Hybridized magnon with orbital and lattice in multiferroics

**Ba$_2$MnGe$_2$O$_7$**

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A spin-driven multiferroics has been extensively studied since the discovery of an enhanced magnetoelectric effect in TbMnO$_3$. 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 Ba$_2$CoGe$_2$O$_7$ 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 Ba$_2$MnGe$_2$O$_7$ is isostructural to Ba$_2$CoGe$_2$O$_7$, where Mn$^{2+}$ 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, $T_1$ for lower and $T_2$ for higher energies. The band energies are consistent with the previous report [3]. A temperature evolution of anisotropy-gap energies, $E_{g1}$ for $T_1$ and $E_{g2}$ for $T_2$ 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 $T_2$ 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 $T_1$ 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 $T_1$ mode is lifted by the spin-nematic interaction, while the $T_2$ one is lifted by the easy-plane single-ion anisotropy. The anomalous temperature dependence of $E_{g1}$ leads to a conclusion that the $T_1$ mode is a hybridized magnon with orbital and lattice.