Phase Transition in Vanadium Dioxide Nanostructures

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Abstract

Strongly correlated materials exhibit a wide variety of extraordinary optical and electronic properties that can serve as a panacea for industrial applications. We present electronic transport measurements of vanadium dioxide nanostructures, a strongly correlated transition metal oxide system, with a unique thermally driven insulator-metal phase transition ~ 340 K. This transition can also be driven by electrical and optical means thereby making this a very useful material for switching applications. The research aims to address issues such as percolation of nanoscale metallic domains at temperatures near the phase transition. The nucleation and propagation of domains across the transition are studied through a high resolution optical microscope for both electrical and thermal stimulations.

Background

- Vanadium Dioxide (VO_2) along with other transition metal oxides represents a class of strongly correlated materials whose applications can be of enormous benefit to society.
- Strongly correlated systems, which correspond to strong electron-electron repulsion, can manifest themselves physically as phenomena such as metal-insulator transitions, colossal magneto-resistance, and superconductivity.
- VO_2 possesses a metal-insulator phase transition (MIT) giving it various applications from switching devices to memory storage.
- The Hamiltonian for strongly correlated electron systems is impossible to solve analytically and we currently don't have an accurate enough theoretical model to predict the materials behavior.
- This turns to experimentalists to explore these phenomena until a rigorous model is developed.

Experimental Setup

- We examined transport characteristics, imaged the phase transition, and examined heating effects.
- Two types of transport measurements were performed on a 5 micron polycrystalline VO_2 thin film:
  - Resistance-Temperature (RT): Cooled down with liquid nitrogen and measured the resistance(AC) from 200K to 380K.
  - Current-Voltage (IV): Measured current(DC) as a function of applied voltage from 0-8 Volts.
- Shown in **Figure 1** is the experimental setup for the transport measurements.
- Shown in **Figure 2** is the circuit diagram which was utilized for the transport, imaging, and heating measurements.
- A resistor was placed in series for both measurements:
  - RT: Maintain constant current
  - IV: Ensure the safety of the film

