Experimental report

09/03/2017

Proposal: **4-01-1499** **Council:** 4/2016

Title: Spin-polarized INS study of dynamic magnetoelastic coupling in CuBr2

Research area: Physics

This proposal is a continuation of 4-01-1467

Main proposer: **Yuan LI**

Experimental team: Biaoyan HU
Philippe BOURGES

Local contacts: Louis Pierre REGNAULT
Paul STEFFENS

Samples: CuBr2

Instrument	Requested days	Allocated days	From	To
IN12	6	6	06/09/2016	12/09/2016
THALES	6	0		

Abstract:

Type-II multiferroic spiral magnets are expected to possess strong magnetoelastic coupling and may host hybrid fundamental excitations that involve both spin and lattice degrees of freedom. A thorough and generic understanding of these effects will benefit from experimental studies of structurally simple material systems. CuBr2 was recently discovered to be a type-II multiferroic spiral magnet with a remarkably simple crystal and magnetic structure. Our previous spin-unpolarized INS measurements on single crystals of CuBr2 have revealed highly pronounced phonon anomalies at the dispersion intersecting points of phonons and spin waves, and our preliminary spin-polarized INS study of the spin waves have indicated a possible local minimum in the spin-flip intensity at the same Q-E locations. These results suggest the opening of a gap in the spin wave spectrum and the presence of strong hybridization between phonons and spin waves. Here we propose to verify the possible existence of a local minimum in the magnetic intensity at the phonon-spin-wave dispersion intersecting point, using the high-flux cold-neutron INS spectrometer ThALES (or IN12, depending on availability).

Spin-polarized INS study of dynamic magnetoelastic coupling in $CuBr_2$

Abstract: Copper (II) bromide $CuBr_2$ is a recently discovered type-II multiferroic material [1], with a high critical temperature T_N = 73 K below which a spiral magnetic order leads to the formation of ferroelectric polarization. An incommensurate spiral magnetic order is stabilized below T_N with a propagating wave vector Q_{AFM} = (1, 0.235, 0.5) [1], which corresponds to spin rotation between nearest neighbors of about 85 degrees. In our recent spin-polarized INS experiments of $CuBr_2$ single crystals performed on IN12, key information about the hybridizing mechanism between the spin and lattice degrees of freedom has been found.

Measurement condition: Large single crystals of $CuBr_2$ are grown from aqueous solution using a slow evaporation method. The co-aligned sample has a total mass of 12 g, mounted with Q vector (2L K L) in horizontal plane. K_f = 1.650 A^{-1} was fixed. Sample was in a cryostat, and the measurement was carried out at 20 K. We performed longitudinal polarization analysis (LPA) with Helmholtz coils at different Q positions. A velocity selector before the monochromator was used.

Calibration measurement: We chose (2L K L) to be set in the horizontal plane. We first tested the mosaic at (2 0 1) and (0 2 0) and the FWHM are both around 4°. Then we tested the SFz channel of (0 2 0) with 0 meV. After comparing, we chose the better mode (kept F2 off) to switch between SF and NSF, which had a flipping ratio of 17. The energy resolution and the momentum resolution were test. The FWHM of the energy scan at (1 0.1 0.5) was 0.24 meV and that of QK scan at (1 k 0.5) with 4.5 meV was 0.072. Comparing with those resolutions got from IN20 and 4F1, the energy resolution of IN12 was much better than the other two, which caters our hope.

	IN20	4F1	IN12
K_f (A^{-1})	2.662	1.97	1.65
E resolution (meV)	1.11	0.81	0.24
K resolution (b*)	0.089	0.043	0.072

Table 1 Comparison of energy and momentum resolution of IN20, 4F1 and IN12.

Experiment result: In our previous time-of-flight neutron experiment of $CuBr_2$, there is a phonon anomaly at around 7 meV where the acoustic phonon crosses the magnetic excitations at Q_{AFM} = (1 0.235 0.5) (Fig. 1a). Thus, the dynamic magnetoelastic coupling effect was expected at around 7 meV. In our previous polarized measurements of CuBr2 both on 4F1 of LLB and IN20, a dip was seen at 7 meV in the curve of energy scan of magnetic excitations at Q_{AFM} (1 0.235 0.5) (Fig 1bc). Because of the large error bars and the poor energy resolutions, neither find of the 7-meV dip was so convincing. That's why we proposed for a cold neutron experiment on IN12 this time, hopefully to get a better energy resolution, to study the magnetoelastic coupling more deeply.

