Deep-UV Transparent Conducting Oxide La-Doped SrSnO$_3$ with a High Figure of Merit

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ABSTRACT: Perovskite stannate SrSnO$_3$ (SSO) is attracting attention as ultraviolet transparent conducting oxides (UV TCOs) due to its ultrawide band gap and high conductivity. Here, we investigate in detail the thickness-dependent electrical, structural, and optical properties of sequentially strain-relaxed La-doped SrSnO$_3$ (SLSO) epitaxial thin films. We find that the SLSO films grow as an orthorhombic $Pnma$ phase with $a^* \alpha^* c^*$ in the $c^*$ direction under the tensile strain. With the strain relaxation, as the films become thicker, vertical grain boundaries are created and the orthorhombic phase becomes reoriented to all three possible orientations. Simultaneously, the conductance starts to deviate from the linear behavior with increasing film thickness. Through the analysis of thickness fringes in optical transmittance, we found that a 120 nm thick nominally 4% La-doped SrSnO$_3$ film has a figure of merit ($\rho_{TC} = 2.65 \times 10^{-3}$ Ω$^{-1}$) at $\lambda = 300$ nm in the deep-UV region, which is the highest value among the well-known candidates for UV TCOs reported to date.

KEYWORDS: ultraviolet transparent conducting oxides, figure of merit, La-doped SrSnO$_3$, ultrawide band gap semiconductors, thickness-dependent properties

1. INTRODUCTION

The need for deep-ultraviolet transparent conducting oxides (DUV TCOs) has been growing steadily. Accordingly, the search for high conductance in ultrawide band gap and high conductivity. Here, we investigate in detail the thickness-dependent electrical, structural, and optical properties of sequentially strain-relaxed La-doped SrSnO$_3$ (SLSO) epitaxial thin films. We find that the SLSO films grow as an orthorhombic $Pnma$ phase with $a^* \alpha^* c^*$ in the $c^*$ direction under the tensile strain. With the strain relaxation, as the films become thicker, vertical grain boundaries are created and the orthorhombic phase becomes reoriented to all three possible orientations. Simultaneously, the conductance starts to deviate from the linear behavior with increasing film thickness. Through the analysis of thickness fringes in optical transmittance, we found that a 120 nm thick nominally 4% La-doped SrSnO$_3$ film has a figure of merit ($\rho_{TC} = 2.65 \times 10^{-3}$ Ω$^{-1}$) at $\lambda = 300$ nm in the deep-UV region, which is the highest value among the well-known candidates for UV TCOs reported to date.

To date, two classes of materials have been proposed as UV transparent conducting oxide candidates. One is correlated metals, which have strong electron−electron interactions, such as SrNbO$_3$,$^{18,19}$ CaVO$_3$, and SrVO$_3$. The other class is doped semiconductors with an ultrawide band gap such as Sn-doped $\beta$-Ga$_2$O$_3$ ($E_g \sim 4.9$ eV),$^{22}$ Sn-doped $\alpha$-Ga$_2$O$_3$ ($E_g \sim 5.3$ eV),$^{23}$ and La-doped SrSnO$_3$ ($E_g \sim 4.6$ eV).$^{24,25}$ Among the materials corresponding to the former class, CaVO$_3$ and SrVO$_y$ perovskite vanadates, have excellent conductivity over 10$^4$ S cm$^{-1}$. This high conductivity comparable to ITO comes from the higher carrier density ($n \sim 10^{22}$ cm$^{-3}$) with high effective mass due to the strong Coulomb interaction between electrons. However, their optical transmittance for energies above ∼3.3 eV is suppressed and the transmittance is under 50% in the deep-UV region.$^{20,26}$ In the case of SrNbO$_y$, there is no direct measurement data yet for optical transmittance, although it has just been calculated to be over 80% from the absorption coefficient through the complex dielectric function obtained by spectroscopic ellipsometry.$^{18,19}$ Sn-doped $\alpha$-Ga$_2$O$_3$ and Sn-doped $\beta$-Ga$_2$O$_3$ are candidates for UV TCOs

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belonging to the latter class. However, they have poor transparency in the deep-UV range at the Sn-doping concentration required for the large conductance.\textsuperscript{27,28} In the case of Sn-doped β-Ga\textsubscript{2}O\textsubscript{3}, the band gap greatly decreases from 4.9 eV for undoped to 4.1 eV at the optimum doping rate of 10\%, resulting in decrease of transmittance from 60 to 5\% at 300 nm.\textsuperscript{27} A similar phenomenon is observed in Sn-doped α-

