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Crystal and magnetic structure of Nd_2CuO_4 at millikelvin temperatures

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We have investigated the crystal and magnetic structure of Nd_2CuO_4 at millikelvin temperatures. No distortion of the tetragonal structure could be detected with the present instrumental resolution. The refinement of the crystal structure with the intensities of 88 independent nuclear reflections measured at 500 mK lead to an agreement factor $R=0.057$. The magnetic structure was refined with 15 independent magnetic reflections measured at 80 mK which lead to an agreement factor of $R=0.136$. The magnetic moments of Nd and Cu atoms were found to be 1.3(1) and 0.47(8)μ_B, respectively, at $T=80$ mK. The magnetic moments of Nd and Cu atoms are parallel to the crystallographic [1 1 0] direction. No evidence for a component of moment parallel to [0 0 1] could be obtained from the present investigations.

1. Introduction

The magnetic properties of high-temperature superconductors have been investigated in great detail and there have been theoretical suggestions that the magnetic properties of these materials play an important role in the underlying superconducting mechanism. Fluctuating two-dimensional antiferromagnetic spin correlations in the CuO_2 planes have been reported to exist up to very high temperatures in these compounds and persist even in the samples which are doped to become superconductors in which the Neél temperature is reduced to zero. Until recently, all the different versions of cuprate superconductors studied were hole doped. Recently, a new series of cuprate superconductors have been discovered of the form $R_{2-x}Ce_xCuO_4$ (R=Pr, Nd, Sm or Eu) and also with the Ce replaced by Th [1–4]. These materials are particularly interesting because electrons rather than the holes, in the CuO_2 planes are suggested to be the charge carriers involved in the high-T_c superconductivity. The magnetic ordering of the copper moments in the undoped compounds Pr_2CuO_4 and Nd_2CuO_4 takes place at $T_N=270$ and 245 K, respectively [5,6]. In Nd_2CuO_4 additional magnetic transitions in which copper moments reorient have been reported at 75 and at 30 K [6,7]. Induced ordering of the rare-earth magnetic moments have been observed in both compounds. However, the details of the spin orientations of the copper and the rare-earth sublattice and ordered magnetic moments at very low temperatures have not been determined properly. We have therefore reinvestigated the magnetic structure of Nd_2CuO_4 at millikelvin temperatures.

2. Experimental

Single crystals of Nd_2CuO_4 were grown by the flux technique using CuO as the flux material. The single crystal used in the present investigation was a plate-shaped crystal of linear dimensions $5\times5\times0.5$ mm^3. Neutron diffraction investigations were performed with the diffractometer D15 of the Institut Laue-Langevin. The crystal was fixed on an aluminium

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plate which was fixed to the cold tip of the ^3He–^4He dilution cryostat. The crystallographic [1 1 0] axis of the crystal was parallel to the ω-axis of the diffractometer. The measurements were made with a calibrated wavelength of 1.176(1) Å and a small two-dimensional position-sensitive detector. The lattice constants were determined from the angles of 15 centered strong nuclear reflections and were found to be $a=3.935(5)$ and $c=12.14(1)$ Å at 80 mK. No distortion of the tetragonal structure could be detected at this low temperature with the present instrumental resolution.

3. Results and discussions

Intensities of all measurable nuclear reflections up to $\sin\theta/\lambda=0.55$ Å^{-1} were determined at 0.5 K which yielded 88 independent nuclear reflections. The crystal structure was refined from these intensities corrected for absorption ($\mu=0.728$ cm^{-1}). Table 1 gives the results of this refinement. The only positional parameter of the structure (space group I4/mmm) viz. the z parameter of the Nd atom was determined to be $z=0.3516(2)$. The conventional R factor of this refinement was $R=0.057$.

Magnetic reflections were detected corresponding to the propagation vector $\boldsymbol{k}=(\frac{1}{2}, \frac{1}{2}, 0)$. Intensities of 15 independent magnetic reflections from two domains ($\boldsymbol{k}_1=(\frac{1}{2}, \frac{1}{2}, 0)$ and $\boldsymbol{k}_2=(\frac{1}{2}, -\frac{1}{2}, 0)$) were measured at 80 mK. These intensities were corrected for absorption and were put on to an absolute scale using the scale factor obtained from the refinement of the nuclear structure. At this millikelvin temperature

Table 1
Refinement of the crystal structure of Nd_2CuO_4 at 0.5 K. Space group I4/mmm (No. 139), Nd (4e) 0, 0, z; Cu (2a) 0, 0, 0; O1 (4c) 0, $\frac{1}{2}$, 0; O2 (4d) 0, $\frac{1}{2}$, $\frac{1}{4}$.

