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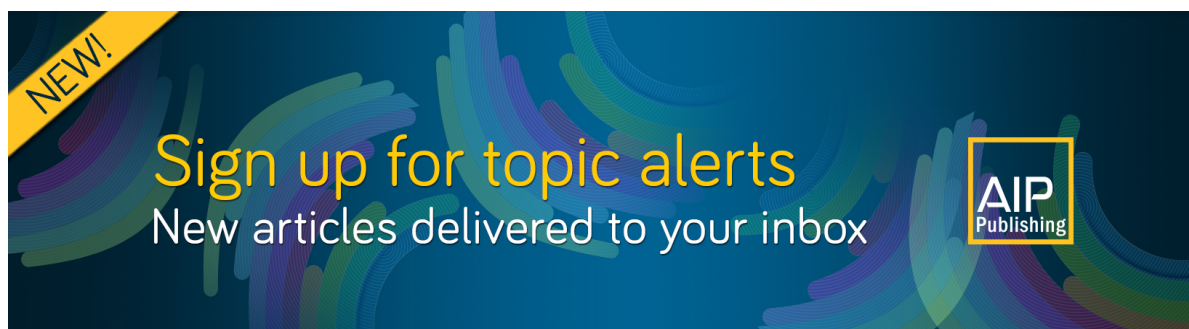
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ABSTRACT

Ruthenium dioxide (RuO₂) is an ideal buffer layer for vanadium dioxide (VO₂) heterostructures due to its high electrical conductivity and matching crystal structure with metallic VO₂. VO₂ thin films were deposited on single crystal TiO₂ (001) substrates with RuO₂ buffer layers via pulsed laser deposition. The metal-insulator transition temperature (T_{MIT}) in VO₂ films can be controlled by the epitaxial strain between the VO₂ film and RuO₂ buffer layer by adjusting the buffer layer thickness (10 – 50 nm). We observed a decrease in the T_{MIT} of VO₂ films from 59 °C to 24 °C as the RuO₂ thickness decreased from 50 nm to 10 nm. Additionally, we show that the RuO₂ buffer layer can sustain an intermediate strain state in VO₂ films up to 100 nm in thickness with a subsequently lower T_{MIT} (30 °C). The 10 nm thick RuO₂ buffer layer can reduce the T_{MIT} in VO₂ films by providing a pathway to relieve the strain through grain boundaries.

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I. INTRODUCTION

Vanadium dioxide (VO₂) undergoes a sharp metal-insulator transition (MIT) above room temperature at ~67 °C, which is associated with a structural phase transformation (SPT) between a low-temperature insulating monoclinic phase and a high-temperature metallic tetragonal phase.¹ The MIT and SPT can be controlled by external parameters such as temperature,² electric field,³ or photo-excitation,⁴ and the switching time of the transition can be on ultrafast timescales (~100 fs) when the transition is induced optically.^{5,6} As the temperature of the VO₂ increases above 67 °C, the electrical resistivity decreases by several orders of magnitude and the infrared transmittance decreases by ~60 %.⁷ These unique properties have made VO₂ an attractive candidate in many promising applications such as ultrafast switches,^{8,9} thermo-optical modulators,¹⁰ field effect transistors,^{11,12} bolometric photodetection,¹³ plasmonic metamaterials,¹⁴ thermal actuators,¹⁵ and smart radiators for spacecraft.¹⁶

The nature of the MIT and SPT in VO₂ has been a long-standing debate. It is generally acknowledged that the mechanism of the MIT in bulk VO₂ is considered to be a collaborative Mott-Peierls transition. The SPT from monoclinic (M1) phase to tetragonal rutile (R) phase is most commonly reported; however, some intermediate phases, such as M2-phase,^{17,18} B-phase,¹⁹ A-phase,¹⁹ have also been recognized during the phase transition. On the other hand, recent reports have revealed that ultrathin VO₂ films deposited on lattice matched TiO₂ substrates show no monoclinic phase at room temperature, suggesting that the VO₂ films are tetragonal rutile in both the insulating and metallic states, i.e., the films undergo an electronic phase transition without the structural phase transition.^{20–22}

