Experimental report

15/07/2016

Proposal: **4-01-1449** **Council:** 10/2014

Title: Magnons and electromagnons in the polar helicoidal phase of CuO

Research area: Physics

This proposal is a new proposal

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Samples: Cupric oxide

Instrument	Requested days	Allocated days	From	To
IN20	8	8	11/05/2015	17/05/2015
			17/06/2015	22/06/2015

Abstract:

Cupric oxide is a high temperature multiferroic. The polar phase is stable between 213 and 230 K, and has a helicoidal magnetic structure. We will measure the magnetic modes in the helicoidal phase at the magnetic zone centre, and we will compare these with the spin wave modes of the low temperature collinear antiferromagnetic phase. We expect to observe spin wave modes characteristic of the magnetic helix together with electromagnons caused by magnetoelectric coupling. Our aim is to elucidate the interactions that stabilise the helicoidal phase and cause the magnetoelectric coupling.

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 exhibits collinear antiferromagnetism without ferroelectricity.

Although the magnetic structures in the two magnetic phases of CuO were established many years ago [2], the important interaction terms in the low energy effective Hamiltonian that describes the magnetic order and magnetoelectric phenomena are not yet known. The experiment reported here is 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. Our earlier polarized neutron study on IN20 [3] and unpolarised measurements at ISIS [4] focused on the collinear phase at low temperatures. We have also detected an electromagnon excitation by THz time-domain spectroscopy [5]. The aim of the present experiment was to learn more about the subtle interactions that stabilise the helicoidal magnetic order in the temperature range 213 to 230K and cause magnetoelectric coupling. We used neutron polarization analysis on IN20 to identify the different low energy magnetic modes (E < 25 meV) of the helicoidal structure, including effects due to spin anisotropy.

CuO has a monoclinic structure (a = 4.68, b = 3.42, c = 5.13 Å, β = 99.5°). A crystal of mass 6 g was mounted on the spectrometer with the a and c axes in the horizontal scattering plane. A constant final wave vector k_f = 2.662 Å^{-1} was used. Most of the measurements were made in the helicoidal phase, in which the spin structure approximates to a transverse helix with ~90° angle between adjacent spins along the chain, i.e. $\uparrow\otimes\downarrow\odot$. The actual propagation vector is incommensurate, $\mathbf{q}_{helix}$ = $\mathbf{q}_{AFM}$ + (0.006, 0, 0.017) [2] where $\mathbf{q}_{AFM}$ is the antiferromagnetic wave vector (0.5, 0, –0.5) and equivalent wave vectors for the C-centred lattice.

Figure 1 presents a series of energy scans recorded on IN20 at four different helicoidal ordering wave vectors. The data quality is excellent, with a very high ratio of magnetic to non-magnetic scattering thanks to a flipping ratio of about 20. The energy spectra contain a lot of information because the direction of the neutron scattering vector **Q** for each energy scan varies relative to $\mathbf{q}_{helix}$, which means that the scans are sensitive to different spin components. In addition, the individual polarization channels are also a filter for the different spin components relative to **Q**.

A qualitative inspection of the data reveals a number of interesting features: (i) magnetic intensity in some channels extending down to the elastic line, indicating a Goldstone mode; (ii) magnetic peaks at 3 meV, 5 meV, 7.5 meV and 12.5 meV, which vary in intensity depending on the channel; (iii) a phonon peak at 16-17 meV, clearly identified by the polarization analysis. The magnetic peaks are expected to derive from a combination of spin anisotropy and oscillation modes (phason and tilt modes) of the helicoidal structure. The 3 meV peak coincides with an electromagnon mode observed in THz spectroscopy [5].

We are currently in the process of test a number of possible spin Hamiltonians against the data to identify the important interactions.

