Eur. Phys. J. B **3**, 163–167 (1998)

THE EUROPEAN PHYSICAL JOURNAL B

New slow and short range magnetic correlations in superconducting $La_{2-x}Sr_xCuO_4$

S. Petit[a], A.H. Moudden, B. Hennion, A. Vietkin, and A. Revcolevschi

Laboratoire Léon Brillouin, CE-SACLAY, 91191 Gif-sur-Yvette, France
and
Laboratoire de Chimie des solides[b], Université Paris-Sud, Orsay, France

Received: 3 September 1997 / Revised: 21 November 1997 / Accepted: 26 January 1998

Abstract. Inelastic neutron scattering measurements have been performed on high quality single crystals of $La_{2-x}Sr_xCuO_4$ in order to study the spin dynamics of this compound. In addition to the well-established incommensurate magnetic response, we show the existence of a new set of low energy excitations present in the whole superconducting region of the phase diagram. This new feature of the dynamical cross section is characterized, below about 10 K, by very short range ($\sim$ 3 lattice spacing) antiferromagnetic correlations and by a low energy scale of $\sim$ 1 meV. At higher temperatures these fluctuations become nearly Q-independent. Different possible origins of these new spin correlations are discussed.

PACS. 74.25.Ha Magnetic properties – 74.72.Dn La-based cuprates – 78.70.Nx Neutron inelastic scattering

1 Introduction

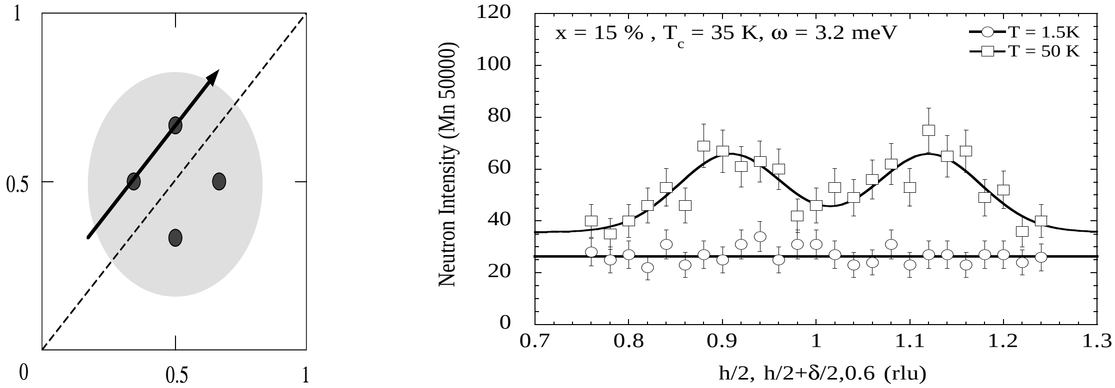
The magnetic properties of cuprates continue to attract considerable attention in the field of superconductivity. Intensive neutron studies have been devoted to the $La_{2-x}Sr_xCuO_4$ system over a wide Sr composition range [1–3,6,7]. It is now generally accepted that the fluctuations of the copper spins which are well described by standard 2D spin wave theory in the antiferromagnetic (AF) insulating state, transform in the metallic state into dynamical quasi-two-dimensional correlations peaked at incommensurate wave vectors given by $Q_\delta = (\frac{1}{2}(1 \pm \delta), \frac{1}{2})$ and $(\frac{1}{2}, \frac{1}{2}(1 \pm \delta))$, when referring to the tetragonal Brillouin zone, (see left panel of Fig. 1). The incommensurability has been discussed at length in the literature and the question whether it corresponds to a nesting property of the Fermi surface of some quasi-particles [8–10] or it is due to the stripe formations [11,12,14,15] is still debated. Its relevance to the problem of high T_c superconductivity is still unclear. This is particularly true when considering the important difference to the spin dynamics in $YBa_2Cu_3O_{6+y}$ [16], the inconsistency of this incommensurate spectrum with the NMR relaxation rates of oxygen [17,18], as well as the fascinating interplay between superconductivity, magnetism and subtle deformations of the CuO_2 planes [19–21].

