## Converse magnetoelectric effect in Co-based Heusler alloy/PMN-PT(011) multiferroic heterostructures

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In the previous reports, we demonstrated a giant converse magnetoelectric (CME) effect in the multiferroic heterostructures of Heusler alloy  $Co_2FeSi$  on ferroelectric  $Pb(Mg_{1/3}Nb_{2/3})O_3$ -PbTiO<sub>3</sub> (PMN-PT) substrate [1,2]. However, the origin of the giant CME effect has not been clarified yet. In this study, we conduct a comprehensive study of multiferroic heterostructures using several Heusler alloys on PMN-PT.

30-nm-thick Co-based Heusler-alloy thin films (Co<sub>2</sub>FeSi, Co<sub>2</sub>FeAl<sub>0.5</sub>Si<sub>0.5</sub>, Co<sub>2</sub>Fe<sub>0.4</sub>Mn<sub>0.6</sub>Si, and Co<sub>2</sub>MnSi) were grown on PMN-PT(011) substrates with the insertion of a 0.3-nm-thick Fe layer by molecular beam epitaxy. As a reference, ferromagnetic Fe<sub>3</sub>Si (Fe<sub>2</sub>FeSi) was also grown on PMN-PT(011). X-ray diffraction measurements exhibit that all Co-based Heusler films are grown in polycrystalline, and the  $L2_1$ -ordered structure is confirmed by the {111} diffraction peaks (not shown here).

The CME effect was characterized by in-plane magneto-optical Kerr-ellipticity  $(\eta)$  measurements and conventional magnetization measurements at room temperature. Prior to the  $\eta$  measurements, we firstly applied an electric field (E) of +8 kV/cm, then released to zeroelectric field for all heterostructures to prepare the same remanent polarization state of the PMN-PT(011). Figure 1 shows the in-plane magnetic-field (H) dependence of  $\eta$  for various Co-based Heusler alloys and Fe<sub>3</sub>Si on PMN-PT at E of 0 and -8 kV/cm. We found that all multiferroic heterostructures using the Co-based Heusler alloys provide the  $90^{\circ}$ changes from a hard axis to an easy axis by applying E within the in-plane magnetic anisotropy. For the Fe<sub>3</sub>Si case, on the other hand, a small variation in  $\eta$ -H curves can be seen where the magnetic easy axis observed at the zero-electric field is weakened at E of -8kV/cm. We will discuss the difference in such behavior between Co-based Heusler alloys and Fe<sub>3</sub>Si multiferroic heterostructures in detail to consider the origin of the giant CME effect in Co<sub>2</sub>FeSi/PMN-PT [1,2].



- [1] T. Usami et al., Appl. Phys. Lett. 118, 142402 (2021).
- [2] S. Fujii, T. Usami et al., submitted.



