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Antiferromagnetic order of Cu in Sm_2CuO_4

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Neutron diffraction techniques have been used to study the magnetic order of the Cu ions in a single crystal of Sm_2CuO_4. The measurements revealed the development of long-range magnetic order of the Cu moments at $T_N = 280 \pm 1$ K, with a relatively simple antiferromagnetic configuration of spins as found in Nd_2CuO_4 and Pr_2CuO_4. However, the spin directions in Sm_2CuO_4 are rotated by 90° from the spin directions in Nd_2CuO_4 and Pr_2CuO_4. The detailed spin structure can be either collinear or noncollinear, and in these tetragonal systems it is not possible to distinguish between them with the present neutron diffraction data on multidomain samples. Our measurements demonstrate that there are no spin reorientations below the Néel temperature, in contrast to the behavior found for Nd_2CuO_4.

The magnetic properties of the high-temperature superconductors have attracted considerable interest since the discovery that the Cu ions in these materials carry an unpaired spin and the consequent possibility that the magnetic fluctuations are responsible for the Cooper pairing. The magnetic properties of $(La_{2-x}Sr_x)CuO_4$ and $\mathscr{R}Ba_2Cu_3O_{6+x}$ ($\mathscr{R}$ = rare earth), where the charge carriers are holes, have been studied by neutron scattering, muon precession, and susceptibility measurements.[1] Superconductivity has also been discovered in a new class of materials, $R_{2-x}Ce_xCuO_{4-y}$ (R = Pr, Nd, Sm, or Eu),[2] where the charge carriers are electrons rather than holes. An investigation of the magnetic properties of these systems could further elucidate the possible relationship between superconductivity and magnetism.

The magnetic properties of Nd_2CuO_4 and Pr_2CuO_4, which are parent materials of the electron superconductors, have been studied recently by neutron diffraction techniques.[3–10] The Cu spins in these compounds order magnetically at higher temperatures (~250 K), with a simple antiferromagnetic configuration in which nearest-neighbor spins within the Cu–O planes are antiparallel. The spin direction between layers, however, can be either collinear or noncollinear; these two models cannot be distinguished with multidomain samples. The collinear spin structure is the same as the one found for La_2NiO_4,[11] where the spin direction **S** is parallel to the antiferromagnetic propagation vector $\mathbf{q}_m$. For Nd_2CuO_4, two spin reorientations of the Cu sublattice are observed at 75 and 30 K,[3,6] and in addition there is a substantial interaction between the Nd and Cu sublattices.[5] However, these spin reorientations do not occur in Pr_2CuO_4.[8] In the present work we report neutron diffraction measurements on Sm_2CuO_4, which is also a parent material of electron superconductors. We observe a similar long-range antiferromagnetic ordering of Cu spins, with a Néel temperature $T_N = 280$ K, while no spin reorientations are found over the temperature range explored (0.75–280 K).

The neutron diffraction experiments were conducted on the BT-2 triple-axis spectrometer at the National Institute of Standards and Technology Research Reactor. A pyrolytic graphite PG(002) monochromator was employed, with a PG filter to suppress higher-order wavelength contaminations, and no analyzer crystal was used. The incident neutron energy was 14.55 meV and the angular collimations before and after the monochromator and after the sample were 60′–60′–40′ (full width at half-maximum), respectively. Single crystals of Sm_2CuO_4 were grown by mixing Sm_2O_3 and CuO powders,[12] and this mixture was heated rapidly to a maximum temperature just above the melting point of the mixture. Extra CuO was used as a flux to reduce the melting point of the starting materials. After a soak of several hours at the maximum temperature, the materials were cooled slowly to room temperature, and good quality free standing crystals were formed with sizes up to $10\times10\times0.5$ mm^3. The very high neutron absorption for samarium made the measurements quite difficult. Hence a thin plate-like single crystal, weighing 66 mg, was used in our measurements to reduce the effects of absorption. The crystal was mounted in the (*hhl*) scattering plane and the data were taken between the temperatures of 0.75 and 300 K. Measurements were made at higher temperatures in a closed cycle helium refrigerator to investigate the ordering of the Cu spins, and in a ^{3}He cryostat to search for the Sm ordering.

The basic crystal structure of Sm_2CuO_4 is tetragonal $I4/mmm$ (T' phase), with lattice parameters of $a = 3.917$ Å and $c = 11.95$ Å at 200 K, and is the same as the structure of Nd_2CuO_4 and Pr_2CuO_4. A transverse scan (sample rotation) of the (004) nuclear Bragg peak is shown in Fig. 1. A single symmetric peak is observed, which demonstrates the high quality of the sample. The full width at half-maximum is about 0.45°, which is entirely instrumental in origin. The intensity of this peak was found to be independent of temperature within experimental error, as would be expected for a nuclear Bragg peak.

