

Microstructure Development and Evolution

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Abstract—We employ a levelset-based geometry tracking software using a “grain continuum” representation, together with models for selected IC manufacturing processes and for microstructural evolution to study the development of grain structures. We consider electroless deposition, physical vapor deposition and grain boundary migration during curvature-driven ripening. We use an “encapsulation technique” to convert atomistic data; e.g., from Monte Carlo simulations of nucleation, to continua for input to deposition studies.

Keywords— microstructure; grain continuum; level set method; computer simulation, encapsulation

I. INTRODUCTION

Grain structures are important to the performance and reliability of many structures, e.g., microelectronic interconnects. As critical dimensions for IC interconnects shrink, Cu deposition techniques in which both conformality and grain size are well controlled are becoming increasingly important. Significant effort has been made to simulate the formation and/or development of grain structures in both two and three dimensions. A variety of methods have been used, including Potts models [1,2], phase field models [3], molecular dynamics methods [4], kinetic lattice Monte Carlo (KLMC) techniques [5-7], and explicit front tracking techniques [8-12].

One successful approach to simulating grain structure in polycrystalline metals involves representing and tracking discrete entities, e.g., atoms or aggregates of atoms. Such discrete representations, while natural at atomic scales, can become unwieldy at larger scales, as the number of discrete particles increases beyond what can be reasonably handled by current computers. We address this computational burden by converting results from discrete-particle simulations into a “grain-continuum” type of representation, in which grains are distinct from each other, but represented as shapes in a continuum [13]. After this “encapsulation” procedure is performed, we continue the evolution of the structure in the grain-continuum representation. At this point, the simulation may account for information taken from other simulations on other scales, such as fluxes of material calculated using plasma reactor and sheath models, or surface diffusion constants calculated using a molecular dynamics code.

We discuss multiple-material levelset methods that we use to both encapsulate discrete particle representations and evolve the resulting grain (grain-continuum) structure. We show sample results from applying grain-continuum methods with our parallel levelset environment for nanoscale topography evolution (PLENTE) to several microstructures. We first address the encapsulations of Monte Carlo simulations, and

then their subsequent growth under a set of simple process models. Finally we demonstrate the grain continuum evolution of microstructure in a polycrystalline interconnect during grain boundary migration.

II. METHODOLOGY

This section briefly discusses the numerical methods used to represent and evolve grain structures in PLENTE. See Ref. 14 for details. PLENTE uses a levelset method to represent and evolve complex topographies that is similar to the methods discussed extensively in Refs. 15 and 16. Levelset methods are a broad class of numerical technique in which an N dimensional manifold surface is embedded as a contour or “levelset” in a field in an $N+1$ dimensional space. The explicit equation of motion of the interface is reduced to a partial differential equation by which the levelset field is integrated in time. When the field is signed distance to the front, the front coincides with the zero contour of the field, the transformed equation of motion, or “levelset equation” is given by

$$\phi_t + F|\nabla\phi| = 0, \quad (1)$$

where F is the perpendicular speed of the front, and ϕ is the levelset field. This implicit method automatically accounts for some of the more difficult issues encountered in explicit front tracking, namely changes in topology.

Equation (1) is of the Hamilton-Jacobi type, and we integrate it using the streamline-upwinding Petrov-Galerkin (SUPG) finite element method described by Barth and Sethian [17] extended to tetrahedral unstructured meshes. We have found this method to be quite stable for most applications, and its use of an unstructured mesh allows for local refinement in areas of interest and in regions where it's particularly important to keep error to a minimum.

PLENTE uses an extension to the above levelset method, as described in Ref. 17, to represent and evolve systems of M phases, where $M > 2$. In this method, there is a one-to-one correspondence between distinguishable phases and levelset functions, ϕ_i . In this work, one phase is associated with one grain. The individual ϕ 's are evolved separately and then brought into agreement through “reconciliation” [18].

Reconciliation can cause distortions of the levelset fields away from the zero contours, which over time can cause instability in the evolution of the structure. To avoid this phenomenon, we use a combination of the “closest point transform” [19] and the “fast marching” method [15] to periodically re-initialize the fields to signed distance from the interfaces. During this re-initialization, we extend the speeds from the surface into the field [15,20].

The speeds that are extended from the surfaces are calculated using an appropriate model representing the process-specific phenomena. In this paper, we show structures resulting from electroless deposition (ELD), two models of physical vapor deposition (PVD), and grain boundary migration (GBM).

