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Thermopower analysis of metal-insulator transition temperature modulations in vanadium dioxide thin films with lattice distortion

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Insulator-to-metal (MI) phase transition in vanadium dioxide (VO2) thin films with controlled lattice distortion was investigated by thermopower measurements. VO2 epitaxial films with different crystallographic orientations, grown on (0001) α-Al2O3, (1120) α-Al2O3, and (001) TiO2 substrates, showed significant decrease of absolute value of Seebeck coefficient (S) from ~200 to 23 μV K⁻¹, along with a sharp drop in electrical resistivity (ρ), due to the transition from an insulator to a metal. The MI transition temperatures observed both in ρ(T) and S(Ts) for the VO2 films systematically decrease with lattice shrinkage in the pseudorutile structure along the c axis, accompanying a broadening of the MI transition temperature width. Moreover, the onset Ts, where the insulating phase starts to become metallic, is much lower than the onset Tc. This difference is attributed to the sensitivity of S for the detection of hidden metallic domains in the majority insulating phase, which cannot be detected in ρ measurements. Consequently, S measurements provide a straightforward and excellent approach for a deeper understanding of the MI transition process in VO2.

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I. INTRODUCTION

Vanadium dioxide (VO2), a strongly correlated oxide, shows an insulator-to-metal (MI) transition when heated to its transition temperature (TMI) of 68 °C [1] due to crystal-structure and electronic-structure changes. In the high-temperature metallic phase at T > TMI, VO2 has a rutile-type tetragonal structure, while in the low-temperature insulating one at T < TMI, the vanadium ions form a dimer along with the position shifting from linear chains to zigzag pattern, resulting in a monoclinic structure [2]. This structural transformation accompanies a dramatic change in the 3d-band configuration in the 3d-band configuration with appearance of bandgap ~0.6 eV [3], yielding abrupt changes of both electrical resistivity on the order of 10⁴–10⁶ and the optical transmission in the infrared region. TMI can be varied over a wide range below room temperature (RT) by aliovalent ion doping [4], external strain [5], and an electric field application [6]. These features of the MI transition in VO2 have proved to be useful in electrical and optical switching devices [7,8].

However, the nature of the MI transition in VO2 is still quite unclear [9,10] and, consequently, potential device applications remain underdeveloped. One reason for this situation is that VO2 has an electronic phase separation at the nanoscale region [11] and a complicated phase diagram with internal strain [12], which significantly makes it difficult to understand and control the MI transition characteristics. In a temperature range around the TMI, the metallic tetragonal and insulating monoclinic phases coexist randomly, where the electronic-transport properties are largely influenced by the domain configuration [13]. Especially in thin films, the polycrystalline domains with epitaxial strains and defects cause discontinuous and irregular phase domain structures, which result in the complex and broad MI transition [14].

The phase domain structures and their electronic behaviors have been experimentally investigated by electrical-transport measurement using nanowires at the nanoscale [15] or nanoscopic imaging on thin-films during the MI transitions [16]. In the electrical-transport measurement on VO2 nanowires, discrete resistivity jump was observed, which suggests the important role of the electronic behaviors of each domain in the electrical properties. The nanoscopic imaging on thin films revealed that both electronic and structural changes exhibit phase coexistence leading to percolation conduction depending on the domain distribution. It is therefore critically important to understand the evolution of coexisting metallic and insulating domains with spatial phase inhomogeneity at the nanoscale, and further characterization that combines several experimental techniques is necessary to unveil and control the MI transition process.

We have focused on thermopower measurement as a physical property to investigate electronic structure change around the MI transition of VO2 [17] because Seebeck coefficient (S) changes significantly due to the electronic structure reconstruction across the TMI. The general expression for the electronic contribution to the S is

\[ S = -\frac{k_B}{e} \int \left( E - E_F \right) \frac{\sigma(E)}{T} \, dE, \]

where \( k_B \) is the Boltzmann constant and \( E_F \) is the Fermi energy [18]. Here, \( \sigma \) is described as an integral over the single carrier without electron correlation effects

\[ \sigma = \int \sigma(E) \, dE = e \int g(E) \mu(E) f(E) [1 - f(E)] dE, \]

where \( g(E) \) is the density of states (DOS), \( \mu(E) \) is the mobility, \( f(E) \) is the Fermi distribution function, and \( \sigma(E) \) is energy-dependent conductivity.

