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"New magnetic coherence effect in superconducting $La_{2-x}Sr_xCuO_4$"

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New magnetic coherence effect in superconducting $La_{2-x}Sr_xCuO_4$

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We have used inelastic neutron scattering to examine the magnetic fluctuations at intermediate frequencies in the simplest high temperature superconductor, $La_{2-x}Sr_xCuO_4$. The suppression of the low energy magnetic response in the superconducting state is accompanied by an increase in the response at higher energies. Just above a threshold energy of ~ 7 meV there is additional scattering present below T_c which is characterised by an extraordinarily long coherence length, in excess of 50Å.

Spin pairing is responsible for dramatic reductions in the magnetic response of metals as they enter the superconducting state. Over the last few decades nuclear resonance [1] and neutron scattering measurements [2,3] have provided convincing evidence for this suppression, thereby validating one of the key features of the BCS wavefunction: the electron spins are hidden because they belong to singlet pairs [4]. Of course, once the pair binding energy is exceeded, the bare electron spins should become visible again. The manner in which this occurs depends on the coherence of excitations from the superconducting ground state, as well as the symmetry of the superconducting order parameter. We have used inelastic neutron scattering to examine the magnetic fluctuations at intermediate frequencies in the simplest high temperature superconductor, $La_{2-x}Sr_xCuO_4$. A key conclusion is that the suppression of the low energy magnetic response in the superconducting state is accompanied by an increase in the response at higher energies. What is more extraordinary is that at energies just above where superconductivity begins to enhance magnetic scattering, the enhancement is very sharp in momentum space. Indeed, the enhancement is resolution limited, corresponding to a coherence length in excess of all other magnetic, superconducting, and electronic length scales, with the exception of the superconducting phase coherence length, for $La_{1.86}Sr_{0.14}CuO_4$.

We performed our experiment using the TAS VI cold neutron spectrometer at Risø National Laboratory in Denmark and the $La_{1.86}Sr_{0.14}CuO_4$ crystals with $T_c = 35$ K employed in previous experiments [3,5]. The scattered neutron intensity is proportional to the two spin correlation function which in turn is proportional to the imaginary part of the wavevector (Q)- and energy ($\hbar\omega$)- dependent magnetic response function multiplied by a thermal population factor, $(n(\omega)+1)\chi''(Q,\omega)$ where $(n(\omega)+1) = (1-e^{-\hbar\omega/k_BT})^{-1}$. Throughout this paper, we use the square lattice notation employed by theorists to locate wavevectors in the reciprocal space for the CuO_2 planes (nearest neighbor Cu-Cu separation=a_o=3.8 $\AA^{-1}$) of $La_{2-x}Sr_xCuO_4$. For $La_{1.86}Sr_{0.14}CuO_4$, $\chi''(Q,\omega)$ peaks at four incommensurate positions, denoted Q_δ in Fig. 1 a), near the magnetic ordering vector, (π,π), of the insulating and antiferromagnetic parent, La_2CuO_4 [6]. These correspond to the four locations equivalent to $(\pi,\pi)+\delta(\pi,0)$ with $\delta = 0.245$. Fig. 2 shows scans along the solid line in Fig. 1 a) through two of the peaks for two energy transfers ($\hbar\omega$). It also shows scans along the dashed line in Fig. 1 a), which are representative of the temperature-dependent and weakly Q-dependent background. For $\hbar\omega = 6.1$ meV, the peak intensities clearly decrease on passing from T = 35 K = T_c to 5 K, while for $\hbar\omega = 9$ meV, they actually increase. The former effect is due to the usual depletion of the electron-hole pair continuum and has been amply documented over the last four years for various samples of $La_{2-x}Sr_xCuO_4$ [3,5,7]. The latter effect is new, and being confined to a region very close to the normal state peak positions, actually corresponds to a dramatic sharpening of the incommensurate fluctuations in the superconducting state. In general, one would expect the low frequency suppression of intensity to be accompanied by an enhancement at higher frequencies due to conservation of spin. Obviously, an increase in signal between 35 K and 5 K could be associated with continuation of an evolution in the normal state intensity as well as with the onset of superconductivity. To check which hypothesis is correct, we have measured the peak intensities as a function of temperature for several $\hbar\omega$. Fig. 3 shows the results. For the lowest frequency, 6.1 meV, we see the well-known reversal of the growth in the normal state signal at T_c. In contrast, for both $\hbar\omega = 9$ meV and 15 meV, the intensities undergo negligible evolution in the normal state, while there is a clear increase which begins on cooling below T_c. We conclude that the rise in incommensurate peak intensity between T_c and 5K is due to superconductivity. For these higher $\hbar\omega$ the temperature dependence of the thermal factor is quite small and is in the wrong direction to produce an increase in the intensity at low temperatures.

