Magnetic Excitation of $Ba_3Yb_2Zn_5O_{11}$ having perfect $S = 1/2$ teterahedra

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Geometrically frustration in magnetic materials disturbs the development of the magnetic long-range order and induces non-trivial and novel states. For example in $S = 1/2$ Heisenberg pyrochlore lattice the theoretical predictions on the ground state are controversial. While pioneering work reported quantum spin liquid exhibiting partial dimer order and associated spin gap [1], later work reported gapless excitation due to collective deformation of the dimers in addition to the massive excitation [2]. Recent work reported possibility of chiral spin liquid [3,4]. One of the popular techniques for the pyrochlore lattice is perturbative expansion [1,2] and its experimental realization can be found in so-called Breathing Pyrochlore lattice [5], where small and large tetrahedra are alternatively arrayed. $Ba_3Yb_2Zn_5O_{11}$ is a model material with the intra-tetrahedra distance of 3.29 Åand the inter-tetrahedra distance of 6.24 Å. Yb^{3+} ions are presumed to carry the effective $S = 1/2$ spins [6]. The magnetic susceptibility of $Ba_3Yb_2Zn_5O_{11}$ at $T >$ 30 K was explained by crystal electric field (CEF) Hamiltonian. The ground state was doublet and the eigenenergy of the first excited state with four-fold degeneracy was 523 K, which means the magnetic properties at low temperatures is dominated by pseudo-spin $S = 1/2$ [6]. Indeed the susceptibility at $T < 30$ K was reproduced by a perfect $S = 1/2$ tetrahedral cluster Hamiltonian with the intra-tetrahedra interaction J = 0.6 meV. Heat capacity measurements on $Ba_3Yb_2Zn_5O_{11}$ showed absence of magnetic long-range order down to 0.3 K.

To identify the CEF Hamiltonian and effective spin Hamiltonian, we performed the inelastic neutron scattering (INS) measurements at MARI in ISIS. By the INS measurements focused on high energy scale, three excitations are observed at 40, 55, and 70 meV and it is qualitatively consistent with the energy spectrum of the point group C_{3v} of the YbO_6 local structure; four Kramer's doublets. The energy of the 1st excited state is consistent with that estimated by the magnetic susceptibility measurement and Yb^{3+} ions are regarded as pseudo-spin $S = 1/2$ at low temperatures. In the INS spectrum at 1.7 K in low energy scale, strong and broad excitation is observed at 0.5 meV and weak and sharp excitation is observed at 0.8 meV. In addition collective-like excitation is observed at $Q \sim 0.8$ Å^{-1} and 1.5 Å^{-1}. The observed spectrum is inconsistent with the perfect $S = 1/2$ tetrahedral cluster proposed by the bulk measurement, since the matrix elements of the neutron cross section for the cluster system have finite values only for the transition between nonmagnetic ground state and the 1st excited state with $S = 1$, i.e., $\hbar\omega = 0.6$ meV in this case, at the base temperature. We tried to analyze the data by the tetrahedral cluster including XXZ-type anisotropy and Dzyaloshinskii-Moriya interaction. The data, however, cannot be reproduced by the cluster model after all. Now we believe that inter-tetrahedra interaction plays important role and the system is regarded as Breathing Pyrochlore.

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