

Field-modulated thermopower in SrTiO₃-based field-effect transistors with amorphous 12CaO·7Al₂O₃ glass gate insulator

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We report transistor characteristics and field-modulated thermopower (S) for single crystal SrTiO₃-based field-effect transistors (FETs). We use 150-nm-thick amorphous 12CaO·7Al₂O₃ glass as the gate insulator of the SrTiO₃-FET. The resulting SrTiO₃-FET exhibits excellent transistor characteristics at room temperature: on-to-off current ratio greater than 10⁶, threshold gate voltage of +1.1 V, subthreshold swing of approximately 0.3 V decade⁻¹, and effective mobility of 2 cm² V⁻¹ s⁻¹. The field-modulated S -value of the SrTiO₃-FET varied from -900 to -580 μV K⁻¹ with electric fields of up to 2 MV cm⁻¹, demonstrating the effectiveness of the FET structure for the exploration of thermoelectric materials. © 2009 American Institute of Physics.

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Thermoelectric (TE) energy conversion technology has attracted a great deal of attention for converting waste heat into electricity by utilizing the Seebeck effect, as well as for refrigerating various devices by means of the Peltier effect.¹ The performance of thermoelectric materials is evaluated in terms of a dimensionless figure of merit ZT , given by $ZT = S^2 \cdot \sigma \cdot T \cdot \kappa^{-1}$, where T , S , σ , and κ are, the absolute temperature, the thermopower, and electrical and thermal conductivities, respectively. Today, TE materials with $ZT > 1$, which is necessary for practical applications, are being rigorously explored, mostly using charge carrier-doped semiconductors with various doping levels. Consequently, a great deal of material is needed to optimize TE properties.

A field-effect transistor (FET) structure on single-crystalline materials, however, would be a powerful tool in exploring TE materials because it provides the charge carrier dependence of both S and σ values simultaneously.² In this letter, we focus on SrTiO₃ to demonstrate the effectiveness of an FET structure for exploring TE materials. SrTiO₃ has attracted much attention as an n -type TE material due to its potential advantages over heavy metallic alloys such as Bi₂Te₃ and PbTe in chemical and thermal robustness.³

In order to unequivocally measure the field-modulated S values, SrTiO₃-FETs with excellent transistor characteristics are required. Although several attempts toward the realization of SrTiO₃-FETs have been made and several gate insulators including MgO,⁴ amorphous (a -)Al₂O₃,⁵ CaHfO₃,^{6,7} and parylene⁸ have been proposed to date, it is still difficult to fabricate SrTiO₃-FET with excellent transistor characteristics. For example, in our preliminary study, we first chose

a -Al₂O₃ as gate insulator because it can be deposited without any substrate heating using the pulsed laser deposition (PLD) process, which may minimize damage layer formation on the SrTiO₃ surface. However, several grain structures were observed in an atomic force microscope (AFM) image of the resulting 200-nm-thick a -Al₂O₃ film, which could cause relatively large gate leakage current. Furthermore, some of the transistor characteristics such as effective mobility (μ_{eff}) and subthreshold swing (S factor) were unsatisfactory, $\mu_{\text{eff}} \sim 0.6$ cm² V⁻¹ s⁻¹ and S factor ~ 2.4 V decade⁻¹ (data not shown).

We then tested various gate insulator materials to obtain SrTiO₃-FETs with superior transistor characteristics and found a -12CaO·7Al₂O₃ (Refs. 9–11) (a -C12A7, dielectric constant $\epsilon_r = 12$) to be an excellent gate insulator for SrTiO₃.¹² The SrTiO₃-FET exhibits excellent transistor characteristics at room temperature: on-to-off current ratio $> 10^6$, threshold gate voltage +1.1 V, subthreshold swing ~ 0.3 V decade⁻¹, and effective mobility ~ 2 cm² V⁻¹ s⁻¹. By applying a gate electric field of up to +2 MV cm⁻¹, the thermopower of the SrTiO₃-FET was modulated from -900 to -580 μV K⁻¹.

