NEUTRON SCATTERING STUDIES OF $YBa_2Cu_3O_{6+x}$ SINGLE CRYSTALS

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Abstract

The magnetic phase diagram of $YBa_2Cu_3O_{6+x}$ system is summarized. Inelastic neutron studies of $YBa_2Cu_3O_{6+x}$ single crystals are reported. In the pure AF phase ($x=0.15$, $T_N=410K$) the 2D character of the spin wave spectrum and the exchange parameters have been established. A small amount of p-holes in the CuO_2 planes ($x=0.37$, $T_N=180$ K $n_h=1.8$ %) strongly modifies the spin dynamics at low-q: strong damping of in-plane spin excitations and renormalization of the spin wave velocity. In the superconducting state ($x=0.45$, $T_c=35$ K) we have found dynamical magnetic correlations and an anomalous decrease of the intensity at low energy in the vincinity of T_c.

Introduction

Since the discovery of superconductivity in lamellar copper oxide materials a huge amount of experimental and theoretical work has been performed but there is no consensus on the mechanism of superconductivity. In order to clarify the physics involved accurate single crystal experiments must be performed. In this context the neutron scattering technique plays an important role because both the spatial and temporal spin fluctuations can be probed. A detailed investigation of

(received November 28, 1989)

the $(La,Sr)_2CuO_4$ system has been carried out at Brookhaven (1) and large antiferromagnetic spin correlations have been found above T_N and in Sr doped samples. At the Centre d'Etudes Nucléaires de Grenoble we have focussed our efforts on the $YBa_2Cu_3O_{6+x}$ system. This system has the advantage that by changing only the oxygen content we can investigate successively different interesting regimes.

In this paper we will first summarize the results obtained by neutron diffraction on the magnetic phase diagram of the $YBa_2Cu_3O_{6+x}$ system (2,3). The results of inelastic neutron scattering studies on crystals with selected oxygen contents characteristic of the undoped antiferromagnetic state (x=0.15), of the magnetic state just below the critical concentration for desappearence of long range ordering (x=0.37) and the non magnetically order and superconducting state (x=0.45) will be presented (4-6).

Experimental

Neutron scattering experiments were performed on single crystals using three-axis spectrometers, mainly IN8, at the Institut Laue-Langevin. Details of the experimental procedure will be given in a more extended paper. We have grown large single crystals of good quality in which the oxygen content can easily and homogenously be changed from x=0 to x=1. The single crystal (0.40 cm^3) was mounted in standard ILL cryostats with the [110] and [001] axes in the scattering plane.

The phase diagram

The phase diagram (temperature, concentration) of $YBa_2Cu_3O_{6+x}$ is reported in figure 1. For any concentration $x < 0.4$ the system develops the same antiferromagnetic ordering to which only the Cu(2) ions, forming two Bravais sublattices at (0,0,z) and (0,0,-z), participate. The wave vector