The ability to tune T_{MIT} is important in many device applications.^{23,24} Doping with high-valence metal ions into the VO₂ lattice is a commonly used method to achieve tuning of the T_{MIT} in VO₂ films.^{25,26} Introducing epitaxial strain between

VO₂ and the substrate has also been realized as an effective way to control the T_{MIT} in VO₂ films. TiO₂ rutile substrates are most commonly used for the growth of epitaxial VO₂ films due to rutile's matching crystal structures and similar lattice parameters with metallic VO₂ (P42/mnm). Muraoka *et al.* reported that ultrathin VO₂ films show a reduced T_{MIT} (299 K) when grown on TiO₂ (001) substrates, while the T_{MIT} increased up to 369 K for VO₂ films grown on TiO₂ (110) substrates.²⁷ In the former case, the compressive strain in the c-axis of the VO₂ films resulted in the reduced T_{MIT} while in the latter, the tensile strain in the c-axis led to the increased T_{MIT} . Fan *et al.* also reported the thickness-dependent interfacial strain dynamics of epitaxial VO₂ films grown on TiO₂ (001) substrates, demonstrating that a large epitaxial strain occurred in the initial growth stage of the VO₂ films and the epitaxial strain was relaxed as the film thickness increased, leading to an increase in the T_{MIT} .²¹ In addition, various buffer layers have been introduced between VO₂ films and substrates in order to control the strain and MIT properties in VO₂ films.^{28–31} Among them, RuO₂ was proposed as an excellent buffer because it has the same crystal structure and space group as both TiO₂ and metallic VO₂.³¹ In this report,³¹ they showed that the T_{MIT} of thin VO₂ films can be changed continuously by epitaxial strain in a buffer layer of varying thickness. Here we report a systematic study on the epitaxial strain of VO₂ films by varying RuO₂ and VO₂ film thickness using X-ray reciprocal space mapping analysis. Furthermore, the a-axis lattice parameter of VO₂ ($a = 4.54$ Å, $c = 2.88$ Å, JCPDS #71-4821) is smaller than the TiO₂ ($a = 4.59$ Å, $c = 2.96$ Å, JCPDS #21-1276) but bigger than the RuO₂ ($a = 4.49$ Å, $c = 3.11$ Å, JCPDS #40-1290), indicating that the VO₂ film can be compressively strained along the in-plane direction when deposited on bulk RuO₂ whereas it would be tensile-strained on the TiO₂ (001) substrate. Thus, the T_{MIT} in VO₂ films can be tuned by adjusting the epitaxial strain of the RuO₂ buffer layer. Another advantage of using a RuO₂ buffer layer is that it can be used as a bottom oxide electrode for VO₂ based devices with out-of-plane configurations, which would considerably reduce the switching voltage and current (compared to VO₂-based planar type devices).

In this work, we demonstrate control of the MIT temperature in VO₂ thin films by adjusting the epitaxial strain in conducting RuO₂ buffer layers. Using a 10 nm thick RuO₂/TiO₂ template, we were able to sustain the intermediate strain states even with large VO₂ film thicknesses (~100 nm), resulting in relatively low T_{MIT} (~30 °C).

II. EXPERIMENTAL METHODS

VO₂/RuO₂ thin films were epitaxially grown on single crystal TiO₂ (001) substrates by pulsed laser deposition. A pulsed laser beam generated by a KrF excimer laser (LPX300, 248 nm, and pulse duration of 30ns) was introduced into a deposition chamber through a quartz window and focused on the target. The energy density of the laser beam was 2 J/cm² at the target surface. A RuO₂ target (American Elements) was used for the buffer layer growth and the VO₂ films were grown from a V₂O₅ target (Kurt J. Lesker). Before thin film deposition, the chamber was evacuated to a background pressure of 10⁻⁵ Torr. The RuO₂ buffer layers (10 - 50 nm) were deposited at 500 °C and at 10 mTorr of oxygen partial pressure, followed by VO₂ layers (20 - 100 nm) grown at 390 °C and at 10 mTorr of oxygen. The structure of deposited films was characterized by x-ray diffraction (XRD) θ -2 θ scans using a Rigaku x-ray diffractometer with Cu K α radiation. The electrical properties of the VO₂/RuO₂ heterostructures were characterized in a probe station equipped with a heating stage (Linkam) at temperatures between 0 and 100 °C using a Keithley 4200 semiconductor characterization system.

