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Notice

Effective August 14, 2003, Eliot Fried moved officially from the University of Illinois at Urbana-Champaign to Washington University in St. Louis. This report closes out the file for work conducted at the University of Illinois at Urbana-Champaign. Hereafter, all further work performed by Eliot Fried will be at Washington University in St. Louis.

In addition to this final report, a renewal application was submitted on September 5, 2003. This report does not take the place of that renewal application.

Contents

1 Overview of key results and contributions

1.1 Defects in oriented media

Disclinations in nematic elastomers

In [D1] and [D6], with UIUC graduate student Russell Todres, we develop a theory for uniaxial nematic elastomers with variable asphericity. As an application of the theory, we consider the time-independent, isochoric radial expansion of a right circular cylinder. Numerical solutions to the resulting differential equation are obtained for a range of radial expansions. For all expansions considered, there exists an isotropic core of material surrounding the cylinder axis where the asphericity vanishes and in which the polymeric chains are shaped as spherical coils. This region, corresponding to a disclination of strength +1 along the axis, is bounded by a narrow transition layer across which the asphericity increases rapidly and attains a non-trivial positive value. The material thereby becomes anisotropic away from the disclination so that the polymeric chains are shaped as ellipsoidal coils of revolution prolate about the cylinder radius. In accordance with the area of steeply changing asphericity between isotropic and anisotropic regimes, a marked drop in the free-energy density is observed. The boundary of the disclination core is associated with the location of this energy drop. For realistic choices of material parameters, this criterion yields a core on the order of 10^{-2} μm, which coincides with observations in conventional liquid-crystal melts. Also occurring at the core boundary, and further confirming its location, are sharp transitions in the behavior of the constitutively determined contributions to the deformational stress and a change in the pressure. Furthermore, the constitutively determined contribution to the orientational stress is completely concentrated at the core boundary. The total energy shows a definitive preference for disclinated states.

[D10], also with R. Todres, focuses on the problem of detecting disclinations in nematic elastomers. To address this problem, we work with a right-circular cylindrical specimen composed of a nematic elastomer that is cross-linked in a uniaxial state, annealed, and then subjected to an isochoric radial expansion. Under these circumstances, numerical solutions show that, above a certain radial expansion, the material has a definitive energetic preference for a state involving a disclination of strength +1 along the cylinder axis. Surrounding such a disclination is a core with radial dimension on the order of 10^{-2} μm, which coincides with observations in conventional nematic melts. On examining the normal stress differences, we find that the first of these depends non-monotically on the extent of radial expansion and possesses a local minimum at the point where a disclination becomes energetically preferred. Based on this non-monotonicity, an experiment designed to measure normal stress differences could be used as a practical means to detect the presence of a disclination and, thereby, our theoretical predictions. This suggests a practical experimental method for testing our theoretical predictions.

[D13], with V. Korchagin and R. Todres, extends the works [D6, D7, D11] to allow for the
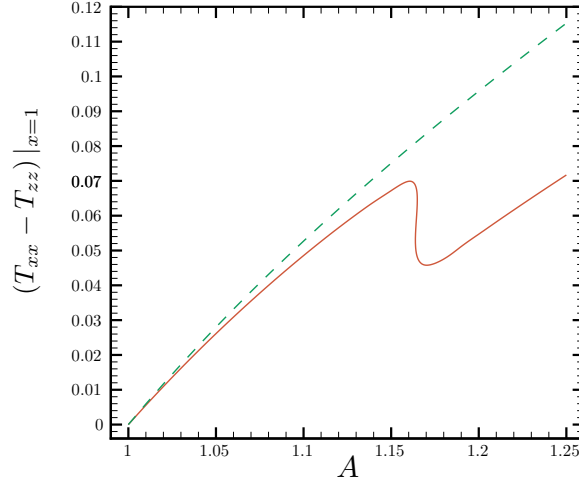
Figure 1: Plots of the value of the (nondimensional) first normal-stress difference (solid line) and its neo-Hookean counterpart (dashed line) at the boundary of the specimen ($x = 1$) as a function of the degree A of radial expansion between 1 and 1.25.

possibility of biaxial molecular conformation. For the problem of a right-circular cylindrical specimen composed of a nematic elastomer that is cross-linked in a uniaxial state, annealed, and then subjected to an isochoric radial expansion, this work shows that disclinated states in which the molecular conformation outside the disclination is biaxial are energetically preferred over those in which the conformation is uniaxial.

