Low-energy incommensurate spin excitations in superconducting $La_{1.85}Sr_{0.15}CuO_4$

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Neutron-scattering experiments with markedly improved signal-to-noise ratios have been performed on single-crystal superconducting $La_{1.85}Sr_{0.15}CuO_4$ ($T_c=33$ K) at energies down to 1.5 meV. The measurements provide quantitative information on the temperature dependence of $\chi''(\mathbf{q},\omega)$. Surprisingly, $\chi''(\mathbf{q}_{\mathrm{peak}},\omega)$ is approximately constant below T_c for the measured energies 1.5 meV $\leq\omega\leq$ 6 meV, that is, for energies much less than the mean-field BCS gap energy of ~ 10 meV, hence implying other than conventional *s*-wave superconductivity.

The superconducting copper oxides $La_{2-x}Sr_xCuO_4$ are well known to exhibit unusual short-range magnetic correlations. Early neutron-scattering experiments established that incommensurate spin correlations[1] persist for superconducting hole concentrations.[2] Further, the structure factor $S(\mathbf{q},\omega)$ in $T_c=33$ K superconducting $La_{1.85}Sr_{0.15}CuO_4$ was found to exhibit a suppression below ~ 100 K for $\mathbf{q}$ near the (π,π) position in reciprocal space (square lattice notation, unit lattice constant) at $\omega=6$ meV, while at $\omega=12$ meV, no such anomaly occurred.[2] However, a complete characterization of the incommensurate scattering geometry and a thorough study of the low-energy excitations were not performed in this early work. Recently, Cheong *et al.*[3] have shown that the positions of the incommensurate peaks are at $[\pi(1\pm\delta),\pi(1\pm\delta)]$. With this information, we have performed experiments that probe the low-energy magnetic excitations.

The high quality of our data with signal to noise ratios of up to ~ 5, which represents a marked improvement on other measurements,[2,4-6] has allowed a detailed characterization of the magnetic excitations for energies less than 6 meV. We find that the superconducting transition influences $S(\mathbf{q},\omega)$ significantly at relatively low energies, $\omega\lesssim 3$ meV. Further, there are substantial spin fluctuations in the superconducting state at energies as low as 1.5 meV. Indeed, on converting the measured $S(\mathbf{q},\omega)$ to $\chi''(q,\omega)$ we find that $\chi''(q_{\mathrm{peak}},\omega)$ is approximately constant below T_c for energies 1.5 meV $\leq\omega\leq$ 6 meV. Our experiments also show, in agreement with the recent work of Mason *et al.*,[4] that the width in momentum space of the incommensurate peaks decreases markedly with decreasing energy.

The two single crystals used in these experiments were grown using the traveling floating-zone method.[7] One of the crystals, sample 1 (labeled KOS-1 in Ref. 2), was examined in previous neutron experiments.[2] Extensive measurements characterizing this sample and, in particular, showing its high degree of microscopic homogeneity are reported in Ref. 2. For the current experiments, a second sample, sample 2, of similar quality to sample 1, was mounted and aligned beside sample 1 in order to increase the scattering intensity. The total sample volume was then ~ 1.0 cm^3, and the combined mosaicity was very good, less than $\sim 0.15°$ half width at half maximum (HWHM). Sample 1 and sample 2 have identical transport and magnetic properties. We emphasize here that both crystals undergo a bulk superconducting transition at 33 K with Meissner fractions approaching 100%. The weak-coupling BCS gap in this material is $2\Delta=3.5k_BT_c=10$ meV so that measurements of the spin fluctuations for $\omega \ll 10$ meV are particularly important.

Neutron-scattering experiments were carried out on the triple-axis spectrometer H-7 at the Brookhaven High Flux Beam Reactor. For most of the experiments the incident neutron energy was fixed at 14.7 meV and the horizontal collimator sequence was 40′-80′-*S*-80′-80′. This gives an energy resolution of ~ 0.5-meV HWHM. Even though the sample is slightly orthorhombic below ~ 200 K, in this paper we shall label reciprocal space using tetragonal units (q_x,q_y,q_z). Here q_x and q_y refer to the directions of the square lattice formed by the CuO_2 sheets, and q_z refers to the layer stacking direction. The magnitude of q_x, q_y, and q_z will be given in units where the lattice constants are set at 1.

