Thermoelectric phase diagram of the SrTiO$_3$–SrNbO$_3$ solid solution system

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I. INTRODUCTION

Thermoelectric energy conversion, in which waste heat is transformed into electricity by the Seebeck effect, is attracting significant research attention as a potential energy harvesting technology.1 Generally, the performance of thermoelectric materials is evaluated in terms of a dimensionless figure of merit, ZT = S²αT/k, where ZT is the figure of merit, S is the thermopower (Seebeck coefficient), α is the electrical conductivity, T is the temperature, and k is the thermal conductivity. Although the ZT values of several commercialized thermoelectric materials such as Bi₂Te₃ (ZT ~ 1@400 K) and PbTe (ZT ~ 0.8@600 K) are roughly 1,2 these heavy metal-based materials present environmental risks due to the toxicity of their constituent elements and their chemical and thermal instability. Recently, transition metal oxides (TMOs) including Na₃CoO₂ (x ~ 0.75),3 Ca₃CoO₂B₆,4 and SrTiO₃5–7 have drawn a high volume of research as high-temperature thermoelectric power generation materials: this group of materials is considered to be particularly suitable for these applications due to their chemical and thermal robustness, as well as the comparatively low environmental risk.8

Among the available TMOs, electron-doped SrTiO₃ has been one of the most extensively studied materials for thermoelectric applications.9 In 2001, Okuda et al.5 synthesized Sr₁₋ₓLaₓTiO₃ (0 ≤ x ≤ 0.1) single crystals by the floating-zone method and reported that they yielded a large power factor (S²α) of 2.8–3.6 mW m⁻¹ K⁻² at room temperature. After this, Ohta et al. reported the carrier transport properties of Nb- and La-doped SrTiO₃ single crystals (carrier concentration, n ~ 10²⁰ cm⁻³) at high temperatures (~1000 K) to clarify the intrinsic thermoelectric properties of these materials. Furthermore, the experimental discovery of unusually large thermopower outputs from superlattices and two-dimensional electron gases in SrTiO₃ spurred substantial research efforts into SrTiO₃ superlattices and heterostructures for thermoelectric applications; for example, a superlattice composed of 1 unit cell (uc) SrTiO₃Nb₂O₅ and 10 uc SrTiO₃ exhibited giant thermopower, most likely due to an electron confinement effect. Although electron confinement is strongly correlated with the electronic structure, a full understanding of the fundamental electronic phase behavior of the SrTi₁₋ₓNbₓO₃ solid solution system has not yet been developed.

In this study, we clarify the thermoelectric phase diagram for the SrTi₁₋ₓNbₓO₃ (0.05 ≤ x ≤ 1) solid solution system, which we derived from the characterization of epitaxial films. We observed two thermoelectric phase boundaries in the system, which originate from the step-like decrease in carrier effective mass at x ~ 0.3 and from a local minimum in carrier relaxation time at x ~ 0.5. The origins of these phase boundaries are considered to be related to isovalent/heterovalent B-site substitution: parabolic Ti 3d orbitals dominate electron conduction for compositions with x < 0.3, whereas the Nb 4d orbital dominates when x > 0.3. At x ~ 0.5, a tetragonal distortion of the lattice, in which the B-site is composed of Ti⁴⁺ and Nb⁴⁺ ions, leads to the formation of tail-like impurity bands, which maximizes the electron scattering. These results provide a foundation for further research into improving the thermoelectric performance of SrTi₁₋ₓNbₓO₃. Published by AIP Publishing.

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decreases with increasing \( \text{Nb}^{4+} \) (x); on the other hand, heterovalent substitution, in which two \( \text{Ti}^{4+} \) or \( \text{Nb}^{5+} \) ions are substituted by adjacent \( \text{Ti}^{3+}/\text{Nb}^{5+} \) ions, can occur as shown in Fig. 1(c). Based on these considerations, we focused on the valence state change of Ti and Nb ions in the SrTi\( _{1-x}\)Nb\( _x\)O\( _3 \) ss.