Schematic device structure and a photograph of the SrTiO₃-FET are shown in Fig. 1. First, 20-nm-thick metallic Ti films, used as the source and drain electrodes, were deposited through a stencil mask by electron beam (EB) evaporation (base pressure $\sim 10^{-4}$ Pa, no substrate heating) onto a stepped SrTiO₃ substrate ($10 \times 10 \times 0.5$ mm³), treated with NH₄F-buffered HF solution.¹³ Second, 150-nm-thick a -C12A7 film was deposited through a stencil mask by PLD (KrF excimer laser, fluence ~ 3 J cm⁻² pulse⁻¹, oxygen pressure ~ 0.1 Pa) using dense polycrystalline C12A7 ceramic as target. Finally, 20-nm-thick metallic Ti films, used as the

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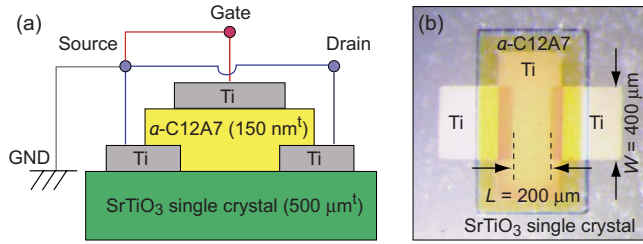


FIG. 1. (Color online) (a) Schematic device structure and (b) photograph of the SrTiO₃-FET. Ti films (20 nm thick) are used as the source, drain, and gate electrodes. A 150-nm-thick *a*-C12A7 film is used as the gate insulator. Channel length (*L*) and channel width (*W*) are 200 and 400 μm, respectively.

gate electrode, was deposited through a stencil mask by EB evaporation. After the deposition processes, the devices were annealed at 200 °C for 30 min in air to reduce the off current.

Halo peaking only at $2\theta \sim 30^\circ$ was observed in the glancing incidence x-ray diffraction (XRD) pattern of the resulting C12A7 film (Cu $K\alpha_1$, x-ray angle of incidence 0.5°) [Fig. 2(a)]. This pattern was very similar to the halo pattern of the C12A7 glass.¹¹ The density of the C12A7 film was $\sim 2.9 \text{ g cm}^{-3}$ as evaluated by grazing incidence x-ray reflectivity (GIXR, ATX-G, Rigaku Co., data not shown) and was in a good correspondence with that of C12A7 glass (2.92 g cm⁻³).¹¹ It should be noted that any grain structure in the as-deposited film was not observed in the topographic AFM image [Fig. 2(a) inset]. Furthermore, no grain structure was observed in the cross sectional high-resolution transmis-

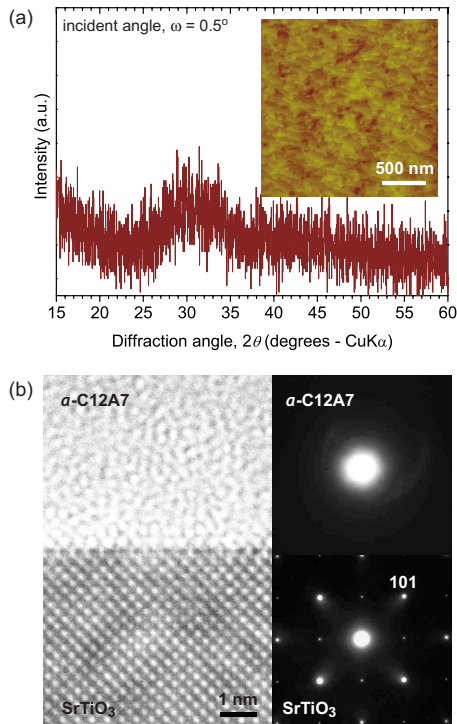


FIG. 2. (Color online) (a) Glancing angle XRD pattern of a 150-nm-thick *a*-C12A7 layer. Broad halo of *a*-C12A7 is seen around $2\theta \sim 30^\circ$. Topographic AFM image ($2 \times 2 \mu\text{m}^2$) of the C12A7 film on the SrTiO₃ surface is also shown in the inset. (b) Cross-sectional HRTEM image of the 150-nm-thick *a*-C12A7/SrTiO₃ heterointerface, showing an abrupt interface of *a*-C12A7/SrTiO₃. Featureless image of *a*-C12A7 clearly indicates that the *a*-C12A7 layer is glass. A broad halo pattern is seen in the selected area electron diffraction patterns of *a*-C12A7 (right).

