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On the crystal and spin structures of Nd_2CuO_4

S. Skanthakumar and J.W. Lynn

Center for Superconductivity Research, Department of Physics, University of Maryland, College Park, MD 20742, and National Institute of Standards and Technology, Gaithersburg, Maryland 20899, USA

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X-ray and neutron diffraction have been used to study the magnetic and structural properties of single crystal Nd_2CuO_4. We previously observed a small distortion of the basic tetragonal structure, whereby peaks such as $(\frac{1}{2}\frac{1}{2}3)$ were observed with both X-ray and neutron diffraction. We explicitly show that these are not due to $\lambda/2$ wavelength contamination, as recently suggested. We also clarify the nature of the magnetic structures observed in this material. In particular, with the present neutron diffraction data on multidomain samples, it is not possible to distinguish between the collinear and noncollinear spin configurations which have been proposed for this system.

The magnetic properties of the Nd_2CuO_4 class of electron superconductors have been studied recently by neutron diffraction techniques [1–7]. Long range antiferromagnetic order of the Cu moments develops in Nd_2CuO_4 at $T_N = 245$ K, with a simple spin configuration in which nearest-neighbors within the Cu–O planes are antiparallel. The coupling between the layers, on the other hand, cancels to a first approximation, and the delicate balance of interlayer interactions appears to lead to a rich behavior as a function of temperature and field. In particular, there are two spin reorientations which occur at 75 and 30 K in this material. In addition, there is a substantial interaction between the Cu and Nd sublattices, and the Nd sublattice orders [4] at $T_N \cong 1.5$ K.

In our original paper [1] we proposed a noncollinear magnetic structure, in which the spins within the Cu–O layers are collinear and nearest neighbors are antiparallel, but the spin direction between layers is rotated by $\pi/2$, and hence the spins point in different directions (i.e. they are noncollinear). The symmetry of the magnetic system in this case is tetragonal [8]. A similar structure was proposed independently by Endoh et al. [2], in which the spin configuration within the layers is the same, but the spins between adjacent layers are collinear rather than noncollinear. The magnetic symmetry in this case is orthorhombic. The two structures are related in that the noncollinear configuration can be thought of as the coherent addition of two separate domains of the collinear structure. The magnetic Bragg intensities calculated for a single domain are different for the two cases, but for a multidomain sample in which the domain populations are equal the domain-averaged intensities are identical. Indeed the data obtained by the authors [1,3] and by Endoh et al. [2] are in excellent agreement, and with the present data it is not possible to distinguish the two spin configurations, contrary to implications made by other authors [9]. It may be possible to prepare a sample with unequal domain populations by the application of a magnetic field, or by applying stress, and measurements of this kind are under way. However, even in this case such experiments may not provide an unambiguous answer. For example, a similar situation has occurred for the spin structure of pure Nd, and the magnetic structure has been the subject of debate for many years [10].

One of the interesting features of the Nd_2CuO_4 system is the spin reorientations which occur at 75 and 30 K. Spin reorientations have not been observed in the sister compounds Pr_2CuO_4 [5,6] and Sm_2CuO_4 [7], and hence the transitions may originate from the particular Nd–Cu interaction. Another possibility is related to the fact that in our original work we observed a small structural peak at

forbidden Bragg positions such as $(\frac{1}{2}\frac{1}{2}3)$, and such a distortion could play an important role in the delicate balance of energies which decides the easy spin direction in this system. However, recently Takada et al. [11] suggested that the peaks we observed might have the trivial explanation that they originate from higher-order wavelength contamination in our diffraction experiments. Here we show that our peaks do not originate from experimental artifacts.