Fig. 4 gives an overview of how the magnetic signal changes on going into the superconducting state in the form of a plot, as a function of both Q and $\hbar\omega$, of the differences between the 5 K and 35 K data. The important features are: (i) that the difference crosses zero at $\hbar\omega \simeq 7$meV, (ii) that it does not appear to return to zero in the range of frequencies (to 15 meV) accessed above 7.5 meV, (iii) that at 9 meV, the difference is a sharp resolution-limited peak centred at the incommensurate position (see also the inset in Fig. 2 a)), and (iv) that at $\hbar\omega = 15$ meV, the difference appears somewhat broader and shifted to seemingly smaller incommensurability. Indeed, fitting Gaussians to the difference spectra, we find that the peak at 15 meV has shifted by 0.04±0.01 $\AA^{-1}$ towards the centre of the scan along the solid line in Fig. 1a) while it is centred at the incommensurate wavevector that characterises the normal state at 9 meV. The peak in the difference spectrum is also broader at 15 meV, with a full width at half maximum of 0.10 ± 0.02 $\AA^{-1}$ compared to a resolution-limited 0.04 ± 0.01 $\AA^{-1}$ at 9 meV. This width indicates a coherence length certainly in excess of 50 Å and most probably larger than 100 Å (~ 26a_o) for the additional 9 meV excitations in the superconducting state. In addition, this is well beyond the 25 Å coherence length characterising the incommensurate magnetic fluctuations above T_c at 9 meV [3,8], as well as the estimated 20-30 Å superconducting pair radius for $La_{1.86}Sr_{0.14}CuO_4$ [9]. The inset to Fig. 2 a) shows the resolution limited Gaussian response (solid line) together with the cross section expected for the 25 Å length scale of the normal state using the lineshape which best describes the normal state response (dashed line) [3,5,8]. Clearly the resolution limited peak provides a better description of the data (indeed models with coherence lengths of 100 and 1000 Å provide equally good fits to the data).

It is possible to obtain a qualitative understanding of our observations by considering Fig. 1 b) and c). In b) we show the Fermi surface most likely responsible for the incommensurate peaks in the magnetic response of $La_{2-x}Sr_xCuO_4$ [10]. Inelastic scattering occurs when neutrons create electron-hole pairs: the arrows correspond to the best nesting vector (i.e. the translation which brings the largest portion of the Fermi surface into good alignment with another parallel portion) and concomitantly the neutron wavevector transfer whereby electrons are removed most efficiently from immediately below the Fermi surface to immediately above in the normal state. Superconductivity eliminates these low frequency electron-hole pair excitations, as illustrated in c). However, c) also reveals that excitations with momenta precisely equal to the nesting vector, $\mathbf{Q}_\delta$, are allowed for an energy transfer equal to the pair binding energy, 2Δ. Indeed, at this energy, the only excitations allowed have precisely the wavevector $2\mathbf{k}_f$, implying that the electron-hole pair excitation peak which one would find for fixed neutron energy transfer, as in Fig. 2, should be narrower in the superconducting than the normal state, where the momentum phase space for possible energy conserving transitions is larger. Of course, as $\hbar\omega$ increases, even for the superconducting state, more phase space becomes available, and there should be a broadening in the enhanced electron-hole pair response function, which is precisely what we see in our experiment as well.

