Experimental report

27/10/2022

Proposal: **CRG-2810**

Council: 10/2020

Title: Search for electromagnons in CuO using polarized neutrons

Research area:

This proposal is a new proposal

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Samples: CuO

Instrument	Requested days	Allocated days	From	To
IN22	7	7	12/03/2021	22/03/2021

Abstract:

Experimental Report for experiment CRG# 2810

Search for electromagnons in CuO using polarized neutrons

- ## Scientific case:

The tenorite (CuO) represents the building block of high-Tc cuprates. It is believed to be the corner stone to the understanding of the properties of many Cu-O based compounds. In addition to its being an antiferromagnetic insulator over a large range of temperature, it presents a zoology of appealing properties. Indeed, it exhibits a high Tc magnetoelectric multiferroicity, which develops in the AF2 phase and remains up to 230K [1]. It was also shown to exhibit charge stripes ordering, which remains a challenging topic in the field of high Tc SC [2].

CuO undergoes, on cooling, a first magnetic transition from a paramagnetic state to an incommensurate helical antiferromagnetic order AF2 at T_{N2}=230K with a propagation vector of k=(0.505,0,-0.483). A second transition to a collinear antiferromagnetic order AF1 occurs at T_{N1}= 213K, where the magnetic moments are aligned along the b-axis, with a propagation vector (-0.5,0,0.5) [3].

Recently, a full inelastic neutron scattering (INS) study of the magnetic excitations spectrum of CuO allowed to reveal a two-spinon continuum of fractionalized spin excitations, in the AF1 phase, and a low-energy spin wave spectrum as the compound becomes more three dimensional [4].

In the context of multiferroicity, it was demonstrated that the application of a strong magnetic field in the AF2 phase leads to a suppression of the helical structure of spins which strongly impacts the electric polarization [5], emphasizing the strong character of the magneto-electric coupling in this compound. The application of an external pressure was further predicted to stabilize the multiferroic phase towards T>300K [6], which makes it a highly promising candidate in view of technological applications. As in manganite multiferroic compounds ($RMnO_3$, RMn_2O_5) [7], the strong coupling between the lattice and spin degrees of freedom results in specific excitations called "electromagnons" or electric-field active magnons that can be measured by means of INS. Indeed, theoretical works [8] predict the existence of two electromagnon modes lying at 3 and 13 meV in CuO. The former results from a Dzyaloshinskii-Moriya mediated mechanism while the second one is triggered by the Exchange- Striction. These modes couple to the electric field along [1,0,1] and [0,1,0], at 3 and 13 meV, respectively, in the AF2 phase.

The low energy mode was already observed using Terahertz (THz) Spectroscopy measurements [9] while no reports on the second mode were made, to date. Interestingly, authors in [4], report two unidentified magnetic optical modes at T=215K, at the same characteristic energies of 3 and 13 meV at Q=(3/2,0,3/2)−ε , with ε=(0.006,0,0.017) that could be ascribed to the magnetic scattering part from the predicted electromagnon modes. The 3 meV mode persists at Q=(1/2,0,3/2)+ε while no footprint of the 13 meV mode was found. This hints at a peculiar Q-dependence or to specific reciprocal space selection rules for this mode, especially since the corresponding electromagnon was initially predicted to couple to the electric field along the direction (0,1,0).

- ## Outcome of Exp CRG#2810

Our THz spectroscopy measurement at the AILES beamline of the SOLEIL synchrotron revealed an electro-active excitation within the expected energy range [10] and satisfying the selection rules for an electromagnon mode as expected from theory [8].

Based on the reported THz and INS measurements in CuO and the related theory of electromagnons in this compound, the aim of this experiment was to perform a full XYZ-longitudinal polarization analysis of the optical modes reported at 3 and 13 meV [4]. This was done by measuring the related scattering in the NSF and SF channels, at various Q-points in reciprocal space (of the form (H,0,L) or mixing (H,0L) and (0,K,0)) and at different temperatures.