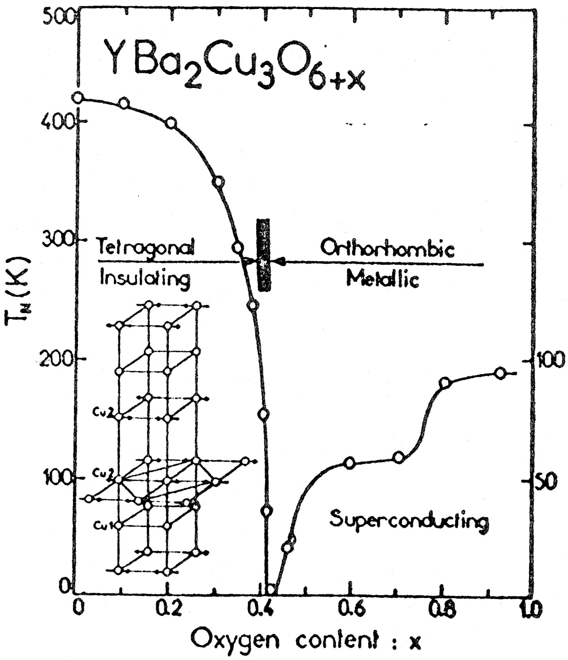

Fig.1 Phase diagram of $YBa_2Cu_3O_{6+x}$

of this ordering $\vec{k} = [1/2,1/2,0]$ corresponds to a (+-) sequence of moments along the a and b directions. The coupling between the two Bravais sublattices is antiferromagnetic and the magnetic moments ly in the (a,b) plane. If the same ordering is observed in the whole antiferromagnetic range some noticable differences occur in the magnetic behaviour allowing to define two distinct regimes. For low oxygen concentrations $x < 0.20$ no effect of the additional oxygen is observed, the Neel temperature T_N and the low temperature ordered moment m_0 keep nearly the same values as in $YBa_2Cu_3O_{6+x}$ (T_N=410 K, m_0=064 μ_B). For larger oxygen contents T_N and m_0 decrease first gradually with x and drop abrubtly to zero on approaching the critical concentration x=0.4 above which no long range magnetic ordering develops. In this concentration range $0.20 < x < 0.40$ some disorder is observed as illustrated in figure 2 for x=0.30 and x=0.37. In this figure we report the temperature dependence of the Bragg intensity corresponding to the ordered part of the magnetic moment and of the diffuse elastic scattering arising along the $(1/2,1/2,\ell)$ ridge which corresponds to a static disorder in the stacking of the CuO_2 planes. The width of this ridge, larger than the experimental resolution, implies also an in-plane disorder. For x=0.30 the variation of the moment is Brillouin like at high temperature but a reentrant behaviour is observed below T=50 K. It is characterized by a decrease of the ordered moment and a simultaneous occurence of the scattering in the rod. In contrast, for x=0.37 the moment variation is no longer Brillouin like and the diffuse scattering is observed up to and even above T_N. The width of the ridge (Γ_q=0.015 r.l.u) corresponds to an in-plane correlation length ξ of 7.5 unit cells. This behaviour originates from the holes created in the CuO_2 planes. For small hole concentrations (n_h) we can expect ξ to be the distance between magnetic defects ($n_h=1/\xi^2$) and then one can estimate a concentration of holes n_h=1.8 %. So the 3D-AF ordering is destroyed for a hole concentration of about 2%, a result quite similar to that found in the $(LaSr)_2CuO_4$ system (1).

The spin dynamics

The study of the wave spectrum allows, in an antiferromagnet, to derive the exchange and anisotropy parameters. In the case of $YBa_2Cu_3O_6$ the main exchange integral J, which couples nearest neigbour Cu ions in a CuO_2 plane, is obtained from the dispersion law $\omega(q)$ along the $(1/2+q,1/2+q,\ell)$ direction and the weak coupling J' between bilayers is deduced from the dispersion along the $(1/2,1/2,\ell)$ direction. The coupling between the two layers J_b is given by the gap of the optical modes while the X-Y anisotropy term $\Delta J/J$ is measured from the gap of the out-of-plane acoustical modes.