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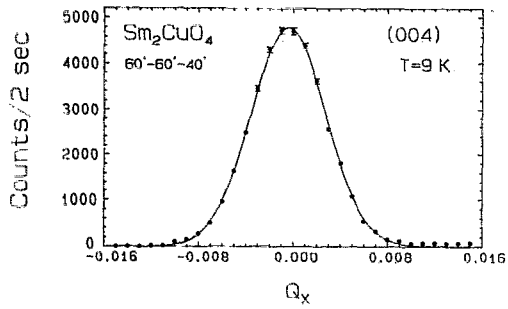

FIG. 1. Transverse scan (sample rotation) of a nuclear Bragg peak at the (004) reciprocal lattice position. The solid curve is a fit to the Gaussian instrumental resolution function. The full width at half-maximum is 0.45°.

A series of magnetic Bragg peaks were also observed below the Néel temperature of $T_N = 280$ K. These peaks can be indexed as $(h/2,k/2,l)$ based on the chemical unit cell, where h and k are odd integers and l is any integer. This is the same indexing as found for the other R_2CuO_4 materials (R = Nd, Pr, and La).[2–11,13] The $(\frac{11}{22}0)$ peak, which is the strongest magnetic peak observed, is shown in Fig. 2. These data were obtained by rotating the sample through the magnetic Bragg peak, and the full width at half-maximum of about 0.34° can again be attributed entirely to the instrumental resolution appropriate for this diffraction angle. Since the first two Miller indices are half-integers, the Cu magnetic unit cell is double the chemical unit cell along both the a and b direction, and this indicates that nearest-neighbor spins within the Cu-O planes are antiparallel. Thus the magnetic spin configuration of the Cu spins within the Cu-O planes in Sm_2CuO_4 is the same as the configuration found in Nd_2CuO_4 and Pr_2CuO_4,[3–10] which in fact is the same configuration observed for all these oxide systems.

The magnetic Bragg peaks with even l were found to have large integrated intensities while those with l odd were found to have small integrated intensities. This behavior is the opposite to that observed for Pr_2CuO_4, and for Nd_2CuO_4 below T_N, and indicates that the spin directions are different. For example, in Pr_2CuO_4, and Nd_2CuO_4 in the high-temperature ordered state, the

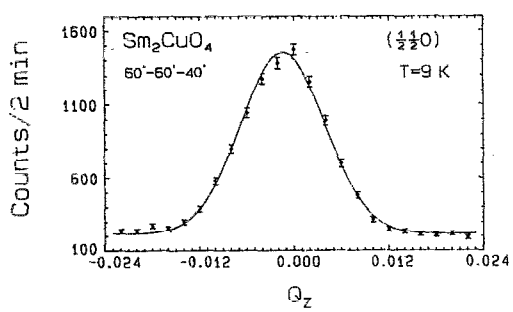

FIG. 2. Transverse scan of a magnetic Bragg peak observed at the $(\frac{11}{22}0)$ reciprocal lattice position at 9 K. The full width at half-maximum is about 0.34°.

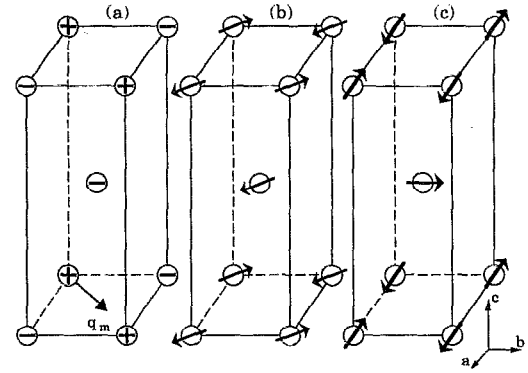

FIG. 3. Basic magnetic spin configuration and possible spin structures of the Cu spins in Sm_2CuO_4. (a) Magnetic spin configuration. The antiferromagnetic propagation vector $\mathbf{q}_m$ is along the [110] direction. (b) Collinear spin structure. There are ferromagnetic sheets of spins in (110) planes while the spins in adjacent sheets are antiparallel. The spin direction is $[\bar{1}10]$, which is perpendicular to the antiferromagnetic propagation vector. (c) Noncollinear spin structure. This structure can be thought of as the coherent addition of two separate domains of the collinear structure.

$(\frac{11}{22}0)$ peak is found to have negligible intensity, while in Sm_2CuO_4 the $(\frac{11}{22}0)$ is the strongest peak observed. The $(\frac{11}{22}1)$ peak, on the other hand, was found to have lower intensity than either the $(\frac{11}{22}0)$ or $(\frac{11}{22}2)$ peaks, in contrast to the situation found for the Pr_2CuO_4 [and Nd_2CuO_4 above the (75 K) spin reorientation transition]. This indicates that the spin directions in Sm_2CuO_4 are rotated by 90° with respect to the spin directions of Pr_2CuO_4, while it is the same as the configuration found for Nd_2CuO_4 between 30 and 75 K, in the spin reorientation regime.