In this paper we focus on detailed neutron studies of the very low energy part of the spectrum in the superconducting and normal state of $La_{2-x}Sr_xCuO_4$ which unambiguously show two distinct types of excitations. One corresponds to the well-known incommensurate response, sharply peaked in momentum space. Our experiment provide very clear evidence for the existence of a spin gap opening below T_c at these incommensurate positions, in the optimally doped sample. In addition, the experiments reported herein show clear evidence of additional slow and short range magnetic correlations represented by the dark region in Figure 1. The newly discovered response corresponds to a very broad signal in the in-plane component of the momentum transfer, centered at about $Q_{AF} = (\frac{1}{2}, \frac{1}{2})$ and which progressively emerges from background as temperature is lowered.

2 Experiments

The neutron experiments have been carried out with the triple axis spectrometer 4F1 installed on the cold source of the Orphée-LLB reactor. In all experiments, the crystals were placed in a cryostat and oriented to give access to the $[h, h, \ell]$ zones. Bent PG002 single crystals were used as monochromator and analyzer and in order to fully benefit from the focusing effects, open collimations were used. The final energy was fixed at 8 meV, resulting in an energy resolution of about 0.5 meV (FWHM), while a graphite filter was set on the scattered beam to remove higher order contaminations. Scans through two equivalent incommensurate positions which do not lie in the scattering plane were achieved using a 3D-mode control program of the triple-axis spectrometer. High quality single crystals with various doping levels ranging from $x = 0.10$ to $x = 0.20$ as

[a] e-mail: spetit@bali.saclay.cea.fr
[b] URA 446 CNRS



(a) (b)

Fig. 1. (a) Brillouin zone of $La_{2-x}Sr_xCuO_4$. The four closed circles indicate the positions of the incommensurate fluctuations. The dashed line and the arrow correspond to scans described in the text. The large dashed circular area corresponds to the new low energy signal. (b) Neutron intensity along $(\frac{h}{2}, \frac{h}{2}+\frac{\delta}{2}, 0.6)$ for $x = 15\%$ and $\omega = 3.2$ meV. Circles correspond to $T = 1.5$ K while the squares correspond to $T = 50$ K. The lines are guides to the eyes.

nominal compositions were grown using the floating zone technique [22]. Three sizable crystals of about 0.4 cm^3 in volume, with $x = 0.10$, 0.15 and 0.20 having T_c's of 25, 35 and 15 K respectively were used in the present work.

2.1 Evidence for a spin gap in the incommensurate spectrum

Figure 1 shows, for the optimally doped sample ($x = 15\%$, $T_c = T_c^{max}$), a representative Q-scan along the direction $(\frac{h}{2}, \frac{h}{2}+\frac{\delta}{2}, 0.6)$, indicated by the arrow in Figure 1. These scans were taken at a fixed energy transfer of 3.2 meV above and below $T_c = 35$ K. At 50 K, two maxima are clearly observed at the expected incommensurate positions. Cooling down to 1.5 K completely suppresses the intensity of these incommensurate peaks at this energy. This reduction of the spectral weight at low energies starting just below T_c, has been ascribed to the opening of a spin gap in the incommensurate component of the fluctuations spectrum. In the left panel of Figure 2, representative energy-scans are shown at low ($T = 1.5$ K) and high temperature ($T = 50$ K) for two constant scattering vectors, at Q_δ for the signal and at $Q_{bg} = (\frac{1}{2}(1+2\delta), \frac{1}{2}, 0.6)$ for background reference. We note that for energies larger than 3.5 meV, the intensity at Q_δ is significantly larger than the intensity at Q_{bg}, while both scans are identical for energies lower than 3 meV. Upon heating through the superconducting transition, magnetic intensity at Q_δ is restored below 3.5 meV, see Figure 2b. From these data we can estimate a spin gap energy value of 3.5 meV consistent with previous findings [3,7].

2.2 Evidence for new excitations

More importantly in the low temperature energy scan of Figure 2b one can easily observe close to the incoherent scattering, a significant signal which is present at both scattering vectors. Notice that Q_{bg} has been chosen far enough from Q_δ to avoid any contribution of the standard incommensurate response. Further, for arbitrary Q_{bg} this quasi-elastic scattering remains clearly visible within the dark area shown in left panel of Figure 1.

