SYMPOSIUM T

Statistical Mechanical Modeling in Materials Research

November 25 – 29, 2001

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* Invited paper

TUTORIAL

**FT T: FUNDAMENTAL METHODS OF
MULTISCALE MODELING**
Sunday, November 25, 2001
9:00 a.m. - 5:00 p.m.
Room 210 (Hynes)

The purpose of this tutorial is to introduce the many facets of multi-length scale modeling. Because of the growing interest in the field of multiple length scale modeling, this tutorial will provide the attendees with a balanced description of the main facets of this field as applied in materials science. The tutorial will consist of four 45-minute lectures that will cover half a day, and the remaining time will be dedicated to a hands-on computer session where the attendees will have access to computers to run actual applications. The areas to be covered are the four fundamental areas that are required for multiple length-scale modeling: electronic structure theory, atomistic modeling (molecular dynamics and Monte Carlo methods), dislocation dynamics, and continuum level modeling. These lectures are meant to provide the attendees with a brief overview of the field and its challenges, and a detailed description of a particular method that will be utilized in the second part of the tutorial. The second half of the tutorial will be a hands-on session where personal computers will be used for the attendees to run illustrative examples in the four topical areas. There will only be a few computers, so attendees are encouraged to bring their laptops for use in the second part of the tutorial.

Instructors:
Ronald Miller, Carleton University
Geroge Gilmer, Lawrence Livermore National Laboratory
Andrew Quong, Lawrence Livermore National Laboratory
Hussein Zbib, Washington State University

SESSION T1: MORPHOLOGICAL EVOLUTION
DURING EPITAXY
Chairs: Vladimir Privman and Da-Jiang Liu
Monday Morning, November 26, 2001
Room 310 (Hynes)

8:30 AM *T1.1
COARSENING OF MOUNDS in EPITAXIAL GROWTH: CRYSTALLINE SYMMETRIES AND CHARACTERISTIC LENGTH SCALES. Martin Siegert, Academic Computing Services, Simon Fraser University, Burnaby, British Columbia, CANADA.

Formation of mounds and the coarsening of such morphologies in molecular beam epitaxy has been found to depend sensitively on the crystalline symmetries of the substrate: the coarsening law differs for deposition on (111) and (001) substrates. Furthermore, the length scale that can be used to characterize the growth law also depends on the substrate symmetry. For triangular symmetries there exists only a single length scale, the pyramid size, and the coarsening law $R(t) = c_0 t^n$ can be found analytically. The solution even provides a very good approximation for the prefactor c_0. For quadratic symmetries there exist two competing length scales related to the densities of pyramid edges and so-called "roof-tops". The ratio of these densities has a crucial effect on the growth law. Finally, in the case of an isotropic substrate there exist several length scales the importance of which is still unclear.

9:00 AM *T1.2
THE LEVEL-SET METHOD FOR MODELING EPITAXIAL GROWTH. C. Ratsch, R.E. Caflisch, M. Petersen, M. Kang, UCLA, Los Angeles, CA; M.F. Gyure, HRL Laboratories, Malibu, CA; D.D. Vvedensky, Imperial College, London, UNITED KINGDOM.

We develop an island dynamics model that employs the level-set technique to describe epitaxial growth. This method is essentially continuous in the plane, yet it retains atomic disrcreteness in the height. In this method, the surface morphology is described by defining the island boundaries as the set $\varphi = 0$ of the so-called level-set function. Islands are nucleated on the surface and their boundaries are moved at rates that are determined by the adatom density, which is obtained from solving the diffusion equation. Reversible aggregation of adatoms can be included without any additional computational cost. Scaled island size distributions in the submonolayer aggregation regime are compared to those obtained from a kinetic Monte Carlo (KMC) simulation for irreversible as well as reversible aggregation. Excellent agreement is obtained. We identify spatial fluctuations in the seeding of islands as the only essential source of noise, while all other stochastic elements can be averaged. We also show that the level-set method can naturally be extended to multilayer growth; here, the set $\varphi = n - 1$ corresponds to the nth

layer. Roughening and coarsening of the surface will be discussed. In particular, we will study the evolution of the step edge density, which is related to the RHEED signal in experiment. A qualtitative and quantitative comparison to KMC simulations will be given.

9:30 AM T1.3
ATOMISTIC MODELING OF MOUND EVOLUTION DURING Ag/Ag(100) HOMOEPITAXY: THREE GROWTH REGIMES. K.J. Caspersen, A.R. Layson, C.R. Stoldt, V. Fournee, P.A. Thiel, J.W. Evans, Iowa State Univ, Ames, IA.

A realistic atomistic model is developed to describe VTSTM observations of mound formation in 25ML Ag/Ag(100) films deposited between 175K and 300K. The model is then used to interpret VTSTM data for kinetic roughening and mound coarsening, and to extend this analysis up to a few 1000ML. Three distinct growth regimes are found: initial transformation from 2D islands to 3D mounds; an extended mound steepening regime (of slow coarsening and rapid roughening); and finally asymptotic slope selection (with complex mound ordering, annihilation, and coalescence dynamics reflecting up-down symmetry breaking). These regimes are compressed at lower temperatures. References:
C.R. Stoldt et al., Phys. Rev. Lett. 85 (2000) 800.
K.J. Caspersen et al., Phys. Rev. B 63 (2001) 085401.

9:45 AM T1.4
LEVEL SETS AND REVERSIBLE ISLAND GROWTH. Max Petersen[1,2], Christian Ratsch[2], Russel Caflisch[2], and Andrew Zangwill[1]. [1]Georgia Tech, School of Physics, GA. [2]UCLA, Dept. of Mathematics, CA.

The level set approach applied to the modeling of epitaxial growth allows a description which is discrete (atomistic) in the height profile and continuous in the lateral dimensions. Therefore, virtues of continuum and discrete models can be combined. The feasibility of this approach in the case of homoepitaxial growth with irreversible aggregation has been successfully shown [1]. In this model, island boundaries evolve with a velocity that is obtained by solving the diffusion equation. Here we present an extension to the island dynamics model to include reversibility, i.e., the possibility of thermal detachment of atoms from island edges. As a consequence, islands do not always grow, and break-up of islands is explicitly allowed in our approach. We make no assumptions about a critical nucleus. The island densities and island size distributions obtained within our model are in very good agreement to those calculated by kinetic Monte Carlo (KMC) simulations. The advantage of our model over the KMC consist in suppressing the simulation of irrelevant attachment-reattachment events by replacing them by their corresponding time average. Thus, an implementation of our model requires essentially no additional computational cost in comparison to the irreversible case. This feature is especially advantageous for the simulation of annealing and ripening. We will discuss the time evolution of the island densities and island size distributions during these processes.
[1] C. Ratsch, S. Chen, M. Kang, M.F. Gyure, and D.D. Vvedensky, Phys. Rev. B **61**, R10598 (2000).

10:30 AM *T1.5
SURFACE MORPHOLOGICAL STABILITY AND PATTERN FORMATION IN MODELS OF EPITAXIAL DEPOSITION. Alberto Pimpinelli, Arnaud Videcoq, LASMEA, Université Clermont-Ferrand 2, FRANCE; Masha Vladimirova, IRRMA-EPFL, Lausanne, SWITZERLAND.

Controlling pattern formation at the surface of epitaxially growing crystals is an important technical and fundamental topic. In particular, morphological instabilities have to be understood to be avoided, or, possibly, exploited. A novel mechanism leading to the onset of surface instabilities during epitaxial growth on vicinal substrates is presented. It is based on the coupling between the surface densities of several diffusing species, in the presence of Ehrlich-Schwoebel barriers at step edges. Such diffusing species may be precursor molecules (e.g. trimethylgallium) and adatoms (e.g. Ga adatoms) in chemical vapour epitaxy, or adatoms (e.g. Ga and As) and molecules (e.g. GaAs) in MBE. We investigate in large details a simple two-particle model accounting for this coupling, and we are able to show that, depending just on growth conditions, step flow growth in the same system may be unstable against either meandering or step bunching, as observed for instance in GaAs deposition on vicinals of GaAs(110). This is contrary to what happens in one-particle models for, say, molecular beam epitaxy, where Ehrlich-Schwoebel barriers always oppose step pairing during step flow growth, and are only able to induce meandering instabilities. This is, to our knowledge, the only model that predicts step bunching during homoepitaxial growth without invoking an unlikely "anti-Schwoebel" effect.

11:00 AM T1.6
COOPERATIVE AND COMPETITIVE MECHANISMS IN ISLAND AND PIT NUCLEATION DURING HETEROEPITAXIAL THIN FILM GROWTH. Mathieu Bouville, Alexandru Riposan, Michael L. Falk, Joanna Mirecki Millunchick, Univ of Michigan, Dept of Materials Science and Engineering, Ann Arbor, MI.

Heteroepitaxially grown semiconductor thin films are known to undergo spontaneous 3D island formation. Recent observations also indicate that pit formation during growth may occur concurrently with the appearance of islands. We have recently observed the nucleation of both islands and pits during the growth of a series of $In_{0.27}Ga_{0.73}As/GaAs$ (001) films using Molecular Beam Epitaxy. While it is generally understood that stress relief in strained films favors both island and pit formation, and that the proximity of islands to pits may be favored by stress-mediated interactions, the kinetics of the island-pit growth process that determine whether pits arise have not been investigated in depth. In order to understand the importance of adatom mediated interactions on pit formation during growth we have devised a theoretical model of island and pit nucleation during the growth of III-V alloy compounds. The model treats the incorporation of adatoms of the group III species as the rate-limiting step for island growth. Using kinetic equations we explicitly model the interactions of adatoms with surface steps and 2D islands in addition to their incorporation into 3D islands. The group V (As) chemical potential determines the relative free energies of islands and pits leading to different rates of nucleation of these features. The predictions of this model for the spontaneous formation of pits will be discussed.

11:15 AM T1.7
CONTINUUM LIMIT OF A STEP FLOW MODEL OF EPITAXIAL GROWTH. R.V. Kohn[1], T.S. Lo[1] and N.K. Yip[2]. [1]Courant Institute of Mathematical Sciences, New York University, New York, NY. [2]Department of Mathematics, Purdue University, West Lafayette, IN.

We examine a class of step flow models of epitaxial growth obtained from a Burton-Cabrera-Frank (BCF) type approach in a (1 1)-dimensional setting. Our goal is to formulate an equivalent continuum model by passing to the continuum limit. Away from peaks and valleys, the continuum law is a *Hamilton-Jacobi equation* (HJE) for the evolution of the surface height. The peaks and valleys are *free boundaries* for this HJE; their evolution must be specified by suitable boundary conditions, reflecting the microscopic physics of the discrete model. At peaks, the governing physics involves nucleation. We study the peak boundary condition by performing numerical simulations of the step flow dynamics, using a simple but reasonable nucleation law. Our simulations reveal the presence of "travelling-wave-like" patterns in the profiles near the peaks. The relationship between these patterns and the continuum equation will be discussed. In particular, we shall address the importance of evaporation for matching the local behavior near the peak to the solution of the continuum equation.

11:30 AM T1.8
QUANTITATIVE INFORMATION ON THE KINETICS OF STEP BUNCHING FROM THE DISTRIBUTION OF TERRACE WIDTHS. Arnaud Videcoq, Alberto Pimpinelli, LASMEA, Univ. Blaise Pascal, Clermont-Ferrand, FRANCE; Masha Vladimirova, IRRMA, EPFL, Lausanne, SWITZERLAND.

Experimentaly, growth on vicinal surfaces is often affected by instabilities such as step meandering and/or step bunching. Beside its interest in the physics of out-of-equilibrium phenomena, being able of predicting the evolution of surface morphology during epitaxial growth is of crucial importance for technological applications. It also means understanding some of the mechanism that rule self-structuring and organisation at surfaces. A simple two-particle (precursors and growth units) model for epitaxial growth has recently been studied both analytically [1] and by Monte Carlo simulations [2]. Assuming only normal Ehrlich-Schwoebel (ES) barriers at step edges, it exhibits all different kinds of surface instabilities in different growth regimes: step meandering, step bunching and mounding. Indeed, other microscopic mechanism accounting for step bunching during growth have been proposed, based on an "inverse" ES effect, or on step-edge diffusion. It is clear that quantitative information are needed to discriminate between the different microscopic mechanisms that lead to step bunching. We study in this work the kinetics of the step bunching by analising the distribution of terrace widths as a function of the deposition time in Monte Carlo simulations. The analysis exploits tools previously developed for equilibrium situations [3], and allows us to obtain a quantitative description of the surface structure and of its morphology.
[1] A. Pimpinelli and A. Videcoq, Surface Sci. Lett.445, L23-28 (2000).
[2] M. Vladimirova, A. Pimpinelli and A. Videcoq, J. Cryst. Growth 220, 631-636 (2000).
[3] T.L. Einstein and O. Pierre-Louis, Surface Sci. Lett. 424, L289-L308 (1999).

11:45 AM T1.9
KINETIC MONTE CARLO SIMULATIONS OF THE GROWTH OF THIN METALLIC FILMS. A.M. Cadilhe and M.D. Ramos, Department of Physics, University of Minho, Braga, PORTUGAL.

We performed kinetic Monte Carlo simulations of a suitable phenomenological lattice-gas model to grow thin metallic films. We are able to extend our simulations to large 1000 X 1000 lattice constants lattices. Specifically, we studied two limiting cases by carefully switching on and off processes involved in surface mass transport. We characterize the simulated films in two different limiting cases, namely, the case of no surface diffusion and in the case with surface diffusion. We observe, in the case of no surface diffusion, a rough surface emerging from our simulations, with many exotic nanostructures present. Physically, this case corresponds to a high activation barrier for bond breaking or to a high incident flux of depositing particles. Allowing surface mass transport to take place, we observe the smoothing out of surface roughness. We characterize the surface and perform a comparative study of both physically interesting situations.

SESSION T2: MORPHOLOGICAL EVOLUTION
DURING SOLIDIFICATION
Chairs: Catherine M. Bishop and Michael L. Falk
Monday Afternoon, November 26, 2001
Room 310 (Hynes)

1:30 PM *T2.1
PHASE-FIELD MODELING OF ELECTROCHEMISTRY. J.E. Guyer, W.J. Boettinger, J.A. Warren, and G.B. McFadden, National Institute of Standards and Technology, Gaithersburg, MD.

