実験報告書様式（プロジェクト利用）

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実験課題番号 Project No. 2009A0073 実験課題名 Title of experiment: Magnetic excitations in quantum spin systems and frustrated magnets 実験責任者名 Name of principal investigator S. Kawamura[1], R. Kajimoto[1], K. Iida[2], K. Nakajima[1], H. Kageyama[3], K. Kakurai[1] 所属 Affiliation [1] JAEA, [2] CROSS, [3] Kyoto Univ.	装置責任者 Name of responsible person K. Nakajima, S. Kawamura 装置名 Name of Instrument/（BL No.） BL14 利用期間 Dates of experiments 2010/4/23 17:00～4/24 9:00; 2010/5/14 11:30-2010/5/18 13:00; 2010/5/18 13:00～5/20 9:00; 2010/6/21 13:00～6/22 13:00; 2010/10/19 23:30-2010/10/23 11:00; 2010/12/10 12:30-2010/12/12 13:00; 2010/11/19 13:00～11/22 13:00; 2011/2/6 13:00～2/8 17:00

1．研究成果概要（試料の名称、組成、物理的・化学的性状を明記するとともに、実験方法、利用の結果得られた主なデータ、考察、結論、図表等を記述してください。

Outline of experimental results (experimental method and results should be reported including sample information such as composition, physical and/or chemical characteristics.

Neutron chopper spectrometer is the best tool to investigate magnetic excitations in polycrystalline sample samples. Equivalent points in Q and E can be integrated so that data from all the detector points in position and time are fully utilized. Hence a very detailed and effective global investigation on magnetic excitations and thus a quick characterization of novel magnetic materials from microscopic dynamical point of view can be performed. To establish and demonstrate the power of the cold neutron chopper spectrometer at J-PARC MLF, we performed a series of ToF inelastic neutron scattering experiments on quantum and frustrated spin systems utilizing the newly installed AMATERAS cold neutron chopper spectrometer at J-PARC.

Magnetic excitations in the quasi-2-D triangular lattice antiferromagnet $CuCrO_2$.

About 3 cm^3 polycrystalline samples of $CuCrO_2$(CCO) and $Cu_{0.85}Ag_{0.15}CrO_2$(CACO) were synthesized by solid-state reaction method. Both compounds have hexagonal structures with an R-3m symmetry. The lattice constants are a=2.977Å and c=17.13 Å for CCO and a=2.982 Å and c=17.35 Å for CACO at RT. The delafossite oxide CCO is a 2D triangular lattice antiferromagnet with S=3/2 spins of Cr^{3+} ions forming the 2D triangular lattice. The Cr layers are separated from each other by Cu layers. The inter-planar coupling between the Cr layers is sufficiently strong in the pure CCO leading to a 3D magnetic ordering below TN=26K. The substitution of Cu ion with Ag ions induces crossover from 3D to 2D low-energy magnetic excitations and the large magnetic specific heat constant in CACO suggests that the 2D low-energy magnetic excitations are not conventional 2D antiferromagnetic magnons. Figure 1 shows a summary of the observed excitation spectra with Ei=15 meV for CCO and CACO. For CCO, the excitation spectrum shows two distinct features at 6K. Firstly it has a steep dispersive component at around Q=1.4 $Å^{-1}$, corresponding to the magnetic Bragg peak position of Q=(0.329,0.329,0) in the ordered phase and extends over 10meV. This dispersive component is attributed to the spin wave excitation originating from the spiral spin structure. Secondly it shows a weakly dispersive excitations at E~5-6meV distributed over a wide Q range. Its intensity decreases as Q increases indicating that it is of the magnetic origin. With increasing T, the dispersive component becomes broad and intense, while the flat component becomes indistinguishable and disappears above T_N. In contrast, CACO has almost no flat component at 5K. The dispersive component is clearly observed at Q=(q,q,0) as in CCO, but is diffusive compared to the pure compound. One should note that the Q-E profile of CACO at 5K resembles that of CCO at 25K. These findings indicate that the ground state of CACO is very similar to that of CCO at $T \sim T_N$. There in CCO, most of the weight of the flat component is transferred to E~0, producing diffusive low energy fluctuations