Disclinations in nematic liquid crystals

In [D3]. with P. Cermeli of U. Torino, the Ericksen–Leslie theory for nematic liquid-crystals is extended to account for the presence of disclinations. To achieve this, we use Gurtin's theory of configurational forces and obtain an evolution equation in which the resultant of the Eshelby stress at a disclination is proportional to the velocity of the disclination with respect to the material. Included among our results is a resolution of Eshelby's paradox concerning the nature of the force acting on a disclination. As an application, we show how surface disclinations can be described using our theory, and how these defects act to curve the free surface of the fluid, in competition with surface tension, and, thus, to create cusps at the surface.

Stripes in nematic elastomers

In [D4], with UIUC graduate student V. Korchagin, we consider a recent experiment of Kundler & Finkelmann [4], who subjected an aligned specimen of nematic elastomer to uniaxial extension and observed the formation of striped domains. In so doing, we apply the general theory for nematic elastomers developed by Anderson, Carlson & Fried [6] and work

2

with an energy density that combines the effects included in the molecular-statistical theory of nematic rubber elasticity with the Oseen–Zöcher–Frank theory of nematic curvature-elasticity. Assuming that the deformation and orientation fields remain in-plane, we arrive at a boundary-value problem which admits solutions corresponding to striped states. We use elementary aspects of bifurcation theory to explore the local stability of these solutions. We also obtain analytical estimates for the energy and thickness of interstripe domain walls as functions of imposed extension and compare these with numerical predictions.
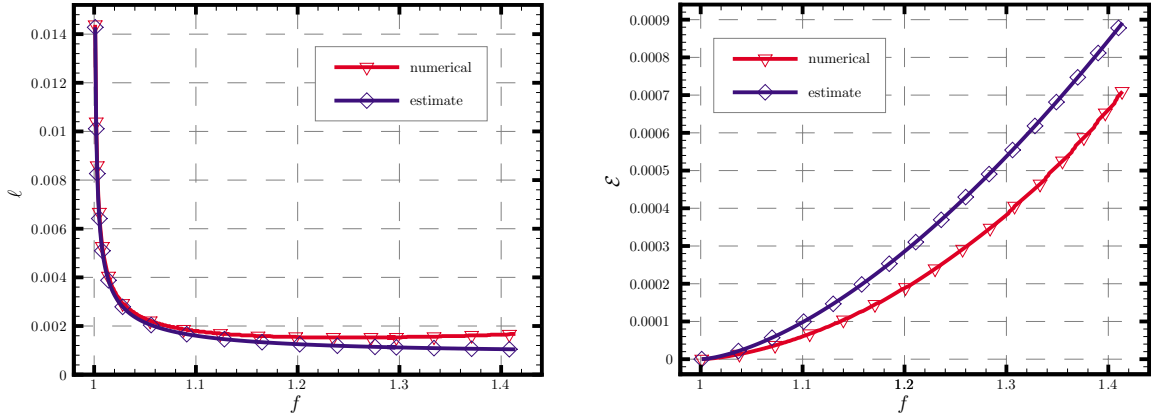

Figure 2: Dimensionless thickness ℓ and dimensionless energy $\mathcal{E}$ of the interstripe wall versus stretch f compared to the predictions of our analytical estimates.