It is remarkable that phase space arguments of the type contained in Fig. 1 c) account for our data. Indeed, our samples are not optimally doped, display considerable pair-breaking in the superconducting state, probably have a Fermi surface with sufficient curvature to broaden the peak beyond what Fig. 1 c) suggests, and are known to host phonons as well as electron-electron interactions which would broaden all sharp momentum space features at finite frequencies or temperatures. Because each phenomenon just enumerated contributes to the width of the intensity enhancement seen at 9 meV below T_c, we must conclude that each phenomenon is separately characterised by a length scale in excess of 50 Å. Thus, the effect we have discovered cannot have anything to do with the disorder which leads to other potentially suboptimal properties (e.g. a T_c of 35 K instead of 38 K) of our sample. Furthermore, it appears that the peak arises from a unique spanning vector across the Fermi surface, implying that its curvature together with the Q-dependent gap function are so arranged that the one-dimensional phase space portrait, shown in Fig. 1 c), is somehow adequate. Finally, the (electron-electron and electron-phonon) interaction - derived mean free path, whose inverse contributes directly to the broadening in momentum space, is indistinguishable from infinity in the superconducting state. This result, obtained directly from scans in momentum space, has been long conjectured in order to explain anomalies in various other spectroscopies of the superconducting state [11]. Because phonons do not disappear below T_c, the scattering of high-frequency quasiparticles from phonons should also not change appreciably on passing through T_c. Because the surplus intensity in the superconducting state is at least a factor of two narrower (after correction for resolution effects) than the underlying incommensurate peaks in the normal state, it is thus improbable that electron-phonon scattering plays much of a role even in the normal state.

No calculation or previous experiment have anticipated the new phenomenon which we have discovered, namely a new length scale, indistinguishable from infinity, characterising a superconductor. Even so, it is worth putting our results in the context of other work on $\chi''(\mathbf{Q},\omega)$ of high-T_c materials. In particular, a 41 meV resonance appears in the superconducting phase of the bilayer compound $YBa_2Cu_3O_7$, and has spawned considerable theoretical activity. Although there are similarities to what we have observed in that there is a pronounced enhancement of the response at finite frequency near the zone boundary there are significant differences. The 41 meV feature $YBa_2Cu_3O_7$ is centered at the commensurate wavevector (π,π), is much broader in momentum space (the corresponding coherence length is 3-6 Å), is a narrow peak (full-width at half maximum < 2.5 meV) in energy [12] rather than the threshold phenomenon observed in $La_{1.86}Sr_{0.14}CuO_4$ where the superconducting to normal state difference does not recover to zero from $\hbar\omega = 7$ meV until at least 15 meV. In addition the 41 meV feature in $YBa_2Cu_3O_7$ shows a clear c-axis modulation which indicates it is related to the CuO bilayers, one of the defining characteristics of that compound. Even so, some of the theories for $YBa_2Cu_3O_7$ also have implications for single-layer $La_{1.86}Sr_{0.14}CuO_4$. Demler and Zhang propose a sharp collective excitation at (π,π) [13] as a feature of all cuprate superconductors. Given that the enhancement which we have discovered is neither sharp in frequency nor centered at (π,π), this is not the origin of the incommensurate signal enhancement which we have discovered for $La_{1.86}Sr_{0.14}CuO_4$ to be sharp in momentum space rather than in energy. Other approaches combine the band theory of metals displaying antiferromagnetic correlations, but not order, with a d-wave pairing state and give rise to features resembling both the 41 meV peak in $YBa_2Cu_3O_7$ and the enhanced scattering we observe in $La_{1.86}Sr_{0.14}CuO_4$ [14–20]. Analogous calculations [21,22] for s-wave pairing can mimic the 41 meV resonance for the bilayer materials, but do not yield the enhancement discovered in the present work for the single layer compound. In spite of their success in anticipating an enhanced high-ω response in the superconducting state of $La_{2-x}Sr_xCuO_4$, the available calculations miss crucial features of the data, most notably the sharpness

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in Q of the enhanced χ''. They also predict a Q-space anisotropy below $\hbar\omega \leq 4$ mev) in χ'' which, in spite of great experimental effort, has yet to be observed in $La_{1.86}Sr_{0.14}CuO_4$ [5,7].

Finally we note that there has recently been great activity on real-space domain pictures yielding incommensurate peaks [23]. It would be interesting to find out whether the strong coupling effects naturally included in such pictures could account for our data.

In conclusion, we have measured the effect of superconductivity on the magnetic response near (π,π) and at intermediate frequencies (6 meV< $\hbar\omega$ < 15 meV) in $La_{1.86}Sr_{0.14}CuO_4$. As the total moment sum rule and the singlet nature of superconductivity suggest, there is a crossing from negative to positive in the difference between magnetic spectra in the superconducting and non-superconducting state as a function of energy. However, for $\hbar\omega$ just above the zero crossing, the spectral weight added by superconductivity is extraordinarily sharp, implying that a new length scale indistinguishable from infinity characterises the superconductivity in this simple and not especially clean high-T_c material.

