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Pressure dependence of the Cu magnetic order in $RBa_2Cu_3O_{6+x}$

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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 $NdBa_2Cu_3O_{6.35}$ and $NdBa_2Cu_3O_{6.1}$. In the high-temperature phase, where the Cu planes order antiferromagnetically, we find that the Néel temperature T_{N1} is *very strongly* dependent on pressure, increasing at the rate of ~ 23 K/kbar. We attribute this phenomenal sensitivity 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 $La_{2-x}Sr_xCuO_4$ (Ref. 1) and $RBa_2Cu_3O_{6+x}$ (Refs. 2 and 3) systems, which is in close proximity to the superconducting regime of the phase diagram at larger x.[4] The energy scale for the magnetic fluctuations is an order of magnitude larger than for the phonons,[5-9] and these fluctuations persist into the superconducting phase.[6,7,9] We have been studying the magnetic ordering of the Cu spins in the semiconducting phase of $NdBa_2Cu_3O_{6+x}$, and have found that the paramagnetic-antiferromagnetic ordering temperature is extraordinarily sensitive to pressure.

The crystals which we have studied have the compositions $NdBa_2Cu_3O_{6.1}$ and $NdBa_2Cu_3O_{6.35}$, and weighed 50 and 9 mg, respectively. These samples have been investigated[3] 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 (T_{N1}) at small x, with a simple spin configuration (inset to Fig. 1). Nearest-neighbor spins both within the tetragonal a-b plane and along the c axis are aligned antiparallel, and the spin direction is in the a-b plane. This structure gives rise to magnetic Bragg peaks of the type $(h/2, k/2, l)$, 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 towards zero. This decrease is associated with ordering of the Cu chain-layer spins, and gives rise to new magnet-

ic Bragg peaks of the type $(h/2, k/2, l/2)$ below T_{N2} (dotted curve). We remark that the antiferromagnetic plane ordering (T_{N1}) and the superconducting phase boundaries as a function of x are well established.[4,10] 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 $NdBa_2Cu_3O_{6.1}$ crystal, and $T_{N1} = 230$ K and $T_{N2} = 10$ K for the $NdBa_2Cu_3O_{6.35}$ crystal.

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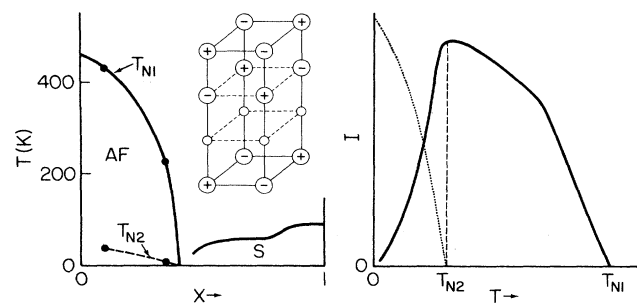

FIG. 1. Shown on the left side is a schematic of the phase diagram as a function of oxygen concentration for $RBa_2Cu_3O_{6+x}$, with the superconducting (S) and antiferromagnetic (AF) phases indicated. A sketch of the magnetic Bragg intensity for the $(h/2, k/2, l)$-type antiferromagnetic peaks (solid 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.

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tor. 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 $(\frac{1}{2} \frac{1}{2} 2)$ 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).[11]

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 ± 3) K/kbar. This rate of increase is more than 2 orders of magnitude higher than the rate of 0.05 K/kbar observed[12] 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[13] 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.[13,14]

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 $(\frac{1}{2} \frac{1}{2} 2)$ peak at a fixed temperature of 260 K. The strength of the scattering rapidly increases at low pressure, and then saturates above $\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
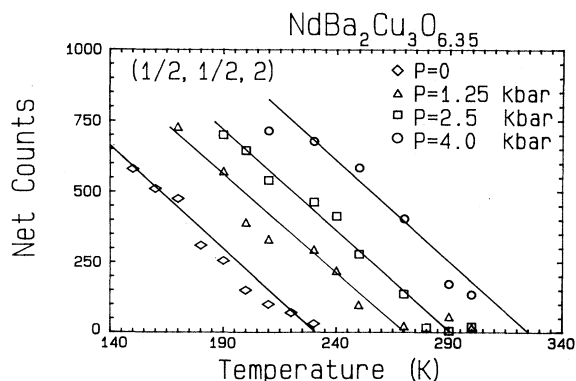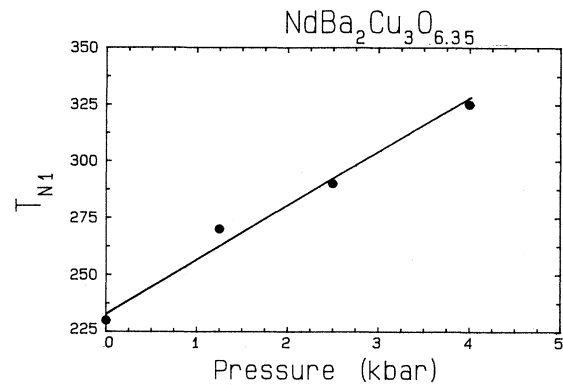

