Incommensurate Spin Ordering and Fluctuations in underdoped La$_{2-x}$Ba$_x$CuO$_4$

S. R. Dunsiger,$^1$ Y. Zhao,$^1$ Z. Yamani,$^2$ W. J. L. Buyers,$^{2,3}$ H. Dabkowska,$^1$ and B. D. Gaulin$^1$,$^3$

$^1$Department of Physics and Astronomy, McMaster University, Hamilton, Ontario, L8S 4M1, Canada.
$^2$Canadian Neutron Beam Centre, NRC, Chalk River Laboratories, Chalk River, Ontario, K0J 1J0, Canada
$^3$Canadian Institute for Advanced Research, 180 Dundas St. W., Toronto, Ontario, M5G 1Z8, Canada.

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Using neutron scattering techniques, we have studied incommensurate spin ordering as well as low energy spin dynamics in single crystal underdoped La$_{2-x}$Ba$_x$CuO$_4$ with $x$=0.095 and 0.08; high temperature superconductors with $T_C$ $\sim$ 27 K and 29 K respectively. Static two dimensional incommensurate magnetic order appears below $T_N$=39.5 $\pm$ 0.3 K in La$_{2-x}$Ba$_x$CuO$_4$($x$=0.095) and a similar temperature for $x$=0.08 within the low temperature tetragonal phase. The spin order is unaffected by either the onset of superconductivity or the application of magnetic fields of up to 7 Tesla applied along the c-axis in the x=0.095 sample. Such magnetic field independent behaviour is in marked contrast with the field induced enhancement of the staggered magnetisation observed in the related La$_{2-x}$Sr$_x$CuO$_4$ system, indicating this phenomenon is not a universal property of cuprate superconductors. Surprisingly, we find that incommensurability $\delta$ is only weakly dependent on doping relative to La$_{2-x}$Sr$_x$CuO$_4$. Dispersive excitations in La$_{2-x}$Ba$_x$CuO$_4$ ($x$=0.095) at the same incommensurate wavevector persist up to at least 60 K. The dynamical spin susceptibility of the low energy spin excitations saturates below $T_C$, in a similar manner to that seen in the superconducting state of La$_2$CuO$_{4+y}$.

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The roles of spin, charge and lattice degrees of freedom have been central to the rich behaviour brought to light over the last 20 years in superconducting lamellar copper oxides$^{1,2,3}$. In particular, the cuprates exhibit phenomena which are a sensitive function of doping, evolving from an antiferromagnetic insulating parent compound into a superconducting phase with increasing hole density. A heterogeneous electronic phase composed of stripes of itinerant charges now appears to be a generic feature of hole doped ternary transition metal oxides$^4$, such as manganites$^{5,6}$ and nickelates$^7$, as well as cuprates. The explanation for these incommensurate spin ordered states is the subject of ongoing debate. In an itinerant picture, the spin dynamics are described in terms of electron-hole pair excitations about an underlying Fermi surface$^{1,2,3,12,13}$. Alternatively, within the stripe picture of doped, two dimensional Mott insulators, the non-magnetic holes in these materials organize into quasi-one dimensional stripes which separate antiferromagnetic insulating antiphase domains$^{14}$. Adjacent antiferromagnetic regions are $\pi$ out of phase with each other giving rise to a magnetic structure with incommensurate periodicity, where the supercell dimension is twice the hole stripe periodicity.

