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ABSTRACT

Transparent amorphous oxide semiconductors (TAOSs) based transparent thin-film transistors (TTFTs) with high field effect mobility (μ_{FE}) are essential for developing advanced flat panel displays. Among TAOSs, amorphous (a-) SnO₂ has several advantages against current a-InGaZnO₄ such as higher μ_{FE} and being indium-free. Although a-SnO₂ TTFT has been demonstrated several times, the operation mechanism has not been clarified thus far due to the strong gas sensing characteristics of SnO₂. Here we clarify the operation mechanism of a-SnO₂ TTFT by electric field thermopower modulation analyses. We prepared a bottom-gate top-contact type TTFT using 4.2-nm-thick a-SnO₂ as the channel without any surface passivation. The effective thickness of the conducting channel was $\sim 1.7 \pm 0.4$ nm in air and in vacuum, but a large threshold gate voltage shift occurred in different atmospheres; this is attributed to carrier depletion near at the top surface (~ 2.5 nm) of the a-SnO₂ due to its interaction with the gas molecules and the resulting shift in the Fermi energy. The present results would provide a fundamental design concept to develop a-SnO₂ TTFT.

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Transparent amorphous oxide semiconductors (TAOSs) based transparent thin-film transistors (TTFTs) with high field effect mobility (μ_{FE}) are essential components for advanced flat panel displays such as transparent organic light-emitting diode (OLED) displays and rollable OLED displays. Currently, amorphous (a-) InGaZnO₄ is widely applied as the TAOS^{1–4} of the TFT channel of commercially available OLED displays; the optical bandgap of a-InGaZnO₄ is ~ 3 eV,⁵ transparent in the visible light region, and the μ_{FE} of a-InGaZnO₄ (~ 10 cm² V^{−1} s^{−1})^{11,2} is two orders of magnitude higher than that of previously used a-Si. However, the use of a-InGaZnO₄ must be reduced because the consumption of rare element such as indium (Clarke number: 1×10^{-5}) is not desirable for maintaining sustainable usage of resources. Therefore, development of post-a-InGaZnO₄ TAOSs showing high μ_{FE} , which are composed of abundant elements, is crucial.

Among several indium-free TAOSs,^{6–8} a-SnO₂ is a promising candidate for overcoming the issues with a-InGaZnO₄. Sn is one of the abundant metal elements with the Clarke number⁹ of Sn which is $4 \times 10^{-3}\%$, 400 times larger than that of In. Moreover, SnO₂ TFT shows

extremely high $\mu_{FE} > 100$ cm² V^{−1} s^{−1},^{10–12} which is one order of magnitude higher than that of a-InGaZnO₄. In 2007, Dattoli *et al.*¹⁰ reported that Ta-doped SnO₂ nanowire TFT exhibits uniform characteristics with average μ_{FE} exceeding 100 cm² V^{−1} s^{−1} at room temperature. In 2009, Sun *et al.*¹¹ fabricated Sb-doped SnO₂ nanocrystal TFTs at room temperature with μ_{FE} of 158 cm² V^{−1} s^{−1}. In 2016, Shin *et al.*¹² reported that an extremely thin (< 4.5 nm) undoped SnO₂ TFT exhibited μ_{FE} of 150 cm² V^{−1} s^{−1} at room temperature in air. In these reports, bottom-gate top-contact type TFTs without any passivation showed high μ_{FE} . Thus, the top surface of SnO₂ channel was exposed to the atmosphere.

SnO₂ is a well-known gas sensing material.^{13,14} It is known that a 3–4-nm-thick depletion layer is formed at the SnO₂ surface.^{15,16} Oxygen molecules in the ambient atmosphere are adsorbed on the surface, seize free electrons near the surface, and form a depletion layer. In a reducing atmosphere, parts of the seized electrons are released back to the depletion layer. Thus, the transistor characteristics of the bottom-gate top-contact type SnO₂ TFTs would be strongly affected by the gases surrounding the exposed channel surface due to their gas sensing property. For this reason, the operation mechanism of the bottom-gate

top-contact type SnO_2 TFTs including the conduction band bending and the effective channel thickness have not been clarified thus far.