We present the first application of phase field modeling to electrochemistry. The phase field technique has previously been applied to the time evolution of complex dendritic, eutectic, and peritectic solidification morphologies. The present work was motivated by the mathematical analogy between the governing equations of solidification dynamics and electroplating dynamics. For example, the solid-liquid interface is analogous to the electrode-electrolyte interface. The various overpotentials of electrochemistry have analogies with the supercoolings of alloy solidification: diffusional (constitutional), curvature, and interface attachment. Dendrites can form during solidification and during electroplating. It is not surprising, however, that we find significant differences between the two systems. The crucial presence of charged species in electrochemistry leads to rich interactions between concentration, electrostatic potential, and phase stability. Further, the inherent non-linear kinetics in electrodeposition is far different from solidification, where problems are generally well-modeled with linear kinetics. The present model properly predicts the charge separation associated with the equilibrium double layer at the electrochemical interface and its extent in the electrolyte as a function of electrolyte concentration. The kinetic behavior of the model will be explored. The goal of the approach is to treat the complex geometry, including void formation, that occurs during plating in vias and trenches for on-chip metallization.

2:00 PM *T2.2
PHASE-FIELD SIMULATIONS OF DENDRITIC GROWTH WITH CONVECTION. X. Tong, C. Beckermann, University of Iowa, Dept of Mechanical Engineering, Iowa City, IA; A. Karma, Northeastern University, Dept of Physics, Boston, MA.

Convective effects on free dendritic crystal growth into a supercooled melt in two dimensions are investigated using the phase-field method. The phase-field model incorporates both melt convection and thermal noise. A multigrid SIMPLE method is used to solve the conservation equations for flow. A multiple time step algorithm is developed that uses a large time step for the flow field calculations while reserving a fine time step for the phase-field evolution. The operating state (velocity and shape) of a dendrite tip in a uniform axial flow is found to be in quantitative agreement with the prediction of the Oseen-Ivantsov transport theory if a tip radius based on a parabolic fit is used. Furthermore, using this parabolic tip radius the ratio of the selection parameters without and with flow is shown to be close to unity, which is in agreement with linearized solvability theory for the ranges of the parameters considered. Dendritic sidebranching in a forced flow is also quantitatively studied. Compared to a dendrite growing at the same supercooling in a diffusive environment, convection is found to increase the amplitude and frequency of the sidebranches. The phase-field results for the scaled sidebranch amplitude and wavelength variations with distance from the tip are compared to linear WKB (Wentzel-Kramers-Brillouin) theory. It is also shown that the asymmetric sidebranch growth on the upstream and downstream sides of a dendrite arm growing at an angle with

respect to the flow can be explained by the differences in the mean shapes of the two sides of the arm.

2:30 PM *T2.3
PHASE-FIELD MODELING OF SOLIDIFICATION AND EPITAXIAL GROWTH. <u>Mathis Plapp</u>, CNRS/Ecole Polytechnique, Laboratoire PMC, Palaiseau, FRANCE; Alain Karma, Northeastern University, Physics Dept, Boston, MA.

The phase-field method, based on continuum equations of Ginzburg-Landau type, has become the method of choice for numerical simulations of solidification patterns. Recent progress, both in mathematical analysis and numerical algorithms, makes it now possible to perform accurate three-dimensional simulations on experimentally relevant length and time scales. This will be illustrated by (i) the dendritic solidification of a pure substance, where the shape and velocity of the dendrite tips can be directly compared to experimental results for both low- and high-speed growth regimes, and (ii) the directional solidification of binary alloys, where the dynamics of cellular and eutectic patterns can be analyzed. Furthermore, in addition to solidification the phase-field method can also be applied to other free-boundary problems. We have developed a phase-field formulation for the classical BCF model of surface growth that is able to simulate the evolution of vicinal surfaces on mesoscopic length and time scales, and that can be used to model epitaxial growth in the step-flow regime. We present results on the growth dynamics of spiral ridges that form around screw dislocations, and discuss the possibility of generalizing the model to include Schwoebel barriers and elastic interactions.

3:30 PM *T2.4
NUMERICAL SIMULATION OF DENDRITIC GROWTH. <u>Jonathan A. Dantzig</u>, Jun-Ho Jeong, University of Illinois, Dept of Mechanical and Industrial Engineering, Urbana, IL; Nigel D. Goldenfeld, University of Illinois, Department of Physics, Urbana, IL.

Understanding pattern selection during dendritic solidification is an important problem for materials scientists and engineers. The microstructure formed during solidification affects the properties of the material in service, and cannot be changed readily by subsequent solid-state processing. In this talk, recent computations using phase-field models to directly simulate dendritic growth are described. In this method, the liquid-solid interface is modeled as a diffuse region whose thickness is characterized by an order parameter, known as the phase field. One of the difficulties encountered when applying the phase field method is the conflicting requirements of high resolution needed to successfully capture the physical phenomena at the interface, and the simultaneous need to fully resolve the diffusion field ahead of the advancing front. We employ an adaptive gridding procedure for solving the phase field equations, where high resolution is available near the interface, and more appropriate grid dimensions are used to resolve the diffusion field. Examples are given for pure, isolated dendrites. Recent work examining the role of fluid flow in the pattern selection process will also be presented. Fully three-dimensional simulations of growth of pure materials are presented, and it is demonstrated that the three dimensional aspects of the flow are essential. Parallel implementation of the code is also described. Direct comparisons of computational and experimental results are presented, demonstrating the fidelity of the simulations.

4:00 PM T2.5
EXPERIMENTAL AND NUMERICAL STUDY OF TWO-PHASE MICROSTRUCTURE FORMATION IN PERITECTIC SOLIDIFICATION. <u>T.S. Lo</u>[1], S. Dobler[2], M. Plapp[3], A. Karma[4] and W. Kurz[2], [1]Courant Institute of Mathematical Sciences, New York University, New York, NY; [2]Department of Materials, Swiss Federal Institute of Technology, EPFL, Lausanne, SWITZERLAND; [3]Laboratoire de Physique de la Matiére Condensée, CNRS/Ecole Polytechnique, Palaiseau, FRANCE; [4]Physics Department and Center for Interdisciplinary Research on Complex Systems, Northeastern University, Boston, MA.

We report the results of a combined experimental and numerical study of two-phase microstructure formation during directional solidification of peritectic alloys. The experiments are conducted in the Fe-Ni system that is ideally suited to investigate microstructural pattern formation in a predominantly diffusion-controlled growth regime due to the small density contrast between solid and liquid that minimizes the amount of convection in the melt. These experiments focus on a regime of large G/V (thermal gradient/growth rate) ratio where the solid-liquid interface of both solid phases is morphologically stable. The two principal microstructures observed are eutectic-like (lamellar and rod) coupled growth structures and new "island banding" microstructures. The latter consist of rows of islands of one solid phase (either peritectic or primary) inside the continuous matrix of the other phase. The latter structures are observed either as transients that seed the formation of coupled growth structures, or as

the final microstructure. On the modeling side, we use a sharp-interface boundary integral model to determine quantitatively the stability limits of coupled growth as a function of lamellar spacing, composition, and G/V ratio, and a diffuse interface phase-field model to investigate qualitatively the complex mechanisms involving both nucleation and growth that control the selection of island banding and coupled growth structures. We find an excellent overall agreement between experimental and numerical results.

4:15 PM T2.6
ONSET OF SIDEBRANCHING DURING DIRECTIONAL SOLIDIFICATION OF DILUTE BINARY ALLOYS. Blas Echebarria and <u>Alain Karma</u>, Northeastern Univ., Physics Department, Boston, MA.

We study directional solidification of dilute binary alloys by means of a phase-field formulation. This avoids the problem of tracking of the interface through the introduction of an order parameter, or phase-field ϕ, which varies smoothly from one value in the liquid to another in the solid across an interface region. However, the resulting equations are stiff, due to the disparity of scales between the interface thickness and the diffusion length. We use a recently derived phase-field model, whose thin interface limit yields a much less stringent restriction on the choice of interface thickness than previous formulations and permits to eliminate non-equilibrium effects at the interface. This allows us to make quantitative comparisons with experiments and study the onset of sidebranching and the cell to dendrite transition for realistic values of the physical parameters. Recently, it has been observed experimentally that increasing the externally imposed thermal gradient reduces the onset velocity and wavelength of sidebranching. We present simulations that confirm this counter-intuitive effect and study the onset of sidebranching as a function of the thermal gradient, pulling velocity and cell spacing.

4:30 PM T2.7
VAPOR-LIQUID EQUILIBRIA OF DENDRITIC POLYMER SOLUTIONS: MOLECULAR THERMODYNAMICS APPROACH. Jeong Gyu Jang, Bong Ho Chang, <u>Young Chan Bae</u>, Hanyang Univ, Div. of Chemical Engineering, Seoul, KOREA.

We investigate vapor-liquid equilibria (VLE) of dendritic polymer (dendrimer & hyperbranched polymer)/solvent systems. We also examine the structure effect of dendritic polymer and the specific interaction due to the difference of interaction energies of endgroup at the periphery of dendritic polymer molecules. In order to predict the activity for systems of dendritic polymer, we use the lattice cluster theory (LCT) with specific interaction consideration and pair method including Monte Carlo sampling method and excluded volume constraint. As a preliminary work to combine the lattice theory for phase behaviors and the molecular simulation, interaction energy parameters are determined by molecular simulations, and then VLE of dendritic polymer solutions are predicted according to the lattice model based on LCT regarding specific interactions with no adjustable model parameters and compared with experimental data. The interaction energy parameters are obtained by the pair method including Monte Carlo simulation method with excluded volume constraint. In general, those parameters are determined by fitting experimental data. Our results show that the specific interactions between the endgroup and the solvent molecule play an important role in determining phase behaviors of the given systems. We obtained all parameters using molecular simulation. We did not fit the experimental data to the model to determine model parameters. However, the difference between the segmental size and the definition of lattice size in lattice model, the rationalization of the coordination number z and the dependence on the force field are needed to be improved to correlate the phase behavior of polymer systems. However, this approach shows that the combination of molecular simulation and the statistical modeling can be the substitute of fully atomistic molecular mechanics or molecular dynamics, which face the difficulties of time and space scale in polymer thermodynamics.

4:45 PM T2.8
MOLECULAR DYNAMICS SIMULATIONS OF REACTIVE WETTING. <u>Edmund B. Webb III</u> and Gary S. Grest; Sandia National Laboratories; Albuquerque, NM.

To examine reactive wetting at the atomistic scale, molecular dynamics simulations of liquid Ag drops on (100) FCC Cu were performed. The interatomic potentials utilized properly describe eutectic behavior for this system so that, at temperatures below melting for pure Cu, extensive alloying occurs. Structural analyses as a function of time, including atomic coordination and wetting angles, permits a description of the kinetics by which the system progresses towards equilibrium. Studying a range in temperature T further elucidates details of the kinetics. In order to establish system size guidelines liquid drops 5, 10, and 20 nm in diameter were simulated. Modeling surface alloying requires substrate atoms be free from

simulation constraints so system sizes must be sufficiently large to avoid artifacts. We demonstrate that stress calculated at an atom according to the virial expression changes for atoms significantly outside the reaction zone, allowing an assessment of necessary system size. Partially alloyed systems were quenched in T to examine nonequilibrium stress states and structural features associated with residual stresses are illustrated. We close with a qualitative discussion of anticipated structural changes given progression towards equilibrium with time.

SESSION T3: COMPLEX PROCESSES AT
SURFACES: SURFACE REACTION,
SEMICONDUCTOR AND COLLOIDAL GROWTH,
ETC.
Chair: Leonard M. Sander
Monday Evening, November 26, 2001
Room 310 (Hynes)

8:00 PM *T3.1
OPTIMIZING THE STRUCTURE OF BIMETALLIC CATALYSTS WITH DYNAMIC MONTE CARLO SIMULATIONS AND GENETIC ALGORITHMS. A.P.J. Jansen, C.G.M. Hermse, Schuit Institute of Catalysis, Eindhoven University of Technology, Eindhoven, THE NETHERLANDS.

We have determined the most the reactive structure of bimetallic catalysts for some simple model systems in which one reactant prefers to adsorb on one component of the catalyst and the other reactant on the other component. We show that there are many structures that can become the most reactive depending on the reaction conditions. The most reactive structure can be predicted without any calculations or simulations in some limiting cases using the concept of the rate limiting step, but in general this is not possible. We then use Dynamic (also called Kinetic) Monte Carlo simulation to describe the kinetics properly, and Genetic Algorithms to optimize the structure of the catalyst.

8:30 PM *T3.2
FROM ATOMISTIC LATTICE-GAS MODELS FOR SURFACE REACTIONS TO HYDRODYNAMIC REACTION-DIFFUSION EQUATIONS. Da-Jiang Liu, Ames Laboratory, Iowa State University, Ames, IA.

Local adsorption, desorption, reaction processes occuring in surface reactions, along with adspecies interactions and diffusion, produce a diverse variety of spatiotemporal pattern formation. Traditionally, this phenomenon has been tackled by mean-field (MF) reaction diffusion equation (RDE). A more fundamental approach is through the use of atomistic lattice-gas models, which can account for effects of adlayer ordering and fluctuations that are neglected in the MF-RDE approach. The pattern formation problem poses a challenge to direct simulations of lattice-gas models. At least for one of the adspecies, the diffusion process happens at a much faster time scale and larger length scale than all the other processes. Our strategy is to develop an appropriate (non-MF) RDE formulation to treat this "hydrodynamic" regime. This requires special simulation procedures, coupled with a correct description of chemical diffusion in mixed reactant adlayers. We apply this approach to a model CO oxidation system that exhibits bistability. The problem of chemical waves and equistability is studied.

9:00 PM *T3.3
GROWTH OF MONODISPERSED COLLOIDS BY NUCLEATION AND AGGREGATION OF NANOSIZE SUBUNITS. Vladimir Privman, Department of Physics and Center for Advanced Materials Processing, Clarkson University, Potsdam, NY.

The importance of well-defined dispersions of particles of different shapes, ranging in sizes from nanometer to colloidal, has been widely recognized in applications and in basic studies of advanced materials. Our program endeavors to advance understanding of formation of uniform particles of simple and composite structure, with focus on synthesis involving nanosize particles and their new unique properties for dimensions smaller than the typical submicron-size colloid scales. Presently, there is convincing experimental evidence that many monodispersed colloids of various shapes, obtained by precipitation in solutions, are formed by aggregation of such nanosize units. Our recent theoretical explanation of this process expands the classical model of formation of uniform particles, by LaMer, and offers an interesting link between nanosize and micrometer size particles. It explains many properties of the latter which could not be previously understood. Thus, the colloid and nano-scale particle dispersions are actually closely related, and we will emphasize uniform aspects of the dynamics of their formation in this presentation.

9:30 PM T3.4
A COMPUTATIONAL INVESTIGATION OF SELECTIVE MOVPE OF $Al_xGa_{1-x}As$ IN THE PRESENCE OF HCl. Maria Nemirovskaya, Carlo Cavallotti, Klavs Jensen, MIT, Dept. of Chemical Engineering, Cambridge, MA.

