Opportunities in Martensite Theory

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Abstract. A workshop has explored interactions of materials science, applied mechanics, physics and mathematics in understanding fundamentals of martensitic transformations. System theory offers a framework for addressing realistic complexity. Theory of invariant-plane kinematics has been extended to multivariant plate groups and hierarchical structures. Electronic total energy calculations explore the origins of martensitic phase stability, and Landau-Ginzberg models for transformations with and without shuffles provide nonlocal continuum theories for treatment of interfacial structure and mobility as well as the competition between classical and nonclassical transformation mechanisms. Quantitative kinetic theory incorporates defect distributions, but requires further analysis of the evolution of mean particle volume. Kinetic theory provides the basis for constitutive relations for transformation plasticity.

1. INTRODUCTION

Similar to a workshop held in 1991 in preparation for ICOMAT 92, a Martensite Theory Workshop was held at Northwestern University in June 1995 to assess developments in materials science, applied mechanics, physics and mathematics offering new insights into martensitic transformations, and to prepare the most effective communication of these ideas to the traditional martensite community at ICOMAT 95. This paper is a summary of those discussions, which covered system theory, kinematics, energetics, interfaces, mechanisms, kinetics, and transformation plasticity.

2. SYSTEM THEORY

While the contributions of science to engineering are widely acknowledged, engineering can make equally profound contributions to basic science if our goal is to understand the real complexity of nature [1]. Engineering system theory [2] provides a general framework for complex dynamic multilevel structure which has already been applied to the integration of scientific knowledge in the conceptual design of martensitic steels [3]. A finer-scale application, treating a martensitic transformation itself as a system is represented by the flow-block diagram of Figure 1. Blocks at the left represent interactive levels of structure both preceding and generated by the transformation, while blocks at the right denote a sequence of dynamic phenomena interacting with these structure levels. As denoted by the interconnections, initial nucleation is sensitive to interfacial defects generated by the processing history, determining the initial number density N_i of heterogeneous nucleation sites and the critical driving force at which they operate. The latter determines the dynamics of unit growth events and influences the degree of dislocation substructure generation determining subunit morphology (plate/lath) and the initial average particle volume $\bar{V}_o$. The character of growth in turn establishes the elastic or plastic fields governing the number density N_A and potency of autocatalytic nucleation sites, which evolve with phase fraction, f. The character of autocatalytic nucleation in turn establishes the initial geometry of variant pairs influencing the cooperative multivariant growth of higher level groups (packet/sheaf) determining the evolution of average particle volume, $\bar{V}(f)$. On the finest scale, solutes and other fine scale defects exert an influence on virtually all higher levels primarily through interfacial friction.

This system view provides a methodology for treating the full-scale path-dependent evolution of a dynamic martensitic microstructure, while providing a framework within which detailed simplifying analysis of fundamental subsystems can be undertaken (and prioritized) without losing sight of interactive connections back to the holistic integrated phenomenon of ultimate interest. Allowing the treatment of detail without the destruction of essence, the systems approach not only offers more accurate science, but fosters the generation of scientific knowledge in a useable form for its integration in the realistic solution of engineering problems of practical utility.