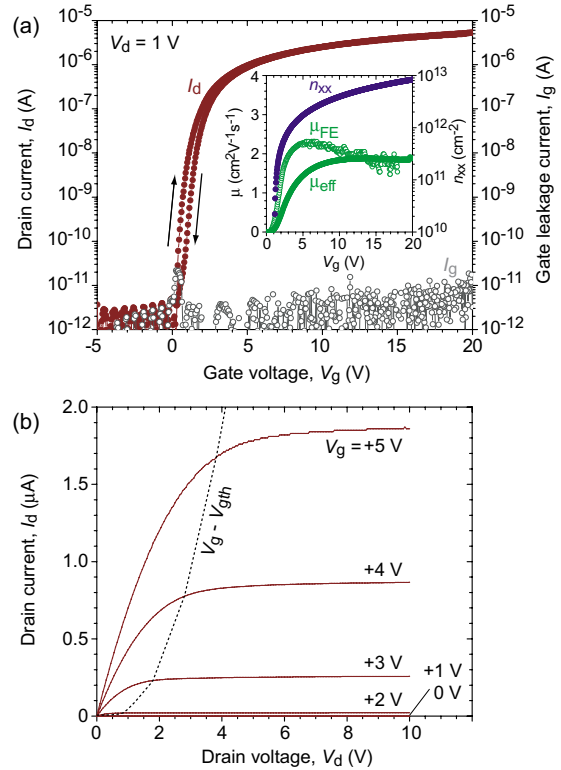


FIG. 3. (Color online) (a) Typical transfer and (b) output characteristics of the SrTiO₃-FET with 150-nm-thick *a*-C12A7 ($\epsilon_r = 12$) gate insulator at room temperature. Channel length (*L*) and channel width (*W*) are 200 and 400 μm, respectively. Effective mobility (μ_{eff}), field-effect mobility (μ_{FE}) and sheet charge density (n_{xx}) vs V_g plots for the SrTiO₃-FET are also shown in the inset of (a). The dotted line in (b) indicates $V_g - V_{\text{gth}}$ value.

sion electron microscope (HRTEM) image of the *a*-C12A7/SrTiO₃ interface region (TOPCON EM-002B, acceleration voltage of 200 kV, TOPCON) [Fig. 2(b)]. A broad halo pattern is seen in the selected area electron diffraction patterns of *a*-C12A7. We thus concluded that the deposited film was *a*-C12A7 glass.

Transistor characteristics of the resultant FETs were measured by using a semiconductor device analyzer (B1500A, Agilent) at room temperature in air. Figure 3 shows typical (a) transfer and (b) output characteristics of the resultant FET. Drain current (I_d) of the FET increased markedly as the gate voltage (V_g) increased, hence the channel was *n*-type, and electron carriers were accumulated by positive V_g [Fig. 3(a)]. A small hysteresis ($\sim 0.5 \text{ V}$) in I_d , probably due to traps (mid- 10^{11} cm^{-2}) at the *a*-C12A7/SrTiO₃ interface, was also seen [Fig. 3(a)]. We observed a clear pinch-off and saturation in I_d [Fig. 3(b)], indicating that the operation of this FET conformed to standard FET theory. The on-to-off current ratio, *S*-factor, and threshold gate voltage (V_{gth}), which were obtained from a linear fit of an $I_d^{0.5} - V_g$ plot (data not shown), are $> 10^6$, $\sim 0.3 \text{ V decade}^{-1}$ and +1.1 V, respectively. We calculated the effective mobility (μ_{eff}) of the SrTiO₃-FETs from $\mu_{\text{eff}} = (e \cdot n_{\text{xx}} \cdot R_{\text{xx}})^{-1}$, where e , n_{xx} , and R_{xx} were elementary charge, sheet charge concentration and sheet resistance, respectively. The n_{xx} values were obtained from $n_{\text{xx}} = C_i (V_g - V_{\text{gth}})$, where C_i was the capacitance per unit area (71 nF cm^{-2}) [Fig. 3(a) inset]. We noted μ_{eff} of the FET increases drastically with V_g and reaches at $\sim 2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, which is $\sim 30\%$ of the room temperature