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FIG. 1. a) The region of reciprocal space probed in this experiment showing the trajectories used to measure the signal (solid line) and background (dashed line). b) Schematic Fermi surface for $La_{1.86}Sr_{0.14}CuO_4$ showing the near nesting condition for the spanning wavevector Q_δ. c) Simple one-dimensional illustration of how the possible spin-flip transitions contributing to $\chi''(Q,\omega)$, with Q spanning the Fermi surface, change upon the development of superconductivity. The dashed lines represent the dispersion of the normal state quasiparticles responsible for the incommensurate peaks shown in a) at low frequencies.

FIG. 2. Intensity of the incommensurate magnetic response measured along the solid line in Fig. 1 a) for $\hbar\omega = 9$ and 6.1 meV above (35 K) and below (4.6 K) T_c. The background (measured along the dashed line in Fig. 1 a)) is shown for the same energies and temperatures. The data have been normalised to a constant incident monitor, the actual counting times used in this experiment varied between 25 and 210 minutes per point. The error bars reflect the actual counting statistics. The inset shows the difference in the intensity measured at the two temperatures for 9 meV. The lines are fits described in the text.

FIG. 3. Temperature dependence of the incommensurate magnetic response at the Q_δ position for 6.1, 9, and 15 meV energy transfer. The background measured at the cross (for which $Q=|Q_\delta|$) in Fig. 1a) has been subtracted from the raw count rates. For energies below 7 meV the magnetic response is suppressed below T_c while for higher energies it is enhanced. The lines are guides to the eye. The open symbols in the bottom panel show the effect of correcting the 6.1 meV data for the thermal factor in the cross-section. For higher energies this correction is much smaller and would further enhance the observed increase in intensity below T_c.

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FIG. 4. Perspective plot of the difference in the scattered intensity in the ($\hbar\omega$=E,Q) plane between the superconducting and normal state. The momentum dependence is expressed as a function of the displacement in Å^{-1} from the incommensurate position, Q_δ. The data shown corresponds to half of the scan of Fig. 2. The measured intensity difference (I(4.6 K) - I(35 K)) is plotted together with the gaussians which best describe the energy and Q dependence observed (see text).