\textsuperscript{2}Ga\textsubscript{2}O\textsubscript{3}; its optical transmittance reduces from 80\% for 0.005\% Sn doping to 50\% at the optimum doping rate of 0.1\%.\textsuperscript{28} Another disadvantage is that they have a high electrical resistivity due to the low dopant solubility limit and degradation of crystallinity with increasing dopant concentration.\textsuperscript{27,28}

Very recently, La-doped SrSnO\textsubscript{3} (SLSO) has emerged as one of the perovskite-structured alkaline stannates which have a potential for transparent conducting oxides due to their superior electrical conductivity and ultrawide band gap.\textsuperscript{29} The conduction band of perovskite stannates originating mainly from the Sn-5s orbital leads to small electron effective mass and high electron mobility in spite of degenerate doping.\textsuperscript{29} La-doped BaSnO\textsubscript{3} single crystals and epitaxial films are shown to have an outstanding electron mobility of 320 and 150 cm\textsuperscript{2}V\textsuperscript{-1}s\textsuperscript{-1}, respectively.\textsuperscript{30,31} SrSnO\textsubscript{3} possesses a wider band gap (E\textsubscript{g} ∼ 4.6 eV) and smaller effective mass (m\textsubscript{e} ∼ 0.23 m\textsubscript{0})\textsuperscript{32} than BaSnO\textsubscript{3} (E\textsubscript{g} ∼ 3.1 eV, m\textsubscript{e} ∼ 0.42 m\textsubscript{0})\textsuperscript{32,33} in bulk powders, an orthorhombic phase (Pnma) is known to be the stable phase at room temperature,\textsuperscript{34} with lattice constants of a\textsubscript{ortho} = 5.703 Å, b\textsubscript{ortho} = 5.709 Å, and c\textsubscript{ortho} = 8.065 Å (its pseudocubic lattice constant a\textsubscript{p} = 4.034 Å).\textsuperscript{35} Trivalent ions such as La\textsuperscript{3+}, Ta\textsuperscript{3+}, Nb\textsuperscript{3+}, and Sm\textsuperscript{3+} were used as effective n-type dopants\textsuperscript{36–46} by substituting for the Sr site without affecting the band structure.\textsuperscript{37} Additionally, pentavalent ions such as Nb\textsuperscript{5+}, Sb\textsuperscript{5+}, and Pb\textsuperscript{5+} in place of Sn were studied as n-type dopants.\textsuperscript{47} It is worth noting that the La dopant creates shallow donor levels in SrSnO\textsubscript{3}, which indicates that extra electrons in La ions are easily activated.\textsuperscript{47} The recently reported 3\% La-doped SrSnO\textsubscript{3} (SLSO) film has the electron mobility as high as 56 cm\textsuperscript{2}V\textsuperscript{-1}s\textsuperscript{-1} with the carrier density (n ∼ 3.26 × 10\textsuperscript{18} cm\textsuperscript{-3}) resulting in the highest conductivity (σ ∼ 3000 S cm\textsuperscript{-1}) ever reported for SrSnO\textsubscript{3}.\textsuperscript{38} The solubility of La ions in SLSO films was reported to be enhanced through the postdeposition vacuum-annealing process.

In this paper, we report systematic studies on the thickness-dependent properties of highly conductive and DUV-transparent La-doped SrSnO\textsubscript{3} films grown on epitaxial SrHfO\textsubscript{3} and BaHfO\textsubscript{3} buffer layers on MgO (001) substrates, all DUV-transparent materials, by pulsed laser deposition (PLD) as illustrated in Figure 1. We used a sequential strain relaxation method using a BaHfO\textsubscript{3} perovskite buffer layer, which has an ultrawide band gap (E\textsubscript{g} ∼ 6.1 eV)\textsuperscript{53} and a lattice constant (a = 4.171 Å)\textsuperscript{52} which matches well with that of MgO (a = 4.21 Å).\textsuperscript{53} SrHfO\textsubscript{3} was chosen as the second buffer layer because it is also an ultrawide band gap (E\textsubscript{g} ∼ 6.1 eV)\textsuperscript{54} perovskite with a lattice constant (a\textsubscript{pc} = 4.087 Å)\textsuperscript{55} between those of BaHfO\textsubscript{3} and SrSnO\textsubscript{3} (a\textsubscript{pc} = 4.034 Å).\textsuperscript{55} The structural properties are examined by a high-resolution X-ray diffractometer (HRXRD) and scanning transmission electron microscope (STEM). We found that as the orthorhombic phase of SLSO films becomes thicker, they begin to change its direction and their conductance starts to deteriorate. The optical transmittance is investigated to maximize the figure of merit by analyzing the thickness fringes displayed by thin-film interference.