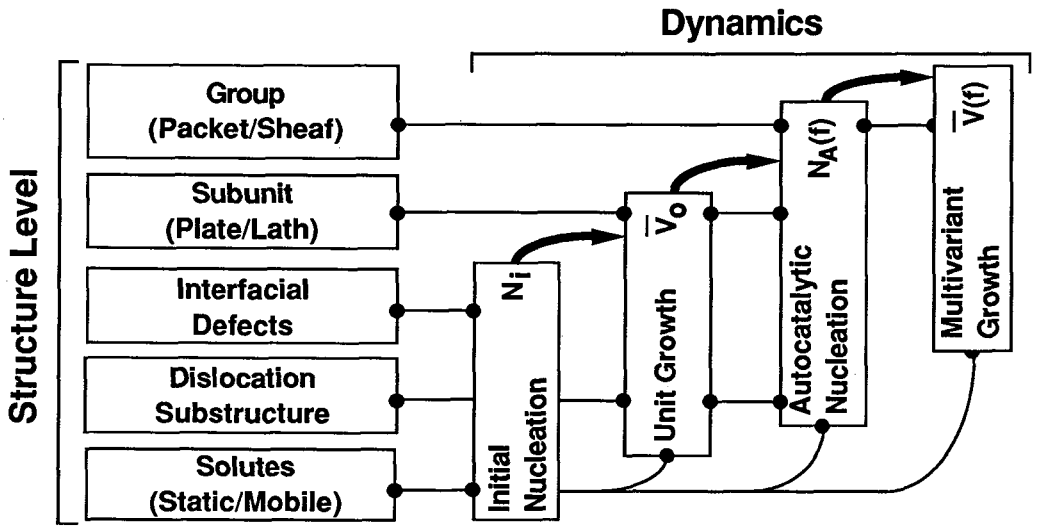

Figure 1: Flow-block diagram of martensitic transformation as a system.

3. KINEMATICS

At the level of a single semicoherent interface or a single enclosed semicoherent particle, the kinematics (net deformations) represent the best understood aspect of martensitic transformations, providing the cornerstone of martensite theory. Originally considered phenomenological, the invariant plane strain (IPS) theory [4,5] was given a fundamental basis in volume elastic energy minimization by Christian [6] and Roitburd [7] in a linear approximation and recently by Ball and James [8] in a geometric nonlinear form.

While the IPS theory gives insight into the start of transformation at the single particle (subunit) level, the nature of final fully transformed multivariant microstructures (the highest level of Figure 1) has been well described by the linear polydomain theory of Roitburd [9] including hierarchically twinned domain structures. A recent extension by James, Kohn and Shield [10] treats details of the "branched needle" microstructure where a set of parallel twins on the K_I plane meets a second array on the nearly-orthogonal K_{II} plane. Linear and geometric nonlinear descriptions treat the precise twin tapering and elastic twin bending in a transition layer between regions of invariance of the K_I and K_{II} planes.

A full kinematic description of the dynamic system of Figure 1 requires treatment of small groups of particles and their interactions in progressively space filling patterns. An important contribution toward this has been the detailed analysis by Bhattacharya [11] of the two-plate "wedge" pair represented in Figure 2. The conditions are treated for simultaneous invariance of the two parent/martensite habit planes and the martensite intervariant interface, identifying lattice parameter conditions for design of thermoelastic alloys with optimal elastic compatibility.

At the highest level of microstructure, the kinematics of shape memory strain recovery in polycrystals has been treated by Bhattacharya and Kohn [12] employing a modified Taylor analysis. From the symmetry of the lattice deformation, the relative degree of recoverable strain is predicted in a wide range of systems, and desirable crystallographic textures are identified.

The range of martensitic behaviors predictable from pure kinematics is remarkable, and the kinematics of multiparticle systems remains a fruitful area for further application of new theoretical tools.

4. ENERGETICS

While martensitic microstructures are highly constrained by kinematics, other aspects, notably

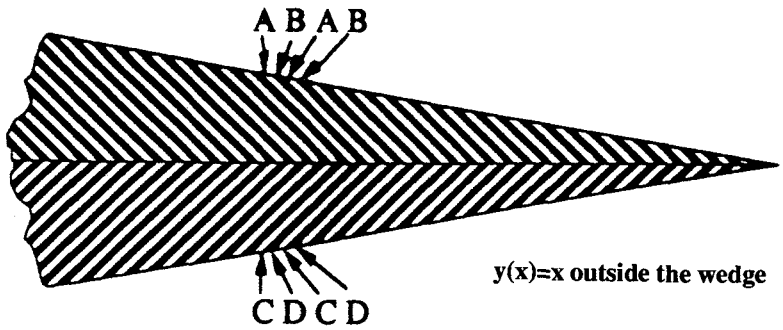

Figure 2: Wedge pair of internally twinned martensite plates with three invariant plane interfaces [11].

microstructural length scales, depend sensitively on the energetic consequences of the deformations. The most important contribution of new atomistic-level theoretical tools has been *ab initio* electronic quantum-mechanical calculations of the energy differences between parent and product lattices (offering insights into the origin of relative phase stability from band structure and bonding) and the physical nonlinear energetics of homogeneous lattice deformations (including internal shuffle displacements) between structures. These calculations have included alkali metals [13,14], Fe and its alloys [15-17], Cu-base alloys [16], and most recently TiNi alloys [18,19]. The lattice deformation calculations define energy density barrier heights important to coherent interfacial energy, and identify conditions where energy surfaces are cusped [15] rather than smooth. Most important to mechanistic issues, calculations as a function of pressure identify critical driving forces for lattice instabilities [15].