The sample is mounted in two configurations. In the first, scans with $q_x=q_y$ and q_z arbitrary can be performed. This allows access to the $(\pi\pi)$ position. In the second, the sample is tilted about the (0,0,1) axis by an angle ϕ, so that scans with $q_x=q_t\cos(45+\phi)$,

$q_y = q_t \sin(45+\phi)$, and q_z arbitrary are possible (we parametrize tilt scans by the variable q_t). When ϕ is set at $\sim 6°$, the scattering near the incommensurate peak positions $[\pi(1-\delta),\pi]$ and $[\pi,\pi(1+\delta)]$ with $\delta \sim 0.22$ can then be probed. The latter configuration is used primarily for the data reported here. The dashed line in the inset of Fig. 1(b) shows a tilt equal to 0° scan through the (π,π) position while the solid line illustrates a tilt scan which passes near the two large black circles which represent incommensurate scattering peaks. The primary advantage of using the tilt geometry over the $(q_x,q_y,0)$ zone geometry[3,4] is that the background is reduced considerably. This is because one can easily change q_z in order to minimize background scattering from phonon and multiple-scattering-type processes, and because the tilt position zone has a low symmetry so that a variety of spurious scattering processes are eliminated.

The ellipse in the inset of Fig. 1(b) represents the resolution function of the spectrometer. We caution that the peak intensity data presented in this paper actually represent an integration over the instrumental resolution function, which is a rather sizable region of the momentum space. One can see this from the figure, where the length of the long axis of the resolution ellipse is about one-third the magnitude of the incommensurability $\delta\pi/a$. Quantitatively, the HWHM of the long axis is ~ 0.06 Å^{-1}, while $\delta\pi/a \sim 0.18$ Å^{-1}. That the peak intensity data are actually integrated over one dimension does not affect any of the conclusions presented in this paper.

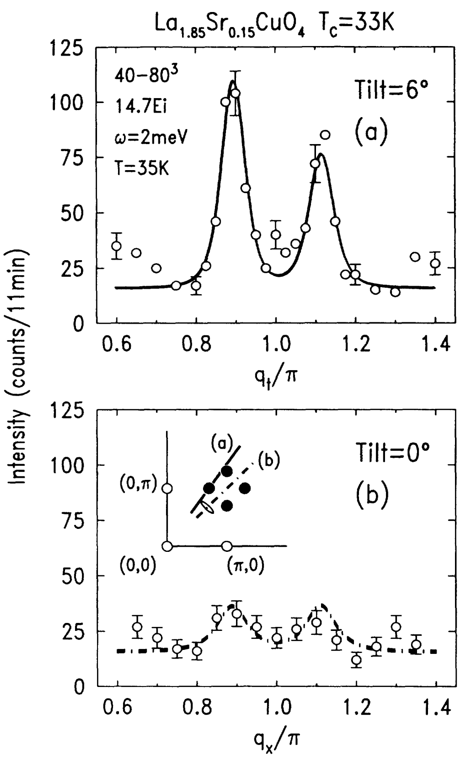

FIG. 1. (a) and (b) Inelastic-neutron-scattering spectra at $\omega=2$ meV and $T=35$ K. Panel (a) shows a tilt $=6°$ scan which follows line (a) in the inset; the trajectory is $(q_t \cos 51°, q_t \sin 51°, -0.6\pi)$. Panel (b) shows a tilt $=0°$ scan which follows line (b) in the inset; the trajectory is $(q_x, q_x, -0.6\pi)$. The solid line in panel (a) is the result of a fit to four squared Lorentzians centered at positions $[\pi(1\pm\delta),\pi(1+\delta)]$ with $\delta=0.22$ convolved with the instrumental resolution function while the dashed line in panel (b) is calculated from this same fit.

