実験報告書様式（一般利用課題・成果公開利用）

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Experimental Report	承認日Date of Approval　2015.11.05 承認者Approver Ryoichi Kajimoto 提出日Date of Report 2015.06.12
課題番号 Project No. 2014B0032 実験課題名 Title of experiment Spin waves in high-temperature multiferroic and model spin-1/2 chain compound CuBr2 実験責任者名 Name of principal investigator Yuan Li 所属 Affiliation Peking University, China	装置責任者 Name of responsible person Kazuki Iida and Kazuya Kamazawa 装置名 Name of Instrument/（BL No.） BL01 実施日 Date of Experiment Apr. 13-19, 2015

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。（適宜、図表添付のこと）
Please report your samples, experimental method and results, discussion and conclusions.　Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.

CuBr2 single crystals, co-mounted on aluminum plates

2. 実験方法及び結果（実験がうまくいかなかった場合、その理由を記述してください。）
Experimental method and results.　If you failed to conduct experiment as planned, please describe reasons.

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_{AF} = (1, 0.235, 0.5) [1], which corresponds to spin rotation between nearest neighbors of about 85 degrees. In our recent INS experiments of $CuBr_2$ single crystals performed at 4SEASONS, comprehensive information about magnetic excitations and the low energy phonon was obtained in the antiferromagnetic phase. Indeed we have found evidence for a strong coupling between magnons and phonons, which indicates the existence of hybrid excitations, evidenced by a sudden splitting of the acoustic phonon when crossing the incommensurate Q vector. This measurement will pave the way to our understanding of magneto-elastic coupling effects in $CuBr_2$.

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 14 g. The measurement was carried out at 15 K, much lower than the antiferromagnetic temperature. Ei was chosen as 150 meV with Fermi chopper frequency of 250 HZ. Because of the multi-Ei method, Ei= 55, 28 and 17 meV can be measured simultaneously.

Experiment result: As for the magnetic excitations, thanks to the high flux and multi-Ei method on 4SEASONS, we have successfully mapped out the spin-wave dispersion throughout the 4D Q-Espace. In Fig.1a-c, the study of the spin excitations in (h 0.235 l) plane reveals the anisotropy of the spin wave dispersion. The X shape in the energy slices (Fig.1a-c) is orginated from the twinning in our sample. The evolution of the X shape in different energy slices indicates different spin wave velocities along and perpendicular to Q_a. Fig.1d shows the magnetic dispersion spectrum along Q_b , integrated for Q_a and Q_c. We combined different energy ranges of data with different E_i and did some smoothing connections. An asymmetric magnetic dispersion can be observed starting from Q_b = 0.235 which is the incommensurability wave vector of the spiral magnetic ordering Two intense branches are labeled by the solid black lines. The left branch ends around 60 meV and the right branch extends to the top of the dispersion which falls into the energy around 80 meV. This is consistent with our linear spin wave calculations (Fig.1e).

Moreover, we have found evidence for a strong coupling between magnons and phonons, which indicates the existence of hybrid excitations. In Fig.2a the magnetic dispersion is observed around (1 0.235 0.5) with relatively high spin wave velocity. Besides the magnetic signal, there are several phonon branches across it. Unexpectedly, a sudden splitting of the 7 meV phonon happens when crossing the spin waves. This magnetism-induced phonon splitting is also observed in higher Brillouin zones, where the spin waves are no longer visible due to the reduction in magnetic form factor (Fig. 2b). More details are shown in Fig.2c with constant Qscans of the dispersion in Fig. 2b. At k = 0.31, there is only one peak around 7 meV. As the k value decreases and passes through 0.235, which is the incommensurability wave vector of the spiral magnetic ordering, the original sharp peak vanishes and is replaced by two peaks on either side, with no continuous evolution in between. These results strongly suggest that the phonon changes its eigen vector and gets possibly mixed with magnetic excitations when crossing the spin waves. This is also supported by our analysis in other parts of the Brillouin zone (not shown here), indicating that the strong coupling between magnons and phonons is associated with the quasi-1D spin correlations.