The S of semiconductors is expressed as

\[ S = \frac{k_B}{e} \left( \frac{E_F - E_C}{k_B T} + A \right), \]

where \( A \) is the Seebeck coefficient at the Fermi level.

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considering Eqs. (1) and (2) in the framework that only
electrons contribute to the $S$, and $E_F$ lies near the conduction
band edge ($E_C$). By using the relation of \( n_e = N_c e^{-\frac{E_F-E_C}{k_BT}} \) in
Eq. (3), the $S$ can be simplified to
\[
S = \frac{k_B}{e} \left( \ln \frac{N_c}{n_e} + A \right)
\]
where $N_c$ is the effective DOS of the conduction band, $n_e$ is
the carrier concentration, and $A$ is a transport constant that
depends on the dominant scattering mechanism [19]. Meanwhile,
$S$ for metals or degenerate semiconductors at low temperature ($E_F \gg \text{thermal energy \ } k_BT$) is basically expressed as
\[
S = \frac{\pi^2 k_B^2 T}{3e} \left( \frac{\partial \log \left( \sigma(E) \right)}{\partial E} \right)_{E=E_F}
\]
in Mott’s equation [20,21].

The $S$ of metals are typically small and keep constant by
reflecting the energy differential of DOS around the $E_F$, but
those of the semiconductors drastically change, depending on
$n_e$, by reflecting the shape of the DOS around the conduction
band bottom due to the $E_F$ shifts by carrier doping [22]; the
doped carriers may be unintentionally caused by the existence
of oxygen vacancies for VO$_2$ thin film [23]. Previously, we
systematically measured the $S$ of electron-doped V$_{1-x}$W$_x$O$_2$
epitaxial films with different doping levels [17] and clarified that
the metallic electronic structure clearly changes into an
insulating electronic structure with a parabolic DOS around
the conduction band bottom, where the $S$ for insulating phase
systematically changes by reflecting the derivative of the DOS
around the $E_F$.

$S$ measurements have been actively performed to in-
vestigate the MI transition characteristics of bulks [24,25],
nanowires [26], and thin-films [27] of VO$_2$, where the $S$
significantly change from 200–400 $\mu$V K$^{-1}$ for the insulating
phase down to $\sim 20 \mu$V K$^{-1}$ for the metallic phase. In addi-
tion, $S$ measurements were recently applied to investigate the
metal-insulator domain arrays in single-crystal microbeams
[26] and the effect of the domain inhomogeneity in thin films
[27], which demonstrates the unusual $S$: the complications
of the phase coexistence near the domain wall may cause a
deviation of the $S$ from a normal behavior in each pure domain.
The ability of sensitive detection of the metallic and insulating
phases around the $T_{MI}$ should make $S$ measurements suitable
to investigate the MI transition process of VO$_2$.

In this paper, we investigate the MI phase transition of VO$_2$
thin films with regulated lattice distortion via $S$
measurements. We controlled the epitaxial orientations of the
VO$_2$ films grown on single crystalline substrates of (0001) \( \alpha\)-Al$_2$O$_3$, (1120) \( \alpha\)-Al$_2$O$_3$, and (001) rutile-TiO$_2$.
Then we compared their MI transition process by simulta-
neously measuring the $S$ and electrical resistivity ($\rho$).

II. EXPERIMENTAL

VO$_2$ epitaxial films were grown on (0001) \( \alpha\)-Al$_2$O$_3$,
(1120) \( \alpha\)-Al$_2$O$_3$, and (001) TiO$_2$ substrates by pulsed laser
deposition. A KrF excimer laser with a laser energy fluence of 3 J cm$^{-2}$ was used to ablate a V$_2$O$_5$ target disk. The growth temperature was fixed at 500$^\circ$C, and the oxygen pressure ($P_{O_2}$) was optimized at 2.0 Pa because the resistivity-change ratio from an insulator to a metal across the $T_{MI}$ is extremely sensitive to $P_{O_2}$ during thin film growth [28]. After deposition at a rate of approximately 2 nm min$^{-1}$, the films were cooled to RT under the same oxygen pressure. The film thickness was set at 30–40 nm, which was characterized by x-ray reflectivity measurements. The crystal structures, including the crystallographic orientation and the mosaicity of the crystallites, were investigated by high-resolution x-ray
diffraction (XRD; anode radiation: monochromatic CuK$_\alpha$, ATX-G, Rigaku Co.) at RT. The film surface structures were
observed by atomic force microscopy (AFM).

