Magnetic excitations in La$_{1.5}$Sr$_{0.5}$CoO$_4$

L. M. Helme,$^1$ A. T. Boothroyd,$^1$ D. Prabhakaran,$^1$ F. R. Wondre,$^1$ C. D. Frost,$^2$ and J. Kulda$^3$

$^1$ Department of Physics, Oxford University, Oxford, OX1 3PU, United Kingdom
$^2$ISIS Facility, Rutherford Appleton Laboratory, Didcot, U.K.
$^3$ Institut Laue-Langevin, BP 156, 38042 Grenoble Cedex 9, France

We report here on polarized- and unpolarized-neutron scattering measurements and magnetometry studies of the half-doped compound La$_{1.5}$Sr$_{0.5}$CoO$_4$, which exhibits a checkerboard pattern of charge ordering below $\sim 800$ K. In the antiferromagnetically-ordered phase below $\sim 40$ K the spins are found to be canted in the $ab$ plane. The spin excitation spectrum includes spin-wave excitations with a maximum energy of 16 meV, and diffuse magnetic modes at energies around 30 meV.

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Layered transition metal oxides have been much studied in an attempt to understand the mechanism of high-$T_c$ superconductivity in the cuprates. Recently there has been speculation that charge stripe correlations may play an important role, leading to investigations of compounds such as La$_{2-x}$Sr$_x$NiO$_4$ which exhibits well defined spin and charge stripe ordering [1].

Here we consider the structurally identical but less studied family, La$_{2-x}$Sr$_x$CoO$_4$. Previous studies [2] of the half-doped member La$_{1.5}$Sr$_{0.5}$CoO$_4$ have concluded that below $T_s \approx 800$ K the holes introduced by Sr doping form a charge ordered phase, with a loosely correlated checkerboard arrangement of Co$^{3+}$ and Co$^{2+}$ ions. It has also been reported that very slightly incommensurate long range magnetic order occurs below $\sim 60$ K [2, 4], with a spin-freezing transition at $T_s \approx 30$ K [2].

We report here on polarized- and unpolarized-neutron scattering measurements and magnetometry studies of single crystals of La$_{1.5}$Sr$_{0.5}$CoO$_4$. The crystals were grown by the floating-zone method in Oxford.

Magnetization data were collected using a superconducting quantum interference device (SQUID) magnetometer with the field parallel to the $ab$ plane. DC field-cooled (FC) and zero-field-cooled (ZFC) data were recorded in a measuring field of 100 Oe.

Figure 1a shows the temperature variation of the in-plane FC and ZFC magnetizations, with the inset showing data up to 350 K. Our results agree with those of Moritomo et al. [4], with a broad maximum in the magnetization centred at about 60 K. However, we also see a definite splitting between the ZFC and FC data at low temperatures indicating a glassy ground-state, and an abrupt change in slope at 30 K. It is possible that this feature corresponds to a spin-reorientation transition as observed in La$_{1.5}$Sr$_{0.5}$NiO$_4$ [5].

To examine the magnetic order further we performed polarized-neutron diffraction on the triple-axis-spectrometer IN20 at the Institut-Laue-Langevin. We studied two rod-shaped crystals of La$_{1.5}$Sr$_{0.5}$CoO$_4$ (taken from one original crystal) with a combined mass of 12 g.

Figure 1b shows the temperature variation of the amplitude of a magnetic Bragg peak. This data confirms that La$_{1.5}$Sr$_{0.5}$CoO$_4$ is magnetically ordered below $\sim 40$ K, but reveals a slow build-up of magnetic correlations with decreasing temperature below $\sim 60$ K. It is likely that this gradual ordering explains the broad maximum in the magnetization data (fig. 1a).

Polarization analysis performed on the magnetic order peaks revealed that below 30 K the spins lie in the $ab$ plane at an angle of $\sim 12^\circ$ to the Co-O bonds. We also found tentative evidence that a small reorientation ($\sim 5^\circ$) occurs above 30 K, which would correspond to the kink in fig. 1a. Such a spin reorientation should cause an anomaly in the temperature variation of the magnetic peak shown in fig. 1a, but no anomaly is evident. This may be because of the small magnitude of the reorientation, or it may imply a different origin for the
magnetization kink.

We studied the spin excitation spectrum both with polarized- and unpolarized-neutron scattering. The latter was performed with the MAPS spectrometer at ISIS on a crystal of mass 35.5 g. The spin correlations were found to be highly two-dimensional (2D), showing no measurable inter-plane correlations for energies above 4 meV. For simplicity, therefore, we describe the scattering with respect to the 2D reciprocal lattice of the square CoO$_2$ planes.

Figure 2 shows a cut through unpolarized neutron data in the ($h$, $-h$) direction. The data were measured at $T = 9.5$ K with an incident energy of 50 meV. The cut has been averaged over $7 \leq E \leq 9$ meV and $0.215 \leq h \leq 0.295$ in the ($h$, $h$) direction. Lorentzian profiles were fitted to the two peaks, which correspond to spin-waves propagating parallel to the ($-h$, $h$) direction away from the ($0.255$, $0.255$) magnetic zone centre.

By fitting (mainly) constant-energy cuts we determined the two-dimensional spin-wave dispersions in the ($h$, $h$) and ($-h$, $h$) directions. These are shown in figure 3. The spin-wave velocity is found to be virtually isotropic in the $ab$ plane. The spin-wave is seen to extend to $\sim 16$ meV, in contrast to $\sim 50$ meV in La$_{1.5}$Sr$_{0.5}$NiO$_4$.[6] This indicates a much weaker exchange interaction in the cobaltates than in the nickelates. A quantitative analysis of the spin-wave dispersion is in progress.

Another feature of the excitation spectrum is a pattern of diffuse scattering between $\sim 25$ meV and $33$ meV, centred close to positions such as $(0, 0)$, $(\pm 0.5, 0)$, $(0, \pm 0.5)$ and $(\pm 0.5, \pm 0.5)$. Figure 4 depicts a cut along $(h, h)$, with data averaged over $25 \leq E \leq 29$. Scattering is seen at 9.5 K but not at room temperature, which indicates that the diffuse features are not due to phonons. Polarised neutron measurements confirmed that the scattering is magnetic.

The origin of this diffuse magnetic inelastic scattering is not clear at present. One possibility is that the Co$^{2+}$ spins have strong planar anisotropy, and that the diffuse feature corresponds to the excitation of the out-of-plane spin component. Another is that we are observing magnetic correlations among the Co$^{3+}$ sites. These possibilities are currently being assessed by modelling of the spin excitations.

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* Electronic address: helme1@physics.ox.ac.uk

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