Here we clarify the conduction band bending and the effective channel thickness of 4.2-nm-thick a- SnO_2 based bottom-gate top-contact type TTFTs without any surface passivation. We analyzed the conduction band bending of the SnO_2 top surface by measuring the TFT characteristics in air and vacuum, and a large threshold gate voltage shift was observed; the Fermi energy in the carrier depletion region at the top surface (~ 2.5 nm) of the a- SnO_2 channel sensitively shifted with the changes in the gas atmosphere. The effective channel thickness (t_{eff}) was analyzed by the electric field thermopower (S) modulation method,^{17–21} and the t_{eff} was $\sim 1.7 \pm 0.4$ nm in air and in vacuum. The present results would provide a fundamental design concept for developing a- SnO_2 TTFT.

The bottom-gate top-contact TTFTs (Fig. 1) were fabricated on 100-nm-thick ITO coated alkali-free glass (thickness: 0.7 mm, Corning® EAGLE XG®) substrates by pulsed laser deposition (PLD, KrF excimer laser, 10 Hz) technique. First, a 300-nm-thick polycrystalline Y_2O_3 gate dielectric film (the dielectric permittivity, $\epsilon_r = 20$ ²²) was deposited at room temperature. The fluence of the KrF laser was ~ 2 J cm^{-2} pulse⁻¹, and the oxygen pressure was kept at 0.4 Pa during deposition. Then, a 4.2-nm-thick SnO_2 film was deposited on the Y_2O_3 /ITO bilayer laminate through a stencil mask at 300 °C [see [supplementary material](#) Figs. S1(a) and S1(b)]. The fluence of the KrF laser was ~ 0.3 J cm^{-2} pulse⁻¹, and the oxygen pressure was kept at 1 Pa during the deposition. The optical bandgap (E_g) of the a- SnO_2 film was ~ 4.3 eV [[supplementary material](#) Fig. S1(c)], which was larger than that of bulk SnO_2 ($E_g \sim 3.6$ eV).²³ This is most likely due to the quantum size effect.²⁴ Finally, 100-nm-thick ITO films, which were used as the source and drain electrodes ($400 \mu\text{m} \times 400 \mu\text{m}$), were deposited at room temperature. The fluence of the KrF laser was ~ 0.9 J cm^{-2} pulse⁻¹ and the oxygen pressure was kept at 3 Pa during deposition. After these PLD processes, the device was annealed at 400 °C for 30 min in air. The channel length L and the channel width

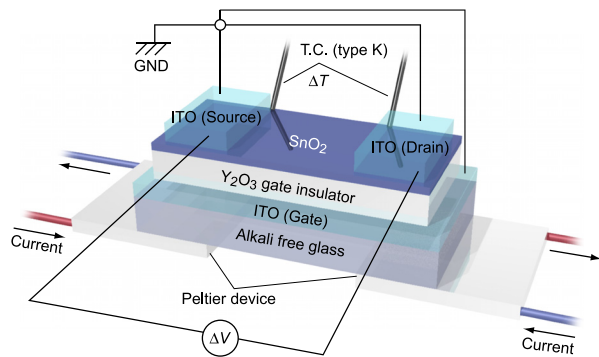


FIG. 1. Electric field thermopower modulation measurement of the bottom-gate top-contact a- SnO_2 TTFT. The top surface of the 4.2-nm-thick SnO_2 channel is exposed to the air. The channel length L is $200 \mu\text{m}$ and the channel width W is $400 \mu\text{m}$. The gate insulator is 300-nm-thick polycrystalline Y_2O_3 ($\epsilon_r = 20$). The SnO_2 channel is placed on the gap (~ 2 mm) between two Peltier devices, which are used to give temperature difference (ΔT) between both edges of the channel. Two K-type thermocouples (T.C.) are located at both edges of the SnO_2 channel to measure the temperature difference between both edges of the SnO_2 channel. The thermoelectromotive force (ΔV) and ΔT are measured simultaneously at a fixed gate voltage (V_g).