III. RESULTS

In order to investigate the effect of the RuO₂ film thickness on the epitaxial strain of VO₂/RuO₂ heterostructures, RuO₂ buffer layers (10 - 50 nm) were prepared on TiO₂ (001) substrates while holding the VO₂ film thickness constant at 50 nm. Figure 1a shows the θ -2 θ XRD patterns of the VO₂/RuO₂/TiO₂ thin films with various RuO₂ film thicknesses (10, 30 and 50 nm). The strong peak at 62.8° is assigned to the TiO₂ (002) substrate whereas the peak at ~60° is indexed

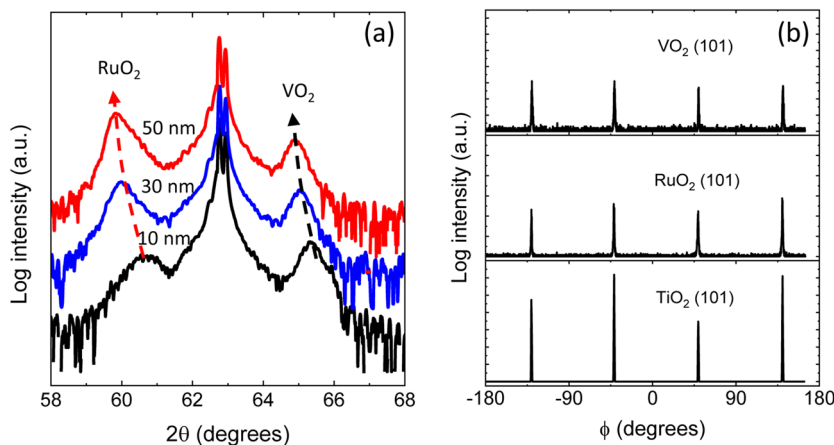


FIG. 1. (a) XRD θ -2 θ patterns of VO₂(50nm)/RuO₂ thin films grown on TiO₂ (001) with three different RuO₂ thicknesses (10, 30, 50 nm). The red and black broken lines represent the trend line of RuO₂ (002) and VO₂ (002) peak positions, respectively. (b) XRD ϕ -scans of VO₂ (101), RuO₂ (101) and TiO₂ (101) peaks for VO₂ (50 nm)/RuO₂ (10 nm)/TiO₂ heterostructure.

to rutile RuO_2 (002), and the peak at $\sim 65^\circ$ is indexed to the tetragonal VO_2 (002) plane. No other peaks were observed, indicating that pure VO_2 was formed during film growth. With increasing RuO_2 buffer layer thickness, the RuO_2 (002) peak moves to lower 2θ angles (approaching the bulk RuO_2 (002) angle) and the VO_2 (002) peak moves to lower 2θ angles (approaching the bulk VO_2 (002) angle). This result suggests that the c -axis lattice parameter of both the RuO_2 buffer layers and the VO_2 thin films increases as the RuO_2 thickness increases, meaning that the epitaxial strain can be adjusted using different thicknesses of the RuO_2 buffer layer.

XRD ϕ -scans were performed to establish the epitaxial relationship in the $\text{VO}_2/\text{RuO}_2/\text{TiO}_2$ layers. Figure 1b shows XRD ϕ -scans on VO_2 (101), RuO_2 (101) and TiO_2 (101) for the 50 nm thick VO_2 film on RuO_2 (10 nm) buffered TiO_2 substrate. The ϕ -scan of the TiO_2 exhibits four peaks separated by a 90° , suggesting fourfold symmetry about the out-of-plane axis. The ϕ -scans of the RuO_2 buffer layer and VO_2 film also show fourfold symmetry with the same azimuth angles of the TiO_2 substrate, thus showing the epitaxy of the VO_2 film on the $\text{RuO}_2/\text{TiO}_2$ template with a relationship of $[100]\text{VO}_2||[100]\text{RuO}_2||[100]\text{TiO}_2$ along the in-plane direction and $[001]\text{VO}_2||[001]\text{RuO}_2||[001]\text{TiO}_2$ along the out of plane direction. Four-fold symmetry of all peaks suggests that all of these materials show their characteristic tetragonal symmetry at room temperature.

