March, 2010

Strain-Induced Phase Transition in VO_2 Thin Films

（VO_2 薄膜における歪み誘起相転移)

Lippmaa lab. 086015: Tatsuya Kikuzuki

Supervisor: Mikk Lippmaa (Associate Professor)

Keywords: strain, phase transition, VO_2

1. Introduction

Phase transitions in strongly-correlated oxides can be induced by a variety of external excitations, such as light, magnetic field, temperature, electric field, etc. These excitations affect structure and electron density. In this work, our focus is on the possibility of using dynamic strain to drive structural phase transitions in oxide thin films. Most strain effect studies focus either on large static strain (~ 100 GPa) or small strain at very high frequencies, as in piezoelectric bulk materials (~ 10 kHz, ~ 10 kPa). However, a dynamic technique that can cover strains comparable to epitaxial mismatch effects (~ 100 MPa) is needed for studying strain-driven phase transitions and properties of oxide heterostructures. As this excitation affects only the structure, we expect to observe property changes independent of electronic effects. We have attempted to solve this problem by constructing a high-frequency crystal bending stage.

A particularly interesting case is the metal-insulator transition in VO_2, where a Peierls-type electronic phase transition is accompanied by a monoclinic to tetragonal structural transition. This electronic phase transition is particularly interesting for various sensor applications because a resistance change of up to 10^4 occurs close to room temperature. It is known that tensile or compressive *c*-axis epitaxial strain in VO_2 thin films can shift the transition temperature from the bulk value of 340 K by up to 50 K.[1] The purpose of this work is to show that it is possible to drive the phase transition in VO_2 thin films grown on TiO_2 (001) substrates by applying dynamic pressure to a thin film sample, bending the substrate and stretching the film. Substrate bending can result in an in-plane strain of 0.05%, which corresponds for a *c*-axis oriented film to a 0.02% compressive strain and 20 MPa stress in out-of-plane.

2. Experimental

VO_2 thin films were grown on $9\times3\times0.2$ mm^3 TiO_2 (001) substrates by pulsed laser deposition (PLD). For comparison, a film grown by excimer laser assisted metal organic deposition (ELAMOD) was also studied. The PLD experiment was done at an oxygen pressure of 4.4 mTorr and at substrate temperature of 440 to 480°C. The ablation laser (KrF, λ = 248 nm) operated at 1 Hz. The laser fluence on the surface of the V_2O_3 ablation target was ~ 0.56 J/cm^2, resulting in a deposition rate of ~ 0.05 Å/pulse. For studying the effect of grain size on strain response, samples with film thickness gradients from 50 to 150 Å were grown by moving a metal mask between the ablation target and the substrate during deposition.

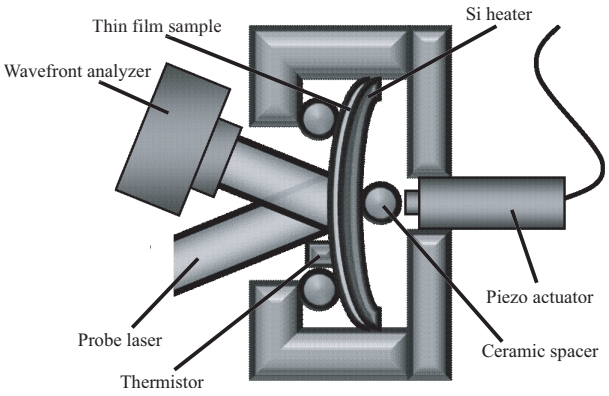

Figure 1: Equipment for measuring dynamic strain effects in thin film samples.

A special high-frequency crystal bending stage was constructed for this work. The dynamic strain effect (frequency response) measurement system is illustrated in Fig. 1. A single-crystal substrate can be bent at frequencies of up to a few kHz with a piezo actuator, while accurately measuring the sample deformation with an optical wavefront analyzer. The change of the *c*-axis length, δ_c, was calculated from Poison ratio estimated from experimental epitaxial strain[2]. The metal-insulator transition in the thin film sample was measured during sample bending by means of electrodes wire-bonded to the sample surface. The piezo voltage and the sample resistance were observed with an oscilloscope (Tek 4032).