The static spin structures in the undoped, parent compounds such as La$_2$CuO$_4$ or YBa$_2$Cu$_3$O$_{6+y}$ have been determined by neutron scattering to be relatively simple two sublattice antiferromagnets characterized by a commensurate ordering wavevector of (0.5,0.5) in reciprocal lattice units within the tetragonal basal plane. On hole doping with either Sr substituting for La in La$_{2-x}$Sr$_x$CuO$_4$ or by adding additional oxygen in YBa$_2$Cu$_{3}$O$_{6+y}$, the magnetic scattering moves out to incommensurate wavevectors, consistent with the stripe ordering picture described above. This incommensurate magnetism can be either static or dynamic, as evidenced by either elastic or inelastic peaks in the neutron scattering respectively and now appears to be a common feature of the La$_{2-x}$Sr$_x$CuO$_4$ family of compounds. Specifically, for lightly doped La$_{2-x}$Sr$_x$CuO$_4$, elastic incommensurate magnetic Bragg features first appear split off from the (0.5,0.5,0) position in diagonal directions relative to a tetragonal unit cell$^{20,21}$. At higher doping in the underdoped superconducting regime, the peaks rotate by 45° to lie along directions parallel to the tetragonal axes or Cu-O-Cu bonds, such that elastic magnetic scattering appears at (0.5$\pm$0.5,0) and (0.5,0.5$\pm$0) and persists$^{23,24}$. For optimal and higher doping the static order disappears, but dynamic incommensurate correlations nevertheless persist$^{23,24}$.

Within the stripe picture, one expects charge ordering associated with the holes to occur at an incommensurate wavevector 26, twice that describing the spin order. Neutron scattering is not directly sensitive to charge ordering per se, but it is sensitive to atomic displacements, such as those associated with oxygen, which arise from charge ordering. An incommensurate nuclear scattering signature is therefore expected to appear at (2$\pm$0,0,0) or (2$\pm$0,0,0) and related wavevectors. Despite extensive efforts, such incommensurate charge related scattering has not been observed in La$_{2-x}$Sr$_x$CuO$_4$ by either neutron or X-ray scattering techniques, although there is indirect evidence of charge stripe excitations from optical measurements$^{25}$. Such scattering has been observed in La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ and La$_{1.875}$Ba$_{0.125}$Sr$_x$CuO$_4$ ($x$=0.0,0.05,0.06,0.075,0.085$^{26}$, motivating discussion as to whether such static charge stripes compete with, rather than underlie, high temper-
FIG. 1: Zero field cooled (ZFC) and field cooled (FC) susceptibilities of La$_{2-x}$Ba$_x$CuO$_4$ (x=0.095 and 0.08) single crystals measured at 0.001 T. Dashed lines indicate the onset of superconductivity at $T_C=27$ and 29 K for x=0.095 and x=0.08 samples respectively.

Surprisingly, La$_{2-x}$Ba$_x$CuO$_4$, the first high temperature superconductor (HTSC) to be discovered, has been much less extensively studied than either La$_{2-x}$Sr$_x$CuO$_4$ or YBa$_2$Cu$_3$O$_{6+x}$ due to the difficulty of growing single crystals, which has only been achieved recently. In this paper, we report neutron scattering signatures of static incommensurate spin order in single crystal La$_{2-x}$Ba$_x$CuO$_4$ (x=0.095, 0.08), consistent with the stripe picture described above, but with interesting complexity not accounted for within present theoretical models. Our results clearly indicate that the spin ordering is insensitive to both the onset of superconductivity and surprisingly the application of a magnetic field. Tranquada et al. and Fujita et al. have reported neutron scattering measurements on an x=0.125 sample of La$_{2-x}$Ba$_x$CuO$_4$. In both La$_{2-x}$Ba$_x$CuO$_4$ and La$_{2-x}$Sr$_x$CuO$_4$ this concentration corresponds to a suppression of $T_C$ as a function of doping, known as the “1/8” anomaly. In La$_{2-x}$Ba$_x$CuO$_4$ the suppression is almost complete and is associated with a structural phase transition at low temperature, from orthorhombic to tetragonal which gives rise to a superlattice peak at (0,1,0) and symmetry related reflections. Samples of La$_{2-x}$Ba$_x$CuO$_4$ near x=0.125 display a sequence of structures on lowering the temperature, going progressively from high temperature tetragonal (HTT, I4/mmm symmetry) to orthorhombic (MTO, mnb symmetry) to low temperature tetragonal (LTT, P4$_{2}$(ncm symmetry). The high temperature tetragonal to orthorhombic transition in particular, and to a lesser extent, the orthorhombic to low temperature tetragonal transition are sensitive indicators for the precise Ba doping level in the material.