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41
12
53

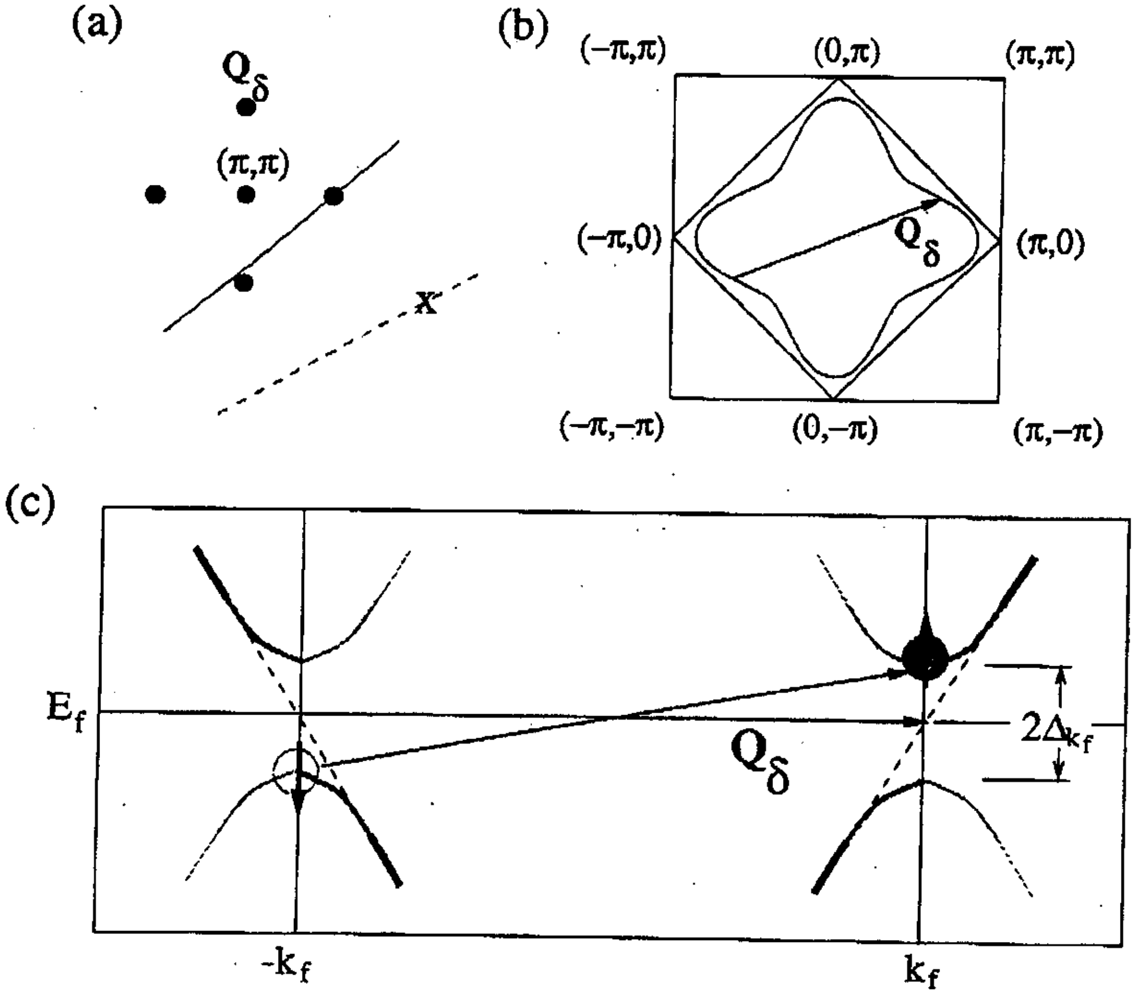
