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Full Length Article

Analyzing optical properties of thin vanadium oxide films through semiconductor-to-metal phase transition using spectroscopic ellipsometry

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ABSTRACT

We investigated the optical behaviors of vanadium dioxide (VO_2) films through the semiconductor-to-metal (STM) phase transition using spectroscopic ellipsometry. Correlations between film thickness and refractive index were observed resulting from the absorbing nature of these films. Simultaneously analyzing data at multiple temperatures using Kramers-Kronig consistent oscillator models help identify film thickness. Nontrivial variations in resulting optical constants were observed through STM transition. As temperature increases, a clear increase is observed in near infrared absorption due to Drude losses that accompany the transition from semiconducting to metallic phases. Thin films grown on silicon and sapphire substrate present different optical properties and thermal hysteresis due to lattice stress and compositional differences.

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1. Introduction

Vanadium dioxide (VO_2) exhibits a remarkable transition between semiconducting and metallic phases, which is accompanied by drastic changes in optical and electrical properties. Above the critical phase transition temperature *Tc*, the material has tetragonal crystal structure with metallic behaviors and high infrared reflectivity. As the temperature goes below *Tc*, the crystal lattice is distorted into energy favorable monoclinic. The material becomes semiconductor. *Tc* of bulk vanadium dioxide is reported to be 68 °C, which can be further reduced through doping, alloying and adjusting underlayers [1,2]. The abrupt changes in film properties at close to room temperature attract great efforts in applying this material for thermochromic windows, optoelectronic devices, field-effect switches, and solid state energy devices in thin film form [3,4].

Reflectance and transmittance measurements [5–7] have long been used to understand the phase transition. In recent years, ellipsometry has been involved in more detailed investigation of optical properties of thin vanadium oxide films [8]. In particular, spectroscopic ellipsometry has been applied in investigating the temperature dependence of the dielectric functions over a wide spectrum range [9,10].

Research on the transition dynamics and substrate dependence may help advance the application of this material in thin film form. Our work using spectroscopic ellipsometry will partially filly this gap by revealing nontrivial differences of this transition on films grown on silicon and sapphire substrates. In this article, we will explore the substrate effects on the optical properties of vanadium oxide thin films. Spectroscopic ellipsometry was applied to dynamically monitor the phase transition process of vanadium oxide thin films. We will discuss correlations between film thickness and refractive index and means to break the correlation and their limitations. Error bars introduced to refractive index due to such correlation will be investigated. Further we will present findings from the film hysteresis behaviors on silicon and sapphire substrates. Effects of band structure and lattice stress will be discussed.

2. Experimental

Thin vanadium oxide films were sputter deposited onto silicon wafer and single side polished c-cut sapphire substrate under carefully optimized pressure conditions at 550 °C [11]. Each sample was heated from 25 °C to 95 °C and then cooled down to 25 °C at a 1 °C/min in a nitrogen-purged temperature cell. A rotating

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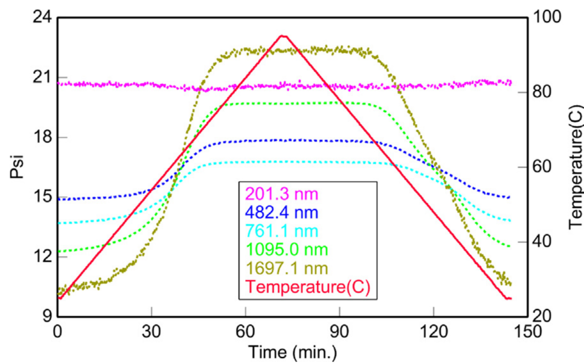

Fig. 1. Spectroscopic ellipsometry data of a thin vanadium oxide film on Si substrate. Psi at 5 wavelengths (out of over 700 wavelengths) is plotted during one temperature cycle from 25 °C to 95 °C. Data was taken every 12 s. Each measurement was set to be 1 s.

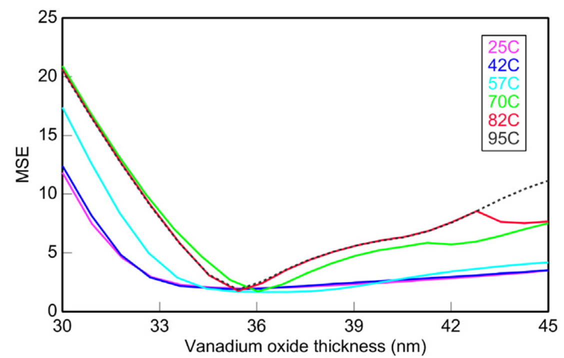

Fig. 3. Thickness sensitivity results on vanadium oxide deposited on silicon substrate. The data above 70 °C shows better sensitivity to film thickness in comparison to the low temperature curves.