FIG. 2: a) Static incommensurate magnetic peaks with $\delta=0.112$ in La$_{1.905}$Ba$_{0.095}$CuO$_4$ at $T=3.8$ K, along (H,0.5,0). b) Representative inelastic scans at T=30 K, also along (H, 0.5, 0) and at $\hbar\omega=2.07$ meV. Parameters characterizing this inelastic scattering are shown in Fig. 4, while the solid line is discussed in the text. c) Elastic scans of the form (H, H/(1-2$\delta$), L), which demonstrate the rod-like, two dimensional nature of the elastic magnetic scattering, as described in the text. The scan at L=2.5 has been displaced by 50 counts upwards for clarity.

I. SINGLE CRYSTAL GROWTH, CHARACTERIZATION AND EXPERIMENTAL PROCEDURE

We have grown high quality single crystals of La$_{2-x}$Ba$_x$CuO$_4$ with x=0.095 and x=0.08 using floating zone image furnace techniques with a four-mirror optical furnace. A small single crystal of La$_2$CuO$_4$ was employed as a seed for the growth, which was performed under enclosed pressures of 165 and 182 kPa of O$_2$ gas for
From Table I: Summary of La$_{2-x}$Ba$_x$CuO$_4$ structural ($T_d1$, $T_d2$), superconducting ($T_c$) and magnetic ($T_N$) phase transition temperatures\textsuperscript{32}

| $x$  | $T_d1$ (K) | $T_d2$ (K) | $T_c$ (K) | $T_N$ (K) |
|------|------------|------------|-----------|-----------|
| 0.125 | 232        | 60         | $\sim 4$  | 50        |
| 0.095 | 272        | 45         | 27        | 39.5      |
| 0.08  | 305        | 35         | 29        | 39        |

\textsuperscript{a}From \textsuperscript{32}.

$x=0.095$ and $x=0.08$ samples respectively. The resulting \~6 gram single crystals of La$_{2-x}$Ba$_x$CuO$_4$ were cylindrical in shape and cut to dimensions 25 mm in length by 5 mm in diameter ($x=0.095$) and 38 mm in length by 5 mm diameter ($x=0.08$). We have determined the Ba concentration from the HTT to MTO transition temperature. The La$_{2-x}$Ba$_x$CuO$_4$ ($x=0.095$) and ($x=0.08$) single crystals displayed HTT to MTO structural phase transitions at $T_d1\sim 272$ K and $T_d1\sim 305$ K, respectively, and MTO to LTT transitions at $T_d2\sim 45$ K and $T_d2\sim 35$ K, respectively\textsuperscript{32}. The bulk superconducting transition temperatures $T_C=27$ K ($x=0.095$) and $T_C=29$ K ($x=0.08$) are identified using the onset of the zero field cooled diamagnetic response of the crystals, as measured using Superconducting Quantum Interference Device (SQUID) magnetometry and shown in Fig. 1. The various structural and superconducting phase transition temperatures are summarised in Table 1, along with the corresponding values for the La$_{2-x}$Ba$_x$CuO$_4$ ($x=0.125$) sample\textsuperscript{32}.

Oxygen stoichiometry in La$_{2-x}$Ba$_x$CuO$_4$+$y$ is more difficult to quantify, especially at the low levels relevant here. Experience with La$_{2-x}$Sr$_x$CuO$_4$+$y$ suggests the oxygen stoichiometry, $y$, is negative in as-grown samples, giving rise to crystals which possess an effective doping level which is lower than that given by the Sr concentration alone; the effective doping level is $x+2y$\textsuperscript{32}. Stoichiometric samples at optimal and underdoped Sr concentrations display a maximum superconducting $T_C$ which cannot be increased by controlled annealing in O$_2$ gas. Excess oxygen can be incorporated into LaCuO$_4$+$y$ crystals, but only through electrochemical doping methods, in which case $y$ can be as high as 0.11 and the interstitial oxygen organizes itself into staged structures\textsuperscript{36}.

Neutron scattering experiments were undertaken on the C5 and N5 triple axis spectrometers at the Canadian Neutron Beam Centre, Chalk River. All experiments were performed with pyrolytic graphite (002) planes as monochromator and analyser, with constant $E_f=14.7$ meV. A graphite filter was placed in the scattered beam to reduce contamination from higher order neutrons. The single crystals were oriented with (H,K,0) in the horizontal scattering plane. Crystallographic indices are denoted using tetragonal notation, where the basal plane lattice constant $a=3.78$ Å at low temperatures.

