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NEUTRON SCATTERING STUDY OF THE ANTIFERROMAGNETIC ORDERING IN $YBa_2Cu_3O_{6+x}$ POWDER AND SINGLE CRYSTAL SAMPLES

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Neutron diffraction experiments have been performed on powder and single crystal (2x5x2x1.5 mm^3) samples of $YBa_2Cu_3O_{6+x}$ compounds. The reduced compound $YBa_2Cu_3O_6$ is found to order antiferromagnetically below $T_N = 420 \pm 10$ K. The magnetic moment is about 0.6 ± 0.05 μ_B on the Cu^{2+} sites and aligned perpendicular to the tetragonal c-axis. The magnetic structure is characterized by an in-plane wave vector $|\frac{1}{2}\ \frac{1}{2}\ 0|$ and an antiferromagnetic coupling between the two CuO_2 planes of the unit cell. The addition of oxygen induces only a decrease of T_N and the moment value (290 ± 10 K, 0.45 ± 0.05 μ_B and 245 ± 10 K, 0.28 ± 0.05 μ_B, for $x = 0.25 \pm 0.03$ and 0.38 ± 0.03, respectively). The long range order disappears abruptly around $x = 0.40$.

INTRODUCTION

Since the discovery of superconductivity in $(La\text{-}Ba)_2CuO_4$ ($Tc \simeq 40$ K) (1) and in $YBa_2Cu_3O_{6.9}$ ($Tc \simeq 90$ K) (2) many theoretical models have been developped to account for these high Tc values. They are based on the classical electron-phonon interaction or on less conventional mechanisms involving charge or spin fluctuations. Therefore one clue for the understanding of the origin of the actual mechanism involved in high Tc superconductors might be found in the study of magnetic properties. In this respect, neutron scattering experiments are of crucial importance to bring more information on the magnetic properties of the copper ions in these superconducting compounds. Magnetic fluctuations in $YBa_2Cu_3O_7$ have been investigated out on a powder sample using a 3-dimensional polarized neutron analysis (3). The total magnetic intensity integrated up to an energy of 20 meV was found to be negligibly small and the authors conclude that mechanisms involving magnetic fluctuations are probably inappropriate to explain superconductivity in $YBa_2Cu_3O_7$. However the discovery of antiferromagnetic ordering in La_2CuO_4 below $T_N \simeq 240$ K (4, 6) demonstrated that d electrons are rather localized in a $3d^9$ configuration as in Cu^{2+} compounds and that magnetism plays a key role in the superconductivity mechanism. Therefore we have undertaken a systematic study of the magnetic scattering in the system $YBa_2Cu_3O_{6+x}$ as a function of the oxygen content. This programme is a part of a more extended one developped in the Département de Recherche Fondamentale of the Centre d'Etudes Nucléaires de Grenoble (7-9). Apart from a higher Tc value, the system $YBa_2Cu_3O_{6+x}$ offers the avantage over the $(LaSr)_2CuO_4$ system that the oxygen amount can be easily changed yielding to a wide variety of properties ranging from the insulating compound $YBa_2Cu_3O_6$ to the best superconducting compound $YBa_2Cu_3O_{6.92}$ (9). This offers the possibility of studying the magnetic properties of the normal compound $YBa_2Cu_3O_6$ before investigating their evolution as a function of the oxygen content.

Powder neutron diffraction experiments performed at Brookhaven (10) and at ILL (11) have established that the simple insulating compound $YBa_2Cu_3O_6$ actually orders antiferromagnetically at rather high temperature ($T_N \simeq 500$ K) with an in-plane coupling ($\vec{k} = (\frac{1}{2}, \frac{1}{2}, 0)$) between Cu^{2+} moments which is similar to that found in La_2CuO_4. Muon spin rotation (μSR) measurements (12) also show evidence for some antiferromagnetic order occuring near room temperature for $x < 0.2$. Single crystal neutron diffraction experiments reported in this paper establish that the antiferromagnetic ordering remains unchanged up to $x \simeq 0.4$ where it desappears abruptly. The only changes are a decrease of T_N from 420 K down to 240 K and a reduction of the ordered moment value from 0.6 μ_B to 0.3 μ_B. We first present the experimental conditions as well as sample preparation and characterization. The results obtained on four oxygen concentrations x = 0, 0.25, 0.38 and 0.42 will be reported and discussed.