Figure 1. Tailoring of deep-UV transparent conducting oxide heterostructure. Schematics show sequential strain-relaxed and thickness-tuned nominally 4\% La-doped SrSnO\textsubscript{3} films, which are highly conductive and transparent in the deep-UV range. SrSnO\textsubscript{3} (SSO) has an orthorhombic perovskite structure with the Pnma space group (a’ a’ c’) at room temperature with a pseudocubic lattice constant (a\textsubscript{pc} = 4.034 Å) and a wide indirect band gap of 4.6 eV.

2. RESULTS AND DISCUSSION

2.1. Doping Dependence of the Electrical Properties of 50 nm Thick SLSO Films. To investigate the effect of a nominal La-doping rate, we measured the electrical conductivity, the carrier density, and the Hall mobility of 50 nm thick Sr\textsubscript{1−x}La\textsubscript{x}SnO\textsubscript{3} thin films (x = 0.003, 0.005, 0.01, 0.02, 0.03, 0.04, 0.05) at room temperature. Figure 2a shows the current–voltage (I–V) data and the schematic for the Van der Pauw measurement used. Figure 2b–d shows the conductivity, Hall mobility, and carrier density, respectively. A significant increase in conductivity was observed at a 0.5\% La-doping rate due to sharp increases in the carrier density (Figure 2d) and the Hall mobility (Figure 2c). As the nominal doping rate increases from 0.5 to 4\%, the conductivity increases almost linearly with the nominal doping rate since there is little change in the Hall mobility, and the carrier density increases linearly. However, exceeding the nominal La-doping rate beyond 4\%, the Hall mobility starts to decrease sharply, while the carrier density maintains the same trend. After a nominal 4\% La-doping rate, scattering mechanisms by ionized and/or unionized La dopants seem dominant. Therefore, we found the optimum La-doping rate to be 4\%; a 50 nm thick 4\% SLSO film shows a high Hall mobility (μ = 44.14 cm\textsuperscript{2}V\textsuperscript{-1}s\textsuperscript{-1}) and large carrier density (n\textsubscript{SD} = 4.25 × 10\textsuperscript{19} cm\textsuperscript{-3}), and above all, the highest electrical conductivity (σ ∼ 3100 S cm\textsuperscript{-1}) among the values of SLSO films reported to date. On the other hand, when the nominal La-doping rate drops from 0.5 to 0.3\%, the Hall carrier density drops quickly from 4.01 × 10\textsuperscript{18} to 8.14 × 10\textsuperscript{18} cm\textsuperscript{-3}, and it was difficult to measure in 50 nm thick 0.25\% SLSO films due to its high resistance. From this, we can infer that the deep acceptor density of our SLSO films is in the same range of the La\textsuperscript{3+} ion dopant density in 0.25\% SLSO films (n\textsubscript{DAA} ∼ 3.81 × 10\textsuperscript{19} cm\textsuperscript{-3}).

2.2. Thickness Dependence of the Electrical Properties of 4% SLSO Films. To examine the electrical properties
buffers on the MgO (001) substrate at room temperature. (a) Voltage conductivity, (c) Hall mobility, and (d) carrier density. A black and 3 of the film. The inset is a schematic of Van der Pauw measurements. Indium was used as a contact. (b) Electrical conductivity, (c) Hall mobility, and (d) carrier density. A black dotted line represents the carrier density when La ions in the films are 100% activated.

Looking at the slope of the conductance up to 145 nm, the conductance does not increase as expected and starts to deviate from the 3000 S cm$^{-1}$ line. The Hall mobility in Figure 3c tends to decrease slightly with the film thickness. The two-dimensional (2D) carrier density follows the blue dotted line representing the carrier density when La ions in the film are 100% activated.