II. ELASTIC NEUTRON SCATTERING IN La$_{2-x}$Ba$_x$CuO$_4$ ($x=0.095$)

A. Meissner State

FIG. 3: a) The temperature dependence of the net elastic incommensurate magnetic scattering in La$_{2-x}$Ba$_x$CuO$_4$ ($x=0.095$) at (0.612,0.5,0) and ($x=0.08$) at (0.5,0.607,0), as well as that of b) the (0,1,0) structural Bragg peak, which marks the orthorhombic to low temperature tetragonal structural phase transition. Note that a constant background has been subtracted in both cases. The superconducting and structural phase transition temperatures are indicated by dashed lines for both samples. All the data for $x=0.08$ have been scaled to the volume of the $x=0.095$ sample by phonon normalization. The counting times refer to the unscaled $x=0.095$ data. c) Temperature dependence of the elastic incommensurate magnetic scattering in La$_{2-x}$Ba$_x$CuO$_4$ ($x=0.095$) in 0 and H=7 T $\parallel$ c.
We discuss the more extensive measurements on the La$_{2-x}$Ba$_x$CuO$_4$ sample with x=0.095 first. Elastic scattering scans at T=3.8 K are shown in Fig 2a), in which Bragg peaks occur at (0.5±δ,0.5,0) δ = 0.112(3), indicating static incommensurate spin order. Analogous magnetic Bragg peaks are observed at (0.5,0.5±0.112,0).

All peak widths are resolution limited with full widths at half maximum, FWHM=0.011 Å$^{-1}$, indicating static spin correlations within the basal plane exceeding 180 Å. By contrast, the spin correlations between planes are very short. To observe this, the crystal was reoriented in the (H,H,L) scattering plane and then tilted $\sim 7^\circ$ at constant L to intersect the incommensurate peak position for H=0.39. Measurements along H of the form (H,H/(1-2δ),L) at fixed L for L=2.5 and 3 are shown in Fig. 2c at T=3.5 K and at L=3 for T=50 K to extract a background. Since the peak intensity is independent of L, the scattering taking the form of an elastic rod along the L direction, the static spin order at low temperatures is two dimensional. Note that the intensity in Fig. 2a) is greater relative to that in Fig. 2c) for the sample oriented in the (H,K,0) plane. This arises because the neutron spectrometer has a broad vertical resolution which integrates the signal in the L direction, perpendicular to the scattering plane.

The temperature dependence of the incommensurate magnetic elastic scattering is illustrated in Fig. 3a. The intensity of the magnetic Bragg peak is proportional to the volume average of the square of the ordered staggered moment. The spin order at (0.612,0.5,0) develops continuously with temperature below $T_N=39.5\pm0.3$ K. The temperature dependence of the (0,1,0) structural Bragg peak indicates the orthorhombic to low temperature tetragonal phase transition occurs at $T_{2D}=45$ K, a transition which is discontinuous in nature$^{27}$. For reference, the superconducting transition at $T_C=27$ K (see Fig. 1) is also indicated on this plot as a dashed line. The onset of spin ordering, $T_N$ correlates most strongly with the completion of the transition to the low temperature tetragonal phase, and the incommensurate spin order coexists with the superconductivity below $T_C$. Associated incommensurate charge ordering has not been observed. The temperature dependence of the spin ordering is qualitatively similar to that observed in the x=1/8 compound$^{22}$, where the superlattice peak intensity becomes non-zero below $\sim 50$ K. Similarly, no anomaly has been observed at $T_C$ in YBCO$_{6.35}$ in the spin order, which has been attributed to robust spin correlations$^{19}$.

