This project studied the dynamic response of metallic materials due to twinning and microstructural evolution. We have studied -- using molecular dynamics -- the nucleation of twinning interfaces, the transition to motion of a static twin interface, the steady kinetics of twinning interfaces. Finally, we have formulated a new continuum multiscale phase-field model that enables us to incorporate prescribed nucleation behavior and complex kinetics.
Final Report for 56137EG Dynamics of Structural Phase Transformations Using Molecular Dynamics

ABSTRACT

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Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

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<td>12/08/2013</td>
<td>8.00 Kaushik Dayal, Chang-Tsan Lu. Linear instability signals the initiation of motion of a twin plane under load, Philosophical Magazine Letters, (04 2011): 0. doi: 10.1080/09500839.2011.552448</td>
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Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

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TOTAL:
Number of Papers published in non peer-reviewed journals:

(c) Presentations
Invited seminars:

-- Eshelby Award Seminar in the Mechanical Engineering Department, University of Houston, October 2013.

-- Workshop on Mathematics and Mechanics in the Search for New Materials at the Banff International Research Station, July 2013.

-- Summer School on Topics in Nonlinear PDEs and Calculus of Variations and Applications in Materials Science at the Center for Nonlinear Analysis, June 2013.

-- Fluids and Materials Seminar in the School of Mathematics, University of Bristol, March 2013.


-- Center for Nonlinear Mechanics Seminar, University of Bath, November 2012.


-- Mechanical Engineering Department Seminar, Florida State University, September 2012.

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-- Center for Nonlinear Studies Seminar, Los Alamos National Laboratory, March 2012.


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-- Materials Interest Group Seminar in the Department of Mechanical Science and Engineering, University of Illinois, November 2011.

-- Mechanical Engineering and Applied Mechanics Department Seminar, University of Pennsylvania, October 2011.

-- Mechanical Engineering Department Seminar, Indian Institute of Science, August 2011.


-- Mechanical Engineering Department Seminar, University of Houston, March 2011.

-- Civil Engineering Department Seminar, Johns Hopkins University, March 2011.

-- Computational and Information Sciences Directorate Seminar, U.S. Army Research Laboratory, March 2011.

-- Mechanical Engineering Department Seminar, Ohio State University, March 2011.

-- Mechanical Engineering Department Seminar, University of Michigan, November 2010.

-- Center for Nonlinear Studies Seminar, Los Alamos National Laboratory, September 2010.

-- Laboratoire de Physique et Mecanique des Materiaux Seminar, University of Metz, June 2010.

Contributed conference proceedings:


-- 2010 SIAM Conference on Mathematical Aspects of Materials Science. Nonequilibrium Molecular Dynamics of Complex Structures using Objective Structures

-- Society of Engineering Science 2011. Thermoelastic Interpretation of Phase Boundary Kinetics in a 1D Chain of Atoms


-- 2013 SIAM Conference on Mathematical Aspects of Materials Science. Unexpected Thermodynamic Properties of Some Exact Far-from-equilibrium Solutions in Molecular Dynamics presented by collaborator RD James

-- 2013 ASME IMECE. Multiscale Modeling of Defect Motion; and Symmetry-Adapted Phophon Analysis of Nanotubes.

**Number of Presentations:** 0.00

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(d) Manuscripts

Received Paper

01/06/2011 1.00 Chang-Tsan Lu, Kaushik Dayal. Linear Instability Signals the Initiation of Motion of a Twin Plane Under Load, Philosophical Magazine Letters (12 2010)

TOTAL: 1

Number of Manuscripts:

Books

Received Paper

TOTAL:

Patents Submitted

Patents Awarded

Awards

NSF CAREER
ARO Young Investigator
AFOSR Young Investigator
ASCE Leonardo Da Vinci Young Investigator Award
Eshelby Mechanics Award for Young Faculty
Graduate Students

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Student Metrics

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The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields: ..... 0.00
The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields: ..... 0.00
Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale): ..... 0.00
Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering: ..... 0.00
The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense: ..... 0.00
The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields: ..... 0.00

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Names of personnel receiving PHDs

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Names of other research staff

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FTE Equivalent:  
Total Number:

Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

Technology Transfer

See Attachment
Grant Title: Dynamics of Structural Phase Transformations Using Molecular Dynamics

Grant Number: 56137-EG, Jul 1, 2010 to Jun 30, 2013

Solid Mechanics Program (Dr. Ralph Anthenien), Army Research Office

PI: Kaushik Dayal, Carnegie Mellon University

Abstract

This is the final report for Grant Number: P-56137-EG from the Army Research Office.

