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Neutron diffraction study of the magnetic ordering of the Cu^{++} spins in $Nd_{1.5}Ba_{1.5}Cu_3O_{6+x}$

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Elastic neutron scattering experiments performed on single crystals of $Nd_{1.5}Ba_{1.5}Cu_3O_{6+y}$ reveal successive antiferromagnetic (AF) ordering of the Cu^{++} spins. The as grown single crystals show an AF structure characterized by a Neél temperature $T_{N1} \sim 390K$ and a magnetic wave vector (1/2 1/2 0) referring to the tetragonal structure of $NdBa_2Cu_3O_6$. As the temperature is lowered below $T_{N2} \sim 150K$, a spin reorientation develops and a second AF ordering with (1/2 1/2 1/2) wave vector is stabilized. When the samples are oxygenated the tetragonal symmetry and the Neél temperature T_{N1} remain unchanged, whereas the spin reorientation at T_{N2} is suppressed. The results indicate that the Nd/Ba substitution increases the stability of the tetragonal structure upon the oxygen content. This may induce new possibilities of local oxygen ordering that favour the presence of holes in the deficient layer.

Introduction

It is well known[1-4] that two types of antiferromagnetic ordering of Cu spins have been observed in single crystals of $YBa_2Cu_3O_{6+y}$ and $NdBa_2Cu_3O_{6+y}$. The first AF transition at T_{N1} is characterised by a wave vector (1/2 1/2 0) referring to the tetragonal chemical cell P4/mmm made of fully oxygenated bilayers of CuO_2 planes separated by oxygen deficient layers also called 'chain' layers. This first transition involves ordered Cu spins in the CuO_2 bilayers and disordered Cu spins in the 'chain' layers. The second AF transition at $T_{N2} < T_{N1}$, is characterized by a magnetic wave vector (1/2 1/2 1/2). It corresponds to an ordering of the Cu spins in the oxygen deficient layer and to a reorientation of the Cu spins in the bilayers. It is currently considered that as y is increased, the oxygen goes randomly into the deficient layer where the Cu^+ are gradually transformed into Cu^{++} leading to a larger average moment per 'chain' Cu and to a stronger coupling. One thus would expect to see an enhancement of the ordering temperature T_{N2} with increasing y. In fact recent studies[3] on $NdBa_2Cu_3O_{6+y}$ have shown that both T_{N1} and T_{N2} are continuously reduced as y is increased but further work[5] on the Co substitution with the Cu chain has revealed an important enhancement of the chain ordering temperature. Moreover samples of $NdBa_2Cu_3O_{6+y}$ have been reported[4] to have the chain and the plane ordering at the same temperature as high as 380 K. This behaviour was suggested[3] to result from possible Nd/Ba substitution which is known[6] to reduce the superconductivity. In this work we present a neutron study of the Nd/Ba substitution effects on the magnetic ordering of the Cu spins in the deficient layer.

