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Incommensurate geometry of the elastic magnetic peaks in superconducting $La_{1.88}Sr_{0.12}CuO_4$

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We report magnetic neutron-scattering measurements of incommensurate magnetic order in a superconducting single crystal of $La_{1.88}Sr_{0.12}CuO_4$. We find that the incommensurate wave vectors which describe the static magnetism do not lie along high-symmetry directions of the underlying CuO_2 lattice. The positions of the elastic magnetic peaks are consistent with those found in excess-oxygen doped La_2CuO_{4+y}. This behavior differs from the precise magnetic order found in the low-temperature tetragonal $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ material for which stripes of spin and charge have been observed. These observations have clear implications for any stripe model proposed to describe the static magnetism in orthorhombic La_2CuO_4-based superconductors.

In the lamellar copper-oxides, neutron-scattering experiments have shown that antiferromagnetic spin correlations are intimately intertwined with the super conductivity.[1–5] Current evidence indicates that static incommensurate magnetism coexists with superconductivity, especially for incommensurabilities near $\frac{1}{8}$ reciprocal-lattice units. This was first observed by Tranquada and co-workers in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ samples[6,7] which are tetragonal at low temperatures. Subsequently, Suzuki *et al.*[8] and Kimura *et al.*[9] have observed spin-density wave (SDW) order in $La_{1.88}Sr_{0.12}CuO_4$, and Lee *et al.*[10] have found a similar SDW order in stage-four La_2CuO_{4+y}. Note that these crystals remain orthorhombic at the lowest measured temperatures. Interestingly, for these latter two systems, the ordering temperature for the magnetism coincides with the superconducting transition temperature.

In $La_{1.88}Sr_{0.12}CuO_4$, a few important features require further clarification. The detailed geometry of the SDW modulation needs to be established. For example, one must check to see if the SDW peaks have modulation wave vectors which are like those in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ or, rather, if they more closely resemble those in La_2CuO_{4+y}. In $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$, a quartet of SDW peaks are observed in a square-shaped arrangement around the antiferromagnetic Bragg position.[6,7] In this case, the incommensurate wave vectors are aligned with the Cu-O-Cu direction. Hence, within a stripe model, the stripes of spin and charge are perfectly collinear with the underlying Cu-O-Cu directions. In contrast, high-resolution neutron-scattering measurements on stage-four La_2CuO_{4+y} (Ref. 10) indicate that the SDW wave vectors are rotated from perfect alignment by $\theta_Y \sim 3.3°$. This observation of rectangular-shifted peak positions indicates that the magnetic ordering need not lie along high-symmetry directions of the underlying CuO_2 plane. The shift of SDW peaks, or the *Y shift*, implies that within a stripe scenario the stripes are slanted. This presents a challenge to existing theoretical descriptions of the magnetism in high-T_c cuprates. It is thus crucial to reexamine the geometry of the elastic magnetic peaks in the $La_{1.88}Sr_{0.12}CuO_4$ material.

In this paper, we report high-resolution neutron-diffraction studies of the incommensurate elastic peaks in a single crystal of $La_{1.88}Sr_{0.12}CuO_4$. We have studied the same crystal ($6\phi \times 35$ mm) previously examined in Ref. 9. The magnetic susceptibility shows bulk superconductivity with $T_c = 31.5(26.5)$ K for the onset (midpoint) temperature of the transition. The lattice constants at room temperature were determined by powder x-ray diffraction on a crushed piece of the single crystal. In the high-temperature tetragonal (HTT) phase, the lattice constants are $a = 3.774$ Å and $c = 13.229$ Å at $T = 298$ K. Upon cooling, the material undergoes a structural transition to the low-temperature orthorhombic (LTO) phase. The structural transition temperature T_{s1} was determined by neutron-diffraction measurements of the structural superlattice reflection. In this crystal, we find $T_{s1} = 240$ K which implies a Sr concentration of $x = 0.12 \pm 0.004$. Further characterization of this crystal is given elsewhere.[9]

Neutron-scattering experiments were performed using the TOPAN triple-axis spectrometer in JRR-3M at the Japanese Atomic Energy Research Institute. The incident neutron energy was fixed at 14.7 meV with pyrolytic graphite (PG) filters inserted before and after the sample for our elastic-scattering measurements. The (002) reflection of a PG crystal was used to monochromatize and analyze the neutrons. We chose very tight horizontal collimations of