Contents

1 Statement of the problem studied

2 Summary of the most important results
   2.1 Nucleation of twin interfaces using atomistics
   2.2 Linear stability to predict the transition from stationary to moving interface
   2.3 A new method to simulate non-equilibrium molecular dynamics
   2.4 Thermodynamic analysis of steadily moving interfaces in molecular dynamics
   2.5 A continuum method that can incorporate atomistically-derived prescribed nucleation and complex kinetics
       2.5.1 Phase-Field Model Formulation
       2.5.2 Preliminary Characterizations of the Dynamic Phase-Field Model

3 University Seminars

4 Interactions with ARL

5 Awards

1Email: kaushik@cmu.edu, Phone: 412-268-2949
1 Statement of the problem studied

This research program aims to understand the dynamics of structural phase transformations in metals using molecular dynamics. This insights will enable multiscale modeling of dynamic deformation in structural and active materials with potential future application to deformation, reliability, and kinetic energy absorption in these materials.

Structural phase transformations in metals occur due to abrupt changes in crystal structure due to applied load or temperature or other changes in environmental conditions. The transformation proceeds by the motion of an interface that separates the different phases. Continuum modeling at the mesoscale requires information about the interface propagation dynamics from the atomic scale. The research effort is to understand how to obtain this information from the atomic scale and provide it to continuum models. Our focus is along three key thrusts: nucleation of interfaces, transition from stationary to moving interface, and steady motion of an interface. Some of our findings challenge the consensus view of these phenomena from the last 2 decades. In addition, we have formulated a continuum model that allows us to readily incorporate the atomistic insights into a multiscale continuum model.

2 Summary of the most important results

The research effort has focused on 5 main thrusts:

1. Nucleation of twin interfaces using atomistics;
2. Linear stability to predict the transition from stationary to moving interface for different crystallographic orientations using atomistics;
3. A new method to simulate non-equilibrium molecular dynamics by identifying a time-dependent invariant manifold;
4. Thermodynamic analysis of steadily moving interfaces in molecular dynamics;
5. A continuum method that can incorporate atomistically-derived prescribed nucleation and complex kinetics

2.1 Nucleation of twin interfaces using atomistics

The key finding is that we can predict the nucleation of twin interfaces using linear stability techniques. This is well-known in dislocation nucleation following the work of Suresh, Miller, Rodney and their co-workers. Our studies show that similar ideas can be fruitfully applied to the nucleation of twinning interfaces. This work is described in detail in the attached thesis by Chang-Tsan Lu (Chapter 5) and an archival publication is being prepared.

2.2 Linear stability to predict the transition from stationary to moving interface

We have applied linear stability analysis to understand the transition of a static interface to a dynamic state. A key finding is that rather than thermodynamic driving force being the controlling factor, the transition to motion is controlled by linear stability. That is, the kinetic curve as a function of driving
force alone will be unable to describe the stick-slip transition of a twinning interface. This work is described in detail in the attached thesis by Chang-Tsan Lu (Chapters 2 and 3) and the attached archival publication *Linear instability signals the initiation of motion of a twin plane under load*. A further archival publication is being prepared.

2.3 A new method to simulate non-equilibrium molecular dynamics

We have formulated a new and accurate method to simulate an extremely large class of non-equilibrium deformations with molecular dynamics. The key insight that enables this is the identification of a time-dependent invariant manifold. The manifold is universal in the sense that it is independent of the behavior of the material, and is hence applicable to deforming solids as well as to flowing liquids and gases. The theory underlying the method is described in the attached archival publication *Design of viscometers corresponding to a universal molecular simulation method*. Applications are described in 3 attached archival publications: *Symmetry-adapted phonon analysis of nanotubes; Tension and twist of chiral nanotubes: torsional buckling, mechanical response and indicators of failure; and Anomalous phonon behavior of carbon nanotubes: First-order influence of external load*. The method is essential to enable the accurate computation (without spurious artifacts that are not physical but to methodology) of the non-equilibrium behavior of the systems of interest in this grant.

