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ANTIFERROMAGNETISM IN $NdBa_2Cu_3O_{6.1}$

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Abstract

Neutron scattering experiments performed on single crystals of $NdBa_2Cu_3O_{6.1}$ reveal antiferromagnetic order for $T < T_N = 385 \pm 2K$ in which the structure is characterized by the magnetic wave vector (1/2 1/2 1/2). The magnetic intensities are accounted for by Cu^{++} spins coupled antiferromagnetically in the CuO_2 planes. The oxygen deficient layers exhibit a small staggered magnetization ϵS which induces the spin ordering -S, $-\epsilon S$, -S, +S, $+\epsilon S$, +S, along the tetragonal axis. The average staggered magnetization evaluated at room temperature is about 0.40 μ_B in the CuO_2 planes and about 0.04 μ_B in the oxygen deficient layers.

Among the various mechanisms proposed to describe the properties of the new high Tc superconductors[1], the magnetic pairing[2,3] appears actually as the most prominent ingredient. In $YBa_2Cu_3O_{6+x}$ the magnetic long range order was first detected by Nishida et al,[4] using μ^+ spin relaxation techniques; using neutron diffraction, Tranquada et al[5] clearly identified the magnetic structure as related to the Cu^{2+} spins coupled antiferromagnetically in the CuO_2 planes and along the tetragonal axis. Assuming no moments in the oxygen deficient layers, the spin direction was found in the tetragonal planes. Similar conclusions have been reported by Li et al[6] in the case of a $NdBa_2Cu_3O_{6+x}$ single crystal with unknown oxygen content. In $YBa_2Cu_3O_{6.35}$ single crystal, Kadowaki et al[7] observed, well below the Neel temperature a second transition to a new magnetic structure characterized by both (1/2 1/2 1/2) and (1/2 1/2 1) wave vectors. The competition between the two types of ordering was then attributed to frustrated spins due to the presence of Cu^{2+} moments in the oxygen deficient layers. In this paper we report on a new magnetic ordering in a single crystal of $NdBa_2Cu_3O_{6.1}$; the structure is uniquely characterized by the magnetic wave vector (1/2 1/2 1/2).

The single crystals were obtained using proper amounts of $BaCO_3$, Nd_2O_3 , and CuO mixed and heated up to 1300°C in a Pt crucible. The mixture was slowly cooled at the rate of 0.5°/min down to 800°C and then quenched. The sizable single crystal of $1.2 \times 1.2 \times 0.2 mm^3$ used in this work was annealed in an Ar gas atmosphere for 100 hours at 800°C . It is a tetragonal single crystal with the lattice parameters a=3.882 Å and c=11.689 Å measured at room temperature using neutrons with well-calibrated energy. The neutron scattering measurements were carried out at the Brookhaven-High-Flux-Beam-Reactor on the H9 triple-axis spectrometer, using 5meV neutrons and a Be filter. Pyrolitic graphite (002) reflection was used for the double monochromator and analyzer. The collimations were set to 60'-40'-60'-40'-80'. For structural calibration a large number of nuclear Bragg reflections were collected on the H7 triple-axis spectrometer using 14.7 meV. All of the scans were performed in the (1 1 0),(0 0 1) horizontal scattering plane.

In Figure(1) we show, at 80K, two sets of scans through the $(1/2\ 1/2\ l)$ magnetic reflections for both integer and half integer l . The data clearly indicate the presence of strong $(1/2\ 1/2\ l)$ peaks with l half integer while the intensity at the scattering vectors $(1/2\ 1/2\ l)$ with integer l, is barely above bakground. The integer l scattering was carefully monitored over a wide temperature range. Although there may be weak $(1/2\ 1/2\ l\ integer)$ fluctuations we find no evidence for a genuine long range ordered component from 80 to 385 K. In figure(2) we show the temperature variation of the integrated intensity of the $(1/2\ 1/2\ 3/2)$ magnetic reflection. As T is increased up to $T_N \approx 385K$, the intensity vanishes continuously.

