Commensurate dynamic magnetic correlations in La$_2$Cu$_0.9$Li$_{0.1}$O$_4$

Wei Bao, R. J. McQueeney, R. Heffner, J. L. Sarrao, P. Dai, J. L. Zarestky

1Los Alamos National Laboratory, Los Alamos, NM 87545
2Oak Ridge National Laboratory, Oak Ridge, TN 37831
3Ames Laboratory, Ames, IA 50011

(May 3, 2021)

When sufficient numbers of holes are introduced into the two-dimensional CuO$_2$ square lattice, dynamic magnetic correlations become incommensurate with underlying lattice in all previously investigated La$_{2-x}$A$_x$Cu$_{1-z}$Bi$_z$O$_{4+y}$ (A = Sr or Nd, B = Zn) including high T$_c$ superconductors and insulators, and in bilayered superconducting YBa$_2$Cu$_3$O$_{6.6}$ and Bi$_2$Sr$_2$CaCu$_2$O$_8$. Magnetic correlations also become incommensurate in structurally related La$_6$NiO$_4$ when doped with Sr or O. We report an exception to this so-far well established experimental “rule” in La$_2$Cu$_{1-z}$Li$_z$O$_4$ in which magnetic correlations remain commensurate.

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$_2$CuO$_4$ or YBa$_2$Cu$_3$O$_6$. 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 $J$. 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$_2$CuO$_4$ 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%, incommensuratedynamic magnetic correlations develop at a quartet of wave vectors $Q = (\frac{1}{2} \pm \delta, \frac{1}{2}, 0)$ and $(\frac{1}{2}, -\frac{1}{2} \pm \delta, 0)$. Even more remarkable is that the incommensurability $\delta$ is a universally increasing function of the hole concentration $n$. Whether the doped material is superconducting or insulating, whether dopant resides on the La, Cu, or the oxygen sites. In the isstructuralinsulating nickelates, the magnetic correlations are found to be incommensurate when a sufficient number of holes are introduced by doping either the La or the O sites. Recently, incommensurate magnetic correlations were also discovered in bilayered superconducting YBa$_2$Cu$_3$O$_{6.6}$. The incommensurability $\delta$ vs. $n$ falls on the same curve as the La$_2$CuO$_4$ system, adding new excitement to the field. There is also experimental evidence indicating incommensurate magnetic correlations in superconducting Bi$_2$Sr$_2$CaCu$_2$O$_8$. Empirically, there are 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 where charge carriers undergo phase separation, to nesting Fermi surface models and doped quantum antiferromagnet models where the carriers remain uniform in the sample.

La$_2$Cu$_{1-z}$Li$_z$O$_4$ remains an insulator for $0 \leq z \leq 0.5$. It has identical in-plane lattice parameters as Sr-doped La$_2$CuO$_4$ at the same hole concentration. The long-range antiferromagnetic order is similarly suppressed by Sr or Li doping. The spin dynamics of La$_2$Cu$_{1-z}$Li$_z$O$_4$, as revealed by local dynamical probes such as nuclear quadrapole resonance (NQR), show an astonishing parallel with La$_2$-Sr$_2$CuO$_4$. 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$_2$Cu$_{1-z}$Li$_z$O$_4$ 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$_2$Cu$_{1-z}$Li$_z$O$_4$ 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 $\delta$, keeping in mind that $\delta$ increases with hole concentration in all other doped La$_2$CuO$_4$ materials. Magnetization measurements show no long-range magnetic order, consistent with previous studies. 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 ($a^\ast = 1.174 \AA^{-1}$ and $c^\ast = 0.4814 \AA^{-1}$ at 15 K) to label the reciprocal space; thus the $(\frac{1}{2}, \frac{1}{2}, 0)$ corresponds to the $(\pi, \pi)$ square-lattice antiferromagnetic wave vector. A single crystal of 0.46 grams with mosaic $< 0.5^\circ$ was used in the Q scans. The energy scans in Fig. (c) were taken with 5 aligned crystals of total mass 1.0 grams and mosaic of 0.9$. Neutron scattering experiments were performed at the HB1A and HB1 triple-axis spectrometers at the HFIR reactor of ORNL. The samples were mounted to the cold finger of a Diplex refrigerator both in the $(h, k, 0)$ and $(h, h, l)$ 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 \(S(Q, \omega)\) [27].

\[
I \propto |F(Q)|^2 \cdot \overline{S}(Q, \omega),
\]

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

\[
\chi''(Q, \omega) = \left(1 - e^{-\hbar \omega / k_B T}\right) S(Q, \omega).
\]

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

\[
S(Q, \omega) = \frac{\hbar \omega}{1 - e^{-\hbar \omega / k_B T}} \frac{\chi_Q \Gamma_Q}{(\hbar \omega)^2 + \Gamma_Q^2},
\]

where \(\Gamma_Q\) is the energy scale for magnetic fluctuations and \(\chi_Q\) is the \(Q\)-dependent magnetic susceptibility which determines the intensity. The maximum of \(S(Q, \omega)\) is at \(\omega = 0\), which is the contribution to quasielastic scattering from dynamic magnetic fluctuations:

\[
S(Q, 0) = k_B T \frac{\chi_Q}{\Gamma_Q}.
\]