In this study, we fabricated high-quality epitaxial films covering the full range of the SrTi\( _{1-x}\)Nb\( _x\)O\( _3 \) ss and analyzed their crystal lattice parameters, as well as their electrical properties such as carrier concentration, Hall mobility, and thermopower, from which the carrier effective masses and relaxation times were derived. During these experiments, we observed that two thermoelectric phase boundaries exist within the system, which originate from the step-like decrease in carrier effective mass at \( x \approx 0.3 \) and from the minimum in carrier relaxation time at \( x \approx 0.5 \). The origin of these phase boundaries was analyzed by considering the cases of isovalent/heterovalent B-site substitution. Parabolic Ti 3d orbitals were found to dominate electron conduction for compositions with \( x \approx 0.3 \), whereas the Nb 4d orbitals became more influential for compositions with \( x \approx 0.3 \). At \( x = 0.5 \), tetragonal distortion of the lattice, in which the B-site was composed of predominantly \( \text{Ti}^{4+}/\text{Nb}^{5+} \) ions, led to the formation of tail-like impurity bands, which maximized the electron scattering. The results obtained in this study may be used as a foundation for further work that seeks to improve the thermoelectric performance of SrTi\( _{1-x}\)Nb\( _x\)O\( _3 \).

II. EXPERIMENTS

Approximately 100 nm thick SrTi\( _{1-x}\)Nb\( _x\)O\( _3 \) (\( x = 0.05, 0.1, 0.2, 0.3, 0.4, 0.5, 0.55, 0.6, 0.7, 0.8, 0.9, \) and \( 1.0 )\) epitaxial films were fabricated by PLD using dense ceramic disks of an SrTiO\( _3 \)–SrNbO\( _3 \) mixture. We selected insulating (001) LaAlO\( _3 \) (pseudo-cubic perovskite, \( a = 3.79 \) Å) as the substrate. Growth conditions were precisely controlled, with a substrate temperature of 850 °C, oxygen pressure of \( \approx 10^{-4} \) Pa, laser fluence of 0.5–1 J cm\(^{-2}\) pulse\(^{-1}\), yielding a growth rate of 0.3 pm pulse\(^{-1}\). Details of the PLD growth process of SrTi\( _{1-x}\)Nb\( _x\)O\( _3 \) developed by our group have been published elsewhere.\(^7,20,21\)

Crystallographic analyses of the resultant films were performed by X-ray diffraction (XRD, Cu K\( _{\alpha} \), ATX-G, Rigaku) and scanning transmission electron microscopy (STEM). TEM samples were fabricated using a cryo ion slicer (IB-09060CIS, JEOL). The thin film was observed using scanning transmission electron microscope (STEM) (JEM-ARM200CF, JEOL Co. Ltd) operated at 200 keV. High-angle annular dark-field (HAADF) images were taken with the detection angle of 68–280 mrad. The electron energy loss spectra (EELS) were acquired in STEM mode by an Enfinium spectrometer (Gatan Inc.) with the energy resolution of 1 eV.

\( \sigma \), carrier concentration (n), and Hall mobility (\( \mu_{\text{Hall}} \)) were measured at room temperature by a conventional d.c. four-probe method, using an In–Ga alloy electrode with van der Pauw geometry. \( S \) was measured at room temperature by creating a temperature gradient (\( \Delta T \)) of \( \approx 4 \) K across the film using two Peltier devices (while using two small thermocouples to monitor the actual temperatures of each end of the SrTi\( _{1-x}\)Nb\( _x\)O\( _3 \) films). The thermo-electromotive force (\( \Delta V \)) and \( \Delta T \) were measured simultaneously, and \( S \)–values were obtained from the slope of the \( \Delta V–\Delta T \) plots.