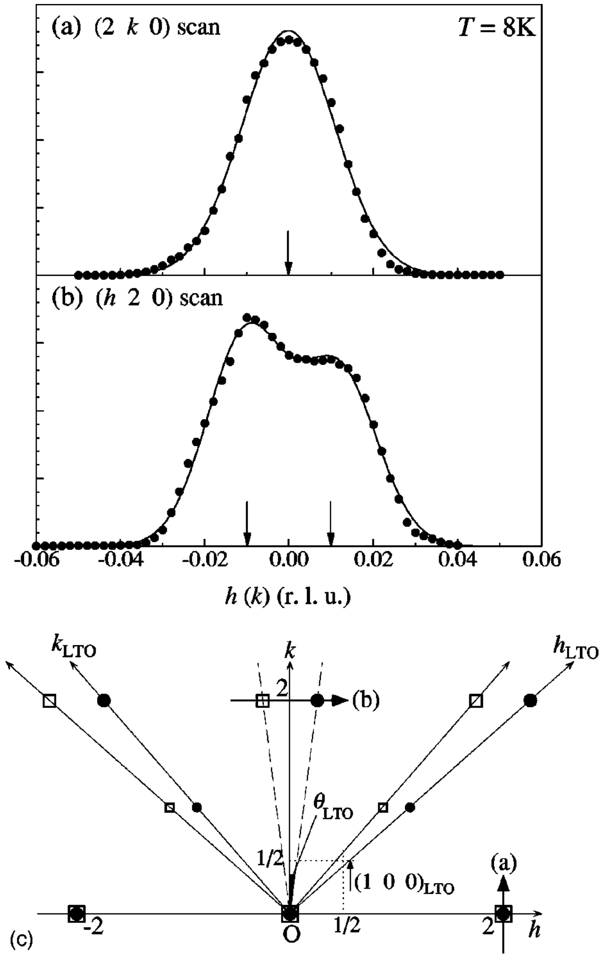

FIG. 1. Transverse scans through (a) the (200) and (b) the (020) nuclear Bragg peaks at $T=8$ K. (c) A schematic diagram of the $(HK0)$ reciprocal lattice depicting the two dominant twin domains. Trajectories for the scans in (a) and (b) are represented by the arrows.

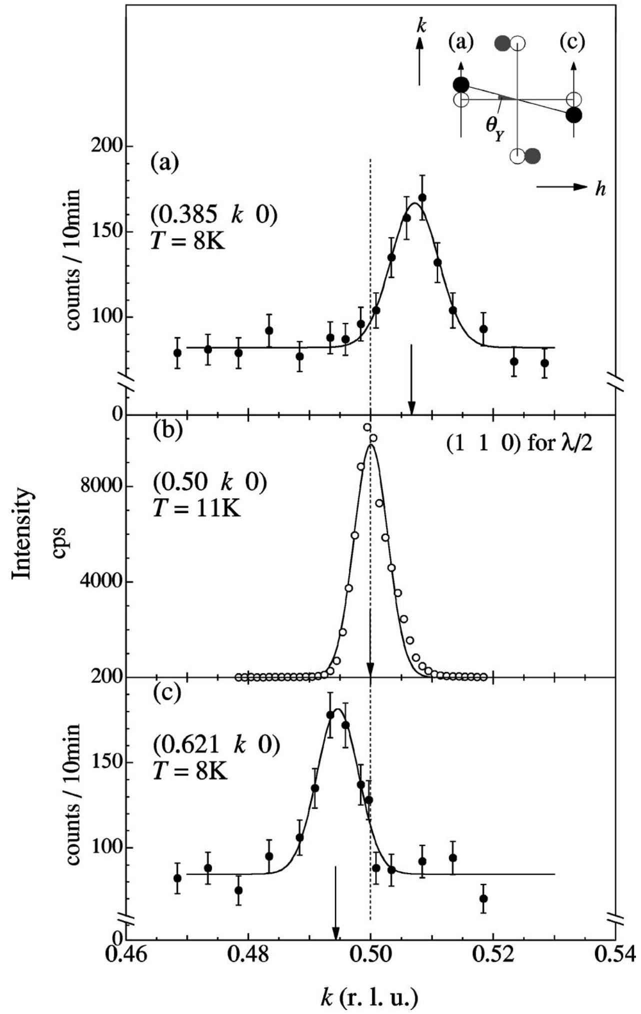

FIG. 2. Scans along K through the elastic magnetic peaks around $(\frac{1}{2}\pm\varepsilon\,\frac{1}{2}0)$ at 8 K are shown in (a) and (c). In (b), the (110) nuclear Bragg peak is shown using λ/2 neutrons from the incident beam. Scan trajectories for (a) and (c) are shown in the inset of (a).

15′-10′-Sample-10′-Blank for the dual purposes of achieving high q-space resolution and improving the signal-to-background ratio. In this paper, we use reciprocal-lattice units for the tetragonal $I4/mmm$ structure, even though the crystal is orthorhombic at low temperatures. This convention is used in order to be consistent with our previous neutron-scattering results[9] on this crystal. The crystal was mounted in the $(HK0)$ scattering zone and cooled using a closed-cycle ^{4}He cryostat.