The selective deposition of $Al_xGa_{1-x}As$ can be performed using MOVPE reactors with the deposition on the mask suppressed by the presence of HCl. The quality of the film and the shape of the features depend strongly on the gas phase composition, temperature and pressure. Multiscale simulations were done in order to improve our understanding of the fundamental processes determining film growth and its morphology. Quantum chemistry calculations were performed to identify the key reactions between HCl and the $Al_xGa_{1-x}As$ surface as well as the gas phase precursors. A gas phase and surface kinetic scheme that is able to predict the growth of AlAs and GaAs in the presence of HCl was thus developed and its validity was tested through the reactor scale simulations of the experimental data. Then the morphological evolution and the onset of the facet formation were studied using kinetic Monte Carlo model. Experimental literature rates of diffusion were evaluated through the simulation of the transition from island to step flow growth. The chemistry of GaCl and AlCl was found to play an important role in the surface evolution. Finally, the linked kinetic Monte Carlo and the reactor scale models were used to study the morphology evolution as a function of the operating conditions. Based on our results we propose a mechanism underlying the morphology evolution of the $Al_xGa_{1-x}As$ films deposited by MOVPE systems.

9:45 PM T3.5
TIME DEPENDENT SURFACE MORPHOLOGY ASSOCIATED WITH GROWTH INTERRUPTS IN GaAs MOLECULAR BEAM EPITAXY. Anders Ballestad, Ben Ruck, Jens Schmid, Tom Tiedje, Univ of British Columbia, Dept of Physics and Astronomy, Vancouver, BC, CANADA.

We have used ex-situ atomic force microscopy and in-situ elastic light scattering experiments to study the time evolution of the surface morphology of GaAs during growth by molecular beam epitaxy. Constant coefficient growth equations describe the surface evolution well during growth [1], but transient effects in the light scattering signal induced by growth interrupts indicate that the coefficients in the growth equation are flux and time dependent. The transition from a constant flux scenario towards a thermodynamical equilibrium on the surface, where islands and step edges settle into a more energetically favourable configuration, is proposed as an explanation for the decaying smoothing rate. We propose a mean-field rate equation approach in order to keep track of incorporation sites and adatoms on the surface during growth and post-anneal. The model extends beyond the dilute sub-monolayer description, and yields qualitative and quantitive agreement with our light scattering experiments.
1. Ballestad, Ruck, Adamcyk, Pinnington and Tiedje, Phys. Rev. Lett. 86}, 2377-2380 (March 12, 2001).

10:00 PM T3.6
ATOMISTIC MODELING OF III/V SEMICONDUCTORS: THERMODYNAMIC EQUILIBRIUM AND GROWTH KINETICS. Frank Grosse[1,2], William Barvosa-Carter[1], Jennifer J. Zinck[1], Mark F. Gyure[1]. [1]HRL Laboratories, LLC, Malibu, CA. [2]Department of Mathematics, University of California, Los Angeles, CA.

The smaller the structural features of III/V semiconductor heterostructures, the more important the atomic nature of surfaces and interfaces. To utilize thermodynamic equilibrium and growth kinetics to design interfaces on the atomic scale makes detailed predictive modeling necessary. We describe a general method of III/V semiconductor modeling on the basis of *ab initio* density functional theory (DFT) calculations, supported by experimental results. Accuracy and predictability is evaluated using the example of a kinetic Monte Carlo (kMC)simulation for the InAs(001) surface. We present results for the phase transition between the $\alpha_2(2\times4)$ and $\beta_2(2\times4)$ reconstruction. Simulated island nucleation in the low coverage regime is in excellent agreement with experiment. Furthermore, relevant microscopic processes for decrease of island number density and decrease of anisotropy with increasing As pressure are revealed: Suppression of effective In adatom density through incoming As leads to reduced island nucleation.
This work is supported by NSF and DARPA through the Virtual Integrated Prototyping Initiative and the NSF Industrial Postdoc Program. The DFT calculations were carried out using the FHI98MD simulation package [1].
[1] Bockstedte, Kley, Neugebauer, Scheffler, CPC 107, 187 (1997). http://www.fhi-berlin.mpg.de/th/fhi98md/

SESSION T4: DEFORMATION-INDUCED
STRUCTURES: ELASTIC AND PLASTIC
BEHAVIOR
Chairs: Alberto M. Cuitino and Anter A. El-Azab
Tuesday Morning, November 27, 2001
Room 310 (Hynes)

8:30 AM *T4.1
CRUMPLED CANDY WRAPPERS AND BENT TAPE MEASURES.
Eric M. Kramer, Physics Dept, Simon's Rock College, Great
Barrington, MA.

We discuss two topics related to the large-deflection behavior of
thin-walled, elastic structures under compression. First, we review the
mechanism of sound production from crumpled sheets of paper and
plastic, and present recent results of high-resolution (200 kHz sample
rate) digital recordings. Next we present a theoretical and
experimental study of the V-shaped kink in a bent tape measure.
Measurements were made of (1) the kink's longitudinal and transverse
profiles and (2) the bending moment as a function of kink opening
angle. Results show good agreement with the predictions of linear
elasticity theory.

9:00 AM *T4.2
STRESS INDUCED INSTABILITIES. Chaouqi Misbah, Klaus
Kassner and Judith Mueller GREPHE, LSP, Univ. J. Fourier,
Grenoble, FRANCE.

We analyze the problem of stress-induced instabilities, with the help
of a phase-field approach. We show that after an initial stage, the
surface corrugation produces grooves that compeete through the
elastic field, the winner of which grow as a fracture. We analyze the
cells coarsening. We reduce dynamics analytically to that of groove
tips and compare them to fracture dynamics models. We show that in
the longwavelength regime dynamics fall into a non-standard
(nonlocal) amplitude equation, which is much efficient for numerical
analyzes. In the grooves the stress accumulation is so strong that
plastic effects become decisive. We analyze this question both from a
macroscopic point of view as well as on the basis of disclocation
dynamics. We show that plastic effects drastically affect dynamics.

9:30 AM *T4.3
THE PHASE FIELD METHOD: MICROSTRUCTURAL
EVOLUTIONS IN ALLOYS AND DISLOCATION DYNAMICS. L.
Proville, Y. Le Bouar, D. Rodney, A. Finel, ONERA, Chatillon,
FRANCE.

The Phase Field Method has been extensively used to study the
dynamical evolution of microstructures inherited from phase
transformations. It has recently been extended to the dynamics of
dislocations and thus to the domain of plastic deformation. We will
present recent results in these two different fields. First, we analyse
the competitive growth of different ordered precipitates in
aluminium-based alloys (Al-Zr). Second, we present a recent extension
of the Phase Field method to dislocation dynamics and plasticity. This
new formalism incorporates simultaneously two different length scales,
the first one associated to the dislocation core radius, and the second
one, needed by the discretization of real space, associated to the long
range scale of slowly varying concentration and long range order fields.
This method will be illustrated of a first approach of alloy hardening.

10:15 AM T4.4
RELATING ATOMISTIC GRAIN BOUNDARY SIMULATION
RESULTS TO THE PHASE FIELD MODEL. Catherine M. Bishop,
W. Craig Carter, Massachusetts Institute of Technology, Dept. of
Materials Science and Engineering, Cambridge, MA.

Atomistic simulations provide detailed results of grain boundary
structures and energies in highly controlled numerical environments.
Mesoscopic models, such as the phase field model, are meant to
extrapolate microscopic information—such as that calculated in an
atomistic model, or measured from HREM—to a continuum and
subsequently be applied to systems that are much larger and more
complex than those that can be studied by direct simulation. We will
present a method of coarse-graining information from atomistic
simulations to order parameters that are required as the energetic
basis of phase field models. The method extracts spatial information
from sets of discrete data and yields a continuously variable order
parameter that can be used to tune a phase field model.

10:30 AM T4.5
STATISTICAL MODELING OF A DISLOCATION PHASE-FIELD
IN DUCTILE SINGLE CRYSTALS. M. Koslowski, M. Ortiz,
Graduate Aeronautical Laboratories, California Institute of
Technology, Pasadena, CA; A.M. Cuitiño, Department of Mechanical
and Aerospace Engineering, Rutgers University, Piscataway, NJ.

A phase-field theory of dislocation dynamics, strain hardening and
hysteresis in ductile single crystals is developed. Specifically, we
consider the motion of large numbers of dislocations within discrete
slip planes through random arrays of point obstacles under the action
of an applied shear stress. A statistical mechanical framework for
dissipative systems was developed to include kinetic and thermal
effects. The energy terms contemplated in the theory include the core
energy of the dislocations, the long-range elastic interactions between
dislocation segments, and the energy of interaction with the applied
resolved shear stress. The simulation is carried out using a path
integral Monte Carlo algorithm with an energy barrier due to the
irreversible interaction between dislocations and point obstacles. The
phase-field representation enables complex geometrical and
topological transitions in the dislocation ensemble, including
dislocation loop nucleation, bow-out, pinching, and the formation of
Orowan loops. The theory predicts a range of behaviors which are in
qualitative agreement with observation, including: hardening and
dislocation multiplication in single slip under monotonic loading;
softening at higher temperatures; Bauschinger effect under reverse
loading; fading memory effect, whereby reverse yielding gradually
eliminates the influence of previous loading; evolution of the
dislocation density under cycling loading, leading to characteristic
'butterfly' curves; and others. The theory permits the coupling
between slip systems; the consideration of obstacles of varying
strengths; and dislocation line-energy anisotropy.

10:45 AM T4.6
CRYSTAL PLASTICITY FROM DISLOCATION DYNAMICS.
Vasily Bulatov, Wei Cai, Tim Pierce, Moon Rhee, Meijie Tang,
Lawrence Livermore National Laboratory, University of California,
CA.

Dislocation Dynamics (DD) models attempt to predict plastic
response of crystalline materials from the collective evolution of large
number of dislocations. The idea is very simple: learn how each
dislocation responds to the forces it sees, throw in a bunch of
dislocations, let them interact with each other, put them under
conditions of stress and temperature that are similar to the laboratory
experiment, sit back and watch what happens. This, seemingly
straightforward approach has not yet delivered on its promise, largely
due to inability of the existing DD codes to deliver computing
performance adequate for handling large enough dislocation groups
and tracing them over long enough time intervals. In this presentation
we discuss the hurdles that need to be overcome for a practical
implementation of the DD method to bring to realization its central
idea, i.e. computing materials strength from dislocation mechanisms.
In particular, we focus on the issues involved in running DD
simulations big (scalable massively parallel computing) and long
(efficient time integration). Results of the first benchmark runs of a
new DD code under development in LLNL will be presented.

11:00 AM T4.7
DISLOCATION CORE AND JUNCTIONS IN BCC METALS.
Wei Cai, Vasily V. Bulatov, Christopher R. Krenn, Lawrence
Livermore National Laboratory, University of California, Livermore,
CA.

Atomistic core mechanisms have been widely recognized as important
factors controlling dislocation mobility and hence crystal plasticity in
BCC metals. While considerable progress has been made along this
direction, more systematic studies are needed for a complete
understanding of the intrinsic dislocation behavior and the effect of
external loading stress. In this presentation, we discuss the effect of
non-glide stress component on the slip preference of 1/2[111] screw
dislocations, as a consequence of degeneracy removal of core
splitting. We also present Molecular Dynamics studies of prismatic glide of
jogged dislocations at different character angles and slip plane
orientations. Mobility of a screw dislocation network consisting of
junctions is also investigated in search for an atomistic understanding
of anomalous slip mechanisms.

11:15 AM T4.8
COMPETITION BETWEEN NUCLEATION OF TWINS AND
DISLOCATIONS: ATOMISTIC SIMULATION OF THEIR
INITIATION DYNAMICS IN BCC-MO. Dongsheng Xu[1,3], Ju Li[1],
Jinpeng Chang[1] and Sidney Yip[1]. [1] MIT, Nuclear Engineering
Department, Cambridge, MA; [2]Institute of Metal Research, Chinese
Academy of Sciences, Shenyang, CHINA.

From finite-T MD simulations using a many-body interatomic
potential, both twinning and dislocation are observed to form in
perfect crystalline bcc-Mo in the aftermath of elastic instabilities
driven by shear. In some cases the twin disappeared in continuing
shear by twinning again in the original place, while in others almost
complete twin transformation achieved by the migration of the
composition plane. The dynamics of their initiations are analyzed,
and the dependence on temperature and confining pressure explored.

We find not only competition but also cooperation in certain loading conditions, where the formation of twin domains facilitates nucleation of dislocations.

11:30 AM T4.9
SIMULATION OF ANELASTIC RELAXATION AND DEFECT INTERACTION IN ALUMINUM. Yoshiaki Kogure, Toshio Kosugi and Masao Doyama, Teikyo University of Science & Technology, Yamanashi, JAPAN.

A large number of mechanical relaxation peaks have been observed in the temperature dependence of internal friction in metallic crystals. The point defects or defect complexes responsible to the peaks are interstitials, impurity pairs, impurity-interstitial pairs and so on. These defects accompany a nonspherical strain field and respond to the external shear stresses. The temperature dependence of internal friction and elastic modulus change are used to be analyzed on the basis of phenomenological anelasticity model developed by Nowick et al., in which the point defect strain per unit concentration is called as "the elastic dipole moment" and denoted by the λ-tensor. According to the model the magnitude of relaxation is proportional to the square of λ tensor. The relaxation time is the function of potential energy at a saddle point, and it can be calculated from the interaction potentials. Both the relaxation strength and the relaxation time for the point defects system in aluminum alloys are calculated in this study. The atomistic displacement, the elastic field around the point defects, and the saddle point energy on the relaxation path are calculated by using the EAM potential based on the molecular dynamics simulation. The interaction of point defects through the strain field is also important, that may result a distribution of relaxation time and a glass-like behavior. The effects of dipole interaction in elastic properties are investigated.

11:45 AM T4.10
THE INFLUENCE OF MANY BODY INTERACTIONS IN THE STRESS-VELOCITY RELATION FOR ONE DISLOCATION IN A 2D LATTICE. M. Robles, Centro de Investigacón en Energía UNAM, Priv. Xochicalco S/N Col. Centro, Temixco, MEXICO; K. Kaski and V. Mustonen, Helsinki University of Technology, Laboratory of Computational Engineering, FINLAND.

The stress-velocity relation of one dislocation moving in a 2-D lattice has been studied using interactive molecular dynamic simulations. A hybrid model coupling the Lenard-Jones(LJ) potential and the Embedded Atom Model (EAM) potential is used to include radial and many body interactions. Both parts are assembled by a parameter that allows to change a pure radial interaction to a strong many body interaction in a continuous way. Setting up a constant-stress scenario, the movement of one dislocation is tracked from the rest, upto a terminal velocity state. The external stress vs. terminal velocity curves are obtained in a subsonic regime for different values of the coupling parameter. Non-linear relations are found in $0.1c_t$ to $0.6c_t$ velocity range, where c_t is the transverse speed of sound. An analysis is done using an augmented Peierls Model to seek the connection between atomic scale, continuum variables and the limiting speed of dislocations.

