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Pressure dependence of the Cu magnetic order in R\(\text{Ba}_2\text{Cu}_3\text{O}_{6+x}\)

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Neutron-diffraction measurements have been carried out as a function of hydrostatic pressure to study the magnetic order of the Cu spins in \(\text{NdBa}_2\text{Cu}_3\text{O}_{6.35}\) and \(\text{NdBa}_2\text{Cu}_3\text{O}_{6.5}\). In the high-temperature phase, where the Cu planes order antiferromagnetically, we find that the \(T_N\) temperature is very strongly dependent on pressure, increasing at the rate of \(\sim 23\) \(\text{K/kbar}\). We attribute this phenomenon to the two-dimensional-like behavior of this magnetic system.

In the low-temperature phase, which is associated with magnetic ordering of the chains, only a small change in the ordering temperature \(T_{N2}\) is observed.

The magnetic properties of the superconducting oxides have been of particular interest since it was discovered that there is an antiferromagnetic phase at small \(x\) for both the \(L_2-x\text{Sr}_{x}\text{CuO}_4\) (Ref. 1) and \(R\text{Ba}_2\text{Cu}_3\text{O}_{6+x}\) (Refs. 2 and 3) systems, which is in close proximity to the superconducting regime of the phase diagram at larger \(x\). 

The energy scale for magnetic fluctuations is an order of magnitude larger than for the phonons, and these fluctuations persist into the superconducting phase. 

We have been studying the magnetic ordering of the Cu spins in the semiconducting phase of \(\text{NdBa}_2\text{Cu}_3\text{O}_{6+x}\), and have found that the paramagnetic-antiferromagnetic ordering temperature is extraordinarily sensitive to pressure.

The crystals which we have studied have the compositions \(\text{NdBa}_2\text{Cu}_3\text{O}_{6.1}\) and \(\text{NdBa}_2\text{Cu}_3\text{O}_{6.35}\), and weighed 50 and 9 mg, respectively. These samples have been investigated thoroughly in the absence of applied pressure, and we summarize the magnetic behavior as follows. There are three Cu layers per chemical unit cell as indicated in Fig. 1. Two layers are fully oxygenated (the Cu-O_2 plane layers), while in the third layer (the chain layer) the oxygen content \(x\) can be varied from zero to one, which affects the magnetic and superconducting properties as shown in Fig. 1. On cooling from the high-temperature paramagnetic state, the Cu moments in the Cu-O_2 plane layers order antiferromagnetically \((TN)\) at small \(x\), with a simple spin configuration (inset to Fig. 1). Nearest-neighbor spins within a Cu layer are aligned antiparallel, and the spin direction is that of the \(a-b\) plane. This structure gives rise to magnetic Bragg peaks of the type \((h/2, k/2, l/2)\), and a sketch of the temperature dependence of the intensity of these peaks is also indicated in the figure (solid curve). At lower temperatures the intensity is seen to decrease rapidly toward zero. This decrease is associated with ordering of the Cu chain-layer spins, and gives rise to new magnetic Bragg peaks of the type \((h/2, k/2, l/2)\) below \(T_{N2}\) (dotted curve). We remark that the antiferromagnetic plane ordering \((T_N)\) and the superconducting phase boundaries as a function of \(x\) are well established. 

The region where the chain ordering has been observed is also shown, although it should be noted that this ordering is not controlled simply by the oxygen content \(x\). For the present samples we have \(T_{N1} = 430\) K and \(T_{N2} = 80\) K for the \(\text{NdBa}_2\text{Cu}_3\text{O}_{6.1}\) crystal, and \(T_{N1} = 230\) K and \(T_{N2} = 10\) K for the \(\text{NdBa}_2\text{Cu}_3\text{O}_{6.35}\) crystal.

The neutron scattering measurements were carried out at the National Institute of Standards and Technology (formerly National Bureau of Standards) research reac-

![FIG. 1. Shown on the left side is a schematic of the phase diagram as a function of oxygen concentration for \(R\text{Ba}_2\text{Cu}_3\text{O}_{6+x}\), with the superconducting (S) and antiferromagnetic (AF) phases indicated. A sketch of the magnetic Bragg intensity for the \((h/2, k/2, l)\)-type antiferromagnetic peaks (solid curve) and the \((h/2, k/2, l/2)\)-type peaks (dotted curve) is shown on the right-hand side. The inset shows the spin configuration in the high-temperature phase. The large circles represent Cu ions in the plane layers, and the small circles represent Cu ions in the chain layer. The spin direction is in the \(a-b\) tetragonal plane.](image-url)
The familiar instrumental setup of pyrolytic graphite monochromator and filter, and a neutron wavelength of 2.346 Å, was employed. The single-crystal samples were placed into an aluminum pressure cell, which was mounted in either a flow-type cryostat, or a furnace. Pressure was applied hydrostatically using helium gas as a medium.