$S$ was measured by giving a temperature difference ($\Delta T$)
of $\sim 4$ K in the film with In-Ga alloy electrodes. Here we
used two Peltier devices placed under the film, where one
side cooled and the other side heated a sample, without
changing the temperature at the center [29]. We used two Cu
leads to measure the thermoelectromotive force ($\Delta V$). Thus, the
observable slope of $\Delta T/\Delta V$ plots should be expressed as
$S_{VO2}\text{film} + S_{Cu}$ (see Supplemental Material [30] for the
calculation of $S_{VO2}\text{film} + S_{Cu}$). Since $S$ of Cu is very small
($\sim +1.5 \mu$V K$^{-1}$) as compared to that of VO$_2$, $S$ of the Cu
leads was ignored in this study. The actual temperatures of
both sides of the VO$_2$ film surface were monitored by
two tiny thermocouples separated by a gap $\sim$7 nm. The
thermoelectromotive force ($\Delta V$) and $\Delta T$ were simultaneously
measured at almost the same positions, and the $S$ were
obtained from the linear slope of the $\Delta V$–$\Delta T$ plots [17].
It should be noted that the sapphire substrate is not suited
for $S$ measurements because of high thermal conductivity, but
we measured the $\Delta V$ at the highly steady state after making
the $\Delta T$ up to 10 K in sapphire substrate, which enables
accurate $S$ measurements of VO$_2$ thin films prepared on
sapphire substrates. Meanwhile, $\rho$ was measured by a dc four
probe method using Au electrodes.

III. RESULTS

A. Epitaxial orientation control of VO$_2$ thin films

Figure 1 shows the (a)–(c) out-of-plane and (d)–(f) in-plane
XRD patterns of VO$_2$ films grown on the substrates of (a)
and (d) (0001) \( \alpha\)-Al$_2$O$_3$, (b) and (e) (1120) \( \alpha\)-Al$_2$O$_3$, and
(c) and (f) (001) TiO$_2$. In addition, Fig. 2 summarizes (a)–(c)
AFM images and reflection high-energy electron diffraction
(RHEED) patterns, and (d)–(f) schematic epitaxial relation
models of each film. For the out-of-plane XRD pattern of
VO$_2$ film grown on (0001) \( \alpha\)-Al$_2$O$_3$ substrate [Fig. 1(a)],
diffraction peaks of 0k0 ($k = 2$ and 4) for monoclinic VO$_2$ are
observed along with those of the (0001) \( \alpha\)-Al$_2$O$_3$ substrate.
The average crystalline tilt angle, full width at the half
maximum (FWHM) value of the 020 diffraction peak [20-fixed
omega scan, inset of Fig. 1(a)], is 0.17$^\circ$. For the in-plane XRD
pattern [Fig. 1(d)], 200 VO$_2$ and 1120 \( \alpha\)-Al$_2$O$_3$ diffraction
peaks are observed, which confirms the epitaxial relationship
of (010)[100] VO$_2$||(0001)[1120] \( \alpha\)-Al$_2$O$_3$. These results are
also consistent with the RHEED pattern [Fig. 2(a)]. However,
the in-plane 20-phi scan of the 200 diffraction peak [inset of
Fig. 1(d)] shows six diffraction peaks instead of the twofold
rotational symmetry of a monoclinic lattice, indicating that

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the VO₂ film has three domain structures with 60° rotational periodicity, presumably due to domain matching between the film and substrate. Meanwhile, the broad 200 diffraction peaks, which indicate a weak in-plane orientation, should originate from the misaligned orientations between (100) VO₂ and (1120) α-Al₂O₃ by 2.1° due to the monoclinic structure, as shown in the schematic epitaxial relation model [Fig. 2(d)].

Reflecting the rotational domain structure, small crystalline grains with size of ~20 nm are observed in the topographic AFM image [Fig. 2(a)].

