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Title
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Permalink
https://escholarship.org/uc/item/5xt8b24m

Journal
Journal of Applied Physics, 67(9)

ISSN
0021-8979

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Publication Date
1990-05-01

DOI
10.1063/1.344859

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Magnetic phase transitions in Nd_2CuO_4

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Polarized and unpolarized neutron scattering techniques along with x-ray diffraction have been used to study the magnetic and structural properties of a single crystal of Nd_2CuO_4. Long-range magnetic order of the Cu moments develops at $T_N = 245$ K, with a simple antiferromagnetic configuration of spins as found in La_2NiO_4, while the spin directions may be either collinear or noncollinear. Additional abrupt transitions are observed at 75 and 30 K, in which spin reorientations take place. Bragg peaks associated with the crystal structure are found at the same positions as the magnetic Bragg peaks, and indicate that a small distortion of the basic tetragonal structure has occurred above 300 K. At low temperatures the Nd moments also order antiferromagnetically ($T_N = 1.5$ K), while an additional transition of a continuous nature is observed at 150 mK.

INTRODUCTION

The magnetic properties of the high-T_c superconducting oxides have attracted considerable interest, both because of the intrinsic interest in magnetism and because of the possibility that the magnetic fluctuations are responsible for the Cooper pairing. It is clear now that the two-dimensional sheets of Cu-O in the $(La_{2-x}Sr_x)CuO_4$ and $\mathscr{R}Ba_2Cu_3O_{6+x}$ ($\mathscr{R}$ = rare earth) systems are intimately involved in both the magnetism and superconductivity, with magnetic long-range order occurring at small x and fluctuating moments existing in the superconducting regime at larger x. The charge carriers are holes in these systems. Superconductivity has been recently discovered[1] in a new class of materials, $\mathscr{R}_{2-x}Ce_xCuO_{4-y}$ ($\mathscr{R}$ = Pr, Nd, or Sm), where trivalent lanthanides are partially substituted by tetravalent lanthanides. Based on chemical valence considerations, XPS data, and Hall effect data, the charge carriers appear to be electrons rather than holes, and the existence of these materials places important constraints on any theory of the oxide superconductors. In this regard it is of central concern to elucidate the nature of the magnetism in this new class of systems. Here we report measurements of the magnetic ordering of the parent material, Nd_2CuO_4.

All of the neutron diffraction measurements were taken on the BT-2 triple-axis spectrometer at the Research Reactor at the National Institute of Standards and Technology. Unpolarized diffraction data were collected using a pyrolytic graphite monochromator and filter, with an incident energy of 14.7 meV. The polarized neutron measurements were performed with a similar incident energy, with a Heusler alloy monochromator and a multilayer polarizing analyzer. Angular collimations before and after the monochromator, and after the sample, were 60′-40′-40′ for the high-temperature data ($T > 4$ K) and 60′-20′-20′ for the low-temperature data. A helium dilution refrigerator was used for the low-temperature measurements down to 80 mK. The sample was a thin platelike single crystal weighing 20 mg.

RESULTS FOR Cu SPIN ORDERING

All the observed magnetic Bragg peaks in this sample can be indexed as $(h/2, k/2, l)$ based on the chemical unit cell, where h and k are odd integers. The $(\frac{1}{2},\frac{1}{2},1)$ and $(\frac{1}{2},\frac{1}{2},2)$

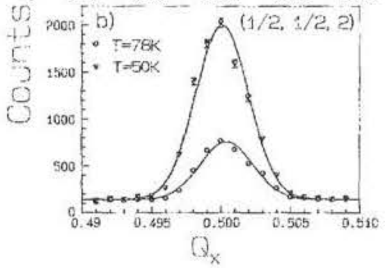

FIG. 1. The $(\frac{1}{2},\frac{1}{2},1)$ and $(\frac{1}{2},\frac{1}{2},2)$ magnetic Bragg peaks above and below the spin reorientation transition at 75 K. The odd-integer peaks decrease in intensity while the even-integer peaks increase in intensity when the transition occurs.