The temperature dependence of the magnetic intensity of the $(\frac{1}{2} \frac{1}{2} 0)$ Bragg peak for the NdBa$_2$Cu$_3$O$_{6.35}$ crystal is shown in Fig. 2 at a series of pressures. At zero pressure the ordering temperature is 230 K, as already noted. With increasing pressure the curves are seen to shift rapidly to higher temperatures, indicating that the ordering temperature $T_{N1}$ is increasing. In this temperature regime the intensity is approximately linearly dependent on temperature as indicated in Fig. 1, and to obtain an estimate of $T_{N1}$ we have simply done a least-squares fit of a straight line to the data (solid curves). 

The transition temperatures which have been obtained from these fits are shown in Fig. 3 as a function of pressure. Up to 4 kbar $T_{N1}$ increases approximately linearly with pressure at the rate of $(23 \pm 3)$ K/kbar. This rate of increase is more than 2 orders of magnitude higher than the rate of 0.05 K/kbar observed for the superconducting transition temperature $T_C$ for YBa$_2$Cu$_3$O$_7$. The behavior for the La$_2$CuO$_4$ system, on the other hand, is just the opposite of RBa$_2$Cu$_3$O$_{6+x}$. The Néel temperature has been found to be approximately independent of pressure up to $\sim 5$ kbar, and then to decrease slowly with a further increase of $P$, while the superconducting transition temperature shows a relatively strong increase with pressure.

Figure 4 shows some of the results obtained on the NdBa$_2$Cu$_3$O$_{6.1}$ sample. The top portion gives the pressure dependence of the intensity of the $(\frac{1}{2} \frac{1}{2} 0)$ peak at a fixed temperature of 260 K. The strength of the scattering rapidly increases at low pressure, and then saturates at $\sim 2.5$ kbar. We believe that this saturation effect only signifies that the order parameter has achieved its full value, and not that $T_{N1}$ is no longer shifting with pressure. Unfortunately, this cannot be checked directly, as the pressure cell failed at higher temperatures, and the sample
was lost. We can, however, make an estimate of the initial 
\( dT_{N1}/dP \) by assuming that the initial increase in intensity 
corresponds to a shift in the order-parameter curve analogous 
to the shift shown in Fig. 2. This yields a value 
\( dT_{N1}/dP \approx 78 \text{ K/kbar} \), a value which is consistent with 
the isobaric data we obtained below 300 K. Even though 
this is a crude estimate, it serves to demonstrate that there 
is a large effect of pressure on \( T_{N1} \) in both samples.

The pressure dependence of the Bragg scattering associated 
with the lower transition \( T_{N2} \), which is where the Cu 
chain spins order, is shown in the bottom portion of Fig. 4. 
There is a small increase in the maximum value of the 
temperature, but there is very little shift in \( T_{N2} \). We believe 
that this weak pressure dependence is representative of the 
fact that when the chain ions order, then the spacing 
between the Cu ions in the \( a, b, \) and \( c \) directions is about 
equal and we have a fully three-dimensional (3D) magnetic 
structure with simple antiferromagnetic nearest-neighbor 
interactions, in contrast to the situation at \( T_{N1} \) as discussed below. At \( T_{N2} \) the magnitude of the pressure 
effect is typical of 3D phase transitions.\(^{15}\)

The Néel temperature \( T_{N1} \) is known to be quite sensitive 
to the oxygen concentration as shown in Fig. 1, so that the 
pressure effect we see might be explained by the removal 
of oxygen from the sample. However, below room 
temperature the oxygen will not reenter the sample, and we 
find no evidence for any irreversible effects in our data. 
Therefore, we discard this as a possibility. We have also 
measured the pressure dependence of the lattice parameters, 
and find a smooth decrease of \( \approx 0.03\%/\text{kbar} \), which 
translates into a compressibility of \( 7.5 \times 10^{-13} \text{ Pa}^{-1} \). We 
don't detect any anomalies or abrupt changes in the lattice over 
the pressure range explored.