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EXPERIMENTAL

Neutron scattering experiments were performed on the three-axis spectrometers IN8 and IN20 at the high flux reactor of the Institut Laue Langevin. Most of the measurements were carried out with a wave length of 2.36 Å given by a Ge (111) crystal monochromator and furthermore P.G. filters were used to reduce higher order contamination at a level as small as 10^{-6}. Bragg intensities were measured in a two axis-configuration with relaxed α_3 collimation. Polarized neutrons were used to establish the magnetic origin of the observed intensities.

The crystal was mounted with a $|1\bar{1}0|$ axis vertical in a standard ILL cryofurnace which allows to vary the temperature from 1.5 K to 550 K. Then any direction in the $|110|$, $|001|$ reciprocal plane can be investigated.

SAMPLE PREPARATION AND CHARACTERIZATION :

Single crystals used in the present neutron scattering experiments were grown at the University of Rennes using a mineralization method. A stochiometric (or nearly) composition is heated at a temperature slightly lower than the peritectic decomposition during 24 hour and cooled down in air at a rate of 100°C/hour. Then an anealing at 450°C under an oxygen flow is performed during 12 hours.

This crystallization method was successful in producing good quality single crystals for neutron diffraction experiments with a mosaic spread $\eta \simeq 0.2\text{-}0.3°$. A large crystal, obtained by this method, of about 2.5 x 1.5 x 1 mm^3 has been selected for the neutron diffraction experiments. This method produces single crystals in which oxygen can be easily introduced or removed. Actually oxygen content up to $x = 0.9$ can easily be achieved as indicated by the high value of $T_c = 90$ K and the sharpness of the superconducting transition $\Delta T_c \simeq 1$ K.

However it is mandatory to introduce a well defined quantity of oxygen in the crystal. This can be achieved in the following way. First the crystal and a weighted amount (∿ 2g) of $YBa_2Cu_3O_y$ powder, placed inside a quartz tube, are reduced to $YBa_2Cu_3O_6$ by heating under vacum at T = 675°C during 12 hours. After cooling, the tube is filled, at room temperature, with a well controled mixture of argon and oxygen gaz. The closed tube is then heated at 700°C during 3 hours and cooled down to room temperature at a rate of 100°C/h.

It is well known now (13, 14, 8) that only the oxygen occupation of the O1 site (0 ½ 0) of the Cu-O plane containing the Cu-O chains can be modified from $x = 0.92$ to $x = 0$ which induces a change in the crystal structure from orthorhombic to tetragonal symmetry. The oxygen gives rise to a decrease of the c-parameter which was interpreted as the consequence of the increase of the Ba^{2+}-$O1^{2-}$ electrostatic interaction (8). Therefore a careful measurement of the lattice parameter c yields a good determination of the oxygen content. The calibration of c versus x was done by using the recently published data of ref. (15) which is in quite good agreement with our own results on $YBa_2Cu_3O_6$ for which X-ray measurements give on powder c = 11.835 ± 0.005 Å and on single crystal c = 11.837 ± 0.001 Å, and on powder samples with x = 0.38 and 0.66. For the various oxygen contents, the lattice parameter c was measured at room temperature both on the powder sample with X-rays and on the single crystal by measuring accuratly the Bragg angle of the (006) Bragg peak with neutrons. The two determinations agree quite well within the experimental accuracy of ±0.005 Å. We found for three samples : 11.812 Å (x = 0.2 ± 0.03), 11.78 Å (x = 0.38 ± 0.03) and 11.775 Å (x = 0.40 ± 0.03). The second method of determining the oxygen amount is to measure integrated nuclear intensities which are sensitive to the occupation of the O1 site. With the crystal orientation only (hhl) Bragg peaks could be measured and their intensities do not depend on the oxygen ordering on sites (½00) and (0½0). The structure analysis of $YBa_2Cu_3O_6$ (8) and $YBa_2Cu_3O_{6.4}$ indicates that the position parameters do not change significantly with oxygen content in this range of concentration. Therefore structure factors can be written as $F(x) = F_0+(1)^h x b_0$ where F_0 correspond to $x = 0$ ($YBa_2Cu_3O_6$) and b_0 is the oxygen scattering length ($b_0 = 0.58.10^{-12}$cm). Some Bragg peaks with weak structure factors, such as (112) or (002), are very sensitive to the oxygen content and a comparison with less sensitive peaks such as (110), yields a rather accurate determination of x (0.28 ± 0.02, 0.38 ± 0.05, 0.44 ± 0.05) in agreement with c-parameter measurements.