From the spacing between the fringes, the BaHfO$_3$ thickness was calculated to be 53 nm, and it agrees with those measured by the stylus profiler and transmission electron microscopy (TEM). As the thickness of the 4% SLSO film increased, the 2θ peak position of 4% SLSO shifted to the left. It is seen clearly between $t = 50$ and 105 nm. Simultaneously, the symmetric peak shape of the 4% SLSO film becomes asymmetric beyond the thickness of 50 nm. Figure 4e–g shows the results of deconvoluting 50, 105, and 290 nm thick 4% SLSO asymmetric peaks using the Voigt functions. The 50 nm thick film peak is well fitted with only one Voigt function and its position is very close to the peak position of the 22 nm thick film in the Supporting Information (Figure S1a). However, in the case of the 105 nm and 290 nm thick films, the additional peak (peak 2) is necessary to fit the experimental data. The sums of the two peaks can be very well fitted to the experimental data (black circles). Therefore, either a new orientation or a new phase, corresponding to peak 2, starts to appear in the thickness range between 50 and 105 nm. The case of 145 nm thick film in the Supporting Information (Figure S1b) is the same as the case of 105 nm. It is highly likely that the tensile strain is relieved by creating a new orientation or a new phase as the 4% SLSO films become thicker. This is in contrast to the previously reported situation, where the compressive strain on GdScO$_3$ (110) substrates is relaxed as the SLSO film becomes thicker. 

**2.3. Structural Properties of 4% SLSO.** To study the phenomena discussed above in detail, we investigate the structural properties of $t$ nm thick 4% SLSO films. Figure 4a,b shows the results of high-resolution X-ray θ–2θ scans and rocking curves of $t$ nm thick 4% SLSO films ($t = 22, 50, 105, 145, 290$) grown on 33 nm thick SrHfO$_3$ and 55 nm thick BaHfO$_3$ buffer layers on the MgO (001) substrate. Figure 3a illustrates the rocking curves for 105 nm 4% SLSO, 33 nm SrHfO$_3$, and 55 nm BaHfO$_3$ and the MgO (001) substrate. The MgO (001) single-crystal substrate has a sharp peak shape with the rocking curves of 145 nm. It suggests that there exists a change in the microstructure in films as the thickness increases, which tends to impede the current flow.

The case of 145 nm thick film in the Supporting Information (Figure S1a). However, in the case of the 105 nm and 290 nm thick films, the additional peak (peak 2) is necessary to fit the experimental data. The sums of the two peaks can be very well fitted to the experimental data (black circles). Therefore, either a new orientation or a new phase, corresponding to peak 2, starts to appear in the thickness range between 50 and 105 nm. The case of 145 nm thick film in the Supporting Information (Figure S1b) is the same as the case of 105 nm. It is highly likely that the tensile strain is relieved by creating a new orientation or a new phase as the 4% SLSO films become thicker. This is in contrast to the previously reported situation, where the compressive strain on GdScO$_3$ (110) substrates is relaxed as the SLSO film becomes thicker. 

**Figure 3.** Thickness-dependent electrical properties of 4% SLSO films grown on 33 nm thick SHO and 55 nm thick BHO buffers on the MgO (001) substrate at room temperature. (a) Voltage measured between 1 and 4 of the film by applying a current between 2 and 3 of the film. (b) Conductance gradually deviates from the red dotted line representing the conductivity of 3000 S cm$^{-1}$ with increasing film thickness. (c) Hall mobility slightly decreases as the film thickness increases. (d) Two-dimensional carrier density follows the blue dotted line representing the three-dimensional (3D) carrier density when La ions in the film are 100% activated.
The rocking curve of 55 nm BaHfO$_3$ consists of two Gaussian functions: narrow and broad Gaussian functions, which suggests that 55 nm BaHfO$_3$ is partially relaxed on the MgO substrate. Typically, 33 nm SrHfO$_3$ and 105 nm 4% SLSO with an orthorhombic structure have wider rocking curves than cubic BaHfO$_3$ and the single-crystal MgO substrate. The full width at half-maximum (FWHM) values of the rocking curves of the 4% SLSO films gradually decrease with increasing the film thickness, as illustrated in Figure 4d.

