CHARGE SEGREGATION AND ANTIFERROMAGNETISM IN HIGH-\( T_c \) SUPERCONDUCTORS

J. M. Tranquada,† N. Ichikawa,‡ K. Kakurai,§ and S. Uchida‡

†Physics Department, Brookhaven National Laboratory, Upton, NY 11973, USA
‡Department of Applied Physics, The University of Tokyo, Yayoi 2-11-16, Bunkyo-ku, Tokyo 113, Japan
§Institute for Solid State Physics, The University of Tokyo, Roppongi, Minato-ku, Tokyo 106-8666, Japan

Abstract

Local antiferromagnetism coexists with superconductivity in the cuprates. Charge segregation provides a way to reconcile these properties. Direct evidence for modulated spin and charge densities has been found in neutron and X-ray scattering studies of Nd-doped \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \). Here we discuss the nature of the modulation, and present some new results for a Zn-doped sample. Some of the open questions concerning the connections between segregation and superconductivity are described.

Keywords: superconductors, neutron scattering, charge-density waves, spin-density waves

1 INTRODUCTION

Neutron-scattering, nuclear-magnetic-resonance (NMR), and muon-spin-rotation (\( \mu \)SR) studies have all provided evidence for the coexistence of local antiferromagnetic spin correlations with superconductivity in the layered cuprates. The nature of this coexistence has been the subject of considerable debate. Neutron scattering studies of certain cuprate superconductors [1,2] have indicated that the magnetic excitations are surprisingly similar to those in the undoped parent compounds, which are both insulating and antiferromagnetic due to strong electronic interactions. One way to accommodate both antiferromagnetic correlations and mobile holes is through charge segregation.
One type of segregation of particular interest involves periodically-spaced charge stripes that act as antiphase domain walls between narrow antiferromagnetic domains. Perhaps the first evidence of such correlations was the measurement of inelastic magnetic scattering peaked at incommensurate wave vectors in La$_{2-x}$Sr$_x$CuO$_4$ with $x \approx 0.15$ [3-5]. Study of the charge modulation has become possible with the discovery that, for $x \approx \frac{1}{8}$, stripes can be pinned by the structural modulation induced by partial substitution of the smaller ion Nd$^{3+}$ for La$^{3+}$ [6,7]. The charge order originally detected by neutron scattering has been confirmed with high-energy x-rays [8], and there is considerable evidence for intimate coexistence of stripe order and superconductivity at $x = 0.15$ [9–11].

Elastic incommensurate magnetic peaks have also been discovered in Zn-doped La$_{2-x}$Sr$_x$CuO$_4$ with $x = 0.14$ [12], and even in samples with no Zn and $x = 0.12$ [13,14]. In samples of La$_{2-x}$Sr$_x$CuO$_4$ with no static order, incommensurate splitting of the inelastic magnetic scattering is nearly identical to that of the elastic peaks in Nd-doped samples with the same Sr concentration [15]. The possibility that the magnetic scattering in underdoped YBa$_2$Cu$_3$O$_{6+x}$ is incommensurate had been considered earlier [16,17], and recently it has been demonstrated that, indeed, at least part of the scattering is incommensurate [18], with modulation wave vectors consistent with the 214 system [19].

In this paper, we briefly discuss several topics related to stripes in the cuprates. In the next section we review the connection between the experimentally observed superlattice peaks, and the real-space modulations of the spin and charge densities. In section 3, we present some new results on a Zn-doped 214 sample. Finally, in section 4 we list some of the open questions concerning stripes and superconductivity in the cuprates.

2 NATURE OF THE SPIN AND CHARGE DENSITY MODULATIONS

There have been questions raised concerning the interpretation of the observed superlattice peaks that suggest some confusion over the distinction between scattering from a 1D system and that from a 2 or 3D system with a 1D modulation. In order to clarify things, first consider a line of atoms with a spacing $a$. In reciprocal space, the scattering from such a 1D system consists of constant-intensity sheets separated by $2\pi/a$.