The application of calculated lattice deformation energetics in higher level problems is aided by the development of analytic functional forms based in Landau theory, which have now been applied to explicitly treat the role of internal shuffle displacements in martensitic transformations [20,21]. An important development pioneered by Barsch and coworkers has been the extension to Landau-Ginzberg theory combining nonlocal gradient energies derived from phonon dispersion curves for specific systems [22-24] and including shuffle contributions [21].

5. INTERFACES

Electronic total energy calculations have been applied to the structure and energy of coherent and "incoherent" twin boundaries [25,26] relevant to martensitic microstructures. Such calculations have traditionally entailed structural relaxation using less precise methods such as interatomic potentials prior to running computationally-intensive rigorous calculations on a single configuration. A recent breakthrough has been the modification of the precise Full Potential Augmented Plane Wave (FLAPW) method to evaluate interatomic forces [27] so that relaxation and final calculation are run within the same method in a self-consistent manner.

Few aspects of martensitic transformations are genuinely atomistic in nature, and continuum methods (incorporating crystal-derived constraints) have proved the most valuable theoretical tools. Barsch and coworkers have applied the Landau-Ginzberg models to compute the detailed core structure of coherent twin boundaries in tetragonal martensites in shuffle-free (proper ferroelastic) systems such as InTl [22] and FePd [23,24]. A typical structure is shown in Figure 3a with the corresponding strain profile plotted in Figure 3b. As an example of a system with a strong shuffle component (improper ferroelastic), the cubic-tetragonal transformation in rare-earth/silver based intermetallic compounds is described by a primary six-component shuffle order parameter, with strain as a secondary order parameter, and precise variational solutions are obtained for the distribution of shuffles across the interface [21]. Such predictions provide a basis for comparison with direct observation by high-resolution electron microscopy (aided by image simulation) of the distribution of shuffles and strain across the parent/martensite interface as obtained thus far in the CuZn system [28]. The coherent BCC/9R transformation in the latter system is a fruitful area for future application of the Landau-Ginzberg approach to the parent/martensite interface.