The most likely explanation for the strong pressure sensitivity 
of \( T_{N1} \) is in terms of the large magnetic anisotropy and 
competing exchange interactions which are present when the 
Cu chain spins are disordered in the \( \text{RBa}_2\text{Cu}_3\text{O}_6+x \) system. The magnetic exchange interaction 
\( J \) within the \( \text{Cu}_2\text{O}_2 \) layers is very large, and thus in 
the vicinity of \( T_{N1} \) there are very strong magnetic correlations 
within the \( \text{Cu}_2\text{O}_2 \) planes. Hence, we have 2D-like magnetic behavior, with the preferred spin direction in the 
tetragonal plane, and since there is no 2D long-range order\(^{5}\) above \( T_{N1} \), an \( x-y \) model (with algebraic decay of the 
correlations) should be appropriate (recall that an Ising 
model orders in 2D). The 3D phase transition is then 
driven by the weak effective interaction \( J' \) between layers.\(^{16}\)
Since the in-plane exchange \( J \) is already very large, 
it is likely that \( J' \) is near a maximum versus ionic separation 
and thus will not be particularly sensitive to pressure. 
On the other hand, the effective interaction \( J' \), which is 
mediated through the spin-disordered Cu chain layer, results 
from an overlap of wave functions on ions which are 
well separated. The overlap integral should then depend 
exponentially on separation, and a substantial increase in 
\( J' \) with pressure can be expected. If this is indeed the 
case, then \( T_{N1} \) should have a much stronger dependence 
on stress applied along the tetragonal \( c \) axis than for stress 
applied in the \( a-b \) direction, and measurements of this 
type are planned. In addition, there is a calculation\(^{17}\) that 
the spin-wave fluctuations gives \( T_{N1} \approx Jn^{-1}(J/J') \) for large 
\( J/J' \). which would yield a linear dependence of \( T_{N1} \) on \( P \) 
as observed. However, \( T_{N1} \) would be a weak function of 
\( J/J' \) and hence this scenario would then require a large 
change in \( J' \) to explain the data. Such a large change 
could be the result of a significant pressure-induced 
change in the electronic structure, similar to the change in 
\( T_{N1} \) caused by oxygen variation, or it could be due to some 
competing interactions caused by the disordered spins on 
the chain layers.\(^{3}\)

Note that the effective interaction between the plane layers which is mediated through the chain layer is quite different depending on whether or not the chain layer is ordered. Below \( T_{N2} \) the plane layers adjacent to the chains (next nearest neighbors) are aligned ferromagnetically, while above \( T_{N2} \) they are aligned antiferromagnetically. It would be interesting in this regard to determine if such a strong pressure sensitivity is observed in \( \text{RBa}_2\text{Cu}_3\text{O}_6+x \) samples which do not exhibit chain ordering at low temperatures.

A second possibility is that the anisotropy within the 
plane increases with pressure, which would increase the 
in-plane correlations and eventually produce a phase transition 
with long-range order in two dimensions.\(^{18}\) Indeed, 
if there were a tendency for pressure to cause an orthorhombic distortion, for example, then the magnetic behavior 
of the layers would cross over to a 2D Ising system (with long-range order), and \( T_{N1} \) could increase dramatically. However, so far we have found no indication experimentally of an orthorhombic distortion.

Finally, we note that in the case of \( \text{La}_2\text{Cu}_4\text{O}_7 \)-type systems 
the magnetic structure is such that there is an approximate 
cancellation of nearest-neighbor interactions between planes, and therefore the effective interplanar interaction is controlled more by the magnitude of the orthorhombic distortion, rather than \( J' \) itself. In the \( \text{RBa}_2\text{Cu}_3\text{O}_6+x \) system, no such cancellation is present.

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4A review of both theory and experiment which pertain to the oxide superconductors is given in *High Temperature Superconductivity*, edited by J. W. Lynn (Springer-Verlag, New York, 1989).


10Over this limited temperature range, and with the statistical accuracy available, we felt that trying to fit a power law to the data in order to extract the critical exponent β was unjustified. We do not expect β to change significantly with pressure.

11See, for example, T. Kaneko, H. Yoshida, S. Abe, H. Morita, K. Noto, and H. Fujimori, Jpn. J. Appl. Phys. 26, L1374 (1987). According to the theory of M. Cyrot [Solid State Commun. 62, 821 (1987)], we should expect \( dT_N/dP = (\frac{3}{2})CT_N^{1.1} \sim 0.1 \text{ K/kbar}. \)

12R. A. Ferrell (private communication).

13For results and discussion on 2D systems, see, for example, R. J. Birgeneau, H. J. Guggenheim, and G. Shirane, Phys. Rev. B 1, 2211 (1970).