Proc. Int. Symposium on Advances in Neutron Scattering Research, Tokai, 2000
J. Phys. Soc. Jpn. **70** (2001) Suppl. A pp. 46–48

Neutron Scattering Studies on Lightly-Doped $La_{2-x}Sr_xCuO_4$

Masaaki MATSUDA, Robert J. BIRGENEAU[1, *], Yasuo ENDOH[2], Masaki FUJITA[3], Haruhiro HIRAKA[2], Marc A. KASTNER[1], Gen SHIRANE[4], Shuichi WAKIMOTO[1,4] and Kazuyoshi YAMADA[3]

Advanced Science Research Center, Japan Atomic Energy Research Institute Tokai, Ibaraki 319-1195, Japan
[1]*Department of Physics and Center for Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, U.S.A.*
[2]*Institute for Materials Research, Tohoku University, Katahira, Sendai 980-8577, Japan*
[3]*Institute for Chemical Research, Kyoto University, Gokasho, Uji 610-0011, Japan*
[4]*Department of Physics, Brookhaven National Laboratory, Upton, New York 11973, U.S.A.*

A review is given of neutron elastic and inelastic scattering studies in the lightly-doped $La_{2-x}Sr_xCuO_4$, which exhibits a spin-glass behavior. The most remarkable behavior in the static spin correlations is that a diagonal spin modulation, which is a one-dimensional modulation rotated away by 45° from that in the superconducting phase, occurs universally across the insulating spin-glass phase in $La_{2-x}Sr_xCuO_4$ ($0.02 \leq x \leq 0.055$). This establishes an intimate relation between the magnetism and the transport properties in the high-temperature copper oxide superconductors. Magnetic excitation spectra suggest that magnetic correlations change from being incommensurate to commensurate at a temperature and an energy.

KEYWORDS: $La_{2-x}Sr_xCuO_4$, neutron scattering, spin-glass

§1. Introduction

The phase diagram of $La_{2-x}Sr_xCuO_4$ shows that the magnetic state changes dramatically with Sr doping. The parent material La_2CuO_4 exhibits three-dimensional (3D) long-range antiferromagnetic order below ~325 K.[1)] When a small fraction of La is replaced by Sr, which corresponds to hole-doping, the 3D antiferromagnetic order disappears and the low temperature magnetic phase is replaced by a disordered magnetic phase in which commensurate two-dimensional short-range antiferromagnetic fluctuations are observed.[2,3)] In the superconducting phase dynamic incommensurate (IC) spin fluctuations persist. It has been known for some time that in $La_{2-x}Sr_xCuO_4$ when the insulator-to-superconductor transition occurs, the instantaneous magnetic correlations change from being commensurate to incommensurate.

In this paper, a review is given of the recent results on the static and dynamic spin correlations in the spin-glass phase of lightly-doped $La_{2-x}Sr_xCuO_4$.

§2. Static Properties

Recently, Wakimoto *et al.* have found that the static magnetic correlations at low temperature are IC in the insulating spin-glass $La_{1.95}Sr_{0.05}CuO_4$.[4)] They have examined the intensity profiles and have shown that there are only 2 satellite peaks along $b_{\rm ortho}$[5)] while in superconducting compounds the IC peaks are located parallel to both the $a_{\rm tetra}$ and $b_{\rm tetra}$ axes. These magnetic correlations in $La_{1.95}Sr_{0.05}CuO_4$ are consistent with diagonal charge stripes, in which the stripes run along the $a_{\rm ortho}$ axis. Actually, such diagonal stripes have been predicted theoretically.[6–10)] Diagonal stripes are also reported experimentally in insulating $La_{2-x}Sr_xNiO_4$.[11)] These results lead to the important conclusion that the static magnetic spin modulation changes from diagonal to parallel at $x = 0.055 \pm 0.005$, coincident with the insulator-to-superconductor transition. Most recently, Matsuda *et al.* showed that the diagonal one-dimensional IC magnetic correlations persist throughout the spin-glass phase down to the critical concentration of x=0.02 for 3D Néel ordering.[12)]

