Commensurate Dynamic Magnetic Correlations in $La_2Cu_{0.9}Li_{0.1}O_4$

Wei Bao,[1] R. J. McQueeney,[1] R. Heffner,[1] J. L. Sarrao,[1] P. Dai,[2] and J. L. Zarestky[3]
[1]*Los Alamos National Laboratory, Los Alamos, New Mexico 87545*
[2]*Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831*
[3]*Ames Laboratory, Ames, Iowa 50011*
(Received 17 September 1999)

When sufficient numbers of holes are introduced into the two-dimensional CuO_2 square lattice, dynamic magnetic correlations become *in*commensurate with underlying lattice in all previously investigated $La_{2-x}A_xCu_{1-z}B_zO_{4+y}$ (A = Sr or Nd, B = Zn) including high T_c superconductors and insulators, and in bilayered superconducting $YBa_2Cu_3O_{6.6}$ and $Bi_2Sr_2CaCu_2O_8$. Magnetic correlations also become *in*commensurate in structurally related La_2NiO_4 when doped with Sr or O. We report an exception to this so-far well-established experimental "rule" in $La_2Cu_{1-z}Li_zO_4$ in which magnetic correlations remain commensurate.

PACS numbers: 75.40.Gb, 74.72.Dn, 75.25.+z, 75.50.Ee

High transition temperature (T_c) superconductivity is realized when charge carriers are introduced to the CuO_2 planes of the insulating parent compound, for example, La_2CuO_4 or $YBa_2Cu_3O_6$. These parent compounds are now well established as two-dimensional (2D) spin $S = \frac{1}{2}$ Heisenberg antiferromagnets with a dominant in-plane exchange interaction [1–4]. The evolution of the magnetic correlations with charge carrier doping is a central issue in high T_c superconductivity research. It has so far been most extensively investigated in the La_2CuO_4 system, due to the availability of large single crystals. These studies show that, when the long-range antiferromagnetic order is suppressed and the doped hole concentration exceeds about 5%, incommensurate dynamic magnetic correlations develop at a quartet of wave vectors $\mathbf{Q} = (\frac{1}{2} \pm \delta, \frac{1}{2}, 0)$ and $(\frac{1}{2}, \frac{1}{2} \pm \delta, 0)$ [5–7]. Even more remarkable is that the incommensurability δ is a universally increasing function of the hole concentration n [8], whether the doped material is superconducting or insulating [9] and whether dopant resides on the La [10,11], the Cu [12], or the oxygen sites [13]. In the isostructural insulating nickelates, the magnetic correlations are also found to be incommensurate when a sufficient number of holes are introduced by doping either the La or the O sites [14]. Recently, incommensurate magnetic correlations were also discovered in bilayered superconducting $YBa_2Cu_3O_{6.6}$ [15]. The incommensurability δ vs n falls on the same curve as the La_2CuO_4 system, adding new excitement to this field. There is also experimental evidence indicating incommensurate magnetic correlations in superconducting $Bi_2Sr_2CaCu_2O_8$ [16]. Empirically, therefore, there is an overwhelming consensus that doping the 2D antiferromagnet on the CuO_2 square lattice eventually makes magnetic correlations incommensurate. There are currently several competing theoretical explanations for the origin of this incommensurability, ranging from stripe models [17–20], where charge carriers undergo phase separation, to nesting Fermi surface models [21] and doped quantum antiferromagnet models [22], where the carriers remain uniform in the sample.

$La_2Cu_{1-z}Li_zO_4$ remains an insulator for $0 \le z \le 0.5$. It has identical in-plane lattice parameters as Sr-doped La_2CuO_4 at the same hole concentration [23]. The long-range antiferromagnetic order is similarly suppressed by Sr or Li doping [23,24]. The spin dynamics of $La_2Cu_{1-z}Li_zO_4$, as revealed by *local* dynamical probes such as nuclear quadrapole resonance, show an astonishing parallel with $La_{2-z}Sr_xCuO_4$ [25]. This suggests a similar temperature dependence in the low energy spin fluctuations. In contrast to the enormous empirical conformity among charge-doped laminar cuprates, however, as will be presented below, $La_2Cu_{1-z}Li_zO_4$ is exceptional in that the dynamic magnetic correlations remain commensurate with the square lattice. This experimental result thus provides a new facet of the rich physics relating antiferromagnetism and charge correlations in charge-doped cuprates.