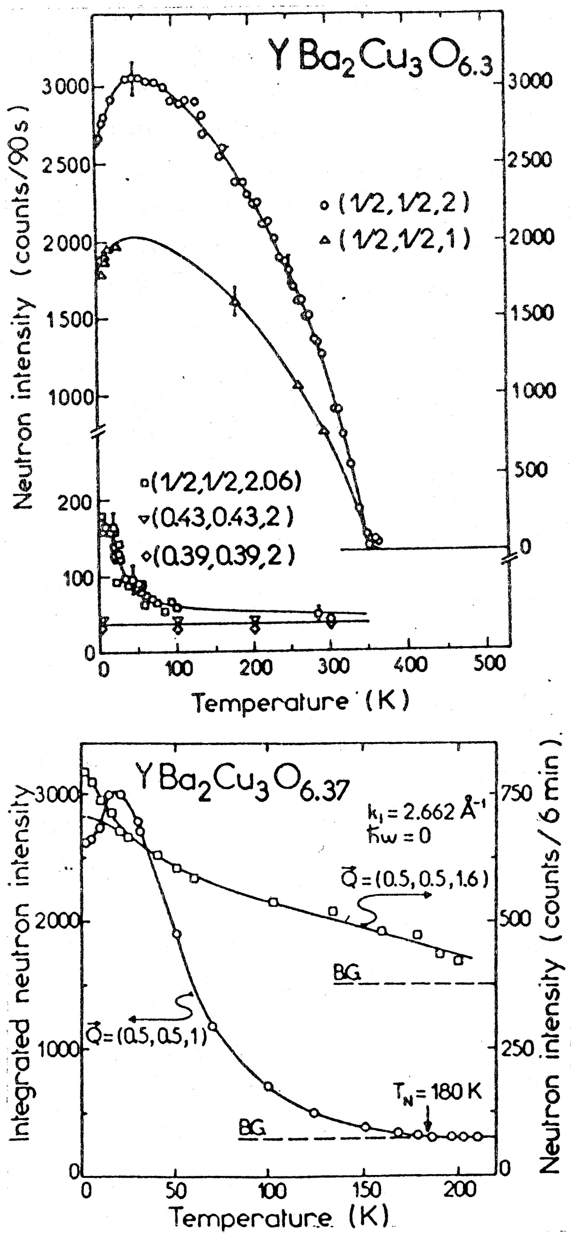

Fig.2 Temperature dependence of the magnetic Bragg intensity and of the diffuse scattering for $YBa_2Cu_3O_{6+x}$ with x=0.3 and 0.37

The pure A-F state : x = 0.15

Energy scans for Q=(1/2,1/2,ℓ) clearly show a double peak structure (Fig.3). The abscence of the high energy peak for large ℓ values establishes that the low energy part of the spectrum can be assigned to in-plane spin excitations and the high energy part to out-of-plane spin excitations. The modulation of the intensity by the structure factor resulting from the AF coupling of the two Bravais sublattices proves the acoustical nature of these excitations.

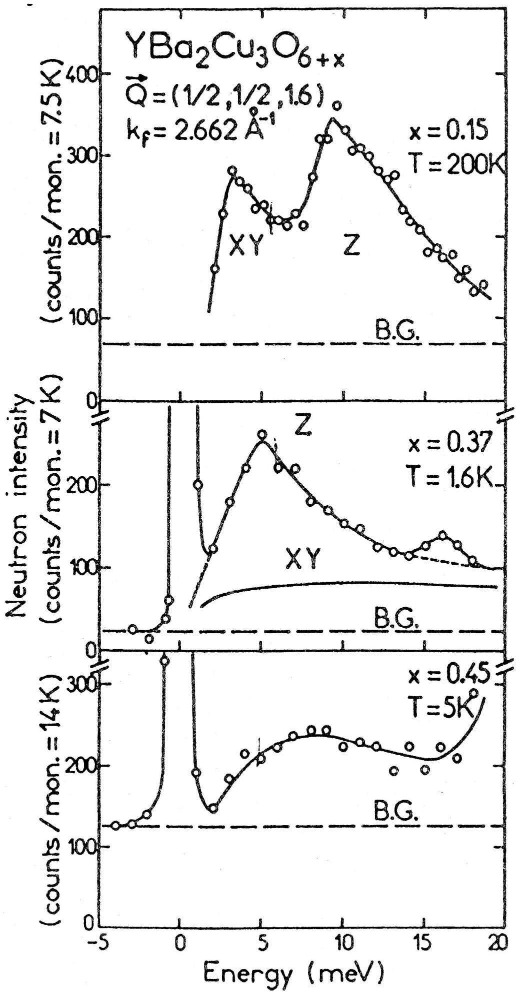

Fig.3 Energy scans performed at Q=(1/2,1/2, 1.6) for $YBa_2Cu_3O_{6+x}$ with x=0.15, 0.37, 0.45

Q-scans for energy transfers up to 50 meV give a single peak; the l-width is close to the experimental resolution (Δq_{res}=0.017 r.l.u.) up to 15 meV (Fig.3a) and only around 30 meV it has a value twice Δq_{res}. The deconvolution of these data yields the dispersion curve given in Fig.4 from which we can deduce an extremely large spin wave velocity $c_o=4\sqrt{2}$ SaJ= =1 ± 0.1 eVÅ assuming no spin wave damping and a classical theory. (Quantum efects are expected to reduce by about 17% this value). So the in-plane Cu-Cu interaction is 2J=1700 K when there is no hole in the CuO_2 planes. The dispersion along (00ℓ) of in-plane excitations (ΔE=1.6 meV) yields an inter-bilayers coupling $J'=10^{-5}$ J ($\Delta E=8JS\sqrt{J'/J}$). The in-plane exitations do not present any observable gap implying a very small in-plane anisotropy. Moreover no optical mode has been detected up to 50 meV which allows only to give a lower limit for the coupling inside the bilayers, $J_b/J > 10^{-2}$ (ΔE_{opt}= $=8JS\sqrt{J_b/J}$). Clearly these results establish that $YBa_2Cu_3O_{6.15}$ is a S=1/2 bilayer Heisenberg antiferromagnet with weak XY anisotropy ($\Delta J/J \simeq 10^{-4}$) and interplanar coupling ($J'/J \simeq 10^{-5}$).