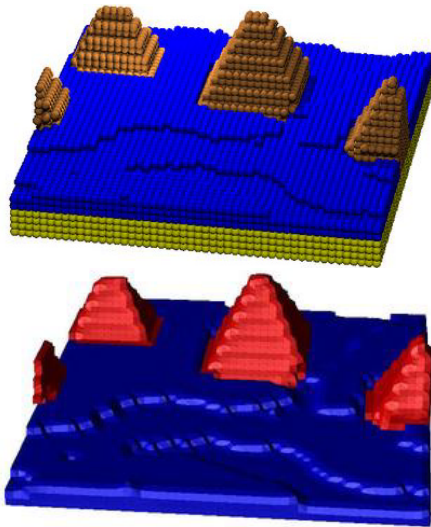


Figure 1. (top) Atomistic structure result from KLMC simulation using ADEPT [6] demonstrating texture competition. (bottom) Encapsulated grain-continuum representation of above result, retaining texture information.

With the exception of GBM, which is calculated from the levelset fields, all the models give velocities based on triangulated surface meshes representing the structures at hand.

Initial structures for several of our simulations are taken from kinetic lattice Monte Carlo (KLMC) simulations, and are represented by discrete particles. We employ a voxel-based encapsulation method and represent both the particles and empty space as a set of classified volume voxels, and determine the minimum signed distances to interfaces between different classes of voxels. For simulations in which a certain minimum smoothness of the encapsulated data is desirable, we apply one of several volume conserving smoothing algorithms [21] to the interface at the same time the signed distance is computed. Fig. 1 shows a two-lattice KLMC simulation performed using ADEPT [6] that exhibits texture competition and the resulting grain continuum representation of the encapsulated structure.

III. RESULTS

A. Electroless Deposition

One simple, but physically realistic process model available is electroless deposition (ELD) of metal in a kinetically limited regime. Tseng et al. [22,23] reported data on a $\text{HNO}_3\text{-CuCl}_2$ bath for Cu ELD, including several SEM micrographs of copper islands before coalescence into a blanket film. Visual inspection indicates that islands are approximately within a

factor of two in radius, indicating an initial period of nucleation followed by nucleation-free growth. We assume an initial set of hemispherical nuclei, approximately normally distributed around an average size, and placed on the substrate randomly, such that no two islands impinge on each other. The kinetic model yields bimodal velocities for each material: the average deposition rate for the free surface, and zero for subsurface material boundaries, such as where a grain meets a substrate, or a grain boundary has formed. The micrographs from Ref. 22 show that many of the resulting grains are close to spherical, and thus we feel that this essentially isotropic model is a reasonable starting point.

We place our simulated islands on a substrate with 5 nm rms roughness, and specify natural boundary conditions at the domain sidewalls. Fig. 2 shows the evolution of this set of islands, using the ELD process model. A small (10%) noise term has been introduced to the surface velocity to mimic local fluctuations in deposition rate. As the islands grow, they begin to impinge on one another and form grain boundaries, until a fully coalesced, blanket film exists. At this point, the resulting grain structure may be the subject of further processing, or used as input to a continuum-based property or performance model.

B. Physical Vapor Deposition

We have also simulated physical vapor deposition (PVD). For deposition systems with high Knudsen numbers, *i.e.*, for structures with length scales of interest that are significantly smaller than the mean free path, transport is by line-of-sight and is dominated by particle-structure collisions (not particle-particle collisions) [24,25]. Thus, flux to the surface can be calculated using distributions from a source volume and a set of “view factors”, or transmission probabilities, calculated solely from the geometry of the structure. In the deterministic low pressure transport and reaction simulator EVOLE [26], the fluxes to each differential surface of the structure are determined by solving integral equations in which fairly arbitrary chemistries can be included.

We use a view factor code that employs a Monte Carlo method to estimate transmission probabilities from a source above the substrate to a mesh representing the surface. We begin our simulations of unity sticking factor deposition by using a KLMC code [27] to form a set of nanoscale islands. These islands are then encapsulated as described in section II and act as the starting structure for our grain continuum simulation. This structure is pictured in Fig. 3a. Using the MC-based view factor code with PLENTE, we apply a Maxwellian flux from a source volume above the substrate, applying

