0$ as $p \rightarrow p_c$ for all Zn concentrations suggests that spin correlations within the upper μSR time threshold of 10^{-9} s die out abruptly beyond p_c leaving only short-lived fluctuations (or none at all) beyond p_c. The fact that T_f and T_g both vanish as $p \rightarrow p_c$ implies that the rate of slowing down diverges at p_c, in the sense that the characteristic time changes from 10^{-9} to 10^{-6} s in smaller and smaller temperature intervals as p_c is approached. In the absence of evidence for long-range order in the normal state, the present observations indicate the existence of a *quantum glass transition* at p_c. We note that if the quantum glass transition is a conventional spin glass transition, then the

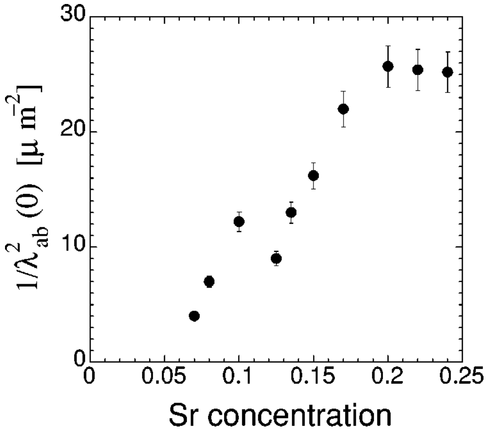

FIG. 4. Doping dependence of the inverse square of the zero temperature in-plane penetration depth for pure $(y=0)$ $La_{2-x}Sr_xCuO_4$ measured by the ac-susceptibility technique.

glass transition at $T=0$ is a quantum critical point.[37] Of course the present behavior could alternatively be driven by the existence of a quantum metal-insulator transition as has been observed in $La_{2-x}Sr_xCuO_4$ near $Sr=0.18$.[38] For $p > p_c$ spin-flip scattering associated with mobile holes could reduce the lifetime of the spins sufficiently to prevent freezing. Either way, the present results demonstrate the disappearance of short-range magnetic order at p_c and a clear link between a quantum transition, the essential physics of superconductivity, and the pseudogap in HTS.

This link is further underscored by our detailed measurements of the doping dependence of $\lambda_{ab}^{-2}(0)$ for pure LSCO shown in Fig. 4. The superfluid density remains constant above p_c but falls abruptly below p_c. A similar doping dependence of the superfluid density has also been observed in Bi-2212.[39] Abrupt changes in the doping dependence of the superfluid density at p_c were also reported for $Y_{0.8}Ca_{0.2}Ba_2Cu_3O_{7-\delta}$ and $Tl_{0.5-y}Pb_{0.5+y}Sr_2Ca_{1-x}Y_xCu_2O_7$[40]. This confirms again that this phase behavior is generic to HTS. Note that the earlier systematic studies of Uemura *et al.*[13] were reported in plots of T_c versus $\lambda_{ab}^{-2}(0)$ which tend to conceal these important changes. In addition, recent penetration depth measurements in LSCO and $HgBa_2CuO_{4+\delta}$ showed that the *c*-axis penetration depth $\lambda_c^{-2}(0)$ exhibits similar behavior.[15] The apparent competition between quasistatic magnetic correlations and superconductivity thus results in a crossover to weak superconductivity characterized by a strong suppression of the superfluid density. This suppression in the underdoped region can also be directly linked to the strong reduction in entropy and condensation energy associated with the pseudogap.[11] It is therefore evident here that the onset of short-range magnetic correlations at the critical point p_c coincides with a change in the superconducting ground state properties in HTS. It separates the phase diagram into two distinct regions: (i) below p_c where T_g, T_f increase rapidly with underdoping and the superfluid density is rapidly suppressed, and (ii) above p_c where T_g, $T_f \rightarrow 0$ and the superfluid density is almost constant, indicating a transition from weak to strong superconductivity, respectively. Moreover, it is at p_c where other fundamental properties such as the superconducting condensation energy and quasiparticle lifetime, change abruptly, the resistivity follows its unusual linear temperature dependence to the lowest temperature and the pseudogap extrapolates to zero.[10,11] These features all indicate that the quantum transition identified here is connected with the fundamental physical properties of HTS.