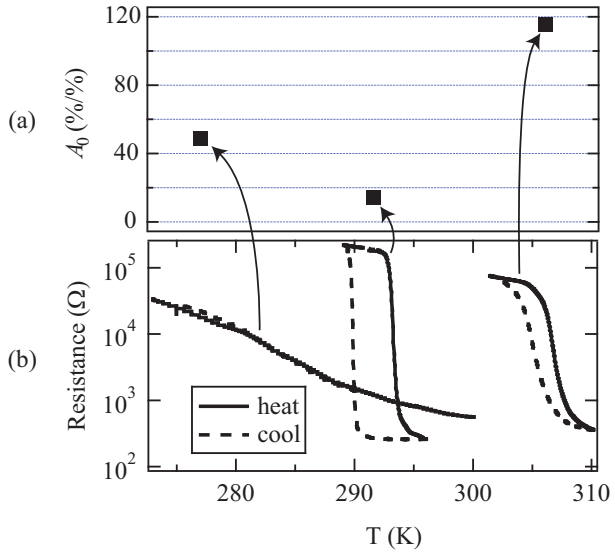

Figure 2: Strain-resistance conversion ratio, A_0 (a) and temperature hysteresis of resistance (b).

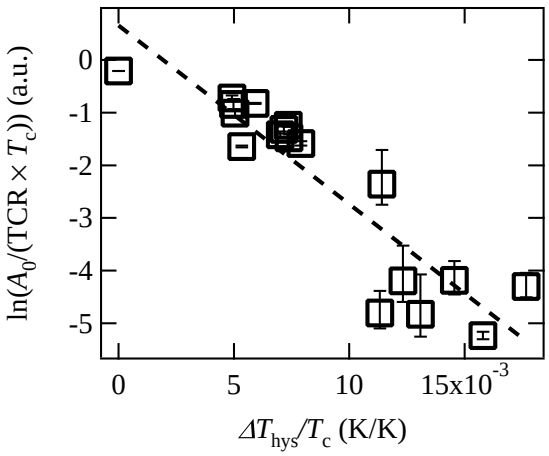

Figure 3: A_0 vs ΔT_{hys} for 18 samples. Liner fit follows Eq. 3.

3. Results and Discussions

The strain-induced resistance change from the maximum resistance, R_{max} was observed in this measurement. For analyzing this effect, two parameters were recorded for each sample as a function of temperature and drive frequency: the phase delay and the amplitude of resistance change. In total, 18 samples have been studied. From the frequency dependence, we can assume the resistance, R to a stepwise strain change would follow a double exponential function as

$$R \propto \alpha \exp\left(-\frac{t}{\tau_1}\right) + (1-\alpha)\exp\left(-\frac{t}{\tau_1}\right), \qquad (1)$$

where t is time, τ_1 and τ_2 are time constants ($\tau_1 < \tau_2$), and α is a scaling factor. Two time constants, τ_1 and τ_2 can be interpreted as domain nucleation and growth, for example. This allows us to calculate a frequency response function f for the peak-to-peak resistance change, ΔR and lattice parameter change, $\Delta\delta_c$ under harmonic excitation as

$$\frac{\Delta R/R_{max}}{\Delta\delta_c} = A_0 f(\omega, \tau_1, \tau_2, \alpha), \qquad (2)$$

where A_0 (%/%) is a dimensionless strain-resistance conversion ratio and ω is the measurement frequency. The relation between A_0 and the width of the hysteresis (0-4 K) and the resistance slope at the transition point are shown in Fig. 2. A figure of merit for strain sensitivity, A_0 can be expressed as a function of hysteresis width, ΔT_{hys} and sharpness of transition as

$$A_0 = C_1 \exp\left(-\frac{k\Delta T_{hys}}{k_B T_c}\right) T_c \left.\frac{\Delta R_{tem}}{\Delta T}\frac{1}{R}\right|_{T=T_c}, \qquad (3)$$

where $\left.\frac{\Delta R_{tem}}{\Delta T}\frac{1}{R}\right|_{T=T_c}$ is the Temperature Coefficient of Resistance (TCR), k is a parameter that relates ΔT_{hys} to the height of the energy barrier separating the metallic and insulating crystallographic phases, and C_1 is an amplification factor that scales the effect of strain relative to temperature on the resistance of the film. Based on Eq. 3, it is possible to estimate k and C_1 values for a VO_2 thin film, as shown in Fig. 3. A linear fit resulted in k = 0.030 eV/K, and C_1=1.9.

