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Neutron-scattering study of static antiferromagnetic correlations in $La_{2-x}Sr_xCu_{1-y}Zn_yO_4$

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Neutron-scattering measurements have been performed to search for possible elastic incommensurate magnetic peaks in superconducting $La_{2-x}Sr_xCuO_4$ with $x=0.10$, 0.12, and 0.15. The most dramatic effects are found for $x=0.12$; in this case, the peak intensity first appears at the onset of superconductivity T_c ($=31$ K). The resolution-limited peak width indicates that the static magnetic correlation length exceeds 200 Å isotropically in the CuO_2 planes. Weak elastic peaks are also observed at low temperatures for $x=0.10$ while for $x=0.15$ any incommensurate elastic scattering is below the limit of detectability. Elastic peaks are observed in Zn-substituted nonsuperconducting $La_{1.88}Sr_{0.12}Cu_{0.97}Zn_{0.03}0_4$. However, in this case, the Zn substitution degrades the magnetic order; the peak appears at lower temperature (17 K) and the correlation length is shorter (80 Å) than that in the Zn-free $x=0.12$ sample. [S0163-1829(99)02909-4]

I. INTRODUCTION

Over the past decade spin fluctuations in the lamellar copper oxides have been studied extensively in insulating, weakly metallic and superconducting samples.[1] Experimental probes include muon spin resonance (μSR), nuclear magnetic resonance (NMR), and neutron scattering. The former two are local probes whereas the latter allows studies as a function of both energy and momentum. These measurements have elucidated the magnetism of the two-dimensional (2D) square lattice quantum Heisenberg antiferromagnet in pure samples including $Sr_2CuO_2Cl_2$ (Ref. 2) and La_2CuO_4.[3] Experiments on lightly doped $La_2CuO_{4+\delta}$ (Refs. 4,5) and $La_{2-x}Sr_xCuO_4$ (Ref. 6) have revealed crossover effects from the antiferromagnetic behavior to that of reentrant spin glass and spin glass behavior. Extensive studies have also been carried out on the incommensurate spin fluctuations in superconducting $La_2CuO_{4+\delta}$ (Ref. 7) and $La_{2-x}Sr_xCuO_4$ (Refs. 1,8) ($x \geqslant 0.05$).

In the vast majority of the neutron studies, the emphasis has been on quasielastic measurements (that is, integrating over energy) or true inelastic measurements. The latter, in particular, show significant effects at and below the superconducting transition temperature, especially for samples of $La_{2-x}Sr_xCuO_4$ with $x \simeq 0.15$, the optimal doping value.[9] Perhaps surprisingly, only a limited number of true elastic neutron measurements probing the static magnetic order have been carried out on the lamellar copper oxides. Early elastic measurements in single crystals of $La_2CuO_{4+\delta}$ with $T_N = 190$ K (Ref. 5) and ~ 100 K (Ref. 4) as well as $La_{2-x}Sr_xCuO_4$ with $x \simeq 0.04$ revealed novel effects at low temperatures. In each case, strong elastic scattering was observed at temperatures below ~ 40 K with intensities which increased progressively with decreasing temperature. In the antiferromagnetic samples, this 2D scattering was quite long ranged, that is, it was resolution limited; it was associated heuristically with the reentrant spin glass behavior[4] albeit with no real microscopic model. In $La_{1.96}Sr_{0.04}CuO_4$ the elastic scattering appeared at temperatures between 40 and 20 K depending on the energy window and had a characteristic length identical within the errors to the measured instantaneous correlation length of ~ 40 Å.[6] In all of these cases, the elastic scattering as well as the quasielastic peaks were commensurate.

Recently, rather dramatic elastic magnetic scattering effects have been observed in samples of $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ with $x=0.12$, 0.15, and 0.20.[10,11] These materials are all superconductors with onset T_c's of ~ 4, 11, and 15 K, respectively. In each case, Tranquada and co-workers[11] observe elastic incommensurate magnetic peaks with onset temperatures of ~ 50, 46, and 15 K, respectively. Recent work[11] has shown that in the $x=0.12$ sample the correlation length reaches its maximum value at temperatures below ~ 30 K. Presumably the larger x samples also order at comparable or lower temperatures. The incommensurabilities are essentially identical to those of the corresponding dynamic fluctuations in samples of $La_{2-x}Sr_xCuO_4$ with the same x.[8] However, there is one important difference between samples of a given x with and without Nd. The former are tetragonal below ~ 70 K whereas the latter retain the orthorhombic structure (labeled LTO) down to the lowest temperatures measured. Tranquada *et al.*[11] speculate that the tetragonal structure [labeled low-temperature tetragonal (LTT) to distinguish it

from the high-temperature tetragonal (HTT) structure] is essential to the static incommensurate magnetic ordering. A contrary interpretation has been given by Goto *et al.*[12] based on their NMR studies of $La_{2-x}Sr_xCuO_4(x\simeq 0.115)$ and $La_{2-x}Ba_xCuO_4(x\simeq 0.125)$.