Band structures for SrTi\( _{1-x}\)Nb\( _x\)O\( _3 \) (\( x = 0, 0.25, 0.5, 0.75, \) and \( 1.0 )\) were calculated based on the projector augmented-wave (PAW) method,\(^22\) as implemented using VASP code.\(^23\) For these calculations, we adopted Heyd-Scuseria-Ernzerhof (HSE) hybrid functionals,\(^24\) and a plane-wave cutoff energy of 550 eV and a \( 6 \times 6 \times 6 k \)-point mesh for cubic-perovskite
were employed in the total-energy evaluation and geometry optimization for the unit cells of SrTiO$_3$ and SrNbO$_3$, while special quasi-random structures (SQSs) were used in the case of SrTi$_{1-x}$Nb$_x$O$_3$ ($x = 0.25, 0.5$, and 0.75). The SQSs with eight Sr or Ti sites were constructed by optimizing the correlation functions of seven types of independent pairs using the CLUPAN code.\textsuperscript{25}

III. RESULTS AND DISCUSSION

Figure 2(a) summarizes the X-ray reciprocal space mappings (RSMs) around the (103) diffraction spot of LaAlO$_3$ (overlaid). Intense diffraction spots from (103) SrTi$_{1-x}$Nb$_x$O$_3$ are seen together with those from the LaAlO$_3$ substrate, indicating that incoherent hetero-epitaxial growth of the target materials occurred for all $x$ compositions. The peak positions of the diffraction spots from each composition correspond well with the cubic line ($q/4q_x = -3$), indicating that no epitaxial strain was induced in the films. It should be noticed that a slight tetragonal distortion was observed in the $x = 0.4$ ($c/a = 1.0057$) and $0.5$ ($c/a = 1.0050$) samples. From the RSMs of the SrTi$_{1-x}$Nb$_x$O$_3$ films, we extracted their lattice parameters using the formula $a = (2\pi nq_x / 2\pi q_x, 6\pi q_x q_y)^{1/3}$. Figure 2(b) plots the lattice parameters of the SrTi$_{1-x}$Nb$_x$O$_3$ film as a function of $x$: we observed an M-shaped trend, along with a general increase in the lattice parameter with increasing $x$. In order to analyze the changes in lattice parameter, we calculated the average ionic radii in the crystal structure and used Shannon’s ionic radii as a comparison.\textsuperscript{36} $\text{Sr}^{2+}$ (60.5 pm), $\text{Ti}^{4+}$ (67.0 pm), $\text{Nb}^{4+}$ (68.0 pm), and $\text{Nb}^{5+}$ (64.0 pm). In the ranges of $0.05 \leq x \leq 0.3$ and $x \geq 0.6$, the observed lattice parameters closely followed the heterovalent substitution line, suggesting that two $\text{Ti}^{4+}$ or $\text{Nb}^{4+}$ ions are substituted by adjacent $\text{Ti}^{4+}$/$\text{Nb}^{5+}$ ions.\textsuperscript{27} On the other hand, at $x=0.4$ and $0.5$, the observed lattice parameter corresponded well with the isovalent substitution line; moreover, at $x=0.5$, the B-site occupation of $[\text{Ti}^{4+}/\text{Nb}^{5+}]$ was almost 100%, as shown in Fig. 2(c).

In order to further clarify the occupation of B-sites by $[\text{Ti}^{4+}/\text{Nb}^{4+}]$, we analyzed the atomic arrangement and electronic structure of the $x=0.5$ film by STEM and EELS. Figure 3(a) shows a cross-sectional HAADF-STEM image of an SrTi$_{0.5}$Nb$_{0.5}$O$_3$ film. Periodical mismatch dislocations with an interval of $\sim 8.5$ nm were seen at the heterointerfaces. If the strain in the thin-film were fully relaxed by such misfit dislocations, it would be possible to calculate the spacing between dislocations ($d$) from $d = b/\delta$, where $b$ is the Burgers vector and $\delta$ is the lattice mismatch between thin-film and substrate.\textsuperscript{28} using the lattice parameters obtained from XRD [$\delta = (q_{x,sub} - q_{x,film}) / q_{x,film} = +0.0435$], a dislocation spacing of $8.7$ nm was estimated, suggesting that the dislocations do fully relax the strain in the film.