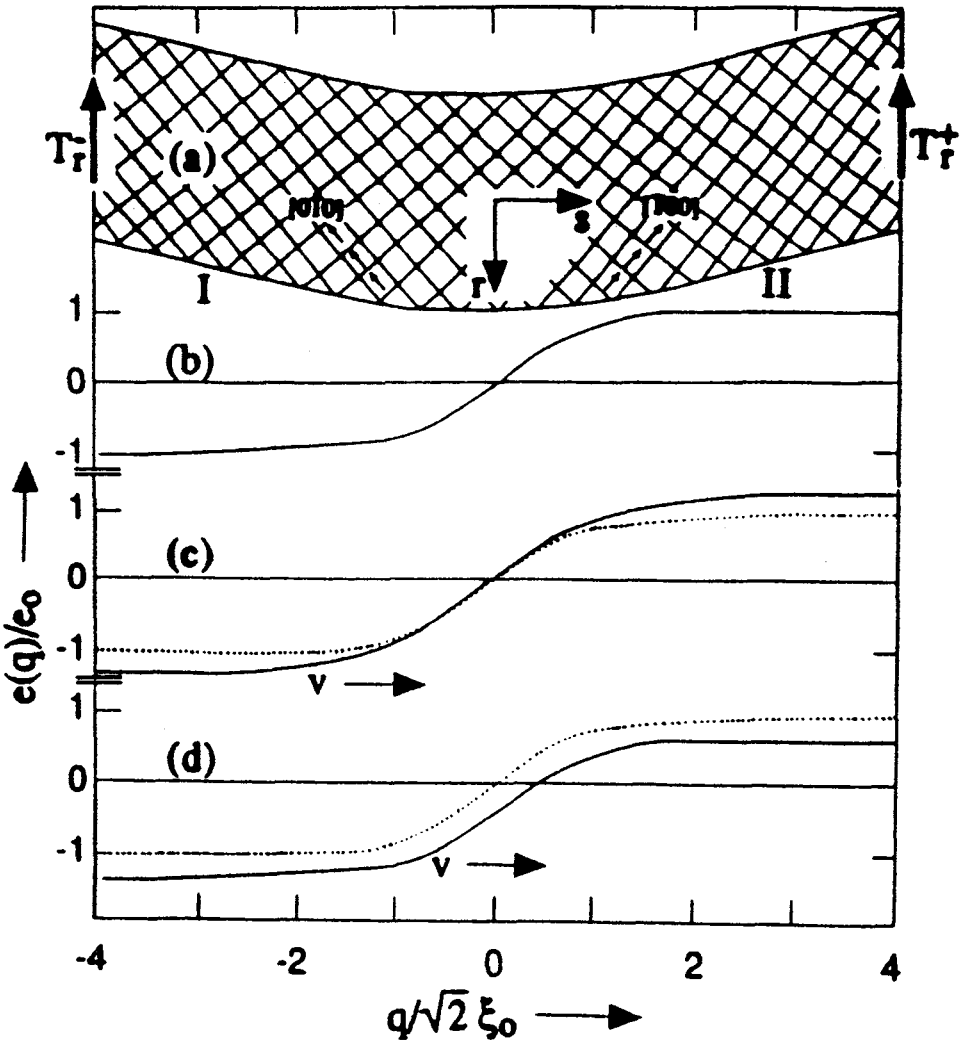

Figure 3: Computed structures of internal twin boundaries in tetragonal martensites, (a) 2D structure, (b) strain profile across static interface, (c) strain profile of interface moving without dissipation (solid) compared to static interface (dotted), (d) strain profile of interface moving with phonon drag (solid) compared to static interface (dotted) [35].

Dislocation-based linear elastic models of parent/martensite interfaces [29,30] have proved effective in accounting for measured mobility in the low-velocity thermal regime [31] controlled by defect elastic interactions including solution hardening [32,33]. Mobility in the high velocity phonon-drag controlled regime for which martensite is famous has been recently investigated experimentally [34] and is ripe for theoretical treatment. An important first contribution is the extension of the Landau-Ginzberg approach to moving twin interfaces in tetragonal V_3Si incorporating a phonon viscosity deduced from available experimental data [35]. The strain variation in Figure 3c represents the computed configuration of such an interface moving without dissipation, while the variation in Figure 3d incorporates the influence of phonon viscosity, allowing prediction of mobility in the high velocity regime.

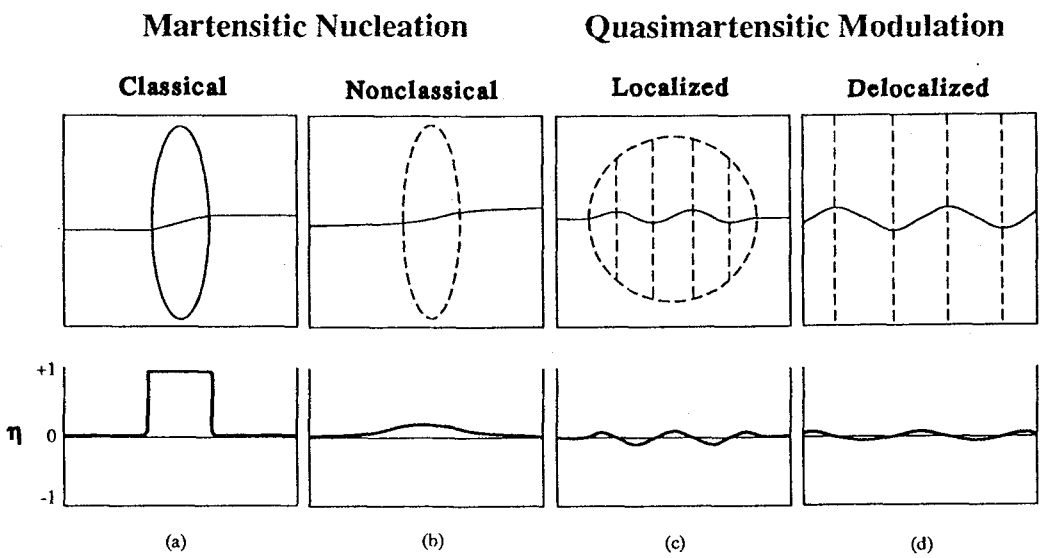

Figure 4: Geometries and displacements (upper) with corresponding strain profiles (lower) for critical events controlling simple shear transformation by (a) classical martensitic nucleation, (b) nonclassical martensitic nucleation, (c) localized quasimartensitic modulation, and (d) delocalized quasimartensitic modulation [36].