2.4 Thermodynamic analysis of steadily moving interfaces in molecular dynamics

The current understanding in the field of interface kinetics implicitly neglects the role of temperature and thermodynamics. This has led to “paradoxes” such as non-unique kinetics of twinning interfaces. Our observation that thermodynamics resolves many of the paradoxes in a simple and physically useful way has been described in the attached thesis by Chang-Tsan Lu (Chapter 4) and an archival publication is in preparation. The key finding is that the kinetics are not just a function of driving force, but rather a function of temperature, driving force, and net heat flux.

2.5 A continuum method that can incorporate atomistically-derived prescribed nucleation and complex kinetics

As this research thrust has not yet been published either in a student thesis or in an archival publication, we describe it here in detail.

2.5.1 Phase-Field Model Formulation

The energetic aspects of our phase-field formulation follow directly the standard approach. The essential difference in our formulation is in the kinetics: the key idea is to combine the advantages of phase-field modeling (diffuse interfaces) with ideas of level-set methods for front motion (Hamilton-Jacobi flows). The main tool that we use to achieve the formulation is to use a geometric balance law as we describe below.
Our current formulation is for microstructural interfaces such as phase boundaries separating various phases. Namely, phase transformations are tracked by a tensor-valued phase field (for instance the stress-free strain in each phase).

The free energy of the material can be written in the linearized kinematic setting:

\[
E[u, \phi] = \int_{\Omega} W(\epsilon(u), \phi) \, d\Omega + \kappa |\nabla \phi|^2 \, d\Omega
\] (2.1)

In the expression above, \( W \) is the elastic energy density, and \( \kappa \) is the regularization parameter that smears out the interface. The fields \( u \) and \( \epsilon \) are respectively the displacement and linearized strain fields.

Standard phase-field evolution methods typically use a gradient flow for the order parameter evolution, i.e.

\[
\mu \dot{\phi} = \frac{\delta E}{\delta \phi}
\] (2.2)

The evolution of the mechanical fields \( F \) either occurs by assuming mechanical static equilibrium at each increment, or by an elastodynamic calculation:

\[
\rho \ddot{u} = \text{div} \frac{\partial W(\epsilon)}{\partial \epsilon(u)}
\] (2.3)

Our formulation makes an important departure in the kinetics of \( \phi \). Our starting point is to note that \( \nabla \phi \) provides, roughly, a measure of the number or “strength” of the interfaces in the \( \phi \) field per unit length. As an example, consider a field \( \phi(x) \) that goes from \( 0 \) to \( 1 \) in a small region near \( x = 0 \). The field \( \nabla \phi \) will have a (non-singular) peak localized around \( x = 0 \) showing that there is a higher “interface density” near the origin. This matches with intuition: there are no interfaces away from the origin, and there is an interface that is located near the origin.

In general, given a field \( \phi(x) \) with localized transitions between constant values, we can readily locate the interfaces in this field using \( \nabla \phi \). Further, if we pick any curve and integrate \( \nabla \phi \) along this curve, the value that we obtain provides a measure of the net number of interfaces that we have traversed, assuming that all interfaces have the same “strength”. If the interfaces have different strengths, we obtain a measure of the net interface strength that we have traversed. This physical picture provides the intuition behind what follows, but it also expresses a simple fact that if we have a single-valued field \( \phi \), then integrating the gradient is simply the difference between \( \phi \) at either end of the curve.

To make this intuitive picture more precise, we appeal to the differential geometric notion of gradients of fields being so-called 1-forms. That is, they are objects that are integrated along curves. Higher-dimensional analogies of this are commonplace in elasticity: e.g., the divergence of a field is a 3-form and is integrated over volumes, as is used in connecting the divergence of the stress to the boundary tractions; the curl of a field is a 2-form and is integrated over surfaces, as is used in the proof of single-valuedness of a deformation field corresponding to \( \text{curl} \, F = 0 \) or in the net vorticity formulas in fluid mechanics.

Given this notion of the interface density field \( \nabla \phi \) a 1-form, we then formulate a balance law (see Fig. 1). Let the interfaces be convected by a velocity field \( v^\phi \); note that this velocity is distinct from the material velocity \( \dot{u} \). Now consider a curve \( C(t) \) in space. This curve “threads” or passes through some number of interfaces. Further, interfaces are entering or exiting at one end and leaving at the other end of the curve due to \( v^\phi \). The balance is simply that the net change in the number of interfaces that are threaded by \( C(t) \)
is a balance between interfaces entering, interfaces exiting, and interfaces being created/destroyed by a source/sink. Making this balance precise provides us with the desired evolution law.