W of the resultant TTFT were 200 and $400 \mu\text{m}$, respectively. The resultant multiple layer was fully transparent in the visible light region ([supplementary material](#) Fig. S2).

The TTFT characteristics such as transfer characteristics (I_d - V_g) and output characteristics (I_d - V_d) were measured using a semiconductor device analyzer (B1500A, Agilent Co.) at room temperature. As shown in Fig. 2(a), the resultant TTFTs showed clear transfer (I_d - V_g) characteristics with the on-to-off current ratios of $\sim 10^5$. All the TTFTs show clear pinch-off in the output characteristics [[supplementary material](#) Fig. S3], indicating that the TTFT operation obeys the standard field-effect theory. It should be noted that the threshold gate voltage (V_{th}), which was evaluated by plotting $I_d^{0.5}$ - V_g relationship, was -14 V in air but shifted dramatically to -23 V in vacuum. The μ_{FE} , calculated from $\mu_{\text{FE}} = g_m \cdot [(W/L) \cdot C_i \cdot V_{\text{d}}]^{-1}$, where g_m is the transconductance $\partial I_d / \partial V_g$ and C_i is capacitance per unit area ($C_i \sim 58$ nF cm^{-2}),²² was ~ 20 $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$ in air and ~ 30 $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$ in vacuum. These confirm that the channel conductance and the V_{th} indeed can be modulated with the gases surrounding exposed a- SnO_2 channel. The SnO_2 TTFT characteristics were summarized in Table I.

The electric field modulated thermopower (S) was measured during the transfer characteristics measurements in the two gas

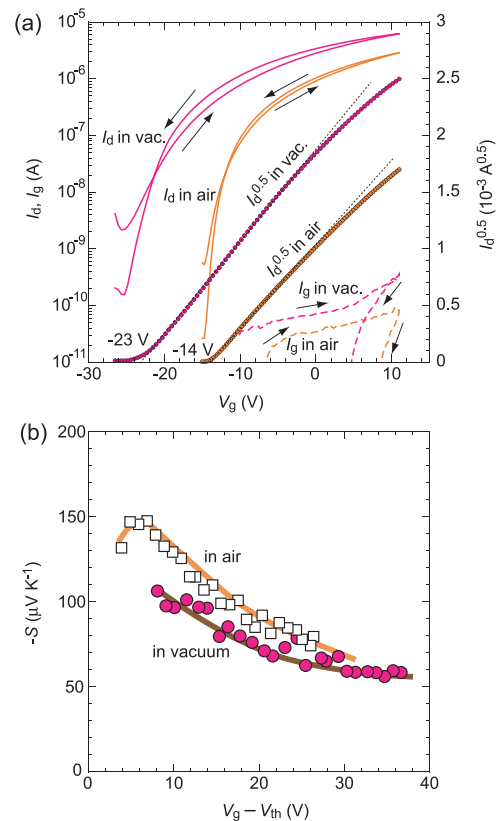


FIG. 2. Transistor characteristics of the bottom-gate top-contact a- SnO_2 TTFT measured in air and in vacuum. (a) Transfer characteristic (I_d - V_g) curve at $V_d = +0.1$ V. Corresponding $I_d^{0.5}$ - V_g and I_g - V_g curves are also shown. The threshold voltage (V_{th}) is -14 V in air and -23 V in vacuum. The gate leakage current (I_g) is < 100 pA. (b) Electric field modulated thermopower (S) at various $V_g - V_{\text{th}}$ ranging from $+4$ V to $+37$ V. The $-S$ gradually decreases with $V_g - V_{\text{th}}$.

TABLE I. On-off current ratio, threshold gate voltage (V_{th}), subthreshold swing factor (S.S.), and field effect mobility (μ_{FE}) for SnO₂ TFT.