In order to understand the epitaxial strain of $\text{VO}_2/\text{RuO}_2/\text{TiO}_2$ heterostructures we performed x-ray reciprocal space mapping (RSM) measurements. Figures 2a–d show the RSMs around the TiO_2 (112) peak for 50 nm thick VO_2 films on RuO_2 buffer layers with various RuO_2 thicknesses (10, 20, 30 and 50 nm). The Q_x and Q_z values of bulk VO_2 are represented with a square symbol and the Q_x and Q_z values of

bulk RuO_2 are marked with a circular symbol. The VO_2 film on a 10 nm RuO_2 buffer layer shows that the Q_z value of the VO_2 peak is slightly larger than the bulk Q_z value of VO_2 , suggesting that the film shows compressive strain in the c -axis. Furthermore, the Q_x value for the VO_2 peak is observed to be slightly smaller than the bulk Q_x value of VO_2 , indicating that the film shows in-plane tensile strain. As the thickness of the RuO_2 buffer layer increases, the Q_x and Q_z values of deposited VO_2 films are approaching the bulk VO_2 . This shift of the diffraction positions suggests that the VO_2 film on thicker RuO_2 is more relieved than the film on thinner RuO_2 . More strain is induced in the VO_2 films grown on thinner RuO_2 buffers (10–20 nm) due to a decrease in the RuO_2 c -axis (compared to 50 nm RuO_2), which leads to a concomitant increase in its a -axis.

In order to investigate thickness effects on the MIT properties of VO_2 , we have prepared VO_2 films with various thicknesses (20 – 100 nm) on 10 nm RuO_2 buffered TiO_2 substrates. Figures 2e–h show the RSMs data for various thicknesses of VO_2 films grown on 10 nm thick RuO_2 buffer layers. For the 20 nm thick VO_2 film, the Q_x values of the VO_2 film, RuO_2 buffer and TiO_2 substrate are almost the same, suggesting that large parts of the VO_2 film and RuO_2 buffer layer are coherently strained to the TiO_2 substrate. As the VO_2 film thickness increases, the Q_x and Q_z values of VO_2 peaks approach those of bulk VO_2 (square symbol), indicating that the thicker VO_2 film (100 nm) is more relaxed than its thinner (20 nm) counterpart but still exhibiting a partially strained state. The streaks present in all maps are due to the saturation of the position sensitive detector. Furthermore, the RSM data for 50 nm VO_2 films grown on TiO_2 substrates with and without a RuO_2 buffer layer is also shown in Fig. S1 in the [supplementary material](#). Notably, the $\text{VO}_2/\text{RuO}_2/\text{TiO}_2$ sample is more strained than the VO_2/TiO_2 sample.