Clearly, it is important to search for possible magnetic ordering in superconducting samples which are orthorhombic rather than tetragonal in order to determine the generality of the effects and to elucidate which features are essential for any such possible magnetic transitions. Of course, such studies will have important implications for any putative microscopic theoretical models. μSR (Ref. 13) and NMR measurements[12] in $La_{2-x}Sr_xCuO_4$ reveal static internal magnetic fields with onset temperatures which are a maximum for $x\simeq 0.12$. This suggests that one should concentrate on this region of the phase diagram. The first such measurements were carried out by Hirota *et al.*[14] on a sample of $La_{1.86}Sr_{0.14}Cu_{0.988}Zn_{0.012}O_{4+\delta}$ which could be made either superconducting [T_c(max)=19 K] or nonsuperconducting by varying δ. They indeed observe incommensurate short range magnetic order with a characteristic length of $\sim$80 Å which appears below $\sim$40 *K*; the general behavior is the same in superconducting and nonsuperconducting samples. Except for the incommensurability, their results are closely similar to those of Keimer *et al.*[6] in $La_{1.96}Sr_{0.04}CuO_4$. More recently, Suzuki *et al.*[15] have obtained evidence for incommensurate magnetic order at low temperatures in $La_{1.88}Sr_{0.12}CuO_4$ although no true elastic neutron-scattering measurements were carried out by them.

In this paper, we report elastic neutron-scattering studies of the magnetic order in $La_{1.88}Sr_{0.12}Cu_{1-y}Zn_yO_4$ both without ($y=0$) and with Zn ($y=0.03$). We also report cursory measurements in samples of $La_{1.90}Sr_{0.10}CuO_4$ and $La_{1.85}Sr_{0.15}CuO_4$. This set of measurements, therefore, serves to elucidate the separate dependences of the magnetic order in $La_{2-x}Sr_xCu_{1-y}Zn_yO_4$ on hole concentration through *x* and Cu^{2+} magnetic dilution through *y*.

The format of this paper is as follows. A description of the sample preparation and characterization and other experimental details are given in Sec II. The results of the neutron-scattering experiments are described in Sec III. Section IV contains a discussion of the results, especially in the context of previous work.

II. EXPERIMENTAL

Each of the single crystals used in the present experiments was grown by the traveling-solvent-floating-zone method. Details of the method of crystal growth are given elsewhere.[16] The sizes of the crystals with $x=0.12$ used in the neutron-scattering measurements are $6\phi\times 35$ mm^3 for $y=0.00$ and $7\phi\times 30$ mm^3 for $y=0.03$. As-grown crystals are annealed under pure oxygen gas flow at 900 °C for 50 h, cooled down to 500 °C at a rate of 10 °C per h, kept for 50 h at 500 °C and then cooled down slowly to room temperature. Some measurements were also performed on samples with $x=0.10$ and 0.15 and $y=0$; the complete characterization of these results is given elsewhere.[8]

The magnetic susceptibilities of the $x=0.12$ samples were measured with a superconducting quantum-interference device (SQUID) magnetometer after cooling in zero field at

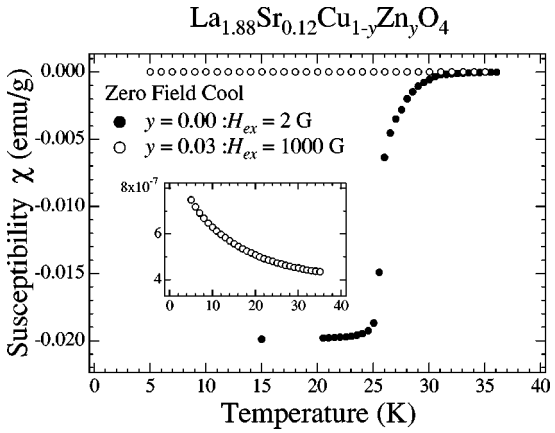

FIG. 1. Magnetic susceptibility measured after cooling in zero field for single crystals with $y=0.00$ (filled circles) and $y=0.03$ (open circles). The inset shows the detailed temperature dependence of the susceptibility for the $y=0.03$ crystal.

magnetic fields of 2 and 1000 G for $y=0.00$ and 0.03, respectively. As shown in Fig. 1, the $y=0.00$ crystal shows bulk superconductivity with $T_c=31.5$ (26.5 K) for the onset (middle) temperature of the transition. Both of these T_c's are almost identical to the values obtained previously on powder samples.[13,17,18] On the other hand, the Zn-substituted sample ($y=0.03$) shows no superconductivity; rather a Curie-Weiss-like upturn appears in the susceptibility (inset of Fig. 1).

The lattice constant for the Zn-free $x=0.12$ sample along the *c* axis was determined with an x-ray power diffractometer to be 13.229 Å at room temperature. The powder sample was prepared from a part of the single crystal used in the neutron-scattering measurements. The tetragonal-to-orthorhombic structural phase transition temperature T_{st} was determined by neutron-diffraction measurements in the single crystal to be 250 K from the temperature dependence of the $(\frac{3}{2},-\frac{3}{2},2)$ superlattice reflection. These values again agree well with previous results from ceramic[19,20] and single crystal[8] samples.