To investigate the structural properties in detail, we measure the asymmetric reciprocal space mapping (RSM) for the (024) plane and (002) plane for $t$ nm thick 4% SLSO, 33 nm thick SrHfO$_3$, and 55 nm thick BaHfO$_3$ and MgO. Figure 5a,b shows the reciprocal space mapping (RSM) results around the (024) plane for 22 and 290 nm thick 4% SLSO, respectively. MgO, BaHfO$_3$, SrHfO$_3$, and 4% SLSO peaks sequentially appear from the bottom. The RSM results for the (024) plane of 28, 50, 70, 105, 145, and 210 nm thick 4% SLSO films are shown in the Supporting Information (Figure S2). The schematic display of the four peaks can be shown in Figure 5a,b. The 290 nm thick 4% SLSO peak consists of a light and a dark green ellipse. In line with the previous $\theta-2\theta$ scan results, the peak positions of BaHfO$_3$, SrHfO$_3$, and MgO do not change, regardless of 4% SLSO thickness. Compared with the 22 and 290 nm thick 4% SLSO peaks, one can notice that their shapes and positions are changed. Even the 22 nm thick 4% SLSO does not grow coherently on the SrHfO$_3$ buffer layer from the fact that their reciprocal space vector $Q_x$ is not the same. Figure 5c,d shows the reciprocal space mapping (RSM) around the (002) plane for 22 and 290 nm thick 4% SLSO films, respectively. One can observe the thickness fringes of a 55 nm BaHfO$_3$ buffer layer. The widths of the four peaks in Figure 5c in the reciprocal space vector $Q_x$ direction seem constant. However, the width of 290 nm 4% SLSO peak in Figure 5d in the $Q_x$ direction is definitely larger. Consequently, the (002) RSM results confirmed again that a new orientation or a new phase is formed as the 4% SLSO thickness increases. Figure 5e shows the in-plane ($a_{ip}$) and out-of-plane ($a_{op}$) lattice constants for 4% SLSO calculated from the RSM (024) data as a function of the 4% SLSO film thickness. As the film thickness increases, the tensile strain by the SrHfO$_3$ buffer gets relaxed and a noticeable drop in the in-plane lattice constant can be observed between 50 and 70 nm thickness. The in-plane and out-of-plane lattice constants eventually merge with each other as the films become thicker.

Since it is clear that the microstructure of the films changes as a function of the SLSO thickness, scanning transmission electron microscopy (STEM) experiments were conducted...
(see the Supporting Information, Figures S3 and S5 for basic structural and compositional characterizations of SrSnO$_3$ films using STEM-energy-dispersive X-ray (EDX) analysis). In Figure 6a, a cross-sectional high-angle annular dark-field (HAADF)-STEM image of the 290 nm thick 4% SLSO film is presented, where the growth of columnar grains with the width of 10$^-50$ nm is noted from the grain boundary contrasts. High-resolution bright-field (BF)-STEM images acquired from the top and bottom regions show distinct patterns (Figure 6b). At the bottom, only a lateral stripe pattern with two-unit-cell periodicity along the z-direction is seen, whereas three dissimilar patterns—lateral stripe, vertical stripe, and cross patterns—in different grains are observed in the top region. STEM images of a 22 nm thick SLSO film show only lateral patterns (see the Supporting Information, Figure S3c), implying the development of different crystal orientations in the SrSnO$_3$ grains with the film growth. Fast Fourier transforms (FFTs) obtained from the grains with the distinct patterns are shown in Figure 6c, where the appearance of spots corresponding to (022) (highlighted with the green circles), (002) (yellow circles), and (020) (violet circles) planes from the cross, lateral, and vertical patterns is witnessed.

At room temperature, SrSnO$_3$ exhibits an orthorhombic phase (space group of Pnma) with the SnO$_6$ octahedral rotation pattern of $\alpha$ $\alpha$ $\alpha$ $\alpha$ $a$ $a$ $a$ in the Glazer notation. Figure 7 shows the atomic model of the Pnma phase SrSnO$_3$ projected in three Cartesian axes along the in-phase ($c$'') (Figure 7a) and out-of-phase ($a$'') (Figure 7b,c) octahedral rotation directions. Here, the projection direction is denoted as the $x$-axis, and the film growth direction is the $z$-axis. The projected image in the $c$'' direction (Figure 7a) shows the in-phase octahedral rotation from corresponding O displacements; however, the image from the $a$'' direction (Figure 7b,c) does not since the rotations in the opposite (clockwise and anticlockwise) directions are overlapped. Atomic-resolution BF-STEM images were obtained to connect the atomic structure viewed in different orientations with the three types of grains. As presented in Figure 7, dark contrast appears at the atomic positions, which enables visualization of light O atoms and subtle atomic displacements in the structure. First, a BF-STEM image producing the cross pattern shows the O displacement corresponding to the octahedral rotation, suggesting that the projection direction of the STEM image is the $c$'' direction, i.e., $c$'' $a$'' $a$''. In the case of the stripe-patterned BF-STEM images, such O pattern is not discernable, but slight Sr displacements along the stripe lines are seen, which is the result of the atomic images projected in the $a''$ direction. A comparison between the atomic model and BF-STEM images reveals that the stripe pattern is generated from an alternating Sr displacement.
plane lattice parameters compared to those of the tetragonal phase and, therefore, excludes the possibility of tetragonal phase formation.