1．研究成果概要（つづき） Outline of experimental results (continued).

in addition to disordered spin waves. This may be the origin of the large magnetic specific heat constant observed in CACO. Recently it has been pointed out that the spin liquid state should have gapless and diffusive excitations, which are similar to the diffuse component found in the present study. The present study thus may indicate the possible realization of a spin-liquid like state by temperature and the doping control of a quasi-2D triangular lattice antiferromagnet coexisting with the spin-ordered phase characterized by magnetic Bragg peaks.

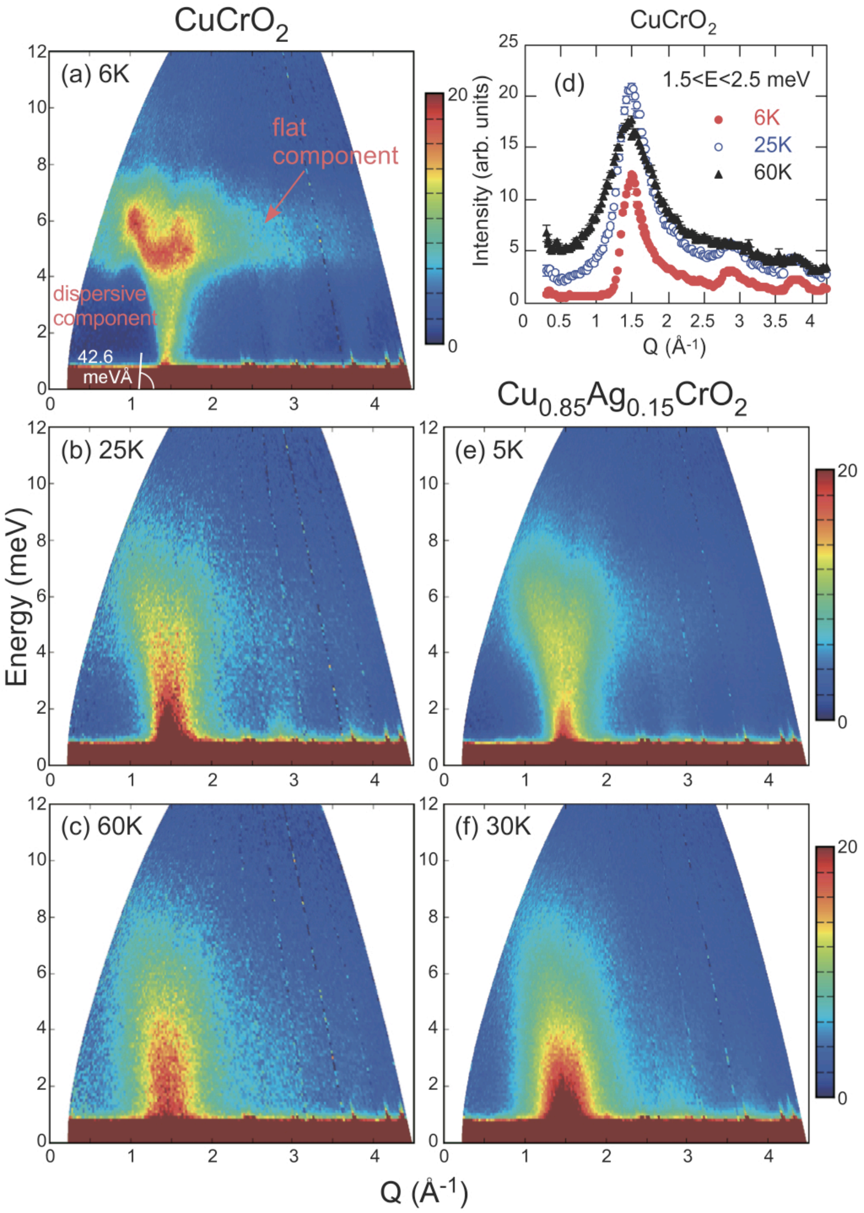

Fig. 1. (Color) Excitation spectra with $E_i = 15$ meV represented as Q–E maps for CuCrO$_2$ at (a) 6, (b) 25, and (c) 60 K, and for Cu$_{0.815}$Ag$_{0.15}$CrO$_2$ at (e) 5 and (f) 30 K. In (a), a line with a slope of 42.6 meV Å is also drawn. (d) Constant-E cuts of (a)–(c) at $E = 2 \pm 0.5$ meV.

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Magnetic excitations in square lattice quantum spin systems (CuCl)LaB_2O_7(B=Nb, Ta)

In (CuCl)LaB_2O_7 systems the magnetic Cu ions form a square lattice within the CuCl layer, separated by the LaB_2O_7 layers. Hence it has been expected that the system can be described by J1-J2 model with the competing nearest and next-nearest exchange interactions. The magnetic ground state of the 2-D system then depends on the ratio of J1 to J2. The ground state