Experimental report

17/08/2022

Proposal:	**4-01-1712**	**Council:**	10/2020
Title:	Temperature dependence of spin wave modes in multiferroic cupric oxide		
Research area:	Physics		

This proposal is a continuation of 4-01-1449

Main proposer: **Andrew Timothy BOOTHROYD**

Experimental team: Viviane PECANHA ANTONIO
Siobhan TOBIN
Andrew Timothy BOOTHROYD

Local contacts: Frederic BOURDAROT

Samples: Cupric oxide, CuO

Instrument	Requested days	Allocated days	From	To
IN22 CPA	0	7	14/09/2021	21/09/2021
IN20	9	0		

Abstract:

The objective is to follow the temperature evolution of the acoustic and optic magnon modes at the magnetic zone centre in CuO. Measurements will be made throughout the collinear antiferromagnetic phase ($T < 213K$) and into the helical phase ($213 < T < 230K$). We shall use longitudinal polarization analysis to identify modes from different spin components, and to separate spin wave scattering from optic phonons. With these systematic measurements we hope to be able to interpret the complex spectrum in the helical phase, and thus identify the interactions which (i) stabilize the helix at high temperatures, and (ii) stabilize the collinear antiferromagnet at low temperatures.

Despite its simple chemical formula, cupric oxide (CuO) has a surprisingly complex magnetic behaviour which remains only partly understood. Interest in CuO has intensified since the discovery in 2008 of improper ferroelectricity (or type-II multiferroic behaviour) in the temperature range 213 to 230K [1] associated with helicoidal magnetic order. Below 213K the magnetic structure transitions to a collinear antiferromagnetic (AFM) phase without ferroelectricity.

The magnetic structures in the two magnetic phases of CuO were established many years ago [2], and the important exchange interactions in the low energy effective Hamiltonian have recently been determined from unpolarised neutron measurements of the spin-wave spectrum at low temperature in the collinear AFM phase [3]. However, the subtler interactions which drive the collinear-to-helicoidal magnetic phase transition and which cause the magnetoelectric coupling are not yet established experimentally.

The experiment reported here was part of a systematic study we are undertaking of the magnetic excitations in CuO in order to develop a quantitative model for the spin Hamiltonian [3-5]. Our earlier polarized neutron study on IN20 [4] probed the low-energy magnetic spectrum at the magnetic propagation vectors $\mathbf{q}_{\mathrm{AFM}}$ = (0.5, 0, –0.5) and $\mathbf{q}_{\mathrm{helix}}$ = $\mathbf{q}_{\mathrm{AFM}}$ + (0.006, 0, 0.017) of the AFM and helicoidal phases, respectively. Figure 1(a) and (b) show such energy scans at T = 2 K and 215 K, respectively. These spectra are seen to be quite different. In particular, the prominent peak at 23 meV in the 2 K spectrum which corresponds to the optic magnon mode is not present in the 215 K spectrum.

We used neutron polarization analysis on IN22 to measure the spectrum at $\mathbf{q}_{\mathrm{AFM}}$ at a series of temperatures between 2 K and 205 K to determine how the 2 K spectrum evolves into the 215 K spectrum. Polarisation analysis is important to separate magnetic from phonon scattering. The crystal was the same 6.7 g crystal used previously on IN20. We measured at several different neutron scattering vectors **Q** for each energy scan, in order to obtain information about different spin fluctuation components.