Single crystals of $La_2Cu_{1-z}Li_zO_4$ were grown in CuO flux, using isotopically enriched ^{7}Li (98.4%) to reduce neutron absorption. The size of single crystals grown in this way decreases with increasing z. We choose $z = 0.10(2)$ for this work to balance the sample size with the detectability of δ, keeping in mind that δ increases with hole concentration in all other doped La_2CuO_4 materials. Magnetization measurements show no long-range magnetic order, consistent with previous studies [23,26]. The sample has orthorhombic *Cmca* symmetry (space group No. 64) at low temperatures. In this paper, it is sufficient to use a simpler tetragonal unit cell ($a^* = 1.174$ Å^{-1} and $c^* = 0.4814$ Å^{-1} at 15 K) to label the reciprocal space; thus the $(\frac{1}{2}, \frac{1}{2}, 0)$ corresponds to the (π, π) square-lattice antiferromagnetic wave vector. A single crystal of 0.46 g with mosaic $<0.5°$ was used in the $\mathbf{Q}$ scans. The energy scans in Fig. 3(c) were taken with five aligned crystals of total mass 1.0 g and mosaic of 0.9°. Neutron scattering experiments were performed at the HB1A and HB1 triple-axis spectrometers at the high flux isotope reactor of ORNL. The samples were mounted to the cold finger of a Displex refrigerator both in the $(h, k, 0)$ and (h, h, l)

 0031-9007/00/84(17)/3978(4)$15.00

scattering planes. The spectrometer configurations are specified in the figures.

The intensity of neutron scattering was measured against a neutron flux monitor placed between the sample and the exit collimator of the monochromator. For magnetic scattering, this intensity directly measures the dynamic magnetic correlation function $S(\mathbf{Q}, \omega)$ [27],

$$I \propto |F(\mathbf{Q})|^2 \cdot \overline{S}(\mathbf{Q}, \omega), \tag{1}$$

where $|F(\mathbf{Q})|^2$ is the magnetic form factor, and $\overline{S}(\mathbf{Q}, \omega)$ is the convolution of $S(\mathbf{Q}, \omega)$ with the spectrometer resolution function. Factoring out the thermal occupation factor, the imaginary part of the generalized dynamic magnetic susceptibility is given by

$$\chi''(\mathbf{Q}, \omega) = (1 - e^{-\hbar\omega/k_B T}) S(\mathbf{Q}, \omega). \tag{2}$$

$\chi''(\mathbf{Q}, \omega)$ is a useful quantity for comparing the magnetic response at different temperatures. Dynamic magnetic fluctuations can be approximated by a Lorentzian model,

$$S(\mathbf{Q}, \omega) = \frac{\hbar\omega}{1 - e^{-\hbar\omega/k_B T}} \frac{\chi_{\mathbf{Q}} \Gamma_{\mathbf{Q}}}{(\hbar\omega)^2 + \Gamma_{\mathbf{Q}}^2}, \tag{3}$$

where $\Gamma_{\mathbf{Q}}$ is the energy scale for magnetic fluctuations and $\chi_{\mathbf{Q}}$ is the $\mathbf{Q}$-dependent magnetic susceptibility which determines the intensity. The maximum of $S(\mathbf{Q}, \omega)$ is at $\omega = 0$, which is the contribution to *quasi* elastic scattering from dynamic magnetic fluctuations:

$$S(\mathbf{Q}, 0) = k_B T \frac{\chi_{\mathbf{Q}}}{\Gamma_{\mathbf{Q}}}. \tag{4}$$

The magnetic form factor $|F(\mathbf{Q})|^2$ in Eq. (1) has its maximum at $\mathbf{Q} = 0$ while the intensity from structural excitations grows as Q^2. This fact is often used in inelastic neutron scattering experiments to distinguish magnetic and structural excitations.