Most of the neutron-scattering experiments were performed on the triple-axis spectrometers HER at a cold neutron guide in JRR-3M in the Japan Atomic Energy Research Institute and SPINS in the National Institute of Standards and Technology (NIST) Center for Neutron Research. The incident (HER) or final (SPINS) neutron energy was fixed at 5.0 meV, and a Be filter was inserted in the incident (HER) or scattered (SPINS) beam to reduce any higher-order contamination. To monochromatize and analyze the energy of the neutron beam, the (0, 0, 2) reflection of pyrolytic graphite (PG) was used for both of the spectrometers. The energy resolution for the elastic measurements in this experiment was about 0.2 meV full width at half maximum (FWHM). Throughout this paper, we use reciprocal lattice units (rlu) for the HTT coordinate system, where the two short axes are defined as the distance between the nearest-neighbor Cu atoms along the in-plane Cu-O bond. At low temperatures the average in-plane lattice constant is $a=3.807$ Å. We mounted both crystals in the $(h, k, 0)$ plane to scan all four peaks at $(\frac{1}{2}\pm\epsilon,\frac{1}{2},0)$ and $(\frac{1}{2},\frac{1}{2}\pm\epsilon,0)$. The crystals were mounted in aluminum containers in which He gas was charged to act as a heat exchanger. A

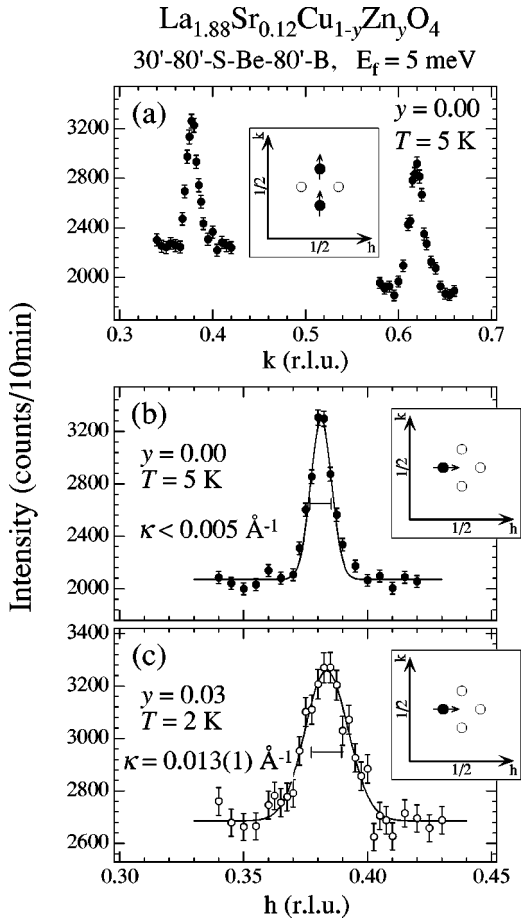

FIG. 2. Scans along $(\frac{1}{2},k,0)$ (a) and $(h,\frac{1}{2},0)$ (b), (c) through the incommensurate magnetic peaks at $\omega=0$ meV; these were measured on single crystals with $y=0.00$ (filled circles) (a), (b) and $y=0.03$ (open circles) (c). Measurements were done at 5 K (a),(b) and 2 K (c). The direction of each scan is shown by the arrow in the inset of each figure. The solid lines in (b),(c) are the results of fits of Gaussians convoluted with the instrumental resolution as discussed in the text; the resolution is shown with a solid short line.

top-loading liquid-He cryostat (ILL cryostat) was used to control the temperature of the crystals between 1.4 and 50 K.

III. RESULTS

Figures 2(a) and 2(b) show the elastic magnetic peaks around $(\frac{1}{2},\frac{1}{2},0)$ for the $x=0.12$, $y=0$ sample for scans along k and h. A scan for the $y=0.03$ sample along h is shown in Fig. 2(c). In order to make it possible to compare the peak-widths precisely, these two sets of data in Figs. 2(b) and 2(c) were taken on the NIST spectrometer SPINS with an identical configuration. Details of the spectrometer setup are shown at the top of all figures throughout this paper. The short horizontal lines denote the instrumental q resolution at the peak positions. The values of the incommensurability, $\epsilon=0.119(1)$ (rlu) for $y=0.00$ and 0.117(1) for $y=0.03$ are almost identical and also equal within the errors to that of the dynamical spin correlations at the same Sr ($x=0.12$) concentration.[8] A Gaussian line profile convoluted with the instrumental resolution together with a constant background is fit to each q spectrum. The solid lines in Figs. 2(b) and 2(c) represent the results of least square fitting. The intrinsic peak width κ_s half width at half maximum (HWHM) is determined to be <0.005 and 0.013(1) Å^{-1}, for the $y=0.00$ and 0.03 samples, respectively. It should be noted that for the Zn-free sample, the observed peak width is close to the instrumental q width. Accordingly, the static correlation length ξ_s defined as the inverse of κ_s is determined to be >200 Å for $y=0.00$, and ~ 80 Å for $y=0.03$.