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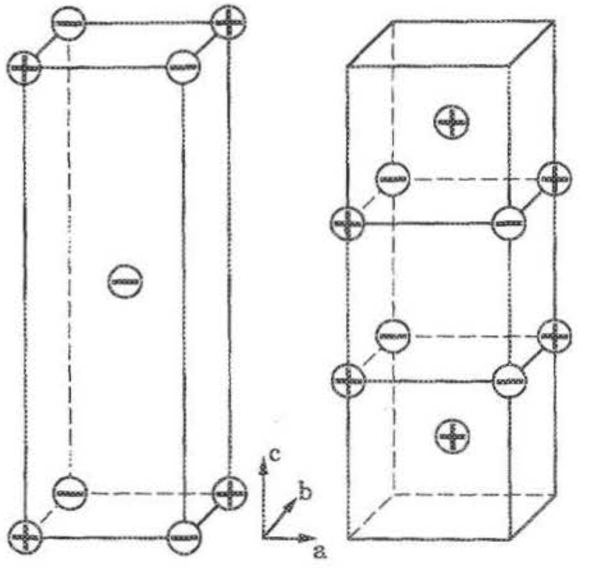

FIG. 2. Magnetic spin configurations for (left) the Cu spins and (right) the Nd spins.

magnetic Bragg reflections at two temperatures are shown in Fig. 1. In general we find that in the higher temperature regime the odd-integer peaks are much stronger in intensity than the even-integer peaks. Since the first two Miller indices are half-integer, the magnetic unit cell is double the chemical unit cell along both the *a* and *b* crystallographic directions. The magnetic spin configuration of the Cu spins is shown on the left-hand side of Fig. 2, and is the same basic structure as found in the other 2-1-4 systems.[2,3]

We observe no intensity for the $(\frac{1}{2},\frac{1}{2},0)$ and $(\frac{3}{2},\frac{1}{2},0)$ reflections in the temperature region above 75 K, which would suggest a La_2NiO_4-type spin structure, with the spin direction along [110] as recently proposed by Endoh *et al.*[5] However, the initial analysis of our polarized beam data suggested modification of the spin directions in which the spins were noncollinear,[4] with one plane of spins being rotated by $\pi/2$ with respect to the next along the *c* axis. The noncollinear structure represents the coherent addition of two domains for the collinear structure, and both structures agree with the data. The same structure has been recently found for the related system Pr_2CuO_4.[6] For both materials, the present data cannot distinguish between the collinear and noncollinear possibilities.

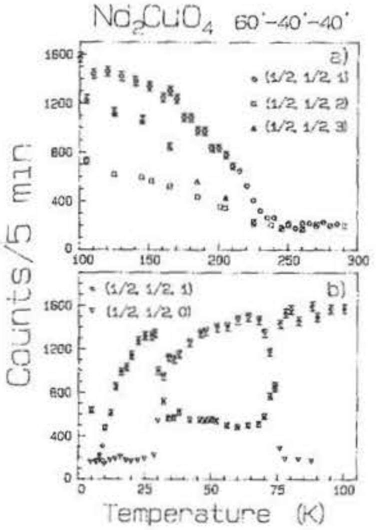

FIG. 3. Temperature dependence of several magnetic Bragg peaks. (a) All the $(\frac{1}{2},\frac{1}{2},l)$ peak intensities increase with decreasing temperature below the Neel temperature of 245 K. (b) shows two spin reorientations, one at 75 K and the other at 30 K.

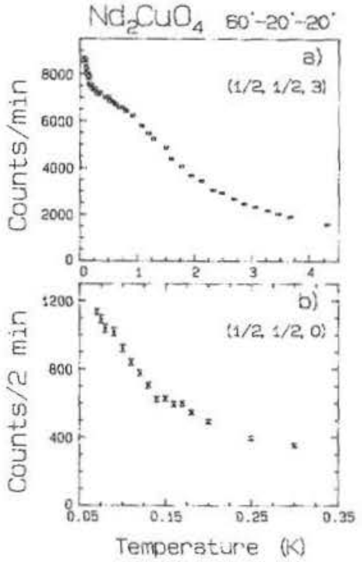

FIG. 4. Temperature dependence of two magnetic Bragg peaks at low temperatures. (a) The $(\frac{1}{2},\frac{1}{2},3)$ peak, which has the strongest intensity when the Nd order. (b) The $(\frac{1}{2},\frac{1}{2},0)$ peak, which shows another transition at 150 mK. This transition is also evident in the $(\frac{1}{2},\frac{1}{2},3)$ reflection (a).