SESSION T5: DEFORMATION-INDUCED
STRUCTURES: STOCHASTIC METHODS
Chairs: Eric M. Kramer and Alphonse Finel
Tuesday Afternoon, November 27, 2001
Room 310 (Hynes)

NOTE EARLY START

1:15 PM *T5.1
STATISTICAL PROPERTIES OF DISLOCATION ASSEMBLIES. Istán Groma, Eötvös Univesity Budapest, Department of General Physics, HUNGARY.

It is well known that during the plastic deformation of crystalline materials the dislocation distribution does not remain homogeneous. In spite of the increasing experimental and theoretical activity on this field we are far from the understanding of this typically self organizatory phenomenon. A crucial open question is whether the long range dislocation-dislocation interactions which do not have an intrinsic range can lead to spatial patterns which may exhibit well-defined characteristic scales. A possible approach for the modeling of these pattern formation processes is to investigate the collective behavior of systems consisting of individual dislocations by computer simulation. However, because of the long range character of the dislocation interaction the direct numerical integration of the equations of motion of dislocations is very computation expensive restricting considerable the affordable dislocation number or simulation volume. The aim of the investigations presented is to

overcome this restriction by taking into account the statistical properties of the dislocation assembly. In order to reduce complexity the behavior of a system of straight parallel dislocations is investigated. This is a strong simplification of a dislocation network developing during plastic deformation, but it is an ideal model system for studying the consequences of long range interaction. It is found by numerical simulation that the internal stress τ created by the dislocations has a stochastic component. In order to describe this stochastic character the form of the probability distribution function of the internal stress is determined. It is shown that the mean value of the distribution function is the self-consistent field created by the dislocation, and the distribution function decays with $1/\tau^3$. On the basis of these analytical findings an $O(N)$ stochastic simulation method is proposed. It is shown that fractal type dislocation pattern formation can be modeled by this approach.

1:45 PM *T5.2
STOCHASTIC METHOD FOR THE EVOLUTION OF MICROSTRUCTURES WITH PARTICULAR REFERENCE TO GRAIN BOUNDARY NETWORKS. Roger W. Minich, Mukul Kumar, James S. Stölken, Christopher Schuh, Lawrence Livermore National Laboratory, Livermore, CA.

Grain boundary engineering has been successfully applied to improve materials properties such as intergranular corrosion and cracking, creep, weldability, and ductility. This has been attributed to an increase in the fraction of special grain boundaries resulting from thermomechanical processing. Our investigations, however, have led us to conclude that the basis for these improvements is most likely to lie in the break-up of the connectivity of random boundary networks. The experimental determination of the network topology has been accompanied by a microstructure modeling effort that is derived from the percolative nature of the network. The problem of grain boundary networks, under crystallographic constraint, has been simulated using a stochastic approach that considers the probability distribution function for each boundary based on the fluctuations in strain energy (from deformation) and intrinsic characteristics such as interfacial energy. The influence of both multiplicative and additive fluctuations toward microstructural evolution during recrystallization and grain growth will be elaborated. Experimentally derived measures to quantify microstructures such as in the grain boundary character and triple junction distributions will be compared to those obtained from simulations.
This work was performed under the auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

2:15 PM T5.3
TOWARDS A PHYSICAL THEORY OF CRYSTAL PLASTICITY. Anter El-Azab, Pacific Northwest National Laboratory, Richland, WA.

The motion of lattice dislocations induces plastic distortion of metallic crystals. Typically, dislocations are very large in number and their motion and interactions have a statistical character. Currently, the statistical aspects of a dislocation system are being probed using simulation models that are based on the fundamental aspects of individual dislocation lines interactions with the lattice stress field and among each other. Such computational models are statistical-mechanical in nature. Development of a formal non-equilibrium statistical mechanics framework however remains a challenge due to the significant fundamental differences between a dislocation system and a classical system of particles. Some recent progress along these lines is reported here. This progress has two basic ingredients. First, a new interpretation of the compatibility condition in a deforming crystal has been found, from which a new physical law has been extracted- "the integral of the dislocation density tensor over the reference crystal volume defines a topological invariant that characterizes a conserved quantity associated with the deformation field." Second, this invariance characteristic of this topological object is combined with the notion of phase space to develop a set of Liouville-type kinetic equations describing the transport and reactions of dislocations on all slip systems of a deforming crystal. Being linear topological objects, dislocations are modeled as a stochastic fiber process. The problem of crystal deformation is then treated in a holistic fashion where the elastic and plastic distortions and the deformation-induced dislocation and deformation patterns are potentially and simultaneously predictable.

2:30 PM T5.4
DISLOCATION PATTERNING IN TWO DIMENSIONS AND SIZE EFFECTS IN PLASTICITY. Robin L.B. Selinger, Brian B. Smith, and Weidong Luo, Catholic Univ., Physics Dept., Washington, DC.

We study the dynamics of screw dislocations in two dimensions to characterize both defect patterning and size effects in plastic deformation. Our model is a close analogue of the XY rotor model under driving boundary conditions. In a homogeneous solid under a

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constant shear strain rate, we observe spontaneous coalescence of screw dislocations into a pattern of slip bands. We also study deformation in strain gradient geometries, looking at the elastic-plastic response of a ductile crack, and at the deformation field around a strong fiber pulled through a ductile solid. In both cases we find clear evidence of size effects and observe scaling behavior. We discuss the origin of the size effect and the emergence of a characteristic length scale in plastic response.

2:45 PM T5.5
A MULTIPHASE MICROSTRUCTURE EVOLUTION MODEL INCLUDING DISLOCATION PLASTICITY. Fabrizio Cleri and Gregorio D'Agostino, Ente Nuove Tecnologie, Energia e Ambiente, Divisione Materiali, Centro Ricerche Casaccia, Roma, ITALY.

We present a recent extension of our stochastic microstructure evolution model including multi-phase domain evolution and dislocation plasticity. The model is implemented by means of numerical simulations based on the velocity Monte Carlo algorithm. It describes the evolution of a two-dimensional microstructure by tracking the motion of triple junctions, i.e. the vertices where three grain boundaries meet. Grain boundaries can be modeled as straight, curved or discretized segments; the misorientation dependence of both grain-boundary energies and mobilities can be included to represent different textures. Nucleation and growth are also accounted by the same approach. The problem of grain size distribution and its temporal evolution is also dealt with by means of a lattice stochastic dynamical system, which may also account for heat flow. Two and three dimensional model systems were studied by means of the latter approach. Consistent results are obtained in two dimensions by the two different methods, thus confirming generality of scaling laws involved.

3:15 PM *T5.6
STATISTICAL MECHANICS OF DISLOCATION MOTION IN DYNAMIC STRAIN AGING ALLOYS. Peter Hähner, European Commission, DG-JRC, Institute for Advanced Materials, Petten, THE NETHERLANDS; Sebastian Bross, Technische Universität Braunschweig, Institut für Metallphysik und Nukleare Festkörperphysik, Braunschweig, GERMANY.

Plastic deformation by dislocation glide in dynamic strain ageing (DSA) alloys interferes with the accumulation of mobile solute atoms in the dislocation cores. In a certain range of applied strain rate and deformation temperature, the repetitive unpinning and recapture of dislocations surrounded by solute clouds may lead to macroscopic plastic instability associated with the Portevin-Le Chatelier (PLC) effect (serrated yielding). Phenomenologically this is known to go along with a negative strain-rate sensitivity of the flow stress (strain-rate softening instability). In the present paper statistical mechanics concepts are used to investigate how DSA and dislocation interactions on the mesoscopic scale can manifest themselves as macroscopic instability. It is shown that strain-rate softening instability arises as a result of a positive feedback between DSA and long-range dislocation interactions. It represents a non-equilibrium phase transition from individual to collective dislocation glide. To substantiate those theoretical results a mesoscopic 2D computer model is introduced to simulate the thermally activated motion of dislocations within a random array of obstacles. DSA of glide dislocations, which are temporarily arrested at the obstacles, is taken into account in addition to the long-range interactions between the dislocations. Simulation results at constant strain rate and at constant stress rate are presented, and the respective statistical aspects of dislocation activation are discussed. The simulations confirm the analytical results according to which the macroscopic plastic instability associated with the PLC effect is induced by the interplay between dislocation interactions and DSA.

3:45 PM *T5.7
DISORIENTATIONS IN DISLOCATION STRUCTURES: FORMATION AND SPATIAL CORRELATION. Wolfgang Pantleon, Risoe National Laboratory, Materials Research Department, Roskilde, DENMARK.

During plastic deformation of metals, individual crystallites loose their initially uniform orientation. Within each crystallite orientation differences develop due to dislocations becoming trapped in dislocation boundaries after carrying the plastic flow. Any orientation difference across a boundary is directly connected with an excess of dislocations of the same sign in the boundary. In some boundaries (incidental dislocation boundaries) excess dislocations accumulate by stochastic reasons only, in other boundary types (geometrically necessary boundaries) additional deterministic contributions arise. Based on the accumulation of excess dislocations the formation of disorientation angles across dislocation boundaries of both types is treated as a stochastic problem. A stochastic evolution equation (Langevin-equation) for the disorientation angle is derived and solved.

The average disorientation angle increases with plastic strain differently for both types of boundaries in good agreement with experimental observations on different materials. An equivalent formulation as Focker-Planck-equation for the distribution function of the disorientation angles allows the interpretation of the problem as diffusion (with or without drift) in disorientation space with a distinct physical meaning of the diffusion coefficient. The resulting distribution functions show scaling behavior i.e. normalization of the disorientation angles by their average angle leads to a master distribution independent of strain, but depending on the degrees of freedom (number of dislocations sets in a boundary). Comparison with experimental data on cold-rolled aluminum shows a preference for a Rayleigh-distribution corresponding to two (dominating) independent dislocation sets in a boundary. An extension of the model allows the description of spatial correlations between the disorientation angles across neighboring boundaries. An anti-correlation arises from the limited free path of mobile dislocations before their trapping leading to a saturation of the disorientation angle across several boundaries. Experimental evidence is presented and opposing interpretations are discussed as well as the consequences on e.g. recovery.

4:15 PM T5.8
COMPUTER SIMULATIONS OF CATASTROPHIC EVENTS DURING MICROSTRUCTURAL EVOLUTION. Mark A. Miodownik, Department of Mechanical Engineering, Kings College-London, UNITED KINGDOM; Elizabeth A. Holm, Sandia National Labs, Albuquerque, NM.

Abnormal grain growth occurs when a small set of grains grow to consume all the other grains in the system. The phenomenon is of considerable industrial and academic interest but due to its stochastic nature it has been very difficult to study experimentally; the abnormal grains consuming all evidence of the their origin. Computer models have advantage here, in that they can run time backwards to the initial point of an abnormal event. However there are various issues about studying and classifying rare stochastic events using a computer model, such as estimating the minimum size of the model that is required to yield a single event. In a system where the number of variables is very large defining the territory where these possible discrete catastrophic events might occur is very important. In this paper we describe our approach with a cellular automaton in which we gradually increases the complexity of the model in order map out the territory in which abnormal grain growth can occur. We compare our results with the geometrically based analytical theories and draw conclusions about the relative merits of the two approaches in predicting these sort of catastrophic events during microstructural evolution.

4:30 PM T5.9
KINETIC MONTE CARLO STUDY OF DISLOCATION GLIDE IN THE PRESENCE OF SOLUTES. Chaitanya S. Deo, Department of Materials Science and Engineering, University of Michigan, Ann Arbor, MI, Princeton Materials Institute, Princeton University, Princeton, NJ, Department of Mechanical and Aerospace Engineering, Princeton University, Princeton, NJ; David J. Srolovitz, Princeton Materials Institute, Princeton University, Princeton, NJ, Department of Mechanical and Aerospace Engineering, Princeton University, Princeton, NJ.

We present a kinetic Monte Carlo (kMC) simulation of the glide of a <111> screw dislocation on the (011) plane in body centered cubic molybdenum in the presence of substitutional tantalum atoms. In the simulation, the dislocation is treated as a series of linear segments. The solute is distributed in 3-d while the dislocation is constrained to move in the glide plane via a kink mechanism. The kMC simulation include the elastic interactions between the dislocation and solutes as well as solute-core interactions. The kink model explicitly includes double kink nucleation, kink migration and kink-kink annihilation. In BCC molybdenum, kink migration is very fast compared with kink nucleation. This, together with line tension, tend to produce relatively straight screw segments. Thus, long times are required for the formation of a stable double kink. Before the dislocation migration simulations start, we pre-calculate the rate of nucleation of stable double kinks based on a stochastic model that includes elastic interactions, the applied stress, thermal fluctuations and the biased random walk nature of kink migration. In addition, the stable double kink nucleation rate also depends on the local solute distribution. The double kink nucleation model is shown to agree with analytical predictions in some simple limits. Incorporation of such a double kink nucleation model is a key to efficient simulations. Simulations are performed at several solute concentrations and a range of stresses. The stress-plastic strain rate relationships are determined by averaging over several simulations. The results are analyzed in terms of percolation theory.

4:45 PM T5.10
DEFORMATION IN ALUMINUM. Robb Thomson, L. Levine,

Metallurgy Div., NIST, Gaithersburg, MD; and Y. Shim, Center for Simulational Physics, Univ. Georgia, Athens, GA.

This paper will summarize progress in our attempt to model the transport of dislocations through the partially ordered structures generated during deformation. The problem is characterized by strain bursts in time associated with the production of fine slip lines on the surface, and the spatial localization of the fine slip into gross slip bands. A percolation model previously developed applies to the fine slip features, but new approaches to what controls the time between bursts and mechanisms for the spatial localization will be presented. The time between percolation events is determined by recovery processes associated with cross glide and the nucleation of new events within a given slip band. The spatial localization is associated with the development of dense dislocation walls parallel to the active slip plane which channel the slip. A feedback mechanism will be presented which stabilizes these walls in their open channel direction, while new channels are activated as strain increases by the nonpercolating activity.

SESSION T6: DEFORMATION-INDUCED STRUCTURES: CRACKS AND VOIDS
Chair: Tom Arsenlis
Tuesday Evening, November 27, 2001
Room 310 (Hynes)

8:00 PM *T6.1
BRANCHING IN BRITTLE FRACTURE. Leonard M. Sander, Jon Efron, Michigan Center for Theoretical Physics, The University of Michigan, Ann Arbor, MI.