Although superspots originating from (111) diffractions are often observed in AB$_{0.5}$B'$_{0.5}$O$_3$ compositions that crystallize in B-site–ordered double perovskite structures,\textsuperscript{29} these were not observed in the SrTi$_{0.5}$Nb$_{0.5}$O$_3$ film [Fig. 3(b)], most likely due to the slight tetragonal distortion of the crystal structure. Figure 3(c) shows the EELS spectra acquired along the Ti L3 (c) and O K edges (d); the reported EELS spectra of $\text{Ti}^{4+}/\text{Ti}^{3+}$ (Ref. 30) and $\text{Nb}^{4+}/\text{Nb}^{5+}$ (Ref. 31) are also plotted for comparison. In the Ti L3 edge spectra (c), $t_{2g}$ and $e_g$ peak splitting was clearly observed for Ti L3, indicating that the dominant valence state of Ti is $4+$. In the O K edge spectra (d), two intense peaks (assigned as A and B) were clearly seen, with the intensity of peak B being higher than A, which was noted to be a characteristic feature of $\text{Nb}^{4+}$ in a previous study;\textsuperscript{31} the peak intensity ratio A/B was calculated to be 0.66, which roughly corresponds with the $\text{Nb}^{4+}$ spectrum (0.66). From these results, we conclude that isovalent substitution of $\text{Ti}^{4+}/\text{Nb}^{4+}$ dominates in the $x=0.5$ composition.

We then measured $\sigma$, $n$, and $\mu_{\text{Hall}}$ by Hall effect measurements. Generally, $\sigma$ was observed to increase with increasing $x$, as shown in Fig. 4(a); additionally, it was noted that there was a sharp increase in $\sigma$ between $0.5 < x < 0.6$, suggesting the existence of an electronic phase boundary at $x \sim 0.5$. $n$ was also measured to increase with increasing $x$, which almost corresponds with the nominal value ($n=x$ in a unit cell) [Fig. 4(b)]. For the mobility, as shown in Fig. 4(c), $\mu_{\text{Hall}}$ for compositions with $x<0.5$ remained almost constant at $\sim 6 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, which is consistent with other reported values.\textsuperscript{32,33} As was observed for $\sigma$, a sharp increase in $\mu_{\text{Hall}}$ from $\sim 5 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ to $\sim 10 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ was observed between compositions with $x=0.5$ and $x=0.6$, after which the values steadily increased to $\sim 12 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at $x=1$ (cf. a value of $\sim 14 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ measured for this composition in a previous study\textsuperscript{76}). From these results, we can conclude that the small $\mu_{\text{Hall}}$ dominates the electronic phase boundary at $x \sim 0.5$. 

![FIG. 2. Crystallographic characterization of the SrTi$_{1-x}$Nb$_x$O$_3$ epitaxial films on a (001) LaAlO$_3$ single-crystal substrate. (a) X-ray reciprocal space mappings around the (103) diffraction spot of the SrTi$_{1-x}$Nb$_x$O$_3$ epitaxial films. The location of the LaAlO$_3$ diffraction spot ($2\theta/2\theta = (-2.64, 7.92)$, corresponds with the pseudo-cubic lattice parameter of LaAlO$_3$ ($a = 3.79\text{nm}$). Red symbols (+) indicate the peak positions of the SrTi$_{1-x}$Nb$_x$O$_3$ epitaxial films. (b) Changes in the lattice parameters of the SrTi$_{1-x}$Nb$_x$O$_3$ films (circles, left axis), with superimposed isovalent/heterovalent substitution lines (black line: isovalent substitution, gray line: heterovalent substitution, right axis), calculated using Shannon’s ionic radii.\textsuperscript{26} (c) Change in the B-site occupation by $[\text{Ti}^{4+}/\text{Nb}^{5+}]$ derived from the data in (b).]
In order to further elucidate the origin of the electronic phase boundary, we took measurements of $S$ across the composition range: by measuring the values of both $n$ and $S$, we were then able to calculate the density of states effective mass ($m^*$), as outlined in equations (1)–(3) below. Not surprisingly, $S$ was found to monotonically decrease with increasing $x$, as shown in Fig. 4(d). We then calculated $m^*$ using the following relation between $n$ and $S$:

