Magnetic correlation and excitation in triangle and hexagonal magnetic molecules

Kazuki Iida, Keisuke Hukaya[A], Hiroshi Ishikawa[A], Gou Kamohara[A], Toshihiro Yamase[A], Taku J Sato

ISSP, [A]Tokyo Institute of Technology

Single molecular magnets attract special attention recently, as they provide a new playground for studying isolated (or weakly coupled) quantum systems. Here, we have investigated four molecular magnets, V_3, Mn_6Sb, Mn_6As and Cu_6Sb using inelastic neutron scattering. These experiments are rather preliminary, as we look for systems of which spin Hamiltonian may be determined by inelastic neutron scattering. Chemical formulae of those systems are $K_{11}H[(VO)_3(SbW_9O_{33})_2] \cdot 27H_2O$ and $K_{12}[(VO)_3(BiW_9O_{33})_2] \cdot 29H_2O$, $[nBuND_3]_{12}[Mn_6Cl_6(SbW_9O_{33})_2] \cdot 6D_2O$, $[nBuNH_3]_{12}[Mn_6Cl_6(AsW_9O_{33})_2] \cdot 6D_2O$ and $[nBuNH_3]_{12}[Cu_6Cl_6(AsW_9O_{33})_2] \cdot 6D_2O$ respectively. V_3 forms a triangle cluster, whereas hexagonal cluster geometry was realized in the other systems. From the bulk magnetic susceptibility measurements, V_3 and Mn_6Sb were found to be anitferromagnetic, whereas physical properties of other systems have not been reported to date.

Powder samples of 0.71 g V_3, 0.34 g Mn_6Sb, 0.36 g Mn_6As, and 0.24 g Cu_6As were used in the neutron experiments. The neutron inelastic scattering experiments were performed at the cold-neutron triple-axis spectrometer ISSP-HER installed at JRR-3, JAEA, Tokai. 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 $E_f = 3$ meV. The V_3 experiment was performed at 0.9 K using the 1 K refrigerator, whereas the other experiments were done at 4 K using the Orange cryostat.

Inelastic spectra from the four samples at $Q = 0.6$ Å^{-1} are shown in Fig. 1. Data acquisition durations for each point were 20 minutes in the V_3 experiment, 4 minutes in the Mn_6Sb, 6 minutes in Mn_6As, 8 minutes in Cu_6As, respectively. In the figure, the spectrum of V_3 is shifted by 800 counts, Mn_6Sb by 800 and Mn_6As by 400. In the V_3 spectra there appears a peak at $\hbar\omega = 0.8$ meV at the lowest temperature. It is found that the peak intensity decreases as Q becomes large, and that it disappears at the high temperature $T = 30$ K. These results indicate that this peak has a magnetic origin. There also appears an slight shoulder at $\hbar\omega = 0.4$ meV, although it is difficult to conclude due to insufficient statistics. For Mn_6Sb, we only observed a symmetric peak around the elastic position, and thus it most likely originates incoherent elastic scattering of hydrogen or a nuclear bragg peak. The spectrum of Mn_6As exhibits a broad shoulder in an energy range of $0.2 < \hbar\omega < 0.5$ meV. Its temperature dependence suggests that this shoulder has also a magnetic origin. The spectrum of Cu_6As shows only incoherent scattering from hydrogen.

A continuous study is apparently necessary to elucidate their spin Hamiltonians. In future, we will focus on V_3 and Mn_6As where existence of the magnetic scattering was suggested in the present results.

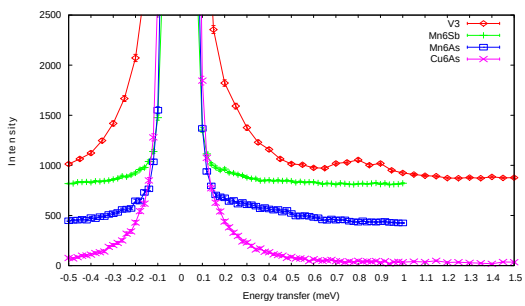

Fig. 1. Inelastic spectra of V_3, Mn_6Sb, Mn_6As, and Cu_6As observed at ISSP-HER. See text for details.