So we can conclude that the crystal was successively prepared with the following oxygen amounts x = 0 ± 0.01, 0.25 ± 0.03, 0.41 ± 0.05 and 0.38 ± 0.05. X-ray patterns from the test powders with the same oxygen content show that the crystal structure is tetragonal for x = 0 and 0.25, whereas for the last two concentrations an orthorhombic distorsion is observed (for x = 0.38 the distorsion is quite small and only a line broadening is observed). The variation of the orthorhombic distorsion (b-a)/b, reported in Fig. 5, indicates that the structure remains tetragonal up to x = 0.30 ± 0.05.

For the latter two concentrations the error bars correspond to the absolute values, actually their relative values are known with a better accuracy and they represent really two distinct concentrations.

MAGNETIC ORDERING IN $YBa_2Cu_3O_6$

The single crystal measurements performed at room temperature have confirmed that $YBa_2Cu_3O_6$ is ordered with an antiferromagnetic structure described by the wave vector $\vec{k} = (\frac{1}{2}, \frac{1}{2}, 0)$, i.e. with a unit cell $(a\sqrt{2}, a\sqrt{2}, c)$. The magnetic intensities, measured without any $\lambda/2$ contamination, are given in table 1. The absence of intensity at $\vec{Q} = (\frac{1}{2}, \frac{1}{2}, 0)$ is accuratly established which confirms : i) the AF coupling between magnetic moments of Cu^{2+} ions in the two Cu(2) Bravais sublattices at (00z) and $(00\bar{z})$, ii) the non-magnetic nature of Cu^{+} ions in Cu(1) site. For such a simple AF structure, shown in Fig. 1, the magnetic intensity can be written as : $I(\vec{H}+\vec{k}) = (0.27 . m f(\Theta)\ 2 \sin^2 \pi L z)^2 \langle \sin^2 \alpha \rangle$ where the parameters have their usual meaning and the average is done over the different moment directions (α) with respect to the scattering vector. The measured intensities (see table 1) establishe without any ambiguity that the AF-direction lies within the basal plane and not along the c-axis as was first proposed from less accurate powder data (11).

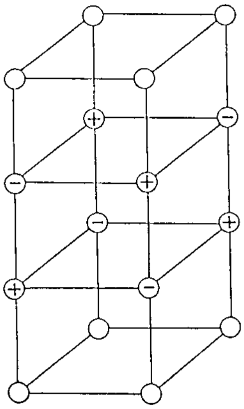

FIGURE 1
Schematic representation of the AF-structure of $YBa_2Cu_3O_{6+x}$ ($x < 0.4$) ; only Cu^{2+} ions in $Cu-O_2$ planes carry a magnetic moment

The variation of the ($\frac{1}{2}$ $\frac{1}{2}$ 1) intensity, reported in Fig. 2, give an ordering temperature $T_N = 420 \pm 10$ K. For this measurement the crystal was mounted inside an evacuated quartz tube to prevent any absorption of oxygen which was confirmed by an accurate X-ray measurement of the c-parameter after the experiment. Additional measurements on a powder sample prepared in the same condition gave the same value of T_N (see Fig. 2). So the actual ordering temperature has a somewhat smaller value than 500 K, a value which was extrapolated from room temperature measurements (11). The low temperature value of the ordered moment is $m_0 = 0.61 \pm 0.05\ \mu_B$.