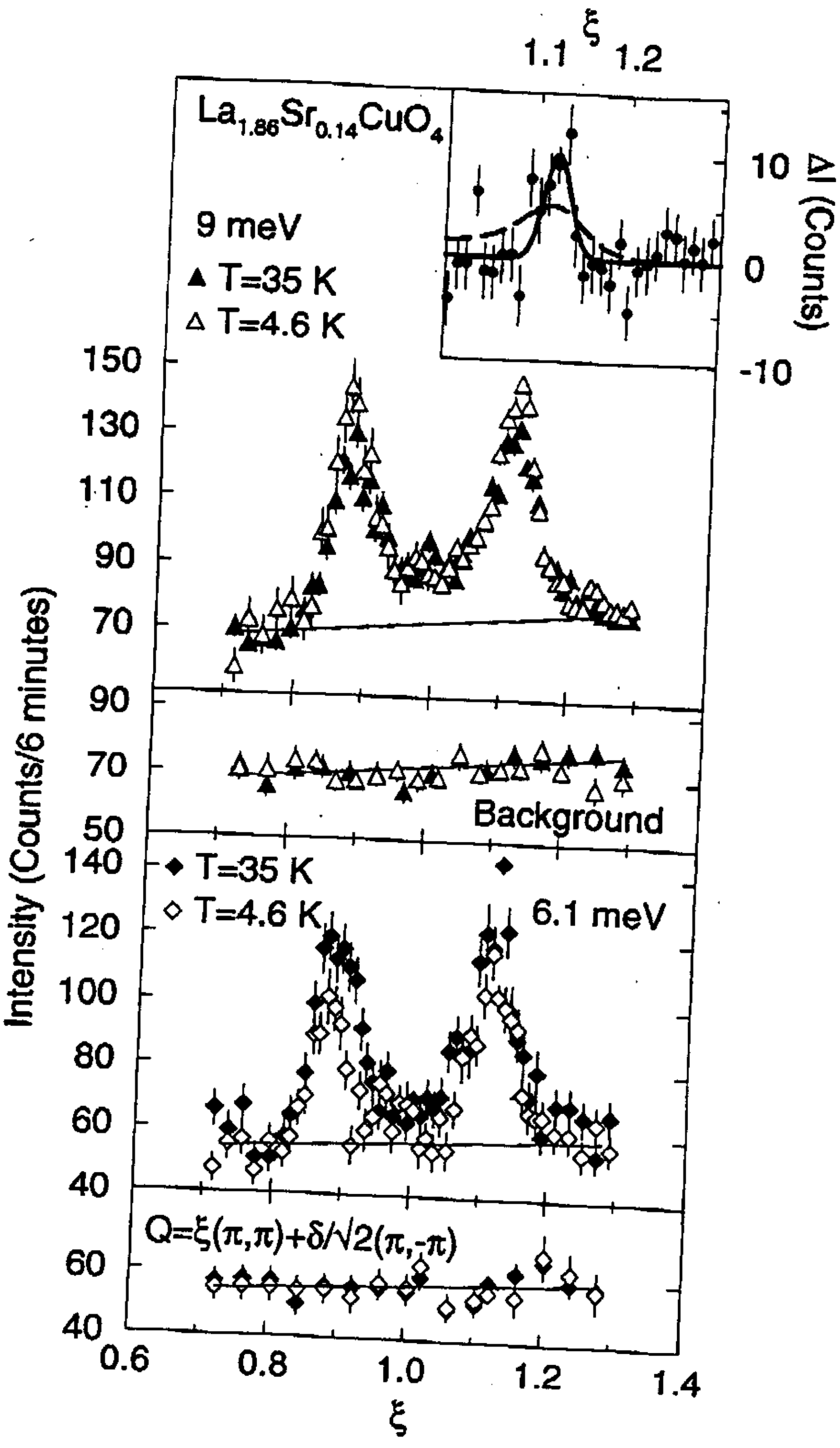
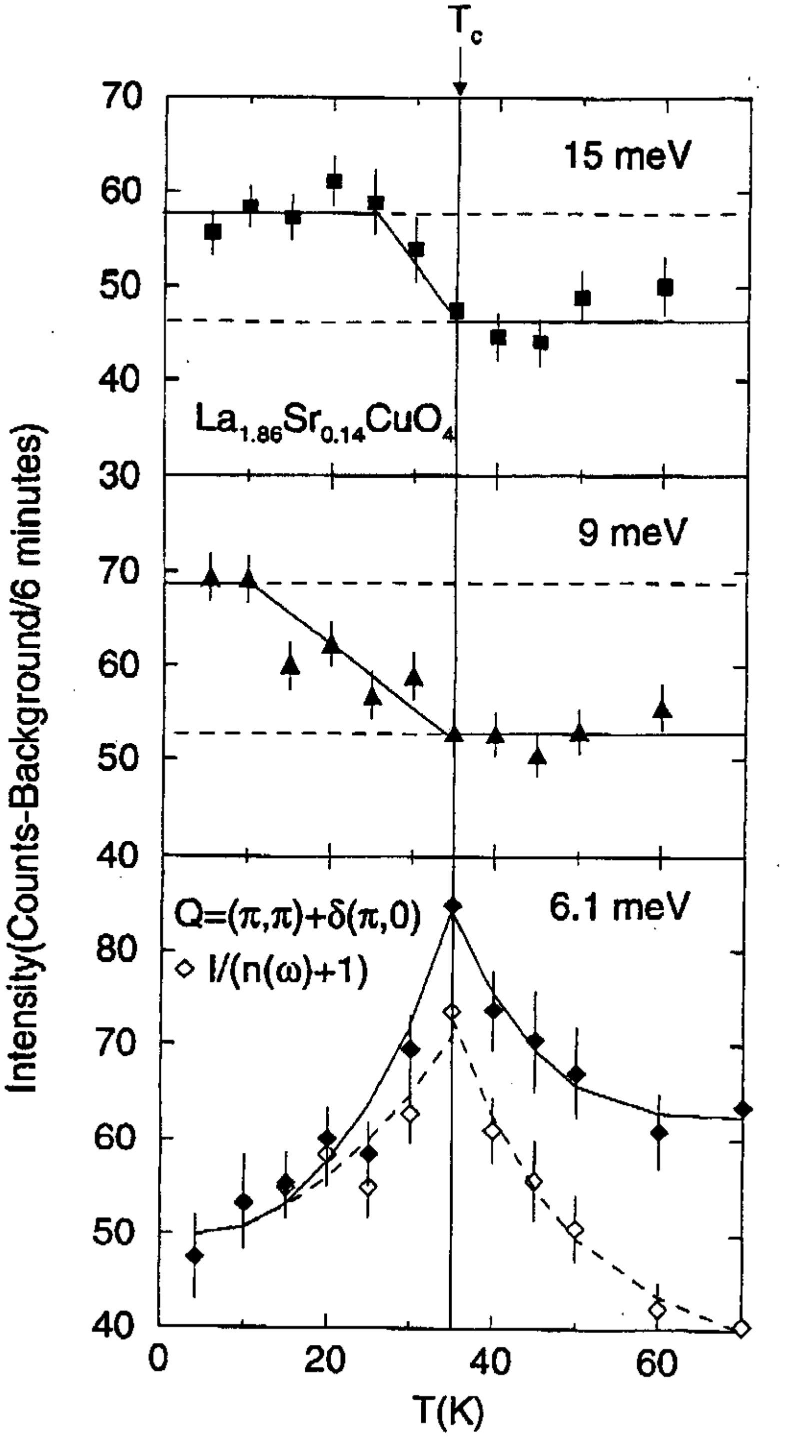