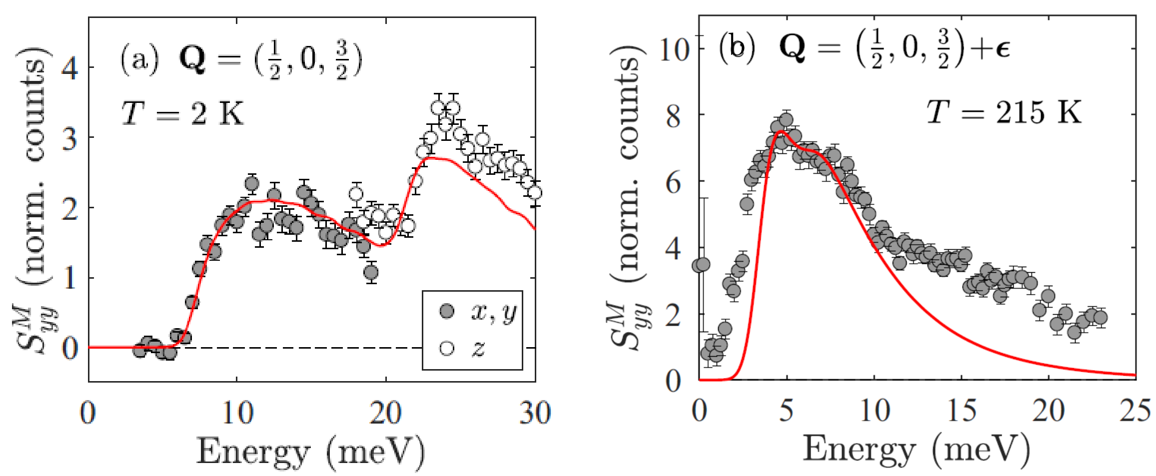

Figure 1. (a) Energy scan at $\mathbf{q}_{\mathrm{AFM}}$ obtained with PA on IN20 in the AFM phase. The mode at 23meV is the optic magnon, and gap below 7.5 meV is due to axial anisotropy [3]. (b) Similar energy scan at $\mathbf{q}_{\mathrm{helix}}$ in the helicoidal phase.

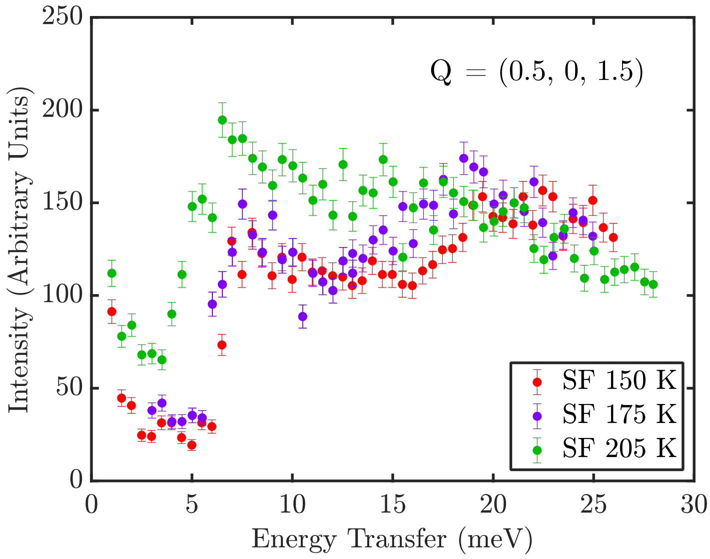

Figure 2. Energy scans at $\mathbf{q}_{AFM}$ recorded in the spin-flip (SF) polarization channel on IN22. Measurements are shown at three temperatures in the AFM phase.

The data enabled us to follow how the optic magnon gap and the anisotropy gap decrease with increasing temperature. This is illustrated in Fig. 2, which shows energy scans measured in the spin-flip polarization channel at $\mathbf{q}_{AFM}$ at T = 150 K , 175 K and 205 K. The anisotropy gap is seen to decrease with increasing temperature from about 7.5 meV at T = 2 K (see Fig. 1) to 5 meV at 205 K. The optic magnon gap decreases more rapidly with increasing temperature, and at 205 K there is no clear gap feature any longer. We have compared the rate at which the optic magnon gap decreases with increasing temperature and found disagreement with what would be predicted by linear spin-wave theory assuming the measured temperature dependence of the sublattice magnetization.

We are currently in the process of testing a model which could explain the temperature dependence of the two magnon gap features measured in the experiment.

References

[1] T. Kimura *et al.*, Nature Materials **7**, 291 (2008).

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