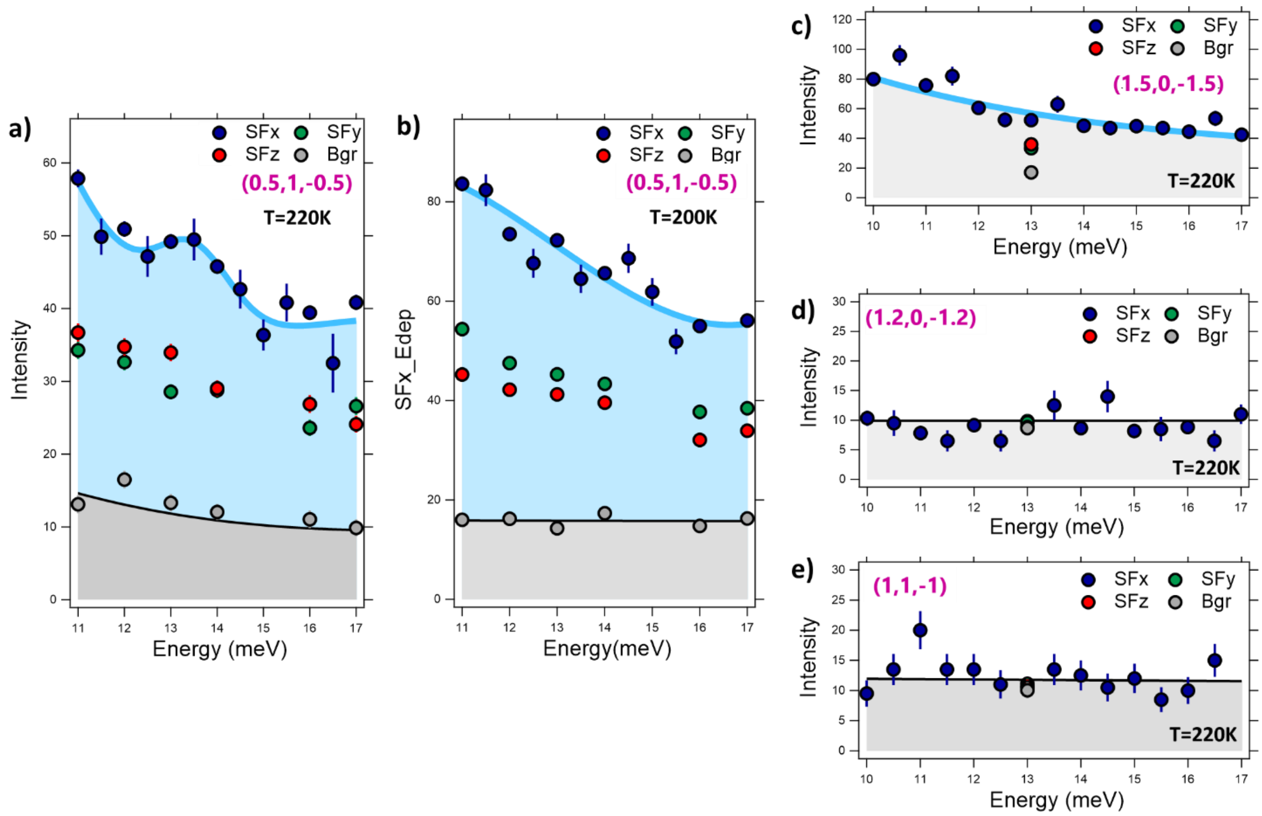

Figure 1. Energy scans at (0.5,1,-0.5) measured at **a)** 220K in the AF2 phase and **b)** at 200K in the AF1 phase. Energy scans at 220K obtained at several Q-points **c)** (1.5,0,-1.5), **d)** (1.2,0,-1.2), **e)** (1,1,-1).

We used a CuO single crystal grown by the travelling solvent floating zone technique, aligned in the (1,0,-1)/(0,1,0) scattering plane. We worked at a final neutron wavevector of k_f=2.662A^{-1}. The incoming and scattered beam polarization was ensured using Heusler crystals. The polarization analysis was achieved using CRYOPAD.

We collected several energy scans in the SF and NSF channels both in the AF1 (T=200K) and AF2 (T=220K) magnetic phases after identification of the related magnetic Bragg peaks.

Figure 1 shows the scans obtained in the SF channel at several Q values: (0.5, 1,-0.5), (1.5, 0,-1.5), (1.2,0,-1.2) and (1,1,-1), at 220K and 200K.

The energy scan at (0.5,1,-0.5) in the SFx channel (**Fig.1.a**) reveals a feature at 13 meV at T=220K in the AF2 phase while no similar feature is seen on scans collected at other Q-values (**Fig.1.c-e**), rather exhibiting a featureless energy dependence. Polarization analysis reveals that the signal is of magnetic origin and sits on top of a magnetic background.

In order to probe the evolution of the feature seen at 13meV as a function of temperature and determine whether it could be associated to the presence of an electromagnon mode, we repeated the same scan in the AF1 phase at T=200K (**Fig.2.b**) where it is expected to vanish. The scan collected at 200K reveals the absence of a similar feature at 13 meV with only a flat magnetic scattering level most probably arising from the two-spinon continuum of the CuO chains identified in earlier studies [4].

Further data is required to complement this study. Owing to the small intensity of magnetic scattering related to the mode at 13 meV, further lying over a strong magnetic background, the full temperature dependence of this mode could not be achieved. Indeed, owing to the strong magnetic background in either of the AF1 and AF2 phases, Q-scans at E=13meV are needed at several temperatures in order to estimate the precise temperature of appearance of this mode.

- **References**

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