The A-F state with p-holes : x = 0.37

Prior experiments on $YBa_2Cu_3O_{6.3}$ (3) have shown that the spin wave velocity is renormalized when x increases. Therefore to study the influence of oxygen p-holes on the spin dynamics in the antiferromagnetic state we have prepared a sample $YBa_2Cu_3O_{6+x}$ with x=0.37 close to the critical concentration x_c=0.40. This sample orders at T_N=180 K and no sign of superconductivity was detected (7).

We can see from Fig.3b that energy scans, performed at T=1.6 K are strongly different from those for x=0.15. Out-of-plane spin excitations have still a propagative character and the anisotropy gap is reduced by a factor two (ΔE=2.5 meV). In-plane spin exitations are now overdamped (diffusive character) with a characteristic energy Γ_ω=17 meV. Such a behaviour is expected because, in this energy range ($\hbar\omega$<20 meV), the

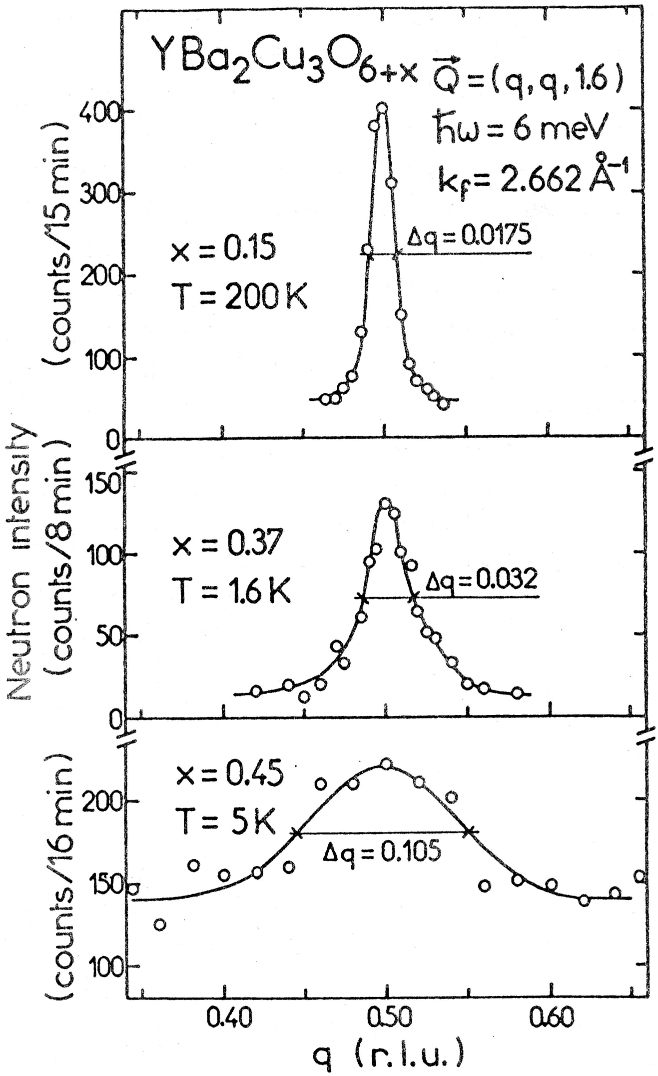

Fig.4 Q-scans performed for an energy transfert $\hbar\omega$=6 meV for $YBa_2Cu_3O_{6+x}$ with x=0.15, 0.37, 0.45. The q-width due to the resolution is Δ=0.017 r.l.u.

wave vectors (wavelength) are smaller (larger) than $\Gamma_q(\xi)$. q-scans performed at energy transfers of 6 and 12 meV show single broadened peaks as shown in Fig.4b (Γ_q=0.015 r.l.u. at $\hbar\omega$=6 meV) indicating a softening of the spin wave velocity. However q-scans at higher energies, around 30 meV, do not show any indication for a double peak arising from the spin