6. MECHANISMS

Current mechanistic issues concern the competition amongst the alternative transformation mechanisms depicted by the schematic critical event structures and associated strain profiles of Figure 4 for the case of a simple shear transformation [36]. Figure 4a represents classical martensitic nucleation, where the critical nucleus has the full transformation strain amplitude and a relatively sharp interface, while Figure 4b depicts a nonclassical critical nucleus with reduced strain amplitude and a diffuse interface. A process of delocalized continuous strain modulation expected for higher-order or very weakly first-order systems is depicted in Figure 4d, while Figure 4c represents a localized continuous modulation packet of self-accommodating variants for the fluctuation-assisted transformation of a weakly metastable system. The latter continuous modulation processes have been termed *quasimartensitic* transformation [37] to distinguish from the more localized process of martensitic nucleation.

The Landau-Ginzberg approach has allowed theoretical comparison of these mechanisms within a single framework. The dynamics of the delocalized modulation process of Figure 4d has recently been analyzed by Umantsev [38,39] for the case of a weakly first-order system in which an isothermal shear modulus has become negative. As discussed elsewhere in connection with transformation classification criteria [40], the analysis identifies conditions under which strain modulation is controlled by transfer of the transformation latent heat, giving a direct displacive analogue of spinodal decomposition. When the adiabatic shear modulus is sufficiently negative, the dynamics become controlled by mechanical relaxation. For the latter case in underdamped systems exhibiting hydrodynamic behavior, a numerical analysis by Reid [41] has shown the behavior represented by the 1-dimensional strain profile of Figure 5. Corresponding to a hybrid of the profiles of Figure 4, inertial effects are found to favor localized nucleation at the free surface at the left, with continuous modulation in the interior. Similar results are obtained in 2-dimensional simulations, with heterogeneous nucleation at corners and faces followed by continuous modulation of the bulk material. Extension of such analyses to the weakly metastable regime where such processes can be aided by thermal fluctuations (e.g., as in Figure 4c) should provide quantitative criteria for the ultimate competition between the primary alternative mechanisms of martensitic nucleation and growth vs. quasimartensitic modulation, thus defining, for weakly first-order systems, where martensite ends and other transformations take over.

The transition of martensitic nucleation behavior between the classical and nonclassical modes of Figures 4a and b has been treated most rigorously in the simplifying context of homogeneous (defect-

