Hybridized magnon with orbital and lattice in multiferroics Ba₂MnGe₂O₇

S. Hasegawa¹, S. Hayashida¹, S. Asai¹, M. Matsuura², and <u>T. Masuda^{1#}</u>

¹ Institute for Solid State Physics, The University of Tokyo, Chiba 277-8581, Japan ²Neutron Science and Technology Center,

Comprehensive Research Organization for Science and Society, Ibaraki 319-1106, Japan

a corresponding author: E-mail masuda@issp.u-tokyo.ac.jp

A spin-driven multiferroics has been extensively studied since the discovery of an enhanced magnetoelectric effect in TbMnO₃. Among microscopic mechanisms of the multiferroics, a spin-dependent *d-p* hybridization mechanism locally determines the relationship between an electric polarization and spin moment on each magnetic site [1]. In this case, a magnetic anisotropy plays a key role in the characterization of the multiferroicity. A remarkable work in Ba₂CoGe₂O₇ demonstrated that the magnetic anisotropy is explained by a spin-nematic interaction which is equivalent to an electric polarization interaction [2]. The interaction may lead to a nontrivial magnon coupled with orbital and lattice.

A multiferroics Ba₂MnGe₂O₇ is isostructural to Ba₂CoGe₂O₇, where Mn²⁺ ions carrying Heisenberg spin S = 5/2 form square lattice [3]. An antiferromagnetic order with a collinear structure occurs at 4 K, and simultaneously an electric polarization appears [4]. The magnetic susceptibility and the inelastic neutron scattering (INS) spectra were explained by a square-lattice classical Heisenberg model, where the magnetic anisotropy was not considered.

Here we study the temperature variation of the magnetic anisotropy by combination of a magnetization measurement and an ultra-high energy resolution INS experiment. We observed a pair of well-defined magnon modes, T₁ for lower and T₂ for higher energies. The band energies are consistent with the previous report [3]. A temperature evolution of anisotropy-gap energies, E_{g1} for T₁ and E_{g2} for T₂ modes were measured. The energy of E_{g2} at 1.8 K is consistent with a conventional easy-plane anisotropy reported in the previous ESR study [5], which means that T₂ mode is lifted by the easy-plane anisotropy. We found that E_{g1} is scaled by the electric polarization, but it cannot be scaled by any power of the sublattice moment. Since E_{g1} is also scaled by the measured spin flop field, the T₁ mode is lifted by an easy-axis anisotropy. E_{g2} is, in contrast, scaled by the sublattice magnetic moment. Through the analysis of INS spectra using spin wave theory, we found that the T₁ mode is lifted by the spin-nematic interaction, while the T₂ one is lifted by the easy-plane single-ion anisotropy. The anomalous temperature dependence of E_{g1} leads to a conclusion that the T₁ mode is a hybridized magnon with orbital and lattice.

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