$$S = \frac{k_B}{e} \left( \frac{r + 2}{r + 1} F_{r+1}(\xi) - \xi \right),$$

where $k_B$, $\xi$, $r$, and $F_r$ are the Boltzmann constant, chemical potential, scattering parameter of relaxation time, and Fermi integral, respectively. $F_r$ is given by

$$F_r(\xi) = \int_0^\infty \frac{x^r}{1 + e^{x-\xi}} dx$$

and $n$ by

$$n = 4\pi \left( \frac{2m^* k_B T}{\hbar^2} \right)^{\frac{3}{2}} F_2(\xi),$$

where $h$ and $T$ are the Planck constant and absolute temperature, respectively. Using the equations (1)–(3), we obtained the values of $m^*$, which are presented in Fig. 4(e): a step-like decrease of $m^*$ was observed with increasing $x$, as shown in Fig. 4(d). We then calculated $m^*$ for compositions with $x < 0.3$ was calculated to be $\sim 1.1 m_0$, which corresponds well with the values reported by Okuda et al., while for compositions with $x > 0.3$, $m^*$ was $\sim 0.7 m_0$. At $x = 1$ (SrNbO$_3$), $m^*$ further decreased to 0.5 $m_0$, agreeing well with the values calculated in another study using spin-polarized DFT calculations (0.4 $m_0$). We
speculate that there is a percolation threshold for compositions with \( x \sim 0.3 \) and that parabolic Ti 3 d orbitals dominate electron conduction when \( x < 0.3 \), whereas the Nb 4 d orbitals determine electron conduction when \( x > 0.3 \). Similar percolation threshold was observed in the other insulating/conducting oxide system. \(^{36}\) Finally, the relaxation time, \( \tau \), was extracted from the \( \mu_{\text{Hall}} \) and \( m^* \) values (\( = e \cdot \tau \cdot m^* \))—these results are shown in Fig. 4(f). The \( \tau \) values in the range \( 0.05 \leq x \leq 0.2 \) are \(~4\) fs, followed by a drop, reaching a minimum value of \(~2\) fs at \( x = 0.5 \). After \( x = 0.5 \), \( \tau \) initially increases sharply to 3.5 fs at \( x = 0.6 \), with a much more gradual increase with \( x \) composition in the range \( 0.6 \leq x \leq 0.9 \). Finally, there was a slight drop in \( \tau \) value to \( 3.2 \) fs for the \( x = 1 \) composition. The trend’s trough shape, in particular, the sharp increase of \( \tau \) from \(~2\) to \(~3.5\) fs in the range \( 0.5 < x < 0.6 \), clearly indicates that \( \tau \) dominates the electronic phase boundary at \( x \sim 0.5 \). Yamamoto et al. reported that \( \tau \) of the SrTi\(_{0.8}\)Nb\(_{0.2}\)O\(_3\) epitaxial films was significantly reduced by Sr-site substitution with Ca or Ba, due to the occurrence of tetragonal lattice distortion. \(^{20}\) Since slight tetragonal distortion was observed in the \( x = 0.4 \) and 0.5 films, as shown in Fig. 2(a), we speculate that the tetragonal distortion also minimizes the \( \tau \) in this set of thin-films.

FIG. 5. Thermoelectric phase diagram for the SrTiO\(_3\)-SrNbO\(_3\) solid solution system. The thermoelectric power factor (\( S^2 \sigma \)) of the SrTiO\(_3\)-SrNbO\(_3\) solid solution system is plotted, alongside the previously reported values. \(^{36}\) The \( x \) dependence of \( S^2 \sigma \) is shown in the inset. The system’s thermoelectric phase boundaries are clearly seen at \( x \sim 0.3 \) and \(~0.5\).