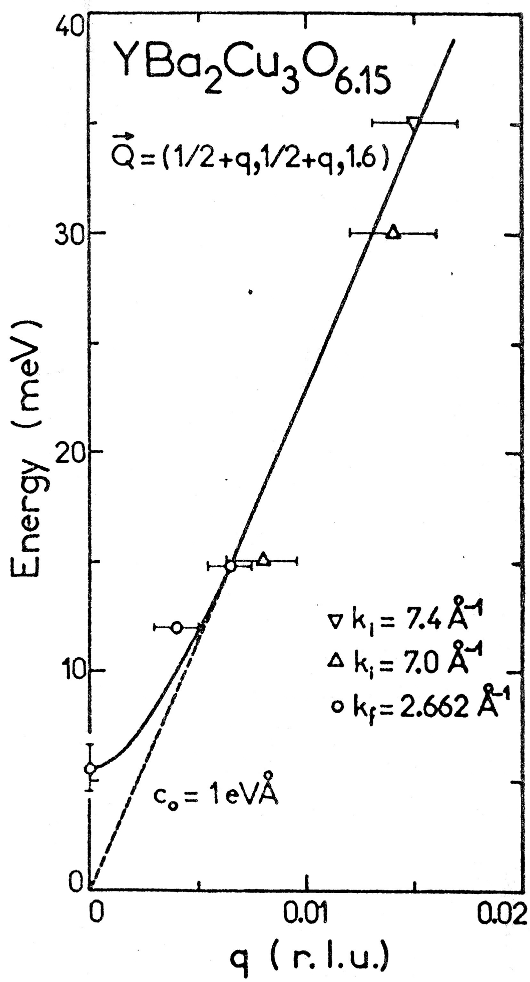

Fig.5 Spin wave energies for small wave vectors around the antiferromagnetic Bragg point of the AF sample $YBa_2Cu_3O_{6.15}$. The dotted line corresponds to a spin wave velocity c_0=1 eV Å.

wave with wave vectors +q and -q. The main reason of this result is that in-plane and out-of-plane excitations behave differently. In order to separate these two contributions the same q-scans have be performed around $\vec{Q}$=(1/2,1/2,1.6) and (1/2,1/2,5.2) for energy transfers of 6 and 12 meV, at higher energy the contribution of the in-plane component is dominant. The obtained q-widths are larger for the in plane component, at $\hbar\omega$=6 meV Δ_{qz}=0.030 r.l.u. and Δ_{qxy}=0.052 r.l.u. (Δ_{qres}= =0.017 z.l.u.). The deconvolution of the data is reported ir Fig.6.

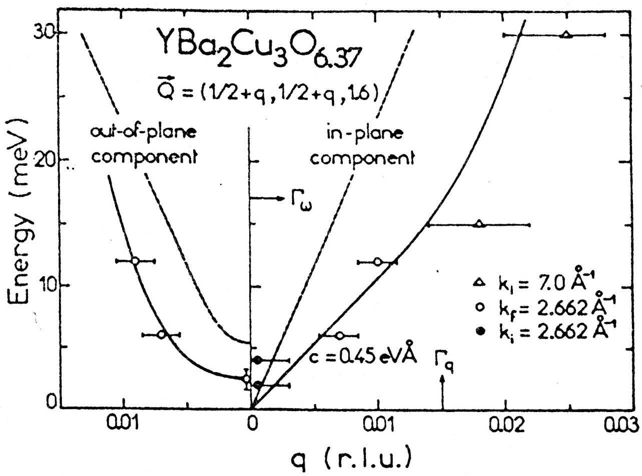

Fig.6 Excitation energies of in-plane and out-of-plane spin components for $YBa_2Cu_3O_{6.37}$. The renormalization of excitation energies is large at small q as shown by the comparison with the undoped sample (dotted line).

For the out-of-plane component no damping was found, whereas a very large damping (Γ_ω=17 meV) was determined for the in-plane contribution. The obtained results clearly indicate that the renormalization of the spin wave energies is q-dependent and reaches a factor two at small wave vectors ($q < \Gamma_q$). The spin wave velocity c_0=0.45 ± 0.1 eVÅ can be deduced indicating a large reduction by a small amount of p-holes. While it is not possible to get experimental data we can anticipate that the renormalization in negligible for $q \gg \Gamma_q$ which means that the Cu-Cu superexchange coupling is not affected. It is worth noting that the damping Γ_ω=17 meV is related to Γ_q=0.015 r.l.u. by the simple relation $\Gamma_\omega = 2.3\, c_0\, \Gamma_q$. Therefore the main effect of the oxygen p-holes at low-T is to produce some local static disorder, i.e. some kind of magnetic polarons. These polarons strongly disturb the propagation of in-plane spin excitation and reduce the spin wave stiffness which is likely to vanish when the hole concentration reaches the critical value $n_h^c \approx 2$ % (x_c=0.40). It must be emphasized that the spin dynamics described above was observed at T=1.6 K when the holes are localized. At higher temperature (T > 10 K) the holes begin to move, as demonstrated by the reentrant behaviour, therefore the spin dynamics becomes more complex and the AF-ordering is reduced.