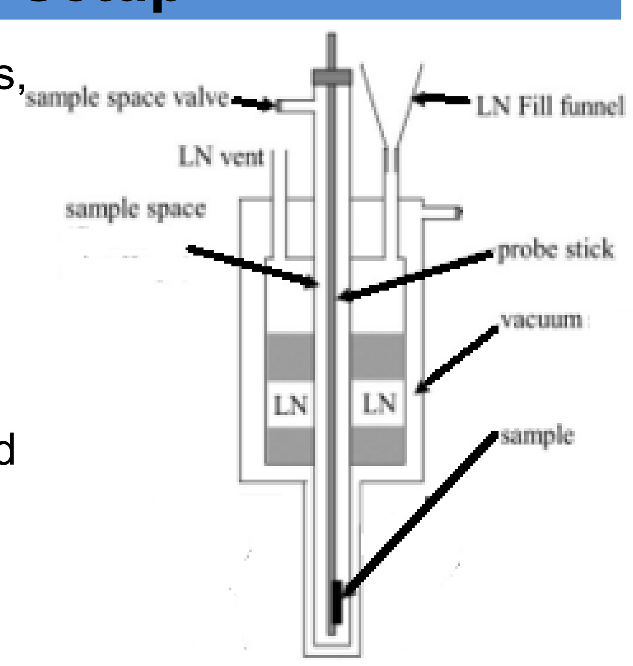

Figure 1: Transport Setup

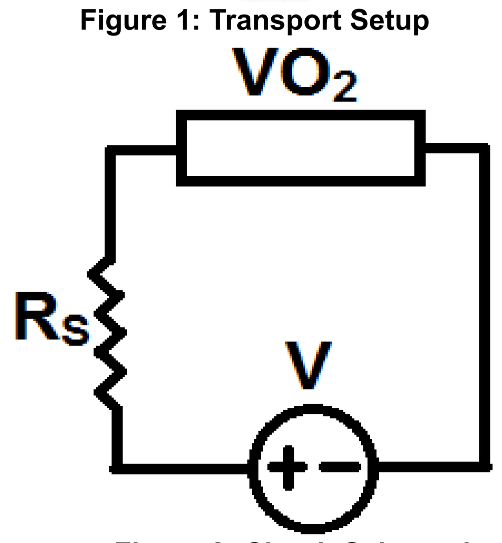

Figure 2: Circuit Schematic

- Following the characterization we imaged the transition under two different mechanisms:
  - Temperature Driven: Excited the MIT using solely thermal stimulation.
  - Voltage Driven: At 330K we applied a voltage to excite the phase transition.
- Following the imaging: Examine heating effects on the film.
- Measuring the current as a function of time near the transition temperature allows us to determine if Joule heating plays a role as a mechanism behind VO_2's MIT.

Theory

- VO_2 is a unique material that has two separate transitions that occur in the same instance:
  - Mott Transition: A metal-nonmetal phase transition. In VO_2 it is characterized by strong electron-electron repulsions.
  - Peierls Transition: A structural phase transition characterized by the distortion of the lattice. In VO_2 this is characterized by a unit cell change of M1 monoclinic to an R rutile phase.
- Both of these transitions are decoupled.
- Under conventional band theory you can't predict the existence of many strongly correlated materials, VO_2 included.
- Depicted in **Figure 3** is the combination of the electronic and structural transition.

Budai, John D. et al. "Metallization of vanadium dioxide driven by large phonon entropy" *Nature* 2014: (5). Online

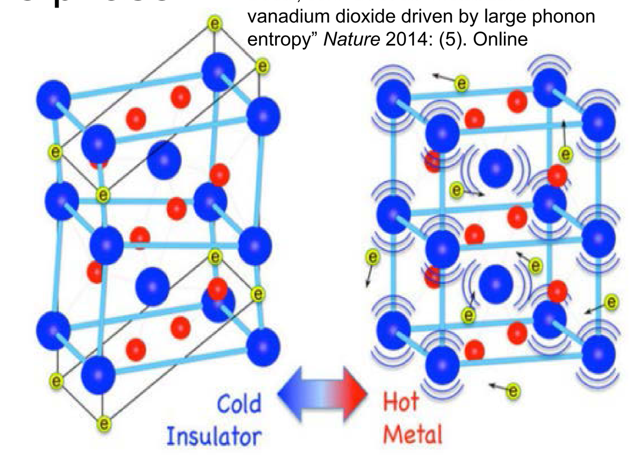

Figure 3: VO_2 Phase Transition

- The structural transition is characterized by the dimerization of the vanadium ions resulting in a lattice distortion.
- The Hamiltonian for any generalized atomic system is given by:

$$\boldsymbol{H} = \sum_i \frac{\boldsymbol{p}_i^2}{2m} + \sum_{i<j} V_{ee}(\boldsymbol{r}_i - \boldsymbol{r}_j) + \sum_I \frac{\boldsymbol{P}_I^2}{2M} + \sum_{I<J} V_{ii}(\boldsymbol{R}_I - \boldsymbol{R}_J) + \sum_{iI} V_{ei}(\boldsymbol{R}_I - \boldsymbol{r}_i)$$

- Given the complexity of the equation very few analytical solutions exist for solid-state systems, the hydrogen atom and an infinite chain of hydrogen atoms are two examples.
- Explicitly the Hubbard model accurately predicts the overlapping of valence and conduction bands during the transition and simplifies the Hamiltonian for Mott insulators like VO_2.
- The Hamiltonian for the Hubbard is given by:

$$\boldsymbol{H} = -t \sum_{\langle i,j\rangle,\sigma} (c_{i,\sigma}^H c_{j,\sigma} + c_{j,\sigma}^H c_{i,\sigma}) + U \sum_{i=1} n_{i+} n_{i-}$$

Results: Transport

- The transport features are displayed in **Figure 4** and **Figure 5**.
- The RT transport shows us the phase transition changes the resistance by many orders of magnitude.
- The hysteresis width of the loop is ~10K.
- This larger hysteresis width is due to the polycrystalline makeup of the film, giving it a smoother transition.
- The IV transport shows the transition appearing at temperatures from 315K to 336K.
- The transition becomes less abrupt at higher temperatures and lower voltages are required to excite the transition.
- **Figure 6** shows a very interesting result. Taking the voltages where the first-order transitions occur and plotting them as a function of temperature we retrieve a linear relation.

