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FINAL REPORT
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This is a brief report on my DOE contract for the period of 8/1/97 until I left to move to Princeton University in 8/99. The report is broken down according to project areas.

Grain Boundary Migration

(*i*) Our two-dimensional simulations have clearly demonstrated that grain boundary velocity is indeed proportional to grain boundary curvature (driving force). This was shown to be valid for half-loops in two-dimensions of widths larger than approximately 20 interatomic spacings. Below this width, some deviations from this predicted proportionality are observed such that the grain boundary velocity is higher than predicted. We attribute this observation to elastic interactions between different segments of the grain boundary.

(*ii*) We have performed simulations for approximately 15 different boundary orientations as a function of temperature and extracted the boundary mobilities. In all cases, the data is well fit by an Arrhenius relationship, in agreement with widely accepted predictions. We extracted both the activation energy for grain boundary migration, as well as the pre-exponential term.

(*iii*) We have examined the mechanisms of grain boundary migration in these two-dimensional calculations. In short, we observe numerous, high frequency single atom hops back and forth across the boundary, followed by much less frequent, irreversible "hops" by larger groups of atoms 3-12. It is these larger group hops that actually contribute to grain boundary migration, rather than the single atom hops that are nearly reversible (they lead to small structural rearrangements that trigger the large group hops). We have decided to put this activity aside until three-dimensional simulations results are available to insure that the additional degree of freedom does not qualitatively change the migration mechanism.

(*iv*) Our quantitative measurements of the grain boundary mobility indicate that the activation energy for boundary migration is substantially lower than found in experiments (after adjustment for the dimensionality and the interatomic potential). Similar results were found by Schönfelder, et al. [Schönfelder] based on simulations using strain energy driven boundary migration. Gottstein, Shvindlerman and I attribute the disagreement to the presence of solute, that slow boundary migration, even with the purest samples used in the experiments. On the other hand, the variation of the activation energy and pre-exponential factor for the grain boundary mobility with misorientation observed in experiment is accurately reproduced by the simulations.

(*v*) We examined the variation of grain boundary mobility as a function of misorientation over a substantial misorientation range. Within this range, two distinct cusps were observed in the activation energy versus misorientation, in excellent agreement with the experiments. The logarithm of the pre-exponential factor varies with misorientation in a similar manner. This suggests that the true variation of the mobility with temperature from boundary-to-boundary (within a given structural class) as suggested by the variation of the activation energy with misorientation is compensated by the variation of the pre-exponential factor. This gives rise to the so-called "compensation effect", which suggests that structurally similar boundaries have identical mobilities at a well-defined compensation temperature. This temperature is approximately the melting point for some boundary structures while noticeably lower for others. A detailed comparison of our results on the simulated misorientation dependence of the mobility with experiment is featured in a paper submitted to Acta Materialia with our experimental collaborators.

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Growth of Multilayer Films

We have completed our study of the growth and stability of multilayer films where material transport is limited to surfaces and interfaces. The post-growth stability analysis was very similar to work we performed earlier on laminated composites and hence was attacked first. We found that misfit between the layers in the film can destabilize a multilayer structure in cases where the thinner layer is elastically stiffer than the thicker layer. The rate at which these instabilities develop increase with increasing misfit and decreasing interfacial energy. Even when there is no misfit between layers, the misfit between the multilayer film and substrate can destabilize the interfaces. This type of instability occurs whether the thinner layers are stiffer or more compliant than the thicker ones. Under certain conditions, adjacent interfaces can be modulated in-phase (i.e., in cross-section, a layer looks a snak) or out-of-phase (i.e., in cross section, a layer looks like an accordion). By appropriate choice of the elastic moduli mismatch between layers and relative layer thicknesses, the presence of an interlayer misfit can suppress the instability caused by the substrate misfit. We obtained stability diagrams that can be used to design stable, multilayer films using all of the degrees of freedom commonly available in multilayer film deposition..

We also had considerable success in analytically extracting the interface morphology evolution during the growth of the periodic, strained multilayer films. As the physical (surface diffusivity, surface energy, and misfit of each layer) and growth (deposition rate and thicknesses of the individual layers) parameters are varied, two distinct regimes of behavior were predicted. The unstable regime, where the amplitude of all perturbations on the interfaces increase with increasing number of layers, is further divided into cases where perturbations to adjacent interfaces are (1) in-phase or (2) out-of-phase or exhibit periods larger than two layers. The out-of-phase perturbations produce lateral composition modulation. While misfitting single layer films are always unstable with respect to perturbations from a flat surface of wavelengths greater than a critical size, we have identified ranges of growth and material parameters over which strained multilayer films have flat interfaces/surface (i.e., are completely stable). This surprising result provides guidelines for the <u>growth</u> of practical multilayer films. The present analysis was performed within the framework of linear stability theory. The elastic fields on the evolving surface had contributions from all of the interfaces buried so far. We developed an analytical recurrence relation that relates the stresses on each layer to the roughness of the few previous interfaces, which reduces to a Fibonacci type series. The resultant stability was determined numerically as a solution to an eigenvalue problem, which showed that surface perturbations grow or shrink exponentially as a given layer is grown, but as a power law (power determined from the eigenvalue problem) over many layers. Given the wide range of physical and growth parameters associated with a multilayer system on a substrate, the present results are quite complex. Part of this work was performed in collaboration with Jerry Tersoff of IBM.