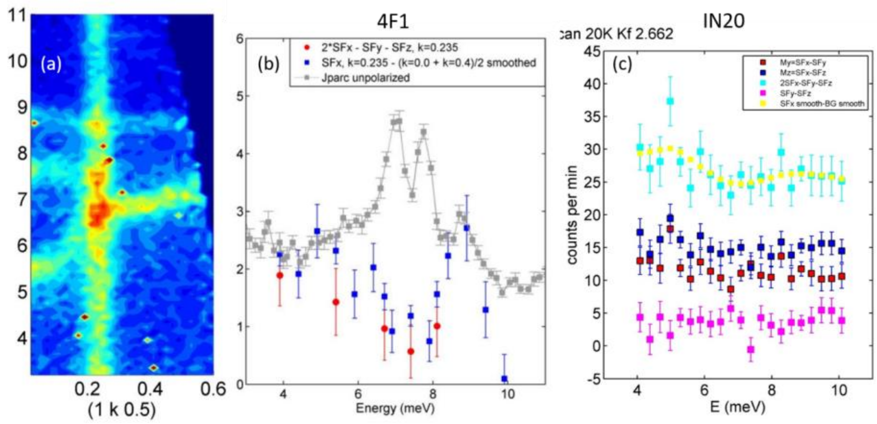

Fig. 1 (a) Unpolarized time-of-flight data got in J-PARC. (b) and (c) Polarization analysis of the data got on 4F1 of LLB and IN20.

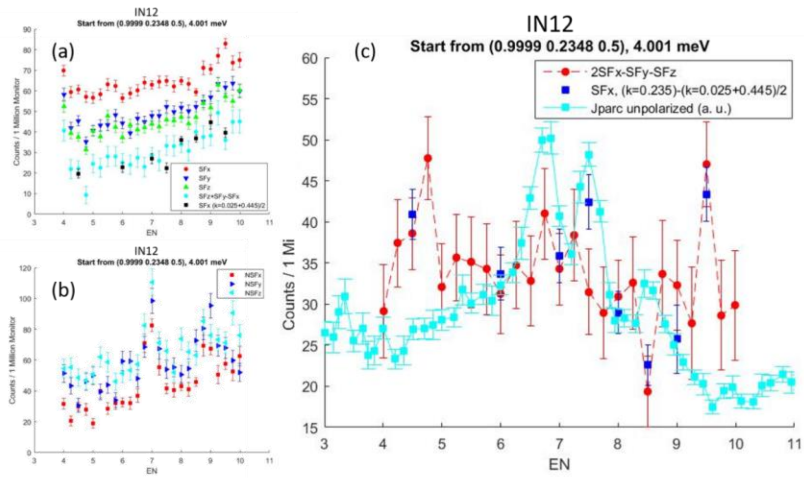

Fig. 2 (a) and (b) Spin-flip and non-spin-flip data of energy scans at Q_{AFM} on IN12. (c) Polarization analysis of the data got on IN12.

We performed energy scans at Q_{AFM} with LPA in the SFx, SFy, and SFz channels (Fig. 2a) and NSFx, NSFy, and NSFz channels (Fig. 2b) on IN12. We didn’t see any obvious dips near 7 meV (red line in Fig. 2c), but a minimum value at 8.5 meV in both two methods to remove the background. Polarized measurements of QK scans at (1 k 0.5) with eight different energies were performed. We focused on the SFx data of that, which are mainly the magnetic signals, and fitted them with Gaussian function (Fig. 3a). In Fig. 3c, we saw the fitting peak intensity is generally consistent with that got from polarization analysis that there is a minimum at 8.5 meV. The fitting FWHM is not the lowest at 8.5 meV but at 7.5 meV. In Fig. 1a, there are crossings of phonons and the magnetic excitations at both 7.5 meV and 8.5 meV other than 7 meV. We calculated the FWHMs time with fitting peak intensities as the approximate integral of magnetic excitations (Fig. 4) and found a dip area from 7.5 meV to 8.5 meV but a large value at 7 meV. Since there are three crossing at 7 meV, 7.5 meV and 8.5 meV, we thought there would be magnon-phonon hybridizations at either of the three energy. At each hybridization, some magnetic signals would break and connect to the phonons, which would reduce the magnetic signal at the middle of the crossing area and increase a little of that on both side of the crossing area that keeps the whole magnetic signal a constant. The result we got on IN12 is very similar to that if the hybridizations at 7.5 meV and 8.5 meV are both strong but that at 7 meV is weak. In that situation, the hybridization at 7 meV might be too weak to make a difference of that being beside a hybridization (7.5 meV) will have a large integral of magnetic excitations. So we think the hybridizations at 7.5 meV and 8.5 meV are both stronger than that at 7 meV. Considering of the error bars, we need more beam time to make our conclusion more convincing.

