Magnetism of S=1/2 Square-Lattice Antiferromagnets (CuX)LaNb2O7 (X=Cl, Br)

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Magnetic systems usually achieve long-range order of spins at low temperatures. However, completely different behaviors can be seen in low-dimensional antiferromagnetic (AFM) quantum spin systems. Due to quantum fluctuations, the systems may not order long range even at 0 K. A characteristic of the spin liquid ground state is an energy gap in the magnetic excitation spectrum. Among them, the S = 1/2 frustrated square lattice AFM system is of special importance in the light of Anderson's resonating valence bond (RVB) concept to explain high-Tc superconductivity in doped cuprates [1].

We discovered that a double-layered Dion-Jacobson phase (CuCl)LaNb2O7 can be a very good candidate for the 2D quantum square lattice system. As shown in Fig. 1, the crystal structure is tetragonal (space group P4/mmm), where 2D Cu square lattices are well separated with each other by nonmagnetic double perovskite slabs. Our bulk susceptibility and zero-field inelastic neutron scattering experiments revealed that this system indeed provides a new class of two-dimensional (2D) Heisenberg spin systems that has a spin-singlet ground state with a finite energy gap of 2.3 meV [2, 3]. The magnetic susceptibility of (CuBr)LaNb2O7 is also prepared from topotactic ion-exchange reactions. In contrast to the spin-liquid behavior in the Cl sample, the Br sample shows an antiferromagnetic ordering at 32 K [4], despite nearly identical structural parameters. The neutron diffraction experiment demonstrated that the magnetic structure is described by a stripe ordering or a collinear ordering with a propagation vector of q = (1/2 0 1/2). This observation implies that the next-nearest bond (J2) is dominant over nearest-neighbor bond (J1), as theoretically suggested for the square-lattice J1-J2 model. This is the 4th experimental example of the stripe ordering. Mixed ferromagnetic nearest-neighbor and antiferromagnetic next-nearest-neighbor interactions are of comparable strength (J1/kB= -35.6 K and J2/kB= 41.3 K), placing the system in a more frustrated region of the stripe (collinear) phase than ever reported.

In order to reveal the origin of the anomalous critical field, we have performed in 2005 and 2006 neutron scattering measurements on the copper chlorine system with fields up to 5 T, using the C1-1 spectrometer installed at the guide hall of JRR-3M and the 5 T superconducting magnet. The scattering intensity as a function of energy transfer shows that the 2.3 mode splits into three mode. This is the first direct experimental observation of the softening of the triplet excitation in this compound. The subsequent experiment at higher magnetic field performed on the DCS at the NCNR further supported this view.

We successfully obtained the solid-solution series between (CuCl)LaNb2O7 and (CuBr)LaNb2O7, (CuCl1-xBrx)LaNb2O7 (0<x<1), which allows us to investigate the phase diagram between the spin liquid state and collinear ordered state. Inelastic neutron scattering experiments on the 5%-Br substitute sample, i.e., CuCl0.95Br0.05)LaNb2O7 were carried out on the ISSP-PONTA triple-axis spectrometer installed at a 5G beam port of JRR-3M at the Japan Atomic Energy Research Institute. A powder sample of 12.6 g was put into an aluminum cylinder. Most of the data were collected using a fixed final energy Ef of 14.7 meV (kf = 2.67 Å-1) and a horizontal collimation of open-40'-

S-80′-80′ in combination with a pyrolytic graphite (PG) filter placed after the sample to eliminate higher-order beam contaminations. As shown in Fig. 1, the neutron diffraction evidences the long-range magnetic ordering of the collinear type. It means that 5% Br substitution for the Cl site is enough to induce the long range ordering. A similar impurity-induced antiferomagnetic order from the spin-singlet ground state has been observed in the 1D spin-Peierls system CuGeO3 and 1D spin-ladder system SrCu2O3, suggesting that this phenomena is a universal behavior, occurring not only for 1D but also 2D system.

This work has been supported by Grants-in-Aid for Young Scientists (A) (No. 17684018) and Science Research on Priority Area (No. 16076210) from MEXT of Japan, and by the 21st Century COE Program, Kyoto University Alliance for Chemistry.

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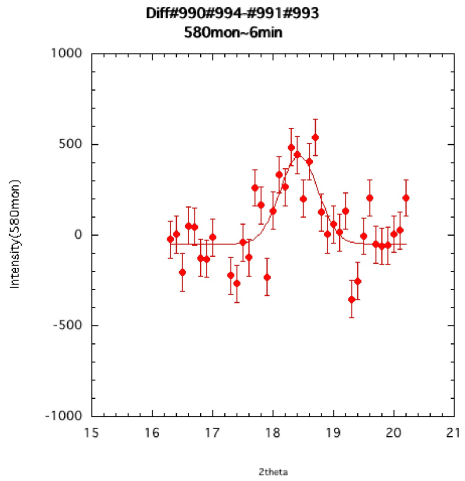

Fig. 1. Neutron diffraction patterns for (CuCl0.95Br0.05)LaNb2O7, suggesting the magnetic reflection at (1/2 0 1/2) at low temperature below 8K.