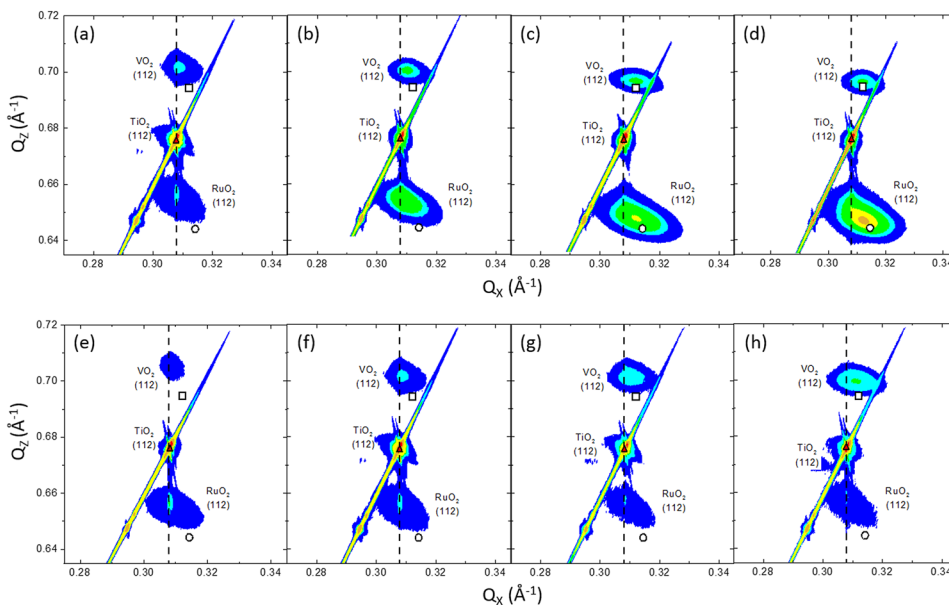


FIG. 2. (a-d) XRD RSMs of VO_2 films (50 nm) deposited on various thicknesses of RuO_2 buffer layers; (a) 10 nm, (b) 20 nm, (c) 30 nm and (d) 50 nm. (e-f) RSMs of various thicknesses of VO_2 films; (e) 20 nm, (f) 50 nm, (g) 75 nm and (h) 100 nm, deposited on 10 nm- RuO_2 buffers. All RSMs are collected around the (112) diffraction spot of TiO_2 at room temperature. The square symbols mark the Q_x and Q_z values for bulk VO_2 (112), the triangle symbols mark the bulk TiO_2 (112), and the circular symbols mark the bulk RuO_2 (112) spot.

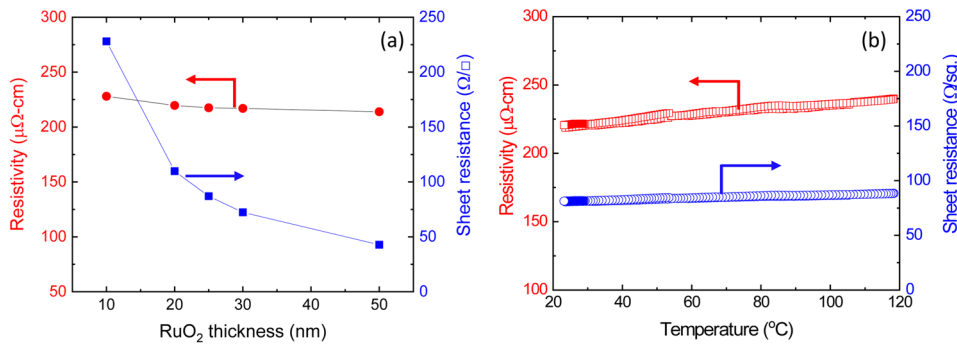


FIG. 3. (a) Room temperature resistivity and sheet resistance of RuO₂ thin films grown on TiO₂ (001) substrates as a function of the film thickness (10 - 50 nm). (b) Resistivity and sheet resistance of the RuO₂ thin film (25 nm) grown on TiO₂ (001) substrate as a function of temperature (heating and cooling).

Thickness-dependent electrical properties of the RuO₂ buffer layers were measured in order to examine the feasibility of the RuO₂ thin films as a bottom electrode. Figure 3a shows the room temperature resistivity and sheet resistance data of the RuO₂ thin films grown on TiO₂ (001) substrates as a function of the film thickness (10 - 50 nm) while the film growth temperature and oxygen pressure were fixed at 500 °C and 10 mTorr, respectively. The room temperature sheet resistance decreases from 225 to 43 $\Omega/\square$ with increasing RuO₂ film thickness from 10 to 50 nm while the film resistivity remains almost constant (~220 $\mu\Omega\text{-cm}$), which is similar to that of commercial indium tin oxide (ITO) electrodes [50 nm-thick ITO films with ~50 $\Omega/\square$; ~250 $\mu\Omega\text{-cm}$]. Thus, the RuO₂ buffer layers with a thickness range from 10 to 50 nm can be used as a bottom electrode for VO₂-based devices with out-of-plane configurations. Figure 3b shows the resistivity and sheet resistance of a typical RuO₂ film (25 nm) as a function of temperature (20 - 120 °C). The resistivity rises slightly from 220 to 240 $\mu\Omega\text{-cm}$ while increasing temperature from 20 to 120 °C. Upon cooling, the film resistivity follows the same line (during heating) with a positive slope, which is typical behavior of metals. The room temperature resistivity remains the same

(220 $\mu\Omega\text{-cm}$) after the cooling process. The electrical properties as a function of oxygen deposition pressure of the RuO₂ thin films are also shown in Fig. S2 in the [supplementary material](#).

