# Vertical Growth Models Outperform Effective Medium Models in Analyzing $VO_2$ Phase Transition

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**Abstract:** We propose vertical growth models as a numerical method to analyze the phase transition in vanadium dioxide thin films, and show that they can outperform effective medium models in predicting temperature-dependent linear optical properties. © 2021 The Authors

## 1. Introduction

Vanadium dioxide ($VO_2$) possesses a metal-insulator phase transition at ~68 °C that attracts a great amount of research interest. Recently, this volatile, near-room temperature phase transition has been explored for a range of nanophotonic and nanoelectronic applications, including neuromorphic circuits and metasurfaces. A key challenge of fully utilizing $VO_2$ in these applications is to accurately model its linear electromagnetic properties, in particular the permittivity, during the phase transition.

The established approach in the research community is to use effective medium theories. Effective medium theories are widely used to predict and interpret the electromagnetic properties of composite materials that consist of multiple different constituent materials. Treating the intermediate states of $VO_2$ as an effective medium, the constituent materials are the two phases, the metallic phase and the insulating phase, of the material. Looyange and Bruggeman models are the two most widely used effective medium models.

Regardless of the wide adoption of these effective medium models in nanophotonic research, here we question their validity. A fundamental assumption of these models is that the different constituent materials/phases are well mixed at the microscopic scale. However, in nanoparticles and thin films, the interfaces play a critical role in material properties due to the finite surface energy. Thermodynamically, the interfaces can dominate the phase transition as they are the favored locations where the phase nuclearization occurs. This breaks the prior fundamental assumption of the effective medium models, and invokes this question: is there any model that outperforms the effective medium models in analyzing the intermediate states of $VO_2$?

## 2. Vertical growth models and a thin film test sample

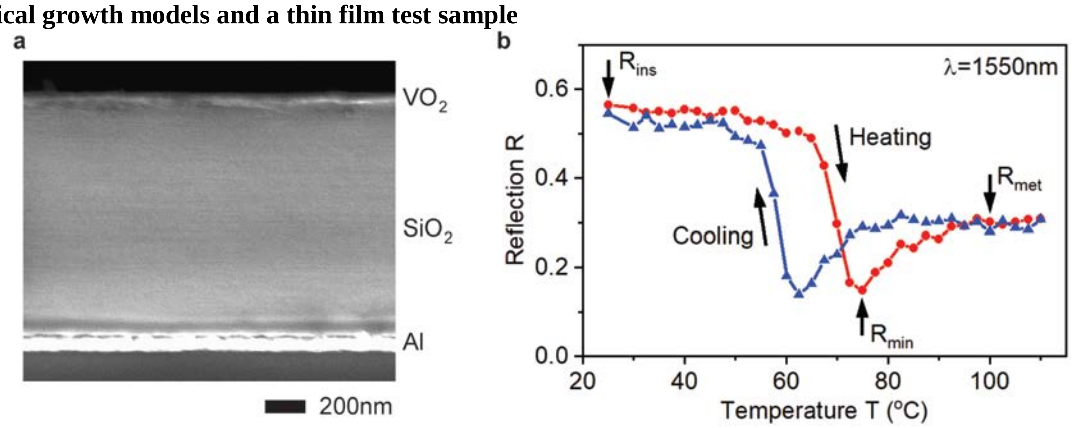


Fig. 1. SEM image and optical reflection of a $VO_2$ thin film. (a) Cross-sectional SEM image of the stack, with the three optically functional layers specified. The scale bar is 200 nm. (b) Light reflection of the film measured in a thermal cycle, with the temperature swept between 25 °C and 110 °C at a step of 2.5 °C in most of the range. The light wavelength is 1550 nm. The three key reflection values ($R_{ins}$, $R_{min}$ and $R_{met}$) required for further analysis are indicated by the three vertical arrows.

In this work, we present microscopic vertical growth models, and demonstrate that they can outperform both Looyange and Bruggeman models in linear optics analysis. The vertical growth models are developed to address the phase transition in $VO_2$ thin films. They are based on the assumption that the interfaces and the interior of a $VO_2$

thin film, due to their differences in thermodynamics, are distinct in phase transition. They consequently approximate the phase transition as a layer-by-layer phase growth process: heating in the phase transition corresponds to the metallic phase growing layer by layer in an insulating thin film, while the cooling corresponds to the layer-by-layer growth of the insulating phase.