TABLE 1
Observed and calculated magnetic intensities (10^{-3} barn/formula unit) of $YBa_2Cu_3O_{6+x}$ for an AF-direction within the basal plane

	$YBa_2Cu_3O_6$		$YBa_2Cu_3O_{6.25}$		$YBa_2Cu_3O_{6.38}$	
h k l	I_{obs}	I_{cal}	I_{obs}	I_{cal}	I_{obs}	I_{cal}
½ ½ 0	0	0	0	0	0	0
½ ½ 1	22±2	18	14±2	12	5±1	5
½ ½ 2	32±3	35	20±3	23	11±1	10
½ ½ 3	9±2	9	5±2	5	3±1	2.6
½ ½ 4	4±2	4	3±2	2	-	-
T(K)	300		100		10	
$m(\mu_B)$	0.54±0.05		0.42±0.05		0.28±0.05	

MAGNETIC ORDERING IN $YBa_2Cu_3O_{6+x}$

Crystals with x = 0.25 and 0.38 have been investigated. The magnetic ordering ($\vec{k} = (\frac{1}{2},\frac{1}{2},0)$) and AF-coupling between the two Cu2 sublattices) is found to remain the same as in $YBa_2Cu_3O_6$ but to occur at lower temperatures : $T_N = 290 \pm 10$ K for x = 0.25 and $T_N = 245 \pm 10$ K for x = 0.38 (see Fig. 2). For x = 0.25 an

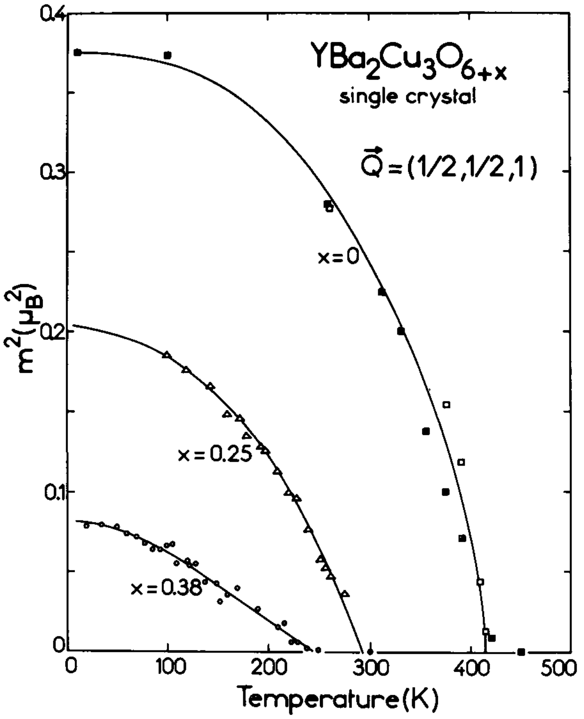

FIGURE 2
Normalized intensities (m^2) of the magnetic Bragg peak ($\frac{1}{2}\frac{1}{2}1$) as a function of temperature for $YBa_2Cu_3O_{6+x}$ compounds

accurate measurement (see Fig. 3) yields the conclusion that the (½½0) peak has still a non detectable intensity. As only the Cu1 site contributes to this peak and taking into account ofthe experimental accuracy we can conclude that at low temperature the ordered moment of copper in Cu1 site is smaller than 0.05 μ_B. A result rather surprising because we can naively expect that a proportion 2x (50 %) of copper ions in Cu1 site are in a Cu^{2+} magnetic state instead of Cu^{+} ($3d^{10}$ non magnetic) as in $YBa_2Cu_3O_6$.

In the Cu2 site the Cu^{2+} ordered magnetic moment is depressed when the oxygen content increases (see Fig. 2 and Fig. 4) : $m_0 = 0.45 \pm 0.05\ \mu_B$ and $0.28 \pm 0.05\ \mu_B$ for x = 0.25 and 0.38, respectively at low temperatures. For x = 0.41, no long range ordering could be observed down to 2K indicating that the antiferromagnetic ordering is abruptly destroyed within narrow range of oxygen concentration around x ≈ 0.40.

Our results are summarized in Fig. 4. The structural transition from the tetragonal to the orthorhombic symmetry appears to take place for an oxygen concentration slightly smaller (x ≈ 0.38) than the magnetic - non magnetic transition(x ≈ 0.40). The linear variation of T_N below x ≃ 0.2 is not well established. From a powder experiment, Tranquada et al. (10) have reported T_N = 400 K for x = 0.15. This would indicate that T_N varies very slowly at low oxygen concentration, and more rapidly for x > 0.2.