Temperature-dependent electrical transport properties were measured for VO₂ (50 nm)/RuO₂ heterostructures with varying RuO₂ buffer layer thicknesses (10 - 50 nm). The effective sheet resistance (R_{eff}) of the VO₂/RuO₂ heterostructures is shown in Fig. 4a. It shows that the T_{MIT} of all VO₂/RuO₂ films is lower than that of typical bulk VO₂ (~67 °C). The T_{MIT} surely decreases from 59 to 24 °C as the RuO₂ buffer layer thickness decreases from 50 nm to 10 nm. This reduction in T_{MIT} can be explained by a difference in epitaxial strain between the VO₂ and RuO₂ films. In general, when VO₂ films have the highest in-plane tensile epitaxial strain, the lowest T_{MIT} in the VO₂ would be expected.^{21,32} Therefore, the 10 nm RuO₂ seems to be an ideal buffer layer to produce the largest epitaxial strain in VO₂ films, showing the lowest T_{MIT} (24 °C). The VO₂ films grown on thicker RuO₂ buffer layers (>10 nm) all showed higher T_{MIT} , indicating that these films are less strained than the VO₂ film on 10 nm RuO₂ buffer. It is noted that the magnitude of resistance change diminishes with increasing RuO₂ thickness. This

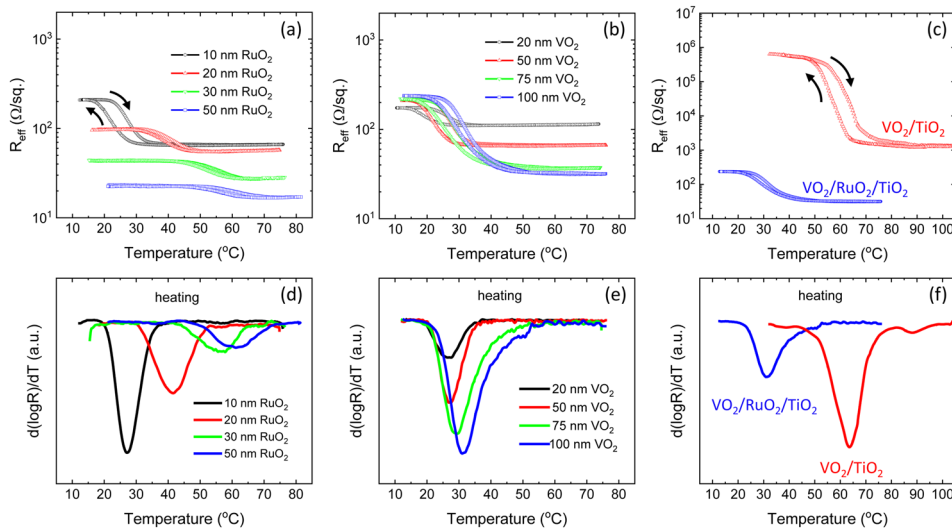


FIG. 4. (a) Effective sheet resistance vs temperature for 50 nm thick VO₂ films grown on RuO₂/TiO₂ as a function of RuO₂ buffer layer thickness (10 - 50 nm). (b) Effective sheet resistance vs temperature for various thicknesses of VO₂ films (20, 50, 75 and 100 nm) grown on RuO₂ (10 nm)/TiO₂ templates. (c) Effective sheet resistance vs temperature for 100 nm VO₂ films grown on TiO₂ substrates with/without RuO₂ buffer layer. Arrows show measurement direction. (d) - (f) Corresponding derivative curves during heating process for plots in (a), (b) and (c), respectively.

TABLE I. Transition temperatures of VO₂/RuO₂/TiO₂ heterostructures during heating and cooling processes. T_{heat} and T_{cool} are the transition temperatures determined from the derivative curves during heating and cooling, respectively. T_{MIT} is determined by the average between T_{heat} and T_{cool} .