Many studies have confirmed that cracks in brittle materials branch when the crack speed exceeds a certain threshold, v_c, but the value of that threshold is not understood. Almost all theoretical calculations overestimate v_c by factors of two or more. We have shown (L.M. Sander and S.V. Ghaisas, "Thermal Noise And The Branching Threshold In Brittle Fracture" Phys. Rev. Lett. 83, 1994 - 1997 (1999); O. Pla, F. Guinea, E. Louis, S.V. Ghaisas and L.M. Sander, "Straight cracks in dynamic brittle fracture" Phys. Rev. E 61, 11472 (2000)) that thermal noise can reduce the threshold by a substantial amount, and we proposed that this effect can account for the discrepancy. In this talk we extend our treatment to account for quenched noise, and, again show that the threshold for branching is very sensitive to this effect.

8:30 PM *T6.2
PHASE-FIELD MODEL OF MODE III DYNAMIC FRACTURE. Alain Karma, Dept. of Physics, Northeastern Univ., Boston, MA; David Kessler, Dept. of Physics, Bar-Ilan University, Ramat-Gan, ISRAEL; Herbert Levine, Dept. of Physics, Univ. of California, San Diego, La Jolla, CA.

We introduce a phenomenological continuum model for mode III dynamic fracture that is based on the phase-field methodology used extensively to model interfacial pattern formation. We couple a scalar field, which distinguishes between "broken" and "unbroken" states of the system, to the displacement field in a way that consistently includes both macroscopic elasticity and a simple rotationally invariant short scale description of breaking. We report two-dimensional simulations that yield steady-state crack motion in a strip geometry above the Griffith threshold.

9:00 PM T6.3
MODELING VOID NUCLEATION IN COPPER BY ULTRA-SHORT PULSE, LASER-DRIVEN SPHERICAL SHOCKS. J.S. Stölken, M.-J. Caturla, B.A. Remington, Lawrence Livermore National Laboratory, Livermore, CA.

The performance of materials under short time scales and high pressures applied through laser interactions is of great interest for the development of targets for inertial confinement fusion applications. Shock propagation has traditionally been studied using continuum models. However, under certain conditions and geometries the boundary conditions applied to the problem can have an impact in the solutions obtained. Such a situation arises when studying spherical shocks, in particular, at the convergence point. One way of overcoming this limitation and understanding the effect of converging shock waves is to use molecular dynamics simulations. We have studied with this method the effect of spherical shocks on a copper target. The pulse width applied was 1 ps, with peak stresses between 1 and 20GPa. The initial condition is a copper sphere 35nm in diameter. The embedded atom method was used as the interatomic potential for this simulation. We observe the propagation of the shock through the material and follow its change in character (compression vs. tension) and structure. As a result of the applied deformation a void is generated at the center of the copper sphere. We analyze the

formation of the void with respect to the type and strength of the shock applied. We correlate the size of the void with the peak stresses, the size of the original system, and compare to known scaling relations.
This work was carried out under the auspices of the US Department of Energy by Lawrence Livermore National Laboratory under contract W-7405-Eng-48.

9:15 PM T6.4
ATOMISTIC STUDIES OF THE STRUCTURAL MODIFICATIONS IN FUSED SILICA DUE TO SHOCK COMPRESSION. A. Kubota, L. Davila, M.-J. Caturla, J.S. Stolken, B. Sadigh, A. Quong, A.M. Rubenchik and M.D. Feit, Lawrence Livermore National Laboratory, Livermore, CA.

We have studied the atomic-scale processes of dynamic shock-loading of fused silica using molecular dynamics simulations. The calculations predict macroscopic properties such as shock velocity, shock pressure and densification, which are in agreement with experiments. In addition, detailed analysis of the microstructure shows structural modifications associated with permanent densification. An increase in the number of 3- and 4-membered rings is observed after shock densification in agreement with Raman measurements. Changes in coordination of silicon and oxygen atoms are also observed, related to the formation of defects. We discuss the structural modifications in terms of ring, coordination, defect and void spatial distributions. These simulations explain some of the underlying mechanisms in high-power laser damage, where a shock wave is expected to develop. Moreover, they can provide quantitative measurements of changes in the strength of the material during dynamic transformations. Parameters such as density changes or viscosity could then be used as input for macroscopic models of damage. In particular, the obscuration and damage growth during continuous irradiation of these materials will depend on the interaction of the modified silica glass with the laser light. Therefore, the optical properties of this modified silica must be studied. We have computed, using first principle methods, the electronic and optical properties of some of the defects formed during shock-loading. These defects were identified in the classical molecular dynamics simulations mentioned above.
This work was carried out under the auspices of the US Department of Energy by Lawrence Livermore National Laboratory under contract W-7405-Eng-48.

9:30 PM T6.5
STRUCTURAL-PHASE TRANSFORMATIONS IN THIN FILM Ni_3Al TAKING PLACE AT THE PRESENCE OF THE CRACK-INITIATOR IN THE DEPENDENCE ON TEMPERATURE AND PRESSURE. M.S. Dmitrievic, A.A. Ovcharov, G.M. Poletayev, D.M. Starostenkov, General Physics Department, Altai State Technical University, Barnaul, RUSSIA.

Structural-phase transformations in two-dimensional thin film of intermetallid Ni_3Al, taking place in α zone of the crack-initiators in the dependence on deformation, temperature and time are investigated. The interactions between Ni-Al are given by the pair interatomic potentials of Morse type. Thin film is presented by the packing of the plain [111] of FCC lattice. The crack-initiator is oriented in the direction <112>. Structural-phase transformations in the system are investigated by the method of the molecular dynamics. The distribution of velocities of the atomic displacements in the dependence on temperature is given according Boltzman statistics. At the expense of the atomic diffusion of alloy components, the development of the crack is accompanied by the appearance of the phase germs of Ni_2Al and Ni_4Al composition. The kinetics of the reaction is regulated by the temperature and the value of applying pressure. The velocities of similar transformations are determined. The zones of segregations of Al and Ni atoms in the head of the crack-initiators are found. The dislocation mechanisms, regulating the development of the crack according the mechanism of transformation from brittle behavior to plastic one are defined. The presence of germs of new phases also influences on the dynamic of the crack growth.

SESSION T7: LIQUIDS, GLASSES, AND SOLID-LIQUID INTERFACES
Chairs: Brian D. Wirth and Susan N. Coppersmith
Wednesday Morning, November 28, 2001
Room 310 (Hynes)

8:30 AM *T7.1
STATISTICAL MECHANICS TOOLS FOR NON-EQUILIBRIUM LIQUIDS. Denis J. Evans, Research School of Chemistry, Australian National University, Canberra, ACT, AUSTRALIA.

We summarize recent developments in the field of non-equilibrium molecular dynamics (NEMD) computer simulations of fluids.

Developments include: the "method of planes" methods for computing the spatial variation of thermodynamic and transport properties within narrow pores; and the use of entirely configurational information to estimate the temperature. The latter permits us to estimate the local temperature of systems undergoing complex, possibly time dependent flows. We also describe new methods that allow us to control the temperature of systems using entirely configurational, as opposed to kinetic, methods.

9:00 AM *T7.2
ATOMIC-SCALE MODELING OF HETEROPHASE INTERFACES IN ALLOYS. <u>Mark Asta</u>, A. van de Walle, Dept. of Materials Science and Engineering, Northwestern University, Evanston, IL; J.J. Hoyt, Sandia National Laboratories, Albuquerque, NM; A. Karma, Northeastern University, Boston, MA.

Static and dynamic properties of heterophase interfaces play a critical role in dictating non-equilibrium structural evolution resulting from phase transformations. Quantitative modeling of phase transformation microstructures requires a detailed understanding of such interface properties which are characteristically difficult to extract directly from experimental measurement. We will describe the application of classical atomic-scale simulation methods to the study of thermodynamic and kinetic properties of heterophase interfaces in alloys. We focus on applications to alloy solidification where the morphology of dendritic microstructure is sensitively influenced by the degree of crystalline anisotropy in solid-liquid interface properties. Simulations are based upon classical many-body potentials of the embedded-atom form where alloy cross-interaction terms are derived from results of first-principles total-energy calculations. Static properties of solid-liquid interfaces in alloys are derived through Monte-Carlo simulations which provide equilibrated structures as input to molecular dynamics simulations. Due to the rapid time scales associated with liquid-phase diffusion and atom attachment/detachment kinetics at rough solid-liquid interfaces in metals, large-scale molecular dynamics simulations can be applied directly to the calculation of equilibrium and non-equilibrium properties of solid-liquid interfaces including (anisotropic) excess free energies, interface mobilities and non-equilibrium partitioning. This work is supported by the U.S. Department of Energy through the Computational Materials Science Network and the Office of Basic Energy Sciences.

9:30 AM *T7.3
THERMODYNAMIC PROPERTIES OF SOLID-LIQUID INTERFACES FROM ATOMISTIC SIMULATIONS. J.J. <u>Hoyt</u>, Sandia National Laboratories, Albuquerque, NM; M. Asta, Dept. of MS&E, Northwestern University, Evanston, IL; A. Karma, Physics Dept., Northeastern University, Boston, MA.

It is well known that during the solidification of alloys, the shape and velocity of a growing dendrite depends critically on the anisotropy of the solid-liquid interfacial free energy, γ. We demonstrate that this small anisotropy can be computed from molecular dynamics simulations by monitoring the statistical fluctuations in the height of the interface. Results are reported for Ag, Au, Cu, Ni and Pb where the embedded atom method is used to describe the interatomic potential. In addition, it will be shown how the interfacial stress, the excess energy and the excess entropy of the solid-liquid boundary can be obtained from atomistic simulations. In particular the excess entropy can be computed in two ways, from separate computations of both the excess energy and γ or from a direct calculation of the temperature dependence of the interfacial free energy. Results of the two methods will be compared and the values of the excess entropy will be discussed in terms of existing statistical mechanics models.

10:30 AM T7.4
AN EXTENDED CAHN-HILLIARD MODEL FOR INTERFACES WITH CUBIC ANISOTROPY. Thennatur Abinandanan, Indian Institute of Science, Bangalore, INDIA; <u>Ferdinand Haider</u>, Univ. Augsburg, GERMANY.

For studying systems with a cubic anisotropy in interfacial energy σ, we extend the Cahn-Hilliard model by including in it a fourth rank term, {viz}, $\gamma_{ijlm}(\partial^2 c/\partial x_i \partial x_j)(\partial^2 c/\partial x_l \partial x_m)$. This term leads to an additional *linear* term in the evolution equation forthe composition field. It also leads to an orientation-dependent effective fourth rank coefficient γ_{hkl} in the governing equation for the one-dimensional composition profile across a planar interface. The main effect of a non-negative γ_{hkl} is to increase both σ and interfacial width w, each of which, upon suitable scaling, is related to γ_{hkl} through a universal scaling function. In this model, σ is a differentiable function of interface orientation $\hat{\mathbf{n}}$, and does not exhibit cusps; therefore, the equilibrium particle shapes (Wulff shapes) do not contain planar facets. However, the anisotropy in the interfacial energy can be large enough to give rise to corners in the Wulff shapes in two dimensions. In particles of finite sizes, the corners get rounded, and their shapes tend towards the Wulff shape with increasing particle size. Using this model, we studied the influence of an anisotropic interfacial energy on spinodal decomposition.

10:45 AM T7.5
SIMULATION OF CRYSTAL-MELT INTERFACES FOR A SYSTEM OF BINARY HARD SPHERES. <u>Rachel Sibug Aga</u> and Brian B. Laird, Univ of Kansas, Dept of Chemistry, Lawrence, KS.

Crystal-melt interfaces of binary hard spheres are investigated using molecular-dynamics simulation. The system, which is a simple model for binary alloys, consists of a two-component fluid of hard spheres of differing size (defined by the diameter ratio α), in equilibrium with the coexisting crystal phase. For a diameter ratio of $\alpha = 0.414$, two crystal phases coexisting with the fluid are possible, depending on the pressure. At low pressures, the liquid coexists with a pure fcc crystal of the larger particle, while at high pressures a 1:1 binary crystal of "NaCl" type is the coexisting phase [Trizac, *et al.*, Mol. Phys. **90**, 675 (1997)]. For both of these systems, we study the structure and dynamics in the interfacial region as functions of interface orientation. In particular, it is observed that as the interface is traversed from fluid to crystal the diffusion constant of the larger particle vanishes before that of the small particle defining a region of the interface where the large particles are frozen in their crystal lattice, but the small particles exhibit significant mobility. This behavior was not observed for binary hard spheres with diameter ratio of 0.9 [Davidchack and Laird, Mol. Phys. **97**, 833 (1999)]. In addition, structural and dynamical relaxation across the interface do not occur over the same region. Traversing the system from the crystal side to the fluid side, structural relaxation is seen to occur first.

11:00 AM T7.6
SIMULATION OF STRESS RESPONSE AND MECHANICAL RELAXATION IN METALLIC GLASSES. <u>Yoshiaki Kogure</u>, Masao Doyama, Teikyo Univ of Science & Tech, Yamanashi, JAPAN.

Materials of the glassy state, a quenched state of liquid, are known to show peculiar mechanical properties, which is related with the disordered configuration of atoms. Elastic and anelastic responses to the external stresses are investigated by calculating the displacement and the relaxation of atoms under the stress based on the molecular dynamics simulation. Metallic glasses of binary alloy, such as Ag-Cu and Ag-Ni, are chosen as model systems. The simulation systems contain several thousands of atoms and the periodic boundary condition is adopted. Embedded atom method potentials for the materials developed by the present authors are adopted for atomic interactions. As an initial condition atomic systems are kept in molten state, then the systems are quenched to make glassy states. The structure of the systems in glassy state is examined through the radial distribution function. The external stress is applied by deforming the simulation cell, and the motion of atom is monitored. The distribution of internal stress is evaluated from the configuration of the nearest neighbor atoms and the potential energy of individual atoms. The temperature of the material is increased and the relaxation of internal stress is monitored. Especially a large relaxation is observed under the shear stress. The crystallization from the amorphous state is one of the useful process to produce a nanophase material. The annealing of the model system is also tried to realize the partial crystallization, and the changes of atomistic structure in the system are investigated.

11:15 AM T7.7
TRANSPORT PROPERTIES OF LIQUID AND AMORPHOUS METALLIC SYSTEMS USING MOLECULAR DYNAMICS TECHNIQUES. <u>Frank J. Cherne</u>, Michael I. Baskes, Los Alamos Natl Laboratory, Los Alamos, NM; Pierre A. Deymier, University of Arizona, Dept of Materials Science and Engineering, Tucson, AZ.

The advanced casting modeler requires accurate viscosity and diffusivity data of liquid metals and their alloys. The general scarcity of this data combined with the experimental difficulties in obtaining this data motivates this work. Utilizing both the equilibrium and non-equilibrium molecular dynamics techniques, we calculate the diffusivity and viscosity of several technologically important metals and their alloys (i.e. aluminum, nickel, zirconium, aluminum-nickel alloys, and nickel-zirconium alloys). We compare the transport properties obtained from both EAM and MEAM potentials to the available experimental data. The results from this work indicate that equilibrium and non-equilibrium molecular dynamics techniques provide reliable and accurate data required by the advanced casting modeler.

