Orbital and Spin Excitations in Cobalt Oxide

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Abstract

By means of neutron scattering we have determined new branches of magnetic excitations in orbitally active CoO (T_N=290 K) up to 15 THz and for temperatures from 6 K to 450 K. Data were taken in the (111) direction in six single-crystal zones. From the dependence on temperature and \( Q \) we have identified several branches of magnetic excitation. We describe a model for the coupled orbital and spin states of Co\(^{2+}\) subject to a crystal field and tetragonal distortion.

Keywords: Transition metal compounds; Spin-waves; Neutron scattering

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].

We have made high-resolution measurements of the magnetic excitations in the (HHL) plane of a high-quality crystal of CoO at 6 K, 320 K and 450 K. In the first set of experiments, with a focusing PG(002) monochromator and flat PG analyzer with \( E_F = 3.52 \) THz, a resolution of 1.2 THz was achieved at 10 THz energy transfer. In a second set of measurements, with a Be(002) monochromator and PG(002) analyzer set to \( E_F = 7.37 \) THz, a resolution of 0.8 THz was achieved for excitations at 10 THz.

With both configurations we resolved four peaks at 6 K in the excitation spectrum between 4 and 12 THz as shown in Fig. 1 for different magnetic zones. In a recent study only two broad modes in the same spectral range were detected [6].

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Fig. 1: The magnetic excitations at different zone centres at 6 K fitted to gaussian profiles

partially magnetic. The peak at 4.8 THz has a substantial phonon component and 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 of Fig. 1, as observed, so that we conclude that the lowest peak carries largely phonon amplitude. The intensities of the three peaks at 6.5, 7.6 and 9.5 THz all have substantial magnetic weight with the lower two showing a mix with vibrational amplitude. In the same frequency region lie the transverse optic and longitudinal acoustic phonons at ~7.6 THz and ~8.3 THz [3].

The temperature dependence of the excitations is shown in Fig. 2. A comparison of the data at 320 K (above \(T_N\)) and 6 K shows that the peak at 9.5 THz disappears on increasing temperature to 320 K. This confirms that this peak is indeed magnetic in origin. The intensities of the other peaks also decrease becoming unobservable relative to the paramagnetic scattering, suggesting they are largely magnetic.

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 suggests that the out-of-plane domains have 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 [6]. Our analysis shows that even a ‘simple’ transition metal oxide can exhibit complex phenomena in its spin excitations.

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