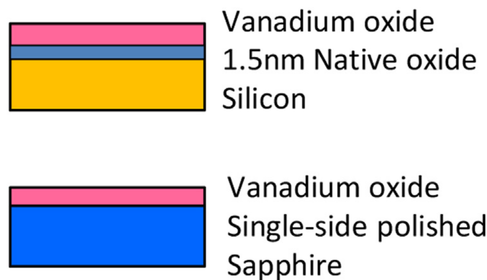

Fig. 2. A three-layer model and a two-layer model were applied to samples on silicon or sapphire substrate, respectively. Optical constants of vanadium oxide and its thickness were defined as variables. Optical properties of the silicon [14], native oxide, and sapphire are fixed to published values. The native oxide thickness was 1.5 nm.

compensator ellipsometer (J. A. Woollam M-2000) was used to dynamically monitor the thermally induced phase transition. Spectroscopic data from 193 nm to 1690 nm were collected at an angle of incidence of 70°. The effects from the temperature cell windows on ellipsometric measurement were pre-calibrated and thickness of native oxide was measured on a similar silicon wafer.

3. Data analysis and results

Fig. 1 shows the raw data of vanadium oxide film on silicon substrate over the entire thermal process. Dynamic Psi at five wavelengths was plotted to illustrate the temperature effect from ultra-violet to near infra-red. Clearly, there are significant changes in four of the Psi curves as the temperature varies. The changes become more pronounced as the probing light shifts from visible further into near Infra-red (NIR) wavelengths. Minimal changes are observed in ultra-violet. Similar trend was observed with the coating on sapphire. The changes are reversible with respect to thermal flux.

The optical models are depicted in Fig. 2. We first analyzed room temperature spectra that were collected outside of the thermal cell at multiple angles of incidence. The results revealed non-zero extinction coefficient, k, at all the investigated wavelengths. Small amount of light absorption was observed even below band gap at the semiconductor state [5,9,10,18]. Correspondingly, we found the vanadium oxide thickness and its dielectric functions were correlated (Fig. 3). Data collected at multiple angles of incidence does not break this correlation. Even though the correlations have been reported as a general challenge in analyzing absorbing films [12], it was hardly fully explored in ellipsometry analysis of vanadium oxide films. Swann and De Smet [8] addressed the correlation through analyzing vanadium oxide films of multiple thicknesses, assuming identical optical constants. We applied the multi-spectra analysis approach by analyzing data at different temperatures using a tunable optical model. Kramers-kronig consistent oscillators [13] were used to describe the dielectric functions of vanadium oxide film on silicon substrate. A Drude and five Gaussian equations were applied such that their resonant energies remain similar during the entire thermal process. Amplitudes and broadenings of individual oscillator were adjusted to match spectroscopic ellipsometry data at different temperatures. Parameters that are not temperature-sensitive are fixed based on the multi-spectra analysis. The goal is to seek the correct thickness by constraining the absorption resonance energies and linking parameters at different temperatures.

Film thickness from the above analysis was found to be in the range of 35.4–36.2 nm. We then verified model sensitivity to this parameter, i.e., thickness uniqueness. We fixed the film thickness to a range of values; for each thickness, the modeling algorithm will vary other variables that describe the film dielectric functions until the best match to the measured data is attained. The differences between the experimental data and model calculated results are quantified through mean square error (MSE) defined as below:

$$MSE = \sqrt{\frac{1}{2N-M}\sum_{i=1}^{N}\left[\left(\frac{\psi_i^{Mod}-\psi_i^{Exp}}{\sigma_{\psi,i}^{Exp}}\right)^2+\left(\frac{\Delta_i^{Mod}-\Delta_i^{Exp}}{\sigma_{\Delta,i}^{Exp}}\right)^2\right]}$$

where N is the number of ψ and Δ pairs, M is the number of variables in the model and σ is the standard deviation for each experimental data point.

The model that produces the minimum MSE corresponds to a best match to experimental data. The lowest MSEs for each thickness at the selected temperatures are plotted in Fig. 3. Similar to what we found from the room temperature data, there are no clearly defined MSE minima at low temperatures. Thickness ranging from 33 nm to 45 nm provides similar MSE up to at least 57 °C, indicating low model sensitivity to the film thickness. However, minima in MSE curves can be much better identified above the empirical phase transition temperature of 68 °C.

Considering the uncertainties in film thickness at lower temperatures, we fix the film thickness to 35.5 nm as determined after the phase transition. Though volume expansion has been reported through the semiconductor to metal transition [8,15], discrepancies exist as the transition involves lattice contraction from monoclinic to tetragonal when transitioning into metals. Regardless, the thermal expansion is negligible in the measured temperature range (less than 0.2%). On the other hand, the crystal structures of the film will theoretically present anisotropy. Different thermal behaviors along a- and c-axis were reported [15,16]. However, the current

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