Shown in Figs. 1(a) and 1(b) are representative inelastic scans at $\omega=2$ meV and $T=35$ K. The scan in 1(a) has the tilt set at 6°, while in 1(b) the tilt is set at 0°. Most importantly, the signal-to-noise ratio of these data is up to 5:1, a vast improvement over previous work[2–4] where all signal-to-noise levels were less than 1:1. The peak on the right-hand side of Fig. 1(a) is smaller than the one on the left-hand side because the tilt scan trajectory does not pass through the center of both peaks. At this energy, the scan through the (π,π) position shown in Fig. 1(b) has barely any detectable scattering. This is because the spectrometer resolution function, represented by the small oval along scan (b) in the inset of Fig. 1(b), does not overlap appreciably with regions of momentum space where the scattering is strong. The solid line through the tilt scan data in Fig. 1(a) is the result of a fit to a two-dimensional (2D) squared Lorentzian profile with a HWHM of 0.025 Å^{-1}. The squared Lorentzian profile in $\mathbf{q}$ approximates the mean field form for $S(\mathbf{q},\omega)$ at fixed ω. The dashed line in Fig. 1(b) is calculated using the results of this tilt fit with no further adjustable parameters. Thus the very weak scattering observed in the (π,π) scan is entirely accounted for by the tails of the scattering from the incommensurate peaks.

We show in Fig. 2 a series of scans at $T=35$ K at energies of 1.5, 2, 3, and 4 meV together with scans at 10 K at 1.5 and 3 meV. The solid lines for energies 1.5, 2 and 3 meV at $T=10$ and 35 K are all calculated using 2D squared Lorentzian profiles with HWHM of 0.025 Å^{-1} convolved with the instrumental resolution function while the scan at 4 meV has a HWHM of 0.04 Å^{-1}. Similar scans at 6 meV (not explicitly shown) have HWHM of 0.06 ± 0.01 Å^{-1} below 50 K. These data contain a number of features which are both important and, in our view, somewhat surprising. First, as already suggested by the recent measurements reported in Ref. 4, the excitations are remarkably sharp in momentum space at low temperatures and energies. Second, this excitation momentum width increases with increasing energy. It is apparent that because of the weak dependence of the peak intensity on energy,[2,4] together with the increasing width with increasing energy, an integration over energy will yield a static structure factor with HWHM much larger than 0.025 Å^{-1}. Thus, the corresponding real-space static correlation lengths are rather in agreement with our earlier work[1] but in disagreement with the conclusions of Ref. 4. Most importantly, from the scans at 10 K in Fig. 2 it is evident that these sharp, low-energy excitations persist in the superconducting state. We shall discuss this further below.

We show in Fig. 3 the intensity at the $[\pi(1-\delta),\pi]$ position as a function of temperature in the range 10 K $\leq T \leq 70$ K for energies 1.5 meV $\leq \omega \leq 6$ meV. We remind the reader that because of the anisotropic resolution function these data in fact represent one-dimensional integrations over the incommensurate peaks. Although there may be some subjectivity in the interpretation of the results shown in Fig. 3, the following seems clear. First, at 1.5 and 2 meV there is a well-defined peak in the intensity at a temperature which to within the errors coincides with $T_c = 33$ K. For $\omega = 3$ meV with decreasing temperature the intensity approaches a constant above T_c and then decreases by about a factor of 3 between T_c and 10 K. However, at 4 and 6 meV the effect of the superconducting transition on the amplitudes of the spin fluctuations is less apparent. In no case is the width in momentum space of the excitation appreciably affected by the superconductivity. We emphasize that the signal-to-noise ratio in these measurements is an order of magnitude better than that in all previous neutron-scattering studies of superconducting copper oxides,[1-6] and accordingly we regard these data as being correspondingly more reliable. Specifically, they provide quantitative information for comparison with predictions of various models for the spin fluctuations in the superconducting state.

