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Treating grain growth in thin films in three dimensions: A simulation study

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ABSTRACT

Historically, metallography has been the two-dimensional characterization of materials microstructures by optical microscopy. Consequential problems have long been known: A two-dimensional section through a three-dimensional object gives us only a very poor idea about size and form of the object. Therefore, many attempts have been made to gain three-dimensional information experimentally. Nevertheless, in simulations and also analytical theories thin films are commonly still treated as two-dimensional objects making comparisons with 3D experimental data rather hard. In the present paper, grain growth in thin films is investigated for the first time by three-dimensional Potts model simulations focusing particularly on the transition from bulk-like growth to columnar microstructures.

1. Introduction

Grain boundaries are microstructural features of polycrystalline materials that are of special interest, since almost all mechanical properties, such as strength or creep resistance, depend on the boundary distance and therewith grain size. Consequently, understanding grain boundary networks and their temporal evolutions during coarsening processes like recrystallization and grain growth is of great significance. Nevertheless, the phenomenon of grain growth seems relatively simple at first glance: At elevated temperatures grain boundaries start to move, and large grains grow at the expense of their smaller neighbors. As a result, the total number of grains decreases, reducing the grain boundary area and therewith also the total Gibbs free energy of the microstructure. However, on closer inspection the real complexity becomes apparent among others in form of an ongoing competition between the preservation of a local balance in the grain boundary network, e.g. regarding the dihedral angles, and the request for total volume conservation.

In all such studies regarding grain coarsening it is often assumed that we are treating bulk materials. Hence, surfaces and interfacial areas can be neglected. The again, in thin films surface effects—such as grooving or pinning of grain boundaries—become important. Whenever during grain growth in films the average grain size reaches the order of the layer thickness, grain growth slows down and comes to a halt. This phenomenon was observed both in relatively thick [1] as well as very thin [2] metal layers. Mullins [3] predicted that the formation of thermal grooves at those points, where the grain boundaries come up at the surface, is responsible for the stagnation of grain growth. Thompson [4,5] summarized important research results on growth and coarsening of thin films, like factors influencing the structural evolution.

Due to the fact that polycrystalline films have numerous applications as, for example, in magnetic storage media, semiconductor technology, and as coatings, the amount of publications has increased strongly in recent times. Publications on grain growth in thin films have increased in the last 20 years by a factor three and in the last 30 years even by a factor of thirteen. Particularly in recent years, materials science has put quite a focus on this topic:

- Wang and Atkinson [6] investigated the microstructure of yttria-stabilized zirconia electrolyte films using scanning electron microscopy and a sintering model. They concluded that it is of great importance to have a defect size smaller than the critical pore size already at an early stage of processing in order to achieve leak-free electrolyte films.
- Novikov [7] modeled grain growth in thin layers assuming that the only driving force for grain growth is a decrease of the total energy of grain boundaries. He found that the main characteristics of the observed microstructure developments are consistent with experimental observations and concluded that abnormal grain growth in thin films can evolve without taking a decreased energy of the film surface into consideration.

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- Karoutsos et al. [8] have studied the film structure, grain characteristics, and surface roughness of nanocrystalline gold films as a function of film thickness using X-ray diffraction, scanning tunneling microscopy as well as transmission electron microscopy analyzing the average grain size and related size distributions.
- Liu and Shen [9] investigated the effect of an amorphous matrix on microstructural evolution in nanostructured films consisting of two phases finding by use of two-dimensional simulations that the amorphous matrix may considerably impede grain growth and therewith reduce the grain size significantly.
- Also using two-dimensional simulations, Hallberg and Olsson [10] analyzed grain growth in thin Cu films by mesoscale level set and ab initio modeling focusing on the influence of grain boundary energy and mobility. They found among others that abnormal grain growth depends largely on the existence of stored energy variations.
- In two outstanding papers, Barmak et al. [11,12] compared the results of experiments in Al and Cu films with 2D simulations of grain growth. They brought together large data sets of grain size distributions as well as data regarding the topology of the structures. To identify the cause of the differences in observed size distributions between simulations and experiments various factors were examined. However, they found that none of the analyzed factors alone could provide an explanation for the observed experimental behavior.
- In addition, Streitenberger and Zöllner [13] successfully compared those data with analytical results obtained by considerations attributing each structural feature of a polygonal grain network a specific finite mobility.