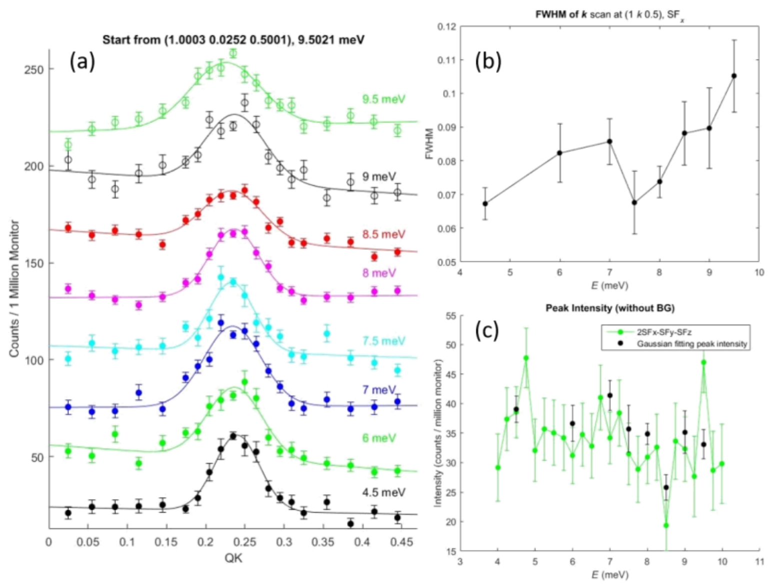

Fig. 3 (a) QK scans of SFx data at (1 k 0.5) with 8 different energies. (b) and (c) Fitting width and height with Gaussian function of the QK scans of different energies at Q_{AFM}.

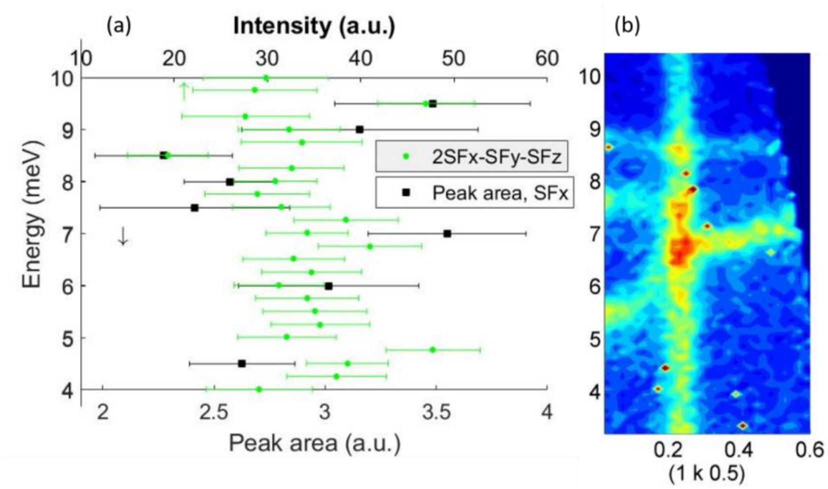

Fig. 4 (a) Polarization analysis of E-scan at (1 0.235 0.5) (green points) and fitted peak areas of the k-scans in Fig.3a (black square points). (b) Time-flight data got from J-PARC.

References:

[1] L. Zhao et al., Adv. Mater. 24, 2469 (2012).