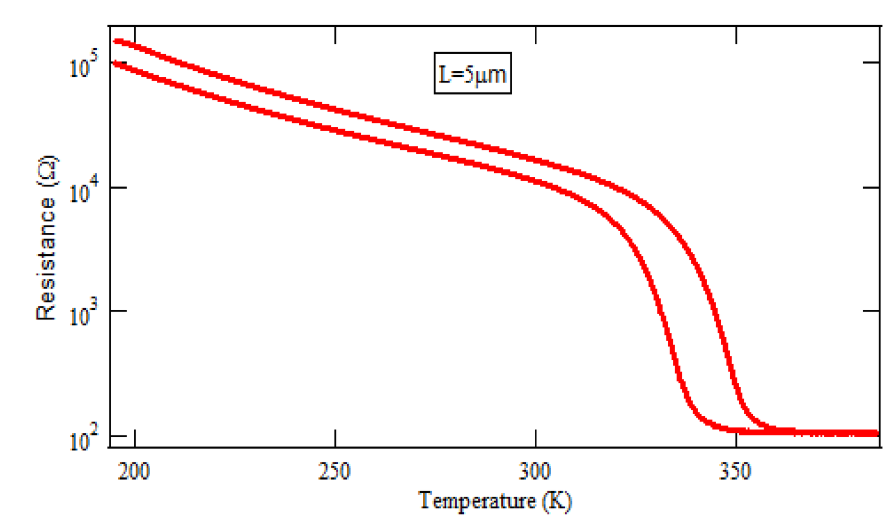

Figure 4: RT Transport

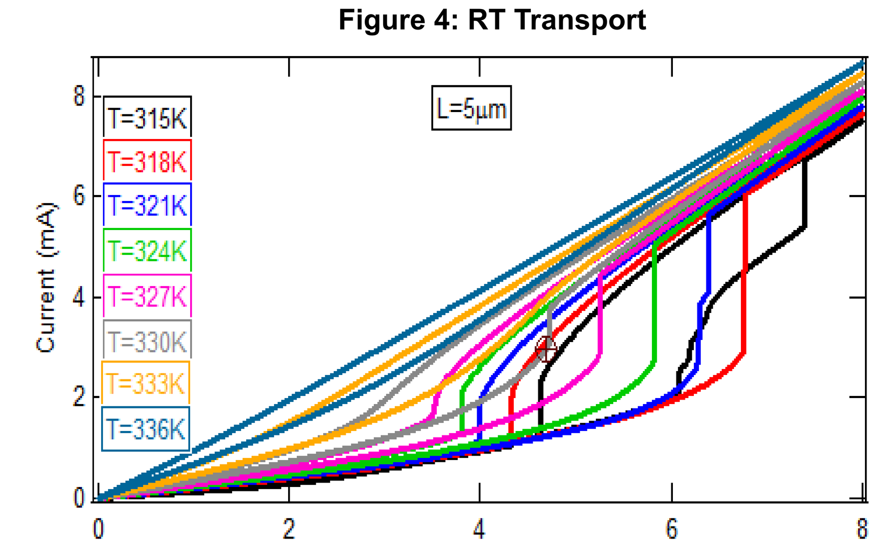

Figure 5: IV Transport

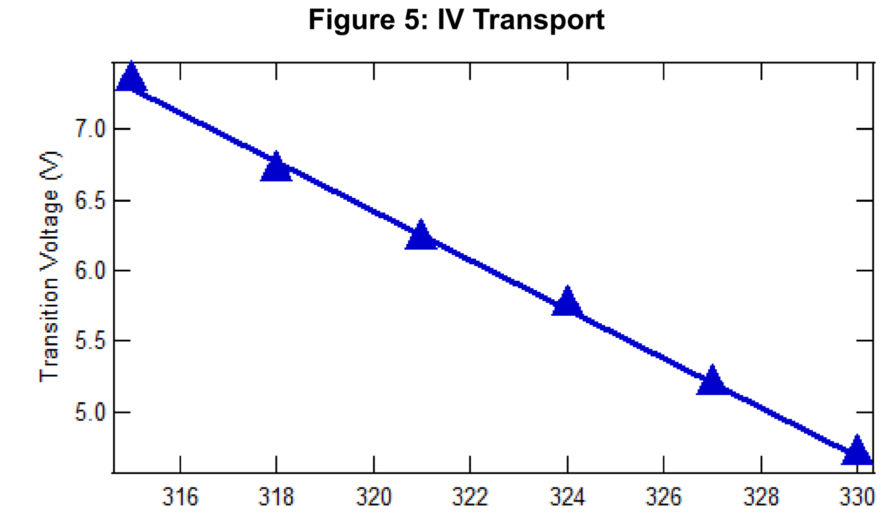

Figure 6: Transition Relation

Results: Imaging

- The temperature-driven image is shown in **Figure 7**. The insulating phase is characterized by the yellow color whereas the green shade displays the domains on opposite sides of the transition. Both images are magnified 25x.