We first separate the effects of structural twinning from the magnetic scattering. The LTO phase is characterized by a twin structure consisting of four possible domains, which give rise to four sets of nuclear Bragg peaks in the $(H,K,0)$ zone. The relative populations of these domains are sample dependent. Thus, we examined the peak profiles of the nuclear Bragg peaks in the LTO phase of this crystal in detail. As shown in Figs. 1(a) and (b), a transverse scan through the (200) Bragg peak position shows a single peak, whereas a transverse scan through the (020) position shows two peaks. This indicates that our crystal is primarily populated by two structural twin domains. These domains give rise to two sets of peaks in reciprocal space which are simultaneously observed, as depicted in Fig. 1(c).

Now that the structural twinning has been well characterized, we turn our focus to the elastic magnetic scattering. In previous work (Ref. 9), the authors assumed that the incommensurate elastic magnetic peaks appear at $(\frac{1}{2}\pm\varepsilon\,\frac{1}{2}0)$ and $(\frac{1}{2}\,\frac{1}{2}\pm\varepsilon 0)$, where $\varepsilon=0.118$. Using high instrumental resolution, we have reexamined the positions of the incommensurate peaks. We find that two of the peak positions are given by $(\frac{1}{2}-\varepsilon\,\frac{1}{2}+\eta 0)$ and $(\frac{1}{2}+\varepsilon'\,\frac{1}{2}-\eta' 0)$, where $\varepsilon=0.115$, $\varepsilon'=0.121$, $\eta=0.007$ and $\eta'=0.006$ with an error bar of ±0.002. The nonzero values of η and η' indicate that the peaks are shifted off of the Cu-O-Cu axes. This is indicated by the filled circles in Fig. 2(a) which form a rectangular quartet of incommensurate peaks (as opposed to a square arrangement). Alternatively, this peak-shift can be characterized by a tilt angle $\theta_Y\simeq3.0°$ between the incommensurate wavevector and the Cu-O-Cu direction, also depicted in the inset of Fig. 2(a).

We can show that the shifted positions of the magnetic peaks are *not* an artifact due to crystal misalignment or the presence of multiple structural twin domains. Figure 2(b) shows a scan along k through the (110) nuclear Bragg re-

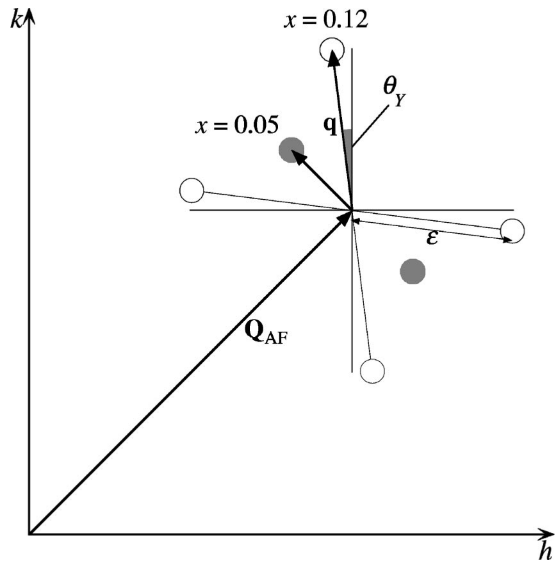

FIG. 3. Schematic drawing of the elastic magnetic peak positions in $La_{2-x}Sr_xCuO_4$ for $x=0.05$ and 0.12.

flection using higher order $\lambda/2$ neutrons from the incident beam. This peak is observed exactly at $(\frac{1}{2}\frac{1}{2}0)$, as expected. This rules out the possibility that the peak shift of the elastic magnetic peak is due to misalignment of the crystal. Measurements of various nuclear Bragg peaks, summarized in Fig. 1(c), indicate that peaks from different twin domains are at most offset from each other by $\sim 0.3°$ (θ_{LTO}). Since $\theta_Y \sim 3.0°$ is ten times larger, the orthorhombicity and/or concomitant twin structure cannot account for the magnitude of the shift in the magnetic peak positions. Furthermore, a similar magnitude for the magnetic peak shift has been observed in La_2CuO_{4+y},[10] even though the orthorhombicity is about twice as great as that in $La_{1.88}Sr_{0.12}CuO_4$. Therefore, we conclude that the shift of the magnetic peaks, i.e., the *Y* shift, is an intrinsic property of the SDW order. While striking, the reduced symmetry of the rectangular quartet of SDW peaks is not completely unexpected since the orthorhombicity has already broken the fourfold symmetry of the underlying CuO_2 lattice.

