Magnetic Excitations in CoO

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The magnetic properties of cobalt compounds have recently attracted attention [1,2] because the Co ion may have transitions between high and low spin states giving rise to charge ordering, magnetic ordering and superconductivity. The recent interest in exotic oxides displaying superconductivity or orbital order emphasizes the importance of understanding simple oxides such as CoO. An earlier study provided an incomplete picture [3].

Cobaltous oxide is a face-centred cubic antiferromagnet in which (1 1 1) ferromagnetic sheets of spins stack antiferromagnetically along [1 1 1] directions below T_N = 291 K with a ~ 1% contraction along a cube edge. At any wave-vector transfer, *Q*, up to as many as four spin-wave modes may arise from the domain and spin structure for any one transition from the ground state to one of the eleven excited spin and orbital states. The nearest neighbour antiferromagnetic exchange is frustrated, contributing no molecular field, and it is the antiferromagnetic next nearest neighbour exchange that breaks the symmetry below T_N. The nature of the order and fluctuations is still controversial [4,5].

In order to determine the nature of the three-dimensional ordering in the presence of the nearest neighbour frustration, we have made detailed measurements of the magnetic excitations in a high quality CoO crystal aligned in the (HHL) plane. Our previous high-resolution study indicated [6] that there are four resolved peaks at 6 K in the excitation spectrum between 4 and 12 THz for different magnetic zone centres. In a recent study, only two broad modes in the same spectral range were detected [7]. To gain more insight into the nature of these peaks, we determined the spectrum at other magnetic zone centres as well as at a few nuclear zone centres. In addition, we investigated these spectra at several temperatures below T_N from 11 K to 150 K.

The inelastic neutron studies were performed at the C5 triple axis spectrometer with a vertically focusing PG(002) monochromator and a flat PG analyzer at a fixed final energy of E_f = 3.52 THz for most of the scans. A fixed final energy of 7.36 THz was used at a few wave vectors either to be able to close the scattering triangle in the desired energy transfer range or to avoid large scattering angles that were not accessible. The horizontal collimations were set to [none, 0.48°, 0.56°, 1.2°] for which an energy resolution of 0.2 THz was achieved at zero energy transfer. One PG filter was used it the scattered side to suppress the higher order contamination.

The excitations observed at magnetic zone centres at 11 K are shown in Figure 1. Scans at lower energies (<4 THz) confirmed that there are no low energy modes of appreciable strength at any measured *Q*. For energy transfers between 4 and 14 THz, four excitations are observed at all zone centres. An analysis of the data including the Co^{2+} magnetic form factor indicates that the peak at 9.5 THz is mainly magnetic in origin. The intensities of the peaks at 6.5 and 7.6 THz decrease with *Q*, but by less than the form factor, suggesting the peaks at 6.5 and 7.6 THz have some magnetic weight. The peak at 4.8 THz has a substantial phonon component. This lower energy ~ 5 THz mode lies close in frequency to the TA zone boundary phonon. This phonon has the same cross-section at (1/2 1/2 3/2), (3/2 3/2 1/2) and (5/2 5/2 1/2) zones centres, hence we conclude that this lowest peak carries largely phonon amplitude.

The temperature dependence of the excitations observed at magnetic zone centres was also measured. This study again supports the conclusion that the excitation at 9.5 THz is mainly magnetic, the 7.4 THz and 6.5 THz modes have some phonon contribution and finally the mode at 4.8 THz is mainly due to phonons, possibly through an incoherent process.

Similar scans at nuclear zone centres were also performed (see Figure 3 for an example). From these scans we find the (002) intensity for some modes is stronger than their intensity at (222). This suggests these modes are likely to be magnetic. In addition since the nuclear Bragg peaks are the same for all domains, the frequencies of the modes are the same irrespective of the magnetic domains. Hence it is concluded that the peaks at 6.7 and 7.6 THz are mainly magnetic and common to all domains.

Within each domain, there are several bands of spin excitations to states controlled by the exchange, spin–orbit and tetragonally distorted crystal field. Numerical calculations of these modes with a Hund's rule model suggest that the two out-of-plane domains give similar frequencies and so the domain structure accounts for three distinct modes. This leads to a different model than that proposed in recent low-resolution experiments [7].

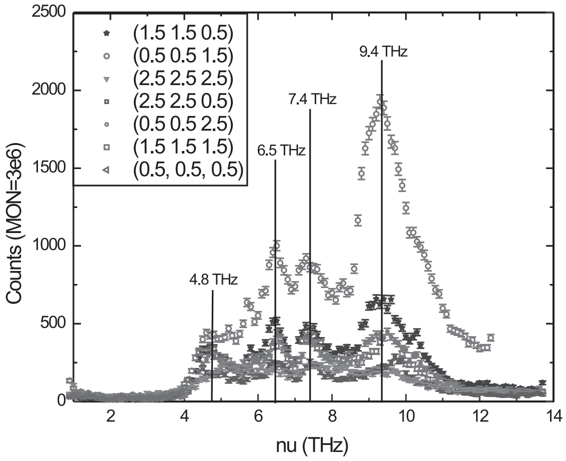

Fig. 1 *The observed excitations at several magnetic zone centres at 11 K. Data were collected at a fixed final energy of 3.52 THz except for (2.5 2.5 2.5) and (0.5 0.5 1.5) zone centres where a final energy of 7.36 THz was used.*

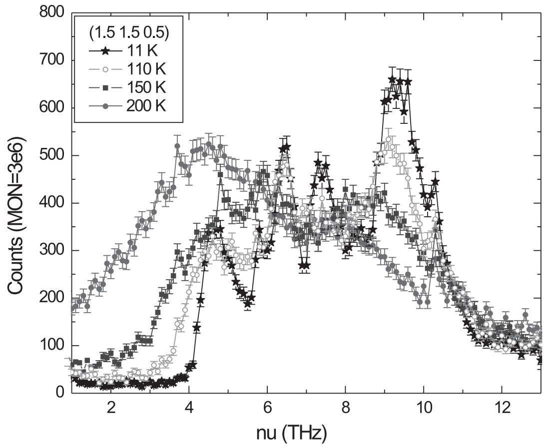

Fig. 2 The temperature dependence of the excitations at (1.5 1.5 0.5) again suggesting that the excitation at 9.5 THz is mainly magnetic whereas the one at ~ 5 THz mainly arises from phonons. The excitations between 6 to 8 THz have some magnetic contributions.

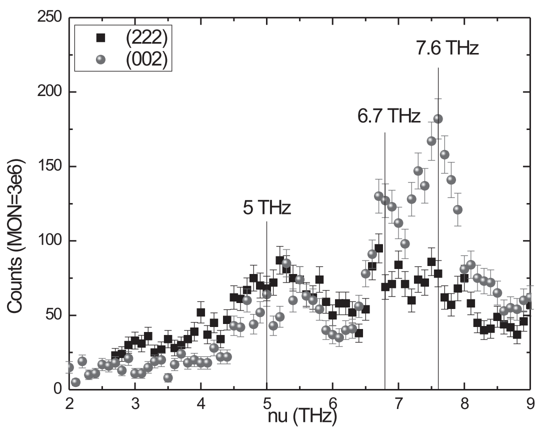

Fig. 3 The observed excitations at nuclear zone centres (002) and (222). Data were collected at a fixed final energy of 3.52 THz and at 11 K.

References

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