In contrast, consider a Bravais lattice of atoms with positions $\mathbf{R}_j$, and suppose that the positions undergo a small sinusoidal modulation of the form

$$\mathbf{u}_j = A \sin(\mathbf{g} \cdot \mathbf{R}_j + \phi),$$

(1)
where $\mathbf{A}$ is the amplitude, $\mathbf{g}$ is the modulation wave vector, and $\phi$ is an arbitrary phase shift that we will ignore. Overhauser [20] has shown that the scattering from such a system is given by

$$I(\mathbf{Q}) = \sum_{G,n} J_n^2(\mathbf{Q} \cdot \mathbf{A}) \delta(\mathbf{Q} - \mathbf{G} - n\mathbf{g}).$$

(2)

When $n = 0$ the scattering corresponds to fundamental Bragg peaks with

$$I(\mathbf{G}) \approx 1 - \frac{1}{2}(\mathbf{Q} \cdot \mathbf{A})^2 \approx e^{-(\mathbf{Q} \cdot \mathbf{A})^2/2},$$

(3)

for $\mathbf{Q} \cdot \mathbf{A} \ll 1$. The modulation causes an intensity reduction with a form similar to a Debye-Waller factor. For $n = 1$, one finds superlattice peaks split about each reciprocal lattice vector $\mathbf{G}$ by $\mathbf{g}$, with

$$I(\mathbf{G} + \mathbf{g}) \approx \frac{1}{4}(\mathbf{Q} \cdot \mathbf{A})^2.$$

(4)

Higher-order peaks will also occur, but they are extremely weak for small $\mathbf{Q} \cdot \mathbf{A}$. For example, $I(\mathbf{G} + 2\mathbf{g}) \approx \frac{1}{4}I^2(\mathbf{G} + \mathbf{g})$.

The configuration of superlattice peaks observed in stripe-ordered La$_{1.6-x}$Nd$_0.4$Sr$_x$CuO$_4$ has been reviewed elsewhere [21]. Briefly, there are two sets of superlattice peaks, which are most easily described in terms of a unit cell with $a \approx 3.8$ Å. One set of peaks is split about the antiferromagnetic wave vector by an amount $\epsilon \times 2\pi/a$ along the [100] and [010] directions, indicating antiphase antiferromagnetic domains. A second set of peaks occurs about nuclear Bragg peaks, split by $2\epsilon \times 2\pi/a$, indicative of charge-order. Given that we have peaks split in two directions, there are two possible interpretations: either we are averaging over domains each of which has a single modulation direction, or the two modulations are superimposed in each region of the sample [22]. The simple stripe picture is based on the former model. Can we rule out the latter?

If there is a stripe grid, then the phase of the antiferromagnetic domains must be modulated in two directions. The arrangement of domains and their relative phase factors forms a checker-board structure, similar to a simple Néel antiferromagnet. The unit cell of such a structure has its axes rotated by $\pi/4$ relative to the modulation directions, and it has an area twice that of a single domain. This means that in reciprocal space, the first superlattice peaks should be rotated by $\pi/4$ relative to the modulation wave vectors. Experimentally, we have checked for magnetic peaks split along [110] and [110], and have found nothing. (In principle, a stripe grid should result in a square lattice of superlattice peaks, so that there should be peaks in these directions regardless of
Fig. 1. Search for charge-order peaks in La$_{1.48}$Nd$_{0.4}$Sr$_{0.12}$CuO$_4$. Upper left indicates scan directions in reciprocal space. Upper right: intensity difference between 7 K and 65 K scans along [010] direction. Lower: similar measurement along [110].