The incommensurability ϵ corresponds to the inverse modulation period of the spin density wave. Here, ϵ is defined in orthorhombic notation so that $\epsilon = \sqrt{2} \times \delta$ where δ is defined in tetragonal units. As shown in Fig. 1, δ follows the linear relation $\delta = x$ reasonably well over the range $0.03 \leq x \leq 0.12$ which spans the insulator-superconductor transition. In a charge stripe model this corresponds to a constant charge per unit length in both the diagonal and parallel stripe phases, or equivalently, 0.7 and 0.5 holes per Cu respectively because of the $\sqrt{2}$ difference in Cu spacings in the diagonal and parallel geometries. The value for x=0.024 definitely deviates from the $\delta = x$ line and instead appears to be close to ~1 hole/Cu as in $La_{2-x}Sr_xNiO_4$ where there is ~1 hole/Ni[11)] along the diagonal stripes. This suggests that as the hole concentration is decreased, in the context of the stripe model, the hole concentration evolves progressively from ~0.5 hole/Cu at x=0.12 to 1 hole/Cu at x=0.024. This behavior is very different from that in $La_{2-x}Sr_xNiO_4$, where the hole density is ~1 hole/Ni over a wide range of hole concentrations in the insulating phase albeit at rather larger hole densities.[13)] We note

* Present address: University of Toronto, Toronto, Ontario, Canada M5S 1A1.

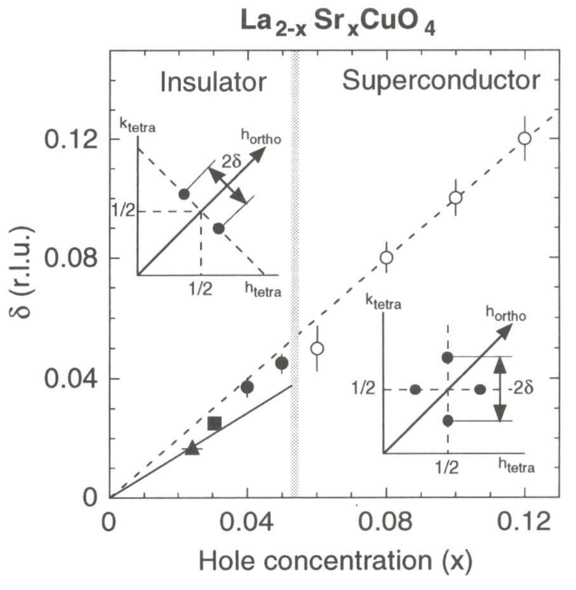

Fig. 1. Hole concentration (x) dependence of the splitting of the IC peaks (δ) in tetragonal reciprocal lattice units. Open circles indicate the data for the inelastic IC peaks reported by Yamada *et al.*[18] Filled circles and square are the data for the elastic IC peaks reported by Wakimoto *et al.*.[5,19] The filled triangle is obtained by Matsuda *et al.*[12] The broken and solid lines correspond to $\delta = x$ and $\epsilon = x$, respectively. The insets show the configuration of the IC peaks in the insulating phase (diagonal stripe) and the superconducting phase (parallel stripe). From Matsuda *et al.*[12]

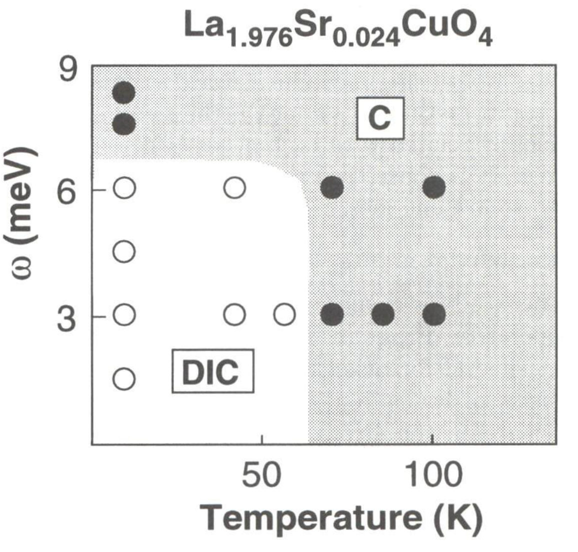

Fig. 2. Energy-temperature phase diagram in $La_{1.976}Sr_{0.024}CuO_4$. Open and filed circles represent that the magnetic correlations are diagonal incommensurate (DIC) and commensurate (C), respectively. From Matsuda *et al.*[12]

that Machida and Ichioka predict 1 hole/Cu throughout the diagonal stripe phase.[14]