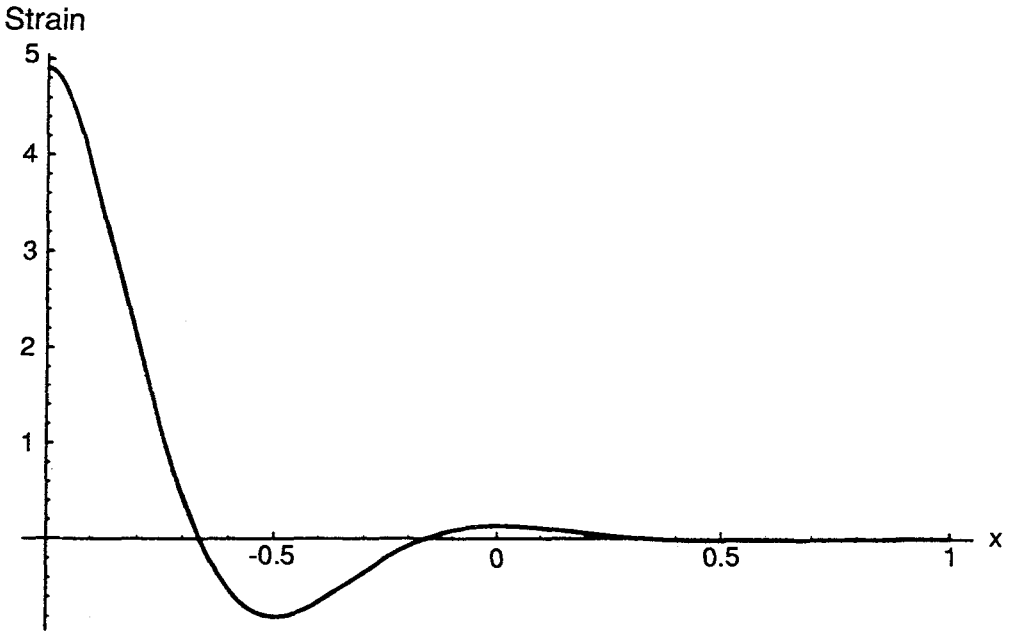

Figure 5: Computed strain profile of transforming unstable hydrodynamic system with free surface at left [41]. Nucleation at surface is accompanied by continuous modulation in interior.

free) nucleation, a regime which has been recently accessed in small particle experiments in the Fe-Co system [42]. Applying the Landau-Ginzberg approach to the related problem of a pure dilational transformation (as in Ce alloys), rigorous 3-dimensional solutions for the evolution of critical nucleus structure and energy with increasing driving force have been obtained by Moran and coworkers [43] employing fine-element methods. The approach has been extended [44] to 2-dimensional solutions for the simple shear transformation of Figure 4, employing a new Element-Free Gallerkin (EFG) method [45] with advantages for treating both large strain deformations and strain gradients. Figure 6 summarizes the evolution of critical nucleus shape and size with increasing α, the driving force normalized to its critical value at lattice instability. A surprising result is the shape of the classical (full strain amplitude) critical nucleus at α=0.6. Instead of the single plate (lath) traditionally assumed [46], the nucleus adopts a star configuration equivalent to two intersecting particles of the same variant. With increasing α, the nucleus strain amplitude diminishes, the size diverges, and the interface becomes more diffuse. Apart from the unexpected shapes, the rigorous calculations confirm the general features of earlier approximate calculations [47,48], with strongly nonclassical behavior requiring α>0.9.

The mechanism of greatest practical interest is the *heterogeneous* nucleation that dominates the behavior of martensitic transformations. As reviewed recently [49], the essential characteristics of heterogeneous martensitic nucleation have been well understood theoretically for the past two decades [50-52] and confirmed by direct electron microscopy observation [53]. More rigorous calculations are of particular interest regarding the degree of nonclassical behavior. Approximate applications of Landau-Ginzberg models so far predict that barrierless nucleation at strong defects is preceded by formation of classical embryos [48,54]. An important advance [55] has been the successful calculation of the structure of a dislocation in the same nonlinear nonlocal medium of Figure 6 employing the EFG method. This opens the way to study of the detailed evolution of embryo structure with increasing driving force at dislocation arrays modeling observed nucleation sites.

7. KINETICS

In a first-order transformation, the transformed volume fraction f is the product of the particle number density N_V and the average particle volume $\bar{V}$. The goal of mechanistically-based kinetic theory is to