2.4. Optical Properties of 4% SLSO Films. Optical transmittance spectra of 4 nm thick 4% SLSO films (t = 50, 70, 105, 145, 210, 290) are measured as a function of wavelength in Figure 8a. As the film thickness increases, the absorption pattern that is perpendicular to the c’ direction. Thus, in the case of grains displaying a lateral stripe pattern, the c’ direction is along the z-axis (growth direction), i.e., a’ a’ c’, while the vertical stripe pattern develops when the c’ axis is the y-axis, i.e., a’ c’ a’. The development and distribution of the reoriented grains were inspected from the Fourier filtered images using the unique spots from each orientation as presented in the Supporting Information (Figure S4a). The reoriented grains starting from ~50 nm above the film-substrate interface are evident, which agrees with the lattice parameter change evidenced in the X-ray diffraction experiments. The BF-STEM image of the boundary between the initial and oriented grains exhibits a continuous but clearly distinct boundary between two regions (see the Supporting Information, Figure S4b).

Figure 8b shows the (ahv)^1/2–photon energy plot to estimate the indirect band gap of t nm thick 4% SLSO films, where α is the absorption coefficient, h is the frequency of the incident photon, and h is the Planck’s constant, since SrSnO3 is known to exhibit an indirect band gap.61,62 The indirect band gap can be estimated by extrapolating the straight-line portion of the plot to the energy axis, illustrated in the inset of Figure 8b. Independent of the film thickness, the indirect band gap of 4 nm thick 4% SLSO is around 4.6 eV, which is similar to the previously reported value of 3% SLSO estimated by assuming a direct band gap.24 Our estimation for the direct band gap by plotting (ahv)^2 vs photon energy is shown in Figure S6, which suggests that the direct band gap is about 4.9 eV.

Finally, we can calculate the figure of merit for transparent conductors63 (φTC = T^0.5/Rs, where T is the transmission and Rs is the sheet resistance) at a wavelength of 300 nm, as shown in the Supporting Information (Table S1), and obtain φTC = 2.65 × 10^3 Ω−1 for 120 nm thick 4% SLSO, which is the highest value reported to date, among the candidates of deep-UV transparent conducting oxides such as Sn-doped β-Ga2O3.

Figure 8. Thickness-dependent optical properties of 4% SLSO films. (a) Optical transmittance spectra vs wavelength of 4% SLSO films with various thicknesses. The inset shows a magnification of the optical transmission spectra in the UV region. A black dotted vertical line represents the wavelength of 300 nm. (b) Optical indirect band gap vs photon energy of 4% SLSO films with various thicknesses. The inset is the results of indirect band gap fitting for 4% SLSO films. At all thicknesses, the indirect band gap is about 4.6 eV.
Conclusions

In conclusion, we have fabricated thickness-tuned 4% SLSO films grown on 33 nm SrHfO3 and 55 nm BaHfO3 buffers on the MgO (001) substrate by employing a sequential strain relaxation method. We have found that excellent conducting properties were obtained in 50 nm thick nominally 4% La-doped films with their high conductivity ($\sigma = 3100 \text{ S cm}^{-1}$), large carrier density ($n = 4.25 \times 10^{20} \text{ cm}^{-3}$), and high Hall mobility ($\mu = 44.14 \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$). As the films become thicker, we found that the tensile strain exerted by a SrHfO3 buffer layer is relaxed by forming grains in all three directions of the orthorhombic phase. At the same time, the conductance starts to deviate from the linear relationship with the film thickness due to the vertical grain boundaries that impede the transport of electrons. Via thickness tuning for the optical transmittance at a wavelength of 300 nm, a 120 nm 4% SLSO film was found to show the highest figure of merit calculated from the sheet resistance and transmittance. We demonstrate the great potential of 4% SLSO films for the next-generation deep-UV (λ = 300 nm) transparent conducting oxide (DUV TCO).