In general terms our results complement a growing body of work pointing to an intimate relation between slow magnetic correlations and superconductivity that seems mutually co-operative in some experiments[36,41,42] and competitive in others (present work and Ref. 26). In the superconducting state an energy gap is observed in the spin spectrum[43] but its anisotropy in **k** space (approximately *d*-wave like) ensures that low energy spin fluctuations may still be present, as observed here. Inelastic neutron scattering[17,36] and nuclear magnetic resonance[35] experiments show that Zn substitution slows spin fluctuations and suppresses long-range order. They are consistent with our observation that Zn doping enhances spin glass behavior. These studies provide a context for the present work but the key new result here is the experimental observation of the disappearance of short-range magnetic correlations at a critical doping suggesting the presence of a quantum transition with an associated change in the superconducting ground state. It follows from our work that the elusive physics of HTS may reduce to the known generic physics of materials near a quantum transition, thus explaining many of their unconventional properties.[1–8]

In conclusion we have performed a comprehensive study of low-energy spin fluctuations and of the superfluid density in several HTS. We found that both low-energy spin fluctuations and the spin-glass state disappear at a critical doping p_c at zero temperature. This provides evidence for a *quantum glass transition* in HTS and indicates quantum critical fluctuations and associated dynamical crossovers may dominate the essential physics of high temperature superconductivity. The identified critical point shows that there are two distinct ground states in the superconducting phase, manifested in a crossover from strong superconductivity for $p > p_c$ to weak superconductivity for $p < p_c$. The identified transition bears a close resemblance to the locally critical two-dimensional quantum phase transitions[2,8] in which the magnetic correlations are localized in space but have unlimited range in time.

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[1] S. Chakravarty, B.I. Halperin, and D.R. Nelson, Phys. Rev. Lett. **60**, 1057 (1988); S. Chakravarty, R.B. Laughlin, D.K. Morr, and C. Nayak, Phys. Rev. B **63**, 094503 (2001).

[2] C.M. Varma, P.B. Littlewood, S. Schmittrink, E. Abrahams, and A.E. Ruckenstein, Phys. Rev. Lett. **63**, 1996 (1989); C.M. Varma, in *Strongly Correlated Electronic Materials*, edited by K.S. Bedell (Addison-Wesley, Reading, MA, 1994); C.M. Varma, Phys. Rev. B **55**, 14 554 (1997); Phys. Rev. Lett. **83**, 3538 (1999).

[3] S. Sachdev and J. Ye, Phys. Rev. Lett. **69**, 2411 (1992); S. Sachdev, Science **288**, 475 (2000).

[4] C. Castellani, C. DiCastro, and M. Grilli, Phys. Rev. Lett. **75**, 4650 (1995).

[5] D. Pines, Physica C **341**, 59 (2000).

[6] A. Abanov, A.V. Chubukov, and J. Schmalian, Europhys. Lett. **55**, 369 (2001).

[7] A.V. Chubukov, D. Pines, and J. Schmalian, Review Chapter to appear in *The Physics of Conventional and Unconventional Superconductors*, edited by K.H. Bennemann and J.B. Ketterson (Springer, Berlin, in press); A.V. Chubukov, D. Pines, and J. Schmalian, cond-mat/0201140 (unpublished).

[8] Q. Si, S. Rabello, K. Ingersent, and J. Smith Lleweilun, Nature (London) **413**, 804 (2001).

[9] N.D. Mathur, F.M. Grosche, S.R. Julian, I.R. Walker, D.M. Freye, R.K.W. Haselwimmer, and G.G. Lonzarich, Nature (London) **394**, 39 (1998).

[10] J.L. Tallon and J.W. Loram, Physica C **349**, 53 (2001).

[11] J.W. Loram, K.A. Mirza, J.R. Cooper, and J. L Tallon, J. Phys. Chem. Solids **59**, 2091 (1998).

[12] C. Panagopoulos, B.D. Rainford, J.R. Cooper, W. Lo, J.L. Tallon, J.W. Loram, J. Betouras, Y.S. Wang, and C.W. Chu, Phys. Rev. B **60**, 14 617 (1999).

[13] Y.J. Uemura *et al.*, Phys. Rev. Lett. **62**, 2317 (1989).

[14] D.L. Feng *et al.*, Science **289**, 277 (2000).

[15] C. Panagopoulos, J.R. Cooper, T. Xiang, Y.S. Wang, and C.W. Chu, Phys. Rev. B **61**, R3808 (2000).

[16] P.G. Radaelli, D.G. Hinks, A.W. Mitchell, B.A. Hunter, J.L. Wagner, B. Dabrowski, K.G. Vandervoort, H.K. Viswanathan, and J.D. Jorgensen, Phys. Rev. B **49**, 4163 (1994).