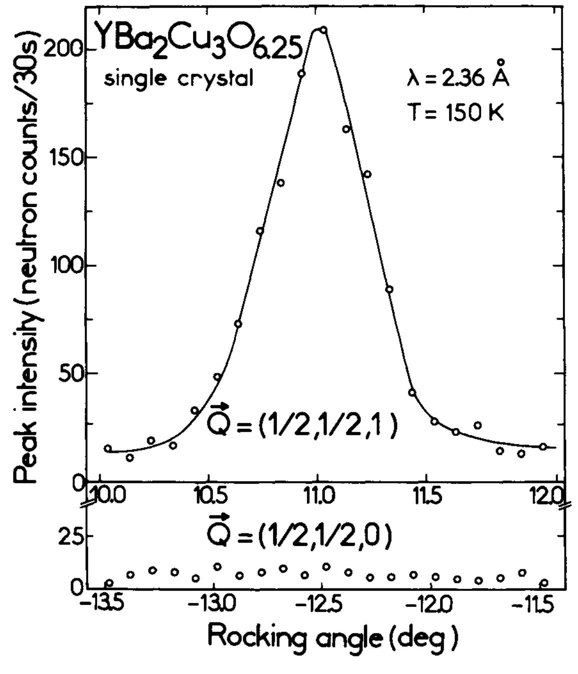

FIGURE 3
Intensities of the (½½0) and (½½1) peaks as a function of the crystal angle for $YBa_2Cu_3O_{6.25}$

It is worth noting that the staggered magnetization (Fig.2) decreases more rapidly, as a function of temperature, when the oxygen content approaches the magnetic-nonmagnetic transition. Actually for a concentration close to the border line the magnetic intensity varies almost linearly with T over a wide range of temperature, a behaviour quite similar to that found in La_2CuO_4 (16). This result is more clearly demonstrated in Fig. 5 which shows the temperature dependence of the moment value in reduced units. Thus, when approaching the magnetic instability there is not only a reduction of the ordered moment but also an increase of magnetic fluctuations. So it would be quite interesting to investigate magnetic excitations and correlations. In $YBa_2Cu_3O_{6.38}$ we performed some Q-scans around (½,½,2) both along and perpendicular to the c-axis (Fig. 6). Above T_N (T = 280 K) we were not able to detect any important two-dimensional

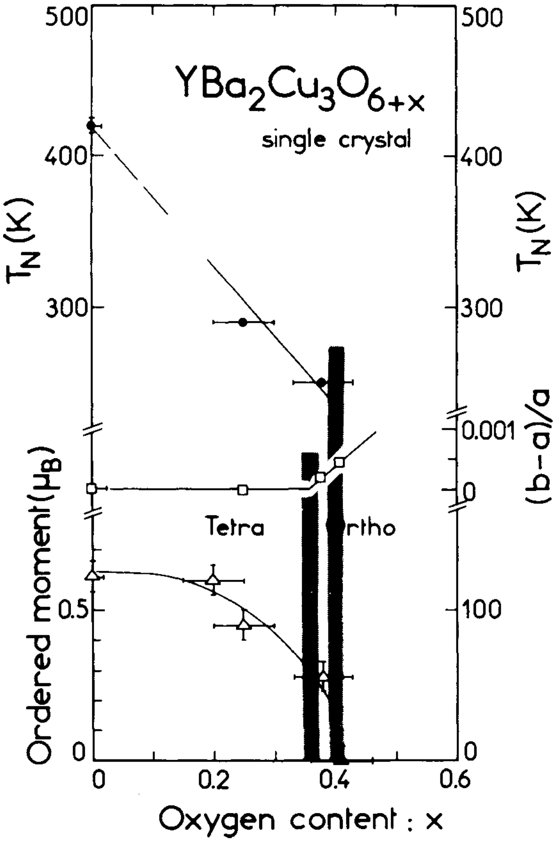

FIGURE 4
Variation as a function of the oxygen content of T_N, the ordered moment value and the orthorhombic distorsion in $YBa_2Cu_3O_{6+x}$