FIG. 3. Pressure dependence of the Néel temperature T_{N1}. The solid line is a fit to the data, and yields a slope 23 K/kbar.
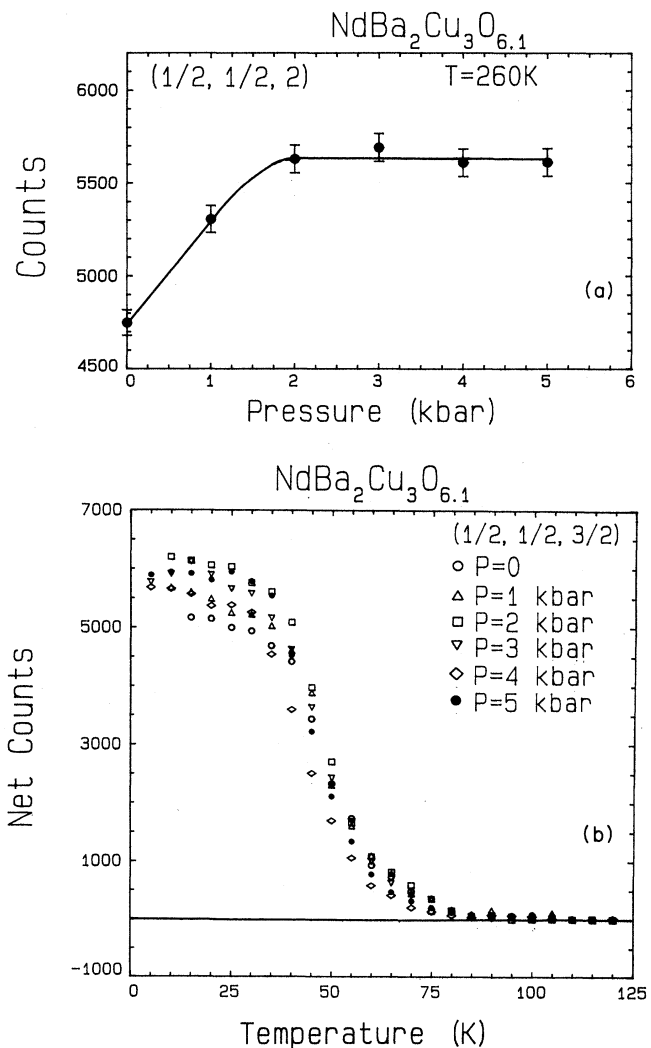

FIG. 4. (a) Pressure dependence of the intensity of the $(\frac{1}{2} \frac{1}{2} 2)$ Bragg peak for the NdBa$_2$Cu$_3$O$_{6.1}$ sample at 260 K. The intensity initially increases rapidly, and then saturates above $\sim$2.5 kbar. (b) Intensity of the $(\frac{1}{2} \frac{1}{2} \frac{3}{2})$ Bragg peak at a series of pressures.

FIG. 2. Temperature dependence of the $(\frac{1}{2} \frac{1}{2} 2)$ Bragg peak intensity in the vicinity of T_{N1} for the NdBa$_2$Cu$_3$O$_{6.35}$ crystal at a series of pressures. The solid lines are fits to the data to obtain T_{N1}.

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 $dT_{N1}/dP \sim 78$ 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 intensity, 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 $\sim 0.03\%$/kbar, which translates into a compressibility of 7.5×10^{-13} Pa^{-1}. We detect no 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 RBa$_2$Cu$_3$O$_{6+x}$ system. The magnetic exchange interaction J within the Cu-O$_2$ layers is very large, and thus in the vicinity of T_{N1} there are very strong magnetic correlations within the Cu-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, re-

sults 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, a calculation[17] based on spin-wave fluctuations gives $T_{N1} \sim J \ln^{-1}(J/J')$ 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 RBa$_2$Cu$_3$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 La$_2$CuO$_4$-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 RBa$_2$Cu$_3$O$_{6+x}$ system, no such cancellation is present.

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