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August 4, 2004

Dr. Timothy Fitzsimmons
Materials Sciences and Engineering Division
Office of Basic Energy Sciences
SC-13/Germantown Building, Room F-418
U.S. Department of Energy
1000 Independence Avenue, SW
Washington, D.C. 20585-1290

Dear Tim,

Enclosed please find the final report for the contract DE-FG02-95ER14566, and a copy of the publications that have resulted from research supported by this contract.

I am very grateful to your program, and to the Department of Energy, for the research support that I have received over the years, dating back to the days of Oscar Manley, followed by Bob Price, and now yourself. I do hope that, in some small measure, I have contributed to the science mission of the U.S. DOE.

As I had mentioned to you, I will be moving to McGill University where I intend to continue research in Materials Science. Needless to say, do not hesitate to call on me if you think I could be of any help. You can reach me at vinals@physics.mcgill.ca

Best regards,

A handwritten signature in black ink, appearing to read "Jorge Viñals".

Jorge Viñals
Program Director and
Associate Professor

**Final Report of Research Progress
for the period
August 2001 - August 2004
Contract DE-FG02-95ER14566**

Theoretical and computational studies of pattern formation

**Jorge Viñals
School of Computational Science and Information Technology
Florida State University
Tallahassee, FL 32306-4120**

This is the final report in connection with the project "Theoretical and computational studies of pattern formation". The objectives of our research are to improve the understanding of the mechanisms underlying the formation of complex spatio temporal patterns in systems that are driven outside of thermodynamic equilibrium, to characterize the macroscopic properties of such states, and to apply the methodology to situations of interest in fluid mechanics, materials processing and engineering.

Topological defect motion in mesophases.

Mesophases are phases which in terms of symmetry are intermediate between disordered liquids and fully ordered crystalline solids. They are characterized by spontaneously broken translational or rotational symmetries along one or more directions, but do not exhibit the full order characteristic of three dimensional crystalline solids. Mesophases are ubiquitous in soft-matter systems, and are being currently investigated from a number of different angles. With the transition to new technologies in fields such as micro electronics or biomedicine, microstructural control of mesophases has become necessary, and increased emphasis has been placed on research on microstructure and self-assembly at the mesoscale. We mention, for example, the use of block copolymers as nanolithographic templates, photonic band gap materials, light emitting diodes or dielectric mirrors, or as artificial cell membranes. The central challenge for widespread applications is how achieve long ranged domain order in a mesophase.

Therefore we have focused on systems that do not exhibit long range macroscopic order, and hence out of equilibrium. Time scales necessary to achieve long ranged order are controlled by the motion of topological defects. Topics of research during the period include the motion of grain boundaries in modulated phases, interactions between grain boundaries and other defects (dislocations and disclinations) that govern long time structure coarsening, the discovery that the so called non-adiabatic corrections to defect motion can lead to pinning and glassy behavior in mesophases, and a generalization of these studies to hexagonal (columnar) phases.

Grain boundary motion in modulated phases has been investigated within the amplitude (or envelope) equation formalism. If the boundary is curved, we have shown the existence of a nonlinear mode that leads to net boundary translation, with a speed that is proportional to the local mean curvature squared, and increases as the onset of the phase (bifurcation or phase transition point) is approached. Coarsening of a macroscopically disordered structure has also been investigated. Grain boundary motion is dominant in a distorted background set by other topological defects (disclinations). Their mutual interaction leads to a coarsening rate of the structure that scales as time to the $1/3$ power.

We have also shown that the amplitude or envelope of a topological defect field (which changes over scales that are much larger than the underlying periodicity of the modulated phase) can couple to that periodicity in a non-analytic way (through an essential singularity in the distance away from onset of the phase). As a consequence, defects can become pinned as the driving force for motion decreases over time, leading to effective coarsening exponents and eventually to a glassy phase. The effect of thermal fluctuations on this phase has also been investigated, and the appropriate length scales for pinning investigated.

The research just described concerns a supercritical bifurcation to a phase of smectic symmetry (broken translational symmetry along only one direction, and hence the term modulated phase). We have next investigated a hexagonal or columnar phase (broken translational symmetry along two directions) which appears subcritically. We have shown that pinning effects are always present in the subcritical case (instead of the essential singularity in the supercritical counterpart), and that

the magnitude of the pinning depends sharply on the misorientation between neighboring grains. In fact, we have provided an asymptotic (analytic) derivation of the classical Peierls stress for the case of a mesophase close to onset. In contrast, the calculation of the Peierls stress for defect motion in a crystalline solid involves a heuristic mixture of atomistic considerations and macroscopic elasticity theory. Our calculation is asymptotically correct within the model of the mesophase employed.