To get more information about the Q-dependence of this new signal, we performed, at high (50 K) and low (1.5 K) temperatures, various Q-scans at a fixed energy of 1.2 meV (an energy chosen to be well below the spin gap at the incommensurate position Q_δ) through the whole Brillouin zone, particularly along the diagonal represented by the dashed line in the left panel of Figure 1. Displayed at this large scale the featureless "background" usually subtracted for better observation of the incommensurate peaks in more restricted Q-scans, shows now a significant Q-dependence. This quasi-elastic signal clearly consists of a very broad peak centered at the AF wavevector. The evolution of the intensity along $[\frac{1}{2}, \frac{1}{2}, \ell]$ has been measured and only a small decrease could be detected for ℓ varying between -2 and 2. This evolution indicates that these correlations are compatible with the copper magnetic form factor but hardly with a pure nuclear origin of the signal. They show in addition that these excitations have a two dimensional character.

The next essential feature we would like to focus on is the temperature dependence of the quasi-elastic signal. Upon heating up to 50 K, the intensity of this low energy signal is considerably reduced and the profile flattens out as illustrated in the Q-scan of the optimally doped sample in the right panel of Figure 2, indicating a nearly Q-independent signal for all concentrations studied. More information is given in Figure 3 where the intensity at Q_{AF} and at a background reference $Q = (0.8, 0.8, 0.6)$ are displayed for $\omega = 1.2$ meV and $x = 10\%$. It is particularly interesting to note that the intensity at Q_{AF} is