The 2D reciprocal space for the CuO_2 planes near the (π, π) point is shown in the inset of Fig. 1(a). The crosses schematically denote incommensurate wave vectors for the low energy spin fluctuations previously found in sufficiently doped La_2CuO_4 and $YBa_2Cu_3O_{6.6}$. A constant energy (const-E) scan along the k direction with $\hbar\omega = 0$ is shown by open circles in Fig. 1(a). The arrow indicates the incommensurate point where a quasielastic peak in the low energy magnetic fluctuations is found in other doped cuprates with 10% hole concentration. Apparently, $La_2Cu_{0.9}Li_{0.1}O_4$ is different from its peers. The only discernible peak is at the commensurate $(\frac{1}{2}, \frac{1}{2}, 0)$, i.e., the (π, π) point. An extended search along the $(h, h, 0)$ direction has also been conducted with a similar result.

The intensity of the superlattice peak at $(\frac{1}{2}, \frac{1}{2}, 0)$ is only 5×10^{-4} of the intensity of the structural Bragg peak (110). By adding more filters, it can be shown that the intensity at $(\frac{1}{2}, \frac{1}{2}, 0)$ is not due to higher-order neutron contamination. The intensity is also insensitive to neutron energy change from 13.5 to 14.8 meV. However, the dominant contribution to this weak superlattice peak is not of magnetic nature, based on its temperature dependence [refer to Fig. 1(b)] and $\mathbf{Q}$ dependence in other Brillouin zones. Further investigation as to its origin is under way.

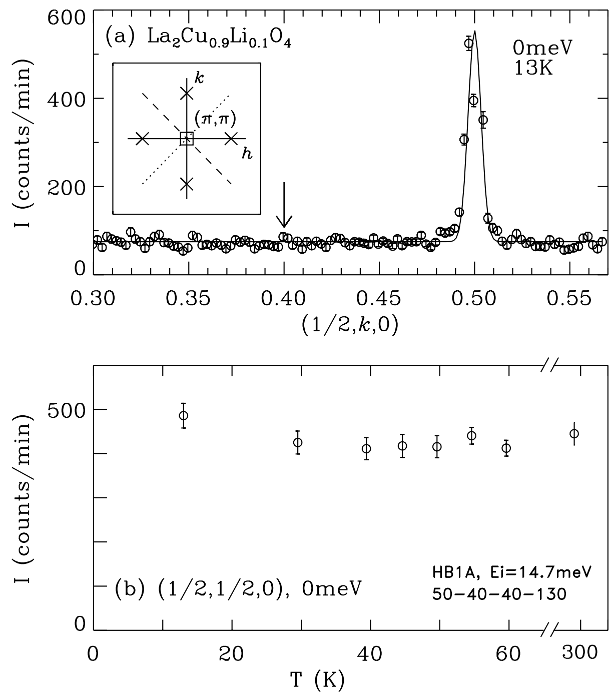

FIG. 1. The inset in (a) shows the 2D in-plane (h, k) reciprocal space. The square marks the commensurate (π, π) point which characterizes dynamic magnetic correlations in $La_2Cu_{0.9}Li_{0.1}O_4$. The crosses mark the quartet of incommensurate wave vectors found for magnetic correlations in other doped cuprates [5–8,10–13,15]. (a) Scans across the (π, π) point along the k direction at $E = 0$. The arrow indicates the incommensurate peak position found in other cuprates of identical hole concentration. (b) Temperature dependence of the $(\frac{1}{2}, \frac{1}{2}, 0)$ peak intensity.