<u>The metallic regime : x = 0.45</u>

In order to understand how the superconductivity develops we have investigated a sample with an oxygen content in the superconducting region but close to the border line: $YBa_2Cu_3O_{6.45}$. No trace of 3D-AF ordering has been found down to T=1.6 K, however a.c. suscpetibility measurements (7) have given evidence of a very sharp (ΔT_c=2K) superconducting transition at T_c=35 K. Therefore superconductivity appears just above the critical concentration x_c=0.40 and T_c increases sharply with x up to a plateau of about 60 K (7). At low temperature (T=5 K) energy scans (Fig.3c) and q-scans (Fig. 4c) give evidence for a magnetic scattering. Clearly propagative spin exitations do not exist any more. q-scans give a q-width Δq=0.11 r.l.u. pratically independent of the energy transfer yielding a value Γ_q=0.050 r.l.u. (0.11 $\mathring{A}^{-1}$), i.e. a correlation length ξ=2.2 a. This value is too small to be used for an estimation of the hole concentration due to the break down of the relation $n_h=1/\xi^2$. High energy scans up to 40 meV indicate that this contribution extends up to about 30-40 meV. Therefore we conclude that in this superconducting regime there are only short range dynamical magnetic correlations. The scattering has been investigated in details as a function of temperature and a quite unusual behaviour has been observed. A typical example is reported in Fig.7 which shows the temperature behaviour of the intensity measured at $\vec{Q}$=(1/2,1/2,1.6) for energy transfers $\hbar\omega$=2 and 6 meV. For

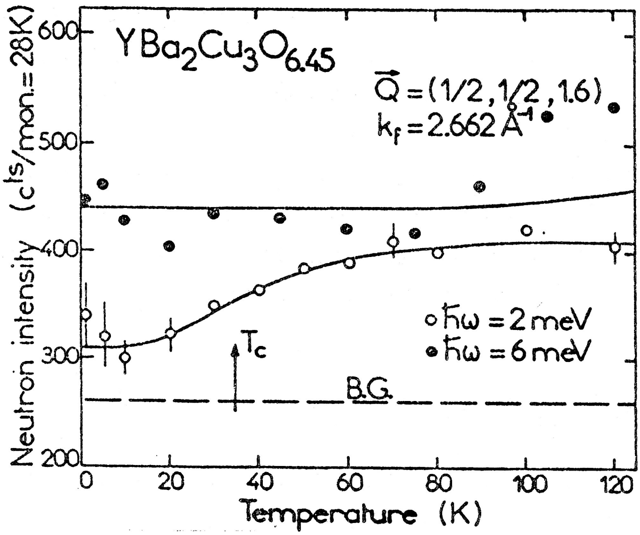

Fig.7 Magnetic intensity as a function of temperature measured at $\vec{Q}=(1/2,1/2,1.6)$ for energy transfers $\hbar\omega=2$ and 6 meV for $YBa_2Cu_3O_{6.45}$

$\hbar\omega > 5$ meV the intensity is almost T-independent whereas for low energy transfers the magnetic scattering gradually decreases when cooling from 60 K down to 10 K. For $T < 10$ K the inelastic magnetic scattering is supressed below $\hbar\omega=2$ meV which may indicate the existence of some magnetic gap in the magnetic excitation spectrum. More details of this study will be reported in a more extended paper.

Conclusion

The above inelastic scattering results have provided the exchange parameters of the pure AF-state and a detailled description of the change in the spin dynamics produced by magnetic defects created around oxygen p-holes : at low q a strong damping of the in-plane spin excitations and a renormalisation of the spin wave velocity. In the superconducting state the magnetic correlations are shorter and the magnetic excitations exhibit an unusual T-behaviour for low energy transfers. Clearly further extensive studies have to be undertaken in a near future in the superconducting states with T_c=60 K (x=0.66) and T_c=92 K (x=0.92).

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