As an experimental benchmark, we fabricated a multi-layered thin film and measured its temperature-dependent reflection (Figure 1). The films were deposited on a Si substrate. Only the top three layers of the stack contributed to light reflection, and they were $VO_2$ (40 nm in thickness), $SiO_2$ (1225 nm) and Al (100 nm). The $VO_2$ layer was formed using atomic layer deposition, which provided a high level of control over film thickness, stoichiometry and uniformity. The film stack was designed to create a large variation in light reflection during the phase transition. Figure 1(b) shows the temperature dependent reflection measured in a full thermal cycle between 25 °C and 110 °C, with the film illuminated by light with a wavelength of 1550 nm at normal incidence. Both the heating and cooling branches show a non-monotonic dependence on temperature, which is utilized as the experimental benchmark for the evaluation of different simulation models.

### 3. Comparison between the vertical growth models and the effective medium models

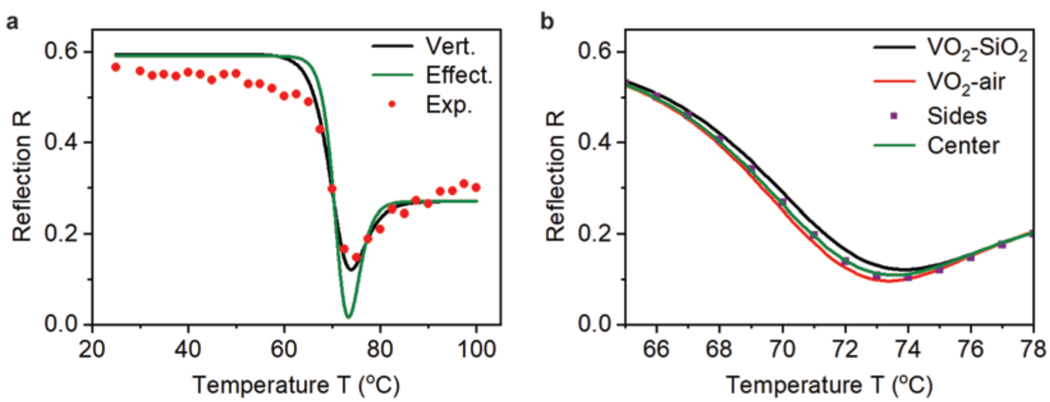


Fig. 2. Comparison between the effective medium models and the vertical growth models, in their reproduction of experimental data. (a) Reflection computed using a representative vertical growth model (black line) and using the effective medium models (green line), overlaid with the experimental reflection. The results of the two effective medium models appear identical at the scale of the figure. (2) Comparison of four possible vertical growth models at temperatures around phase transition. The black lines in the two panels are based on the same data.

Figure 2 compares different theoretical models, by using the reflection observed in the heating branch of the experiment as the benchmark. Here all the models use the same permittivity values for the pure phases, which are obtained from temperature-controlled ellipsometry measurement. They consequently produce the same values $R_{ins}$ and $R_{met}$, the reflection of the pure insulating, low-temperature state and the pure metallic, high-temperature state, respectively. The main contrast among the models is in the reproduction of the minimum reflection value $R_{min.}$

Figure 2(a) shows the reflection calculated using the two effective medium models, which at the scale of the figure give identical results, with a representative vertical growth model. In this growth model, the metallic phase is assumed to grow from the $VO_2$-$SiO_2$ interface. These two types of theoretical models produce reflection curves that are almost identical, expect for a range of ~30 °C around phase transition. Using the experimental value of $R_{min}$ as the benchmark, Figure 2(a) clearly indicates that the vertical growth model outperforms the effective medium models. For completeness of analysis, four different types of vertical growth models are adopted to calculate the reflection and the results are shown in Figure 2(b). They correspond to the growth of metallic phase from (I) the $VO_2$-$SiO_2$ interface, (II) the $VO_2$-air interface, (III) both interfaces at an equal rate to the center, and (IV) from the center to the two interfaces at an equal rate. The four vertical models show small differences in the calculated reflection, and they all outperform the effective medium models using the same benchmark of $R_{min}$.

Similar measurements and analysis are subsequently performed in a broad wavelength range. We observe that for a wavelength range of ~500 nm around the $\lambda = 1550$ nm shown above, the vertical growth models proposed here outperform the widely used effective medium models.