Magnetic excitation in the Ni_4 magnetic cluster

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The molecular magnet $[Mo^{V}_{12}O_{30}(\mu_2\text{-}OH)_{10}H_2\{Ni^{II}(H_2O)_3\}_4]\cdot 14H_2O$ (Ni_4) has four magnetic Ni^{2+} ($S = 1$) ions forming an isolated tetrahedral cluster. Earlier macroscopic experiment suggests dominant antiferromagnetic intracluster interactions between the Ni^{2+} spins, with negligible single-ion anisotropy [1]. Thus the Ni_4 was expected to be a good realization of Heisenberg antiferromagnet with strong geometrical frustration effect due to tetrahedral spin arrangement. Owing to the small S, small number of spins involved, and the geometrical frustration, strong quantum effects may be expected in the Ni_4 magnetic cluster.

Two grams of deuterated powder sample of Ni_4 was prepared following the prescription given in the earlier report [1]. The neutron inelastic scattering experiments were performed at the cold-neutron triple-axis spectrometer ISSP-HER installed at JRR-3, JAEA, Tokai. Superconducting magnetic was used to apply vertical magnetic field up to 5 T. The lowest attainable temperature was 1.5 K. A brand-new horizontally focusing analyzer was used with all seven blades operational, and a Be filter is placed in front of the sample to remove the higher order harmonics. Several final energies were used to collect the inelastic spectra depending on the contradicting necessity for the energy resolution and intensity; typically $E_f = 4.5$ meV.

Figure 1 shows inelastic neutron scattering spectra at $Q = 0.6$ Å^{-1} under the various magnetic fields. (Each spectrum is shifted by 400 counts to increase visibility.) In a zero filed spectrum, there is a peak at $\hbar\omega = 0.6$ meV appearing as a sholder of the huge elastic peak due to the incoherent scattering of the remaining hydrogen. Also seen is a peak at 1.3meV accompanied by a sholder peak at 1.6 meV. The magneic origin for these peaks was confirmed by measuring their temperature and Q dependence. Applying the magnetic field, the peak at 0.6 meV becomes broad, the peak at 1.3 meV shifts to the lower energy side, though the peak at 1.6 meV exhibits negligible change. These chagnes can hardly be explained consistently by the Zeeman effect.

We have also performed inelastic neutron scattering experiments using the DCS spectrometer at NIST Center for Neutron Research. Combining the all $S(Q, \hbar\omega)$ obtained at the two spectrometers, we are now working on the determination of the spin Hamiltonian in this system.

[1] A. Muller it et al., Inorg. Chem. bf 39 (2000) 5176.

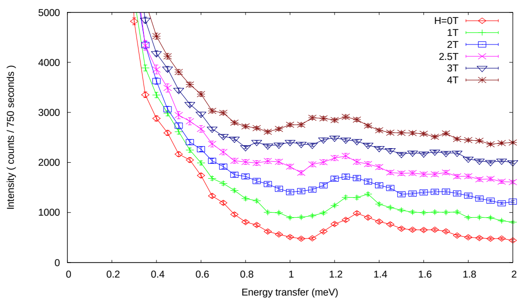

Fig. 1. Inelastic spectra of Ni_4 under various magnetic field observed at ISSP-HER. See text for details.