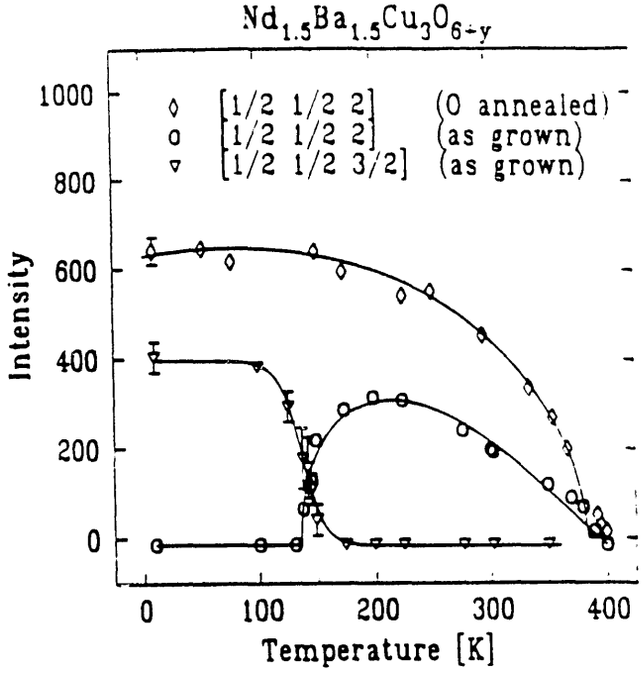
Experiments

The neutron scattering experiments were carried out at Saclay's Orphee Reactor. The nuclear structures were determined on the four-circle diffractometer 5C2 using neutron wavelength of 0.833 Å. The magnetic measurments were performed on the triple-axis spectrometers 1T and 4F2 using 13.6 meV incident neuton energy. Pyrolytic graphite PG(002) was used as monochromator and analyzer with appropriate collimations and PG filters. The spectrometers were equiped with a displex refregirator or a vaccum furnace for low or high temperature studies. The single crystals were grown using the appropriate starting composition of $BaCO_3$, Nd_2O_3 and CuO . The mixture was heated up to about 1200°C in a Pt crucible. In the present work we focus on the particular stable composition[6] 336 i.e $Nd_{1.5}Ba_{1.5}Cu_3O_{6+y}$. We have used two singles crystals from the same batch. Sample A of about $0.6 \times 1.2 \times 2.4$ mm^3 has been annealed in oxygen at 550°C for 40 hours while sample B was used as grown.

Results and discussions

To characterize the Nd/Ba substitution and the oxygen content of our samples, we have determined carefully the nuclear structures and established the chemical compositions. In the refinement of the structural parameters the Nd site was found fully occupied, while the occupation of the Ba site was found to be larger than one. We have also found a high occupancy of the O(1) site in comparison with the largest occupancy in the tetragonal phase of YBCO. These results clearly indicate that regarding the oxygen content, the tetragonal phase has a larger stability in $Nd_{1.5}Ba_{1.5}Cu_3O_{6+y}$ than it has in YBCO. The tendency to form Cu-O chains at large y is reduced by the Nd/Ba substitution. From these measurements we have established with good reliability factors $R = 0.052$ and $R_w = 0.058$, the compositions for crystal A as $Nd(Ba_{1.46}Nd_{.54})Cu_3O_{6.88}$ and for crystal B as $Nd(Ba_{1.50}Nd_{.50})Cu_3O_{6.39}$. Regarding the Nd/Ba substitution, both crystals are found very close to the nominal composition 336, however they have quite different oxygen content as expected from the heat treatment.

We have searched for magnetic Bragg peaks of the kind (1/2 1/2 l) with both integer and half integer l. We have found that both samples show a long range antiferromagnetic order. However the oxygenated sample A exhibits integer l magnetic Bragg peak only below the Neel temperature at about 390 K and down to 9 K, whereas the as grown crystal B show integral l peaks at high temperature and half integral l peaks at low temperature. A continuous transfer of the magnetic intensity from integral to half integral l peaks occurs around $150K$. In the figure we show the temperature variation of the integrated intensity of the magnetic Bragg peak (1/2 1/2 2) for the oxygenated sample A and for the as grown sample B we report on the same figure the variation of integrated intensity of both (1/2 1/2 2) and (1/2 1/2 3/2) magnetic peaks. At 9K we have collected a list of magnetic reflections for the two samples and solve the magnetic structures. For sample A, these magnetic intensities are readily accounted for with collinear spins as in YBCO. The resulting moment is found on the Cu(2) sites only and is of the order of $0.55 \pm 0.05\mu_B$ with the spin lying in the CuO_2 planes. For sample B the magnetic structure is found to be similar to that reported in reference 4 with an average moment per Cu plane of the order of $0.6 \pm 0.05\mu_B$ and an average moment per Cu chain of the order of $0.09 \pm 0.05\mu_B$.



Temperature variation of the integrated intensity of the magnetic Bragg reflections in the as grown and O_2-annealed $Nd_{1.5}Ba_{1.5}Cu_3O_{6+y}$.

We have no evidence of an ordering of the rare earth moment Nd^{3+}, which could influence the Cu spin reorientation as reported[7] for Nd_2CuO_4. Clearly these results, along with the fact that crystalline structure remains unchanged under oxygenation, indicate that the mechanism associated with the drastic reduction of the Cu spin reorientation must be found in the partition of the charge carriers between the plane and the chain layers. We suggest that the enlarged stability of the tetragonal structure, for some local O_2 ordering, favours the presence of holes in the deficient layer. These holes would reduce the average moment per Cu chain and then the stability of the second AF ordering.

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