B. Magnetic Field Dependence

The most surprising result of this study is that the incommensurate spin structure shows no magnetic field dependence up to 7 T, applied vertically along the c* axis. Neither cooling nor warming the sample in a magnetic field has an effect on either the temperature dependence of the spin ordering, or the Bragg intensity in La$_{2-x}$Ba$_x$CuO$_4$ (x=0.095), as shown in Fig. 3c. This result is in marked contrast with the behaviour of underdoped and optimally doped La$_{2-x}$Sr$_x$CuO$_4$ where pronounced field dependent effects are observed. For the optimally doped La$_{2-x}$Sr$_x$CuO$_4$ compound (x=0.163), the application of a magnetic field enhances the dynamical spin susceptibility but does not induce static order$^{28}$. Most dramatically, in a slightly underdoped sample (x=0.144), Khaykovich et al$^{29}$ report the development of a static incommensurate spin structure above a critical field of 2.7 T. The authors therefore argue that La$_{2-x}$Sr$_x$CuO$_4$ (x=0.144) may be tuned through a quantum critical point, at which there is a magnetic field induced transition between magnetically disordered and ordered phases. Their results are interpreted in terms of a Ginzburg-Landau model due to Demler et al$^{30}$, which assumes a microscopic competition between spin and superconducting order parameters. The predicted magnetic intensity increases as $\Delta I \sim H/H_C \ln(H_C/H)$, which is consistent with experiments on La$_{2-x}$Sr$_x$CuO$_4$.

Note that the intensity changes most rapidly with magnetic field at low fields on a scale set by $H_C$. In the La$_{2-x}$Ba$_x$CuO$_4$ family of compounds, the lower critical field is $H_{C1} \sim 0.04$ T, while the upper critical field $H_{C2}$ is in excess of 40 T$^{11}$. The upper critical field is of the same order of magnitude in optimally doped La$_{2-x}$Sr$_x$CuO$_4$. Thus an applied magnetic field of 7 T should be sufficiently large to see an effect in La$_{2-x}$Ba$_x$CuO$_4$.

For sufficiently underdoped La$_{2-x}$Sr$_x$CuO$_4$ (x=0.12,0.10), the ordered magnetic moment associated with preexisting static spin order is enhanced on application of a magnetic field$^{33,41,42}$. The spin order within the vortex state of La$_{2-x}$Sr$_x$CuO$_4$ (x=0.10)$^{41}$ indicates long in-plane correlation lengths, greater than both the superconducting coherence length and the intervortex spacing at 14.5 T. As the coherence length is a measure of the size of the vortices, Lake et al argue the static magnetism must therefore reside beyond the extent of the vortices themselves$^{41}$. Whereas the La$_{2-x}$Ba$_x$CuO$_4$ (x=0.095) correlation length for static spin order is similarly long, the underlying physics is clearly different and the spins appear to order independent of vortex creation.