After normalizing the data by sample volume, we find that the peak intensity is stronger for the Zn-free sample by a factor of about 2.5 compared with that for the Zn-substituted crystal while the integrated intensities are comparable. A rough evaluation of the size of the ordered moment, accurate to about a factor of 2 gives $0.1\mu_B$/Cu for both samples. This value overlaps that inferred from NMR measurements ($\sim 0.08\mu_B$/Cu), but appears to be smaller than that deduced from μSR measurement ($\sim 0.3\mu_B$/Cu). Complete three-dimensional scans as well as normalization of the doped sample magnetic Bragg scattering to that in pure La_2CuO_4 is necessary to determine the ordered moment more precisely. Such experiments are planned in the future.

In order to confirm that the elastic scattering is magnetic, we also carried out a cursory study of the scattering around the position $(\frac{3}{2},\frac{3}{2},0)$ at 8 and 50 K in the sample with $x=0$. Sharp peaks are observed at 8 K at the positions $(\frac{3}{2},\frac{3}{2}\pm\epsilon,0)$ with $\epsilon=0.12$ while at 50 K no scattering is observable above the background. The peak intensities observed at 8 K at $(\frac{3}{2},\frac{3}{2}\pm\epsilon,0)$ are about a factor of 4 less than those at the equivalent positions around $(\frac{1}{2},\frac{1}{2},0)$. This is consistent with the diminution expected from geometrical and magnetic form factors. By contrast, for nuclear scattering the structure factor should increase as Q^2 which is $\sim$ 9 between $(\frac{1}{2},\frac{1}{2}\pm\epsilon,0)$ and $(\frac{3}{2},\frac{3}{2}\pm\epsilon,0)$. We conclude, therefore, that the observed incommensurate elastic scattering is indeed magnetic in origin.

Figure 3 shows the temperature dependence of the $(\frac{1}{2}-\epsilon,\frac{1}{2},0)$ peak intensity in both $x=0.12$ samples. The peak first appears around $T_m=30$ K for the Zn-free sample, identical within the errors to the onset temperature T_c of the superconductivity. This onset temperature for the magnetic order agrees approximately with the result from NMR (Ref. 15) but is slightly higher than that inferred from μSR.[13] On the other hand, for the Zn-substituted nonsuperconducting sample, the broadened peak becomes observable below about 17 K.

Q scans of the elastic magnetic peak were performed for the $La_{1.88}Sr_{0.12}CuO_4$ sample to probe any anisotropy of the peak shape in the a^*-b^* plane. The arrows in the right panels of Fig. 4 represent the four directions of the scans. As in Fig. 2, the solid lines represent the results of least square fits of a Gaussian line shape convoluted with the instrumental resolution. The inverse correlation lengths parallel to the a and b axes so obtained are $\kappa_a=0.0055(11)$, and $\kappa_b=0.0070(7)$ Å^{-1} while those along the two diagonal directions are $\kappa_{[110]}=0.0017(46)$ and $\kappa_{[1\bar{1}0]}=0.0054(25)$ Å^{-1}. Therefore, the q width of the elastic peak is close to being resolution limited for all scan directions. We note that any possible effects of the twin structure on the peak profiles are not included in the fits. Thus, these values for κ_s represent

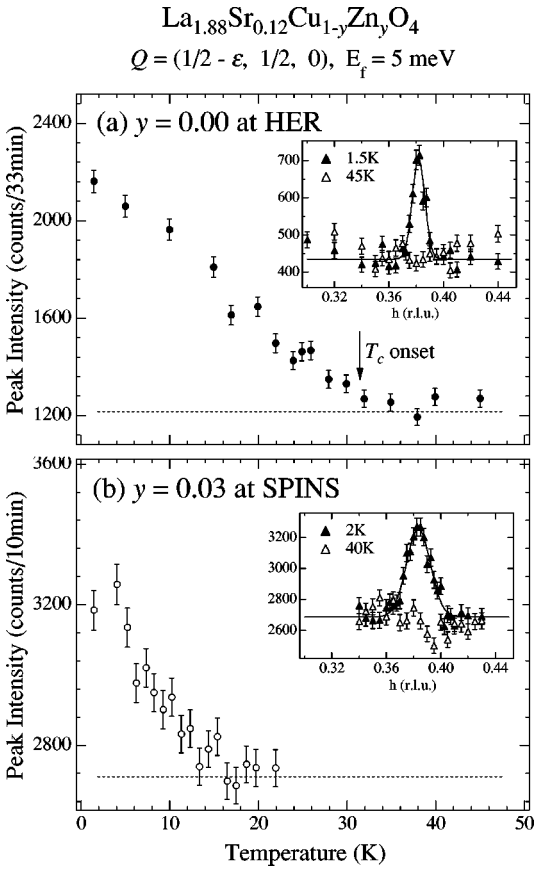

FIG. 3. Temperature dependence of the incommensurate elastic magnetic peak intensity at $Q=(\frac{1}{2}-\epsilon,\frac{1}{2},0)$ for (a) $y=0.00$ (filled circles) and (b) $y=0.03$ (open circles) crystals. Note that two different spectrometers, that is, HER and SPINS were used for the $y=0.00$ and $y=0.03$ samples, respectively. The inset in each figure shows peak profiles around $(\frac{1}{2}-\epsilon,\frac{1}{2},2)$ at high and low temperatures.