- The gold pieces overlaid on the film are the electrical contacts used for transport measurements.

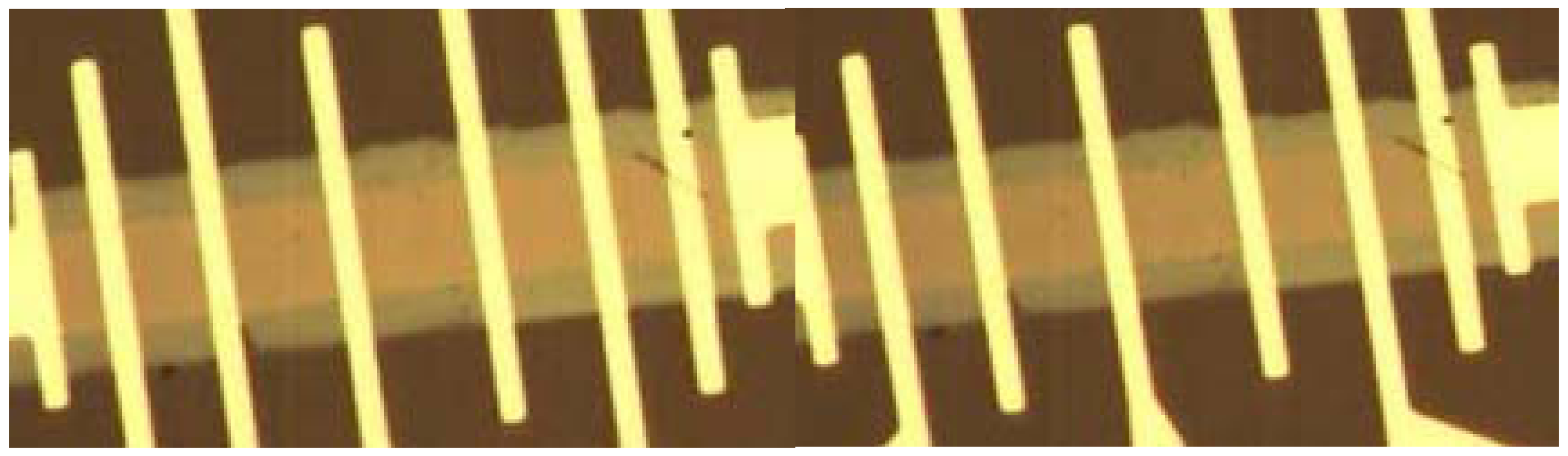

Figure 7: Insulating 25x (left)/Conducting 25x (right)

- The voltage-driven transition is shown in **Figure 8**. The insulating phase of the five micron piece imaged is shown in detail as the same color as the insulating phase for the temperature-driven.
- The left and center images are magnified at 100x whereas the right most image displays the same exact domain formation magnified at 50x.