11:30 AM T7.8
SIMPLE ATOMIC MODEL WITH UNUSUAL THERMO-DYNAMICS. <u>P. Keblinski</u>, R.K Dash, Material Science and Engineering Department, Rensselaer Polytechnic Institute, Troy, NY; M.Z. Bazant, Department of Mathematics, Massachusetts Institute of Technology, Cambridge, MA; and M.M. Treacy, NECI, Princeton, NJ.

Using molecular dynamics simulations we studied the thermodynamic behavior of a recently proposed model of a single-component covalent material with atomic interactions described by an empirical environment-dependent-interatomic potential (EDIP). The model exhibits a range of unusual properties typically found in more complex materials. They include negative thermal expansion coefficients for the crystalline and low-temperature amorphous structures, and the existence of two structurally distinct disordered phases. Structural differences between the two disordered phases lead to a first-order transition between them and imply the existence of a second critical point. Such behavior is analogous to that believed to exist for amorphous forms of frozen water.

11:45 AM T7.9
"PSUEDO-CHARGE-TRANSFER" SIMULATION METHOD FOR LIQUID AMORPHOUS AND METALLIC SYSTEMS.

Frank J. Cherne, Ricardo B. Schwarz, Michael I. Baskes, Los Alamos National Laboratory, MST-8, Los Alamos, NM; Pierre A. Deymier, University of Arizona, Dept. of Materials Science and Engineering, Tucson, AZ; Srivilliputhur G. Srinivasan, Los Alamos National Laboratory, T-11, Los Alamos, NM.

Molecular dynamics has proven a useful tool for understanding the structure of liquids and glassy structures. Although these simulations predict bulk properties of these amorphous structures reasonably well, they fail to reproduce more detailed changes in properties with composition (i.e. radial distribution functions, viscosity, and shear moduli). It has been observed that current potentials used to simulate metals predict certain structural information for the binary alloy vastly different from experiment. The limitations of these simulations may reside in the fact that the potentials ignore the role of charge transfer between the elements. A strong charge transfer is expected between pairs of elements with largely negative heats of mixing. We have incorporated charge transfer effects in our potentials using two methodologies: by a direct evaluation of Coulombic charge transfer and by changing the atomic radius of an atom depending on its atomic environment. We will present the results for two binary metallic systems with rich phase diagrams, namely, aluminum-nickel and nickel-zirconium. These model systems were chosen primarily due to the complexity of their phase diagrams. For the first alloy we are interested in reproducing the value and composition dependence of the liquid viscosity, whereas in the second alloy we are interested in reproducing the measured changes in the shear modulus with composition in its amorphous phase. It will be shown through an analysis of a variety of properties that each of these "pseudo-charge-transfer" techniques can provide valuable insight into the structure and formation of metallic glasses.

SESSION T8: INTERFACES IN POLYCRYSTALS
AND ALLOYS
Chairs: Sidney Redner and Jeffrey J. Hoyt
Wednesday Afternoon, November 28, 2001
Room 310 (Hynes)

1:30 PM *T8.1
GRAIN BOUNDARY AND DISLOCATION MIGRATION IN ALLOYS. <u>David J. Srolovitz</u>, Mikhail I. Mendelev, Chaitanya S. Deo, Princeton Materials Institute and Dept. of Mechanical & Aerospace Engr, Princeton University, Princeton, NJ.

The motion of extended defects, such as grain boundaries and dislocations, in crystalline materials generally occurs by nucleation and propagation of kinks on one-dimensional defects and islands on two-dimensional defects. Models for defect motion based upon this mechanism have been studied for decades. In this presentation, we discuss the motion of extended defects in crystalline materials in the presence of solute. Solute atoms can have both long range and short range interactions with extended defects. In most cases, it is the short range interactions that have the largest effect on defect migration. We present a series of simulation studies based upon generalized Ising models that examine the influence of static and diffusing impurities on the motion of extened defects. In particular, we focus on the cases of grain boundaries, where the impurity interactions are short-range only and on dislocations where there are both long- and short-range interactions. We also develop analytical models for the mobility of these defects as a function of solute concentration, solute diffusivity and the heat of solute segregation.

2:00 PM T8.2
MONTE-CARLO STUDIES OF ORDERING KINETICS IN THE LONG-PERIOD SUPERLATTICE ALLOY Cu-Pd. X. Wang, <u>K.F. Ludwig, Jr.</u>, Boston University, Dept. of Physics, Boston, MA; X. Flament, R. Caudron, ONERA, Chatillon, FRANCE.

Monte-Carlo studies of the 1-d long-period superlattice (LPS) ordering kinetics in Cu-Pd have been performed at the 21 at.% Pd composition. Long-range effective pair interaction potentials for the simulations were derived from x-ray diffuse scattering measurements. The simulations reproduce the equilibrium LPS structure of the alloy as well as the salient kinetics features observed in our time-resolved x-ray scattering studies. The important kinetics phenomena observed include a delayed growth of the central superlattice peaks relative to the satellite peaks and a nonmonotonic evolution of the satellite peak positions with time. The simulations show that, following a quench of the disordered alloy into the 1-d LPS region of the phase diagram, compact structures first form that are modulated in more than one direction. Both conservative and non-conservative antiphase boundaries are initially present. Only later do these compact modulated regions grow anisotropically, eliminating the nonconservative antiphase boundaries, and forming the equilibrium 1-d modulated LPS structure.

2:15 PM T8.3
MICROSTRUCTURE EVOLUTION IN ELASTIC MATERIALS UNDERGOING STRUCTURAL TRANSFORMATIONS. <u>Turab Lookman</u>, Avadh Saxena, Subodh Shenoy and Dorian Hatch, Theoretical Div, Los Alamos Natl Lab, Los Alamos, NM.

We show how a unified understanding of both displacive and reconstructive phase transformations is possible from a mesoscopic Ginzburg-Landau free energy entirely in strain variables. An important feature is a long-range anisotropic interaction in the order parameter strain components that mediates different elastic textures under various thermodynamic conditions. For instance, local stress inhomogenieties can effect structures on a global scale. The formalism is readily applied to all major crystal systems in 2D and 3D. We will illustrate it for structural transitions of the square and triangular lattices in 2D. We present microstructure results of dynamic simulations relevant for materials such as FePd and lead-orthovanadate.

2:30 PM T8.4
AN ATOMICALLY-BASED CONTINUUM FORMULATION FOR DIFFUSIONAL PHASE TRANSFORMATIONS. <u>Krishna Garikipati</u>, Univ of Michigan, Dept. of Mechanical Engr, Ann Arbor, MI.

Diffusional transformations in crystalline materials are modelled. A continuum formulation is developed, that nevertheless accounts for the atomic-level processes of exchange, agglomeration, generation and annihilation of species. Such mechanisms are considered and, accompanied by local states of stress and strain, provide the thermodynamic basis for this approach. Densities of internal energy, work and entropy are thus constructed explicitly and combined into the Gibbs free energy density. Formal methods of continuum mechanics then yield constitutive equations that reflect the coupling of composition and mechanics. Balance laws, consisting of continuity equations coupled between diffusing species and the equation of quasistatic stress equilibrium, complete the continuum description. This formalism has been previously been applied to stress-mediated self-diffusion in polycrystalline metals. The addition of surface/interface energies, composition-dependent properties, nucleation conditions and mathematically and physically-motivated methods for interface evolution will be discussed. These extensions allow the modelling of diffusional phase transformations that follow the kinetics of nucleation-and-growth or spinodal decomposition. Applications of interest include phase precipitation and clustering during dopant diffusion—a stage in semiconductor manufacture, and strain-based self-assembly of micro/nano-structures. Void nucleation in metal interconnect lines, due to self diffusion of metal atoms mediated by stress and electric fields, has many features in common with problems of phase transformation and can be formally treated using these methods. The spatial scales involved in these processes are in the micron to deep submicron range, motivating the atomically-based methods. The theory and formulation will be discussed with special emphasis upon the aspects that are crucial to diffusional phase transformations. Some special solutions will be demonstrated and, time permitting, numerical implementation and examples will be presented.

2:45 PM T8.5
COMPUTATIONAL EVALUATION OF THE PHASE DIAGRAM OF THE Au-Ni BINARY ALLOY. DETERMINATION OF THE *SOLVUS*, *LIQUIDUS*, AND *SOLIDUS* LINES AS PREDICTED BY AN EAM POTENTIAL. E. Ogando[1], M. Caro[2,3], and <u>A. Caro</u>[2,3], [1]Elektrika eta Elektronika Saila, UPV-EHU 644 P.K., Bilbo, SPAIN; [2]Centro Atomico Bariloche, Bariloche. ARGENTINA; [3]Atomistic Simulation Group, Queen's University Belfast, NORTHERN IRELAND, UNITED KINGDOM.

The description of binary alloys using classic interatomic potentials and computer simulations are often based on a scheme that first adjusts the pure elements, and then both dilute heats of solutions, which are fit using additional degrees of freedom that do not alter the properties of the pure elements. In this way the nature of the phase diagram that is predicted by the model is usually unknown. In this work we evaluate the temperature dependence of the Gibbs free energy of the liquid and solid phase of the simple Au-Ni solid solution. Using molecular dynamics simulations with an Embedded Atom Potential available in the literature, the switching Hamiltonian method, and thermodynamic integration, we obtain what we believe is the first evaluation of the entire phase diagram predicted by this type of models. We observe that, although the solid state properties of the solid solution, i.e. the location of the solvus line and the critical temperature of the miscibility gap, is in good agreement with the experiments, the properties of the liquid depart from the experiments in the sense that the predicted excess enthalpy of mixing is negative. This in turn locates the temperature of the congruent melting well below the experimental point. Within the regular approximation the systems appears then to be a peritectic, while the exact calculation of the free energies in the entire compositional range gives a phase diagram in qualitative agreement with the experiments.

3:30 PM T8.6
MODELING CASCADE AGING IN DILUTE Fe-Cu ALLOYS. Brian D. Wirth, Lawrence Livermore National Laboratory and G. Robert Odette, University of California, Santa Barbara, CA.

The continued safe operation of nuclear reactors and their potential for lifetime extension depends on ensuring reactor pressure vessel integrity. Reactor pressure vessels and structural materials used in nuclear energy applications are exposed to intense neutron fields that create highly non-equilibrium defect concentrations, consisting of a shell of self-interstitial atom and clusters surrounding a vacancy-rich core, over picosecond time scales. This spatially correlated defect production initiates a long chain of events responsible for microstructure evolution and hence irradiation embrittlement. In this paper, we describe the combined use of molecular dynamics (MD) and kinetic lattice Monte Carlo (KMC) to simulate the long-term rearrangement (aging) of displacement cascades in dilute Fe-Cu alloys. The simulations reveal the formation of a continuous distribution of three dimensional cascade vacancy-Cu cluster complexes and demonstrate the critical importance of spatial, as well as short and long-time correlated processes that mediate the effective production of primary defects. Finally, this approach can generate production cross-sections for vacancy-Cu clusters that can then be used in rate theory type models of long term global micro and microstructural evolution.

3:45 PM T8.7
NONEQUILIBRIUM PHASES SYNTHESIZED BY IRRADIATION: EFFECTIVE DRIVING FORCES AND FLUCTUATIONS. Raúl A. Enrique and Pascal Bellon, Univ. of Illinois at Urbana-Champaign, Dept. of Materials Science and Engineering and Frederick Seitz Materials Research Laboratory, Urbana, IL.

Alloys under irradiation are continuously driven away from equilibrium. Every energetic particle colliding with the atoms of the solid produces a perturbation, very localized both in space and in time. The accumulation of such events can, in the long-time regime, alter the structure of the material, and lead the system to nonequilibrium phases and microstructures. In particular, heavy-ion irradiation has been established as a processing tool to synthesize unstable and metastable phases. An example is the preparation of solid solutions from immiscible elements, a phenomenology relevant, as well, to the stability of second phases in structural components subject to irradiation. In this work, we go beyond existing deterministic models and formulate an effective thermodynamics description of this far-from equilibrium process that includes both an effective free energy (which drives the dynamics and is minimum at steady state), and a measure of the nonequilibrium fluctuations. We have recently carried out studies of ion-beam mixing of immiscible binary alloys, both theoretically, based on kinetic Monte Carlo simulations (KMC) and Cahn-Hilliard-type modeling, and experimentally, based on electron microscopy. We have found that when irradiation mixing is in dynamical balance with thermal decomposition, the alloy can spontaneously form self-organized compositional patterns at the nanometer scale. Furthermore, our KMC results show that a solid solution stabilized by irradiation can exhibit pair correlations (as measured by the structure factor) that are intrinsically different from those of equilibrium solid solutions. To investigate this problem theoretically, here we formulate an extension to the stochastic Cahn-Hilliard-Cook equation that includes the effects of irradiation mixing. From this equation we obtain the expression for the nonequilibrium structure factor, which is in agreement with the KMC simulations. We propose that these results can be confronted experimentally to small-angle scattering analysis of solid solutions synthesized by ion irradiation. Moreover, we use our formulation to study of the effect of fluctuations on the wavelength selectivity of self-organized compositional patterns.

4:00 PM T8.8
DYNAMICAL SELF-ORGANIZATION UNDER IRRADIATION OF ALLOYS UNDERGOING PHASE SEPARATION AND ORDERING. Jiwen Liu, Pascal Bellon, Materials Research Laboratory, University of Illinois at Urbana-Champaign, IL.

Experiments on Ni-Al have suggested long ago that, under appropriate conditions, ion irradiation can induce the stable coexistence at a finite, nanometer scale of a chemically ordered phase (here an L12 ordered phase) with an fcc solid solution (A1 phase). However, the origin of such a mesoscopic phase coexistence, as well as the wavelength selection rules, have remained so far unclear. For the simpler case of an alloy exhibiting a miscibility gap between two disordered solid solutions (e.g., Cu-Ag), our recent theoretical and simulation work indicates that spontaneous organization can be due to the irradiation forced atomic jumps take place at a finite range in displacement cascades, whereas thermally activated diffusion proceeds by short range jumps. In the present work, we extend this approach and we consider the case of an alloy that can both undergo phase separation and phase ordering. We introduce a time-dependent Ginzburg-Landau model that describes the evolution of the alloy under irradiation. Various models are discussed for the forced mixing: short range, finite range, or arbitrary range mixing. Atomistic kinetic Monte Carlo simulations, using a vacancy-based mechanism for thermal diffusion and various forced mixing models, are performed on an fcc lattice and compared to analytical predictions. A linear stability analysis of the continuum model indicates that for moderate irradiation-induced jump frequency, the alloy may self-organize into patterns of coexisting L12 and A1 phases. This self-organization is driven by the impact of the finite range forced mixing on the composition field. This prediction is confirmed by our kinetic Monte Carlo simulations: under moderate irradiation conditions phase coexistence of L12 ordered phase and A1 disordered phase saturates at the nanometer. These results corroborate the idea that irradiation can be used as a method to synthesize nanocomposite materials containing chemically ordered phases.