Unlike the magnetic peaks, the charge-order peaks from such a grid of diagonal stripes should not be rotated. The first charge order peaks should appear at $(\epsilon, \epsilon, 0)$, so that the peaks we have observed at $(0, 2\epsilon, 0)$ would involve the sum of the two modulation wave vectors. To test this possibility, we performed neutron scattering measurements on the PONTA (5G) triple-axis spectrometer at the JRR-3 reactor at JAERI. The $x = 0.12$ Nd-doped crystal characterized previously [7] was used, and elastic scans in the appropriate directions were performed (using 14.7-meV neutrons and relatively open collimation) at two temperatures: 7 K and 65 K. We subtracted the data at 65 K from the 7 K measurement in order to isolate any signal that might appear only at low temperature. The results are shown in Fig. 1. The peak found in the [010] direction is consistent with previous work [7]: however, only noise is present along [110]. Thus, a 2D grid interpretation appears to be incompatible with experiment.
static modulations in Zn-doped La$_{2-x}$Sr$_x$CuO$_4$

It is of interest to search for evidence of charge-stripe order in other cuprate systems. So far, charge-order superlattice peaks have only been observed in Nd-doped La$_{2-x}$Sr$_x$CuO$_4$, where the stripes are pinned by the low-temperature-tetragonal lattice structure. One obvious candidate system is Zn-doped La$_{2-x}$Sr$_x$CuO$_4$. Elastic magnetic peaks have been observed by Hirota et al. [12] in a superconducting crystal with $x = 0.14$.

With the intention of looking for charge order, a single crystals ($\approx 0.45$ cm$^3$) of La$_{1.88}$Sr$_{0.12}$Cu$_{0.98}$Zn$_{0.02}$O$_4$ was grown with an infrared image furnace at the University of Tokyo. Again, we made use of the PONTA (5G) triple-axis spectrometer, with an incident energy of 14.7 meV and a pyrolytic graphite filter before the sample. Relatively tight collimation was used to measure the lattice parameters, yielding $a = 5.2657$ Å and $b = 5.3042$ Å ($b - a = 0.0385$ Å) at 40 K. Although the crystal is orthorhombic at low temperature, we chose to work in tetragonal coordinates ($a \approx b = 3.74$ Å) to search for elastic magnetic peaks. Opening the horizontal collimations to 40′-40′-80′-80′, we scanned along $Q = (\frac{1}{2}, \frac{1}{2} + \xi, 0)$ and found peaks at $\xi = \pm 0.121 \equiv \pm \epsilon$. An example is shown in Fig. 2. The peak width is roughly 40% greater than that found under the same conditions in La$_{1.48}$Nd$_{0.4}$Sr$_{0.12}$CuO$_4$, with no correction for resolution. The temperature dependence of the peak intensity is presented in Fig. 3. The disordering temperature of $\sim 20$ K is intermediate with respect to those ($T_m \sim 30$ K and 17 K) found in crystals of La$_{1.88}$Sr$_{0.12}$Cu$_{1-y}$Zn$_y$O$_4$ with $y = 0$ and 0.03 (respectively) by Kimura et al. [14]. Initial attempts to observe charge-order peaks were unsuccessful.

$\mu$SR [23] and NMR [24] studies indicate that Zn suppresses superconductivity locally, resulting in electronic inhomogeneity. One might expect that if Zn serves to pin stripes, this effect would also be inhomogeneous. Hence,
Fig. 3. Temperature dependence of magnetic peak intensity in La$_{1.88}$Sr$_{0.12}$Cu$_{0.98}$Zn$_{0.02}$O$_4$. The dashed line is a linear fit to the background measurements.

it is of interest to compare the magnetic peak intensity with that found in La$_{1.48}$Nd$_{0.4}$Sr$_{0.12}$CuO$_4$, where $\mu$SR has shown the magnetic order to be relatively uniform [11]. We measured the same magnetic peaks in the latter compound under identical conditions, except that we worked at a temperature of 7 K in order to avoid any significant contribution from the Nd moments. The relative crystal volumes were determined by phonon measurements. Normalizing by volume, and assuming no substantial difference in $l$ dependence of the scattering, the magnetic intensity in the Zn-doped crystal was found to be just $(22 \pm 6)\%$ of that in the Nd-doped crystal. If this represents the volume fraction showing stripe order, then it is not surprising that charge-order peaks were not observed.

