MAR15-2014-000444

Abstract for an Invited Paper
for the MAR15 Meeting of
the American Physical Society

Giant spin-driven ferroelectric polarization and magnetoelectric effect in perovskite rare-earth maganites under high pressure

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The discovery of ferroelectricity in $TbMnO_3$ triggered extensive studies on a type of multiferroics, “spin-driven ferroelectrics.” Unlike conventional ferroelectrics such as $BaTiO_3$, spin-driven ferroelectrics exhibit remarkable magnetoelectric (ME) effects. However, the ferroelectric polarization P in spin-driven ferroelectrics ever reported ($<10^{-1}\mu$C/cm^2) is much smaller than that in conventional ferroelectrics (typically $10^0 \sim 10^1\mu$C/cm^2). Thus, the quest for robust magnetically-controllable P comparable to that in conventional ferroelectrics is still a major challenge in the research on multiferroics. In this study, we utilized the “high-pressure” to attain a magnetically-controllable spin-driven P with its magnitude being comparable to that in conventional ferroelectrics [T. Aoyama *et al.*, Nature Commun. 5, 4927 (2014)]. With a home-made high-pressure measurement system with a diamond anvil cell, we investigated high-pressure effects on ME properties of perovskite RMnO$_3$ ($R =$ Gd, Tb, and Dy). Our study revealed that these manganites exhibit a pressure-induced ME phase transition and that the high-pressure phase shows the largest P (e.g., 1 μC/cm^2 in $TbMnO_3$) among spin-driven ferroelectrics ever reported. Moreover, P is further enhanced by applying a magnetic field. Our study demonstrates that it is possible to attain giant spin-driven ferroelectric polarization which comes close to that in conventional ferroelectrics, and to control it magnetically.

This work has been done in collaboration with T. Aoyama, K. Yamauchi, A. Iyama, S. Picozzi, A. Miyake, and K. Shimizu.