1.2 Strain-gradient viscoplasticity

Single-crystals and bicrystals

[D2] develops a gradient theory of single-crystal plasticity that accounts for geometrically necessary dislocations. The theory is based on classical crystalline kinematics; classical macroforces; microforces for each slip system consistent with a microforce balance; a mechanical version of the second law that includes, via the microforces, work performed during slip; a rate-independent constitutive theory that includes dependences on a tensorial measure of geometrically necessary dislocations. The microforce balances are shown to be equivalent to *nonlocal* yield conditions for the individual slip systems. The field equations consist of the yield conditions coupled to the standard macroscopic force balance; these are supplemented by classical macroscopic boundary conditions in conjunction with nonstandard boundary conditions associated with slip. As an aid to solution, a weak (virtual power) formulation of the nonlocal yield conditions is derived. To make contact with classical dislocation theory, the microstresses are shown to represent counterparts of the Peach–Koehler force on a single dislocation. In [D8], with E. Bittencourt and A. Needleman of Brown U. and E. Van der Giessen of U. Groningen, discrete dislocation simulations of two boundary value problems
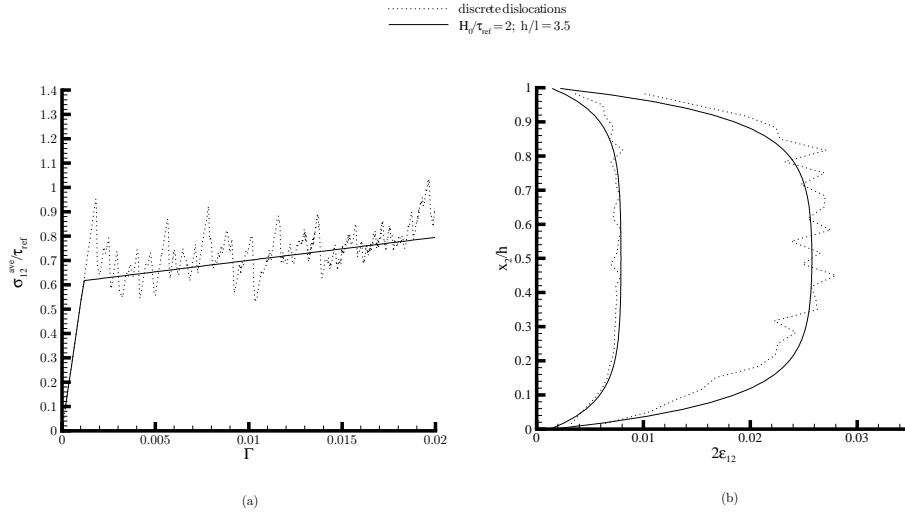
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Figure 3: Comparison of discrete dilsocation and strain-gradient crystalline plasticity solutions for a sheared strip assuming double slip. (a) Average shear stress response to the imposed shear strain Γ. (b) Shear strain distribution at $\Gamma = 0068, 0218$. The discrete dislocation data is from Shu, Fleck, Van der Geissen, and Needleman [1].

are used as numerical experiments to explore the extent to which the nonlocal crystal plasticity theory developed in [D2] can reproduce their predictions at micron length scales. In one problem simple shear of a constrained strip is analyzed, while the other problem concerns a two-dimensional model composite with elastic reinforcements in a crystalline matrix subject to macroscopic shear. In the constrained layer problem, boundary layers develop that give rise to size effects. In the composite problem, the discrete dislocation solutions exhibit composite hardening that depends on the reinforcement morphology, a size dependence of the overall stress-strain response for some morphologies, and a strong Bauschinger effect on unloading. In neither problem are the qualitative features of the discrete dislocation results represented by conventional continuum crystal plasticity. The nonlocal plasticity calculations reproduce the behaviors seen in the discrete dislocation simulations in remarkable detail (Figures 3 and 4).