On the other hand, in the case of the VO₂ film on the (1120) α-Al₂O₃ substrate, h00 (h = 2, 3, and 4) diffraction peaks of monoclinic VO₂ are observed along with intense peaks from the (1120) α-Al₂O₃ substrate in the out-of-plane XRD pattern [Fig. 1(b)]. Because the double lattice spacing (h = odd numbers) along the a axis of monoclinic VO₂ originates from the formation of the vanadium-ion dimer, the existence of the 300 diffraction peak confirms that the structural transition temperature (monoclinic-to-tetragonal phase transition) is above RT. The average crystalline tilt angle is 0.16° [inset of Fig. 1(b)]. The in-plane XRD pattern [Fig. 1(e)] shows clear diffraction peaks of 020 VO₂ and 0006 α-Al₂O₃. The in-plane φ scan of the 020 diffraction shows a twofold rotational symmetry with 180°, originating from the monoclinic symmetry of the VO₂ lattice. These results substantiate that the VO₂ film is grown on the (1120) α-Al₂O₃ substrate with an epitaxial relationship of (100)[010] VO₂ || (1120)[0001] α-Al₂O₃ [Fig. 2(e)], which is also confirmed in the RHEED pattern [Fig. 2(b)]. For the film surface structure of the VO₂ film on the (1120) α-Al₂O₃ substrate, reflecting the anisotropic crystal structure of VO₂ along the [102] direction, rectangular shaped grains are observed in the AFM image [Fig. 2(b)].

For the out-of-plane XRD pattern of VO₂ film grown on (001) TiO₂ substrate [Fig. 1(c)], the 402 diffraction peak of VO₂ is observed along with the 002 diffraction peak of TiO₂. The 402 diffraction corresponds to 002 diffraction of tetragonal VO₂, which has the same rutile structure with the TiO₂ substrate. For the in-plane XRD pattern [Fig. 1(f)], the 022 diffraction of monoclinic VO₂, corresponding to the 220 diffraction peak of the TiO₂ substrate, overlaps with the 220 diffraction peak of the TiO₂ substrate. The asymmetric φ scan for the 420 (monoclinic) [222 (tetragonal)] diffraction peak, shown in the inset, exhibits a fourfold rotational symmetry with 90°, confirming that VO₂ film grows on isostructural TiO₂ (001) substrates with an epitaxial relationship between the film and TiO₂.
Here we normalized \( \rho \) to the transition temperatures observed in undoped VO2 [31]. In general, \( T_\rho \) depends on the \( c \) axis length of the rutile structure in VO2 because the V-V chain length along the \( c \) axis plays an important role in the MI transition [5,32]. It should be noted that the transition from an insulator to a metal in bulk VO2 simultaneously induces shrinkage by 1% along the \( c \) axis of the rutile structure [23,33], indicating that the lattice shrinkage would drive the electronic phase toward the metal.

To visualize the MI transitions clearly, Figs. 3(c) and 3(d) plot the temperature derivative curves of \( d[\log \rho/\rho_{35\,\text{C}}]/dT \) and \( dS/dT \), respectively. The arrows in Fig. 3(c) denote the MI transition temperatures observed in \( \rho(T_\rho) \), which are defined as the peak position in the \( d[\log \rho/\rho_{35\,\text{C}}]/dT \) versus \( T \), for the VO2 films on the substrate [i.e., \( T_\rho \) of the VO2 film on (0001) \( \alpha\text{-Al}_2\text{O}_3 \) and (001) TiO2]. The difference in \( T_\rho \) should be related to the interfacial strain effect between the films and the substrates because it has been reported that strains imposed on VO2 films by substrates significantly affect \( T_\rho \) [31].

FIG. 2. (Color online) (a)–(c) Topographic AFM images and (d)–(f) schematic epitaxial relations of monoclinic (m-) VO2 epitaxial films grown on substrates of (a) and (d) (0001) \( \alpha\text{-Al}_2\text{O}_3 \), (b) and (e) (1120) \( \alpha\text{-Al}_2\text{O}_3 \), and (c) and (f) (001) TiO2. RHEED patterns of each VO2 film are superimposed in (a)–(c).