III. INELASTIC NEUTRON SCATTERING IN La$_{2-x}$Ba$_x$CuO$_4$ (x=0.095)

The magnetic excitations were studied in constant energy transfer scans performed through the incommensurate ordering wavevectors. Horizontal collimation sequences of 0.54°-0.48°$^\circ$S-0.54°-1.2° and 0.54°-0.79°$^\circ$S-0.85°-2.4° were used at energy transfers of 2.07 and 3.1 meV respectively, yielding corresponding energy resolutions of $\sim 1$ and $\sim 1.5$ meV FWHM. The representative scan along (H,0.5,0) and $\hbar \omega=2.07$ meV at $T=30$ K in Fig. 2b, shows that the low energy dynamic spin response peaks up at the same wavevector, (0.5±0.112,0.5,0), as
and \( Q_{\delta,n} \) represents the four incommensurate wave vectors \((\frac{x}{2} \pm \delta, \frac{y}{2}, 0)\) and \((\frac{x}{2}, \frac{y}{2} \pm \delta, 0)\). This assumes the magnetic excitations consist of four rods of scattering running along the \( c^* \) axis. The extracted temperature dependences of \( \chi''(h\omega) \), \( \delta \) and \( \kappa \) are plotted in Figs. 4a, b and c respectively. \( \chi''(h\omega) \) is proportional to the integral of \( \chi''(Q, \omega) \) over \( Q \) in the (H,K,0) scattering plane; \( \delta \) is the incommensurability, while \( \kappa \) is the inverse of the static correlation length in the basal plane, defined as the peak half width at half maximum. For reference, both the spin ordering transition at \( T_N \sim 39.5 \pm 0.3 \text{ K} \), and the superconducting transition near \( T_C \sim 27 \text{ K} \) are indicated on this plot. At both 2.07 meV and 3.1 meV, the dynamical susceptibility, \( \chi''(h\omega) \), increases continuously as the temperature is reduced below \( \sim 60 \text{ K} \), becoming roughly constant and non-zero below \( T_C \sim 27 \text{ K} \). This is similar to measurements in both overdoped \( \text{La}_2\text{CuO}_4+y \), where a levelling off of the dynamic incommensurate spin response has been reported below \( T_C \sim 42 \text{ K} \) and also in \( \text{La}_{2-x}\text{Ba}_x\text{CuO}_4 \) (\( x=0.125 \)) in the normal state\(^{52} \). In the latter compound, as a function of frequency there is relatively little change in \( \chi''(h\omega) \) at low temperature (8 K), whereas it drops rapidly in the present \( x=0.095 \) sample. As the temperature is raised, \( \chi''(h\omega) \) varies linearly with frequency at lower energy transfers below 10 meV in the \( x=0.125 \) sample for \( T>65 \text{ K} \), whereas it declines with increasing \( \omega \) in the \( x=0.095 \) for all \( T<60 \text{ K} \). Note that the signal intensity has been corrected for the monitor sensitivity to higher order incident neutrons, as described in Ref.\(^{47} \). These low energy excitations have some characteristics of the spin waves observed in the parent compound \( \text{La}_2\text{CuO}_4 \) as one warms through the Néel temperature, where instantaneous spin correlations with the character of the Néel state persist into the paramagnetic regime\(^{49} \). The form of \( \chi''(h\omega) \) varies dramatically as a function of doping in the related \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) compounds. In optimally and slightly overdoped \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4(x=0.15,0.18) \) there is a characteristic energy of \( \sim 7 \text{ meV} \) below which the dynamic susceptibility is dramatically reduced in the superconducting state - the opening up of a spin gap. However, on the underdoped side of the superconducting dome there is finite spectral weight in the spin response at all low energy transfers\(^{52,53} \). The bottom two panels of Fig 4 show that the incommensurability, \( \delta \), and the inverse correlation length, \( \kappa \), correlate most strongly with the disappearance of the static spin order near \( T_N \sim 39 \text{ K} \), which is not surprising. Above \( T_N \) the increase in \( \kappa \) may indicate that stripe correlations are weakened by thermal fluctuations that broaden the hole distribution about antiphase domain boundaries.
IV. ELASTIC NEUTRON SCATTERING IN La$_{2-x}$Ba$_x$CuO$_4$ (X=0.08)

Qualitatively, the magnetic and superconducting properties of La$_{2-x}$Ba$_x$CuO$_4$ (x=0.08) ($T_C$=29 K) are very similar to those of the higher doped x=0.095 ($T_C$=27 K) sample. Elastic neutron scattering measurements were carried out under the same conditions as described earlier, with the single crystal oriented with (H,K,O) in the horizontal scattering plane. The elastic scattering scans at T=8 K (see Fig. 5) show that the incommensurate wave vector has decreased from $\delta = 0.112(3)$ in the x=0.095 sample to 0.107(3). Surprisingly, the magnetic scattering is roughly a factor of eight less intense (Fig. 3a). In Figs. 3a) and b) the intensities have been scaled to the sample volume using the integrated intensity of an acoustic phonon measured near a strong nuclear Bragg peak at (2,0.15,0). We do not understand the reduced intensity for x=0.08, since extinction does not play a role.

The temperature dependence of the elastic magnetic signals at the incommensurate wavevectors (0.5,0.607,0) for La$_{2-x}$Ba$_x$CuO$_4$ (x=0.08) and at (0.612,0.5,0) for La$_{2-x}$Ba$_x$CuO$_4$ (x=0.095) are reproduced in Fig. 6, where the intensities have been scaled so that their functional form may be directly compared. As can be seen, both the temperature dependence of the order parameter and the phase transition temperatures are very similar despite the difference in the strength of the elastic magnetic Bragg scattering. We therefore conclude the two electronic energy scales for these crystals at x=0.08 and x=0.095, set by the superconducting $T_C$ and $T_N$, are surprisingly similar. It is not clear why a 4% decrease in $\delta$ should produce an eight-fold decrease in the spin Bragg intensity. A search revealed no additional magnetic intensity in diagonal directions. As in x=0.095, no incommensurate peaks due to charge ordering were observed in the x=0.08 sample.