Time constants variation between different measurement resistance (temperature) can be seen in Fig. 4 based on Eq. 1. The slower time constants (domain growth), τ_2 is little affected by temperature. This is because domain growth is mainly affected by thermal energy, but the temperature variation in this data set is only a few degrees. Different from τ_2, domain nucleation time constant, τ_1 showed a large change with temperature. This indicates that domain nucleation is visible only in the insulating phase. This data is supported by a previous study[3] where the free energy of the metallic and insulating phases of VO_2 at different temperatures were calculated. Moreover, conducting-tip AFM (Fig. 5) also sup-

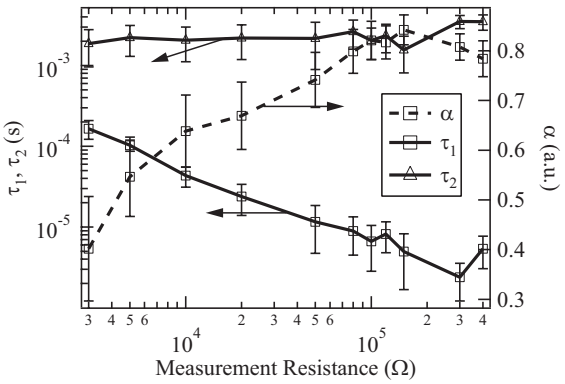

Figure 4: Time constant and scaling factor dependence on measurement resistance besed on Eq. 1.

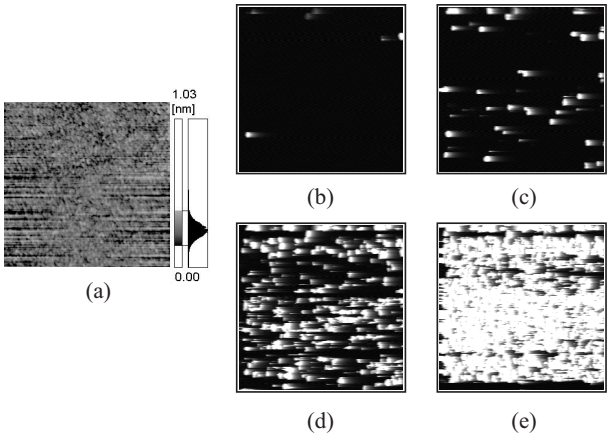

Figure 5: VO_2 sample surface image (1×1 μm^2) measured by conductive AFM. Topography (a) and current map at 304.8 K (b), 306.2 K (c), 306.6 K (d), and 307.0 K (e). White color indicates the metallic phase.

ports this data. By analyzing domain size in Fig. 5, we can estimate the scaling factor, α, giving $\alpha \sim 0.7 - 0.8$ at low temperature and $\alpha \sim 0.5$ at high temperature. These values match well with α value measured from strain-induced phase transition in Fig. 4.

4. Conclusion

A high-frequency crystal bending stage was successfully constructed and used to measure the strain response in VO_2 thin films. Dynamic-strain-induced phase transition was observed. The maximum value of amplification from strain to resistance, A_0 was 157 in VO_2 film with thickness of 125 Å, while it is 4 in usual metal strain gauges. The activation energy $k\Delta T_{\mathrm{hys}}$ for the dynamic strain-induced metal-to-insulator transition was measured, giving $k = 0.030$ eV/K. Close to T_c, the effect of strain on resistance is larger than a pure thermal change by a factor of nearly 2 ($C_1 = 1.9$).

The data includes two time constants, corresponding to domain nucleation process ($\tau_1 \sim 10^{-5}$ s) and domain growth process ($\tau_1 \sim 10^{-3}$ s). τ_1 is increasing with temperature because the energy barrier height separating the two phases becomes larger at higher temperature, while τ_2 is constant because it is mainly affected by thermal energy.

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Papers and conferences

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