of (CuCl)LaNb_2O_7 has been suggested to be a spin-singlet state. On the other hand (CuCl)LaTa_2O_7 shows an antiferromagnetically ordered ground state with collinear spin structure below T_N=7K. The spin dynamical characterization of these two compounds should provide important information on the nature of the different ground states in these quantum spin systems. Since these samples are synthesized by the ion-exchange reaction, large volume single-crystalline samples for inelastic neutron scattering experiments are not available. For the experiments 10g and 17 g of powder samples of (CuCl)LaNb_2O_7 and (CuCl)LaTa_2O_7 , respectively, were packed in the cylindrically shaped Al foils of 10 mm diameter and 50mm height. Figure 2 shows the excitation spectra of (CuCl)LaNb_2O_7 and (CuCl)LaTa_2O_7 for different temperatures taken with E_i=7.7meV. In the spectra of (CuCl)LaNb_2O_7 a non-dispersive excitation with an energy gap of ~2meV and a bandwidth of ~1meV is observed indicating the existence of weakly coupled singlet ground state dimers. The analysis of the Q-dependent integrated intensity modulation using isolated dimer model yields an intra-dimer separation of ~8.7Å, corresponding to the fourth neighbor distance of 8.67Å. In the spectra of (CuCl)LaTa_2O_7 some what broader band-like excitation with inner structure and excitation branches extending down to lower energy region at wave vectors corresponding to the magnetic ordering vectors.