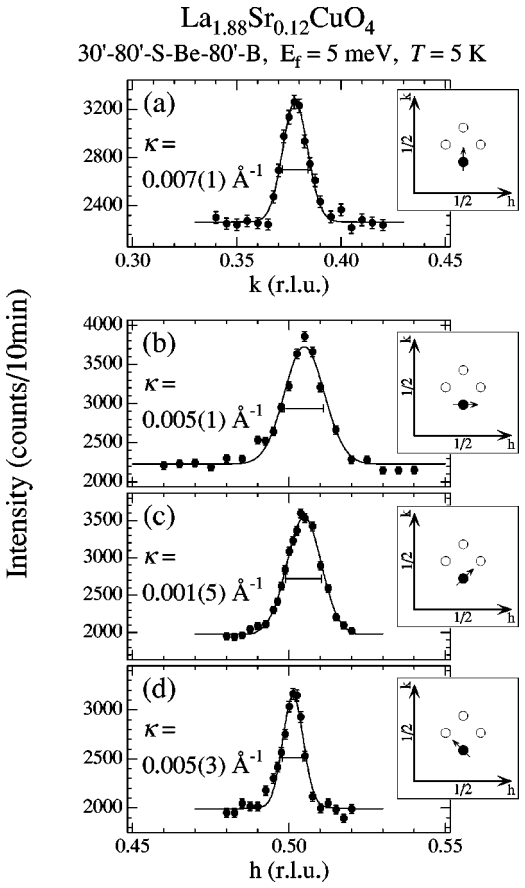

FIG. 4. q dependence of the incommensurate magnetic peak in the crystal with $y=0.00$ which was measured through the peak at $Q=(\frac{1}{2},\frac{1}{2}-\epsilon,0)$ at $T=5$ K. The arrow in the inset of each figure shows the direction of each scan. The solid line in each figure shows the result of fitting a Gaussian profile convoluted with the instrumental resolution as discussed in the text. The solid short line in each figure shows the width of the resolution at the peak position.

maximum possible values for the inverse correlation lengths. Specifically, our results are consistent with true long range order in all directions.

We searched for possible superlattice peaks associated with any concomitant charge ordering as was observed for $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ with $x=0.12$.[10,21,22] However, no well-defined peaks were found around $(1,\ 1-2\epsilon,l)$ and $(2-2\epsilon,\ 0,\ l)$ for $l=0$, and ± 3.5. These are charge peak positions corresponding to the magnetic peaks at $(\frac{1}{2}-\epsilon,\frac{1}{2},\ 0)$. Of course, any such scattering due to charge ordering could simply be below our level of detectability. We also carried out two-axis energy integrating measurements along l. These also gave no detectable signal above the background.

Finally, we carried out some cursory measurements to determine if any of the $La_{1.88}Sr_{0.12}CuO_4$ sample had transformed from the LTO to the LTT phase at low temperatures.[22–25] In the LTO phase, the reflections (1, 0, 0) and (0, 1, 0) are both forbidden although there may be some measurable intensity at these positions due to contamination from double scattering or from higher order neutrons. In the LTT phase these two reflections, which become identical for tetragonal symmetry, are allowed and the calculated relative intensity $I(1,\ 0,\ 0)/I(2,\ 0,\ 0)$ is $\sim 9.2\times 10^{-5}$. Note that here we continue to use the high-temperature tetragonal unit cell with lattice constant $a\simeq 3.807$ Å. The HTT to LTO structural phase transition temperature is 250 K. Using a coarse collimation configuration we find peak intensities $I(1,\ 0,\ 0)=3.33$ counts/sec at 270 K and $I(1,\ 0,\ 0)=3.75$ counts/sec at 8 K compared with $I(2,0,\ 0)=4.4\times 10^5$ counts/sec. This corresponds to a volume fraction of any LTT phase of at most 1%. Using a tight collimation configuration at 8 K, we find for the relative integrated intensity $I(0,\ 1,\ 0)/I(2,\ 0,\ 0)=8.7x10^{-7}$. If all of this scattering were from the putative LTT phase, this would correspond to a LTT volume fraction of $\sim 1\%$. In fact, the observed (0, 1, 0) scattering at 8K most likely originates primarily from double scattering and from higher order neutrons in the incident beam so that any LTT fraction must be much less than 1%.