The basic symmetry of the magnetic spin configuration for the Cu spins in Sm_2CuO_4 is shown in Fig. 3(a), and this configuration is the same as the spin configurations found for the other 2-1-4 systems. The detailed spin structure, which entails assigning a spin direction to each site, turns out to contain an ambiguity; there are two possible descriptions which may occur for the present case where the crystal structure has tetragonal symmetry, and it is not possible to distinguish between them with neutron diffraction data on multidomain samples. An identical situation occurs for both Nd_2CuO_4[14] and Pr_2CuO_4.[8] One possibility is a collinear spin structure as shown in Fig. 3(b), which is the same structure observed in (orthorhombic) La_2CuO_4.[13] The antiferromagnetic propagation vector $\mathbf{q}_m$ is along the [110] direction, while the spin direction $\mathbf{S}$ is along the $[\bar{1}10]$ direction. The structure then consists of ferromagnetic sheets in the (110) plane, with the spins in adjacent sheets antiparallel, and $\mathbf{q}_m \perp \mathbf{S}$. This differs from the high-temperature collinear structures proposed for Nd_2CuO_4 and Pr_2CuO_4, where $\mathbf{q}_m \| \mathbf{S}$. The magnetic symmetry for this collinear structure is orthorhombic. A second possibility is the noncollinear spin assignments shown in Fig. 3(c). In this structure the spins within each Cu-O plane are again collinear and antiferromagnetically coupled, but the spins between adjacent planes are rotated by

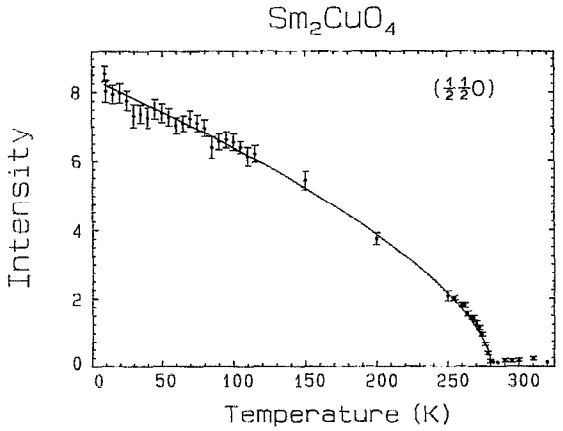

FIG. 4. Intensity of the $(\frac{11}{22}0)$ magnetic Bragg peak as function of temperature. Below 250 K integrated intensities are plotted, while above 250 K peak intensities are plotted. The solid curve is a guide to the eye. The Néel temperature is 280 K.

90° and hence are noncollinear. The basic magnetic symmetry in this case is tetragonal rather than orthorhombic. We remark that these magnetic structures are closely related in that the noncollinear structure can be obtained from the collinear structure by the *coherent* addition of two separate domains of the collinear structure. For multidomain samples, the observed intensities for these two structures are identical as already mentioned. Further discussion can be found elsewhere.[4,8,14]

The temperature dependence of the intensity of the $(\frac{11}{22}0)$ peak is shown in Fig. 4. Recall that this intensity is proportional to the square of the sublattice magnetization, which is the magnetic order parameter in this case. Most of these data were obtained from measurements of the integrated intensity, while near the Néel temperature the integrated intensities were supplemented by peak intensity data in order to determine the ordering temperature more accurately. The peak intensities have been scaled to the integrated intensities by normalizing the data at 250 K. The data reveal a continuous and reversible phase transition, with a Néel temperature $T_N = 280 \pm 1$ K. The data also demonstrate that there are no spin reorientations evident over the temperature range explored (0.75–280 K), in contrast to the behavior found for the Nd_2CuO_4 material.

One interesting feature of the order parameter data shown in Fig. 4 is that the $(\frac{11}{22}0)$ peak intensity does not saturate in intensity until well below 10 K. This has been observed in other Cu–O systems, and is thought to originate from the two-dimensional quantum fluctuations present in these highly anisotropic antiferromagnets. The estimate of the ordered moment we obtain from these data is $(0.38 \pm 0.04)\ \mu_B$, although there is some uncertainty in this value due to the rather large absorption corrections as well as the uncertainty in the coherent nuclear scattering amplitude for Sm.

Another interesting observation we have made is that we do not observe any significant change in the intensities of $(h/2, h/2, l)$ peaks, or development of new peaks in this scattering plane, down to 0.75 K. It is known from specific heat and magnetic susceptibility measurements, however, that the Sm ions order antiferromagnetically at 5.95 K.[15,16] If the Sm ions in Sm_2CuO_4 order like the Nd ions in Nd_2CuO_4,[4,5] then we would have expected substantial changes in these peak intensities. These data indicate that the ordered Sm moment must either be less than $0.1\ \mu_B$, or (the more likely case) that the Sm spin arrangement is fundamentally different. This may have important implications for the nature of the interaction of the rare-earth and Cu sublattices in these materials,[5] and for the effect the rare-earth sublattice has on the superconducting properties.[16] The ordering of the Sm ions will be the subject of a separate study.

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