4:15 PM T8.9
PHASE TRANSFORMATIONS INDUCED BY ARSENIC IMPLANTS INTO SILICON. James P. Lavine, David D. Tuschel, Donald L. Black, Eastman Kodak Company, Rochester, NY.

Micro-Raman spectroscopic investigations of arsenic-implanted silicon show lines characteristic of silicon crystallites [1] even at implant doses above the amorphization threshold. The intensity and frequency of occurrence of the lines increase with the implanted dose. The present polarization/orientation Raman studies rule out silicon in the diamond phase as the source of the 505 to 510/cm line and indicate the 520/cm optical phonon line is not due to the substrate silicon. Monte Carlo simulations of the arsenic ion energy loss suggest each arsenic ion deposits sufficient energy to locally melt the silicon lattice. This is supported by detailed molecular dynamics calculations for low energy implants [2], which find the localized heating dissipates in picoseconds. It is probable that upon cooling a phase transformation leads to silicon crystallites in both the diamond phase (Si-I) and the hexagonal phase (Si-IV), as the observed Raman lines are consistent with these phases. Hexagonal silicon was found by Tan et al. in diffraction studies of arsenic-implanted silicon [3]. This work will briefly outline the Raman spectroscopic evidence for the two phases of silicon. This will be followed by a careful examination of the time scales for heat dissipation and arsenic ion cascade overlap. Simple estimates for the former based on the thermal diffusivity of silicon lead to nanoseconds, while the latter factor brings in dose effects. These time scales are important for estimates of the crystallite size resulting from the phase transformations. The energetics of the formation of Si-IV versus Si-I will also be addressed.
1. D.D. Tuschel and J.P. Lavine, MRS Symp. Proc. 588, 227 and 149 (2000).
2. M.-J. Caturla et al., Phys. Rev. B54, 16683 (1996).
3. T.Y. Tan et al., Philos. Mag. A44, 127 (1981).

4:30 PM T8.10
MOTION OF BOUNDARIES BY DISCRETIZED MEAN CURVATURE IN 2D AND 3D SPIN MODELS. Elizabeth A. Holm, Materials and Process Modeling, Sandia National Laboratories, Albuquerque, NM; Mark A. Miodownik, Department of Mechanical Engineering, King's College London, London, UNITED KINGDOM.

Spin models, such as kinetic Monte Carlo Ising and Potts models, are statistical mechanics models widely used to study grain growth, Ostwald ripening, magnetic domain coarsening, and other phenomena driven by surface energy minimization. However, the relationship between motion of a discretized boundary and motion of a continuum

boundary has not been elucidated. We show that under appropriate conditions, a discretized boundary moves with velocity proportional to its discretized mean curvature. Thus, boundaries in spin models obey a discrete version of the continuum boundary motion law. The presence of domain junctions and lattice anisotropy also influence boundary motion in spin model systems.

4:45 PM T8.11
CURVATURE DRIVEN GRAIN BOUNDARY MIGRATION IN ISING MODEL. <u>Mikhail I. Mendelev</u>, David J. Srolovitz, Princeton Materials Institute and Dept. of Mechanical & Aerospace Eng., Princeton University, Princeton, NJ.

The evolution of polycrystalline microstructures is determined by the relative ease with which grain boundaries can move. The boundary mobility is the ratio of the boundary velocity and the driving force for migration. In order to test whether the mobility of the boundary is independent of the type of driving force and the migration geometry, we have performed a series of simulations within the framework of a simple, Ising model. In particular, we simulate boundary migration on a 2-d triangle lattice with non-conserved spins and Glauber dynamics. Two types of boundary motion are considered: motion under an applied driving force (magnetic field) and curvature driven grain boundary migration. In the first case, we consider boundary motion in two distinct crystallographic directions. It is found that the mobilities for these two direction of motion are not only differ in magnitude but exhibit fundamentally different temperature dependences. The mobility of the boundary with an applied driving force can be satisfactorily described using a kink model. Curvature driven boundary migration is examined in both half-loop and circular grain geometries. The mobility is found to depend only very weakly on the half-loop orientation. While the boundary mobilities were very similar for all cases where the motion is driven by curvature, they are much different than those found in the presence of an externally applied force. The mobilities measured in the curvature driven cases were found to be a much weaker function of temperature than those measured in the presence of an applied force. The shape of the half-loop is in excellent agreement with an analytical, continuum model.

SESSION T9: DISORDERED MATERIALS
Chair: Babak Sadigh
Wednesday Evening, November 28, 2001
Room 310 (Hynes)

8:00 PM *T9.1
HOW LONG DOES IT TAKE FOR A FILTER TO CLOG?
<u>Sidney Redner</u>, Boston Univ, Dept of Physics, Boston, MA.

In depth filtration, a dirty fluid is "cleaned" by trapping dirt particles within the pore space during flow through a porous medium. These trapped particles block individual pores until the filter is clogged and no additional flow occurs. This leads to a self-generated gradient percolation process which exhibits a power law distribution for the density of trapped particles at downstream distance x from the input. A deterministic model for the pore space is introduced which predicts this trapped particle distribution. The time evolution of the clogging process is also studied. The most interesting situation is when the particle and pore size distributions overlap substantially. By developing a simple extremal model for the history of the clogging process and exploiting basic ideas from extreme value statistics, it is shown that the distribution of clogging times generically has a power-law long-time tail. These results are in excellent agreement with simulations of filtration on lattice networks. Some consequences of this theory for optimal filter operation are also discussed.

8:30 PM T9.2
MIXING PHASE FIELD AND FLUID-STRUCTURE INTERACTIONS. <u>Adam Powell</u>, David Dussault, MIT, Dept of Materials Science and Engineering, Cambridge, MA.

A phase field model is presented for transport of elastic solids in a fluid, using a mixed elastic-viscous stress formulation. Elastic modulus is set to zero in the fluid, and viscosity to zero in the solid, with intermediate values of both in the diffuse interface. The model presented here uses a simple Cahn-Hilliard free energy function, but extension to more complex free energies is straightforward, including anisotropic functions where local orientation is updated according to the vorticity. An illustration of the model using finite differences in two dimensions is presented. Numerical difficulties such as interface erosion are discussed.

8:45 PM T9.3
GRANULAR "GLASS" TRANSITION. <u>Leonardo E. Silbert</u>, Sandia National Laboratories, Albuquerque, NM; Deniz Ertaş, Corporate Strategic Research, ExxonMobil Research and Engineering,

Annandale, NJ; Gary S. Grest, Sandia National Laboratories, Albuquerque, NM; Thomas C. Halsey, Corporate Strategic Research, ExxonMobil Research and Engineering, Annandale, NJ; Dov Levine, Department of Physics, Technion, Haifa, ISRAEL.

We study, through large-scale simulations, the transition from a flowing to a static state of a dense packing of granular particles flowing down an inclined plane. As the tilt angle θ, is reduced to the angle of repose θ_r, the angle at which flow ceases, the system cools and densifies. We present results for the distribution of normal forces $P(F)$. At large F, $P(F)$ decays exponentially and is finite as $F \to 0$. As θ approaches θ_r from above, $P(F)$ develops a hump at small forces. For $\theta < \theta_r$, a peak emerges in $P(F)$. This is accompanied by the growth of the first peak and splitting of the second peak in the radial distribution function $g(r)$, suggesting compaction and increased short-range ordering. Furthermore, we observe a significant change in the mechanical state of the system: although a finite fraction of interparticle contacts are sliding (at yield) for all flowing piles, the contacts become elastic (away from yield) when the pile becomes static. We define a staticity index that determines how close the system is to marginal rigidity, and observe how the system approaches this limit from the flowing to the non-flowing state, comparing the static case with dense packings generated through alternative methods.

9:00 PM *T9.4
COMPARING THE LOW ENERGY LANDSCAPES OF A CLASSICAL AND A QUANTUM SPIN GLASS. James W. Landry and <u>S.N. Coppersmith</u>, James Franck Institute, University of Chicago, Chicago, IL.

The Ising $\pm J$ spin glass is a canonical example of a random system in which competing interactions lead to frustration and thus large numbers of configurations with the same energy. We investigate how the low energy landscape of this model changes when small additional terms are added to the Hamiltonian. We have studied a two-dimensional model in which the spins are coupled to lattice degrees of freedom (a compressible model) as well as a model with a single-site quantum tunneling term. Our method first finds *all* ground states of the $\pm J$ spin glass, and then incorporates the the effects of compressibility and of quantum tunneling using lowest order perturbation theory. We find that the low energy states of the compressible system and of the quantum system differ qualitatively. The quantum eigenstates are surprisingly complex, typically being superpositions of a macroscopic fraction of all the classical ground state configurations of the system.

9:30 PM T9.5
ACTIVATED FRONT PROPAGATION IN RANDOM MEDIA. Rune Skoe, <u>Damien Vandembroucq</u> and Stéphane Roux, Unité Mixte CNRS/Saint-Gobain Aubervilliers, FRANCE.

We present a numerical study of the propagation of an elastic chain in a random pinning potential at finite temperature. Depending on the range of the elastic interaction this model can describe vortices motion in type II superconductors, charge density waves in solids, wetting or crack front propagation. We focus on the creep regime obtained below the depinning transition. We study the dependence of the velocity-force characteristics and the front roughness statistics on the temperature and on the range of the elastic interactions.

9:45 PM T9.6
EFFECTS OF RANDOM STRAIN FIELD ON FIRST ORDER PHASE TRANSITION. <u>Mo Li</u>, Department of Materials Science and Engineering, The Johns Hopkins University, Baltimore, MD.

A phenomenological theory is proposed to explain the effects of frozen defects and the associated random strains on first order phase transitions. Through a linear coupling term between the primary order parameter and the strain field, the Landau theory shows that first order phase transitions, such as melting, solid state amorphization and some displacive phase transitions, could become continuous. The primary reason for this crossover phenomenon is the enhancement of the fluctuations caused by the presence of the defect-induced random strain field. The transition temperature is shown to be supressed and transition is smeared with decrease in the latent heat and the amplitude of the order parameter when the critical point is approached. The elastic modulii are shown to decrease with increase of defect density and exhibit cusps as the critical defect density is reached at the tricritical point.

10:00 PM T9.7
MODELLING THE POISSON'S RATIO OF CELLULAR AND MICROPOROUS SOLIDS. <u>Ken Evans</u>, Chris Smith, Dept of Engineering, University of Exeter, Exeter, UNITED KINGDOM; Joseph Grima, University of Malta, Msida, MALTA.

Cellular or microporous solids have many advantages over solid materials, one of which is the ease in which a negative Poisson's ratio can be engineered. Materials with negative Poisson's ratios (auxetic materials) have many potential advantages over similar but positive Poisson's ratio materials, such as increased indentation resistance, sound absorption and increased shear modulus. Understanding of the mechanisms giving rise to the auxetic behaviour is crucial for the engineering of manufacturing processes. A three dimensional model is presented for the behaviour of several classes of microporous polymers and cellular solids. The model is based upon the observable geometry of micro/structure of the materials and predicts macroscopic behaviour form that of a repeating unit cell. Three concurrent elastic and plastic deformation mechanisms are considered. Experimental data from a variety of conventional and auxetic materials are compared with model predictions and conclusions drawn.

SESSION T10: NEWLY DEVELOPED TOOLS
Chairs: Arthur F. Voter and A.P.J. Tonek Jansen
Thursday Morning, November 29, 2001
Room 310 (Hynes)

8:30 AM *T10.1
FLEXIBILITY IN MOLECULES, GLASSES AND PROTEINS.
M.F. Thorpe, Michigan State Univ, East Lansing, MI.

The rigidity of networks can be determined using an exact constraint counting algorithm that maintains all nearest neighbor bond lengths and bond angles [1,2] plus other constraints as required [3,4]. We analyze our results in terms of the rigid region decomposition of the network, and identify the stressed regions and also the hinge joints between rigid clusters, that lead to flexibility. We quantity the floppiness of the network by defining a local flexibility index as the density of floppy modes. We also quantify the amount of stress in the network via the stability index, which is the density of redundant bonds. We give three examples of this approach. In the first we show how the flexibility of macro-molecules containing rings can be approached using these techniques. In the second, we show that in glasses, these results relate to the intermediate phase [2] where self-organization is thought to occur. Finally we show how protein unfolding corresponds to a first order phase transition in which the rigidity of the protein core is lost [3,4].
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[2] M.F. Thorpe, D.J. Jacobs, M.V. Chubynsky and J.C. Phillips, J. Non-Cryst. Sols. 266-269, **859-866** (2000).
[3] M.F. Thorpe, Ming Lei, A.J. Rader, Donald J. Jacobs and Leslie A. Kuhn Proteins **44**, 150 - 165, (2001).
[4] M.F. Thorpe, Ming Lei, A.J. Rader, Donald J. Jacobs and Leslie Kuhn Journal of Molecular Graphics and Modelling **19**, 60 - 69, (2001).
Work done in collaboration with Leslie A. Kuhn, Ming Lei, N. Chubynsky, A.J. Rader and B. Hespenheide and supported in part by the National Science Foundation.

9:00 AM T10.2
COARSE-GRAINED MOLECULAR DYNAMICS FOR NANOMECHANICAL RESONATOR MATERIALS. Robert E. Rudd, Condensed Matter Physics Division, Lawrence Livermore National Laboratory, Livermore, CA; Jeremy Q. Broughton, formerly of Naval Research Laboratory, Washington, DC.

Coarse-Grained Molecular Dynamics (CGMD) is a modeling methodology we have developed to simulate inhomogeneous, multiscale systems. [1,2] In particular, it is designed for systems in which nanoscale atomistic dynamics in a small region of the system is strongly coupled to continuum elastic fields that extend for microns into the surrounding material. The model is formulated by using statistical mechanical projection techniques to remove irrelevant short distance degrees of freedom in the periphery. In the central atomistic region, the motion of millions of atoms is fully resolved–there is no coarse-graining. In the surrounding material which would encompass billions of atoms, coarse-graining reduces the number of degrees of freedom to a few hundred thousand. The resulting increase in computational efficiency allows us to model the molecular dynamics to micron-scale systems. We have used this methodology to search for deviations from continuum mechanical predictions in NanoElectro-Mechanical Systems (NEMS). [3,4] In particular, we have studied atomistic surface effects in the nanomechanics of sub-micron resonators. We discuss simulations of the vibrational behavior of micron-scale oscillators, and we focus on unconventional dissipation mechanisms at the nanoscale.
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The work was performed in part under the auspices of the U.S.

