論 文 の 内 容 の 要 旨

論文題目　Novel magnetic excitations in spin systems investigated by neutron scattering
（中性子散乱によるスピン系の新奇な磁気励起の研究）

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1. Preface

The importance of a spin system is an easy access to the verification of theories for non-trivial phenomena. The effective experimental tool is neutron scattering which probes static and dynamical states of matter. In this thesis, I focus on two spin systems; (1) singlet ground state magnet $CsFeCl_3$ and (2) Kagome antiferromaget $NaBa_2Mn_3F_{11}$. $CsFeCl_3$ exhibits a pressure-induced quantum phase transition (QPT), and Higgs mode is expected in the spin spectrum. $NaBa_2Mn_3F_{11}$ is a classical Kagome antiferromagnet, and a zero-energy mode is expected.

2. Pressure-induced quantum phase transition in singlet ground state magnet $CsFeCl_3$

Introduction

$CsFeCl_3$ is an easy-plane type antiferromagnet having a quantum disordered state. Magnetic Fe^{2+} ions form the 1D chain along the crystallographic *c*-axis, and the chains form the triangular lattice in the *ab*-plane. The low-energy excitation is described by a pseudo-spin s = 1 due to the cubic crystal field and spin-orbit coupling. Moreover, the pseudo-spin s = 1 is split into $s^z = 0$ singlet ground state and $s^z = \pm 1$ doublet state due to the easy-plane type single-ion anisotropy $D(s^z)^2$. The system, thus, realizes the singlet ground state. Inelastic neutron scattering (INS) study at ambient pressure and at zero magnetic field revealed that the spin system was ferromagnetic chains that were weakly coupled by antiferromagnetic interchain-interaction [1]. Very recently, it was found that the pressure induced a magnetic long-range order at low temperatures [2]. The result indicated that applying pressure effectively enhanced the spin interaction, suppressed the anisotropy gap, and induced a QPT.

In the quantum field theory, collective excitation near the quantum critical point (QCP) in the ordered phase is described by the phase and amplitude fluctuations of an order parameter. While the phase fluctuation is known as Nambu-Goldstone (NG) mode, the amplitude one is called Higgs mode, which is analogue to Higgs boson in particle physics. I aim to observe the Higgs mode and study the quantum criticality in $CsFeCl_3$ by neutron scattering technique under pressures. In contrast with excitations in pressure-induced ordered state in a spin-dimer compound $TlCuCl_3$ [3], unconventional excitation including non-degenerated NG- and Higgs-modes are expected [4].

Results

Neutron diffraction (ND) experiments under pressures were carried out at the single crystal diffractometer ZEBRA in PSI. New peaks are observed in addition to the nuclear Bragg peaks. Their intensities increase with the decrease of the temperature, meaning that they are the magnetic Bragg peaks. The peaks are indexed by a magnetic propagation vector $\boldsymbol{k}_{\mathrm{mag}}$ = (1/3,1/3,0). A 120° structure which is proposed by the representation analysis gives a good agreement with the experimental data. The estimated size of the magnetic moment at 1.3 GPa and at 1.5 K is 2.6 μ_B. The critical exponent of the order parameter is estimated to be β = 0.20(1), which is consistent with the one for a XY stacked triangular antiferromagnet β = 0.22 [5]. This criticality belongs to the universality class of U(1)×Z_2, in which the chiral liquid state is expected [6]. $CsFeCl_3$ can be, thus, a candidate to realize the chiral liquid state.

INS experiments under pressures were performed at the high resolution chopper spectrometer HRC in J-PARC/MLF and the cold-neutron triple axis spectrometer CTAX in ORNL. In the INS spectrum at ambient pressure, a dispersive magnetic excitation having a gap is observed, which is consistent with the previous report in Ref. [1]. The low-lying excitations are suppressed by applying pressure and softened toward the critical pressure P_c ~ 0.9 GPa. Calculation of the dispersion in the disordered phase evidences that applying pressure tunes the easy-plane anisotropy and spin interactions. At 1.4 GPa in the ordered phase, a well-defined excitation with a gap of 0.6 meV and a gapless continuum-like excitation are observed at (1/3,1/3,0). The magnetic excitation in the ordered phase is calculated by the bond operator theory [7]. The calculation exhibits the mixed mode of transverse and longitudinal fluctuations (T+L-mode) in addition to purely gapless transverse (T) and gapped longitudinal (L) modes. The T- and L-modes correspond to the NG- and Higgs-mode, respectively. The T+L-mode is caused by the non-collinearity of the magnetic structure. The careful comparison with the experimental results reveals that the observed well-defined gapped excitation corresponds to the non-trivial T+L-mode, and the gapless continuum-like excitation is the NG-mode.