We emphasize, here, that only a one-dimensional spin modulation has been observed in $La_{2-x}Sr_xCuO_4$ to-date; any associated charge ordering has not yet been detected. As pointed out by Tranquada, Ichikawa and Uchida,[15] in Nd-doped $La_{2-x}Sr_xCuO_4$ widths of incommensurate magnetic and nuclear peaks become broader in the presence of disorder originating primarily from a random distribution of stripe spacings and orientations. Especially, nuclear peak widths are more sensitive to the disorder. Since incommensurate magnetic peaks are broad throughout the diagonal stripe phase, nuclear peaks due to the lattice distortion should be much broader so that these peaks are very difficult to be observed. Further studies are needed in order to clarify the existence of the charge stripe.

§3. Dynamic Properties

Another important point is to clarify the nature of the magnetic excitations in the diagonal IC state. Intensive studies of the inelastic magnetic spectra in insulating $La_{2-x}Sr_xCuO_4$ were performed by Keimer *et al.*[3] They studied the energy and temperature dependences of the Q-integrated susceptibility and found that it follows a scaling function of ω/T in a wide range of ω and T. However, the Q-dependence of the excitation spectra was not discussed since the detailed peak profile was not known. Now that the static magnetic correlations have been elucidated, the excitation spectra can be analyzed qualitatively.

Matsuda *et al.* studied inelastic magnetic spectra in $La_{1.976}Sr_{0.024}CuO_4$.[12] Figure 2 represents a summary of their neutron inelastic measurements. The open and filled circles signify that the magnetic correlations are diagonal IC and commensurate, respectively. The diagonal IC phase exists below ω ~7 meV and T ~70 K (~6 meV). Above these energy and temperature the magnetic correlations are similar to those in pure La_2CuO_4 although the range of order in the CuO_2 plane is much shorter in $La_{1.976}Sr_{0.024}CuO_4$. This result indicates that the characteristic energy for the diagonal IC structure is 6-7 meV. It is noted that similar behavior is also observed in $La_{1.98}Sr_{0.02}CuO_4$.[16] In this compound, broad elastic magnetic peaks are observed at (π, π). These peaks can be interpreted to be either broad along the b_{ortho} or slightly IC along the b_{ortho}, which is consistent with the diagonal stripe model.

It is interesting to consider the relation between this result and the scaling behavior with ω/T. As reported by Matsuda *et al.* and Keimer *et al.*, deviation from the scaling function is observed at low energies and low temperatures in $La_{1.98}Sr_{0.02}CuO_4$[17] and $La_{1.96}Sr_{0.04}CuO_4$.[3] In both compounds, the Q-integrated susceptibility follows the scaling function above 3-4.5 meV. On the other hand, below 3-4.5 meV, it is suppressed from the scaling function at low temperatures. It was argued that the susceptibility is suppressed at low energies due to the out-of-plane anisotropy. However, as reported by Matsuda *et al.*, an excitation gap due to the out-of-plane anisotropy is lower than 1 meV in $La_{1.976}Sr_{0.024}CuO_4$.[12] From the new results, it appears to be possible that the observed susceptibility is suppressed at low energies and low temperatures because the IC peaks from 2 or 4 magnetic domains were not properly integrated in the experiments in the $(H0L)$ scattering zone.[12] Since the energies at which the deviation from the scaling function is observed in $La_{1.98}Sr_{0.02}CuO_4$ (3-6 meV) and $La_{1.96}Sr_{0.04}CuO_4$ (3-4.5 meV) are similar to the one at which the spin correlations change from being incommensurate to commensurate in $La_{1.976}Sr_{0.024}CuO_4$ (~7 meV), the two behaviors could be closely related with each other. If this speculation is correct, it is considered that the scaling behavior holds down to low energies.