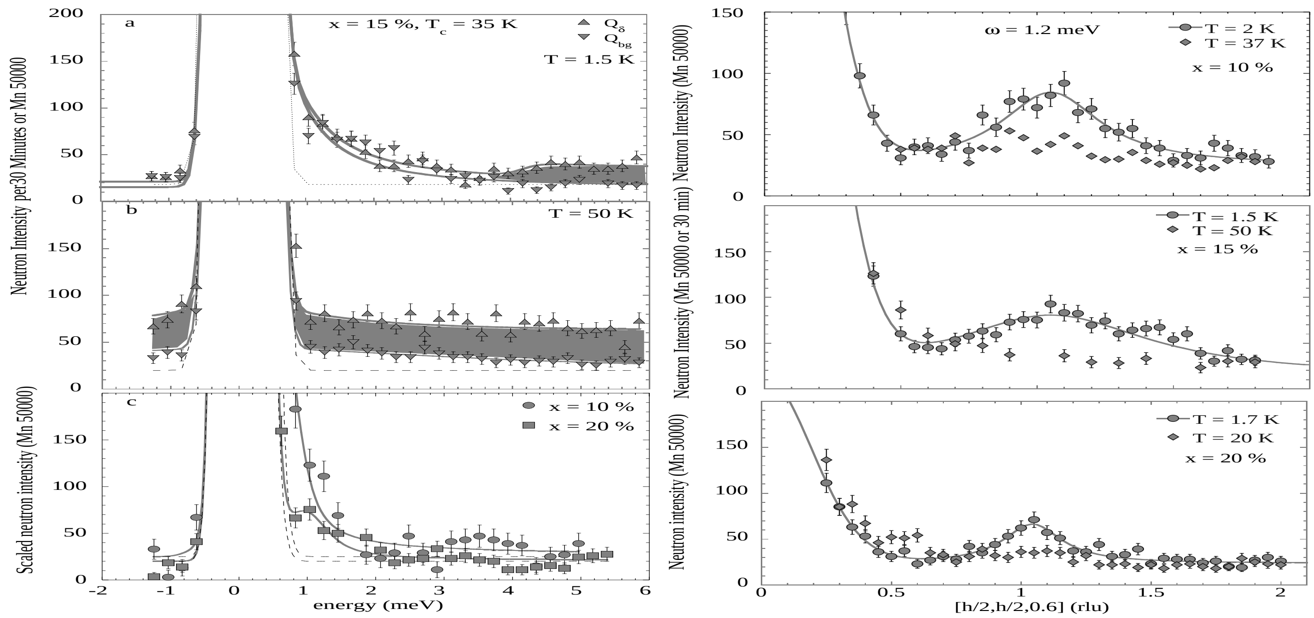


Fig. 2. Left panel: Figures a and b: neutron intensity as a function of energy transfer for $x = 15\%$, $T_c = 35$ K, corresponding to data taken at 1.5 K and 50 K respectively. The upper triangles correspond to Q_δ and the lower triangles to Q_{bg}. The hatched area shows the incommensurate fluctuation spectrum. The lines are guides to the eyes. Figure c corresponds to data, scaled to the volume of the used single crystals, taken as a function of energy transfer at Q_{AF} for $x = 10\%$, and 20% at 1.5 K. Right panel: Scaled neutron intensity, according to the sizes of the crystals, along $[\frac{h}{2}, \frac{h}{2}, 0.6]$ for $x = 10, 15$ and 20% respectively and $\omega = 1.2$ meV. Open circles correspond to $T = 1.5$ K while squares correspond to $T = 50$ K. The lines are fitted to a Gaussian profile describing the incident beam plus equation (1).

systematically above the background reference but raises substantially, below about 10 K. The emergence of the large peak centered at Q_{AF}, means that the correlations become more effective below this temperature. Note that, within the accuracy of our measurement, no particular effect is observed at T_c.

This signal is, however, not specific to the 15% doped crystal since it has been found in other compositions, in the underdoped ($x = 10\%$) as well as in the overdoped ($x = 20\%$) regimes. The right panel of Figure 2 illustrate this point, displaying the Q-dependence through the $(\frac{h}{2}, \frac{h}{2}, 0.6)$ direction at 1 meV and 1.5 K, for these two Strontium contents. The energy dependence for these dopings is shown in the left panel of the same figure. Again, we observe an asymetry between negative and positive energies in constant-Q scans, and a broad peak centered at the AF wavevector in constant energy scans. Note that the Q-width is somewhat smaller than in the optimally doped case.

3 Discussion

Because of their Q_ℓ and temperature dependence, it is reasonable to propose a magnetic origin for these excitations. This signal could be the manifestation of slowly relaxing and locally AF correlated spins. This point is now developed on the basis of simple form for the neutron scattering function that reproduces our experimental observations:

$$S(q,\omega) = (1 + n(\omega))\frac{A(T)}{(q - Q_{AF})^2 + \kappa^2}\frac{\Gamma\omega}{\Gamma^2 + \omega^2} \quad (1)$$

$$n(\omega) = \frac{1}{e^{\hbar\omega/(k_B T)} - 1} \cdot \quad (2)$$

Such a response describes the local relaxation of weakly interacting spins via an AF interaction, whose memory is lost over a coherence length $\xi = \frac{1}{\kappa}$ and after a characteristic correlation time $\tau = \frac{1}{\Gamma}$. It is remarkable that the smallest coherence length ξ is observed in the optimally doped samples while, in both underdoped and overdoped regimes they are significantly larger. This points out the singular role of optimal doping where the decay of the memory of the coupling between spins is the strongest. Thus, these data provide strong evidence for a two-component magnetic response implying two different regimes of the copper spin dynamics: the incommensurate mode and a comparatively slower and shorter range AF relaxation. To our knowledge, this novel type of fluctuations of the spectrum has not been reported so far in $La_{2-x}Sr_xCuO_4$.

One might think that these low excitations are due to extrinsic impurities trapped in the crystals, but we think that this is not the case: while a spin gap has been observed in $YBa_2Cu_3O_{6+y}$ for a long time, clear evidence for a similar feature in $La_{2-x}Sr_xCuO_4$ has only been shown recently [3,7]. The blurred gap reported by previous experiments, was thought to be caused by impurities trapped in the crystals [3,6]. This point of view is supported by recent

