Spin dynamics in novel Rare-earth based single-molecule magnets

Takashi Kajiwara[1], Motohiro Nakano[2], Maiko Kofu[3], Osamu Yamamuro[3]
[1]*Nara Women's Univ.*, [2]*Osaka Univ.*, [3]*ISSP, Univ. of Tokyo*

Single-molecule magnets (SMMs) are a class of metalorganic compounds, which exhibit hysteresis of magnetization upon external magnetic field even though they have no long-range cooperative interactions. This behavior originates in a large magnetic moment and uniaxial magnetic anisotropy ($D < 0$), which gives rise to a double-well potential of the spin-up and the spin-down states and relaxation phenomenon between them. In recent years, not only thermal activation processes but also quantum tunneling ones are reported in SMMs. However, the mechanism of quantum tunneling process has not been fully understood yet.

To date, SMMs containing multiple transition metal atoms such as Mn, Fe, and Ni, have been intensively studied. Most recently, a new series of rare-earth based SMMs was discovered and attracts much attention. Because of large contribution of angular momenta, lanthanide complexes can become SMMs containing only one or two magnetic ions, being simpler than the transition metal SMMs and suitable for fundamental studies. Recently, we have succeeded to synthesize SMMs consisting of Tb ($J = 6$) and Cu ($S = 1/2$) ions (see Fig. 1(a)) and found that the system switch from SMMs (complex 1, 2, 4) to non-SMMs (complex 3, 5) by slight structural modification around the Tb ions. From dc magnetic susceptibility data, we suppose that non-SMM complexes do not have easy-axis anisotropy but easy-plane one. Our purposes are (i) to determine the parameters of Hamiltonian by inelastic neutron scattering (INS) measurements and (ii) to investigate the magnetic relaxation by quasi-elastic neutron scattering (QENS) measurements.

We have performed INS measurements for complex 1 on AGMES in the standard mode (FWHM = 0.12 meV). As seen in Fig. 1(b), a clear excitation peak was observed at 1.7 meV. We expect that this peak corresponds to the excitation between $|6, 1/2\rangle$ and $|6, -1/2\rangle$ states. Assuming an exchange coupling between Tb and Cu spins, the exchange coupling is estimated to be 0.28 meV. The INS experiments for non-SMM complex are now in progress.

We have also carried out QENS measurements for complex 1 in the high-resolution mode (FWHM = 0.049 meV). Above 50K, we observed a QENS component. In order to confirm whether the QENS results from the magnetic relaxation, further experiments will be performed.

[1] T. Kajiwara, *et. al.*: Inorg. Chem., 47 (2008) 8604.

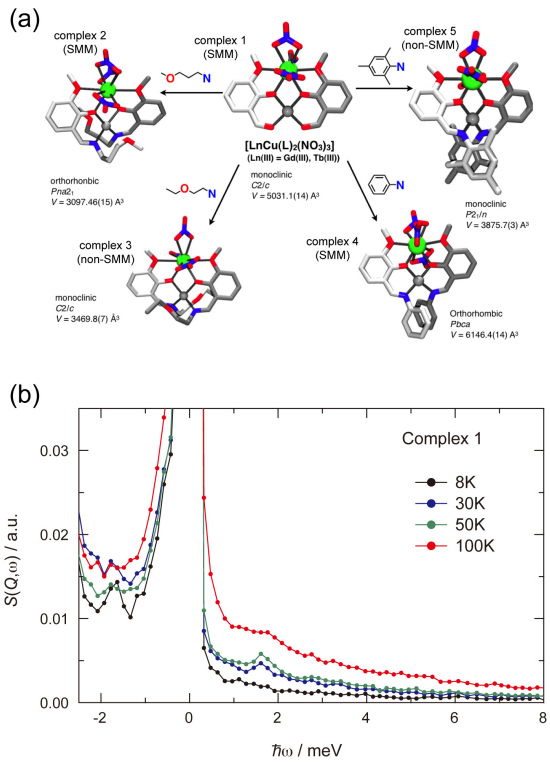

Fig. 1. (a) Molecular structure of Tb-Cu based compounds. (b) S(Q,ω) of complex 1 taken on AGNES.