[17] M.A. Kastner, R.J. Birgeneau, G. Shirane, and Y. Endoh, Rev. Mod. Phys. **70**, 897 (1998).

[18] B. Nachumi *et al.*, Phys. Rev. B **58**, 8760 (1998).

[19] C. Panagopoulos, J.R. Cooper, T. Xiang, G.B. Peacock, I. Gameson, and P.P. Edwards, Phys. Rev. Lett. **79**, 2320 (1997).

[20] R.F. Kiefl *et al.*, Phys. Rev. Lett. **63**, 2136 (1989).

[21] Ch. Niedermayer, C. Bernhard, T. Blasius, A. Golnik, A. Moodenbaugh, and J.I. Budnick, Phys. Rev. Lett. **80**, 3843 (1998).

[22] Y.J. Uemura, T. Yamazaki, D.R. Harshman, M. Senba, and E.J. Ansaldo, Phys. Rev. B **31**, 546 (1985).

[23] R. Cywinski and B.D. Rainford, Hyperfine Interact. **85**, 215 (1994).

[24] I.A. Campbell, A. Amato, F.N. Gyrax, D. Herlach, A. Schenck, R. Cywinski, and S.H. Kilcoyne, Phys. Rev. Lett. **72**, 1291 (1994).

[25] A. Kanigel, A. Keren, Y. Eckstein, A. Knizhnik, J.S. Lord, and A. Amato, Phys. Rev. Lett. **88**, 137003 (2002).

[26] P.M. Singer and T. Imai, Phys. Rev. Lett. **88**, 187601 (2002).

[27] D.R. Harshman *et al.*, Phys. Rev. B **38**, 852 (1988).

[28] J.I. Budnick, B. Chamberland, D.P. Yang, C. Niedermayer, A. Golnik, E. Recknagel, M. Rossmanith, and A. Wedinger, Europhys. Lett. **5**, 65 (1988).

[29] F.C. Chou, N.R. Belk, M.A. Kastner, R.J. Birgeneau, and A. Aharony, Phys. Rev. Lett. **75**, 2204 (1995).

[30] S. Wakimoto, S. Ueki, Y. Endoh, and K. Yamada, Phys. Rev. B **62**, 3547 (2000).

[31] V.J. Emery and S.A. Kivelson, J. Phys. Chem. Solids **59**, 1705 (1998).

[32] J. Zaanen, J. Phys. Chem. Solids **59**, 1769 (1998).

[33] C.M. Smith, A.H. Castro Neto, and A.V. Balatsky, Phys. Rev. Lett. **87**, 177010 (2001).

[34] C. Panagopoulos, J.L. Tallon, C.A. Scott, and B.D. Rainford (unpublished).

[35] M.-H. Julien, T. Feher, M. Horvatic, C. Berthier, O.N. Bakharev, P. Segransan, G. Collin, and J.F. Marucco, Phys. Rev. Lett. **84**, 3422 (2000).

[36] H. Kimura *et al.*, Phys. Rev. B **59**, 6517 (1999).

[37] S. Sachdev, *Quantum Phase Transitions* (Cambridge University Press, Cambridge, 1999), Chap. 16.

[38] G.S. Boebinger, Y. Ando, A. Passner, T. Kimura, M. Okuya, J. Shimoyama, K. Kishio, K. Tamasaku, N. Ichikawa, and S. Uchida, Phys. Rev. Lett. **77**, 5417 (1996).

[39] W. Anukool, Cambridge University, Ph.D. thesis, 2002.

[40] C. Bernhard, J.L. Tallon, T. Blasius, A. Golnik, and C. Niedermayer, Phys. Rev. Lett. **86**, 1614 (2001).

[41] M. Fujita, K. Yamada, H. Hiraka, P.M. Gehring, S.H. Lee, S. Wakimoto, and G. Shirane, Phys. Rev. B **65**, 064505 (2002).

[42] B. Lake, H.M. Ronnow, N.B. Christensen, G. Aeppli, K. Lefmann, D.F. McMorrow, P. Vorderwisch, P. Smeibidl, N. Mangkorntong, T. Sasagawa, M. Nohara, H. Takagi, and T.E. Mason, Nature (London) **415**, 299 (2002).

[43] K. Yamada *et al.*, Phys. Rev. Lett. **75**, 1626 (1995).