Realization of giant magnetoelectricity in helimagnets

Sae Hwan Chun,[1] Yi Sheng Chai,[1] Yoon Seok Oh,[1] Deepshikha Jaiswal-Nagar,[1] So Young Haam,[1] Ingyu Kim,[1] Bumsung Lee,[1] Dong Hak Nam,[1] Kyung-Tae Ko,[2] Jae-Hoon Park,[2] Jae-Ho Chung,[3] and Kee Hoon Kim[1]
[1] *FPRD, Department of Physics and Astronomy, Seoul National University, Seoul 151-747, South Korea*
[2] *Department of Physics & Division of Advanced Materials Science, POSTECH, Pohang 790-784, South Korea*
[3] *Department of Physics, Korea University, Seoul 136-713, South Korea*

We show that low field magnetoelectric (ME) properties of helimagnets $Ba_{0.5}Sr_{1.5}Zn_2(Fe_{1-x}Al_x)_{12}O_{22}$ can be efficiently tailored by Al-substitution level. As x increases, the critical magnetic field for switching electric polarization is systematically reduced from ~1 T down to ~1 mT, and the ME susceptibility is greatly enhanced to reach a giant value of 2.0 $\times$ 10^4 ps/m at an optimum $x = 0.08$. We find that control of nontrivial orbital moment in the octahedral Fe sites through the Al-substitution is crucial for fine tuning of magnetic anisotropy and obtaining the conspicuously improved ME characteristics.

PACS numbers: 75.80.+q, 77.80.Fm, 71.45.Gm, 75.30.Gw

In recent years, extensive researches on multiferroics have been performed with motivations to understand the nontrivial cross-coupling mechanism between magnetism and ferroelectricity as well as to search for new materials applicable in next-generation devices [1–8]. Numerous studies have focused particularly on the class of so-called magnetic ferroelectrics [1–3, 9, 10], in which ferroelectricity is induced by magnetic order through either inverse Dzyaloshinskii-Moriya effects [6–8] or the exchange striction mechanism [10]. Although dramatic variation of electric polarization P with magnetic field B, often realized in the magnetic ferroelectrics [3], might be useful for application, the phenomena occur mostly at low temperatures [1, 2, 9, 10] and related ME susceptibility is yet too small [5]. Hence, it is a longstanding challenge in the research of multiferroics to improve both the operating temperature [11, 12] and the ME sensitivity [13–15].

The hexaferrite $Ba_{0.5}Sr_{1.5}Zn_2Fe_{12}O_{22}$ (BSZFO) with helical spin order is currently a unique candidate that can show the B-induced ferroelectricity above room temperature up to ~340 K [12]. However, at 300 K, its ferroelectric (FE) phase is expected to emerge at B ~1 T, which is too high for memory applications. Moreover, its ME coupling is rather weak; the MES $\alpha_{\rm ME} \equiv \mu_0 \frac{dP}{dB}$ at 30 K shows a maximum value of ~1.3 $\times$ 10^3 ps/m at B ~400 mT [12], which is one or two orders of magnitude smaller than the highest $\alpha_{\rm ME}$~10^4 - 10^5 ps/m realized in heterogeneous films [13] or strain-coupled composites [5, 14]. When Zn is replaced by Mg to form a Mg_2Y-type hexaferrite, $Ba_2Mg_2Fe_{12}O_{22}$, the critical magnetic field for inducing P becomes extremely low, ~30 mT [15, 16]. On the other hand, a maximum operation temperature of the Mg_2Y-type hexaferrite is expected to be lower than 195 K. Moreover, a microscopic understanding for the lowered critical magnetic field remains unclear. Therefore, systematic studies are further required to understand the origin of the intricate ME effect and improve multiferroic properties in the hexaferrite system.

In this letter, we demonstrate that Al-substituted BSZFO, i.e. $Ba_{0.5}Sr_{1.5}Zn_2(Fe_{1-x}Al_x)_{12}O_{22}$ greatly improves the multiferroic characteristics, resulting in the highest $\alpha_{\rm ME}$ of single-phase multiferroics near-zero magnetic field. We find that predominant substitution of Al ions into octahedral Fe sites with nontrivial orbital moment is crucial for fine tuning of the magnetic anisotropy and thus for tailoring the ME coupling.

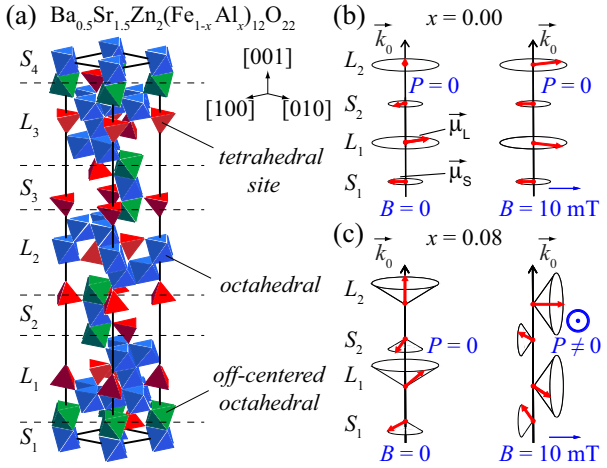

FIG. 1. (a) Crystal structure of the Zn_2Y type-hexaferrite $Ba_{0.5}Sr_{1.5}Zn_2(Fe_{1-x}Al_x)_{12}O_{22}$ that has alternating stacks of magnetic S and L blocks along the c-axis. Schematic illustration of rotating magnetic moments in the L ($\vec{\mu}_L$) and S blocks ($\vec{\mu}_S$) in the (b) helical ($x = 0.00$) and (c) heliconical ($x = 0.08$) phases under in-plane $B = 0$ T and 10 mT. $\vec{k}_0$ is the spin modulation wave vector parallel to [001].

Single crystals of $Ba_{0.5}Sr_{1.5}Zn_2(Fe_{1-x}Al_x)_{12}O_{22}$ were grown from Na_2O-Fe_2O_3 flux in air [17]. Crystals were cut into a rectangular form for electric polarization P measurements along the ab-plane while B was applied along the direction normal to the P vector in the ab-plane. Before the ME current measurements, each specimen was electrically poled in its paraelectric state, for