We also report here some preliminary elastic measurements on samples of $La_{2-x}Sr_xCuO_4$ with $x=0.10$ and $x=0.15$. These measurements were carried on the SPINS triple-axis spectrometer at NIST. The final energy was fixed at 5 meV and the spectrometer configuration was $30'-80'-S-\mathrm{Be}-80'-B$ for $x=0.10$ and $30'-40'-S-\mathrm{Be}-40'-B$ for $x=0.15$. Here S denotes the sample, Be the beryllium filter, and B a blank collimator. The triple axis spectrometer was set for zero energy transfer, that is, it was set to

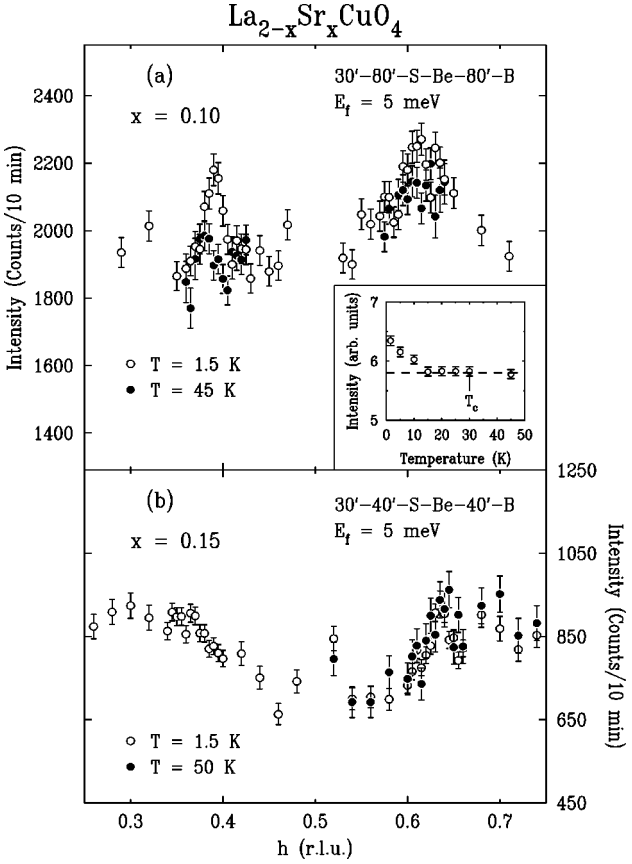

FIG. 5. Scans along $(h,\ h,\ 0)$ in $La_{2-x}Sr_xCuO_4$ in samples with $x=0.10$ (a) and $x=0.15$ (b) at $T=1.5$ K and at $T=45$ K($x=0.10$) or $T=50$ K($x=0.15$). The spectrometer configurations are given in the figures. The inset in (a) gives the temperature dependence of the integrated intensity measured with five-point scans.

detect elastic scattering with energy resolutions of 0.3 meV FWHM ($x=0.10$) and 0.2 meV FWHM ($x=0.15$). The samples were mounted in the $(h,k,0)$ zone and scans were carried out along $(h,\frac{1}{2},0)$.

Results of scans at $T=1.5$ K and $T=45$ K for the $x=0.10$ sample with $T_c=30$ K are shown in Fig. 5(a). As is evident from the figure, weak incommensurate elastic scattering is observed at 1.5 K. The temperature dependence of the integrated intensity of the peak (five-point scans) is shown in the inset of Fig. 5(a). The onset temperature of the elastic scattering appears to be near 15 K although more complete and accurate data will be needed to determine T_m precisely. Similar data for the $x=0.15$ sample with ($T_c=38$ K) are shown in Fig. 5(b). In this case, the background is quite irregular so it is difficult to make a strong statement. At this stage, we can only say that no significant incommensurate elastic magnetic scattering is observed in the $x=0.15$ sample in the superconducting state.

IV. DISCUSSION

These experiments have evinced a number of results which appear to be surprising. First, we have confirmed the inference of Suzuki *et al.*[15] that there is long range incommensurate static magnetic order at low temperatures in the superconducting state in $La_{1.88}Sr_{0.12}CuO_4$. Notably, the onset temperature T_m for the spin density wave order coincides with the superconducting transition temperature T_c to within the errors. At the minimum, this demonstrates that the spin density wave and superconducting states are not in competition with each other, barring of course, the unlikely coincidence that $La_{1.88}Sr_{0.12}CuO_4$ is exactly at a bicritical point or some other type of multicritical point in the phase diagram.

Second, we find that $La_{1.88}Sr_{0.12}CuO_4$ remains in the LTO phase down to the lowest temperatures measured and that any LTT phase, either short or long range ordered, must be present in concentrations below the 1% level. This confirms the assertion of Goto *et al.*[12] that magnetic order may occur whether the material is orthorhombic or tetragonal. This implies, therefore, that the incommensurate spin density wave ordering in the superconducting state in these monolayer high-T_c superconductors is a general phenomenon which does not depend on specific microscopic structural details. Concomitantly, any theory of the combined superconducting and magnetic order must be quite general.