Defining the interface density $\alpha := \nabla \phi$, we have:

$$\left. \alpha v^\phi \right|_{C^+} = \int_{C(t)} \text{grad}(\alpha \cdot v_\alpha) \, dC = \frac{d}{dt} \int_{C(t)} \alpha \, dC - \int_{C(t)} S \, dC$$

where $S$ is the velocity gradient. Assuming small deformations for simplicity and localizing, we obtain

$$\nabla \phi \cdot v^\phi + G = \dot{\phi}$$

The appropriate boundary conditions for typical crack settings is to allow the flux of $\phi$ to be unconstrained. In addition, we append the standard elastodynamic equation $\rho \ddot{u} = \text{div} \partial W / \partial \epsilon$. The conservation form for the crack kinetics provides important advantages for robust numerical implementations such as a natural weak form.

We note two important aspects of our approach. First, the field $v^\phi$ is constitutively prescribed and can be a function of stress, strain, as well as any other relevant quantity, e.g. the work conjugate to $\phi$ (the Eshelby force). This makes it trivial to obtain complex kinetics; for instance, if the interface is pinned below a critical value of the stress or Eshelby force, we simply prescribe that $v^\phi$ is zero at all spatial points where the stress or Eshelby force is below the critical value. Similarly, other kinds of nonlinear and complex kinetics can be readily incorporated. Second, the source term in the balance law...
provides precise control on nucleation of new interfaces. For instance, we can prescribe that a source is activated only beyond some critical stress; thus a shock wave can nucleate a new interfaces in a material if the physics so dictates. In addition, the activation of the source can be completely heterogeneous and vary vastly from point ot point. That enables a powerful description of cracking in composites and polycrystalline media whereby we can prescribe different nucleation stresses near grain boundaries and in the grain interiors.

2.5.2 Preliminary Characterizations of the Dynamic Phase-Field Model

We have tested the dynamic phase-field model in the case of microstructural twinning interfaces, where $\phi$ is replaced by a tensor-valued phase-field. These preliminary characterization provide confidence in the ability of the model to (1) capture complex kinetics as prescribed, (2) capture complex nucleation as prescribed, and (3) behave appropriately in the near-sonic regime. We briefly describe here the relevant predictions of the model.

In Fig. 2, we examine 2 choices for $v^\phi$. We use linear and quadratic relations between $v^\phi$ and the local value of the thermodynamic work-conjugate to $\phi$. We perform computations with far-field applied stresses and observe the velocity of the defect produced by our model. We find that the velocity of the defect has a linear/quadratic relation respectively with the classical driving force that would be computed using the far-field stresses. We also note that in the sonic regime, near Mach number tending to 1, the kinetics of the interface is dominated by the rate of energy transfer to the moving defect that is constrained by elastodynamics. However, cracks can grow faster than the shear wave velocity. In this context, it is important to note that the kinetic relations that we present relate to the normal velocities for interfaces in the phase-field field $\phi$, i.e., the crack faces; while the velocities of the faces are constrained by the Hugoniot relations, the crack tip location and velocity are related to the intersection of the crack faces and the evolution of the intersection point. As has been studied in the literature by P. Rosakis and Tsai, even if the normal velocities of the faces are subsonic, the intersection point can nonetheless be supersonic.

![Figure 2](image)

**Figure 2**: Left: We specify a linear kinetic relation between $v^\phi$ and the work-conjugate driving force. We then apply our model and examine the computed defect velocity. The curve clearly the linearity up to $\sim 0.75$ Mach number as a function of far-field applied driving force. In the sonic regime, the kinetics is dominated by the rate of energy transfer to the defect in turn limited by elastodynamics. Right: We specify a quadratic kinetic relation between $v^\phi$ and the work-conjugate driving force at relatively low Mach number.

Fig. 3 presents further calculations with complex and spatially-dependent kinetics. In particular, we...
provide a clear demonstration that stick-slip kinetics is readily achievable by our model by the simple device of using a threshold value in the constitutive response chosen for \( v^\phi \). A contrast with a standard phase-field model (whether elastostatic or elastodynamic) is also shown where the defect velocity is always finite for a finite value of the configurational force. In addition, we show that motion of an interface as it interacts with an inclusion in which the kinetics requires much greater driving force for a given velocity as compared to the matrix. The interface is pinned at the inclusion and breaks free at a larger applied far-field load.