The importance of the orthorhombicity becomes obvious by noting that the SDW peak positions are most simply described using orthorhombic notation. Recall that using tetragonal notation, we found that the incommensurabilities for the magnetic peaks, ε and ε', were significantly different. Using orthorhombic *Bmab* notation, we find that the quartet of incommensurate peaks are precisely centered at $(100)_{ortho}$, where the low-temperature lattice constants are $a=5.3184$ Å, $b=5.3450$ Å, and $c=13.175$ Å. As shown in Fig. 3, only two parameters are required to describe the SDW peak positions centered at $(100)_{ortho}$: a single wavevector magnitude $\varepsilon \simeq 0.118$ and a tilt angle $\theta_Y \simeq 3.0°$. Note that in orthorhombic notation, the four SDW peaks are located at $(1\pm q_x \ \pm q_y \ 0) \equiv \mathbf{Q}_{AF}+\mathbf{q}$ in Fig. 3. This description for the SDW is equivalent to that found by Lee *et al.*[10] in stage-four La_2CuO_{4+y} for which $\theta_Y \simeq 3.3°$, a tilt angle almost identical to the present result for $La_{1.88}Sr_{0.12}CuO_4$ within the errors. Our present results for $La_{1.88}Sr_{0.12}CuO_4$ have clear implications within the context of stripe models. In the limit that the kinks all occur in the same direction, a tilt angle of $\theta_Y \simeq 3.0°$ corresponds to one kink every ~ 19 Cu sites on the charge domain walls. The observed wave vector magnitude $|\mathbf{q}| \simeq 0.118$ is smaller than the commensurate value of $\frac{1}{8}$. This indicates that the average separation between the charge walls consists of a mixture of three-spin and four-spin regions. The population ratio of the four-spin regions to the three-spin regions is approximately 1:18. The close match between this ratio and the kink density is consistent with a simple picture in which each kink on the charge walls is accompanied by a four-spin segment of intervening spins; whereas, away from the kinks, the usual three-spin spacing dominates. However, since direct diffraction from any charge order is below our detection limit, we cannot rule out alternative models for the SDW. For example, a grid pattern of orthogonal stripes oriented along the two orthorhombic directions adequately describes our data as well. Further similarities to the La_2CuO_{4+y} material are found. In La_2CuO_{4+y}, the incommensurate elastic peaks are centered at $(10l)_{ortho}$ (for even l) and $(01l)_{ortho}$ (for odd l), implying the same local spin stacking arrangement as that in the undoped La_2CuO_4 insulator. Also, the ordered spin direction of the SDW in La_2CuO_{4+y} is deduced to be along the orthorhombic *b* axis, again, identical to that in La_2CuO_4.[11] We have preliminary measurements of the *L*-dependence of the elastic peaks in $La_{1.88}Sr_{0.12}CuO_4$. We observed broad and weakly *q*-dependent peaks (not shown), implying short-ranged magnetic correlations along the *c* axis. The result is qualitatively consistent with that for La_2CuO_{4+y},[10] implying a similar stacking arrangement and spin direction for the ordered spins in $La_{1.88}Sr_{0.12}CuO_4$. Since the orthorhombicity of $La_{1.88}Sr_{0.12}CuO_4$ is significantly smaller than that of La_2CuO_{4+y}, it is difficult to resolve the various contributions of the superposed structural twins to the *L*-dependent scattering. Therefore, further focused studies are required to give a more precise determination of the spin correlations along the *c* axis in this material. In La_2CuO_{4+y}, the ordered moment had been estimated to be $0.15\pm0.05\mu_B$. Comparing data taken on the same spectrometer under identical conditions, the ordered moment of the SDW in $La_{1.88}Sr_{0.12}CuO_4$ is approximately the same as that in La_2CuO_{4+y}, within the errors.

In summary, we have established that the SDW peaks in superconducting $La_{0.88}Sr_{0.12}CuO_4$ have an identical geometry, within error, to that in La_2CuO_{4+y}, namely the *Y* shift. That is, the SDW peaks are centered around the $(100)_{ortho}$ position with a peak-shifted rectangular arrangement. This is in contrast to the SDW peaks in $La_{1.6-x}Nd_{0.4}Sr_xCuO_4$ which retain square symmetry and do not show this peak shift. In general, the existence of short or long-range ordered SDW peaks appears to be a common feature of the La_2CuO_4-type superconductors for incommensurabilities near $\frac{1}{8}$. However, the *orthorhombic* superconductors, $La_{1.88}Sr_{0.12}CuO_4$ and stage-four La_2CuO_{4+y}, exhibit unique characteristics for the SDW order. At base temperature, the static spins are correlated over very long ranges: greater than 200 Å in $La_{1.88}Sr_{0.12}CuO_4$ (Ref. 9) and greater than 400 Å in stage-four La_2CuO_{4+y}.[10] We now have established that the SDW modulations have a reduced-symmetry peak shift in both systems. A clear implication is that, in a stripe model, the stripes are slanted. Any possible correspondence between slanted stripes and high superconducting T_c's requires further work.

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