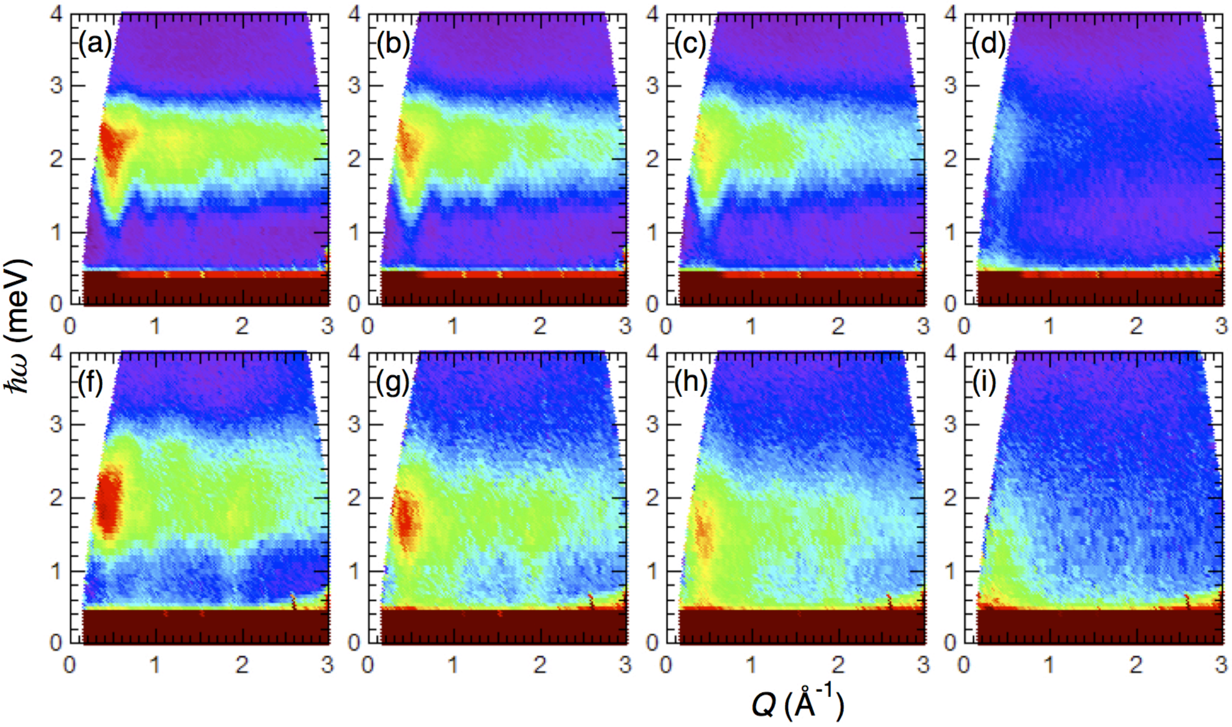

Fig 2 Excitation spectra in Q-E maps for (CuCl)LaNb_2O_7 measured at (a) T=4.7, (b) 7, (c) 9, and (d) 30 K, and for (CuCl)LaTa_2O_7 measured at (f) T=4.7, (g) 7, (h) 9, and (i) 30 K

The calculated spectrum for two dimensional coupled dimer systems with Heisenberg exchange interaction with parameters adjusted to describe the observed spectrum in (CuCl)LaNb_2O_7 is shown in Fig.3 a). Figures 3 b) and c) show the quantitative comparison of the observed and calculated spectra by looking at the energy and Q-dependence of the intensities integrated over Q range of 0.5 to 1.5 Å^{-1} and over energy range of 1.2 to 2.8 meV, respectively.

Correspondingly Fig. 4 a) displays the calculated spectrum of a two dimensional linear spin wave theory with parameters adjusted to the observed spectrum in (CuCl)LaTa_2O_7 with the ordered collinear antiferromagnetic structure where the magnetic propagation vector is k=(1/2,0). Figures 4 b) and c) show the quantitative comparison of the observed and calculated spectra by looking at the energy and Q-dependence of the intensities integrated over Q range of 0.5 to 1.5 Å^{-1} and over energy range of 1.2 to 2.8 meV, respectively.

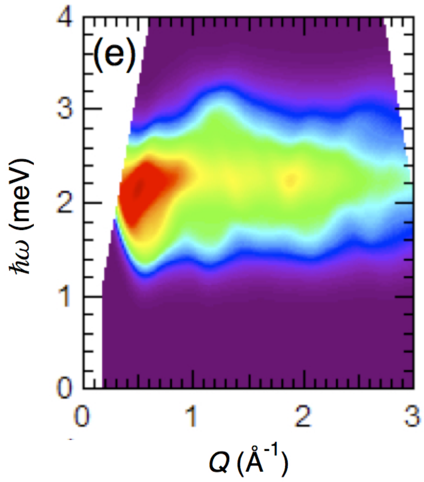

Fig 3 a)

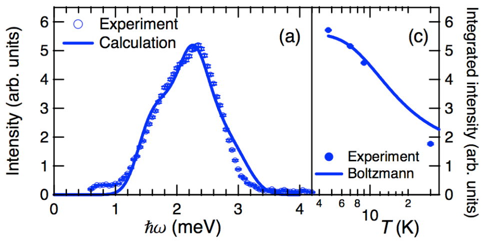

Fig 3b)