To detect the dynamic magnetic correlations, we have repeated the k scan at a finite energy, $\hbar\omega = 1.8$ meV, that avoids the elastic superstructure contribution. Results are shown in Fig. 2(a). The const-E scan measures a peak at $\mathbf{Q} = (\frac{1}{2}, \frac{1}{2}, 0)$ in the dynamic magnetic correlation function $S(\mathbf{Q}, \omega)$. Scans along two other symmetrically inequivalent directions are shown in Figs. 2(b) and 2(c), further supporting the conclusion that the magnetic correlations are commensurate. Using measured phonon intensity at a similar energy and temperature near (110) (8 counts per minute), the Q^2 scaling factor in the phonon scattering cross section, and the Bragg intensity ratio between the $(\frac{1}{2}, \frac{1}{2}, 0)$ and (110), the estimated acoustic phonon contribution near $(\frac{1}{2}, \frac{1}{2}, 0)$ is negligible.

In the left frames of Fig. 3, the dynamic magnetic correlations are compared at 14 and 295 K in an identical const-E scan. The data have been converted to χ'', using Eq. (2). The shorter correlation length of the dynamic magnetic correlations at the higher temperature is reflected

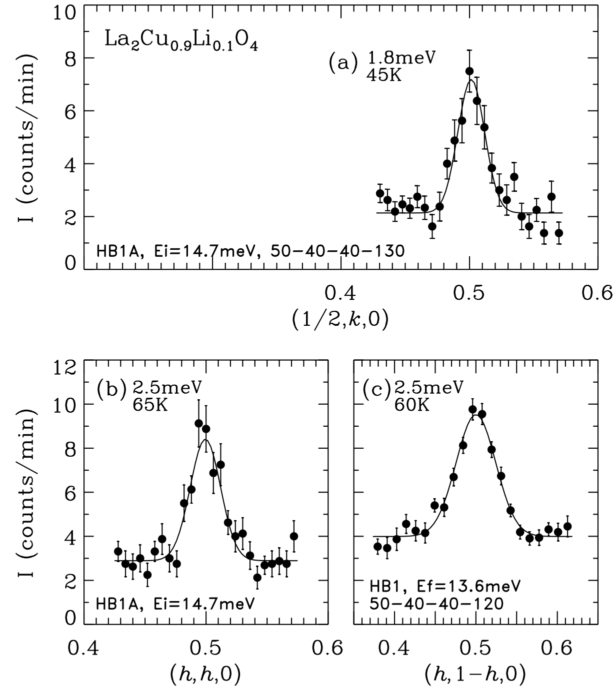

FIG. 2. Const-E scans across the (π, π) point, showing commensurate dynamic magnetic correlations. (a) Scans along the k direction (refer to the vertical solid line in the inset of Fig. 1). (b) Scan along the (h, h) direction (refer to the dotted line in the inset of Fig. 1). (c) Scan along the $(h, -h)$ direction (refer to the dashed line in the inset of Fig. 1).

in the broader peak width at 295 K. The energy dependence of the commensurate magnetic correlations is shown in Fig. 3(c), with const-**Q** scans at three different temperatures. The upturning data points above the dotted curves