From the data of Fig. 3 one may immediately extract $\chi''(\mathbf{q},\omega)$ via the fluctuation dissipation theorem, that is, $\chi''(\mathbf{q},\omega) = (1 - e^{-\omega/T}) S(\mathbf{q},\omega)$. The results so obtained are shown in Fig. 4. The data above T_c are too limited to draw strong conclusions. Generally, however, they are qualitatively consistent with the ω/T scaling model developed by Keimer *et al.*[8] in their study of the spin fluctuations in nonsuperconducting and commensurate $La_{1.96}Sr_{0.04}CuO_4$, that is, χ'' increases with decreasing temperature at a rate which increases with decreasing energy. To within the errors, this growth is arrested at T_c and $\chi''(\mathbf{q},\omega)$ with $\mathbf{q} = [\pi(1-\delta), \pi]$ is approximately constant below T_c. This holds for energies from 1.5 to 6 meV, except possibly at $\omega = 3$ meV (~ 35 K in temperature units) where $\chi''(\mathbf{q},\omega)$ appears to decrease somewhat below T_c. Because the width in $\mathbf{q}$ does not change significantly with temperature near and below T_c, this behavior holds for both the peak and integrated values.

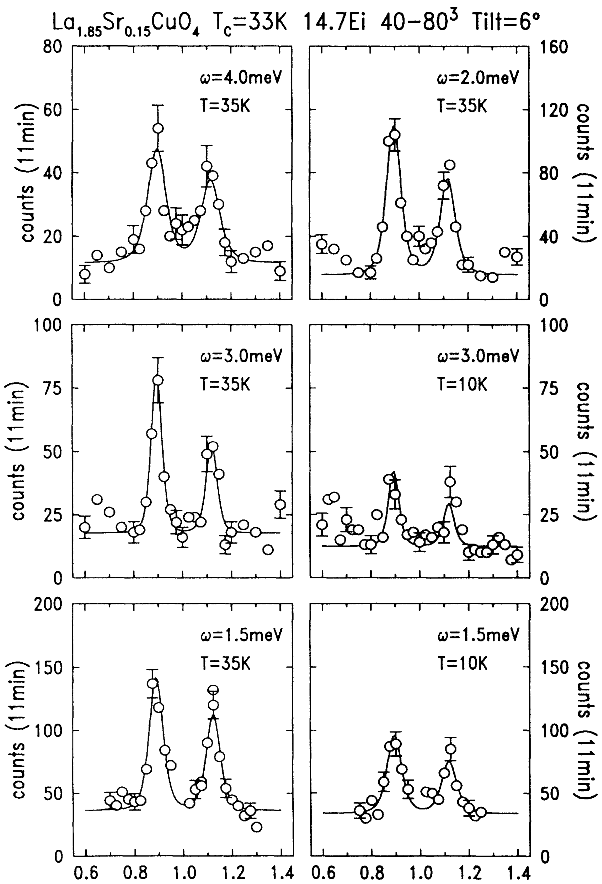

FIG. 2. Inelastic-neutron-scattering spectra at $T = 35$ and 10 K for various energies. All of the scans were taken in a tilt $= 6°$ configuration which has the trajectory $(q_t \cos 51°, q_t \sin 51°, q_z)$. Here $q_z = -0.6\pi$ at 4 meV, and $q_z = -0.8\pi$ at 2 meV. The spectrometer had fixed incident (14.7 Ei) neutron energy. The lines are the results of fits to squared Lorentzians centered at positions $[\pi(1\pm\delta), \pi(1\pm\delta)]$ convolved with the instrumental resolution function as discussed in the text.