FIG. 6. Calculated density of states (DOS) for SrTi\(_{1-x}\)Nb\(_x\)O\(_3\) ss (\( x = 0, 0.25, 0.5, 0.75, \) and 1). (a) Total DOS, (b) and (c) orbital-projected DOS. The energy origin was set at the Fermi level, \( E_F \), indicated by the dotted line.
Then, we plotted the thermoelectric power factor \((S^2\sigma)\) of the \(\text{SrTi}_{1-x}\text{Nb}_x\text{O}_3\) ss together with values reported in the previous studies (Fig. 5). In the region around \(x \sim 0.05\) where the observed data (red circles/red line) overlap with the reported values from \(10\) (white circles/grey line), the two data sets match well. There are two local maxima in \(S^2\sigma\)—first at \(x \sim 0.05\left(\sim 2.5 \text{ mW m}^{-1} \text{ K}^{-2}\right)\) and second at \(\sim 0.55\) \((\sim 0.9 \text{ mW m}^{-1} \text{ K}^{-2})\). We also observed two thermoelectric phase boundaries in the system, occurring in the regions around \(x \sim 0.3\) and \(x \sim 0.5\), as shown in the inset. Since \(S\) is strongly correlated with \(m^*\), the sharp decrease in \(m^*\) at \(x \sim 0.3\) [Fig. 4(e)] accounts for the concomitant reduction in \(S\). In addition, the small value of \(\tau\) at \(x \sim 0.5\) (where \(\tau\) was measured to reach a minimum) contributed greatly to the observed reduction in \(S\). In conclusion, in order to maximize the thermoelectric performance of the \(\text{SrTi}_{1-x}\text{Nb}_x\text{O}_3\) ss, the use of compositions with \(x \leq 0.2\) would be the most suitable.

Finally, we calculated the band structures for \(\text{SrTi}_{1-x}\text{Nb}_x\text{O}_3\) \((x = 0, 0.25, 0.5, 0.75, \text{ and } 1)\) in order to elucidate the origin of the two electronic phase boundaries using \(2 \times 2 \times 2\) supercell model structures. Figure 6 summarizes the resultant total and orbital-projected density of states (DOS) of the \(\text{SrTi}_{1-x}\text{Nb}_x\text{O}_3\) \((x = 0, 0.25, 0.5, 0.75, \text{ and } 1)\) solid solution crystal. Moreover, the analyses of the B-site ion valence states in this study provide valuable information that may be partly originated from the restriction of the supercell size, which may be insufficient to represent the random B-site ionic arrangements. Larger supercell model would help to understand the origin of the two electronic phase boundaries.

### IV. SUMMARY

In summary, we have clarified the thermoelectric phase diagram for the \(\text{SrTi}_{1-x}\text{Nb}_x\text{O}_3\) \((0.05 \leq x \leq 1)\) solid solution system through characterization of epitaxial thin-films. We observed two thermoelectric phase boundaries in the system, which originate from the combination of a step-like decrease in carrier effective mass at \(x \sim 0.3\) with the local minimum carrier relaxation time at \(x \sim 0.5\). The origins of these electronic phase boundaries were analyzed in the context of isovalent/heterovalent B-site substitution. Parabola-shaped Ti 3 d orbitals dominate electron conduction for compositions \(0 < x < 0.3\), whereas the Nb 4d orbitals dominate when \(x > 0.3\). At \(x \sim 0.5\), a tetragonal distortion of the lattice occurs, in which the B-site is composed of predominantly \([\text{Ti}^{4+}/\text{Nb}^{4+}]\) ions, leading to the formation of tail-like impurity bands in the density of states, which maximizes electron scattering.

These findings may prove useful in further improving and optimizing the thermoelectric performance of \(\text{SrTi}_{1-x}\text{Nb}_x\text{O}_3\). Moreover, the analyses of the B-site ion valence states in this study provide valuable information that could be used in the design of other functional materials based on transition metal oxides.

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