We have also completed the analysis of domain coarsening for columnar phases. Because they appear subcritically and pinning effects do not vanish near onset, it is expected that no asymptotic self similarity and universal coarsening exponents will exist for these phases. We have obtained analytically the equations governing grain boundary motion, and explicitly derived expressions for the (now anisotropic) grain boundary energies and mobilities as a function of misorientation. As expected, an effective coarsening exponent is obtained, function of the degree of subcriticality. Two surprising issues have emerged. First, the effective coarsening exponent is seen to increase with a decrease in subcriticality, but our results are not consistent with an extrapolation to the value of $1/2$, the expected exponent for normal grain growth in polycrystalline materials. Second, we find a self similar distribution of grain orientations, in contradiction with current simple models of coarsening in anisotropic systems. Whether the disagreement can be attributed to a failure of the theory, or lack of accuracy in our calculations is still an open question. A manuscript detailing these results is currently in preparation.

Faraday waves.

We have considered an extension of current nonlinear theory of parametrically driven surface waves (Faraday waves) to accommodate the effects of mean flows. Close to onset, Faraday waves bifurcate to standing waves of different symmetries (including patterns of quasi-crystalline symmetry, as described earlier in this contract), but the region of existence of standing waves is expected to be small. Secondary instabilities lead to pattern modulation, and very soon to spatio temporal chaos. Our results show that mean flows lead to weak destabilization of the base state against Eckhaus and Transverse Amplitude Modulation instabilities, and that it introduces a new longitudinal oscillatory instability which is absent without the coupling to mean flows. We have introduced a phenomenological model to describe the coupling, which we hope will allow a more systematic investigation of the spatio temporal dynamics of this complex system.

Microstructure evolution in defected crystalline solids.

We have developed a unified framework for the calculation of defect energies for those defects that can be represented as a superposition of isolated dislocations and, as examples, obtained both self and interaction energies of combinations of grain boundaries and cracks. We introduce the dislocation density tensor as the basic mesoscopic quantity to describe the defect structure, and use the functional representation of the elastic energy in terms of the dislocation tensor to compute interaction energies between sets of extended defects. We can recover in special limits several well known quantities such as the energy of a low-angle tilt boundary, but have also derived new results concerning boundary/boundary and crack/boundary interaction energies.

We have also addressed the issue of abnormal grain growth in polycrystalline materials. We show that abnormal grain growth follows from grain boundary pinning in a model that considers lower and upper thresholds to the driving force for boundary migration. Therefore anisotropy of grain boundary energy or mobility is not required for abnormal grain growth.

Publications.

We list below those publications that have resulted from research supported by this contract, and that acknowledge this support. Related to the research funded by this contract, I was invited to participate in the program "Complex and Macromolecular Fluids" in the Spring of 2002 as a member of the Institute of Theoretical Physics of the University of California, Santa Barbara, and again in the Fall of 2003 for the program "Pattern formation in Physics and Biology".

- D. Boyer and J. Viñals. "Grain boundary motion in layered phases". *Phys. Rev. E* **63**, 061704 (2001).
- F. Drolet and J. Viñals. "Adiabatic elimination and reduced probability distribution functions in spatially extended systems with a fluctuating control parameter". *Phys. Rev. E* **64**, 026120 (2001).
- D. Boyer and J. Viñals. "Domain coarsening of stripe patterns close to onset". *Phys. Rev. E* **64**, 050101(R) (2001).
- D. Boyer and J. Viñals. "Grain boundary pinning and glassy dynamics in stripe phases". *Phys. Rev. E* **65**, 046119 (2002).
- W.W. Mullins and J. Viñals. "Linear bubble model of abnormal grain growth". *Acta Materialia* **50**, 2945 (2002).
- D. Boyer and J. Viñals. "Weakly nonlinear theory of grain boundary motion in patterns with crystalline symmetry". *Phys. Rev. Lett.* **89**, 055501 (2002).
- J.M. Vega, S. Rüdiger, and J. Viñals. "A phenomenological model of weakly damped Faraday waves and the associated mean flow". *Phys. Rev. E*, in press.
- J. Rickman, J. Viñals, and R. LeSar. "Unified framework for dislocation-based defect energies". Submitted to *Phil. Mag. A*.
- S. Rüdiger and J.M. Vega. "Mean flow effects in model equations for Faraday waves". To appear in "Dynamics and Bifurcation of Patterns in Dissipative Systems", G. Dangelmayr and I. Oprea Eds., World Scientific Series on Nonlinear Science, Series B. World Scientific 2004.
- F. Drolet, D. Boyer and J. Viñals. "Domain coarsening at a weakly subcritical bifurcations", in preparation.

Department of Energy
Washington, DC 20585

Michele,
for your files.
Jorge

September 27, 2004

Professor Jorge Viñals
McGill Institute for Advance Materials
and Department of Physics
McGill University
215 Rutherford Building
3600 University Street
Montreal, QC H3A 2T8
Canada

Dear Professor Viñals:

We have received your final report for Grant No. DE-FG02-95ER14566 and have authorized our Chicago Operations Office to proceed with the close-out of the grant. We hope that your latest results will be prepared for publication in the open literature since the final report is not given broad distribution.

We have enjoyed working with you and the Florida State University in support of this project. We trust that you will continue following your research interests.

Sincerely,

Timothy J. Fitzsimmons
Division of Materials Sciences
and Engineering
Office of Basic Energy Sciences

cc:
Chicago Operations Office