2. 実験方法及び結果（つづき） Experimental method and results (continued)

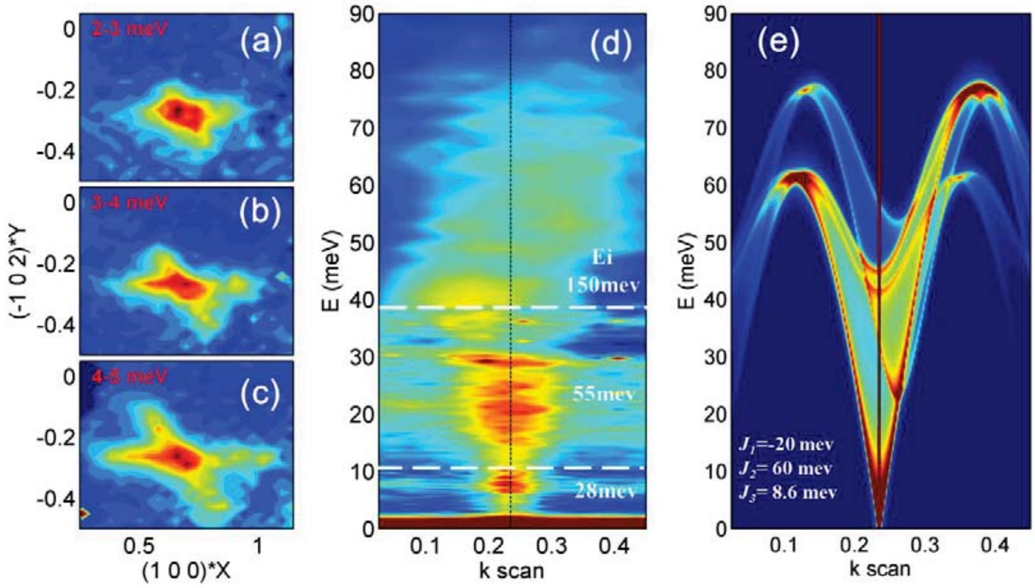

Fig. 1. (a),(b),(c) Energy slices of spin-wave dispersion with k = 0.235 for Q = (h,k,l), (d) Spin-wave dispersion along k integrated over a certain range in h and l, using data from measurements with different Ei. The intensity is multiplied by $\sqrt{E}$ to make the signal clearer for the high energy magnetic excitations and some smoothing is done at the connection of different Ei. (e) Linear spin-wave calculations integrated along h and l, with intrachain nearest-neighbour exchange J1 = −20 meV, next-nearest-neighbour exchange J2 = 60 meV and interchain spin exchange J3 = 8.6 meV. The measured spin waves are seemingly broader than in the calculation, probably due to pronounced quantum fluctuations in this spin-1/2 low dimensional system.

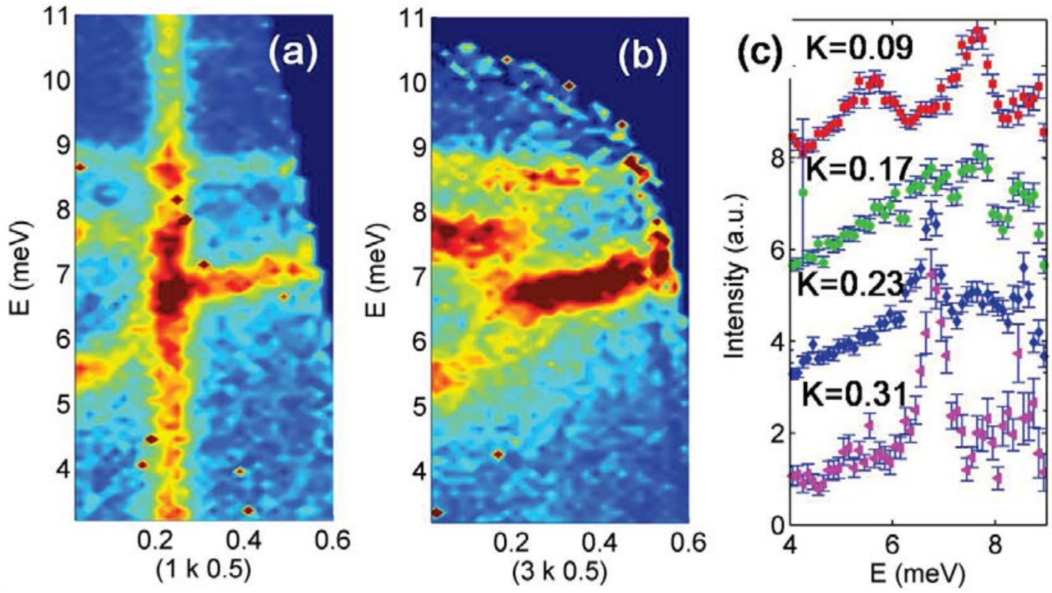

Fig. 2. (a) and (b) Time of flight data plotted as a function of E and Q, (c) Constant Q scans of Fig. 2b, offset for clarity.

References:

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