



## Editor's Choice

## Phase-field models for simulating physical vapor deposition and grain evolution of isotropic single-phase polycrystalline thin films

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## ABSTRACT

Two models are presented based on the phase-field methodology to simulate thin film growth during physical vapor deposition (PVD), including subsurface microstructure evolution, for isotropic single-phase polycrystalline materials. The first model couples previous phase-field modeling efforts on ballistic deposition of single-phase materials and grain orientation evolution in polycrystalline materials in a sequential simulation algorithm. The second model incorporates both PVD and grain evolution dynamics into a single free energy functional for use in a phase-field model. To illustrate the capability of the proposed models in capturing combined thin film growth and subsurface grain evolution, PVD simulations of a generic single-phase polycrystalline metal are performed on substrates with different grain sizes. In both models, when the initial substrate grain sizes are smaller than the expected surface features, the thin film grains coarsen via grain boundary (GB) migration until the GBs become aligned with the valleys between the columnar surface features. Thus, each columnar feature is associated with a distinct subsurface grain, in qualitative agreement with experimental observations. Differences between the models arise when initial substrate grain sizes are larger than the surface columnar features. For example, when using the single free energy functional approach, grains contain noticeable internal low-angle variations, which are not captured using the coupled model.

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## 1. Introduction

Thin films are the focus of significant research due to their variety of applications, such as in optoelectronic and microelectronic devices, nanoelectromechanical systems, and optical and protective coatings. Generally, thin films have thicknesses and feature sizes in the nanoscale to microscale range and are usually grown using vapor deposition methods [1]. Here, the focus of this modeling and simulation work is on physical vapor deposition (PVD) in a vacuum environment because (i) the composition of the ejected target material is conserved during transport to the deposition substrate and (ii) the vacuum or low-pressure environment allows physical processes to dominate over gas-phase reactions and collisions during transport [2,3]. Therefore, chemical reactions and composition changes can be neglected in the development of a computational model for PVD. During PVD, incident atoms may not impinge on the substrate surface in a spatially and/or temporally uniform manner, thus creating fluctuations that give rise to surface roughness. These fluctuations compete with surface diffusion, which attempts

to smooth the surface, resulting in nano to microscale surface features [1]. Furthermore, it is known that shadowing and re-emission effects can dominate surface diffusion and growth fluctuations by way of their nonlocal effects, leading to columnar growth. Therefore, to elucidate the fundamental dynamics of thin film growth during PVD, surface tension, diffusion, shadowing and re-emission effects must be taken into consideration [1–3].

The surface features and the details of the underlying microstructure that arise during PVD processing often influence the physical, electrical and chemical properties of the thin film. The deposition conditions and materials primarily determine the resulting surface morphology and microstructure. For example, a material with negligible mobility combined with a low substrate temperature or large incident vapor flux may promote growth of a disordered system [2,3]. On the other hand, many thin films are polycrystalline where the microstructure consists of nanometer to micron-sized domains that differ in crystal structure and/or grain orientation [1–7]. As such, in order to predict thin film properties, it is necessary to consider grains and grain boundaries (GBs) and their evolution during PVD [6,8], including potential connections between surface morphology and subsurface GB evolution [8,9]. Experimentally, structure zone diagrams (SZDs) have

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been developed to help predict the microstructures that may form as a function of deposition conditions, which are useful in determining initial deposition parameters [2]. While SZDs are useful, they are purely phenomenological and predictive only if deposition is performed using the exact same overall process. Furthermore, SZDs cannot account for or describe the underlying nanoscale mechanisms influencing microstructure evolution or the effects of multiple phase evolution and distribution on microstructure dynamics [2]. Therefore, by using modeling and simulation techniques, the aim is to develop a predictive model that contains the underlying physics necessary to describe critical aspects of PVD and microstructure evolution to provide a tool for (i) discovery studies of microstructure evolution during PVD and (ii) guiding experimentalists regarding deposition parameters for the development of advanced materials with designed properties [10].

The purpose of this work is to present two different two-dimensional (2D) modeling approaches based on the phase-field method to simulate PVD growth and subsurface grain evolution within the microstructure of an isotropic single-phase polycrystalline thin film. To the author's knowledge, there has been no attempt in the literature to address concurrent PVD and grain evolution in a comprehensive phase-field model. The phase-field method is a popular simulation technique based on the Ginzburg-Landau theory of phase transitions that has been successfully applied in materials science to study many complex phenomena: dendritic growth in the solidification of an undercooled liquid, solid-solid phase transitions, GB motion, pattern formation in alloys, etc. (cf. [4–6,11–14]). In the phase-field theory, a field variable is assumed to exist, which describes the phase or state of a system at every location within a model. The free energy of the system is assumed to be a functional of the field variable, thermodynamic variables, such as temperature and composition, and their gradients. Evolution of the system is then governed by Cahn-Hilliard or Allen-Cahn dynamics for conserved and non-conserved phenomena, respectively [4,5,11]. In this work, two phase-field modeling approaches are developed for simulating PVD of a single-phase polycrystalline material by leveraging previous modeling efforts on ballistic deposition of single-phase materials [15] and grain evolution in single-phase polycrystalline materials [14]. The first simulation approach couples the equations of motion for PVD and grain evolution models in a sequential simulation algorithm. The second approach incorporates the relevant energetic information for capturing PVD and grain evolution into a single free energy functional. These two models are then applied to simulate PVD of a generic single-phase polycrystalline metal on a substrate with different initial grain sizes to (i) study the influence of initial grain sizes on the evolution of a polycrystalline microstructure and (ii) illustrate and compare the utility and capability of these PVD models in capturing solid film growth and subsurface polycrystalline evolution.