The magnetic form factor \(|F(Q)|^2\) in Eq. (1) has its maximum at \(Q = 0\) 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 \((\pi, \pi)\) point is shown in the inset in Fig. 1(a). The crosses schematically denote incommensurate wave vectors for the low energy spin fluctuations previously found in sufficiently doped La\(_2\)CuO\(_4\) and YBa\(_2\)Cu\(_3\)O\(_6.6\). A constant energy (const-\(E\)) scan along the \(k\) direction with \(h \omega = 0\) 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\(_2\)Cu\(_{0.9}\)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 \((\pi, \pi)\) 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 \(5 \times 10^{-4}\) of the intensity of the structural Bragg peak \((110)\). By adding more filters, it can be shown that the intensity at \((\frac{3}{2}, \frac{3}{2}, 0)\) 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 \(Q\) dependence in other doped cuprates with identical hole concentration. Further investigation as to its origin is underway.

To detect the dynamic magnetic correlations, we have repeated the \(k\) scan at a finite energy, \(h \omega = 1.8\) meV, that avoids the elastic superstructure contribution. Results are shown in Fig. 1(a). The const-\(E\) scan measures a peak at \(Q = (\frac{1}{2}, \frac{1}{2}, 0)\) in the dynamic magnetic correlation function \(S(Q, \omega)\). Scans along two other symmetrically inequivalent directions are shown in Fig. 1(b) and (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. 1, the dynamic magnetic correlations are compared at 14 K and 295 K in an identical const-\(E\) scan. The data have been converted to \(\chi''\), using FIG. 1. The inset in (a) shows the 2D in-plane \((h, k)\) reciprocal space. The square marks the commensurate \((\pi, \pi)\) point which characterizes dynamic magnetic correlations in La\(_2\)Cu\(_{0.9}\)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 \((\pi, \pi)\) 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.
the doped insulating nickelates [14] and are recently de-
Incommensurate magnetic correlations are also found in
the universal function for the single-layered system [15].

Prior to this study, a unified picture of magnetic correl-
arions was emerging, namely

that this linear relation provides an important clue to
the origin of the high transition temperature supercon-

Eq. (2). The shorter correlation length of the dynamic
magnetic correlations at the higher temperature is re-
flected 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 differ-
ent temperatures. The upturning data points above the
dotted curves 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-Q
scans [Fig. 3(a) and (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$_2$Cu$_{0.8}$Li$_{0.2}$O$_4$ are magnetic.

Prior to this study, a unified picture of magnetic correl-
arions for the La$_2$CuO$_4$ 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 [3, 11, 12, 13]. The
sole double-layered material in which incommensurabil-
ity has so far been observed, YBa$_2$Cu$_3$O$_{6.6}$, also follows
the universal function for the single-layered system [1].
Incommensurate magnetic correlations are also found in
the doped insulating nickelates [4] and are recently de-
valent 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)$_4$O$_7$ and hole-doped La$_2$NiO$_4$, both of which are insulating.

In summary, we have shown by neutron scattering that dynamic magnetic correlations in La$_2$Cu$_{0.9}$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$_2$CuO$_4$ and YBa$_2$Cu$_3$O$_{6+y}$ 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, 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, B. Chakoumakos for their hospitality at ORNL. Work at LANL and Ames Lab. was conducted under the auspices of US Department of Energy, at ORNL supported by US DOE under contract No. DE-AC05-96OR22464 with Lockheed Martin Energy Research Corp.

[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, 14034 (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. Yoshizawa, 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); ibid. 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 C 282-287, 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 B 241-243, 817 (1998); H. Kimura, et al., Phys. Rev. B 59, 6517 (1999).
[13] B. O. Wells, Y. S. Lee, M. A. Kastner, R. H. Christiansen, 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 C 221, 355 (1994); J. M. Tranquada, P. Wochner and D. Buttrey, Phys. Rev. Lett. 79, 2133 (1997).
[15] H. A. Mook et al., Nature 395, 580 (1998); P. Dai et al., Phys. Rev. Lett. 80, 1738 (1998).
[16] H. A. Mook, F. Degau and B. C. Chakoumakos, eprint cond-mat/9811101 (1998).
[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. B 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. Tomyama et al., ibid., 59, R11649 (1999); A. L. Chernyshev, A. H. Castro Neto and A. R. Bishop, cond-mat/9909128.
[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, 10641 (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, 12014 (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 C 247, 327 (1995).
[27] G. L. Squires, Introduction to the Theory of Thermal Neutron Scattering (Cambridge University Press, Cambridge, 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. Hass, F-C. Zhang, F. Mila and T. M. Rice, Phys. Rev. Lett. 77, 3021 (1996).