Figure 8: Insulating 100x (left)/ Conducting domain 100x (center)/ Conducting domain 50x (right)

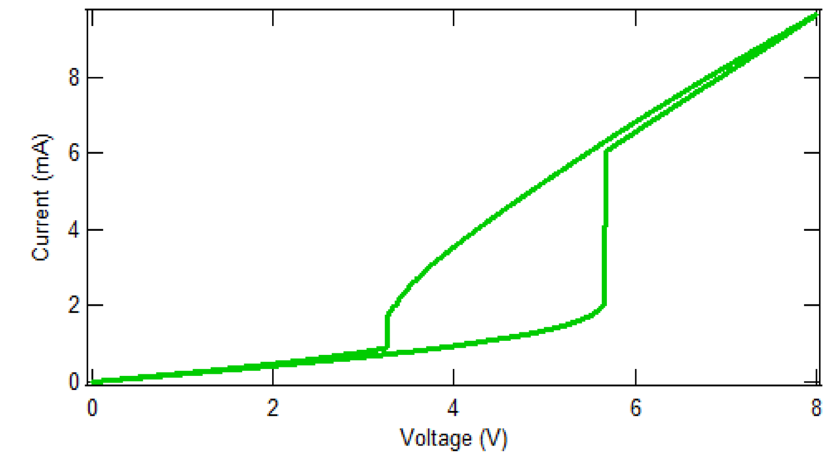

Figure 9: IV Transport for imaging

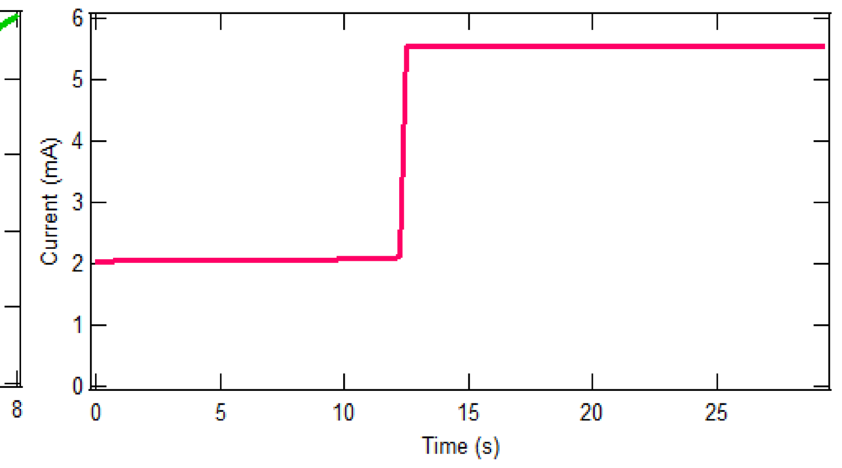

Figure 10: Heating

- **Figure 9** shows the IV transport characteristics for the voltage sweep at 330K.
- Notice that it takes on both the qualitative and quantitative behavior from **Figure 6**.
- Note that this is different from the temperature profile because it the slope is infinite at the transition points.

- **Figure 10** shows the current as a function of time at a constant 5.35 Volts.
- The phase transition occurring at a constant voltage tells us that the transition must occur from the Joule heating of our sample.

Conclusions

- The VO_2 MIT occurs around a temperature of 340K but can also be excited by applying a voltage across.
- When the voltage-driven transition is excited at various temperatures it is found empirically that the transition voltage is a linear function of temperature
- The voltage-driven transition imaged results in a green metallic domain between the electrical contacts
- When the current is monitored at a temperature near the transition it is observed that the transition occurs due to heating effects due to the time-dependence.

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