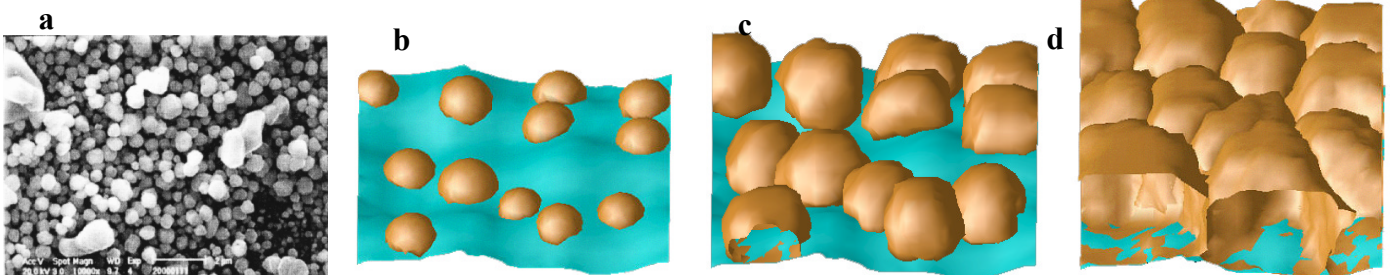


Figure 2. (a) Micrograph [22] showing spherical nuclei during electroless deposition of copper. (b) Initial structure for ELD simulation, showing distribution of hemispherical nuclei on 5 nm RMS rough surface. (c) Onset of coalescence during ELD simulation as described in the text (d) Fully coalesced grain structure.

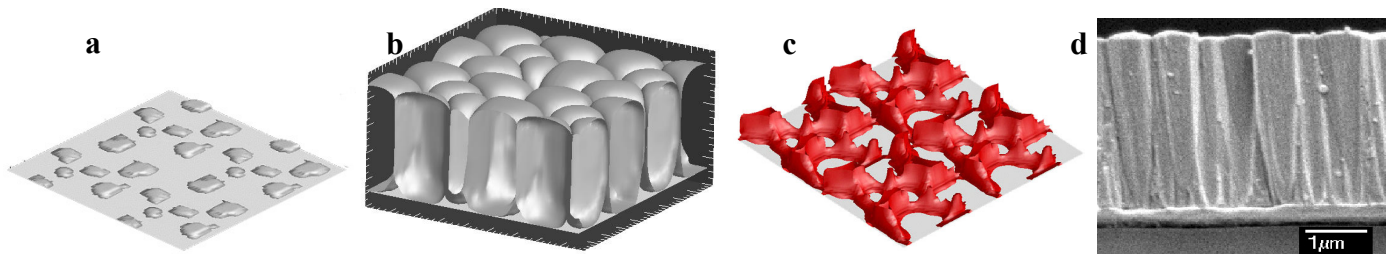


Figure 3. (a) Encapsulated grain-continuum representation of KLMC simulation result. (b) PVD simulation result for unity sticking factor using (a) as starting structure. (c) Voids at base of simulated film, formed around nucleation points due to shadowing. (d) SEM of tantalum oxide film deposited by reactive sputtering showing similar voiding.

periodic boundary conditions at the sides of the computational domain. Fig. 3b shows the resulting, mostly coalesced structure. Free space is left in the coalesced structure, due to shadowing of the sides of grains by other grains. This can be seen clearly in Fig. 3c, which shows the void shapes that develop at the base of the grains due to shadowing.

This simulation compares qualitatively with the SEM in Fig. 3d of a reactive-sputtered tantalum oxide film. Although the simulation was not performed with the TaO_x system in mind, it does share many of the characteristics of the sputtering system, including high sticking factor and negligible surface diffusion. Both films exhibit voids near the base of the grains that are triangular in cross section and grains that become wider until they coalesce, growing in a highly columnar structure. The flux of Ta from the sputtering target is probably not completely Maxwellian, which may account for the difference in shapes of the grains. In particular the tops of the TaO_x grains are less domed than those of the simulation. Additionally, the KLMC simulation was not calibrated to produce a nucleation density for the tantalum oxide system. This directly affects the shape and size of the columnar grains produced.

C. Grain Boundary Migration

One important type of microstructural evolution is grain boundary migration, either due to electromigration during use, or to grain boundary curvature. Such curvature-driven evolution can occur during annealing at high temperatures [2] or, as in the case of electroplated copper, at room temperature [28]. This evolution is a key motivation for

performing microstructural evolution simulations.

We simulated electroless deposition, as above, into an aspect ratio 3 trench with a tapered cross section, onto a set of small deposition nuclei distributed randomly on the trench walls and bottom. We then used the resulting grain structure, pictured in Fig. 4, as a starting structure for a simulation of grain boundary migration under mean curvature.

The adatom concentration along a grain's boundary was assumed to be a linear function of the local mean curvature, with high (concave) curvatures corresponding to high concentrations and low (convex) curvatures corresponding to low concentrations of adatoms. Resistance to mass transfer across grain boundaries was assumed not to depend on grain boundary angle nor on total adatom concentration. The mass flux of material parallel to grain boundaries and along the copper-barrier interface was considered to be small compared to motion transverse to the grain boundaries, due to the large difference in areas available for transport.