[D11], with P. Cermelli of U. Torino, develops a gradient theory of small-deformation single-crystal plasticity that accounts for geometrically necessary dislocations (GNDs). The resulting framework is used to discuss grain boundaries. The grains are allowed to slip along the interface, but growth phenomenona and phase transitions are neglected. The bulk theory is based on the introduction of a microforce balance for each slip system, and includes a *defect energy* depending on a suitable measure of GNDs. The microforce balances are shown to be equivalent to *nonlocal* yield conditions for the individual slip systems, yield conditions that feature backstresses resulting from energy stored in dislocations. When applied to a grain boundary the theory leads to concomitant yield conditions: relative slip of the grains is
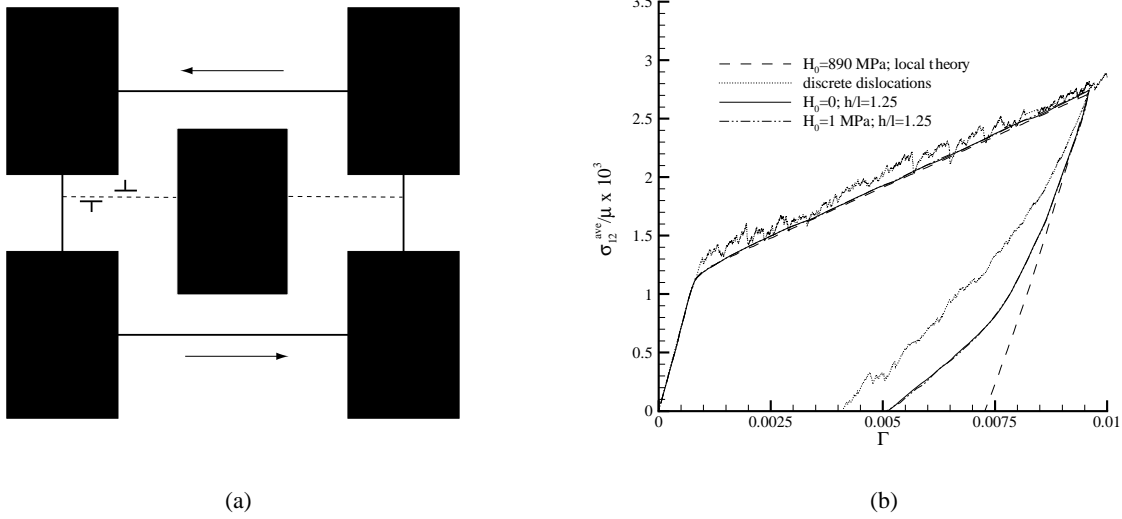
4

Figure 4: (a) Shear of a unit cell of a composite material with a doubly periodic array of elastic particles. (b) Comparison of discrete dislocation and strain-gradient crystalline plasticity solutions for the composite. The discrete dislocation data is from Cleveringa, Van der Geissen, and Needleman [2].

activated when the shear stress reaches a suitable threshold; plastic slip in bulk at the grain boundary is activated only when the local density of GNDs reaches an assigned threshold. Consequently, in the initial stages of plastic deformation the grain boundary acts as barrier to plastic slip, while in later stages the interface acts as a source or sink for dislocations. We obtain an exact solution for a simple problem in plane strain involving a semi-infinite compressed specimen that abuts a rigid material. We view this problem as an approximation to a situation involving a grain boundary between a grain with slip systems aligned for easy flow and a grain whose slip system alignment severely inhibits flow. The solution exhibits large slip gradients within a thin layer at the grain boundary.

Macrotheory

[D12] develops a general theory for small-deformation viscoplasticity based on: a system of microforces consistent with its own balance; a mechanical version of the second law that includes, via the microforces, work performed during viscoplastic flow; a constitutive theory that allows for dependences on plastic strain-gradients. The microforce balance and the constitutive equations — suitably restricted by the second law — are shown to be together equivalent to a flow rule that accounts for variations in free energy due to flow. When this energy is the sum of an elastic strain energy and a defect energy quadratic, isotropic, and positive definite in the plastic-strain gradients, the flow rule takes the form of a second-order parabolic PDE for the plastic strain coupled to the usual PDE arising from the standard macroscopic force balance and the elastic stress-strain relation. The classical macroscopic

5

boundary conditions are supplemented by nonstandard boundary conditions associated with viscoplastic flow. As an aid to solution, a weak (virtual power) formulation of the nonlocal flow rule is derived.