4 OPEN QUESTIONS

While there is substantial evidence for stripe correlations in La$_{2-x}$Sr$_x$CuO$_4$, the issue of whether charge stripes are common to all superconducting cuprates remains controversial. The recent observations [18,19] of incommensurate magnetic scattering in YBa$_2$Cu$_3$O$_{6.6}$ provide an important connection. One would also like to see evidence of related charge correlations, but this is more difficult. Mook and coworkers have made some progress in this direction using a special energy-integrated technique [25]. It may also be necessary to investigate phonon anomalies, such as the high-energy longitudinal optical branch in La$_{1.88}$Sr$_{0.15}$CuO$_4$ that has been studied by Egami and coworkers [26].

Another issue in YBa$_2$Cu$_3$O$_{6+x}$ concerns the so-called resonance peak. The resonance peak is centered on the antiferromagnetic wave vector, and the energy at which it is centered increases with $x$. Bourges [27] has noted that the ratio of the resonance-peak energy to the superconducting transition temperature,
is roughly constant. In the BCS model for superconductivity, the ratio of the superconducting gap to $T_c$ is also a constant. This similarity might lead one to suspect a connection between the resonance-peak energy and the superconducting gap. However, measurements of the gap in Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ by tunneling [28–30] and photoemission [31,32] indicate that the superconducting gap increases while $T_c$ decreases on the underdoped side. While similar measurements are not yet available for YBa$_2$Cu$_3$O$_{6+x}$, infrared conductivity studies of the latter system suggest that the size of the gap does not decrease as $x$ is reduced from 1 [33]. What is the possible relationship between the resonance peak and stripe correlations?

So far, we have discussed only hole-doped superconductors. Might stripes be relevant to electron-doped superconductors? Moving beyond cuprates, charge stripes are already known to be important in nickelates and certain manganates. Do stripes occur in other transition-metal oxide systems? Clearly, there is a great deal of work left to do, and neutron scattering will be a prominent tool in this effort.

5 ACKNOWLEDGMENT

Work at Brookhaven is supported by Contract No. DE-AC02-98CH10886, Division of Materials Sciences, U.S. Department of Energy.

References

[1] S. M. Hayden, G. Aeppli, H. A. Mook, T. G. Perring, T. E. Mason, S.-W. Cheong, and Z. Fisk, Phys. Rev. Lett. 76, 1344 (1996).
[2] P. Bourges, H. F. Fong, L. P. Regnault, J. Bossy, C. Vettier, D. L. Milius, I. A. Aksay, and B. Keimer, Phys. Rev. B 56, R11 439 (1997).
[3] 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).
[4] T. E. Mason, G. Aeppli, and H. A. Mook, Phys. Rev. Lett. 68, 1414 (1992).
[5] T. R. Thurston, P. M. Gehring, G. Shirane, R. J. Birgeneau, M. A. Kastner, Y. Endoh, M. Matsuda, K. Yamada, H. Kojima, and I. Tanaka, Phys. Rev. B 46, 9128 (1992).
[6] J. M. Tranquada, B. J. Sternlieb, J. D. Axe, Y. Nakamura, and S. Uchida, Nature 375, 561 (1995).
[7] J. M. Tranquada, J. D. Axe, N. Ichikawa, Y. Nakamura, S. Uchida, and B. Nachumi, Phys. Rev. B 54, 7489 (1996).
[8] M. v. Zimmermann, A. Vigliante, T. Niemöller, N. Ichikawa, T. Frello, S. Uchida, N. H. Andersen, J. Madsen, P. Wochner, J. M. Tranquada, D. Gibbs, and J. R. Schneider, Europhys. Lett. 41, 629 (1998).

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

[10] J. E. Ostenson, S. Bud’ko, M. Breitwisch, D. K. Finnemore, N. Ichikawa, and S. Uchida, Phys. Rev. B 56, 2820 (1997).

[11] B. Nachumi, Y. Fudamoto, A. Keren, K. M. Kojima, M. Larkin, G. M. Luke, J. Merrin, O. Tchernyshyov, Y. J. Uemura, N. Ichikawa, M. Goto, S. Uchida, M. K. Crawford, E. M. McCarron, D. E. MacLaughlin, and R. H. Heffner, Phys. Rev. B 58, 8760 (1998).