In the $La_{2-x}Ba_xCuO_4$ (Refs. 22–24) and $La_{2-y-x}Nd_ySr_xCuO_4$ (Ref. 25) systems which are tetragonal at low temperatures the superconductivity is suppressed around $x\simeq 0.12$. At the same time, the spin density wave moment and ordering temperatures have their maximum values for $x\simeq 0.12$. This led previous investigators naturally to the conclusion that having the hole concentrations near $p\simeq\frac{1}{8}$ as well as a tetragonal crystal structure were both essential for these phenomena. As noted above, our results suggest that the tetragonality does not play a crucial role. However, it seems that having a hole concentration near $p=\frac{1}{8}$ is indeed fundamental for the static magnetic ordering. This was also presaged by the work of Yamada *et al.*[8] who showed that the low-energy dynamical coherence length was a maximum for $p\simeq\frac{1}{8}$. The interplay of the magnetic order with the superconductivity is less clear. Although the onset temperature of the superconductivity in $La_{1.88}Sr_{0.12}CuO_4$ is intermediate between that for $x=0.10$ and $x=0.15$, the midpoint temperature of the superconducting transition is slightly suppressed. One possible interpretation of these data is that there is a sharp minimum in T_c very close to $x=0.12$ (Ref. 12) and that the higher onset temperature is due to the small gradient in the Sr concentration.[8] This would suggest some subtle deleterious effect on the superconductivity for hole concentrations near $p=1/8$.

We note that the incommensurate wave vector of the magnetic ordering for the $x=0.12$ sample is $\epsilon=0.119$ (1) rlu which is close to but not identical to the high order commensurate value $\epsilon=0.125$. In $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$, Tranquada and co-workers[11] also found incommensurabilities close to, but not identical to 1/8 while in $La_{1.86}Sr_{0.14}Cu_{0.988}Zn_{0.012}O_{4+\delta}$ Hirota *et al.*[14] find $\epsilon=0.132$ (1) rlu. Clearly, therefore, $\epsilon\sim 1/8$ is most stable, but strict commensurability is not required. Current data for the orthorhombic materials are not precise enough to allow one to make any statement about any possible anisotropy in the incommensurability in the two orthorhombic directions.

Our results in the Zn-doped sample $La_{1.88}Sr_{0.12}Cu_{0.97}Zn_{0.03}0_4$ are similar to those of Hirota *et al.*[14] in $La_{1.86}Sr_{0.14}Cu_{0.988}Zn_{0.012}0_{4+\delta}$. Specifically, the Zn vitiates the spin density wave transition to long range order. Instead, for $x=0.12$ and $y=0.03$ below ~ 17 K an interme-

diate range, ~80 Å, spin density wave state develops. A Zn concentration of 0.03 is also sufficient to destroy the superconductivity entirely. In the $x=0.14$, $y=0.012$ sample, the magnetic behavior is similar to that for $x=0.12$, $y=0.03$ while Hirota *et al.*[14] find that the superconductivity in the 1.2% Zn crystal depends on the oxygen stoichiometry. Presumably, varying the oxygen concentration δ changes both the hole concentration and the integrity of the CuO_2 planes. In brief, therefore, replacement of Cu by Zn is destructive of both the superconductivity and the incommensurate spin density wave long range order. As argued by Hirota *et al.*,[14] Zn substitution may nevertheless serve to stabilize a short range order incommensurate static spin density wave state which might otherwise be purely dynamic. The short range magnetic order is naturally explained by noting that the Zn-doped system should correspond to a two-dimensional XY model in a random field.

One question which we have not yet addressed is the relationship of the incommensurate elastic magnetic scattering observed in the superconducting samples with the commensurate elastic scattering observed in samples with hole concentrations less than ~0.05. For the latter, the onset temperature depends explicitly on the energy window; for example, in Keimer's $x=0.04$ sample, the apparent transition temperature varies from ~40 K as deduced from the staggered susceptibility with a 0.9 meV energy window[6] to ~7.2 K for dc susceptibility measurements.[26] This is prototypical behavior for a glass transition. The one unusual feature in the experiment of Keimer *et al.*[6] is that the elastic component correlation length coincides with the instantaneous correlation length.[27] In $La_{1.88}Sr_{0.12}CuO_4$, it appears that there is a regular second order mean field transition to true long range magnetic order. μSR or NMR measurements on the explicit sample studied in our neutron experiments would serve to test this assertion. Specifically, the transition temperature should not depend on the energy window. It is possible that the Zn doping will change the character of the transition to be more glasslike. This will be the subject of future research.

As noted in Sec. III, we do not have any direct evidence for charge ordering although more extensive and precise measurements are required to rule it out. Our results show unambiguously that spin density wave and superconducting order may be concordant. On the other hand, as emphasized by Emery and co-workers,[28] in their stripe model charge density wave ordering and superconductivity should be discordant. Clearly, therefore, further exploration of the charge density degrees of freedom is required. Finally, in $La_{1.88}Sr_{0.12}CuO_4$ the superconducting and magnetic transitions coincide. This has several possible explanations: first, it may be a simple coincidence, second, it could imply that this sample sits at a special point in the phase diagram, or third, this may be generic behavior for incommensurate spin density wave ordering in relatively clean samples with hole concentrations near 1/8. Measurements on other systems such as low stage $La_2CuO_{4+\delta}$ will be necessary to probe the generality of this result.