1.3 Phase transitions

Epitaxial growth

In [D9], we discuss the epitaxial growth of an elastic film, allowing for stress and diffusion within the film surface as well as nonequilibrium interactions between the film and the vapor. Our approach, which relies on recent ideas concerning configurational forces, is based on: (i) standard (Newtonian) balance laws for forces and moments together with an independent *balance law for configurational forces*; (ii) atomic balances, one for each species of mobile atoms; (iii) a mechanical version of the second law that accounts for temporal changes in free-energy, energy flows due to atomic transport, and power expended by *both standard and configurational forces*; (iv) thermodynamically consistent constitutive relations for the film surface and for the interaction between the surface and the vapor environment. The normal component of the configurational force balance at the surface represents a generalization, to a dynamical context involving dissipation, of a condition that would arise in equilibrium by considering variations of the total free-energy with respect to the configuration of the film surface. Our final results consist of partial differential equations that govern the evolution of the film surface.

When the interfacial energy is a nonconvex function of orientation, the anisotropic curvature flow equation becomes backward parabolic. To overcome the instability thus generated, a regularization of the equation that governs the evolution of the interface is needed. In [D7], with M. Jabbour of U. Kentucky we develop a regularized theory of curvature flow in three-dimensions that incorporates surface diffusion and bulk-surface interactions. The theory is based on a superficial mass balance; configurational forces and couples consistent with superficial force and moment balances; a mechanical version of the second law that includes, via the configurational moments, work that accompanies changes in the curvature of the interface; a constitutive theory whose main ingredient is a positive-definite, isotropic, quadratic dependence of the interfacial energy on the curvature tensor. Two special cases are investigated: (i) the interface is a boundary between bulk phases or grains, and (ii) the interface separates an elastic thin film bonded to a rigid substrate from a vapor phase whose sole action is the deposition of atoms on the surface.

Superelasticity of crystalline shape-memory materials

[D19], with L. Anand of MIT, develops a three-dimensional theory for the superelastic response of single-crystal, shape-memory materials. Since energetic considerations play a major role in the phase transformations associated with the superelastic response, we have devel-

oped the theory within a framework that accounts for the laws of thermodynamics. We have implemented a special set of constitutive equations resulting from the general theory in a finite-element computer program, and using this program have simulated the superelastic response of a single crystal Ti-Ni shape-memory alloy under both isothermal and thermo-mechanically coupled situations. Both manifestations of superelasticity — stress-strain response at fixed temperature and strain-temperature response at fixed stress — are explored. The single-crystal constitutive-model is also used to discuss the superelastic response of a polycrystalline aggregate with a random initial crystallographic texture. The overall features of the results from the numerical simulations are found to be qualitatively similar to existing experimental results on Ti-Ni.

Unified theory of evolving interfaces with deformation and atomic transport

[D17] is a (book-length) invited paper for Advances in Applied Mechanics. This paper reviews and unifies diverse theories of phase transitions involving deformation and atomic transport. Specifically, we develop a complete theory of evolving interfaces using a format based on:

(a) Standard (Newtonian) balance laws for forces and moments that account for standard stresses in bulk and within the interface.

(b) An *independent balance law for configurational forces* that accounts for configurational stresses in bulk and within the interface.

(c) Atomic balances, one for each atomic species. These balances account for bulk and surface diffusion.

(d) A mechanical version of the second law that accounts for temporal changes in free-energy, energy flows due to atomic transport, and *power expended by both standard and configurational forces*.

(e) Thermodynamically consistent constitutive relations for the interface and for the interaction between the interface and its environment.

