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### **Quantitative investigation of magnetoelectric coupling in various forms of multiferroics<sup>1</sup>**

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Magnetoelectric susceptibility (MES) is probably the most direct way of estimating the magnitude of magnetoelectric coupling in many forms of magnetoelectric and/or multiferroic materials. Historically, the MES has been measured in numerous existing magnetoelectric materials in broad field, frequency, and temperature ranges and their MES values have been tabulated [1]. With growing interest worldwide toward applications of multiferroics for novel memory and sensor devices, however, there have been ever-increasing demands to measure quantitatively the MES of multiferroic thin films. Yet, the measurements of thin film MES become challenging in spite of its large MES value because the magnetoelectric voltages, proportional to the film thickness, usually get too small to be measured reliably. Herein, we introduce a highly sensitive magnetoelectric susceptometer that can detect the charge variation down to  $\sim 10^{-17}C$  in a few gauss oscillating magnetic field. Using this specific setup, we could measure the MES of multiferroic thin films or single crystals with unprecedented accuracy and sensitivity in cryogenic (down to 2 K) and magnetic field (up to 9 T) environments. In this talk, we summarize a number of key results based on this technique; (1) MES of a 300 nm BiFeO<sub>3</sub>-CoFe<sub>2</sub>O<sub>4</sub> nanopillar structure as well as those of a 250 nm BiFeO<sub>3</sub> film and of a BiFeO<sub>3</sub> single crystal. (2) MES of (Pb,Zr)TiO<sub>3</sub>-NiFe<sub>2</sub>O<sub>4</sub> nanocomposite films, and (3) temperature- and field-dependent MES in representative multiferroic crystals/films including TbMn<sub>2</sub>O<sub>5</sub>, GaFeO<sub>3</sub>, and Cr<sub>2</sub>O<sub>3</sub>. In particular, we demonstrate that the MES of the film with the nanopillar structure is enhanced by approximately one order of magnitude reaching  $210^{-10}$  s/m at room temperature, compared with those of a pure BiFeO<sub>3</sub> film and a single crystal. Furthermore, based on detailed field and temperature dependent MES studies, we show that magnetoelectric coupling in TbMn<sub>2</sub>O<sub>5</sub> has been mediated and amplified by the large magnetoelastic effect.

[1] G. A. Smolenskii and I. E. Chupis, Sov. Phys. Usp. 25, 475 (1982); F. W. Hehl *et al.* Phys. Rev. A 77, 022106 (2008).

<sup>1</sup>In close collaboration with Yoon Seok Oh, S. Crane, I. Takeuchi, R. Ramesh, T. W. Noh, S.-W. Cheong, and Albert Migliori.