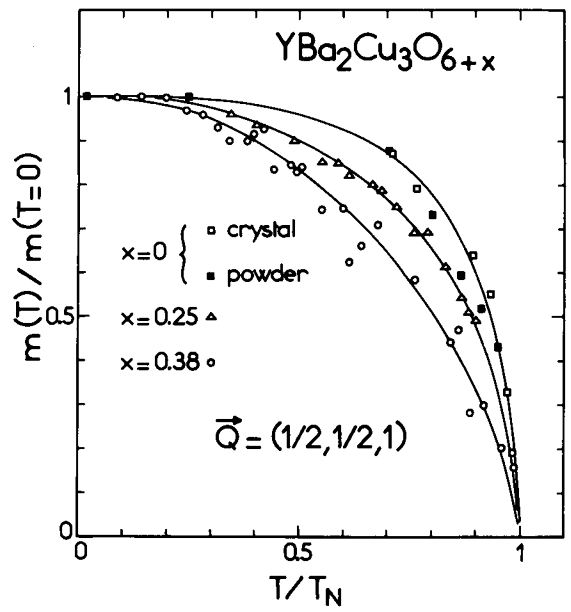

FIGURE 5
Temperature dependence of the ordered magnetic moment in reduced units for three oxygen contents in $YBa_2Cu_3O_{6+x}$

correlations. However in the AF state a diffuse magnetic contribution can be observed in addition to the Bragg peak, and disappears above T_N. These preliminary results require further work on larger crystals.

In classical quasi 2D-Heisenberg antiferromagnets the 3D magnetic ordering occurs when the in-plane correlation length reaches a critical value which depends on J'or on a weak anisotropy. The observed decrease of the diffuse magnetic scattering when approaching T_N indicates that $YBa_2Cu_2O_{6+x}$ does not exhibit this classical behaviour. More likely the phase transition is driven by large in-plane fluctuations.

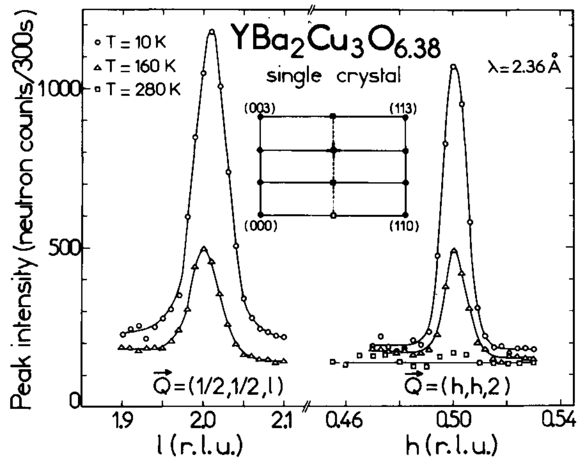

FIGURE 6
Q-scans performed at zero energy transfer and different temperatures around $\vec{Q}$ = (½½2) for $YBa_2Cu_3O_{6.38}$

DISCUSSION

The first striking feature is the high value of the ordering temperature (T_N = 420 K) found in $YBa_2Cu_3O_6$ although the crystals structure and therefore the magnetic couplings exhibit a pronounced two-dimensional character. This T_N value is much larger than the one found in La_2CuO_4 (5) ($T_N \simeq$ 230 K) despite the fact that both crystal structures contain the same CuO_2 quadratic layers with nearly the same Cu-O distance in La_2CuO_4 and $YBa_2Cu_3O_6$ (1.90 Å and 1.96 Å, respectively). Therefore it is very unlikely that this factor two in T_N values can arise from a difference in the in-plane exchange interaction J. The reason has to be found in a larger interplanar coupling J' in $YBa_2Cu_3O_6$ than in La_2CuO_4 while in both compounds magnetic couplings have a quasi two-dimensional character (J' << J). In $YBa_2Cu_3O_6$ the coupling between planes involves two types of exchange integrals. First a direct coupling J_p between nearest neighbouring Cu^{2+} planes, distant of 3.3 Å, defining a double-layer with a weak antiferromagnetic coupling between layers. The coupling J' between double-layers, distant of 8.5 Å, takes place via a linear superexchange path Cu^{2+}-O^{2-}-Cu^{+}-O^{2-}-Cu^{2+}. There are two reasons why the AF exchange integral J' is expected to be larger in $YBa_2Cu_3O_6$ than in La_2CuO_4 ; first, in the latter compound, the exchange paths are more complex, and second, due to the lattice translation (½½0), a cancellation of the interplanar coupling occurs for such an AF in-plane structure. In fact a change of J'/J from 10^{-5}-10^{-4} to 10^{-3}-10^{-2} can well explain the increase by a factor two of T_N from La_2CuO_4 to $YBa_2Cu_3O_6$ because in such compounds the Cu^{2+} ground state exhibits a very weak single ion anisotropy as shown by single crystal susceptibility measurements (17). In contrast to Ising or XY-systems, for a quasi-two dimensional Heisenberg system, the ordering temperature depends more sensitively on J' according to the approximate formula :