The temperature dependence of several peaks is shown in Fig. 3(a), and reveals a Neel temperature of $T_N = 245$ K for our sample. The temperature dependence of the $(\frac{1}{2},\frac{1}{2},l)$-type peaks shown in Fig. 3(b) reveals an abrupt transition at 75 K. At this transition we find that the odd-integer peaks drop in intensity while the even-integer peaks jump in intensity, so that the even peaks are now strong and the odd peaks weak. We also observe a reversal in the polarized beam spin-flip scattering ratios: Above 75 K the odd-integer peaks have a large flipping ratio and the even-integer peaks have a flipping ratio of unity, while below 75 K the situation is reversed. Also note that the $(\frac{1}{2},\frac{1}{2},0)$ peak now has a measurable intensity. These data indicate that a spin reorientation has occurred, in which the spins rotate by $\pi/2$. We obtain an ordered moment of 0.37 μ_B at 78 K, and 0.44 μ_B at 50 K. Figure 3(b) also shows that another spin reorientation transition occurs at 30 K, where the spins rotate back to the original direction. The relative intensities and spin-flip ratios taken below this second spin reorientation transition are similar to those of the high-temperature phase (75 K $< T <$ 245 K). These spin reorientation transitions are not observed in the Pr_2CuO_4 system.[6]

Nd SPIN ORDERING

At temperatures below the second spin reorientation transition (<30 K), the intensities of the magnetic Bragg peaks continue to evolve with temperature, as shown in Figs. 3 and 4. These changes are related at least in part to the Nd moments, where the Nd sublattice is polarized by interaction with the ordered Cu moments.[7] The temperature dependence of the $(\frac{1}{2},\frac{1}{2},3)$ and $(\frac{1}{2},\frac{1}{2},0)$ peaks at low temperatures is shown in Fig. 4; the $(\frac{1}{2},\frac{1}{2},3)$ peak has by far the strongest intensity when the Nd sublattice orders. Figure 4(a) suggests that the Nd spins order at about ~ 1.5 K as indicated by specific heat,[8] but the transition appears "smeared" due to the coupling of the Nd and Cu sublattices; no such smearing is observed[7] in superconducting $Nd_{1.85}Ce_{0.15}CuO_4$, where the Cu sublattice is magnetically disordered.

Our measurements suggest a simple antiferromagnetic structure for the Nd spins as shown on the right-hand side of Fig. 2. The Nd magnetic unit cell is double the chemical unit cell along both the *a* and *b* directions as it is for the Cu. We also note that another transition occurs at about 150 mK, which is most easily observed in the $(\frac{1}{2},\frac{1}{2},0)$ peak as shown in Fig. 4(b). This transition is also evident in the temperature dependence of the $(\frac{1}{2},\frac{1}{2},3)$ peak [Fig. 4(a)]. The nature of this additional ordering is unknown at present, but it could originate from nuclear spin ordering. Thus, altogether we find five distinct magnetic phase transitions in this system.

STRUCTURAL DISTORTION

Above the Neel temperature of 245 K, we found that there are small Bragg peaks still observed at positions such as $(\frac{1}{2},\frac{1}{2},2)$ and $(\frac{1}{2},\frac{1}{2},3)$. These peaks have been observed with both neutrons and x rays, and correspond to a zone-boundary charge-density wave-type distortion of the system. The overall distortion must be quite small since the intensities of these peaks are small, and considerable additional data will be needed to quantify the nature of the distortion. It is essential, of course, to understand the structural distortion before a complete understanding of the origin of all the magnetic phase transitions can be achieved.

ACKNOWLEDGMENTS

The research at the University of Maryland was supported by the NSF, Contract No. DMR 86-20269 and DMR 89-21878. The research at Los Alamos was supported by the Department of Energy.

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