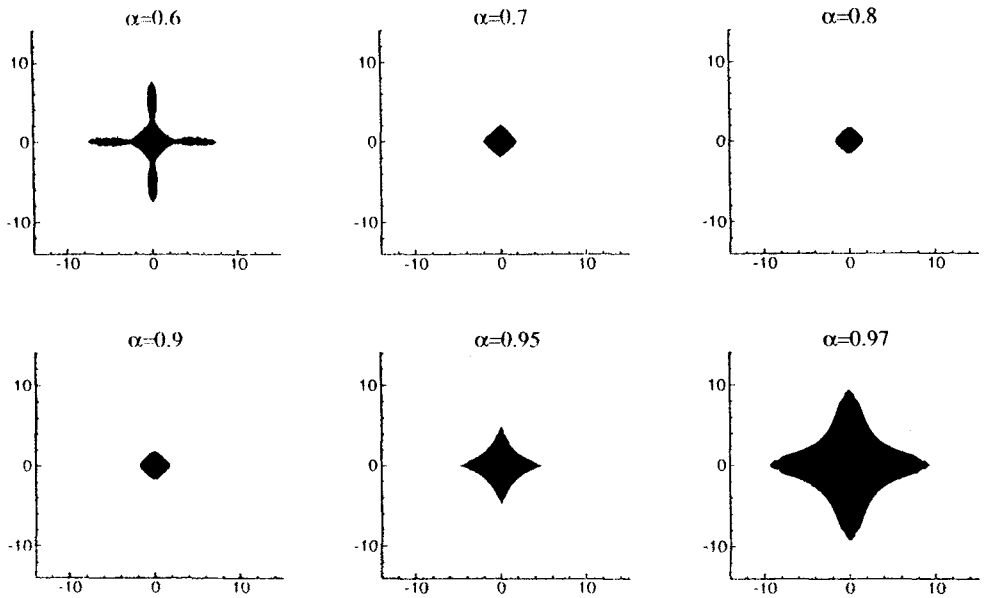

Figure 6: Evolution of critical nucleus size and shape for 2D shear transformation with increasing normalized driving force, α [44].

describe the evolution of both these contributions, as governed by the various phenomena summarized in Figure 1.

Employing classical heterogeneous nucleation theory [50], the evolution of N_V is well described by an exponential potency distribution of initial nucleation sites derived from small-particle [56] and bulk acoustic-emission [57] experiments in metals and ceramics [58], combined with a normal distribution of autocatalytic sites whose amplitude scales with f, as derived from quantitative stereological studies [59]. Incorporating the influence of the thermal component of interfacial nobility [30,33], the defect potency distribution gives a distribution of activation energies [59] predicting observed transitions between athermal and isothermal kinetic behaviors [e.g., 60]. Detailed comparison of transformation kinetics in Fe-Ni and Fe-Pt alloys [61] reveals equivalent nucleation site potencies in thermoelastic and nonthermoelastic transformations. A model system for study of distributed heterogeneous nucleation is the stress-induced twin conversion in CuAlNi martensites where the detailed analysis of the "branched needle structure" described earlier [10] can allow prediction of observed heterogeneous twin nucleation ("tip splitting") governing observed stress-strain curve shapes [62]. A remaining opportunity is a merger of microscopic (ergodic) and macroscopic (nonergodic) statistical theories to account for observed forms of defect distribution functions [e.g., 63] which often exert a dominant influence on macroscopic behavior.

The evolution of average particle volume $\overline{V}$ with f has been well studied in nonthermoelastic plate martensites [64]. An initially constant $\overline{V}$ is associated with grain-to-grain spreading of transformation, followed by a monotonic decrease during "filling in" of partially transformed grains, described by an empirical power law [59]. In contrast, the behavior of $\overline{V}(f)$ is nonmonotonic in thermoelastic systems [61] due to an initial increase in $\overline{V}$ attributed to plate rearrangement for better self-accommodation promoting thicker plates at the thermoelastic equilibrium condition [65]. The decrease in $\overline{V}$ at high f associated with microstructural partitioning is similar in thermoelastic and nonthermoelastic systems, but more gradual than the original geometric partitioning model of Fisher [66]. The observed fractal character of plate martensite microstructures [67] may provide the basis for an improved partitioning model. Finding theoretical forms for $\overline{V}(f)$ is a worthy goal for full simulations of the evolution of martensitic microstructures [68]. The $\overline{V}(f)$ behavior has an important role in the incomplete transformation or "stasis" behavior of martensitic and other displacive transformations.

8. TRANSFORMATION PLASTICITY

The transformation plasticity phenomena exhibited in both thermoelastic and nonthermoelastic alloys have attracted the interest of the applied mechanics community [69] resulting in a number of phenomenological constitutive models. The kinetic theory of martensitic transformations provides the basis for more realistic mechanistic models. Analysis of the broadening of the effective potency distribution by the orientation distribution of heterogeneous nucleation sites predicts a smooth yield locus for stress-assisted transformation under multiaxial stress [57,70], and a self-consistent treatment accounting for the noncentrosymmetric transformation shear systems prescribes differences in tension vs. compression [71]. The development of kinetics-based constitutive relations for both the stress-assisted and strain-induced modes of transformation and their application in enhancement of ductility and toughness have been reviewed recently [72].

9. CONCLUSIONS

The application of both traditional and new theoretical tools remains a rewarding activity for gaining insight into the diverse phenomena associated with martensitic transformations. Continuing analysis not only offers a deeper basic understanding, but the parallel development of methodologies of integrative synthesis for incorporating knowledge in the solution of problems of realistic complexity promises a science-based engineering with broad impact over the array of materials technologies which martensite pervades.

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