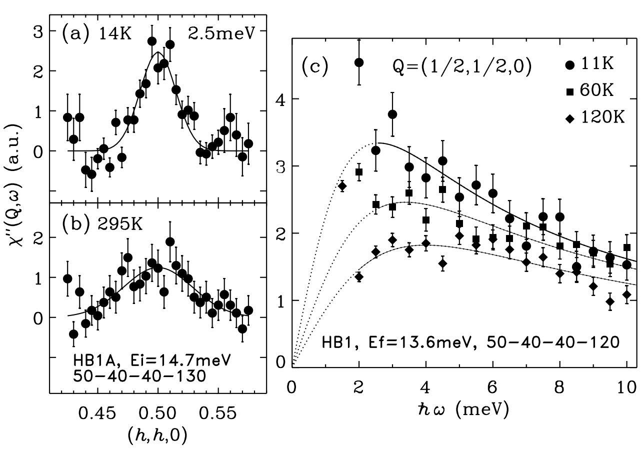

FIG. 3. Const-E scans at 14 K (a) and 295 K (b). Correlation length of the dynamic magnetic correlations is reduced by temperature, as evidenced by a broader peak width at 295 K. The peak intensity of $\chi''(\mathbf{Q}, \omega)$ is also reduced by temperature. (c) Dynamic magnetic susceptibility at the (π, π) point as a function of energy at three temperatures. The temperature dependence shows the expected behavior for dynamic magnetic correlations.

at low energy contain elastic contributions due to the finite energy resolution of the spectrometer. The energy scale, indicated by the peak position of χ'', increases with rising temperature. In both the const-E scans [Figs. 3(a) and 3(b)] and const-**Q** scans [Fig. 3(c)], the magnitude of χ'' decreases with rising temperature. All of these features are as expected for short-range dynamic magnetic order. They reinforce the observation that the commensurate dynamic correlations at (π, π) we found in $La_2Cu_{0.9}Li_{0.1}O_4$ are magnetic.

Prior to this study, a unified picture of magnetic correlations for the La_2CuO_4 system was emerging, namely, that the incommensurability follows a universal function of hole concentration. This universality occurred whether or not the material is superconducting, and whether or not the doping was in the CuO_2 plane [5–8,10–13]. The sole double-layered material in which incommensurability has so far been observed, $YBa_2Cu_3O_{6.6}$, also follows the universal function for the single-layered system [15]. Incommensurate magnetic correlations are also found in the doped insulating nickelates [14] and are recently detected in superconducting $Bi_2Sr_2CaCu_2O_8$ [16]. This robust occurrence of incommensurate magnetic correlations in charge-doped laminar materials, especially the independence on transport properties, has been used to support the stripe picture over the nesting Fermi surface picture, since there is no Fermi surface in the insulating materials. Commensurate magnetic correlations ($\delta = 0$) reported here for $La_2Cu_{0.9}Li_{0.1}O_4$, which has a doped hole concentration of 10%, provide a first exception to this empirical rule. Another attempt to unify magnetic behavior in cuprates is to plot the incommensurability versus the superconducting transition temperature. A linear relation between them is found in $La_{2-x}Sr_xCuO_4$ [10] and possibly also in $YBa_2Cu_3O_{6+y}$ [28]. It is argued that this linear relation provides an important clue to the origin of the high transition temperature superconductivity in cuprates [29]. Our result on $La_2(Cu,Li)O_4$ may be fitted into this scheme as a trivial limiting case: $T_c = 0$ at $\delta = 0$. However, we note that measurements on $(La,Sr)_2CuO_4$ that are codoped with isovalent Zn substituted for Cu seem to violate this rule [12].

Mobility of the doped holes may be another dimension which needs to be included in the picture. When a hole is bound to a dopant, such as in the Li-doped case, the Skyrmion, a long-range topological disturbance for spins surrounding the dopant, is favored as shown theoretically by Haas *et al.* [30]. It is expected that the long-range antiferromagnetic order may be destroyed with a dilute Skyrmion concentration and the short-range magnetic correlations remain commensurate. This is consistent with our neutron scattering data for the Li-doped cuprate. In Zn codoped $(La,Sr)_2CuO_4$, holes are introduced by Sr dopants and they are known to be more mobile. The isovalent Zn dopants may serve as impurity pinning centers for the incommensurate structure formed by the holes. It remains interesting to understand the differences in magnetic correlations between $La_2(Cu,Li)O_4$ and hole-doped La_2NiO_4, both of which are insulating.

In summary, we have shown by neutron scattering that dynamic magnetic correlations in $La_2Cu_{0.9}Li_{0.1}O_4$ are commensurate with the CuO_2 square lattice. This is different from all other previously investigated materials in the hole-doped La_2CuO_4 and $YBa_2Cu_3O_{6+y}$ systems, which remarkably follow a universal dependence on hole concentration.