FIG. 5: Static incommensurate magnetic peaks with $\delta = 0.107$ in La$_{1.92}$Ba$_{0.08}$CuO$_4$ at T=8 K, along (0.5,K,0).

FIG. 6: The temperature dependence of the elastic incommensurate magnetic scattering in La$_{2-x}$Ba$_x$CuO$_4$ (x=0.095) at (0.612,0.5,0) and (x=0.08) at (0.5,0.607,0). The net intensity for x=0.08 has been multiplied by a factor of 7.7 to match that of x=0.095.

V. DISCUSSION

Whether magnetism and superconductivity coexist in the same microscopic regions of the CuO$_2$ planes or are phase separated is a topical subject of research. The issue of microscopic spatial segregation has been examined using a combination of neutron scattering and $\mu$SR techniques in LaCuO$_{4+y}$. As a local probe $\mu$SR is sensitive to heterogeneous structures. The magnetic ordering in LaCuO$_{4+y}$ (y=0.11) and La$_{2-x}$Sr$_x$CuO$_4$ (x=0.12) has been reported to occur in reduced magnetic volume fractions of 40 and 18% respectively. Khaykovich et al argue an applied magnetic field enhances spin ordering primarily in the nonmagnetic regions, consistent with the above observations. By contrast, the magnetic volume fraction in La$_{2-x}$Ba$_x$CuO$_4$ (x=0.095) is much larger, approaching 100%. We speculate that no magnetic field dependence has been observed in La$_{2-x}$Ba$_x$CuO$_4$ x=0.095 because the non-magnetic volume fraction is too low. A systematic study of the variation of the spin ordering with magnetic field is therefore of interest, with emphasis on the correlations between this effect and the magnetic volume fraction.

An interesting difference between the x=0.08 and the 0.095 La$_{2-x}$Ba$_x$CuO$_4$ samples is that the spin ordered state in the x=0.095 sample grows within a fully developed LTT structure, as $T_N$=39.5 K and $T_{d2}$= 45 K. By contrast, in the x=0.08 La$_{2-x}$Ba$_x$CuO$_4$ sample, the MTO to LTT structural phase transition begins near $T_N$ on decreasing temperature and is only completed at temperatures below ~ 20 K. The situation for x=0.125 La$_{2-x}$Ba$_x$CuO$_4$ is similar to the x=0.095 case, as $T_N$=50 K, at which temperature the MTO-LTT transition for x=0.125 is largely complete. The first order nature of the MTO-LTT structural phase transition implies co-existing structures over the temperature regime at which the spin order forms for x=0.08 La$_{2-x}$Ba$_x$CuO$_4$. It is then possi-
results for the x=0.08 and x=0.095 samples give the same value for δ, in related La$_{2-x}$Sr$_x$CuO$_4$ systems. However, we also note that variability in the elastic magnetic Bragg intensity has been reported from La$_{2-x}$Sr$_x$CuO$_4$ samples to sample with similar nominal doping levels of x~0.1 and the La$_{2-x}$Sr$_x$CuO$_4$ system does not display the LTT phase at low temperatures.

We have observed the coexistence of static, two dimensional incommensurate spin order and superconductivity in La$_{2-x}$Ba$_x$CuO$_4$ with x=0.095 and x=0.08. This result is in broad agreement with other well studied high temperature La-214 cuprate superconductors, such as La$_{2-x}$Sr$_x$CuO$_4$ or La$_2$CuO$_{4+y}$, which show signatures of the abrupt nature of the x=1/8 anomaly in the LBCO system is clear.