	ON/OFF	V_{th} (V)	S.S. (V decade ⁻¹)	$\mu_{FE,max}$ (cm ² V ⁻¹ s ⁻¹)
Air	$\sim 10^5$	-14	0.65 ± 0.02	20
Vacuum	$\sim 10^5$	-23	1.57 ± 0.2	30

atmospheres (air, vacuum) to analyze the t_{eff} . Details of the electric field modulated S measurement are described elsewhere.^{17–20} Figure 2(b) shows the changes in $-S$ as a function of the effective gate voltage ($V_g - V_{th}$). The S values were always negative, consistent with the fact that the SnO₂ film is n -type semiconductor. The absolute values of S decrease gradually with increasing $V_g - V_{th}$ in both atmospheres due to an increase in the sheet carrier concentration (n_s), which was deduced from $n_s = C_i \cdot (V_g - V_{th}) \cdot e^{-1}$. Although a small variation in S was observed due to the gate leakage current (I_g), the observed S could be used to analyze the t_{eff} since the variation is less than 10%, and the difference in the air and vacuum atmospheres is noticeably clear.

In order to extract the t_{eff} , we plotted $-S$ as a function of n_s [Fig. 3(a)]. An almost linear relationship with a slope of $\sim 120 \mu V K^{-1}$ decade⁻¹ was observed in the $-S$ vs $\log n_s$ plot when n_s exceeded $2.5 \times 10^{12} \text{ cm}^{-2}$ in air, and a slope of $\sim 84 \mu V K^{-1}$ decade⁻¹ was observed in the same plot when n_s exceeded $2.9 \times 10^{12} \text{ cm}^{-2}$ in vacuum. We also plotted the three-dimensional carrier concentration (n_{3D}) dependence of $-S$ measured from separately prepared SnO₂ thin films [Fig. 3(b), Table S1]. From the $-S$ vs $\log n_{3D}$ relationship, the carrier effective mass (m^*) of the SnO₂ films was extracted to be $0.47 m_0$ using the following equations:

$$s = \frac{k_B}{e} \left(\frac{\left(r + \frac{5}{2}\right) F_{\left(r + \frac{3}{2}\right)}(\eta)}{\left(r + \frac{3}{2}\right) F_{\left(r + \frac{1}{2}\right)}(\eta)} - \eta \right), \quad (1)$$

$$n_H = \frac{1}{eR_H} = \frac{8\pi(2m_d^*k_B T)^{\frac{3}{2}}}{3h^3} \frac{\left(r + \frac{3}{2}\right)^2 F_{\left(r + \frac{1}{2}\right)}(\eta)}{\left(2r + \frac{3}{2}\right) F_{\left(2r + \frac{1}{2}\right)}(\eta)}, \quad (2)$$

$$F_n(\eta) = \int_0^\infty \frac{x^n}{1 + e^{x-\eta}} dx, \quad (3)$$

where $F_n(\eta)$ is the n th order Fermi integral, η is the reduced fermi energy, r is the scattering factor, h is the Planck constant, and k_B is the Boltzmann constant. Because acoustic phonon scattering is commonly the main scattering mechanism for this material, the scattering factor r could be considered as $-1/2$. S only depends on the slope of the electronic density of states at the Fermi level, which depends on n_{3D} . Since the n_s measured from the transistor characteristics represents the projected n_{3D} within the t_{eff} , we can directly compare the $-S$ vs $\log n_s$ relationship with the $-S$ vs $\log n_{3D}$ relationship. The slope of $-S$ vs $\log n_{3D}$ plot [Fig. 3(b)] is comparable to that of the $-S$ vs $\log n_s$ plot [Fig. 3(a)], indicating that the $E-k$ relation at the bottom of the conduction band is parabolic, and t_{eff} can be extracted as n_s/n_{3D} .

Several S -values around the lower n_s did not follow the straight line, probably due to that the non-parabolic shaped tail states just