Sub-Projects

Several minor projects related to the above were performed as part of the DOE BES sponsored research. These include studies of:

- Defect interactions on surfaces (with L. Shilkrot)
- The effects of interfaces on elastic boundary conditions (with L. Shilkrot)
- Shapes of hollow dislocation cores in complex materials (with N. Sridhar, JP Hirth and JW Cahn)
- Effect of externally imposed boundary mobility inhomogeneities on microstructural evolution (with N. Zacharopoulos and EA Holm)
- Twinning in ferroelectrics (with N. Sridhar and JM Rickman)
- Dislocation motion with diffusing impurities (with Y. Wang, RA LeSar and JM Rickman)
- Stress development in the early stages of film growth (with RC Cammarata)
- Dislocation cell structures for recrystallizaiton (with M Miodownick, EA Holm and P Smereka)

List of Publications associated with this BES contract from 1997-2000

1. N.Sridhar, J.M.Rickman, and D. J.Srolovitz, "Microstructural Stability Of Stressed Lamellar and Fiber Composites", Acta Materialia **45** [7], 2715-2733 (1997).

2. N.Sridhar, J.M.Rickman, and D. J.Srolovitz, "Multilayer Film Stability", Journal of Applied Physics **82** [10], 4852-4859 (1997).

3. L. Shilkrot and D. J. Srolovitz, "Adatom-Step Interactions: Atomistic Simulations And Elastic Models", Physical Review B **55** [7], 4737-4744 (1997).

4. L. E. Shilkrot and D. J. Srolovitz, "Anisotropic Elastic Analysis And Atomistic Simulation Of Adatom-Adatom Interactions On Solid Surfaces", Journal of the Mechanics and Physics of Solids **45** [11/12], 1861-1873 (1997).

5. N. Sridhar, J.M. Rickman and D. J. Srolovitz, "Microstructural Stability Of Stressed Lamellar Eutectics", Thermodynamics and Kinetics of Phase Transformations (Mater. Res. Soc., Pittsburgh, PA, 1997), pp. 445-450.

6. M. Upmanyu, R. W. Smith and D. J. Srolovitz, "Atomistic Simulation of Curvature Driven Grain Boundary Migration", Interface Science **6**, 41-58 (1998).

7. L. E. Shilkrot and D. J. Srolovitz, "Elastic Analysis of Finite Stiffness Bimaterial Interfaces: Application to Dislocation-Interface Interactions", Acta Materialia **46** [9], 3063-3075 (1998).

8. E. A. Holm, N. Zacharopoulos, and D. J. Srolovitz, "Nonuniform and Directional Grain Growth Caused by Grain Boundary Mobility Variations", Acta Materialia **46** [3], 953-964 (1998).

9. D. J. Srolovitz, N. Sridhar, J. P. Hirth and J. W. Cahn, "Shape of Hollow Dislocation Cores: Anisotropic Surface Energy and Elastic Effects", Scripta Materialia **39** [4-5], 379-387 (1998).

10. M. Upmanyu, D. J. Srolovitz, G. Gottstein and L. Shvindlerman, "Vacancy Generation During Grain Boundary Migration," Interface Science **6** [4], 287-298 (1998).

11. N. Sridhar, J. M. Rickman and D. J. Srolovitz, "Defect Model of Twinning in Ferroelectrics," Acta Materialia **47** [4], 1325-1336 (1999).

12. M. Upmanyu, D. J. Srolovitz, L. S. Shvindlerman, and G. Gottstein, "Misorientation Dependence of Intrinsic Grain Boundary Mobility: Simulation and Experiment", Acta Materialia **47** [14], 3901-3914 (1999).

13. M. Upmanyu, D. J. Srolovitz, L. S. Shvindlerman, and G. Gottstein, "Triple Junction Mobility: A Molecular Dynamics Study", Interface Science **7** [3-4], 307-319 (1999).

14. Y. Wang, D. J. Srolovitz, J. M. Rickman and R. LeSar, "Dislocation Motion in the Presence of Diffusing Solute: A Computer Simulation Study"", Acta Materialia, **48** [9], 2163 (2000).

15. L. E. Shilkrot, D. J. Srolovitz and J. Tersoff, "Dynamically Stable Growth of Strained-Layer Superlattices", Appl. Phys. Lett. **77** [2], 304-306 (2000).

16. R. C. Cammarata, T. M. Trimble and D. J. Srolovitz, "Surface Stress Model for Intrinsic Stress in Thin Films", Journal of Materials Research **15** [11], 2468-2474 (2000).

17. L. E. Shilkrot, D. J. Srolovitz and J. Tersoff, "Morphology Evolution During the Growth of Strained Layer Superlattices", Physical Review B **62** [12], 8397-8409, (2000).

18. M. A. Miodownik, P. Smereka, D. J. Srolovitz and E. A. Holm, "Scaling of Dislocation Cell Structures: Diffusion in Orientation Space", Proc. Roy. Soc. (Lond) in press.

19. D. J. Srolovitz, M. I. Mendelev, M. Upmanyu, G. Gottstein, D. A. Molodov and L. S. Shvindlerman, "Grain Boundary Mobility in Metallic Alloys: Recent Simulations and Experiments ", in *Proceedings of the 21th Risø International Symposium on Materials Science: Recrystallization - Fundamental aspects and relations to deformation microstructure*, ed. N. Hanser, X. Huang, D. Juul Jensen, E. M. Lauridsen, T. Leffers, W. Pantleon, T. J. Sabin and J. A. Wert (Risø National Laboratory, Roskilde, Denmark, 2000), pgs. 157-178.

20. David J. Srolovitz and Monnesh Upmanyu, "Anisotropic Grain Boundary Properties for Modeling Grain Growth Phenomena," in *Grain Boundary Engineering in Ceramics* (American Ceramic Society, Westerville, OH, 2000), pp 89-96; Ceramics Transactions **118**, 89-96 (2000).