$$T_N^{3d} = 8\pi S^2/|\mathrm{Ln}(J'/J)|$$

i.e. for S = ½ : $T_N^{3d}/T_N^{MF} = \pi/|\mathrm{Ln}(J'/J)|$

where T_N^{MF} = 2J is the mean field ordering temperature.

The experimental values are consistent with the above J'/J values and with J ≃ 400 K ($T_N^{MF} \simeq$ 800 K).

This large value can explain the low and nearly constant value for the susceptibility of $YBa_2Cu_3O_{6+x}$ below room temperature (9). $YBa_2Cu_3O_6$ being an insulating compound (9) the magnetic susceptibility is expected to be similar to that of other quasi-2D Heisenberg Cu^{2+} systems (18) which exhibit a broad peak at $T_{max} \simeq T_N^{MF}$ and a constant value $X_0 \simeq X(T_{max})/2$ below $T < T_{max}/3$.$X(T_{max})$ can be estimated from a mean field calculation :

$$X(T_{max}) = (N\mu_B{}^2/3k)g^2S(S+1)/2T_N^{MF}$$

For $S = \frac{1}{2}$ this leads to $X_0 \simeq 1/20$ J (in e.m.u.). So the large value J = 400 K accounts quite well for the small observed susceptibility $X_0 \simeq 10^{-4}$ e.m.u./Cu^{2+}. Since in superconducting samples ($x \simeq 0.9$) the susceptibility is almost the same, it can be proposed that the susceptibility is not of Pauli type but arises from some AF pairing.

In $YBa_2Cu_3O_{6+x}$ it is also puzzling to observe a reduction of T_N for higher x. The addition of oxygen in Cu-O planes is expected to increase the number of Cu^{2+} ions and then to reinforce the interplanar coupling, which is not observed.

Furthermore no detectable ordered magnetic moment was found in the Cu1 site. It seems that Cu^{2+} moments in the Cu1 site remain paramagnetic, which could be a consequence of the linear chain structure.

Another feature is the abrupt magnetic-non magnetic transition occuring around $x = 0.40$. This behaviour can be understood from resistivity data (19) which show a semiconducting behaviour for $x < 0.4$ and the occurence of superconductivity around $x = 0.45 \pm 0.05$.Therefore the magnetic-non magnetic transition in $YBa_2Cu_3O_{6+x}$ is associated, as in (La-Ba or $Sr)_2CuO_4$ compounds, with an insulating-metallic transition. This transition can be related to the appearance of some holes in O(2p) orbitals within CuO_2 planes. This would indicate that up to $x = 0.4$ only the valence state of copper is modified within the Cu-O plane ($Cu^+ \rightarrow Cu^{2+}$) and holes in CuO_2 planes are created only for $x > 0.4$. Then $YBa_2Cu_3O_{6+x}$ compounds would behave quite similarly to $(La-Sr)_2CuO_4$ materials with an abrupt AF-superconducting transition. The question arises as to why T_c is larger in $YBa_2Cu_3O_{6+x}$ than in $(La-Sr)_2CuO_4$. A partial answer could be that the crystalline structure allows larger amounts of holes or a larger interlayer coupling. This last speculation is supported by the fact (may be accidental) that the AF ordering temperatures of $YBa_2Cu_3O_6$ and La_2CuO_4 (420 K and 230 K, respectively) scale with the superconducting transition temperatures in $YBa_2Cu_3O_{6.92}$ and $La_{1.85}Sr_{0.15}CuO_4$ (90 K and 40 K,respectively).

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