Transition temperature	50 nm-VO ₂ grown on TiO ₂ with various thicknesses of RuO ₂ buffer				Various thicknesses of VO ₂ films grown on TiO ₂ with 10 nm RuO ₂ buffer				100 nm-VO ₂ on TiO ₂ with/without 10 nm RuO ₂ buffer	
	10 nm	20 nm	30 nm	50 nm	20 nm	50 nm	75 nm	100 nm	With RuO ₂	Without RuO ₂
T_{heat} (°C)	28.3	41.6	56.2	60.7	26.7	27.3	29.1	31.2	31.2	63.6
T_{cool} (°C)	22.9	38.4	52.4	57.1	19.6	21.3	26.6	28.4	28.4	56.8
T_{MIT} (°C)	25.6	40.0	54.3	58.9	23.2	24.3	27.8	29.8	29.8	60.2

is primarily due to the low resistivity ($\sim 220 \mu\Omega\text{-cm}$) of the RuO₂ layer and the associated current shunting through the RuO₂ layer as the RuO₂ sheet resistance decreases from 225 to 43 $\Omega/\square$ with increasing RuO₂ thickness from 10 to 50 nm (shown in Fig. 3a).

The electrical transport properties were also affected by the VO₂ film thickness. Figure 4b shows temperature-dependent sheet resistance plots of VO₂ films with varying thickness (20, 50, 75 and 100 nm) while the thickness of the RuO₂ buffer layers was fixed at 10 nm. As the film thickness increases from 20 nm to 100 nm, the resistance change amplitude increases threefold, while the T_{MIT} of VO₂ films slightly increases from 23 °C to 30 °C. For comparison, the sheet resistance curves of 100 nm VO₂ films on TiO₂ substrates with and without the RuO₂ buffer layer are plotted in the same figure (Fig. 4c). Without the RuO₂ buffer layer, the T_{MIT} of the VO₂ film increases from 30 °C to 60 °C. It is clear that the 10 nm RuO₂ buffer layer is responsible for lowering the T_{MIT} by preventing relaxation of the strain on the 100 nm thick VO₂ film. Furthermore, this low T_{MIT} (30 °C) for 100 nm thick VO₂ films is ideal for operating switching devices because the transition occurs near room temperature. The T_{MIT} of VO₂ films are deduced from the derivative curves in Fig. 4d-f and are summarized in Table I.

IV. DISCUSSION

The T_{MIT} in VO₂ has been observed to be dependent on epitaxial strain in thin films.^{27,33} In general, compressive strain along the c-axis of VO₂ (i.e., in-plane tensile strain) leads to a reduced T_{MIT} in epitaxial VO₂ films. Furthermore, since the a-axis lattice parameter of RuO₂ ($a = 4.49 \text{ \AA}$) is smaller than TiO₂

($a = 4.59 \text{ \AA}$), the initial epitaxial RuO₂ film layers are subject to tensile-strains at the RuO₂/TiO₂ interface. As the thickness of RuO₂ increases, its lattice parameter monotonically decreases due to relaxation by misfit dislocations. Thus, the T_{MIT} in VO₂ films can be modified efficiently by the epitaxial strain in RuO₂ buffer layers. Figure 5a shows the axial ratio (c/a) for 50 nm-thick VO₂ films with varying RuO₂ thickness. The a- and c-axis lattice parameters of VO₂ films were calculated from RSM data. Clearly, the tendency of the c/a ratio is similar to that of the T_{MIT} in strained VO₂ films. The c/a ratio in VO₂ films decreases as the RuO₂ thickness decreases which, in turn, results in a lower T_{MIT} . The lowest T_{MIT} (24 °C) is observed in the VO₂ film having the smallest c/a ratio (for a 10 nm RuO₂ buffer layer), indicating that the VO₂ film is under tensile strain along the in-plane axis and under compressive strain along the out-of-plane axis. However, as the RuO₂ buffer layer thickness increases, its epitaxial strain becomes more relaxed and the induced strain in the VO₂ film is relieved, thus increasing the VO₂ c/a ratio, leading to an increased T_{MIT} (59 °C), closer to the bulk VO₂ value (67 °C).