Acknowledgements

This study was supported in part by the U.S.-Japan Cooperative Program on Neutron Scattering, by a Grant-in-Aid for Scientific Research from the Japanese Ministry of Education, Science, Sports and Culture, by a Grant for the Promotion of Science from the Science and Technology Agency, and by CREST. Work at Brookhaven National Laboratory was carried out under Contract No. DE-AC02-98CH10886, Division of Material Science, U.S. Department of Energy. The research at MIT was supported by the National Science Foundation under Grant No. DMR97-04532 and by the MRSEC Program of the National Science Foundation under Award No. DMR98-08941.

1) D. Vaknin, S. K. Sinha, D. E. Moncton, D. C. Johnston, J. Newsam, C. R. Safinya and H. King: Phys. Rev. Lett. **58** (1987) 2802.
2) B. J. Sternlieb, G. M. Luke, Y. J. Uemura, T. M. Riseman, J. H. Brewer, P. M. Gehring, K. Yamada, Y. Hidaka, T. Murakami, T. R. Thurston and R. J. Birgeneau: Phys. Rev. B **41** (1990) 8866.
3) B. Keimer, N. Belk, R. J. Birgeneau, A. Cassanho, C. Y. Chen, M. Greven, M. A. Kastner, A. Aharony, Y. Endoh, R. W. Erwin and G. Shirane: Phys. Rev. B **46** (1992) 14034.
4) S. Wakimoto, G. Shirane, Y. Endoh, K. Hirota, S. Ueki, K. Yamada, R. J. Birgeneau, M. A. Kastner, Y. S. Lee, P. M. Gehring, and S. H. Lee: Phys. Rev. B **60** (1999) R769.
5) S. Wakimoto, R. J. Birgeneau, M. A. Kastner, Y. S. Lee, R. Erwin, P. M. Gehring, S. H. Lee, M. Fujita, K. Yamada, Y. Endoh, K. Hirota and G. Shirane: Phys. Rev. B **61** (2000) 3699.
6) K. Machida: Physica C **158** (1989) 192.
7) M. Kato, K. Machida, H. Nakanishi and M. Fujita: J. Phys. Soc. Jpn. **59** (1990) 1047.
8) D. Poilblanc and T. M. Rice: Phys. Rev. B **39** (1989) 9749.
9) H. Schulz: J. Phys. (Paris) **50** (1989) 2833.
10) J. Zaanen and O. Gunnarsson: Phys. Rev. B **40** (1990) 7391.
11) J. M. Tranquada, D. J. Buttrey and V. Sachan: Phys. Rev. B **54** (1996) 12318.
12) M. Matsuda, M. Fujita, K. Yamada, R. J. Birgeneau, M. A. Kastner, H. Hiraka, Y. Endoh, S. Wakimoto and G. Shirane: Phys. Rev. B **62** (2000) 9148.
13) H. Yoshizawa, T. Kakeshita, R. Kajimoto, T. Tanabe, T. Katsufuji and Y. Tokura: Phys. Rev. B **61** (2000) R854.
14) K. Machida and M. Ichioka: J. Phys. Soc. Jpn. **68** (1999) 2168.
15) J. M. Tranquada, N. Ichikawa and S. Uchida: Phys. Rev. B **59** (1999) 14712.
16) M. Matsuda, Y. S. Lee, M. Greven, M. A. Kastner, R. J. Birgeneau, K. Yamada, Y. Endoh, P. Böni, S.-H. Lee, S. Wakimoto and G. Shirane: Phys. Rev. B **61** (2000) 4326.
17) M. Matsuda, R. J. Birgeneau, Y. Endoh, Y. Hidaka, M. A. Kastner, K. Nakajima, G. Shirane, T. R. Thurston and K. Yamada: J. Phys. Soc. Jpn. **62** (1993) 1702.
18) K. Yamada, C. H. Lee, K. Kurahashi, J. Wada, S. Wakimoto, S. Ueki, H. Kimura, Y. Endoh, S. Hosoya, G. Shirane, R. J. Birgeneau, M. Greven, M. A. Kastner and Y. J. Kim: Phys. Rev. B **57** (1998) 6165.
19) S. Wakimoto and S.-H. Lee: private communication.