Nevertheless, from a historical point of view metallography has been the two-dimensional characterization of materials microstructures by optical microscopy and later also by electron beam. Of course, a consequential problem has long been known: A two-dimensional section through a three-dimensional object gives us often only a very poor idea about size and form of the object. Therefore, many attempts have been made to gain three-dimensional information in the form of, e.g., serial sectioning, X-ray tomography, or the use of soap foams as see-through equivalents to polycrystalline grain networks. In recent years, also three-dimensional experiments by X-ray diffraction or transmission electron microscopy have been developed that yield indeed 3D microstructural information.

The problems that arise from analyzing, explaining, and comparing such experimental results with computer simulations and analytical theories lie in the simple fact that particularly thin films are commonly treated as two-dimensional objects still today [9–12]. It is assumed that the films are interfused purely by a columnar grain microstructure as in Fig. 9 (bottom images). However, during the early stage of grain growth the average grain size, e.g., the average grain diameter $\langle D\rangle$ or average grain radius $\langle R\rangle$, can be (distinctly) smaller than the film thickness H (Fig. 9 (top image)). It follows that the growth kinetics for $\langle D\rangle \ll H$ could be similar to the growth kinetics in a bulk material, whereas the microstructure for $\langle D\rangle \gg H$ could be columnar and the associated growth kinetics can be approximated as two-dimensional.

In the present paper grain growth in thin films is investigated for the first time by three-dimensional Potts model simulations focusing particularly on the transition from bulk-like growth to columnar microstructures.

2. Potts model

The Monte Carlo Potts model [14–17] is a lattice based model. The polycrystalline grain structure is mapped onto a lattice, for which generally the number of lattice points per grain should be large enough to enable a morphological characterization of the individual grains. Grain boundaries are defined to be between two neighboring lattice points of unlike orientation. Triple and quadruple junctions are resulting structural features.

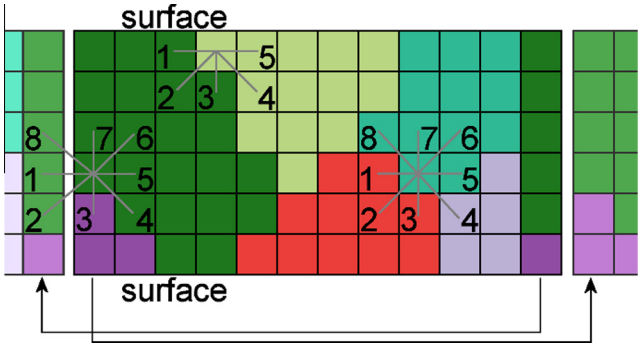

Fig. 1. Two-dimensional scheme of a quadratic simulation lattice indicating the boundary conditions and nearest neighboring lattice points for three examples.

The simulated growth kinetics has to be independent of the underlying lattice type (e.g., quadratic or hexagonal in 2D). For this purpose, the simulation temperature is of great importance, since it influences the grain boundary roughness as well as the triple junction angles therewith ultimately effecting the growth kinetics as shown in [18]. While the simulation temperature is a purely technical parameter, only two real materials parameters are generally used in the simulations, namely the grain boundary mobility m and the grain boundary energy γ. The latter is used to calculate the energy of the microstructure on the lattice by the Hamiltonian and is given as the specific boundary energy per unit length. It is a function of the misorientation angle between the crystallographic orientations of two neighboring grains, which also holds for the grain boundary mobility. The boundary mobility m is a major factor influencing the final selection of a new orientation of a lattice point under consideration. Both, m and γ, are usually considered in relation to their maximum values such that in case all grain boundaries are characterized by the maximum values a simulation of normal grain growth results.

For any such two- or three-dimensional simulation of bulk materials behavior commonly periodic boundary conditions are used assuming an infinite continuation of the structure (see left and right hand side in scheme of Fig. 1). In contrast, in the present work the top and bottom layers are treated as free surfaces. This means that all points on those two layers of a three-dimensional cubic lattice have 17 nearest neighbors (5 first nearest neighbors, 8 s nearest neighbors, and 4 third nearest neighbors), whereas all the other lattice points within the bulk have 26 neighboring lattice points (6 first nearest, 12 s nearest, and 8 third nearest neighbors). The two-dimensional equivalent is shown in the scheme in Fig. 1, where the nearest neighboring lattice points are indicated for three examples.

Apart from that a common implementation is used, of which the details can be found in, e.g. [17,18].

3. Results and discussion

In the following we will analyze at first individual three- and nine-sided grains on two-dimensional lattices with free boundary conditions at the top and bottom lattice layer, then we will extend this simulation to a polycrystalline 2D grain microstructure, and finally investigate the influence of the free surface and layer thickness on the growth kinetics from bulk-like to columnar growth in a corresponding three-dimensional simulation.