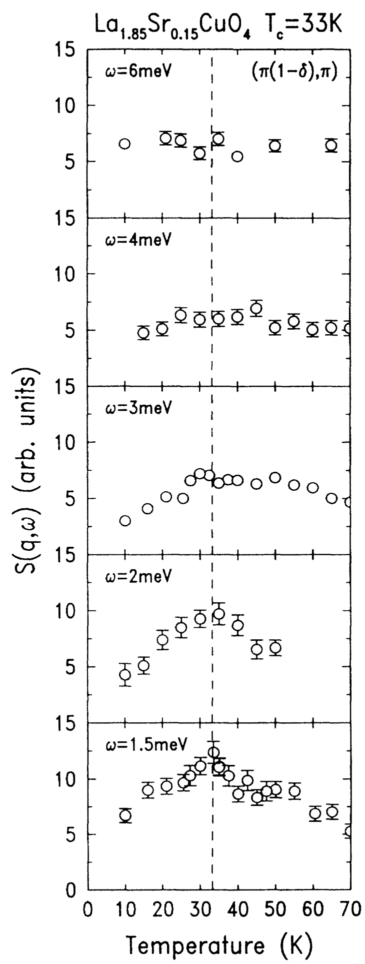

FIG. 3. (a) Temperature dependence of the peak intensity at the $[\pi(1-\delta), \pi]$ position for energies of 1.5, 2, 3, 4 and 6 meV. Higher-order and resolution corrections have been applied, and smooth, weakly temperature-dependent backgrounds subtracted.

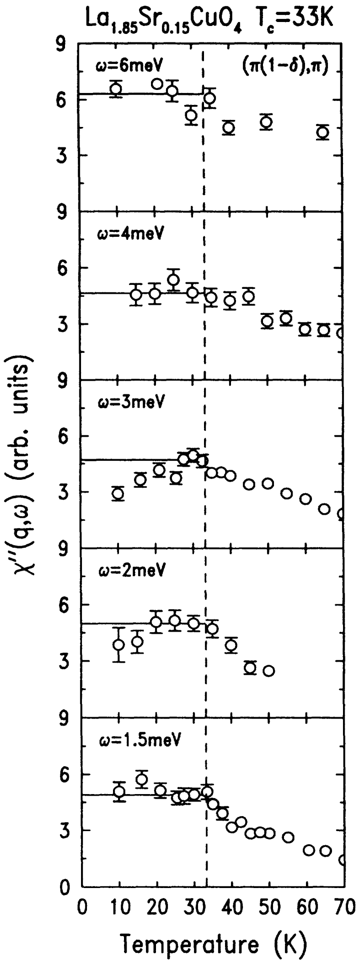

FIG. 4. Temperature dependence of $\chi''(\mathbf{q},\omega)$ at 1.5, 2, 3, 4, and 6 meV for $\mathbf{q}=[\pi(1-\delta),\pi]$. Corrections for higher-order contamination and instrumental resolution have been made to these data. The solid lines below T_c are simply constants.

These results differ significantly from the expectations based on the analysis in Ref. 4. Specifically, we find that at low T, $\chi''(\mathbf{q},\omega)$ depends only weakly on ω below 6 meV whereas Mason *et al.*[4] predict a marked diminution with decreasing energy. We should note, however, that our results at 3 meV are similar to the 4-meV data in Ref. 4, the latter being the lowest energy for which detailed results are presented in that paper.

We shall now discuss some of the implications of this work. First, there is no simple correspondence between our data and NMR measurements of $(T_1T)^{-1}$ at the copper sites;[9] this latter quantity, which is proportional to an integral of $\chi''(\mathbf{q},\omega)$ over $\mathbf{q}$ weighted by form factors, shows a maximum somewhat above T_c and then decreases gradually to zero with further decrease in temperature. Clearly, further theory is required to relate our measured $\chi''(\mathbf{q},\omega)$ at nonzero ω to that inferred for the $\omega \rightarrow 0$ limit from the NMR data.[9] Second, the behavior we have found for the temperature dependence of $\chi''(\mathbf{q},\omega)$ in $La_{1.85}Sr_{0.15}CuO_4$ is qualitatively similar to that observed by Sternlieb *et al.*[5] in $YBa_2Cu_3O_{6.6}$ ($T_c=53$ K) albeit with the energy scale renormalized by a factor of 2 and with some minor differences in the behavior below T_c. This suggests that the superconductivity has the same basic character in the two materials. Third, these data seem to exclude conventional *s*-wave BCS models for the superconductivity in these materials. Our results may be compatible with a *d*-wave description of the superconducting order parameter although establishment of this will require detailed predictions for $\chi''(\mathbf{q},\omega)$ in the superconducting state. Such models must, of course, account for the incommensurability in $La_{1.85}Sr_{0.15}CuO_4$ and the near commensurability in $YBa_2Cu_3O_{6.6}$.

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