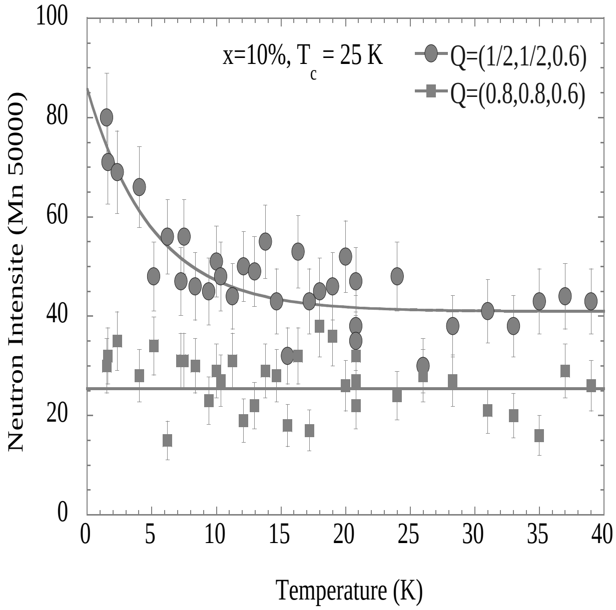


Fig. 3. Neutron intensity for $x = 10\%$ and $\omega = 1.2$ meV, as a function of temperature. Circles correspond to $Q = (\frac{1}{2}, \frac{1}{2}, 0.6)$ while squares to a background position $Q = (0.8, 0.8, 0.6)$. The lines are guides to the eye.

measurements showing that zinc doping in $La_{2-x}Sr_xCuO_4$ creates incommensurate very low energy magnetic fluctuations in the normal and superconducting state of this material [4]. A similar effect is observed in zinc-doped $YBa_2Cu_3O_{6+y}$ [5]. Considering these results, it seems that zinc-doping restores low energy fluctuations, but does not affect the Q-dependence of the spectrum. In reference [7], we showed that no spin gap at Q_δ could be observed in a underdoped $La_{2-x}Sr_xCuO_4$ single crystal, down to 1.5 K and 1.2 meV. Again, one might think that the absence of spin gap is due to impurities, but one has to emphasize that other types of perturbations can reduce the mobility of the carriers and fill the spin gap at Q_δ. Indeed, this feature might be due to the strong perturbation induced by the charge density wave instability, accompanied by the occurrence of the LTT phase, and that this competes with superconductivity around $x \sim 0.115$, *i.e.* in the underdoped regime. In the present experiment, antiferromagnetic but not incommensurate low energy fluctuations are observed. Moreover, their temperature dependence is quite different from what is observed, for example, in zinc-doped $YBa_2Cu_3O_{6+y}$. Thus, we see that the new response is quite different from what is expected in the zinc-doped case. This leads us to the conclusion that both types of low energy fluctuations do not necessarily originate from the same physical mechanism. Knowing that these low energy fluctuations are observed on different samples, we think that they correspond to an intrinsic effect, which is not related to scattering by impurities trapped in the crystals.

The interpretation of these new low energy fluctuations is certainly not unequivocal, nevertheless we propose two possible schemes. First of all, several theoretical approaches claim that the incommensurate mode may be ascribed to a particle-hole excitation arising because of nesting properties of the Fermi surface [8–10]. To explain the new low energy excitations by such approaches seems rather difficult, since the large range of Q-vectors of the new low energy fluctuations is in apparent contradiction with the sharpness of the nesting condition. It seems more appealing to suggest that these correlations correspond to the relaxation of intrinsically localized spins, AF correlated on short distances. This nearly localized band would be sensitive to the itinerant carriers as revealed by the evolution of ξ with doping: as shown, the range of the correlations reaches a minimum at the optimal doping, where concomitantly T_c and the superfluid density reach their maximum [23]. This could be interpreted as a screening effect by the itinerant carriers. The narrow band may result either in disorder-induced localized states around randomly distributed Sr-atoms [24], either to a band involving the apical oxygens [27], or to local occurrence of the LTT phase which favors charge localization restoring AF correlations: indeed, short range AF slow correlations have been observed by muon rotation studies [26] and ESR [25] in the LTT phase.

Alternatively, while the extension of the stripe picture [12,11] to the metallic regime is still debated, a second interpretation can be suggested. It has been argued that in the stripe phase observed in Nd-doped $La_{2-x}Sr_xCuO_4$ and $La_{2-x}Sr_xNiO_4$ single crystals [12,13], holes tend to condense into one-dimensional arrays separating antiphase AF domains. It is commonly assumed that if these stripes exist as well in the metallic compounds, they should exist on shorter time scales only. At sufficiently high energies, the incommensurate fluctuations in superconducting samples might be attributed to a spin-wave-like collective mode in the AF domains, while at low energies, the scattering is expected to be dominated by the slow motion of spins in the vicinity of these stripes, meandering and progressively slowing down as the temperature is lowered. In this sense, our new signal could well correspond to such a meandering. New data are required to distinguish between these two possibilities. Particularly, a clear nuclear response signalling the segregation of the charge in stripes would be an experimental test in favour of the stripe picture.