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

[2] M. Greven, R. J. Birgeneau, Y. Endoh, M. A. Kastner, M. Matsuda, and G. Shirane, Z. Phys. B **96**, 465 (1995).

[3] R. J. Birgeneau, A. Aharony, N. R. Belk, F. C. Chou, Y. Endoh, M. Greven, S. Hosoya, M. A. Kastner, C. H. Lee, Y. S. Lee, G. Shirane, S. Wakimoto, B. O. Wells, and K. Yamada, J. Phys. Chem. Solids **56**, 1913 (1995).

[4] Y. Endoh, K. Yamada, R. J. Birgeneau, D. R. Gabbe, H. P. Jenssen, M. A. Kastner, C. J. Peters, P. J. Picone, T. R. Thurston, J. M. Tranquada, G. Shirane, Y. Hidaka, M. Oda, Y. Enomoto, M. Suzuki, and T. Murakami, Phys. Rev. B **37**, 7443 (1988).

[5] C. J. Peters, R. J. Birgeneau, M. A. Kastner, H. Yoshizawa, J. M. Tranquada, G. Shirane, Y. Hidaka, M. Oda, M. Suzuki, and T. Murakami, Phys. Rev. B **37**, 9761 (1988).

[6] 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**, 14 034 (1992).

[7] 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).

[8] 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**, 6165 (1998).

[9] K. Yamada, S. Wakimoto, G. Shirane, C. H. Lee, M. A. Kastner, S. Hosoya, M. Greven, Y. Endoh, and R. J. Birgeneau, Phys. Rev. Lett. **75**, 1626 (1995).

[10] J. M. Tranquada, J. D. Axe, N. Ichikawa, Y. Nakamura, S. Uchida, and B. Nachumi, Phys. Rev. B **54**, 7489 (1996).

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

[12] T. Goto, S. Kazama, K. Miyagawa, and T. Fukase, J. Phys. Soc. Jpn. **63**, 3494 (1994).

[13] K. Kumajai, K. Kawano, I. Watanabe, K. Nishiyama, and K. Nagamine, J. Supercond. **7**, 63 (1994).
[14] K. Hirota, K. Yamada, I. Tanaka, and H. Kojima, Physica B **241-243**, 817 (1998).
[15] T. Suzuki, T. Goto, K. Chiba, T. Shjinoda, T. Fukase, H. Kimura, K. Yamada, M. Ohashi, and Y. Yamaguchi, Phys. Rev. B **57**, R3229 (1998).
[16] S. Hosoya, C. H. Lee, S. Wakimoto, K. Yamada, and Y. Endoh, Physica C **235-240**, 547 (1994).
[17] H. Takagi, T. Ido, S. Ishibashi, M. Yota, S. Uchida, and Y. Tokura, Phys. Rev. B **40**, 2254 (1989).
[18] T. Nagano, Y. Tomioka, Y. Nakayama, K. Kishio, and K. Kitazawa, Phys. Rev. B **48**, 9689 (1993).
[19] H. Takagi, R. J. Cava, M. Merezio, B. Batlogg, J. J. Krajewski, W. F. Peck, Jr., P. Bordet, and D. E. Cox, Phys. Rev. Lett. **68**, 3777 (1992).
[20] E. Takayama-Muromachi and D. E. Rice, Physica C **177**, 195 (1991).
[21] M. V. Zimmerann, A. Vigiliante, T. Niemoller, N. Ichikawa, T. Frello, J. Madsen, P. Wochner, S. Uchida, N. H. Andersen, J. M. Tranquada, L. D. Gibbs, and J. R. Schneider, Europhys. Lett. **41**, 629 (1998).
[22] J. D. Axe, A. H. Moudden, D. Hohlwein, D. E. Cox, K. M. Mohanty, A. R. Moodenbaugh, and Y. Xu, Phys. Rev. Lett. **62**, 2751 (1989).
[23] T. Suzuki and T. Fujita, J. Phys. Soc. Jpn. **58**, 1883 (1989).
[24] G. M. Luke, L. P. Le, B. J. Sternlieb, W. D. Wu, Y. J. Uemura, J. H. Brower, T. M. Riseman, B. Ishibashi, and S. Uchida, Physica C **185-189**, 1175 (1991).
[25] M. K. Crawford, R. L. Harlow, E. M. Macarron, W. E. Farneth, J. D. Axe, H. Chou, and Q. Huang, Phys. Rev. B **44**, 7749 (1991).
[26] F. C. Chou, N. R. Belk, M. A. Kastner, R. J. Birgeneau, and A. Aharony, Phys. Rev. Lett. **75**, 2204 (1995).
[27] R. J. Gooding, N. M. Solem, R. J. Birgeneau, and F. C. Chou, Phys. Rev. B **55**, 6360 (1997).
[28] S.A. Kivelson, E. Fradkin, and V. J. Emery, Nature (London) **393**, 550 (1998).