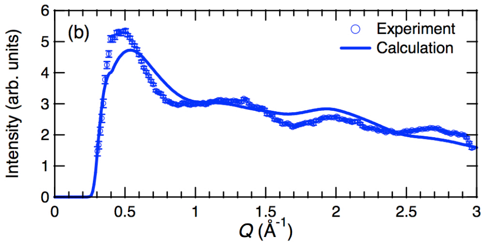

Fig. 3c)

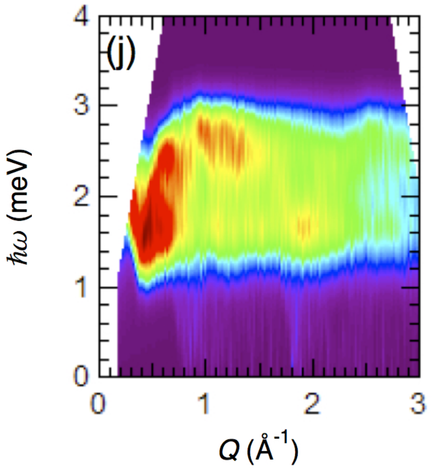

Fig 4a)

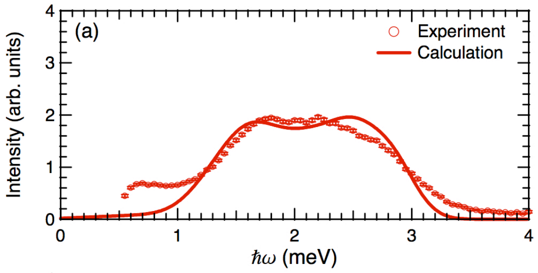

Fig 4b)

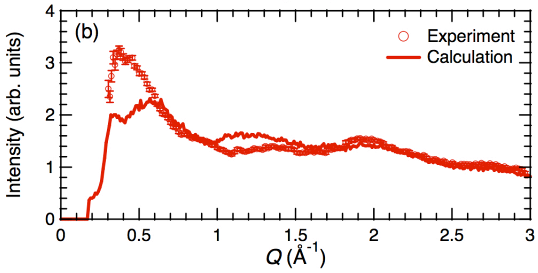

Fig. 4c)

These results clearly indicate that the main features of the spin fluctuations in the two compounds with different ground states are well captured in the powder averaged inelastic neutron scattering results and that the relevant exchange parameters can be derived from these measurements. At the same time it is obvious that the enhanced low energy fluctuations at the ordering wave vector, as observed in the 2-D triangular spin lattice antiferromagnet $CuCrO_2$, is not visible here, indicating that the quantum criticality in $(CuCl)LaB_2O_7$ is apparently not due to the change in the degree of frustration in the 2-D square lattice, as have been originally proposed. It is rather of a three dimensional character, i.e. the transition from the weakly coupled dimer system with singlet ground state to spin ordered state due to 3-D inter-dimer coupling without changing the degree of frustration. The difference in the temperature dependence may reflect the different origins of the quasi-band gap. In $(CuCl)LaNb_2O_{7,}$ the gap originates from a true spin gap singlet ground state with strong intradimer interaction, while in $(CuCl)LaTa_2O_7$ the center of the gravity of the quasi-band is determined by the relatively weak three-dimensional exchange energies of the order of T_N, leading to a considerable renormalization in the temperature range investigated.

The above examples clearly demonstrate the power of the inelastic magnetic scattering experiment with polycrystalline samples with advanced ToF spectrometer at pulsed neutron source such as J-PARC/MLF. The overall characterization of novel magnetic systems, especially the parametric studies for substitution effects, can be very efficiently and effectively performed utilizing the vast coverage of Q and E space with multiplex incident energy data collections realized in the modern ToF chopper instrument.