Figure 5b shows the axial ratio (c/a) for VO₂ films as a function of film thickness while the RuO₂ thickness is fixed at 10 nm. As the VO₂ thickness increases from 20 to 100 nm, the c/a ratio in the VO₂ films slightly increases from 0.621 to 0.628, which is still much smaller than that of a fully relaxed VO₂ film (0.634). This low c/a ratio results in a reduced T_{MIT} (23 - 30 °C) for 20-100 nm thick VO₂ films, indicating that a significant portion of strain is still present even in 100 nm thick VO₂ films. In general, the VO₂ film is coherently strained below its critical thickness, and above the critical thickness the strained film is relaxed by the formation of misfit dislocations until it becomes fully relaxed.³⁴ VO₂ films grown

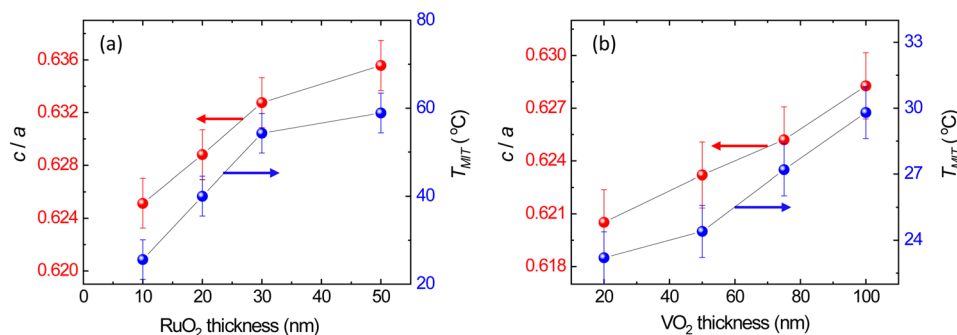


FIG. 5. (a) The axial ratio (c/a) and MIT temperature (T_{MIT}) for 50 nm VO₂ films grown on RuO₂/TiO₂ templates with various thicknesses of RuO₂ buffers (10, 20, 30 and 50 nm). (b) The axial ratio (c/a) and T_{MIT} for various thicknesses of VO₂ films (20, 50, 75 and 100 nm) grown on 10 nm RuO₂/TiO₂ templates.

on single crystal TiO_2 (001) substrates are well explained by this strain relaxation mechanism: where below 15 nm the VO_2 film is fully strained but when its thickness reaches 100 nm it is completely relaxed due to the formation of misfit dislocations.²¹ However, it is difficult to explain our results on VO_2 films grown on $\text{RuO}_2/\text{TiO}_2$ templates using this classical strain mechanism. Instead, the strain relaxation behavior in our films can be explained by grain boundaries. The grain size of the RuO_2 buffer layer was determined by atomic force microscopy to be $\sim 30 - 40$ nm as shown in Fig. S3 in the [supplementary material](#). The boundary between RuO_2 grains is a favorable region for dislocation nucleation. Thus, high density boundaries in RuO_2 buffer layers can provide an alternative pathway to relieve the strain at the VO_2/RuO_2 interface. This explains why our VO_2 films can sustain intermediate strain states for thicker films up to 100 nm. Similar results (strain relaxation through grain boundaries) have been reported for other epitaxial oxide systems.^{29,35–38} It is worth pointing out that 100 nm-thick VO_2 films grown on conducting RuO_2 electrodes can still provide a lower T_{MIT} ($\sim 30^\circ\text{C}$) presumably due to residual strain induced by the RuO_2 buffer layer. Hence, VO_2 based devices with an out-of-plane configuration can be realized at near room temperature using 100 nm thick VO_2 films and 10 nm thick RuO_2 electrodes.

V. CONCLUSIONS

Epitaxial VO_2 thin films were deposited on conductive $\text{RuO}_2/\text{TiO}_2$ templates by pulsed laser deposition. The MIT temperature of $\text{VO}_2/\text{RuO}_2/\text{TiO}_2$ heterostructures can be tuned from 59°C to 24°C by adjusting the strain state of the films by decreasing the RuO_2 thickness from 50 nm to 10 nm. The boundaries between RuO_2 structures are favorable regions responsible for the nucleation of dislocations, which can partially relieve the strain in the film, thereby sustaining intermediate strain states even with large thicknesses (~ 100 nm). This allows lowering of the MIT temperature to near room temperature. These results suggest that the strain generated by the RuO_2 buffer layer can provide an effective way for tuning the T_{MIT} of VO_2 films and provide a route to realizing out-of-plane electrical switching devices.

SUPPLEMENTARY MATERIAL

See [supplementary material](#) for the RSMs data, electrical properties and AFM images.

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