B. Metal-insulator transition characteristics

Figures 3(a) and 3(b) summarize \( \rho - T \) curves normalized by \( \rho \) at 35 \( ^\circ \text{C} \) (\( \rho/\rho_{35\,\text{C}} \)) and \( S - T \) curves of the VO2 epitaxial films, respectively, where it should be noted that there has been no report on the comparison of the MI transition characteristics for the VO2 films with different crystallographic orientations. Here we normalized \( \rho \) to show the change of \( T_{\text{MI}} \) clearly, because \( \rho \) of the VO2 epitaxial films were scattered as shown in the inset of Fig. 3(a), where \( \rho \) should be influenced by grain-boundary scattering due to different grain sizes and different epitaxial orientations of the VO2 films. The average \( \rho_{35\,\text{C}} \) was 1.95 \( \Omega \) cm with a standard deviation of 1.12 \( \Omega \) cm. The average \( \rho \) was confirmed to be consistent with previously reported values of VO2 films [27]. Meanwhile, \( S \) is a physical property that is not affected by grain boundary scattering and makes it easy to directly compare MI transition characteristics. Both \( \rho - T \) and \( S - T \) were measured along the [010], [010], and [011] directions for monoclinic VO2 films grown on (0001) \( \alpha\text{-Al}_2\text{O}_3 \), (1120) \( \alpha\text{-Al}_2\text{O}_3 \), and (001) TiO2, respectively. As the temperature increases, all the VO2 films show a sharp drop (a three-digit decrease) in \( \rho - T \) [Fig. 3(a)] due to the transition from an insulator to a metal. For the \( S - T \) curves [Fig. 3(b)], \( |S| \) of all the VO2 films start to decrease from 220 \( - 240 \mu \text{V K}^{-1} \) for the insulating phase down to 23 \( \mu \text{V K}^{-1} \) for the metallic phase [17]. The \( |S| \) of the insulating phase at low temperature around RT increases linearly with temperature, indicating that the insulating phase is a degenerate semiconductor, while the constant \( S \) for the metallic phase (23 \( \mu \text{V K}^{-1} \)) agrees well with the previously reported \( S \) of 20 \( \mu \text{V K}^{-1} \) for those of undoped VO2 bulks [24,25], microbeams [26], and thin films [27].

To estimate the \( c \) axis length, the monoclinic structure was converted into the pseudorutile structure. Figure 4(a) depicts the relations between \( T_\rho \) and the \( c \) axis lattice parameters estimated from the XRD patterns of the VO2 films (Fig. 1) as well as that of bulk VO2 [34]. \( T_\rho \) of VO2 films with different crystallographic orientations monotonically decreases as the \( c \) axis lattice parameters shrink in the pseudorutile structure compared to those of bulk VO2. Because the film on (0001) \( \alpha\text{-Al}_2\text{O}_3 \) should be structurally relaxed as a result of the large lattice mismatch and different crystallographic symmetry, \( T_\rho \) is comparable to that of bulk VO2. On the other hand, the films on (1120) \( \alpha\text{-Al}_2\text{O}_3 \) and (001) TiO2 are partially strained, and \( T_\rho \) consequently decreases. Meanwhile, the transition temperatures observed in \( S(T_S) \), which are determined as peak positions in \( dS/dT \) versus \( T \) [Fig. 3(d)], also decreases as the \( c \) axis length decreases [Fig. 4(b)]. These results indicate that the distortion along the \( c \) axis of the rutile

\[ T_{\text{MI}} \]
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FIG. 3. (Color online) (a) and (b) Temperature dependences of electrical resistivity ($\rho$) and Seebeck coefficient ($S$) for VO$_2$ epitaxial films grown on (0001) $\alpha$-Al$_2$O$_3$ (blue), (1120) $\alpha$-Al$_2$O$_3$ (red), and (001) TiO$_2$ (green), respectively. (a) $\rho-T$ curves normalized by $\rho$ at 35°C ($\rho_{35°C}$) and (b) $S-T$ curves measured by increasing $T$ from RT. Temperature derivative plots of (c) $d\log(\rho/\rho_{35°C})/dT$ and (d) $dS/dT$. The arrows indicate the positions of $T_r$ and $T_S$. The onset and offset temperatures of the transition from an insulator to a metal are marked by open and closed triangle symbols, respectively, and the color bars indicate the temperature range of the MI transition.

IV. DISCUSSION

To more clearly show the MI transition process, onset and offset temperatures of $T_r$ and $T_S$ for the transition from an insulator to a metal are denoted by open and closed triangles, respectively, in Figs. 3(c) and 3(d). Color bars indicate the temperature range of the MI transition width where the metallic and insulating phases should coexist. It is noteworthy that the MI transition temperature width becomes broader as $T_r$ and $T_S$ decrease, presumably due to the distortion of the $c$ axis length in the rutile structure of the films, which leads to an inhomogeneous domain distribution. Although the offset $T_r$ and $T_S$ are almost the same, the onset $T_r$ is much higher than the onset $T_S$, (i.e., the transition temperature where the insulating phase begins to transform into metallic phase is higher for the $\rho$ measurement compared to that for the $S$ measurement).