## 2. Previous modeling methodologies

In this section, the requisite components of the equations of motion for modeling PVD and grain evolution in single-phase materials are presented. These equations of motion form the foundation of the phase-field models developed in this work. Full discussions of the motivating literature can be found in [14,15].

### 2.1. Equations of motion for physical vapor deposition

The dynamics and morphology of a growing solid film during PVD is based on the interfacial growth model of Keglinski et al.

[15]. This model was demonstrated to naturally capture prominent physical processes during PVD such as (i) arbitrary surface morphology formation, (ii) surface tension and diffusion, and (iii) non-local shadowing effects. To do this, two field variables were introduced. The first field variable,  $f(\mathbf{r}, t)$ , describes the growing solid where  $f(\mathbf{r}, t) \approx 1$  defines a solid region,  $f(\mathbf{r}, t) \approx -1$  defines a region of vacuum or no solid, and  $f(\mathbf{r}, t) \approx 0$  naturally defines the solid-vapor interface. The second field variable,  $g(\mathbf{r}, t) \geq 0$ , describes the local density of the incident vapor, with  $g(\mathbf{r}, t) = 0$  specifying a region of no incident vapor. The evolution equations of these fields are given by the following non-dimensional expressions,

$$\frac{\partial f}{\partial t} = \nabla^2 \frac{\delta F_{VD}}{\delta f} + B(\nabla f)^2 g + C \sqrt{(\nabla f)^2} g \eta \quad (1)$$

$$\frac{\partial g}{\partial t} = \nabla[D\nabla g - \mathbf{A}g] - B(\nabla f)^2 g \quad (2)$$

In Eq. (1), the first term is the Cahn-Hilliard evolution expression, which provides the model with the ability to form arbitrary surface structures and captures surface diffusion during solid growth. The second term couples the solid evolution to the vapor evolution and serves as a source term that leads to growth at the expense of the incident vapor, where  $B$  is a parameter to modulate the growth rate. The last term provides surface fluctuations through a Gaussian distribution,  $\eta(\mathbf{r}, t)$ , where the amplitude is proportional to the square root of the aggregation rate and  $C$  is a parameter to control the overall noise strength. In Eq. (2), the first term is the diffusion equation modified by the presence of an external force,  $\mathbf{A}$ . In this equation,  $D$  is the diffusion coefficient and  $\mathbf{A}$  provides a flux strength and direction to the incident vapor. The second term, which is the negative of the second term in Eq. (1), is a sink that removes vapor in regions that have been converted to solid. An additional parameter,  $-1 < b < 0$ , is also defined within this model. The purpose of this parameter is to prevent solid growth in regions away from the interface, i.e.,  $f(\mathbf{r}, t) < b$ , which is physically motivated by the absence of gas-phase reactions occurring in PVD as mentioned in Section 1. The free energy functional,  $F_{VD}$ , is a function of  $f(\mathbf{r}, t)$  and its gradient as shown in Eq. (3), providing a double well energy barrier between the equilibrium vacuum and solid phases. The parameter  $a$  is the surface tension coefficient.

$$F_{VD} = \int \left( -\frac{1}{2}f(\mathbf{r}, t)^2 + \frac{1}{4}f(\mathbf{r}, t)^4 + a(\nabla f(\mathbf{r}, t))^2 \right) d\Omega \quad (3)$$

### 2.2. Equations of motion for grain evolution

The evolution of subsurface grains, including GB motion and grain rotation, is based on the phase-field model of single-phase polycrystalline materials developed by Warren et al. [14]. This polycrystalline model was shown to capture grain impingement and coarsening during microstructure evolution through GB migration and grain rotation. To accomplish this, a free energy functional is constructed with two field variables. The first field variable,  $\phi(\mathbf{r}, t)$ , distinguishes between the structurally ordered field within a grain interior ( $\phi(\mathbf{r}, t) \approx 1$ ) and structurally disordered regions characteristic of a liquid or vapor ( $\phi(\mathbf{r}, t) \approx 0$ ) or at a GB ( $0 < \phi(\mathbf{r}, t) < 1$ ). The second field variable,  $\theta(\mathbf{r}, t)$ , measures the local grain orientation from a chosen global axis (the  $x$ -axis in Warren et al. [14]) and varies between 0 and  $2\pi/N$ , where  $N$  is the rotational symmetry of the crystal. With this approach, the isotropic free energy functional is now dependent on grain misorientation and is given as,

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