Department of Energy by the University of California, Lawrence Livermore National Laboratory, under Contract No. W-7405-Eng-48.
[1] R.E. Rudd and J.Q. Broughton, "Coarse-Grained Molecular Dynamics and the Atomic Limit of Finite Elements," Phys. Rev. B 58, R5893 (1998).
[2] R.E. Rudd and J.Q. Broughton, "Concurrent Coupling of Length Scales in Solid State Systems," Phys. Stat. Sol. (b) 217, 251 (2000).
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9:15 AM T10.3
ATOMISTIC TRANSITION PATH SAMPLING OF EXTENDED-DEFECT KINETICS IN CRYSTALLINE SOLIDS.
Maurice de Koning, Lawrence Livermore National Laboratory, Livermore, CA; Vasily V. Bulatov, Lawrence Livermore National Laboratory, Livermore, CA.

One of the fundamental bottlenecks in the molecular dynamics (MD) simulation of complex many-body systems concerns the accessibility of relevant time scales. In order to guarantee a stable finite-difference solution of the equations of motion, the integration step is restricted to the time scale of characteristic atomic/molecular motion, typically $\sim 10^{-15}$ seconds in solid-phase systems. Unfortunately, many microscopic phenomena operate on time scales vastly larger than those associated with typical atomic motion. The according unit mechanisms underlying such processes are rare events on the time scale of atomic motion and are completely inaccessible to direct MD simulation. State-of-the-art approaches that attempt to bridge this time-scale gap include the static location of saddle-point configurations and the artificial acceleration of effective process rates. In this paper we explore the application of an alternative approach to the atomistic study of rare-event processes in complex many-body systems. The Transition Path Sampling (TPS) framework, developed by D. Chandler and coworkers, focuses entirely on a direct statistical sampling of the ensemble of reactive trajectories using a Monte Carlo random walk in the space of dynamical transition pathways. In contrast to other techniques, the TPS formalism is fully dynamic in nature and is well-suited for sampling processes involving several reactive mechanisms. Although very successful, the TPS method has so far been limited to problems in the field of chemistry. Here, we explore the applicability of the TPS formalism in the context of materials science. As a particular example we focus on the characterization of rare-event processes involved in the kinetics of extended defects in crystalline solids.

9:30 AM T10.4
TEMPERATURE ACCELERATED DYNAMICS: A POWERFUL METHOD FOR CRYSTAL-GROWTH SIMULATIONS.
F. Montalenti, J.A. Sprague, and A.F. Voter, Los Alamos National Laboratory, Los Alamos, NM.

Crystal growth is an extremely complex non-equilibrium process. Experiments alone are not sufficient to reveal all relevant microscopic processes occurring during growth. Computer simulations can help in interpreting the data, but their reliability is often questionable. One of the main problems is the extremely long time scale involved in crystal-growth experiments since a single layer is usually deposited in ~ 10 s. Kinetic Monte Carlo simulations can reach long time scales, but a list of possible diffusion mechanisms is required in advance, so that unexpected mechanisms that may occur when the system evolves toward complex configurations may be missed. If a reliable semiempirical potential is available, molecular dynamics (MD) simulations can be used to study the system evolution without needing any *a priori* information. Unfortunately only ns time scales are reachable. We show that the temperature accelerated dynamics (TAD) method [1] allows one to extend the time scale by several orders of magnitude enabling fully atomistic simulations of crystal growth where experimental deposition fluxes are matched, without needing any *a priori* information on the diffusion mechanisms.
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9:45 AM T10.5
MULTISCALE SIMULATION OF CLUSTER GROWTH AND DEPOSITION PROCESSES BY DIRECT SIMULATION MONTE CARLO METHOD. Hiroshi Mizuseki, Kenta Hongo, Yoshiyuki Kawazoe, Institute for Materials Research, Tohoku University, Sendai, JAPAN; Luc T. Wille, Department of Physics, Florida Atlantic University, Boca Raton, FL.

Magnetic films obtained by cluster deposition have attracted strong attention both as a new manufacturing technique to realize high density magnetic recording media (1) and to create systems with unique magnetic properties. Since the film's features are influenced by the cluster properties during the flight path, the relevant physical scale to be studied is as large as centimeters. Moreover, since the

behavior of segregation processes on the substrate is not negligible, the smallest physical scale is of the order of 10 nm. Although a complete analysis of this multiscale problem is extremely difficult, it provides a typical research subject in the dynamics of a complex system consisting of interacting subsystems with very different scales. In this paper a new model of cluster growth and deposition processes based on a combination of Direct Simulation Monte Carlo (DSMC) (2) and Monte Carlo (MC) methods is introduced to examine the effects of experimental conditions on film formation by an adiabatic expansion process. From the macroscopic viewpoint, we simulate the behavior of clusters and inert gas in the flight path under different experimental conditions. The internal energy of the cluster, which consists of rotational and vibrational energies, is limited by the binding energy which depends on the cluster size. This binding energy is obtained by classical molecular dynamics. Several types of size distributions of generated clusters under various conditions are obtained by the present model. The results of the simulations show that the size distribution is strongly related to the experimental conditions and in turn has a major effect on the film properties.
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10:30 AM T10.6
SIMULATION OF VAPOR DEPOSITION AND POSTANNEALING OF AMORPHOUS NANOSTRUCTURES. V.M. Burlakov, A.P. Sutton, G.A.D. Briggs, Department of Materials, University of Oxford, UNITED KINGDOM; Y. Tsukahara Technical Research Institute, Toppan Printing Co Ltd., JAPAN.

A recently developed model for the defect network structures [1] is used in Monte Carlo simulations of vapor deposition and annealing of the amorphous Si, C, SiO_x, and SiO_2-Si-SiO_2 nanostructures. The problem of the pore formation in amorphous Si and SiO_x is studied in detail. Rapidly deposited structures turned to be more disordered but less porous than those deposited quasistatically. Their subsequent annealing results in an increase of porosity, and the pore formation/ growth is driven by the decrease in the total energy of topological defects (oxygen and silicon dangling bonds). Comparing a set of SiO_x and Si structures simulated under similar conditions, including a hypothetical 3-fold coordinated Si shows that the ultimate porosity is determined by the effective atomic coordination number, bond-bending force constant, and the dangling bond energies. Simulated annealing of the SiO_2-Si-SiO_2 nanostructures reveals a tendency toward crystallization within the amorphous Si layer, in accordance with the experimentally observed formation of nanocrystals in thin Si layers confined in silica. The model is extended onto simulation of vapor deposition and posttreatment of amorphous carbon.
[1] V.M. Burlakov, G.A.D. Briggs, A.P. Sutton, and Y. Tsukahara, Phys. Rev. Lett. **86**, 3052 (2001).

10:45 AM T10.7
MODIFIED LANGEVIN THERMOSTAT FOR CONSTANT TEMPERATURE - CONSTANT PRESSURE MD SIMULATIONS OF FINITE SYSTEMS. A. Caro[1,2], J. Kohanoff[2], and M. Finnis[2]. [1]Centro Atomico Bariloche - Instituto Balseiro, Bariloche, ARGENTINA. [2]Atomistic Simulation Group, Queen's University Belfast, N. Ireland, UNITED KINGDOM.

The Parrinello-Rahman algorithm is one of the standard algorithms for constant pressure molecular dynamics (MD) simulations on periodic systems. Its strategy is based on the incorporation of the degrees of freedom of the simulation box (both size and shape) as additional dynamical variables, which evolve according to the resultant of the external and internal stresses. For finite non-periodic systems, like small clusters, a recent paper by Martonak *et al.* [1] proposes a model version of the actual experimental procedure, i.e. the use of a fluid as a conveyor of the thermal and pressure external variables. This fluid, modeled by a simple pair potential interaction with known equation of state $f(T,P,\rho)=0$ undergoes collisions with the atoms in the cluster as a consequence of the simultaneous integration of the equations of motion of the cluster and fluid particles. This algorithm clearly mimics the way pressure is applied to clusters in real experiments and does not represent an increase of the computational effort if the number of particles in the fluid is small, or if the equations of motion of the cluster atoms come form expensive ab-initio total energies. Weaknesses of this approach are: (1) the number of particles in the fluid can not be too small if they are to behave as a T-P reservoir, and (2) equilibration of the fluid is necessary prior to the physical contact with the system under study. In this work we present a new algorithm based on a different approach. Atoms in a cluster come to equilibrium through interaction with surrounding degrees of freedom that carry information about the T - P conditions. However, the description of the collision process with these degrees of freedom does not need the integration of their equations of motion. The bath

is characterized by a well-defined distribution of kinetic variables, which is the only relevant physical information for the cluster. These are equivalent to the hypothesis of Brownian motion as described by stochastic Langevin equations. In these equations, random forces represent the collisions and a constant viscous force represents the drag of the center of mass of the cluster immersed in the fluid. Both forces are given in terms of a single constant, which measures the strength of the coupling to the bath, and are related by the fluctuation-dissipation theorem. When the random forces apply in all directions, we recover the usual Langevin thermostat of isothermal MD simulations. Conversely, if the forces apply only to the exterior of a finite body, we prove that, additionally, they carry information about the pressure of the thermal reservoir. The connection between force and pressure is made through the definition of the surface elements of a cluster (or a liquid in an open container). We use the Delaunay triangulation scheme, which is the dual of the Wigner-Seitz cells of the cluster, to associate each surface atom with several surface triangles, and to determine the surface element associated with each atom, both in magnitude and direction. This, in turn, determines the strength and direction of the Langevin random force. As examples of the applications of this method, we use the algorithm to study structural transformations and melting of clusters under pressure, with the interatomic interactions described by classical potentials.
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11:00 AM T10.8
BETA RELAXATION OF ATOMIC-LEVEL STRESS IN POLYMERIC FLUIDS. Catalin Picu, Cristian Pavel, Rensselaer Polytechnic Institite, Dept of Mechanical Engineering, Troy, NY.

The mechanisms of stress production and relaxation in polymer melts are discussed at the atomic scale within the newly developed 'intrinsic framework'. In this formulation, stress is computed from atomistic models of melts in a mobile coordinate system tied to a generic bond. In the linear response regime, such as during the late relaxation period (alpha relaxation), the 'intrinsic' atomic-level stress is independent of melt deformation and the system-level deviatoric stress is induced by the preferential orientation of the intrinsic frames during melt deformation. Deviatoric stress relaxation in this regime is therefore associated with the randomization of bond orientation. It is generally believed that early relaxation regimes (beta relaxation) cannot be classified as being 'linear response'. In these conditions, the intrinsic stress is not deformation independent and other stress production/relaxation mechanisms are dominant. In this presentation we discuss the structural changes occurring during beta relaxation and the associated time scale of the process, all viewed within the intrinsic framework. Furthermore, we show that beta relaxation modes may be divided in two categories, one of which belonging to the linear response regime. The first beta mode corresponds to the relaxation of the system from far non-equilibrium conditions. This relaxation occurs very fast and is associated with a significant stress drop. Due to its small time constant however, this regime is not directly relevant for the larger scale rheology of the fluid. The second beta mode is shown to belong to the linear response regime. The corresponding mechanism is identified to be diffusion-like. Furthermore, once the mechanism is determined, the time constant of this mode may be determined from equilibrium based on the fluctuation-dissipation theorem.

11:15 AM T10.9
PLATFORM FOR DESIGNING HIGH FUNCTIONAL MATERIALS. Masao Doi, Takashi Taniguch, Yuichi Masubuchi, Nagoya Univ, Dept of Computational Science and Engineering, Nagoya, JAPAN; Toshihiro Kawakatsu, Tohoku Univ, Dept of Physics, Sendai, JAPAN; Jun-ichi Takimoto, Yamagata Univ, Dept of Polymer Science and Engineering, Yonezawa, JAPAN; Takeshi Aoyagi, Hiroo Fukunaga, Ryuichi Hasegawa, Takashi Honda, Naoki Kobayashi, Hiroya Kodama, Akiyoshi Kuroda, Hiroshi Morita, Masahiro Nishimoto, Masahiro Noda, Jiunn-Ren Roan, Fumihiro Sakano, Makoto Sasaki, Fumio Sawa, Tatsuya Shoji, Hiroyasu Tasaki, Shinzi Urashita, Tatsuya Yamaue, Katsuyuki Yokomizo, Japan Chemical Innovation Institute, Research and Education Center, Nagoya Univ, Nagoya, JAPAN.

We have developed a computer-aided engineering system named as OCTA for intervening regions between microscopic and macroscopic regions for especially polymer system, which mainly consists of four sets of simulators and a graphical user interface program. (1) a molecular dynamics simulator having flexible architecture for any coarse-grained model, (2) a polymer dynamics simulator for predicting rheological properties with the slip-link model, (3) an interfacial dynamics simulator for polymer systems with any topology and polydispersity based on the dynamic mean-field theory, (4) a multiphase dynamics simulator which can deal with phase ordering dynamics of multi-component polymeric system under shear flow and/or external electric fields and deformation dynamics of polymer solid system. These programs run on a common graphical user interface, which has facilities for manipulating any data written in a

certain text format (called UDF; User Definable Format). The GUI program controls any simulation programs and can analyze simulation results by using the Python language. New programs can be easily incorporated into the OCTA system by using the UDF text format. This work is supported by the national project, which has been entrusted to the Japan Chemical Innovation Institute (JCII) by the New Energy and Industrial Technology Development Organization (NEDO) under METI's Program for the Scientific Technology Development for Industries that Creates New Industries.

11:30 AM *T10.10
GOING THROUGH ROUGH TIMES: FROM NON-EQUILIBRIUM SURFACE GROWTH TO ALGORITHMIC SCALABILITY.
G. Korniss, RPI, Troy, NY; M.A. Novotny, Missisipi State Univ., MS; P.A. Rikvold, Florida State Univ., FL; Z. Toroczkai, LANL, NM.

Modeling and simulation of the evolution of natural and artificial complex systems are of fundamental importance in both science and engineering. In a large class of systems the underlying dynamic is inherently asynchronous. Examples of such systems include magnetization dynamics in condensed matter, cellular communication networks, and the spread of epidemics. Constructing a parallel scheme for simulating the time evolution of these systems, in which the local changes in the configuration are asynchronous, leads to at least two difficult questions.
First, to put it simply, "How to design parallel algorithms for non-parallel dynamics?". Second, if there is a faithful parallel algorithm for the problem, "Is it scalable?". We describe novel parallel Monte Carlo algorithms for asynchronous systems with both the computation and the measurement part being asymptotically scalable. In particular, we discuss the intimate connection [1] between algorithmic scalability and non-equilibrium surface growth phenomena, which has led us to the engineering and fine-tuning of fully scalable massively parallel schemes.
Supported by NSF, NERSC-DOE, CSIT-FSU, MSU, and RPI.
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