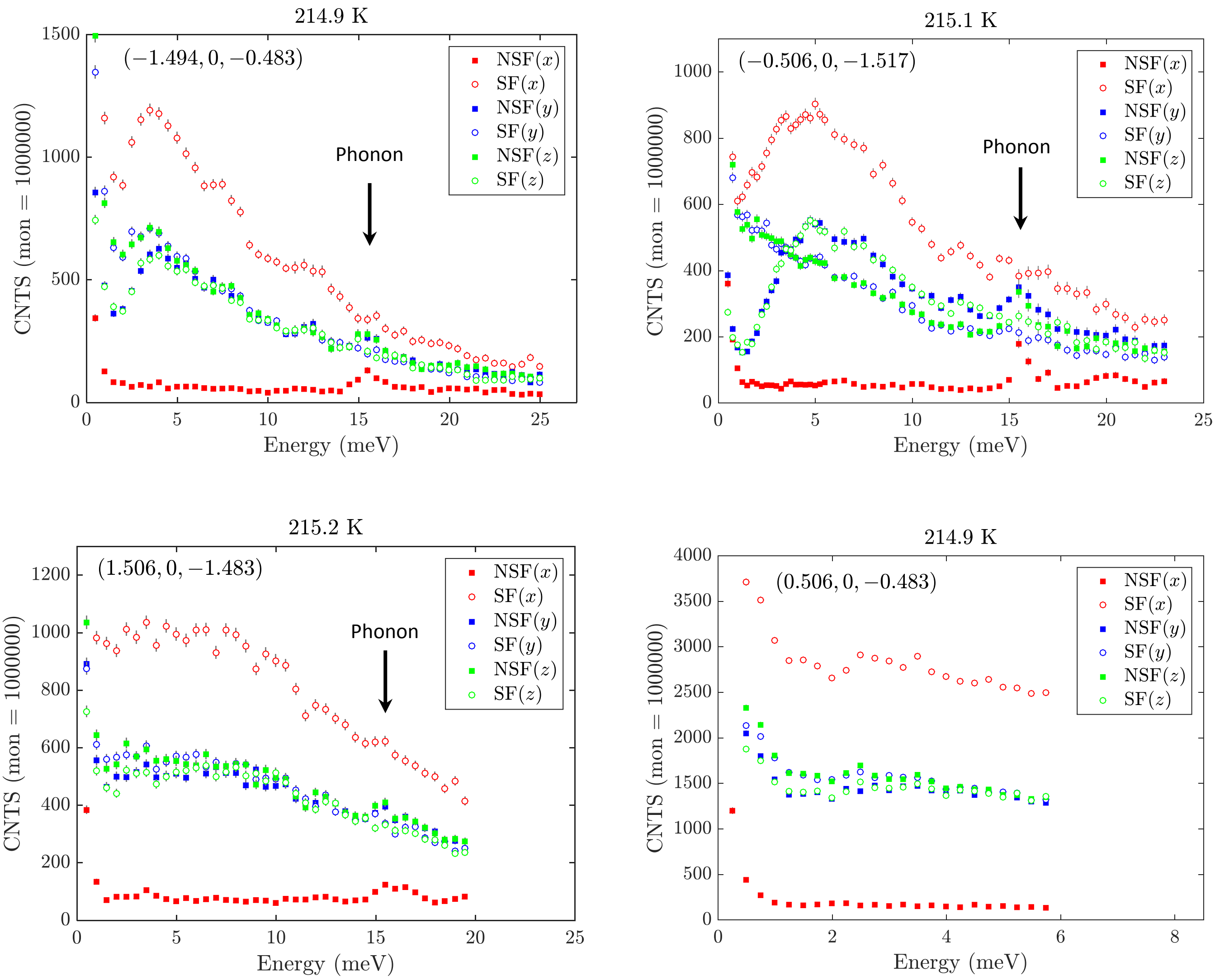

Figure 1. Neutron inelastic scattering with polarization analysis from CuO measured on IN20. The four panels show energy scans recorded at 215 K for four different helicoidal ordering wave vectors. A phonon peak at 16-17 meV is marked.

References

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

[2] J.B. Forsyth *et al.*, J. Phys. C **21**, 2917 (1988); P.J. Brown *et al.*, J. Phys. Condens. Matter **3**, 4281 (1991); M. Ain *et al.*, JPCM **4**, 5327 (1992).

[3] ILL Experimental Report on 4-01-1195.

[4] A. T. Boothroyd, Physica B **234–236**, 731 (1997); S.M. Gaw, DPhil Thesis, University of Oxford (2014).

[5] S.P.P. Jones *et al.*, Nature Comm. **5**, 3787 (2014).