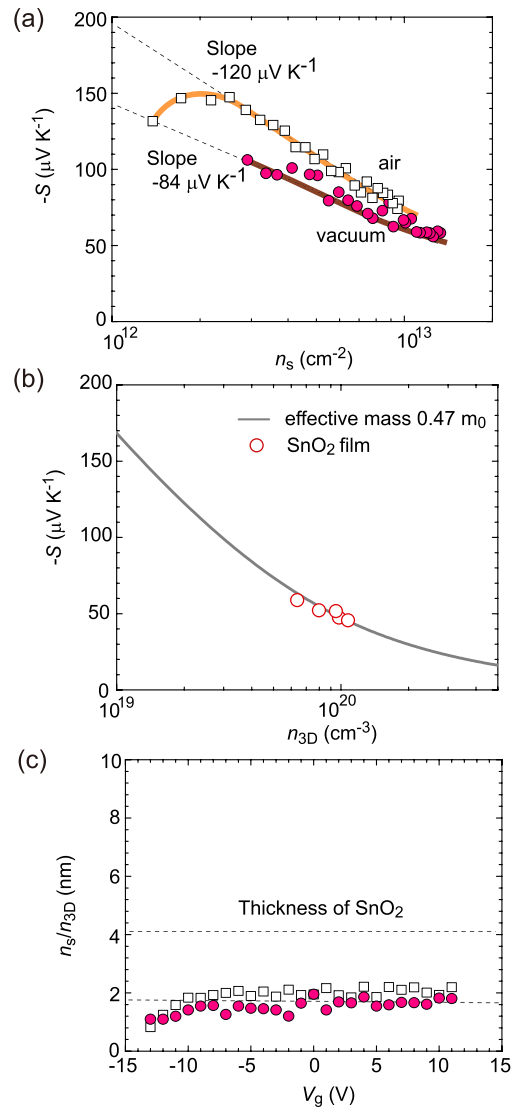


FIG. 3. Electric field thermopower modulation analyses of the bottom-gate top-contact a-SnO₂ TTFT. (a) Change in $-S$ as a function of the sheet carrier concentration (n_s). The slope of the $-S$ - n_s relationship is $-120 \mu V K^{-1}$ decade⁻¹ in air and $-84 \mu V K^{-1}$ decade⁻¹ in vacuum (dotted line). (b) Three-dimensional carrier concentration (n_{3D}) dependence S of the SnO₂ films. We calculated the carrier effective mass (m^*) of the SnO₂ film around $0.47 m_0$. (c) The effective thickness (t_{eff}), which is defined as n_s/n_{3D} , as a function of V_g . The t_{eff} is always $\sim 1.7 \pm 0.4$ nm, insensitive to the atmosphere.

below the original conduction band bottom.^{18,22,25} The S and n_s were modulated from $(-150 \mu V K^{-1}, 2 \times 10^{12} \text{ cm}^{-2})$ to $(-80 \mu V K^{-1}, 1 \times 10^{13} \text{ cm}^{-2})$ in air and from $(-110 \mu V K^{-1}, 2.9 \times 10^{12} \text{ cm}^{-2})$ to $(-60 \mu V K^{-1}, 1.3 \times 10^{13} \text{ cm}^{-2})$ in vacuum with increasing positive electric field in the a-SnO₂ channel. The difference between the measurements in air and in vacuum is attributed to the gas sensing property of SnO₂, where O₂ molecules are adsorbed in air and released in vacuum. The $t_{eff} \equiv n_s/n_{3D}$ of the conducting a-SnO₂ channel were always $\sim 1.7 \pm 0.4$ nm, insensitive to the gas atmosphere [Fig. 3(c)].