Nd	z	0.3516(2)
	B (Å^2)	0.12(7)
Cu	B (Å^2)	0.28(8)
O1	B (Å^2)	0.48(8)
O2	B (Å^2)	0.39(8)
R		0.057
$N(hkl)$		88

moments of both Cu and Nd atoms are ordered. No magnetic intensity could be detected at $\boldsymbol{Q}=(\frac{1}{2}, \frac{1}{2}, 0)$ and $(\frac{3}{2}, \frac{3}{2}, 0)$ suggesting that the magnetic moments lie in the (1 1 0) plane and are parallel to the crystallographic [1 1 0] direction. The relative intensities of the magnetic reflections suggested that the Nd magnetic moments align parallel to the Cu moments which are nearest neighbours along the c-axis. This magnetic structure model is illustrated in fig. 1. Refinement of the magnetic moments of Nd and Cu atoms by using the measured magnetic intensities with the magnetic structure model of fig. 1 gave 0.9(1) and $0.33(6)\mu_B$ for Nd and Cu atoms, respectively. The form factor for the Cu^{2+} and Nd^{3+} were taken from Brown [8]. In the case of Nd^{3+} the coefficient c of the form factor $\langle j_0\rangle + c\langle j_2\rangle$ was refined to allow for crystal field effects. The agreement factor for this refinement was $R=0.136$. The results of this refinement are given in table 2. Assuming equally populated two domains the magnetic moments of Nd and Cu atoms are determined to be 1.3(1) and $0.47(8)\mu_B$, respectively. The magnetic moments of both Nd^{3+} and Cu^{2+} are considerably reduced from the single ion values of 3.27 and $1\mu_B$, respectively. The moment reduction of the Nd^{3+} is obviously related to crystal field effects. The moment reduction of Cu^{2+} is due to the spin fluctuations characteristic of the two-dimensional Heisenberg system. The presently determined value of the Cu^{2+} magnetic moment is the same as that obtained in La_2CuO_4 which is $0.5\mu_B$ [9]. Recently Matsuda et al. [7] have

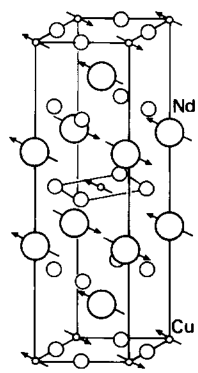

Fig. 1. The proposed magnetic structure model of Nd_2CuO_4 at 80 mK. The magnetic moments of both Cu and Nd atoms are parallel to [1 1 0]. The Nd magnetic moments are parallel to the Cu magnetic moments which are nearest neighbours along the c-axis.

Table 2
Refinement of the magnetic structure of Nd_2CuO_4 at 80 mK.

h	k	l	F_{obs}	F_{cal}
$-\frac{3}{2}$	$\frac{1}{2}$	0	68.7	62.5
$\frac{1}{2}$	$\frac{1}{2}$	0	0	0
$\frac{3}{2}$	$\frac{3}{2}$	0	0	0
$-\frac{5}{2}$	$\frac{1}{2}$	1	16.9	18.6
$\frac{3}{2}$	$\frac{1}{2}$	1	8.6	12.7
$\frac{1}{2}$	$\frac{1}{2}$	2	0	5.2
$\frac{3}{2}$	$\frac{3}{2}$	2	11.5	2.2
$\frac{1}{2}$	$-\frac{1}{2}$	3	68.6	70.8
$\frac{3}{2}$	$\frac{1}{2}$	3	34.7	40.7
$-\frac{3}{2}$	$\frac{1}{2}$	4	34.4	34.3
$\frac{1}{2}$	$\frac{1}{2}$	4	39.1	36.3
$\frac{3}{2}$	$\frac{3}{2}$	4	16.5	17.9
$\frac{1}{2}$	$-\frac{1}{2}$	5	6.0	12.1
$\frac{3}{2}$	$\frac{1}{2}$	5	8.3	7.8
$\frac{1}{2}$	$\frac{1}{2}$	6	50.1	47.2
R		0.136		
χ^2		22.3		
μ_{Nd} ($\times\mu_B$)		1.3(1)		
μ_{Cu} ($\times\mu_B$)		0.47(8)		

determined the magnetic moments of Nd^{3+} and Cu^{2+} in Nd_2CuO_4 to be 0.28 and 0.4 μ_B, respectively, at 8 K and 1.3μ_B for Nd^{3+} at 0.4 K. Our results agree completely with these.

It is to be noted that the present neutron diffraction investigation cannot distinguish between the single-k multidomain structure and the double-k magnetic structure. This has also been discussed by Matsuda et al. [7]. Recently Petitgrand et al. [10] have performed neutron diffraction measurements on Nd_2CuO_4 under a magnetic field parallel to [1, −1, 0] and conclude that the magnetic structure of Nd_2CuO_4 is non-collinear in zero field.

4. Conclusions

In conclusion, we have refined the crystallographic and magnetic structures of Nd_2CuO_4 from single-crystal neutron diffraction at millikelvin temperatures. We have determined the positional and thermal parameters of Nd_2CuO_4 at 500 mK and the magnetic moments of Nd^{3+} and Cu^{2+} at 80 mK.

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