We wish to thank P. C. Hammel, D. Vaknin, K. Hirota, A. V. Balatsky, S. A. Trugman, A. Moreo, D. Scalapino, A. L. Chernyshev, J. M. Tranquada, T. M. Rice, and F. C. Zhang for discussions and communications. W. B., R. M., and R. H. also wish to thank J. A. Fernandez-Baca, S. Nagler, M. Yethiraj, H. Mook, and B. Chakoumakos for their hospitality at ORNL. Work at LANL and Ames Laboratory was conducted under the auspices of the U.S. Department of Energy, at ORNL supported by the U.S. DOE under Contract No. DE-AC05-960R22464 with Lockheed Martin Energy Research Corporation.

[1] D. Vaknin, S. K. Sinha, D. E. Moncton, D. C. Johnston, J. M. Newsam, C. R. Safinya, and H. E. King, Jr., Phys. Rev. Lett. **58**, 2802 (1987).

[2] G. Shirane, Y. Endoh, R. J. Birgeneau, M. A. Kastner, Y. Hidaka, M. Oda, M. Suzuki, and T. Murakami, Phys. Rev. Lett. **59**, 1613 (1987); B. Keimer *et al.*, Phys. Rev. B **46**, 14 034 (1992).

[3] S. M. Hayden, G. Aeppli, R. Osborn, A. D. Taylor, T. G. Perring, S-W. Cheong, and Z. Fisk, Phys. Rev. Lett. **67**, 3622 (1991).

[4] K. B. Lyons *et al.*, Phys. Rev. B **37**, 2353 (1988).

[5] S-W. Cheong, G. Aeppli, T. E. Mason, H. Mook, S. M. Hayden, P. C. Canfield, Z. Fisk, K. N. Clausen, and J. L. Martinez, Phys. Rev. Lett. **67**, 1791 (1991); T. E. Mason, G. Aeppli, S. M. Hayden, A. P. Ramirez, and H. A. Mook, *ibid.* **71**, 919 (1993).

[6] H. Yashizawa, S. Mitsuda, H. Kitazawa, and K. Katsumata, J. Phys. Soc. Jpn. **57**, 3686 (1988).

[7] T. R. Thurston *et al.*, Phys. Rev. B **46**, 9128 (1992); **40**, 4585 (1989); G. Shirane *et al.*, Phys. Rev. Lett. **63**, 330 (1989); R. J. Birgeneau *et al.*, Phys. Rev. B **38**, 6614 (1988).

[8] K. Yamada *et al.*, Physica (Amsterdam) **282C–287C**, 85 (1997).

[9] For transport data of insulating $(La,Sr)_2(Cu,Zn)O_4$, see Y. Fukuzumi *et al.*, Phys. Rev. Lett. **76**, 684 (1996); M. Z. Cieplak *et al.*, Appl. Phys. Lett. **73**, 2823 (1998).

[10] K. Yamada *et al.*, Phys. Rev. B **57**, 6165 (1998).

[11] J. M. Tranquada, J. D. Axe, N. Ichikawa, A. R. Moodenbaugh, Y. Nakamura, and S. Uchida, Phys. Rev. Lett. **78**, 338 (1997).

[12] K. Hirota *et al.*, Physica (Amsterdam) **241B–243B**, 817 (1998); H. Kimura *et al.*, Phys. Rev. B **59**, 6517 (1999).

[13] B. O. Wells, Y. S. Lee, M. A. Kastner, R. H. Christianson, R. J. Birgeneau, K. Yamada, Y. Endoh, and G. Shirane, Science **277**, 1067 (1997); Y. S. Lee *et al.*, Phys. Rev. B **60**, 3643 (1999).

[14] S. M. Hayden *et al.*, Phys. Rev. Lett. **68**, 1061 (1992); K. Yamada *et al.*, Physica (Amsterdam) **221C**, 355 (1994); J. M. Tranquada, P. Wochner, and D. Buttrey, Phys. Rev. Lett. **79**, 2133 (1997).