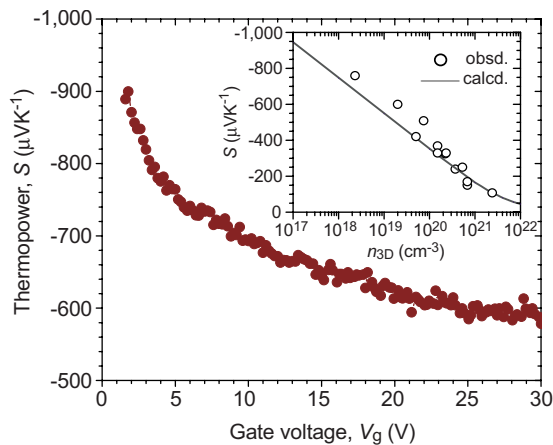


FIG. 4. (Color online) Field-modulated thermopower (S) for the SrTiO₃-FET channel. S - n_{3D} (log scaled) plots (Ref. 14) for electron doped SrTiO₃ are also shown in the inset as a reference.

Hall mobility of electron-doped SrTiO₃ ($\mu_{\text{Hall}} \sim 6 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) [Fig. 3(a) inset].

We measured field-modulated thermopower (S) of the SrTiO₃-FET as follows. First, a temperature difference ($\Delta T = 0.2$ – 1.5 K) was introduced between the source and drain electrodes by using two Peltier devices. Then, thermoelectromotive force (V_{TEMF}) was measured during the V_g sweeping. The values of S were obtained from the slope of $V_{\text{TEMF}} - \Delta T$ plots (data not shown). Figure 4 shows $S - V_g$ plots for the SrTiO₃-FET. The S values are negative, confirming that the channel is n -type. The $|S|$ value gradually decreases from 900 to 580 $\mu\text{V K}^{-1}$, which corresponds to an increase of the volume charge density (n_{3D}) from $\sim 2 \times 10^{17}$ to $\sim 8 \times 10^{18} \text{ cm}^{-3}$ (see Fig. 4 inset¹⁴), due to the fact that electron carriers are accumulated by positive V_g (up to +30 V).

In summary, we have shown transistor characteristics and field-modulated thermopower S of the SrTiO₃-based FET. The SrTiO₃-FET with 150-nm-thick amorphous 12CaO·7Al₂O₃ ($\epsilon_r = 12$) gate insulator exhibits excellent transistor characteristics at room temperature: on-to-off current ratio $> 10^6$, threshold gate voltage +1.1 V, sub-

threshold swing $\sim 0.3 \text{ V decade}^{-1}$, and effective mobility $\sim 2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. While the electric field was increased of up to 2 MV cm^{-1} , S was clearly modulated from -900 to $-580 \mu\text{V K}^{-1}$, demonstrating that the FET structure was indeed effective for the exploration of thermoelectric materials.

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¹²In our preliminary experiments, the dielectric constant (ϵ_r) of a -C12A7 was determined to ~ 12 , which was measured using a Ti/ a -C12A7/Ti capacitor deposited on SrTiO₃ substrate by a precision LCR meter (Model 4284A, Agilent). Typical breakdown field of the a -C12A7 film was $\sim 3 \text{ MV cm}^{-1}$, which corresponds to $n_{\text{ex}} \sim 2 \times 10^{13} \text{ cm}^{-2}$. The breakdown behavior of a -C12A7 is similar to that of PLD-deposited a -Al₂O₃ and a -LaAlO₃; S. Yaginuma, J. Yamaguchi, K. Itaka, and H. Koinuma, *Thin Solid Films* **486**, 218 (2005).

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