We show that this framework encompasses a wide variety of classical theories for interface propagation, classical theories derived on a variety of physically appealing but ad hoc assumptions. One of the more interesting outcomes of the format we use is an explicit relation for the configurational surface tension σ in terms of other interfacial fields; viz.

$$\sigma = \psi - \delta^{\alpha}\mu^{\alpha} - \bar{\sigma}\varepsilon. \tag{1}$$

Here, ψ is free-energy density of the interface, δ^{α} and μ^{α} are the density and chemical potential of atoms of species α on the interface, $\bar{\sigma}$ is the interfacial stress, and ε is the interfacial tensile strain. This relation, a direct consequence of the free-energy imbalance
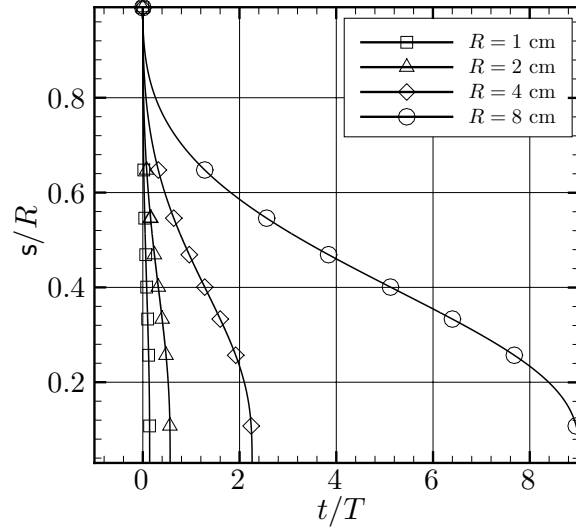
Figure 5: Normalized interface position with time for spherical specimens with radii of $R = 1, 2, 4$, and 8 cm. These plots bear a strong qualitative resemblance to the experimental results of Gehrke, Agrawal and Yang [3].

applied to the interface, is a basic relation valid for all isothermal interfaces, independent of constitutive assumptions and hence of material; it places in perspective the basic difference between the configurational and standard surface tensions, σ and $\bar{\sigma}$. There is much confusion in the literature concerning surface tension and its relation to surface free-energy. By (1), we see that these two notions coincide if and only if standard interfacial stress as well as interfacial atomic densities are negligible.

Swelling of hydrogels

[D16], with John Dolbow and graduate student Huidi Ji of Duke University, presents a theory for chemically-induced volume transitions in hydrogels. Consistent with experimental observations, we account for a sharp interface separating swelled and collapsed phases of the underlying polymer network. The polymer chains are treated as a solute with an associated diffusion potential and their concentration is assumed to be discontinuous across the interface. In addition to the standard bulk and interfacial equations imposing force balance and solute balance, the theory involves an ancillary interfacial equation imposing configurational force balance. Motivated by experimental observations, we specialize the theory to the situation where the time scale associated with the interface motion is slow compared to those associated with diffusion in the bulk phases. We present a hybrid eXtended-Finite-Element/Level-Set Method for obtaining approximate solutions to the equations arising under this specialization. As an application, we consider the swelling of a spherical specimen whose boundary is traction-free and is in contact with a reservoir of uniform chemical potential. Our numerical results exhibit good qualitative comparison with experimental ob-

8

servations (Figure 5) and predict characteristic swelling times that are proportional to the square of the specimen radius. Our results also suggest several possible synthetic pathways that might be pursued as a means to engineer hydrogels with optimal response times.

1.4 Amorphous materials

Molecular-statistical theory

By relaxing the assumption that the end-to-end vectors of molecules transform as macroscopic material line elements, we arrive in [D9] at a generalization of the molecular-statistical theory of rubber elasticity. This generalization includes as special cases continuum-mechanical theories proposed by Mooney and by Rivlin and Saunders as improvements upon the classical neo-Hookean theory.