Fig. 5 shows side views of the evolving grain structure, in the plane where it meets the barrier layer, at several times. Here, small grains are quickly swallowed by large grains, which in turn grow even larger. The white arrow indicates a grain which is completely swallowed by surrounding grains by the last frame of the series, while the black arrow indicates a grain that has appeared at the copper-barrier interface as other, highly curved grains have pulled away, due to a "thrust fault" configuration. The resulting grain structure is non-equiaxed, as grain are constrained by the width of the trench, but are basically free to grow in directions along the length of the line, and at this grain size, along the depth of the trench.

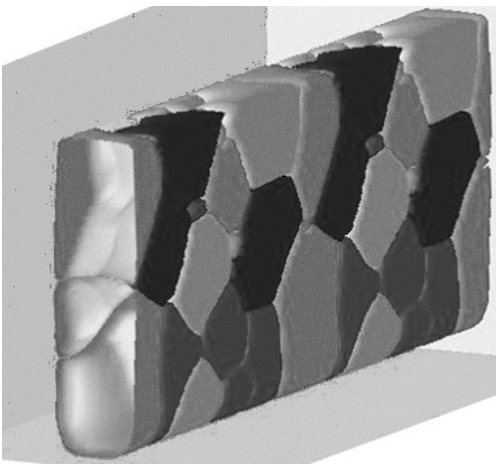


Figure 4. Angle view of starting microstructure deposited into aspect ratio 3 trench for grain boundary migration simulation.

IV. CONCLUSIONS

Grain-continuum representations and levelset methods can be used to track microstructure as it develops during processing and subsequent evolution. Appropriate process and evolution models are still needed to include more detailed physics of the evolving systems, but this class of numerical methods can be applied to several different types of evolving microstructures to track their changing geometry. Qualitative observations of trends in our simulated grain structures can be made even using simple process models. Our simulations of PVD show how interconnected networks of voids can develop when sticking factors are high and surface diffusion is limited. Our grain boundary migration simulations show results of coarsening constrained by a trench structure. Further enhancements to these process models are possible, while using the same overall simulation method for tracking the evolving geometry.

Input structures for these grain scale simulations can be constructed theoretically, or can be “encapsulated” versions of experimental data or discrete particles from other simulations. The details of the starting structure were found to be very important to the resulting shape during PVD simulations. One use for the outputs of these simulations is as inputs to property and performance calculations. The structures are available as triangulated surface meshes with information regarding the interfaces they represent retained on a face-by-face basis.

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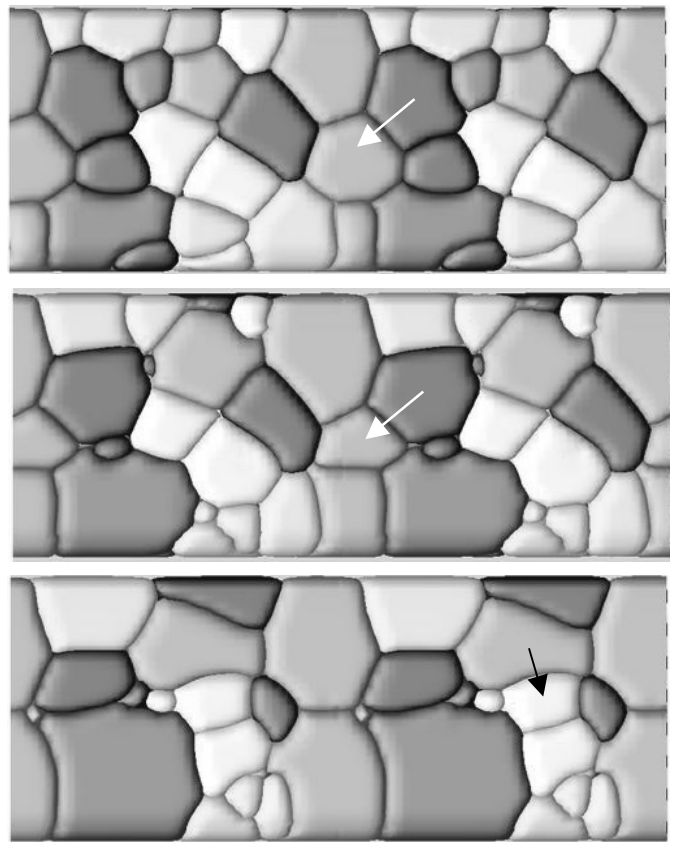


Figure 5. Time series showing side view of microstructure in trench evolving under curvature. Time increases from top to bottom in equal intervals, and different shades of grey indicate distinct orientations. The white arrow indicates a grain which is being “swallowed” by surrounding grains. The black arrow indicates a grain “revealed” as other grains retreat along thrust fault shaped boundaries.