![Figure 3: Left: A plot of defect kinetics in our phase-field formulation. A clear indication of stick-slip kinetics is observed in our formulation and the critical value can be directly specified. Right: A 2D calculation of a twin interface interacting with an inclusion, shown in terms of the strain field. A needle-like twin nucleation is observed.](image)

Finally, we present an example characterizing the nucleation behavior of the model. We use a source term \( \dot{G} \) located at the center of the simulation domain and that becomes “active” above a non-dimensional stress of \( \sigma_0^\pm = \pm 0.03 \) with the appropriate sign chosen depending on whether the material at that point is in the high- or low-strain variant. The activity of the source is readily modeled using a step function with argument \( \sigma - \sigma_0^\pm \). We subject this model specimen to a stress-strain cycle as shown in Fig. 4. We find that nucleation occurs precisely at the prescribed value! Further, there is finite rate-independent hysteresis in the limit. The direct control of nucleation behaviors – without using noise or mechanical inhomogeneities as is typical – represents a transformative advance in the modeling of phase transformations, and PDE models of defects broadly. There is a clear separation between energetics, defect kinetics, and defect nucleation. The powerful implications of this model for cracks are similar.
3 University Seminars

The PI has given a number of invited seminars and talks at top national and international mechanics workshops and universities over the current reporting period of this grant. These include:

- Eshelby Award Seminar in the Mechanical Engineering Department, University of Houston, October 2013.
- Workshop on Mathematics and Mechanics in the Search for New Materials at the Banff International Research Station, July 2013.
- Summer School on Topics in Nonlinear PDEs and Calculus of Variations and Applications in Materials Science at the Center for Nonlinear Analysis, June 2013.
- Fluids and Materials Seminar in the School of Mathematics, University of Bristol, March 2013.
- Center for Nonlinear Mechanics Seminar, University of Bath, November 2012.
- Mechanical Engineering Department Seminar, Florida State University, September 2012.
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- Center for Nonlinear Studies Seminar, Los Alamos National Laboratory, March 2012.
- Army Research Laboratory Workshop on Complex Oxide and Multiferroic Thin Film Materials Science, Technologies, and Applications, January 2012.
- Materials Interest Group Seminar in the Department of Mechanical Science and Engineering, University of Illinois, November 2011.
- Mechanical Engineering and Applied Mechanics Department Seminar, University of Pennsylvania, October 2011.
4 Interactions with ARL

The PI has closely interacted with ARL over the past few years.

One interaction was with the functional materials effort at ARL through Dr. Melanie Cole (ARL Fellow). She organized the *Army Research Laboratory Workshop on Complex Oxide and Multiferroic Thin Film Materials Science, Technologies, and Applications* in January 2012 and I was an invited speaker at that event. She is organizing the *Symposium on Multiferroic Materials and Multilayer Ferroic Heterostructures* in the Electronic Materials and Applications Conference in January 2014, and she has asked me to be an invited speaker at this event.

Another interaction has been with the CIS directorate. I have visited the materials group there and interacted with Drs. Jaroslaw Knap, John Clayton, Misha Grinfeld, and Peter Chung (now not at ARL). I presented at the Computational and Information Sciences Directorate Seminar, U.S. Army Research Laboratory in March 2011. Based on the common interests between me and Dr. Jarek Knap, my PhD student Jason Marshall has spent the summers of 2012 and 2013 collaborating with Dr. Knap. Jointly with Dr. Knap, we are developing the first strongly-coupled multiscale atomistic code for defects in solids with long-range electrostatic interactions. It is intended that this collaboration is maintained at this high level or increases further. Dr. Knap and I plan to have 2 PhD students from CMU visit in 2014 to work with him.

5 Awards

The PI received a number of research awards during the period of the grant. These include:

- 2012 NSF CAREER Award
- 2012 AFOSR Young Investigator Awards
- 2012 ARO Young Investigator Awards
- Junior chair from Carnegie Mellon University, 2013-2016
- Eshelby Award for Young Mechanics Faculty, 2013
My PhD student Jason Marshall was selected for an ARL Summer Internship for 2012 and 2013. In 2013, he was Winner of Computational and Information Sciences Directorate, and ARL-wide Bronze Medalist, at 2013 ARL Summer Graduate Student Symposium.