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Neutron diffraction study of a $La_2CoO_{4.15}$ single crystal

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Abstract

A single crystal of $La_2CoO_{4.15}$ has been grown by the floating zone method and characterised extensively by bulk magnetisation and neutron diffraction studies. Long range antiferromagnetic order (AFM) is observed below (227.7 ± 0.5) K and at (103 ± 2) K a new AFM phase appears. The low temperature phase transition is associated with a first-order structural phase transition from a high-temperature orthorhombic phase to a low-temperature tetragonal phase.

Keywords: Neutron diffraction; Superconductivity; La_2CoO_4

1. Introduction

The magnetic properties of the La_2MO_4 (M = Cu and Ni) systems have attracted much interest in connection with high temperature superconductivity. The antiferromagnetic structure of the two differ only by a 90° rotation of the moments about the magnetic propagation vector [1]. In the cuprate structure, the antiferromagnetic propagation vector, τ, is perpendicular to the spin direction, $\boldsymbol{S}$, and in the nickelates τ is parallel to $\boldsymbol{S}$. The discovery of a superconducting phase in superoxygenated $La_2CuO_{4+\delta}$ ($T_c = 38$ K; $\delta = 0.13$ [2]) has also spurred on studies into oxygen rich cuprates and related compounds. La_2CoO_4 is related to the cuprates since they have similar crystal structures (K_2NiF_4 type) and magnetism (antiferromagnetic insulators) [1]. However, La_2CoO_4 can incorporate much more excess oxygen than the cuprate due to the extreme ease with which the Co^{2+} oxidises to Co^{3+}. Superconductivity has not reliably been observed in either the nickelate or cobaltate systems.

To understand the physical properties of $La_2CoO_{4+\delta}$, single crystals of different stoichiometries have been used in bulk property and microscopic studies. This paper reports the findings from one crystal.

2. Experimental details

Single crystals of $La_2CoO_{4+\delta}$ have been grown by the floating zone method using a double elliptical image furnace. The composition and crystal parameters were determined by EDAX and XRD. TGA in a CO/CO_2 atmosphere was used to determine the oxygen concentration. The magnetic properties parallel and perpendicular to the Co–O planes were measured by AC susceptibility (mutual inductance) and DC magnetisation (SQUID) techniques. A neutron diffraction experiment was carried out on the four circle diffractometer at Risø National Laboratory, between 300 and 20 K.

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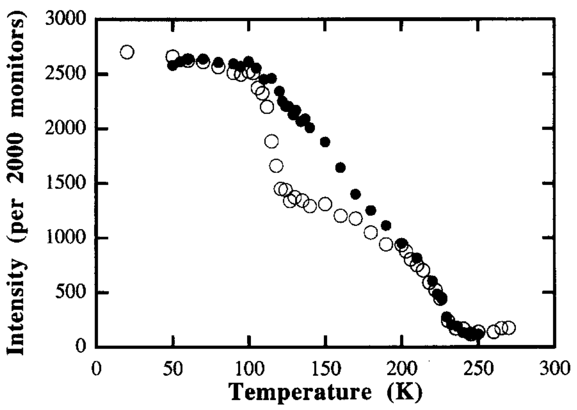

Fig. 1. The temperature dependence of the magnetic (1 0 0) Bragg peak. The solid and open symbols represent warming and cooling respectively. The error bars are no bigger than the size of the symbols.

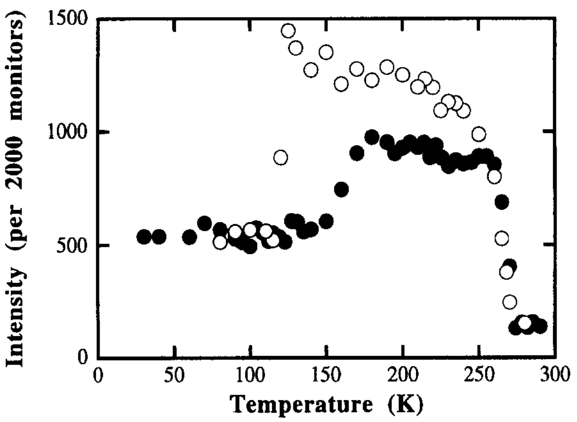

Fig. 2. The variation in intensity with temperature of the magnetic (0 1 1) Bragg Peak. The solid and open symbols represent warming and cooling respectively. The two magnetic transitions are clearly observed. The error bars are no bigger than the size of the symbols.

3. Results and conclusions

A 0.08 cm^3 single crystal of $La_2CoO_{4.15}$ (space group Cmca at room temperature) was used in the magnetic and neutron diffraction study in order to determine its magnetic properties. Magnetisation measurements (both AC and DC techniques) with the applied field perpendicular to the Co–O plane results in one peak at the Néel temperature, $T_N = (227.7 \pm 0.5)$ K. A second magnetic transitions at $T_2 = (103 \pm 2)$ K is only observed when the applied field is in the planes.

Neutron diffraction results showed the crystal to be twinned in the CoO_2 planes and at 103 K an orthorhombic to tetragonal phase transition occurs with decreasing temperature (cf $La_{2-x}Ba_xCuO_4$). From 103 to 230 K the magnetic moments align parallel to τ [1 0 0] (nickelate-type) and below 103 K the spins rotate within the plane, $\boldsymbol{S} =$ [0 0 1], (cuprate type).

Fig. 1 shows the temperature dependence of the magnetic (1 0 0) Bragg peak. It is clear from the hysteresis that the transition at 103 K is first order. The doubling of the intensity at 103 K is an indication that the twinned *a* and *c* axis become equal in length at the structural transition. The temperature dependence of the (0 0 1) peak is the same as the (1 0 0) within experimental error. Fig. 2 shows the temperature dependence of the (0 1 1) magnetic Bragg peak, again hysteresis clearly signals the first-order transition at T_2. The drop in the intensity at 103 K is associated with the 90° rotation of the moment.

Several distinct differences can be seen between our results and those reported by Yamada et al. from a La_2CoO_4 single crystal. First, we observe the Néel temperature at (227.7 ± 0.5) K rather than 275 K. Secondly, the first-order transition at (103 ± 2) K was observed at 135 K by Yamada et al. and finally, we observe the magnetic Bragg reflection (1 0 0) at all temperatures below the Néel temperature, unlike Yamada et al. who only observed it below 135 K.

Acknowledgements

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