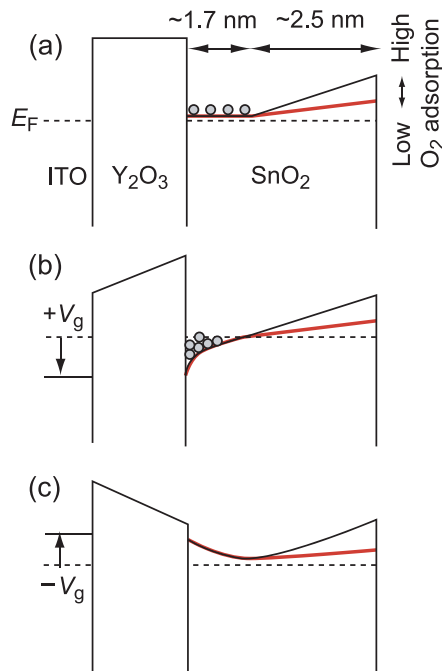


FIG. 4. Electric field modulation mechanism of the bottom-gate top-contact a-SnO₂ TTFT. (a) Without any gate voltage (V_g) application. Conduction band minimum (CBM) around the surface is lifted due to oxygen gas adsorption. Black: higher oxygen atmosphere (air). Red: lower oxygen atmosphere (vacuum). The 2.5-nm-thick surface region is the surface depletion layer. The conducting channel (~ 1.7 nm) remains at the Y₂O₃/a-SnO₂ interface. The sheet carrier concentration (n_s) is $\sim 4 \times 10^{12} \text{ cm}^{-2}$. (b) Under positive V_g application. The 1.7-nm-thick 2DEG layer is formed at the Y₂O₃/a-SnO₂ interface. The n_s increases up to $\sim 1 \times 10^{13} \text{ cm}^{-2}$. The surface region does not change. (c) Under negative V_g application, the interface electrons are completely depleted, resulting in off states.

Here, we would like to discuss the operation mechanism of a-SnO₂ TTFT [Fig. 4]. Without any V_g application [Fig. 4(a)], the conduction band minimum (CBM) at the surface is lifted due to the adsorption of oxygen when the TTFT is exposed to air (higher oxygen atmosphere, black line). When the TTFT is exposed to vacuum, the CBM at the surface is lowered (lower oxygen atmosphere, red line). Due to the oxygen adsorption, a 2.5-nm-thick depletion layer, which is similar to the Debye length of SnO₂ (~ 3 nm),²⁶ is formed near the surface region. As a result, a conducting channel ($\sim 1.7 \pm 0.4$ nm) remains at the Y₂O₃/a-SnO₂ interface, and the n_s is $\sim 4 \times 10^{12} \text{ cm}^{-2}$ ($n_{3D} \sim 2 \times 10^{19} \text{ cm}^{-3}$). Under positive V_g application [Fig. 4(b)], the carriers accumulate at the Y₂O₃/a-SnO₂ interface, and the E_F locates above the CBM. As a result, the 1.7-nm-thick 2D electron gas (2DEG) is formed at the Y₂O₃/a-SnO₂ interface. The n_s increases up to $\sim 1 \times 10^{13} \text{ cm}^{-2}$ ($n_{3D} \sim 4 \times 10^{19} \text{ cm}^{-3}$). The surface region does not change. Under negative V_g application [Fig. 4(c)], the conduction electrons at the Y₂O₃/a-SnO₂ interface are completely depleted, showing off states in the transistor characteristics.

In summary, we have clarified the operation mechanism including the conduction band bending and the effective channel thickness (t_{eff}) of 4.2-nm-thick a-SnO₂ based bottom-gate top-contact type TTFTs without any surface passivation. We analyzed the conduction band bending of SnO₂ top surface by measuring the TFT

characteristics in air as well as in vacuum and found the large threshold gate voltage shift; the Fermi energy in the carrier depletion region at the top surface (~ 2.5 nm) of the a-SnO₂ sensitively shifted depending on the gas atmospheres. We also analyzed the t_{eff} ($\equiv n_s/n_{3D}$) by the electric field thermopower modulation method. t_{eff} was $\sim 1.7 \pm 0.4$ nm in air and in vacuum.

From the thickness of the a-SnO₂ film (4.2 nm) and the t_{eff} (1.7 ± 0.4 nm), the carrier depletion depth at the top surface of the a-SnO₂ film is estimated to be 2.5 nm, which is similar to the depletion length reported in other studies.^{15,16} When a-SnO₂ bottom-gate top-contact TTFTs are exposed to air, oxygen gas would be adsorbed on the surface of SnO₂ film as electron accepting species, which is accompanied by the formation of a depletion layer inside the film. The present results may provide a fundamental design concept for utilizing a-SnO₂ TFT in device applications.

See the [supplementary material](#) for additional crystallographic analyses of the SnO₂ thin film, optical transmission of the bottom-gate top-contact a-SnO₂ TTFT, transistor characteristics of the bottom-gate top-contact a-SnO₂ TTFT, and optical absorption spectrum of the polycrystalline Y₂O₃ thin film deposited on the SiO₂ glass substrate.

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