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Citation: J. Appl. Phys. 83, 7345 (1998); doi: 10.1063/1.367635
View online: http://dx.doi.org/10.1063/1.367635
View Table of Contents: http://jap.aip.org/resource/1/JAPIAU/v83/i11
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Tilted antiferromagnetic ordering of Mn in Nd_{0.62}Ca_{0.38}MnO_{3}

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The magnetic ordering of the Mn spins in polycrystalline Nd_{0.62}Ca_{0.38}MnO_{3} has been investigated by means of neutron diffraction and ac magnetic susceptibility measurements. Three peaks around 230, 90, and 40 K were observed in the temperature dependence of the in-phase component of the ac susceptibility, $\chi'(T)$. Neutron diffraction measurements show that the peak at 90 K is associated with the ordering of the Mn spins, and that at 40 K is due to the reorientation of the Mn spins. Both ferromagnetic and antiferromagnetic coupling between the Mn spins were observed. The spins order at $T_N \approx 130$ K, with a spin structure consisting of ferromagnetically coupled, tilted antiferromagnetic sheets. No evidence was found from the neutron diffraction data to indicate that the peak around 230 K in $\chi'(T)$ is of magnetic origin, which suggests that it is associated with charge ordering. © 1998 American Institute of Physics. [S0021-8979(98)27411-1]

The discovery of colossal magnetoresistance (CMR) in distorted rare-earth containing manganites has sparked renewed interest in this class of materials. CMR as large as ten thousand fold has been observed in many systems. This phenomenon is now known to arise from the occurrence of a ferromagnetic transition accompanied by an insulator-metal transition, in which the double exchange (DE) interaction involving charge and spin exchanges is the main mechanism believed to be operating. The ferromagnetic metallic state is achieved by partially substituting trivalent rare-earth ions with divalent alkaline-earth ions. So far, the La-based compounds have been intensely investigated, whereas fewer studies have concentrated on the Nd-based systems. In this article, we present studies made on a Ca-doped Nd-based compound using neutron diffraction and ac magnetic susceptibility measurements. Charge ordering, tilted antiferromagnetic ordering of the Mn spins, and spin reorientation were observed.

Polycrystalline sample was prepared by the standard solid-state reaction techniques. Detailed sample fabrication procedures can be found elsewhere. The sample fabricated was characterized by a complete structural analysis using neutron powder diffraction and Rietveld analysis. High-resolution neutron diffraction patterns taken at eight temperatures were collected on BT-1, the 32-detector powder diffractometer at the U.S. NIST Research Reactor. A conventional setup of Cu(311) monochromator, pyrolytic graphite (PG) filter, and collimations of 15°-20°-7° full width at half maximum (FWHM) acceptance was employed. The high-resolution diffraction patterns were analyzed using the General Structure Analysis System (GSAS) program. The refinements were carried out assuming the symmetry of space group Pbnm. There are essentially no unexpected peaks present, showing that the sample is practically single phase. We estimated the impurity level to be less than 1%. Careful analysis of the occupancy factors shows that 38% of the Nd sites are occupied by Ca atoms, while the O sites are almost fully occupied. Hence a chemical formula of Nd_{0.62}Ca_{0.38}MnO_{3} is obtained. The compound crystallizes into an orthorhombic phase, with a crystal structure that may be viewed as a stacking of MnO$_2$-(Nd/Ca)O layers ($a-b$ planes) along the $c$ axis direction. Lattice constants that we obtained at $T=180$ K are $a=5.4041(2)$, $b=5.4526(2)$, and $c=7.6274(4)$ Å.

The ac susceptibility measurements were performed on a conventional susceptometer, with which both the in-phase component $\chi'$ and the out-of-phase component $\chi''$ can be measured. The temperature dependence of the ac susceptibility data were collected using a driving field of frequency 100 Hz and rms strength 1 Oe. Shown in Fig. 1 is the variation of $\chi'$ with temperature. Three peaks are seen over the temperature range studied. Using an expanded $y$ scale, shown as an inset of Fig. 1, a small but definitive peak is clearly revealed around 230 K. This peak is not of magnetic origin (see below), and we believe it originates from charge ordering. The main feature seen in Fig. 1 is of course the huge but broad peak around 90 K. This peak is associated with the ordering of the Mn spins (see below), and it has an antiferromagnetic character as a cusp is present. At 40 K another peak is clearly seen in $\chi'(T)$, which is due to the change of the moment direction of the Mn spins (see below). This behavior is much
reduced as a dc field is applied. No ac losses were detected, as essentially zero values were obtained for $\chi''$ at all temperatures studied.