Polymers and metallic glasses

[D18], with Lallit Anand of MIT, develops a continuum theory for the elastic-viscoplastic deformation of amorphous solids such as polymeric and metallic glasses. Introducing an internal-state variable that represents the local free-volume, we are able to capture the highly non-linear stress-strain behavior that precedes the yield-peak and gives rise to post-yield strain-softening. Our theory explicitly accounts for the dependence of the Helmholtz free energy on plastic deformation in a thermodynamically consistent manner. This dependence leads directly to a backstress in the underlying flow rule, and allows us to model the rapid strain-hardening response after the initial yield-drop in monotonic deformations, as well as the Bauschinger-type reverse-yielding phenomena typically observed in amorphous polymeric solids upon unloading after large plastic deformations. We have implemented a special set of constitutive equations resulting from the general theory in a finite-element computer program. Using this, we apply the specialized equations to model the large-deformation response of the amorphous polymeric solid polycarbonate, at ambient temperature and pressure. We show numerical results to some representative problems, and compare them against corresponding results from physical experiments (Figure 6).

1.5 Compaction and segregation in flowing granular materials

In [D15], with K. Hutter of T. U. Darmstadt, we have developed a continuum mechanical theory for the compaction of granular aggregates and the segregation by particle-type (e.g., by size) within such aggregates. Our theory characterizes the microphysical mechanisms that drive these macroscopic processes via a framework that treats the medium as a mixture whose individual constituents are particle types and voids. While continuum theories have been developed for monodisperse granular media, to our knowledge no such approach been applied to the study of segregation and compaction during the flow of polydisperse media.
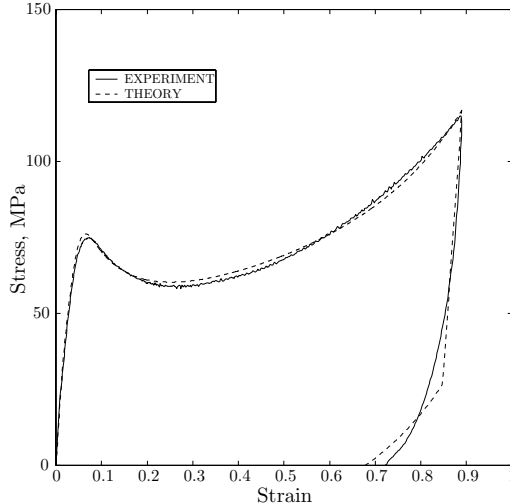
Figure 6: Stress-strain response of polycarbonate in simple compression showing reverse yielding upon unloading due to the development of a back stress. The calculated response shows the same phenomenon.

The resulting theory leads to a partial differential equation for the macroscopic motion of the material coupled to a system of partial differential equations for the volume fractions of the individual particle types. We present an explicit solution for an arbitrary loosely packed mixture of small and large particles in a fixed container under the influence of gravity. We show that a layer of large particles forms at the free surface and grows with time, while a close-packed mixture of large and small particles forms and grows from the base of the container; the final solution, attained in finite time, consists of a layer of close-packed large particles above a close-packed mixed state. At the mundane level of everyday experience, this solution at least qualitatively explains why in a can of mixed nuts, Brazil nuts are generally found at the top.

When segregation is ignored, so that the focus is compaction, this system is replaced by a scalar partial differential equation that closely resembles equations arising in descriptions of traffic flow.

[D14], with UIUC graduate student Bidhan Roy, uses a theory recently developed by Fried, Gurtin and Hutter [D15] to study the gravity-induced segregation of granular mixtures. In particular, we consider a one-dimensional initial-value problem for a granular mixture of $K > 2$ discrete sizes bounded below by an impermeable base, above by an evolving free surface, and subject to gravity. We demonstrate the existence of a solution in which the medium segregates by particle size. For a mixture of small and large particles ($K = 2$), we use methods of Smoller [7] to show that the segregated solution is unique. Further, for a mixture of small, medium, and large particles ($K = 3$), we use LeVeque's [8] CLAWPACK to construct numerical solutions and find that these compare favorably with analytical predictions. The issue of uniqueness for $K > 2$ particle types remains open.

