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Growth of *b*-axis oriented VO_2 thin films on glass substrates using ZnO buffer layer

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ABSTRACT

VO_2 thin films are grown on glass substrates by pulsed laser deposition using vanadium metal as a target. In this study, a ZnO thin film was used as a buffer layer for the growth of VO_2 thin films on glass substrates. X-ray diffraction studies showed that the VO_2 thin film had *b*-axis preferential orientation on a *c*-axis oriented ZnO buffer layer. The thickness of the ZnO buffer layer and the oxygen pressure during VO_2 deposition were optimized to grow highly *b*-axis oriented VO_2 thin films. The metal-insulator transition properties of the VO_2 film samples were investigated in terms of infrared reflectance and electrical resistance with varying temperatures.

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1. Introduction

Past studies have reported that vanadium dioxide (VO_2) has a thermally induced semiconductor-to-metal transition at 341 K, accompanied by a structural change [1]. Below this transition temperature, VO_2 is a narrow gap (0.7 eV) semiconductor. Above 341 K, it has metallic properties. VO_2 is highly transparent in the infrared spectral band between 2.5 and 11.5 μm, below its transition temperature. The metallic phase at high temperatures, however, strongly attenuates incident electromagnetic radiation at all frequencies [2]. These dramatic changes in the physical properties of VO_2 allow application in optical switching devices, optical recording devices, infrared sensors, switchable/tunable microwave devices, un-cooled microbolometers, and energy efficient smart windows for buildings [3–5].

The desired properties of VO_2 thin films are high resistance and reflectance change during phase transition and a narrow hysteresis width. It has been proven that using single crystal substrates is effective to obtain VO_2 thin films with a large electrical and optical change during phase transition and a narrow hysteresis width because metal-insulator domain wall propagation of highly oriented VO_2 is faster. Orientation control is an interesting topic in thin film growth; highly oriented VO_2 films have been obtained on sapphire and TiO_2 single crystals [6,7]. However, single crystal substrates are expensive and difficult to prepare on a large scale. These aspects impede application in smart windows. The phase transition properties of VO_2 thin films deposited on various substrates reported in the literature are shown in Table 1. As can be seen, (0 2 0) oriented VO_2 thin film shows better metal-insulator transition properties [6–10]. In order to obtain orientation controlled VO_2 thin films on glass substrate, this study tries to find a suitable buffer for VO_2 growth. It has been show that *c*-axis preferentially orientated ZnO thin films can be prepared on glass substrates [11]. The surface structures of c-plane ZnO are similar to sapphire and have a hexagonal plane. This study used a *c*-axis, which is a highly preferentially oriented ZnO thin film, as a buffer layer to control the growth orientation of VO_2 thin films on glass substrates.

2. Experimental details

VO_2 thin films and ZnO buffer were deposited on glass substrates by focusing a frequency-quadruped Nd-YAG laser ($\lambda = 266$ nm). The laser beam, for ablation, was focused by a quartz lens to a fluence of approximately 1.0 J/cm^2 and directed at an angle of 40° on the targets. The targets used for deposition were pure V metal and ZnO ceramic pellets. These targets were located on multiple target holders, and thus all layers were prepared in a single run process. The glass substrate, placed parallel to the target at a distance of 40 mm, was kept on a substrate holder and rotated during deposition. During deposition, the substrate was heated by an infrared lamp heater.

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Table 1
The phase transition properties of VO_2 thin films deposited on various substrates reported in the literature.

Deposition method	Substrate	Preferential orientation of VO_2	Phase transition temperature (°C)	Hysteresis width (°C)	Thickness (nm)	Ref.
	VO_2 single crystal		68	0.1	–	[8]
PLD (NA)	TiO_2(0 0 1)	(0 0 2)	96	7[a]	10–15	[7]
	TiO_2(1 1 0)	(110)	27	10[a]	10–15	
		NA	~68	NA	100	
PLD (ArF)	Sapphire(0 0 0 1)	(0 2 0)	65(70)[b]	3	NA	[6]
	Sapphire(1 0 −1 0)	(0 2 0)	55(60)[b]	1	NA	
PLD (KrF)	MgO(1 0 0)	NA	70	2[a]	500	[9]
	Fused quartz	(0 1 1)	68	11[a]	500	
Sputtering	Glass	(0 1 1)	60	15	130	
Alkoxide sol–gel		(0 1 1)	60	10	NA	[10]
Aqueous sol–gel		(0 1 1)	56	7	700	
PLD	Glass	(0 1 1)	72	11	58	This study
(Nd-YAG)	ZnO/glass	(0 2 0)	70	7	58	

[a] Estimated from the figures shown in literature.
[b] From the figures shown in the literature, the phase transition temperature, midpoint of hysteresis loop, is about 70 and 60 °C, respectively.

The deposition chamber was initially evacuated by a turbo molecular pump to achieve a base pressure of the order of 5×10^{-6} Pa. It was then backfilled with O_2 to obtain a suitable pressure for the deposition of each film. ZnO layers were deposited at a pressure of 1.33 Pa in O_2. During deposition of both ZnO and VO_2, the substrate temperature was kept at 500 °C. The deposition rates of the ZnO and VO_2 thin films were determined by SEM cross section images. The deposition rates of ZnO and VO_2 under 1.33 Pa in O_2 are 10.12 and 0.96 nm/min, respectively.