[12] K. Hirota, K. Yamada, I. Tanaka, and H. Kojima, Physica B 241–243, 817 (1998).

[13] T. Suzuki, T. Goto, K. Chiba, T. Shinoda, T. Fukase, H. Kimura, K. Yamada, M. Ohashi, and Y. Yamaguchi, Phys. Rev. B 57, R3229 (1998).

[14] H. Kimura, K. Hirota, H. Matsushita, K. Yamada, Y. Endoh, S.-H. Lee, C. F. Majkrzak, R. Erwin, G. Shirane, M. Greven, Y. S. Lee, M. A. Kastner, and R. J. Birgeneau, (preprint, BNL-65817).

[15] K. Yamada, C. H. Lee, K. Kurahashi, J. Wada, S. Wakimoto, S. Ueki, Y. Kimura, Y. Endoh, S. Hosoya, G. Shirane, R. J. Birgeneau, M. Greven, M. A. Kastner, and Y. J. Kim, Phys. Rev. B 57, 6165 (1998).

[16] B. J. Sternlieb, J. M. Tranquada, G. Shirane, M. Sato, and S. Shamoto, Phys. Rev. B 50, 12915 (1994).

[17] J. M. Tranquada, Physica C 282–287, 166 (1997).

[18] P. Dai, H. A. Mook, and F. Doğan, Phys. Rev. Lett. 80, 1738 (1998).

[19] H. A. Mook, P. Dai, S. M. Hayden, G. Aeppli, T. G. Perring, and F. Doğan, Nature 395, 580 (1998).

[20] A. W. Overhauser, Phys. Rev. B 3, 3173 (1971).

[21] J. M. Tranquada, in Neutron Scattering in Layered Copper-Oxide Superconductors, edited by A. Furrer (Kluwer, Dordrecht, The Netherlands, 1998).

[22] J. M. Tranquada, Physica B 241–243, 745 (1998).

[23] B. Nachumi, A. Keren, K. Kojima, M. Larkin, G. M. Luke, J. Merrin, O. Tchernyshov, Y. J. Uemura, N. Ichikawa, M. Goto, and S. Uchida, Phys. Rev. Lett. 77, 5421 (1996).

[24] A. V. Mahajan, H. Alloul, G. Collin, and J. F. Marucco, Phys. Rev. Lett. 72, 3100 (1994).
[25] H. A. Mook, F. Doğan, and B. C. Chakoumakos, cond-mat/9811100.

[26] R. J. McQueeney, Y. Petrov, T. Egami, M. Yethiraj, G. Shirane, and Y. Endoh, (preprint).

[27] P. Bourges, in *The Gap Symmetry and Fluctuations in High Temperature Superconductors*, edited by J. Bok, G. Deutscher, and D. Pavuna (Plenum, New York, 1998).

[28] M. Oda, K. Hoya, R. Kubota, C. Manabe, N. Momono, T. Nakano, and M. Ido, Physica C *281*, 135 (1997).

[29] C. Renner, B. Revaz, J.-Y. Genoud, K. Kadowaki, and O. Fischer, Phys. Rev. Lett. *80*, 149 (1998).

[30] N. Miyakawa, P. Guptasarma, J. F. Zasadzinski, D. G. Hinks, and K. E. Gray, Phys. Rev. Lett. *80*, 157 (1998).

[31] J. M. Harris, Z.-X. Shen, P. J. White, D. S. Marshall, M. C. Schabel, J. N. Eckstein, and I. Bozovic, Phys. Rev. B *54*, R15 665 (1996).

[32] H. Ding, T. Yokoya, J. C. Campuzano, T. Takahashi, M. Randeria, M. R. Norman, T. Mochiku, K. Kadowaki, and J. Giapintzakis, Nature *382*, 51 (1996).

[33] D. N. Basov, R. Liang, B. Dabrowski, D. A. Bonn, W. N. Hardy, and T. Timusk, Phys. Rev. Lett. *77*, 4090 (1996).