Neutron magnetic diffraction was performed also at the NIST Research Reactor. Data were collected on the BT-9 triple-axis spectrometer operated in double-axis mode, using a conventional setup with PG monochromator and filter. Neutrons of wavelength $\lambda = 2.351$ Å and angular collimations of 40'-48'-48' FWHM acceptance were used. A pumped $^4$He cryostat was used to cool the sample. Diffraction patterns covering a range in scattering angle $2\theta$ from 10° to 65° were taken at several temperatures ranging from 1.8 to 325 K. No structural changes were observed over the temperature range studied. Detectable nuclear peaks appear at $2\theta$ greater than 30°, while the main magnetic peaks occur below 20°. Figure 2 shows the low scattering-angle portions of the diffraction patterns collected at four temperatures. At 160 K, which is above the temperature at which the most pronounced peak in $\chi'(T)$ appears, only background intensities are present in this $2\theta$ range. At lower temperatures, several new peaks develop, while the background intensities become lower as the magnetic scattering evolve into Bragg peaks.

Based on the nuclear unit cell, the two peaks shown in Fig. 2 may be indexed as the $\{00\} + \{0\bar{2}\}$ and $\{1\bar{2}\}$ Bragg reflections that originate from the ordering of the Mn spins. Clearly, the intensities of the $\{00\} + \{0\bar{2}\}$ and $\{1\bar{2}\}$ reflections vary differently with temperature, especially at temperatures below 15 K. The ratio between the $\{00\} + \{0\bar{2}\}$ and $\{1\bar{2}\}$ integrated intensities changes from being equal to 1.09 at 50 K, to 1.28 at 15 K, then to 1.40 at 1.7 K. These data indicate that the moment directions of the Mn spins vary with temperature. Besides the two peaks shown in Fig. 2, additional magnetic peaks that may be indexed as the $\{1\bar{1}\}$, $\{\bar{3}\bar{1}\}$, and $\{1\bar{2}\}$ reflections were also observed (not shown).

The temperature dependence of the $\{00\} + \{0\bar{2}\}$ and $\{1\bar{2}\}$ integrated intensities are shown in Fig. 3. Both peaks begin to develop around 150 K. The ordering temperature of the Mn spins, as determined by the inflection point, is $T_N \approx 130$ K. Above 50 K, these two peaks follow essentially the same temperature-dependent curve, showing that the spin direction does not change over this temperature regime. Below 50 K, the intensity of the $\{00\} + \{0\bar{2}\}$ peaks increase faster than that of the $\{1\bar{2}\}$ peak, implying that the spin directions are changing with temperature. We note that a corresponding peak, which may be completely smeared by an applied dc field of 1 kOe, appears in the $\chi'(T)$ data. The intensity of the $\{1\bar{2}\}$ peak saturates at 15 K. This is the temperature at which the Mn moment is essentially saturated. However, the intensity of the $\{00\} + \{0\bar{2}\}$ peak continues to increase as the temperature decreases further. This intensity is still not saturated even at 1.7 K, the lowest temperature studied. If this further increase of the $\{00\} + \{0\bar{2}\}$ intensity is due to the reorientation of the Mn spins, it must be accompanied by decreases of the intensities of other magnetic peaks. However, no such evidence was found. Instead, the intensity of the $\{301\} + \{0\bar{2}\} + \{0\bar{2}\}$ peak was found to increase with reducing temperature as well. Another possible origin is the ordering of the Nd spins, and the observed data indeed may be explained quite well if one assumes the Nd spins are ordered with a simple antiferromagnetic arrangement. However, Nd spins usually order at quite low temperatures, lead-
ing us to propose that the moments on the Nd are induced by the ordered Mn moments. More data taken at lower temperatures are needed to make this argument conclusive.

At high temperatures, on the other hand, the intensity ratios did not vary significantly. Shown as an inset in Fig. 3 is the integrated intensity measured at high temperatures. No evidence was found to support the argument that the peak observed in $\chi'(T)$ around 230 K is due to a ferromagnetic transition induced by the Ca substitution.

Based on the data presented above, we propose the following magnetic structure and its variation with temperature for the Mn spins in Nd$_{0.62}$Ca$_{0.38}$MnO$_3$: The coupling of the Mn spins in the $a-b$ plane is basically antiferromagnetic, while the interlayer coupling is ferromagnetic. Figure 4 shows the arrangement of the Mn spins in the $a-b$ plane. It consists of antiferromagnetic chains along the $b$ axis, with the moment directions of every fourth chain tilted by a small angle with respect to that of other chains. The tilting is necessary to explain the appearance of the $\{\frac{1}{2}l\}$-type ($l$=integer) reflections that accompany the $\{\frac{1}{2}\frac{1}{2}l\}$-type reflections, which we believe is caused by the Ca substitution. Our powder neutron data cannot resolve the $a$ axis from the $b$ axis due to their similarity. The specific moment directions in the $a-b$ plane hence cannot be determined from our data. For clarity, in Fig. 4 we choose the moments to be basically along the $b$ axis. Above 50 K the tilt angle is about $5^\circ$. Below 50 K this tilt angle increases with decreasing temperature, and reaches $15^\circ$ as the Mn moments saturated at $T \approx 15$ K, with $\langle \mu_z \rangle = 2.07(3) \mu_B$.

The research at the NCU was supported by the National Science Council of the Republic of China under Grant No. NSC-87-2112-M-008-017.