3. Ground state and magnetic excitation in Kagome antiferromaget $NaBa_2Mn_3F_{11}$

Introduction

Geometrically frustrated magnets have attracted great interest because of the exotic orders at low temperatures. In case of the classical Kagome Heisenberg antiferromagnet, the highly-frustrated geometry causes macroscopic degeneracy and a 120° structure is realized by order-by-disorder mechanism [8]. In addition, the large degeneracy of the 120° structure at the ground state generates continuous arrangements of spins, and it forms excitation modes of zero energy [9], which is called zero-energy mode. In real compounds, the zero-energy mode appears as an excited state lifted by additional terms including magnetic anisotropy and/or Dzyaloshinskii-Moriya interaction. To my knowledge, the zero-energy mode in Kagome antiferromagnets was reported only in a potassium iron jarosite $KFe_3(OH)_6(SO_4)_2$ [10], and further study in different material is important.

$NaBa_2Mn_3F_{11}$ is a new model compound for a Kagome antiferromagnet. Mn^{2+} ion carries spin S = 5/2 and the MnF_7 pentagonal bipyramids form a Kagome lattice in the crystallographic *ab*-plane. The heat capacity and magnetic susceptibility exhibited antiferromagnetic transition at T_N = 2 K [11]. The Curie-Weiss temperature θ_{CW} was estimated to be -32 K which is smaller than those of most Kagome lattice magnets. The exchange interactions in $NaBa_2Mn_3F_{11}$ are, thus, relatively small, and the magnetic dipole-dipole (MDD) interaction may be important. Recent theoretical study in a classical Kagome antiferromagnet having the MDD interaction demonstrated the unusual 120º structure with a tail-chase geometry and the existence of the zero-energy mode [12]. In order to discuss the role of the MDD interaction in $NaBa_2Mn_3F_{11}$, I investigate the magnetic structure and magnetic excitation by the ND and INS technique.

Results

Powder ND experiments were carried out at the powder neutron diffractometer ECHIDNA in ANSTO and the long-wavelength time-of-flight (TOF) diffractometer WISH in ISIS. At least eight additional peaks are observed below T_N = 2 K in the ND experiments. The peak intensities increase with the decrease of the temperature. This means that a magnetic long-range order occurs at T_N = 2 K, which is consistent with the previous heat capacity measurement. From indexing the magnetic peaks, three magnetic propagation vectors are found; commensurate vector $\boldsymbol{k}_0$ = (0, 0, 0), and incommensurate (IC) vectors $\boldsymbol{k}_1$ = (0.3209, 0.3209, 0) and $\boldsymbol{k}_2$ = (0.3338, 0.3338, 0). In the magnetic structure analysis, I tested the models of the magnetic structures based on the representation analysis. As the result of the Rietveld refinements, I find that the 120º structure of the tail-chase geometry, which is modulated by the IC vectors, is realized.

To calculate the ground state of the system including the MDD interaction, I use the Luttinger-Tisza-type theory, and investigate the eigenvalues and eigenvectors of the interaction matrix in the wave vector space. From the calculation, the minimum eigenvalue is found at $\boldsymbol{k}$ = 0, and six degenerated states are lifted by the MDD interaction. The calculated structure of the ground state is the 120º structure of the tail-chase geometry. The magnetic structure with $\boldsymbol{k}_0$ in $NaBa_2Mn_3F_{11}$ is, therefore, selected by the MDD interaction.

A powder INS experiment was performed at the cold-neutron TOF spectrometer IN6 in ILL. A magnetic excitation is observed below 2 meV. The spectrum shows a strong excitation at 0.2 meV and weak excitation at 1.3 meV. The spectrum at 0.2 meV is comparatively non-dispersive, and has a strong intensity at 1.0 Å^{-1}, which is coincides with the position of the magnetic Bragg peak. The peak is split at high energy region, implying that a dispersive spin wave excitation occurs from 1.0 Å^{-1}. A magnetic excitation based on the linear spin wave theory is calculated, where the nearest neighbor exchange interaction and the nearest neighbor MDD interaction are considered. The calculation exhibits the low energy non-dispersive mode in addition to the two dispersive modes. Comparison with the obtained spectrum in the experiment reveals that the observed non-dispersive spectrum is quantitatively explained by the zero-energy mode lifted by the MDD interaction. I, thus, found interesting roles of the MDD interaction in the Kagome antiferromaget; the selection of the tail-chase 120º structure and the lift of the zero-energy mode.

4. Conclusions

In $CsFeCl_3$, the ND experiments under pressures evidenced that the magnetic long-range order of the 120º structure was induced by applying pressure. In the INS experiments under pressures, I succeeded in the observation of softening of the gap excitation. Combination of the experiment and calculation revealed that the non-trivial T+L-mode and the gapless continuum-like excitation do exist in the XY triangular antiferromagnet in the vicinity of QCP.

In $NaBa_2Mn_3F_{11}$, the 120º structure selected by the MDD interaction was identified through the powder ND experiment and the calculation of the ground state. The INS spectrum suggested the existence of the zero-energy mode lifted by the MDD interaction. It is concluded that $NaBa_2Mn_3F_{11}$ is a rare material realizing the classical Kagome antiferromagnet having the MDD interaction.

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