The bottom panel of Fig. 7 shows the incommensurability δ vs Ba-content, x and once again we compare our new results on x=0.08 and x=0.095 La$_{2-x}$Ba$_x$CuO$_4$ with the x=0.125 La$_{2-x}$Ba$_x$CuO$_4$ results and those of several underdoped La$_{2-x}$Sr$_x$CuO$_4$ samples at Sr concentrations above x=0.05, where the incommensurate spin ordering is consistent with a picture of parallel (as opposed to diagonal) stripe ordering. In this Sr-content regime, δ tracks x well, assuming stoichiometric oxygen content. One can see that the incommensurability in x=0.08 and x=0.095 La$_{2-x}$Ba$_x$CuO$_4$ shows relatively little x-dependence. Indeed, we observe δ values which are only ~9% less than that displayed by x=0.125 La$_{2-x}$Ba$_x$CuO$_4$, thereby departing significantly from the approximately linear δ vs x relation characterizing the underdoped La$_{2-x}$Sr$_x$CuO$_4$ studies.

It is possible that this difference between underdoped La$_{2-x}$Ba$_x$CuO$_4$ and La$_{2-x}$Sr$_x$CuO$_4$ also arises due to the MTO-LTT structural phase transition, occurring in La$_{2-x}$Ba$_x$CuO$_4$ but absent in La$_{2-x}$Sr$_x$CuO$_4$. It is also conceivable that it arises due to some small oxygen off-stoichiometry, such that our samples have the composition La$_{2-x}$Ba$_x$CuO$_{4+y}$ with oxygen stoichiometry greater than 4. Such excess oxygen would give rise to an effective hole doping given by x$_{eff}$ = x+2y. To bring the δ values for x=0.08 and 0.095 back onto the linear relationship between δ and x$_{eff}$ seen in La$_{2-x}$Sr$_x$CuO$_4$, small, but positive values of y: 0.013 and 0.0075 for the x=0.08 and x=0.095 La$_{2-x}$Ba$_x$CuO$_{4+y}$ samples respectively would be required. This is too small to be detectable and runs counter to what is concluded in La$_{2-x}$Sr$_x$CuO$_{4+y}$. In underdoped La$_{2-x}$Sr$_x$CuO$_{4+y}$, the superconducting T$_c$ is maximized by annealing in oxygen, at which point the measured δ vs Sr concentration, x, lie on the straight line. Consequently, as grown La$_{2-x}$Sr$_x$CuO$_{4+y}$ tends to be oxygen deficient (y<0) and annealing in oxygen results in stoichiometric La$_{2-x}$Sr$_x$CuO$_4$. This is also expected to be true for underdoped La$_{2-x}$Ba$_x$CuO$_{4+y}$, which would imply that the deviation of δ vs x from a linear relationship is intrinsic to stoichiometric La$_{2-x}$Ba$_x$CuO$_4$, a surprising result.

**VI. SUMMARY AND CONCLUSIONS**

We have observed the coexistence of static, two dimensional incommensurate spin order and superconductivity in La$_{2-x}$Ba$_x$CuO$_4$ with x=0.095 and x=0.08. This result is in broad agreement with other well studied high temperature La-214 cuprate superconductors, such as La$_{2-x}$Sr$_x$CuO$_4$ or La$_2$CuO$_{4+y}$, which show signatures of...
either incommensurate static spin ordering or dynamic spin correlations. One significant finding of this study is the field independence of the incommensurate magnetic order in the x=0.095 sample, in marked contrast with other superconducting La-214 cuprates. Studies of the spin ordering as a function of magnetic field in other superconducting systems with large magnetic volume fractions should prove illuminating. In addition, while the dependence of the superconducting \( T_c \) on the incommensuration of the magnetic structure, \( \delta \), in La\(_{2-x}\)Ba\(_x\)CuO\(_4\) is the same as that observed in La\(_{2-x}\)Sr\(_x\)CuO\(_4\), the x-dependence appears to be substantially weaker than that seen in La\(_{2-x}\)Sr\(_x\)CuO\(_4\), where a linear relationship is observed over this range of concentration. Now that crystal growth breakthroughs have resulted in the availability of large, high quality single crystals of La\(_{2-x}\)Ba\(_x\)CuO\(_4\), fuller experimental characterization of the original family of high temperature superconductors is clearly warranted.

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