Recently, Walstedt *et al.* and Berthier *et al.* explicitly discussed the necessity of a Q-independent contribution to the susceptibility to describe the behaviour of the planar ^{17}O relaxation rate [17,18]. It is interesting to note that though quite difficult to extract in the high temperature range, the new quasi-elastic signal becomes weak and almost Q-independent. The experiments suggest that the evidence for an additional response in the low energy range, coexisting with the incommensurate energy fluctuations, should modify the Q-dependence of $\chi''(q,\omega)$ at the NMR energy scale. We suspect that this might restore the correct temperature dependence of this relaxation rate and might reconcile the two types of measurements.

4 Conclusion

In conclusion, we have shown clear evidence for the existence of a quasi-elastic signal developing at low temperatures and centered at the AF point. The persistence of this type of fluctuation from the underdoped to the overdoped samples and its coexistence with a clear spin gap at the incommensurate Q_δ, give very strong support in favour of an intrinsic origin, excluding scattering from possible impurities trapped in the structure. Possible interpretations have been proposed, addressing the origin of these new low-energy fluctuations.

We would like to thank C. Berthier, V. Emery, F. Onufrieva, L.P. Regnault for stimulating discussions, M. Braden for additional studies on his 15% doped crystal and P. Baroni for its technical support.

References

1. R.J. Birgeneau *et al.*, PRB **39**, 2868 (1989).
2. T.R. Thurston *et al.*, PRB **46**, 9128 (1994).
3. K. Yamada *et al.*, PRL **75**, 1626 (1995).
4. K. Yamada *et al.*, submitted to PRL.
5. Y. Sidis *et al.*, PRB **53**, 6811 (1996); A.V. Mahajan *et al.*, PRL **72**, 3100 (1994); K. Ishida *et al.*, Physica B **186-188**, 1015 (1993).
6. G. Aeppli *et al.*, PRL **62**, 2052 (1991).
7. S. Petit *et al.*, Physica B **234 236**, 800-802 (1997).
8. F. Onufrieva, S. Petit, Y. Sidis, PRB **54**, 12464 (1996); F. Onufrieva *et al.*, PRB **52**, 7572 (1995).
9. P.B. Littlewood, PRB **48**, 487 (1993).
10. J.P. Lu *et al.*, Physica C **179**, 191 (1991).
11. V.J. Emery, S. Kivelson, Physica C **209**, 597 (1993); V.J. Emery, S. Kivelson, PRB **46**, 10812 (1992).
12. J. Tranquada *et al.*, Nature **375**, 561 (1995); J. Tranquada *et al.*, PRB **54**, 7489 (1996).
13. J. Tranquada *et al.*, PRL **79**, 2133 (1997).
14. H.J. Schultz, J. Phys. France **50**, 2833 (1989).
15. D. Poilblanc *et al.*, PRB **39**, 9749 (1989).
16. for a review, see J. Rossat-Mignot *et al.*, in *Selected Topics in Superconductivity* (World scientific, 1992), p. 265.
17. R.E. Walstedt *et al.*, PRL **72**, 3610 (1994).
18. C. Berthier *et al.*, J. Phys. France **6**, 2205 (1997).
19. J. Axe *et al.*, PRL **62**, 2751 (1989).
20. B. Büchner *et al.*, Euro. Phys. Lett. **21**, 953 (1993).
21. L. Vasiliu-Doloc *et al.*, Physica B **213**, 63 (1995).
22. A. Revcolevschi, J. Jegandez, in *Coherence in high T_c superconductivity*, edited by G. Deutescher, A. Revcolevschi (World Scientific, 1996), p. 19.
23. Y.J. Uemura *et al.*, PRL **66**, 2665 (1991).
24. L. Martin, PRL **75**, 744 (1995).
25. V. Kataev *et al.*, PRB **55**, 3394 (1997).
26. K. Kumagai *et al.*, J. Superconductivity **7**, 63 (1994).
27. International Workshop on *Phase Separation in Cuprates Superconductors*, Cottbus, 4-10 September 1993, edited by K.A. Muller, E. Sigmund. See references therein.