10

1.6   Papers arising from work under this grant

[D1] E. Fried & R. E. Todres, Prediction of disclinations in nematic elastomers, *Proceedings of the National Academy of Sciences of the United States of America* **98** (2001) 14773–14777.

[D2] M. E. Gurtin, A gradient theory of single-crystal viscoplasticity that accounts for geometrically necessary dislocations, *Journal of the Mechanics and Physics of Solids* **50** (2002) 4–32.

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[D13] E. Fried, V. Korchagin & R. E. Todres, Biaxial disclinated states in nematic elastomers, *Journal of Chemical Physics* **119** (2003) 13170–13179.

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[D15] E. Fried, M. E. Gurtin & K. Hutter, A void based description of segregation and compaction in flowing granular materials, *Continuum Mechanics and Thermodynamics* **16** (2004), 199–219.

[D16] J. Dolbow, E. Fried & H. Ji, Chemically-induced swelling of hydrogels, *Journal of the Mechanics and Physics of Solids* **52** (2004) 51–84.

[D17] E. Fried & M. E. Gurtin, A unified treatment of evolving interfaces with deformation and atomic transport with emphasis on grain-boundaries and epitaxy, *Advances in Applied Mechanics*, in press.

[D18] L. Anand & M. E. Gurtin, A theory of amorphous solids undergoing large deformations, with application to polymeric materials, *International Journal of Solids and Structures*, in press.

[D19] L. Anand & M. E. Gurtin, Thermal effects in the superelasticity of shape memory materials, *Journal of the Mechanics and Physics of Solids*, in press.

2 Interaction with DOE laboratories

Gurtin visited Lawrence Livermore Laboratories in September, 2002; there he gave a seminar:

- *Plasticity of single crystals at the micron level: a gradient theory that accounts for geometrically necessary dislocations.*

During this visit Gurtin had a detailed discussion with TOM ARSENLIS concerning strain-gradient theories of crystal plasticity; more specifically on the characterization of geometrically necessary dislocations (GNDs) through: (i) the use of evolution equations for dislocation densities, and (ii) through a free-energetic dependence on a tensorial measure of GNDs (Nye tensor). Gurtin also had detailed discussions with JAMES STOLKEN on strain-gradient crystal-plasticity theories and more specifically on possible experiments that may be used to compare strain-gradient theories. Gurtin also met with DAVE LASILLAS, CHRIS KRENN, MONICA BARNEY, RICH BECKER, and BRUCE REMINGTON.

Fried and Gurtin visited Sandia Albuquerque National Laboratories in July, 2003; there Fried gave a seminar:

- *Disclinations in nematic elastomers*

and Gurtin gave a seminar:

- *Micron-level, single- and poly-crystal plasticity: gradient theories that account for geometrically necessary dislocations.*

During this visit, Fried and Gurtin had detailed discussions with STEWART SILLING on his peridynamic theory for solids. Also, Fried discussed foams and block copolymers with ANDREW KRAYNIK and Gurtin discussed topics related to geometrically necessary dislocations and strain-gradient plasticity with ART COUNTS. In addition, Fried and Gurtin met with STEVEN PLIMPTON to discuss the application of molecular dynamics simulations to the study of polymers.

3 Collaborators at other universities

- Lallit Anand, MIT

- Paolo Cermelli, University of Torino

- John Dolbow, Duke University

- Kolumban Hutter, Technical University of Darmstadt

- Alan Needleman, Brown University

- Erik van der Geissen, University of Gronigen

4 Graduate students and postdoctoral research associates

- Huidi Ji, Duke University

- Vladimir Korchagin, University of Illinois

- Bidhan Roy, University of Illinois

- Russell Todres, University of Illinois

- Michel Jabbour, University of Kentucky

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