In Table I we give a set of integrated intensities of $(H/2\ H/2\ L/2)$ peaks with H, L odd integers. These intensities were collected at room temperature using $\theta/2\theta$ scan mode. The evolution of these measured magnetic structure factors with the L component of the scattering vector is clearly compatible with spins lying in the tetragonal planes. Assuming collinear spins with the same magnitude $< S >$ on the CuO_2 layers and antiferromagnetic nearest neighbour ordering of the CuO_2 planes, we write the magnetic structure factor as $F_M \sim < S > [\epsilon + 2cos(2\pi l u)]$, where l is half integer and ϵ is the fraction of magnetization lying in the oxygen deficient planes; u is the z coordinate of the CuO_2 plane in units of c. We used ϵ and $< S >$ as two adjustable parameters to fit the observed magnetic intensities of 10 reflections listed in Table I. Making use of the magnetic form factor f(Q) of the Cu^{2+} ions determined experimentally in La_2CuO_4 by Freltoft et al[8], we obtained the best fit of our data for the positive value of $\epsilon = 0.10 \pm 0.02$ and the ordered moment on the CuO_2 planes of $< S >= 0.40 \pm 0.02$. For these values the calculated intensities are reported in Table I as well as the corresponding results for the case where no staggered magnetization is present in the oxygen deficient layers. In this fit u was fixed equal to the value 0.355 ± 0.002 which we determined on the same sample using structural refinement of 20 independent nuclear reflections. The resulting magnetic structure consists of alternating 3 spins(+) and 3 spins(-) along the tetragonal direction. Surprisingly, this sequence seems to be more energetically favoured than the simpler sequence of + - + - + - which one would obtain with exclusively antiferromagnetic interplanar interactions.

In conclusion, by showing the existence of a different type of magnetic ordering in $NdBa_2Cu_3O_{6.1}$ we have shown that the oxygen deficient layer can have a magnetic moment, and its presence is of considerable importance for inter-layer competition of interactions. *This work was supported by the U.S.-Japan Cooperative Neutron Scattering Program, and a Grant-In-Aid for Scientific Research from the Japanese Ministry of Education, Science and Culture. The work at Brookhaven is supported by the Division of Materials Science, U.S. Department of Energy under contract no.* DE-AC02-76CH00016. *Research at MIT was supported by the National Science Foundation under contract nos. DMR85-01856 and DMR84-18718.*

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Figure captions:

Fig. 1. Neutron scans, at 80 K, through the magnetic reflections (1/2 1/2 l) with half integer l, compared to the scans made, with the same statistic, through (1/2 1/2 l) reflections with integer l

Fig. 2. Temperature variation of the integrated intensity of the(1/2 1/2 3/2) peak, proportional to the square of the staggered magnetization.

Table I
Integrated intensity of magnetic reflections $(H/2\ H/2\ L/2)$ observed and calculated for single crystal of $NdBa_2Cu_3O_{6.1}$ assuming collinear spins lying in the tetragonal planes with $\epsilon = 0.10$ and $\epsilon = 0$

H	L	Q($Å^{-1}$)	Io	Ic($\epsilon = 0.1$)	Ic($\epsilon = 0$)
1	1	1.18	56 (21)	54	45
1	3	1.40	186 (22)	190	240
1	5	1.77	91 (21)	98	87
1	7	2.20	6 (12)	5	1
1	13	3.68	0 (18)	1	1
3	1	3.44	4 (18)	7	6
3	3	3.53	53 (18)	26	29
3	5	3.69	28 (18)	16	14
3	7	3.92	0 (20)	1	0
3	9	4.20	23 (24)	17	22
$\sum \lvert Ic - Io \rvert / \sum Io$			=	.14	.26

