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ANTIFERROMAGNETIC ORDER OF THE Cu IN $\mathcal{R}\text{Ba}_2\text{Cu}_3\text{O}_{6+x}$

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Abstract. – Neutron diffraction measurements have been taken on single crystals of $\mathcal{R}\text{Ba}_2\text{Cu}_3\text{O}_{6+x}$ to explore the nature of the magnetic order as a function of oxygen concentration x . The Cu-O planes order antiferromagnetically at T_{N1} , with T_{N1} ($x \approx 0$) ~ 450 K. At lower temperatures the oxygen-deficient Cu planes also order magnetically, with a substantial low temperature moment ($\sim 1/2 \mu_B$) and a simple antiferromagnetic sequencing along the c -axis. Both ordering temperatures decrease with increasing x , with T_{N2} being much more sensitive to x than T_{N1} .

The magnetic properties of the oxide superconductors have been of particular interest due to the intrinsic interest in magnetism as well as because of the possibility that the magnetic fluctuations are responsible for the superconducting pairing [1]. The $\mathcal{R}\text{Ba}_2\text{Cu}_3\text{O}_{6+x}$ (\mathcal{R} = rare earth) system orders antiferromagnetically [2-8] for small x ($x \lesssim 0.4$), where the system is tetragonal and nonsuperconducting. The magnetic structure consists [3, 4] of a simple antiparallel arrangement of Cu spins both within the Cu-O planes as well as along the tetragonal c -axis, while the oxygen deficient Cu planes possess no ordered moment. The three-dimensional ordering can be quite high (T_{N1} ($x \approx 0$) ~ 450 K), indicating that the magnetic interaction energies are large. The 3-d transition temperature is also very sensitive [2] to the oxygen concentration x , decreasing rapidly with increasing x and approaching zero for $x \sim 0.4$.

A second magnetic phase transition has recently been observed [5, 6], in which the oxygen-deficient Cu "chain" layers also order magnetically. We have been investigating the nature of this ordering and the effect of oxygen concentration on T_{N2} and have now obtained results on a number of single crystal specimens of $\text{NdBa}_2\text{Cu}_3\text{O}_{6+x}$. We find that at small x and low T the basic magnetic structure consists of a simple antiferromagnetic arrangement of spins both within the planes as well as along the c -axis. In particular, we find an unexpectedly large magnetic moment on the Cu chain sites. At intermediate temperatures the spin arrangement is a superposition of the high T and low T configurations, and is noncollinear in nature. We also find that both transition temperatures decrease with increasing x , with the lower transition T_{N2} being much more sensitive to x than the higher transition T_{N1} .

The neutron measurements were carried out at the National Bureau of Standards Research Reactor using standard triple-axis instruments. Unpolarized diffraction data were collected with an incident energy of either 14.8 or 13.7 meV using a pyrolytic graphite

PG(002) monochromator. Polarized neutron measurements were then taken to establish that the new peaks are magnetic in origin, and this procedure has been described previously [4]. The Bragg peak corresponding to the $\left(\frac{1}{2} \frac{1}{2} \frac{3}{2}\right)$ reflection is shown in figure 1. Even though the Cu moments are quite small, the crystal is sufficiently large (~ 50 mg) to obtain an excellent signal-to-noise ratio. Polarized neutron measurements were employed to establish that the observed half-integral peaks are wholly magnetic in origin. The half-integral value for the third index demonstrates that the magnetic structure is doubled along all three crystallographic directions in comparison to the chemical unit cell. This contrasts with the spin configuration observed [3, 4] in the higher temperature magnetic phase, where the l index is integral and the magnetic and chemical unit cells are the same size along the tetragonal c -axis.

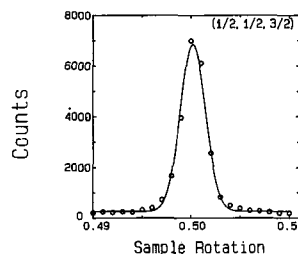


Fig. 1. – Observed magnetic Bragg peak for the $\left(\frac{1}{2} \frac{1}{2} \frac{3}{2}\right)$ peak at low temperatures.

The temperature dependence of the $\left(\frac{1}{2} \frac{1}{2} \frac{3}{2}\right)$ peak is shown in the bottom portion of figure 2. For this value of oxygen concentration ($x \approx 0.1$) the lower phase transition occurs at $T_{N2} \approx 80$ K. The top portion of the figure compares the intensity of the whole-integral peaks such as $\left(\frac{1}{2} \frac{1}{2} 2\right)$, which are associated

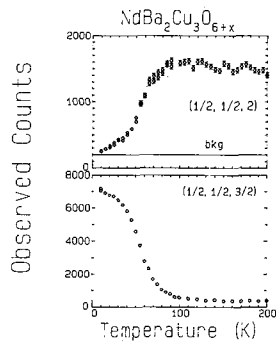


Fig. 2. - Magnetic intensities for the $\text{NdBa}_2\text{Cu}_3\text{O}_{6.1}$ crystal. The downturn of the intensity of the $\left(\frac{1}{2} \frac{1}{2} 2\right)$ peak coincides with the development of half-integral peaks such as the $\left(\frac{1}{2} \frac{1}{2} \frac{3}{2}\right)$ peak shown in the bottom portion of the figure.

with the higher temperature phase transition: For the present sample $T_{N1} \simeq 430$ K. There is a dramatic downturn in the intensity of the whole-integral reflections at low temperatures which is associated with the development of the intensities of the half-integral peaks. It is clear that the half-integral peaks dominant at low T , and hence they correspond to the ground state spin configuration of the system. The $T = 0$ structure consists of a simple collinear arrangement of spins, with the spin direction in the tetragonal plane. The nearest-neighbor spins within the Cu-O planes (designated M_P , for planes) are aligned antiparallel as they are in the high T phase. However, along the c -axis the oxygen-deficient Cu moment (designated M_C , for chains) is nonzero, and aligned antiparallel to the spins in the Cu-O layers. The doubling of the unit cell along the c -axis direction results because the sense of the spins is reversed in going from one chemical unit cell to another.

The values of the ordered moments we obtain at low temperatures are $M_P = 0.97 \pm 0.09 \mu_B$ for the Cu-O planes (assumed equal), and $M_C = 0.46 \pm 0.06 \mu_B$. This compares with $M_P \sim 0.6 \mu_B$ for the maximum moment found [4] above T_{N2} . The essential difference between the two structures is of course the moment M_C . We remark that the conventional viewpoint for the $\text{RBa}_2\text{Cu}_3\text{O}_{6+x}$ class of materials is that the Cu valence in the Cu - O₂ planes is 2 +, with an associated magnetic moment, while the Cu "chain" layers contain Cu¹⁺ which is nonmagnetic. The unexpectedly large ordered moment which we have observed on the Cu "chain" layers demonstrates that the Cu¹⁺ state is not an appropriate description of the electronic configuration of the "chain" layers in these materials. It is reasonable to assume that the chain-layer moment is a result of strong hybridization of the (band) electronic wave functions within this (as well as the Cu-O) Cu

plane, rather than as a result of a perturbation caused by the remnant oxygen in the layer as has been suggested [6]. Further work on the magnetic properties of these systems as a function of concentration, magnetic field, and pressure is underway.

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