[15] H. A. Mook *et al.*, Nature (London) **395**, 580 (1998); P. Dai *et al.*, Phys. Rev. Lett. **80**, 1738 (1998).

[16] H. A. Mook, F. Dogan, and B. C. Chakoumakos, e-print cond-mat/9811100.

[17] J. M. Tranquada, B. J. Sternlieb, J. D. Axe, Y. Nakamura, and S. Uchida, Nature (London) **375**, 561 (1995); S. A. Kivelson, E. Fradkin, and V. J. Emery, *ibid.* **393**, 550 (1998).

[18] D. Poilblanc and T. M. Rice, Phys. Rev. B **39**, 9749 (1989); J. Zaanen and O. Gunnarsson, *ibid.* **40**, 7391 (1989); H. J. Schulz, Phys. Rev. Lett. **64**, 1445 (1990).

[19] V. J. Emery, S. A. Kivelson, and H. Q. Lin, Phys. Rev. Lett. **64**, 475 (1990); M. Grilli, R. Raimondi, C. Castellani, and C. Di Castro, *ibid.* **67**, 259 (1991); W. O. Putikka, M. U. Luchini, and T. M. Rice, *ibid.* **68**, 538 (1992).

[20] E. Dagotto *et al.*, Phys. Rev. B **49**, 3548 (1994); T. Tohyama *et al.*, *ibid.* **59**, R11 649 (1999); A. L. Chernyshev, A. H. Castro Neto, and A. R. Bishop, cond-mat/9909128; C. Buhler, S. Yunoki, and A. Moreo, Phys. Rev. Lett. **84**, 2690 (2000).

[21] Q. Si *et al.*, Phys. Rev. B **47**, 9055 (1993); P. B. Littlewood *et al.*, *ibid.* **48**, 487 (1993).

[22] S. Schmitt-Rink, C. M. Varma, and A. E. Ruckenstein, Phys. Rev. Lett. **60**, 2793 (1988); B. I. Shraiman and E. D. Siggia, *ibid.* **62**, 1564 (1989); H. J. Schulz, *ibid.* **64**, 1445 (1990); T. Giamarchi and C. Lhuillier, Phys. Rev. B **42**, 10 641 (1990); S. Sarker, C. Jayaprakash, H. R. Krishnamurthy, and W. Wenzel, *ibid.* **43**, 8775 (1991).

[23] J. L. Sarrao, D. P. Young, Z. Fisk, E. G. Moshopoulou, J. D. Thompson, B. C. Chakoumakos, and S. E. Nagler, Phys. Rev. B **54**, 12 014 (1996).

[24] L. P. Le, R. H. Heffner, D. E. MacLaughlin, K. Kojima, G. M. Luke, B. Nachumi, Y. J. Uemura, J. L. Sarrao, and Z. Fisk, Phys. Rev. B **54**, 9538 (1996).

[25] B. J. Suh, P. C. Hammel, Y. Yoshinari, J. D. Thompson, J. L. Sarrao, and Z. Fisk, Phys. Rev. Lett. **81**, 2791 (1998); J. H. Cho, F. C. Chou, and D. C. Johnston, *ibid.* **70**, 222 (1993).

[26] A. I. Rykov, H. Yasuoka, and Y. Ueda, Physica (Amsterdam) **247C**, 327 (1995).

[27] G. L. Squires, *Introduction to the Theory of Thermal Neutron Scattering* (Cambridge University Press, Cambridge, England, 1978).

[28] A. V. Balatsky and P. Bourges, Phys. Rev. Lett. **82**, 5337 (1999).

[29] A. V. Balatsky and Z.-X. Shen, Science **284**, 1137 (1999).

[30] S. Haas, F-C. Zhang, F. Mila, and T. M. Rice, Phys. Rev. Lett. **77**, 3021 (1996).