EXPERIMENTAL SECTION

4.1. Thin-Film Growth. All La-doped SrSnO3 (SLSO) epitaxial thin films grown on 33 nm SrHfO3 and 55 nm BaHfO3 buffers on the MgO (001) single-crystal substrate by the PLD method (KrF excimer laser, λ = 248 nm, coherent). Two side-polished MgO (001) substrates (5 mm × 5 mm × 0.5 mm) were provided by MTI Korea. All targets of BaHfO3, SrHfO3, SrSnO3, and 0.5, 1, 4, 8% La-doped SrSnO3 were provided by Toshiba manufacturing Co., Japan. First, a 55 nm BaHfO3 buffer layer is grown on the MgO (001) substrate by PLD (energy influence = 1.44 J cm$^{-2}$ and repetition rate = 10 Hz). The substrate temperature and oxygen partial pressure in the chamber were kept at 750 °C and 100 mTorr, respectively. The distance between the target and substrate is 51 mm. Second, the 33 nm SrHfO3 buffer layer is grown on the 55 nm BaHfO3 buffer layer by PLD (energy influence = 1.44 J cm$^{-2}$ and repetition rate = 10 Hz). The substrate temperature and oxygen partial pressure in the chamber were kept at 750 °C and 100 mTorr, respectively. The distance between the target and substrate is 45 mm. Third, SLSO is grown by PLD (energy influence = 1.55 J cm$^{-2}$ and repetition rate = 4 Hz). The substrate temperature and oxygen partial pressure in the chamber were kept at 750 °C and 150 mTorr, respectively. The distance between the target and substrate is 55.5 mm. After deposition, the SLSO films were cooled in vacuum to 400 °C and in 600 Torr oxygen thereafter.

4.2. Structural Property Measurement. The structural properties of the SLSO films were analyzed by θ–2θ scan, rocking curves, and reciprocal space mapping (RSM). θ–2θ scan and rocking curves were conducted using a high-resolution X-ray diffractometer (D8 Discover, Bruker) at IBS Center for Correlated Electron Systems, Seoul National University. The reciprocal space mapping (RSM) is performed using SmartLab with a Cu Kα source (λ = 1.5406 Å; Rigaku, Japan) at room temperature. An X-ray CBO system, a Ge (220) 2-bounce monochromator, and a one-dimensional semiconductor array detector (hybrid photon counting detector; HyPix-3000) were used for high-resolution crystallographic analysis. A scanning transmission electron microscopy (STEM) experiment was conducted using a FEI Titan G2 60-300 (S)TEM equipped with a CEOS DCOR probe corrector, a Schottky extreme field emission gun, and a super-X energy dispersive X-ray (EDX) spectrometer. The STEM was operated at 200 kV with a beam current of ~30 pA, and the probe convergence angle was 25.5 mrad. High-angle annular dark-field (HAADF)-STEM images were acquired with the annular dark-field (ADF) detector inner angle of 93 mrad, and bright-field (BF)-STEM images were acquired with the BF detector outer angle of 19 mrad.

4.3. Electrical Property Measurement. The electrical properties of SLSO films were measured using a Keithley 4200 semiconductor characterization system. Indium was used as the contact. The Van der Pauw method was used in a 4-point geometry for determining the sheet resistance of the films. The Hall carrier density and mobility were determined by applying the magnetic field perpendicular to the films while measuring the change in resistance.

4.4. Optical Property Measurement. The optical transmittance of SLSO films was measured by a grating spectrometer (Cary 5000, Bruker) over 200–3000 nm (0.6–6.2 eV). The spectrometer has a QL lamp light source for 2000–350 nm and a deuterium UV lamp light source for below 350 nm. Samples are mounted on a holder with a 3 mm diameter hole. Absorbance is calculated as the minus logarithm of the transmittance, and the absorption coefficient $\alpha$ is calculated by accounting for the sample thickness. We removed the absorption of the substrate and two buffers by measuring their optical absorption separately and subtracting it from the absorbance. From the optical absorption measurement, we plotted the Tauc’s plot of (ehv)$^{1/2}$ vs photon energy, where $\alpha$ denotes the absorption coefficient.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsaelm.2c00581.

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