One plausible reason for this difference should come from the domain distribution of the metallic and insulating phases at the nanoscale [15]. Figure 5 schematically depicts the domain configurations at each temperature (a)–(d) around the MI transition according to a previous report [15] and the corresponding changes in $\rho-T$ and $S-T$ as functions of temperature. As the temperature increases from (a) to (d), the majority phase changes from (a) an insulator to (d) a metal, where the metallic and insulating domains coexist at (b) and (c); metallic domains first form inside the insulating phase, then some domains connect with each other, and finally, the entire region becomes a metallic phase. It has been reported that VO$_2$ films show percolation conduction, depending on the domain configuration, and $\rho$ significantly decreases when the metallic domains are connected with each other at (c)
starts to decrease from (b). Since the MI transition [15], and corresponding changes in $\rho$–$T$ and $S$–$T$. Majority phase changes from (a) an insulator to (d) a metal with increase of temperature, through the distribution of (b) locally separated metallic domains and (c) connected metallic domains in insulating domains. Open and closed triangle symbols indicate onset and offset temperatures of the transition from an insulator to a metal. Since $S$ can sensitively detect the metallic phase remaining in the majority insulating phase, which cannot be detected in resistivity, $|S|$ starts to decrease from (b).

[15], i.e., even though the metallic domain exists in the dominant insulating domain in (b), the contribution of the metallic domain to the conduction is extremely small due to the huge difference in resistivity between metallic and insulating phases in the order of $10^4$–$10^5$. $S$ measured in the VO$_2$ film at the temperature around $T_{MI}$ should also come from the combined voltages developed across the metallic and insulating domains. Here we roughly assume the domain distribution at (b) and (c); the metallic and insulating domains are mainly connected in series at (b), while they are linked in parallel at (c), in order to briefly discuss the effect of coexistence of metallic and insulating domains on $S$. It should be pointed out that the $S$ is complex in the multiphase materials, where the complexity originates from conductivity-weighted $S$ of the components for parallel-path arrangements and thermal conductivity for series arrangements. Thermal conductivity of VO$_2$ films increases by 60% in the metallic phase from the insulating one [35], which could be comprehended as the contribution of the increased electrical conductivity, assuming that the lattice contribution of thermal conductivity remains the same across the MI transition. Although there is no such simple description for $S$ in the multiphase materials, we take no account of the difference of thermal conductivity for the discussion. The $S$ at (b) can be simply expressed by the equation of $|S| = x \cdot |S_M| + (1 - x) \cdot |S_I|$ in the series model, where $x$ is defined as the effective volume fraction of metallic domains against the insulating one. When neglecting the contribution from the metal-insulator domain walls, the total $S$ are a sum of the contributions from the metallic and insulating domains, but in the case of $|S|$, the difference between those of the metal $23 \mu$V K$^{-1}$ and the insulator ($\sim 200 \mu$V K$^{-1}$) is not large, which should allow the hidden metallic phase to be detected in the majority insulating phase at (b) using $S$ measurements. Meanwhile, the $S$ at (c) can be expressed by the equation of $|S| = (\sigma \cdot |S_M| + (1 - \sigma) \cdot |S_I|) / (\sigma \cdot \sigma_M + (1 - \sigma) \cdot \sigma_I)$ in the parallel model. Since the $S$ largely depends on the each conductivity ($\sigma$) in the parallel circuit, the total $S$ is more sensitive to that of metallic domains when they are connected with each other at (c). These considerations suggest that the MI transition process is much clearer when following the measurement of $S$ than that of $\rho$.

V. CONCLUSION

In summary, we investigated the MI phase transition of VO$_2$ thin films with controlled lattice distortion by $S$ measurements. VO$_2$ epitaxial films with different crystallographic orientations were fabricated on (0001) $\alpha$-Al$_2$O$_3$, (1120) $\alpha$-Al$_2$O$_3$, and (001) TiO$_2$ substrates. Insulator-to-metal transitions are observed both in $\rho$–$T$ and $S$–$T$, and the $T_p$ and $T_S$ systematically decrease, along with the broadening of the MI transition temperature width, due to the distortion of the $c$ axis length in the pseudorutile structure for each VO$_2$ film. Moreover, the onset $T_S$, which is where the insulating phase starts to become a metallic one, is much lower than the onset $T_p$. This difference is attributed to the more sensitive detection of the hidden metallic domains that remain in the majority insulating phase via $S$ measurements, which cannot be detected in $\rho$ measurements. These results indicate that the $S$ measurements provide a straightforward and excellent technique to investigate the MI transition process of VO$_2$ in detail.

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