As reported in our original work, these peaks were observed with both neutron diffraction and X-ray diffraction. For the case of neutrons we used the standard technique of employing a pyrolytic graphite (PG) filter to suppress higher order wavelengths. The standard filter length is 5.1 cm, and results in a suppression of $\lambda/2$ contamination of $\sim 10^{-3}$. After we observed the extra structural peaks, we added an additional 3.8 cm of filter material, and the ratio of the $(\frac{1}{2}\frac{1}{2}3)$ peak to the fundamental nuclear reflections did not change. Hence we immediately ruled out the possibility that these peaks originated from wavelength contamination. We also remark that they cannot originate from such sources as multiple Bragg scattering.

The peaks in our X-ray diffraction measurements also do not originate from wavelength contamination, as shown in fig. 1. All of our data were taken with a wavelength of 1.54 Å (Cu K_α radiation) and a PG(002) monochromator. In the case of X-ray scattering the source spectrum may be adjusted by varying the tube accelerating voltage. In particular, if the accelerating voltage drops below the threshold for $\lambda/2$ photons of 16.08 kV, then there can be no $\lambda/2$ photons in the spectrum. The top portion of the figure shows the ratio of the $(\frac{1}{2}\frac{1}{2}3)$ peak to the (220) fundamental peak, for the single crystal of Nd_2CuO_4 used in our previous measurements. The ratio is seen to be independent of accelerating voltage, and thus cannot originate from $\lambda/2$ X-rays. The bottom portion of the figure shows data taken on a single crystal of silicon. The ratio of the $(\frac{1}{2}\frac{1}{2}\frac{1}{2})$ peak observed via $\lambda/2$ photons to the (111) fundamental peak is shown as a function of voltage. The ratio is seen to fall to zero below 16 kV, as expected.

The data show that the $(\frac{1}{2}\frac{1}{2}3)$ type peaks are not spurious. Indeed a structural distortion of the type we first reported recently has been confirmed via electron diffraction and microscopy as well as X-ray scattering [12–14]. The detailed nature of the distortion, and the relation (if any) to the spin rotations observed in Nd_2CuO_4 are still open questions.

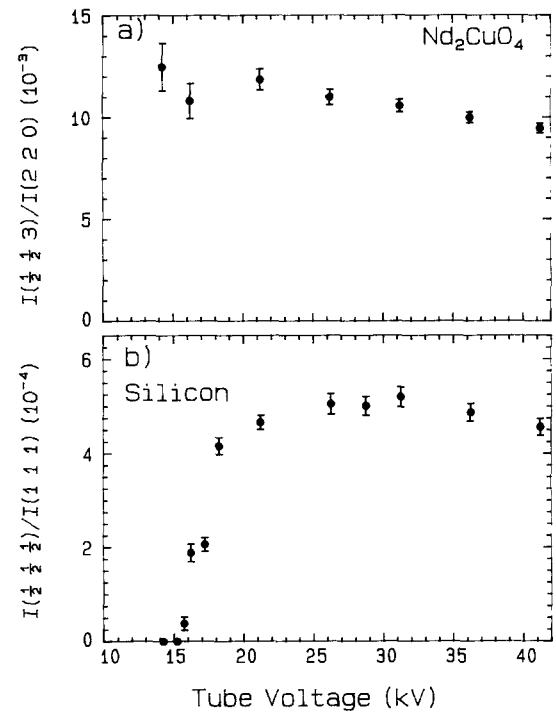

Fig. 1. The ratios of the X-ray peak intensities of the extra peaks to the fundamental charge peaks as a function of tube voltage. (a) For Nd_2CuO_4, the ratio of the $(\frac{1}{2}\frac{1}{2}3)$ and (220) peaks does not depend significantly on voltage even below 16.08 kV, the cut off for $\lambda/2$. This demonstrates that the $(\frac{1}{2}\frac{1}{2}3)$ is not due to higher order wavelength contaminations. (b) For Si, the ratio of the $(\frac{1}{2}\frac{1}{2}\frac{1}{2})$ and (111) peaks depends on tube voltage and drops to zero below 16.08 kV. This shows that the $(\frac{1}{2}\frac{1}{2}\frac{1}{2})$ peak is due to the $\lambda/2$ contamination, as expected.

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