## Nonlinear Electric Field Dependence of Electrocaloric Effect in (001)-epitaxial (Ba, Sr) TiO<sub>3</sub> Thin Films

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Recently, electrocaloric (EC) effect in ferroelectric thin films has been intensively studied as emerging cooling devices. Theoretically, the pyroelectric coefficient, which is a main factor determining the magnitude of EC effect, depends not only on the temperature but also on the electric field applied to the films. Therefore, EC effect will not be just proportional to the applied electric field. However, to the best of our knowledge, it has not been experimentally documented yet. In this study, we fabricated high-quality (001)-epitaxial (Ba, Sr) TiO<sub>3</sub> thin films, and experimentally estimated their EC effect through the measurements of pyroelectric coefficient for wide temperature and electric field ranges.

(001)-epitaxial  $Ba_{0.3}Sr_{0.7}TiO_3$  (BST30/70) and  $Ba_{0.5}Sr_{0.5}TiO_3$  (BST50/50) thin films with thickness of 570 nm and 610 nm were grown on SrRuO<sub>3</sub>/SrTiO<sub>3</sub>(001) at 700°C by radio frequency (RF) magnetron

sputtering. The phase transition temperature was estimated by the temperature dependence of the dielectric constant measured at 100 kHz. In order to precisely evaluate the pyroelectric coefficient, we measured the electric field dependence of polarization at various temperatures by using positive-up-negative-down method, which can eliminate the influence of leakage current. Based on the Maxwell relation [1], the temperature change caused by EC effect was calculated.

Figure 1 shows the temperature change,  $\Delta T$ , in BST50/50 thin film by EC effect as a typical result. As theoretically expected, the temperature  $T_{\text{max}}$  showing the largest  $\Delta T$  was close to the phase transition temperature  $T_0^*$ , and slightly increased with increasing applied electric field change  $\Delta E$  when  $\Delta E$  was below 115 kV/cm. However, it was found that  $T_{\text{max}}$  decreased with increasing  $\Delta E$  when  $\Delta E$  was above 148 kV/cm.



Fig. 1 Temperature dependence of temperature change,  $\Delta T$ , by various electric field changes,  $\Delta E$ , in BST50/50 thin film. Filled circles show the temperature  $T_{\text{max}}$  showing the largest  $\Delta T$ .

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