(a)
Q_δ
(π,π)
x
(b)
$(-\pi,\pi)$
$(0,\pi)$
(π,π)
$(-\pi,0)$
Q_δ
$(\pi,0)$
$(-\pi,-\pi)$
$(0,-\pi)$
$(\pi,-\pi)$
(c)
E_f
Q_δ
$2\Delta_{k_f}$
$-k_f$
k_f

Figure 1


La$_{1.86}$Sr$_{0.14}$CuO$_4$
9 meV
▲ T=35 K
△ T=4.6 K
1.1
ξ
1.2
10
0
-10
ΔI (Counts)
150
130
110
90
70
90
70
50
Background
140
120
100
80
60
40
◆ T=35 K
◇ T=4.6 K
6.1 meV
Q=ξ(π,π)+δ/√2(π,-π)
60
40
Intensity (Counts/6 minutes)
0.6
0.8
1.0
1.2
1.4
ξ


T_c
70
60
50
40
30
70
60
50
40
80
70
60
50
40
15 meV
$La_{1.86}Sr_{0.14}CuO_4$
9 meV
6.1 meV
$Q=(\pi,\pi)+\delta(\pi,0)$
◇ $I/(n(\omega)+1)$
Intensity(Counts-Background/6 minutes)
0
20
40
60
T(K)

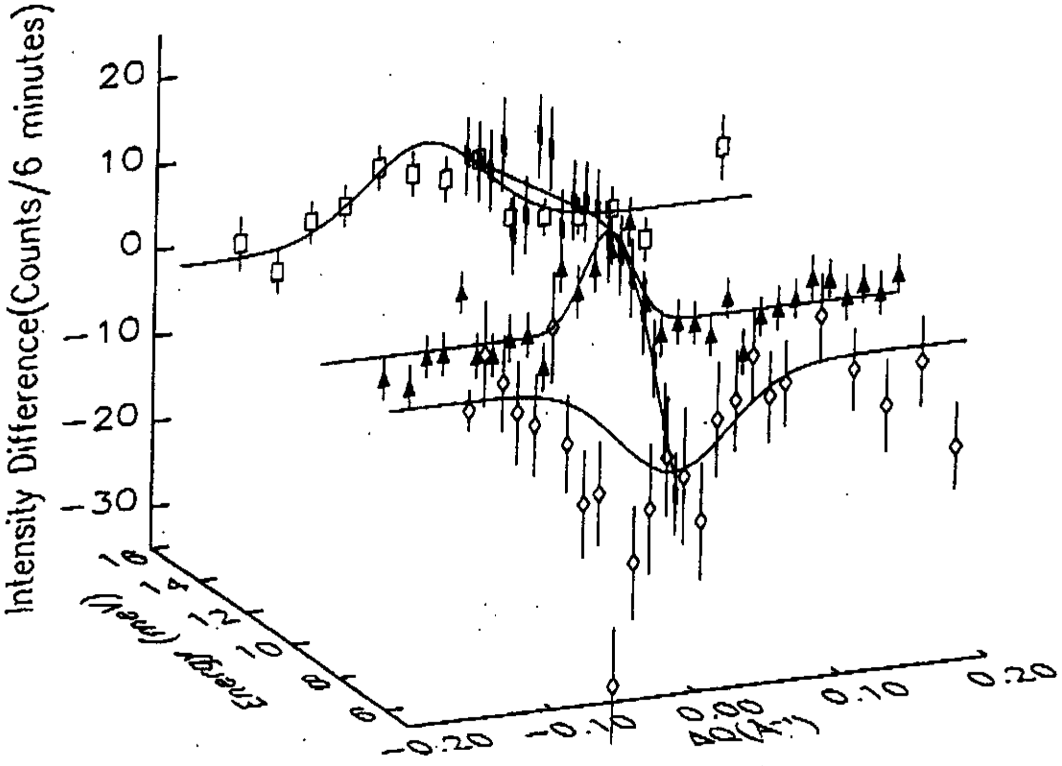


Figure 4

This one has color overlay which doesn't fax