The crystalline phases of the films were identified with an X-ray diffractometer (XRD) (X' Pert MPD, PANalytical, Netherlands). The electrical resistance of the films was measured by the four-point probe method. During the measurement of electrical resistance the films were heated from room temperature to 80 °C in air using a hotplate. The infrared reflection spectrum of the sample was measured by an FT-IR spectrometer (BRUKER TENSOR27) with a reflectivity measurement accessory (BRUKER A517/Q, beam incident angle 30°). A gold mirror (BRUKER) was used as the reflectance reference, and the background spectra was measured without a sample on the sample holder. A plate shaped heater was placed over the sample to heat the sample to above its transition temperature.

3. Results and discussion

3.1. Dependence on O_2 pressure during deposition

Because of its half-filled d-shell, vanadium possesses a set of valence states and forms a number of oxides such as VO, V_2O_3, VO_2, and V_2O_5. Thus, the forming condition of VO_2 films prepared from V metal target by reactive laser ablation is very sensitive and has a strong dependence on oxygen pressure. First, 100-nm thick ZnO thin films were prepared on glass substrates. The VO_2 films were then deposited continuously on the ZnO film under various O_2 pressures at 500 °C for 60 min. Fig. 1 shows the XRD patterns of the VO_2 films deposited on glass substrates, using a 100-nm thick ZnO buffer with various O_2 partial pressure. ZnO (0 0 2) peaks were observed in all O_2 pressures. A peak indicating VO_2 (0 2 0) was observed on the film samples deposited under 1.33 Pa and 2.67 Pa, respectively. However, peaks indicating $Zn_2V_2O_7$ (0 0 2) and (0 0 4) were also observed. In XRD patterns of film deposited under 4.00 Pa, there appeared peaks indicating $Zn_2V_2O_7$ (0 0 2), (0 0 4), and VO_2 (0 1 1); under 5.33 Pa and 6.67 Pa, peaks indicating VO_2 (0 1 1) and $Zn_3(VO_4)_2$ (0 0 2) were observed. The results indicate that *b*-axis oriented VO_2 film could be prepared under 1.33 Pa in O_2 partial pressure using a ZnO buffer layer. However, some of the deposited VO_2 reacted with ZnO to form *c*-axis preferentially oriented $Zn_2V_2O_7$.

3.2. Thickness dependence of ZnO buffer layer

To examine the effect of the thickness of a ZnO buffer layer, the thickness of the ZnO film was varied, and VO_2 films were continuously deposited under 1.33 Pa in O_2 partial pressure for 60 min. The XRD patterns of the films deposited, with various thicknesses of ZnO films, are shown in Fig. 2. XRD patterns of the VO_2 film, deposited directly on glass substrate and with 1.6-nm thick ZnO buffer layer, appear to have peaks indicating VO_2 (0 1 1). When the ZnO film was thicker than 10 nm, $Zn_2V_2O_7$ peaks were observed. VO_2 (0 2 0) peaks were observed only in the VO_2 film deposited on a 5-nm thick ZnO film. The stack structure of VO_2 films deposited on glass substrate, using ZnO films with various thicknesses, is shown in Fig. 3. With direct deposition of VO_2 films on the glass substrate, polycrystalline VO_2 films were formed on the amorphous substrate. When the ZnO film was only 1.6 nm, the glass surface was not wholly covered; the deposited vanadium reacted with ZnO to form another compound, possibly $Zn_2V_2O_7$, and some polycrystalline VO_2. In this case, only small VO_2 (0 1 1) peaks were observed.

When the ZnO buffer layer was as thick as 5 nm, only peaks indicating *b*-axis oriented VO_2 were observed in the XRD pattern. The broad peak of the glass substrate may have hidden the small fraction of $Zn_2V_2O_7$ peak. Peaks indicating VO_2 (0 2 0) and $Zn_2V_2O_7$

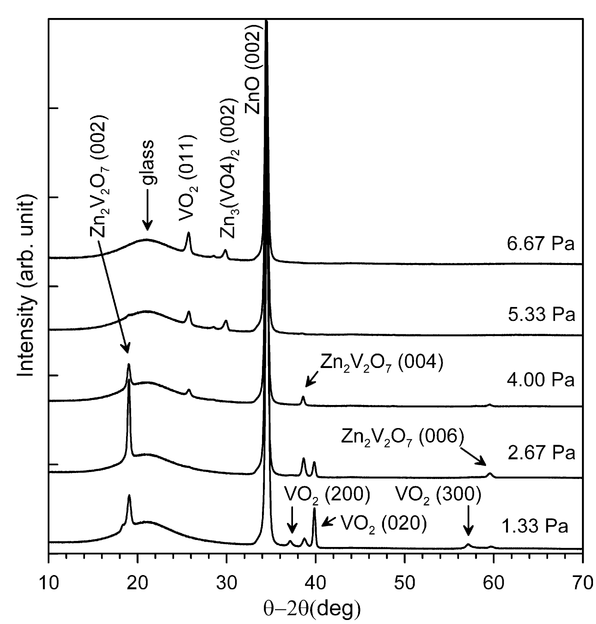

Fig. 1. XRD patterns of the films deposited with 100-nm thick ZnO layer under various O_2 pressures.

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