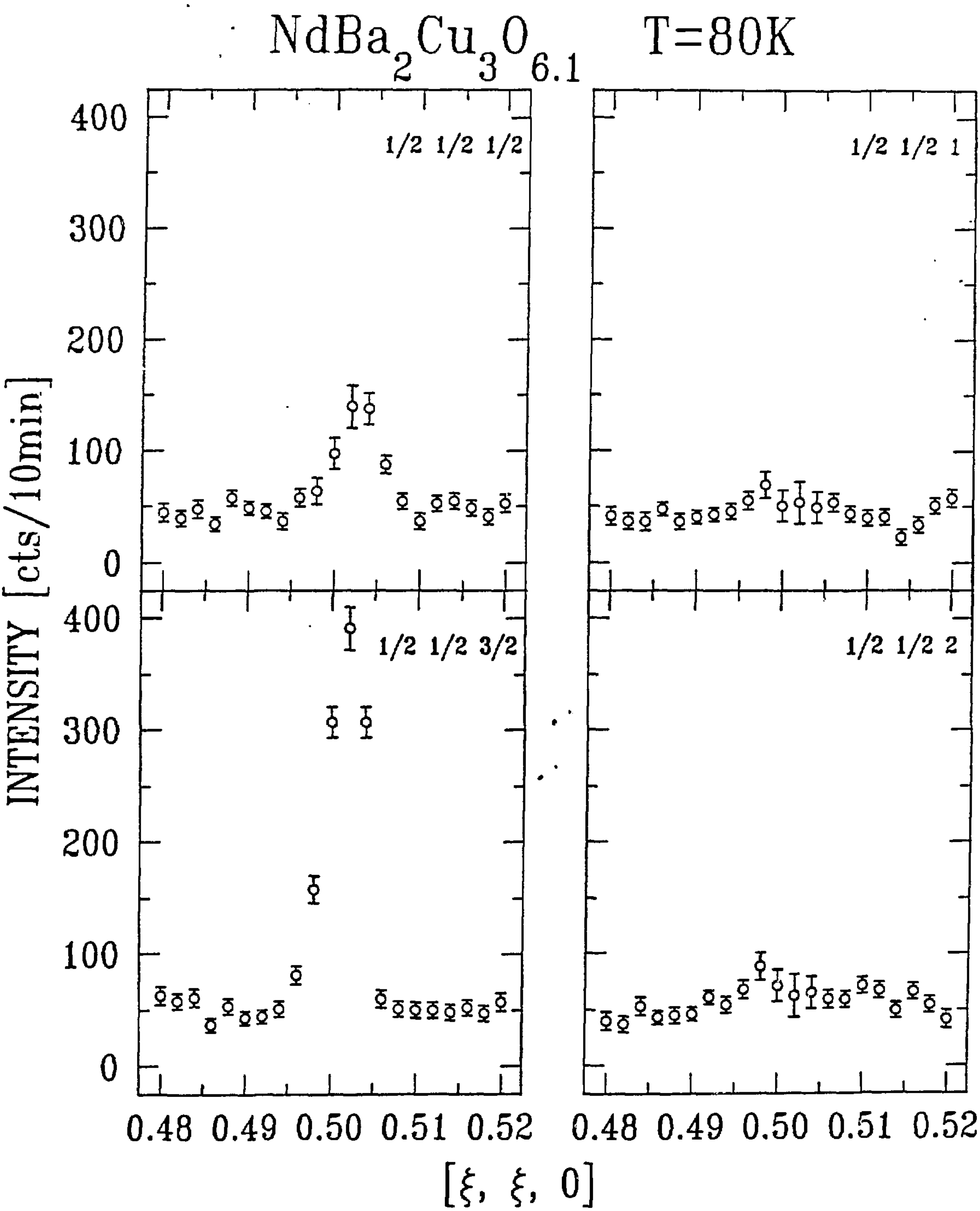
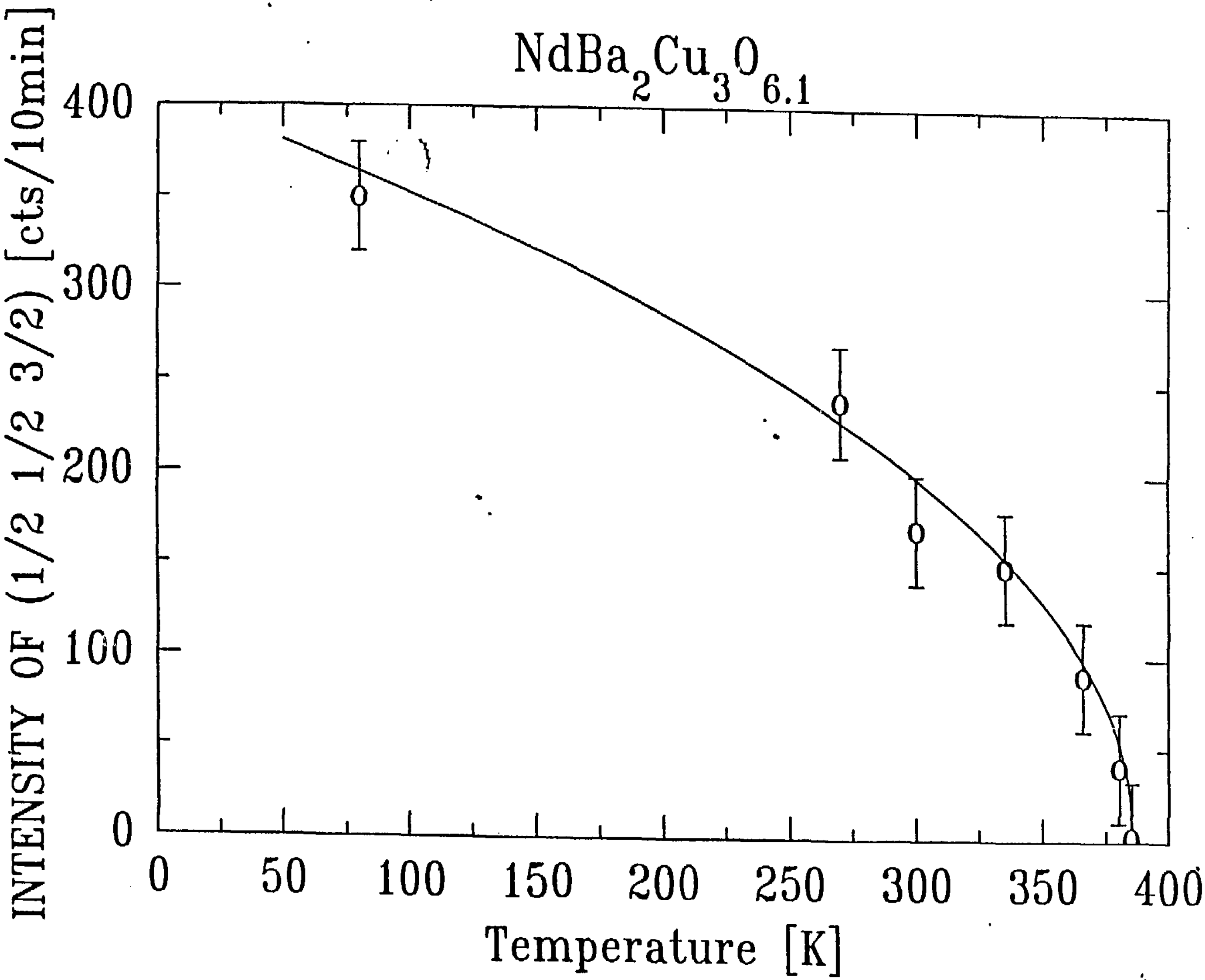

NdBa2Cu3O6.1 T=80K
INTENSITY [cts/10min]
1/2 1/2 1/2
1/2 1/2 1
1/2 1/2 3/2
1/2 1/2 2
400
300
200
100
0
0.48 0.49 0.50 0.51 0.52
[ξ, ξ, 0]

FIGURE 1


NdBa2Cu3O6.1
INTENSITY OF (1/2 1/2 3/2) [cts/10min]
Temperature [K]
0
100
200
300
400
0
50
100
150
200
250
300
350
400