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Significance of slip propensity determination in shape memory alloys

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ABSTRACT

Performance degradation in a shape memory alloy (SMA) occurs due to dislocation activities at any stage of its thermo-mechanical deformation history. Knowing the physical energy barriers to activate slip could prove instrumental in devising strategies to engineer superior resistance thereto. Given the multi-elements, the SMA slip propensity is intrinsically decided by inherent bonding landscapes. In essence, the problem of theorizing SMA slipping tendency reduces to a quantum mechanical one. Consequently, atomistic calculations can provide invaluable experimentally verifiable material trend as well as potentially pave the way for advancing novel SMAs.

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1. Introduction

Shape memory alloys (SMA) can recover large deformations on removal of applied thermal, mechanical or magnetic loads [1,2]. This unique healing ability specializes SMAs as versatile multifunctional actuators in diversified industrial settings [3,4]. During the entire service lifetime, an SMA-made component is expected to undergo multitudes of shape change cycles [5]. Underlying the constitutive responses is the reversible transformations of crystal phases at the sub-structural scale triggered in response to thermomechanical or magnetic stimuli. Crucial to its functionality is the retention of this phase reversibility with a complete absence of plasticity [6,7] irrespective of the loading pathways. Performance compromise would be instigated at any stage of thermomechanical history provided dislocation slip starts accumulating [8–10] often facilitated by highly stressed local sites (e.g. grain or phase boundaries). Once started, growing slip activities would lead to the loss of expected SMA properties over time/cycles. Preserving the desired functionality of SMAs is construed as a complete recovery of large strains, which can be manifested in several forms (Fig. 1).

The *shape memory* behavior at low temperature is related with the twinned martensite transitioning to a detwinned one when stressed incrementally (green loop). The residual strain that remains in the unstressed material can be recovered by applying isobaric heating followed by cooling (blue loop). At a higher temperature, the strained material can recuperate isothermally if mechanical loads are simply removed, an attribute known as *superelasticity* (red loop). From material to material, various shapes/sizes of the constitutive curves and the associated thresholds of structural transformations are noted in the stress–strain-temperature space. A persistent challenge in maintaining the foregoing SMA functionalities is the prevention of dislocation slip during these transitions. The precise evaluation of material dislocation characteristics remains a topic of rigorous research to date. Conventionally, the empirical plastic attributes are established by painstaking trial-and-error based experimental approaches. Modeling endeavors in this regard has been undertaken within the framework of continuum based crystal plasticity assumptions [11,12]. Undoubtedly, it would be quite advantageous to have a priori method for assessing the vulnerable slip systems for plastic deformation. Such information would be useful in developing more comprehensive theories spanning a broad spectrum of shape memory behaviors.

2. Motivation for strategizing a rapid evaluation method

Over the last decade, rigorous efforts have been afoot in the form of expanding the horizon of SMA usage. The slip issue remains a particularly pressing problem in view of enforcing more demanding operating conditions [13,14] (e.g. withstanding extreme temperatures while restoring large strains, say, in hot-section components). Of particular concern is the fact that the material slip resistance continues to decrease with rising temperature. At elevated temperatures, an enhanced tendency of thermal activation would foster an environment of easy glide. Combined with the possibility of triggering localized dislocation sources (i.e. at interfacial defects), this poses an immediate design constraint that limits the high temperature operational ceilings of SMAs. From fabrication standpoint, one of the most desirable traits to accomplish in an SMA therefore would be to impart a very high slip resistance both in austenitic and martensitic crystals [15] (Fig. 1). Other attractive and concurrently looked-for features include a low transformation stress as well as a high recoverable strain [16]. The material

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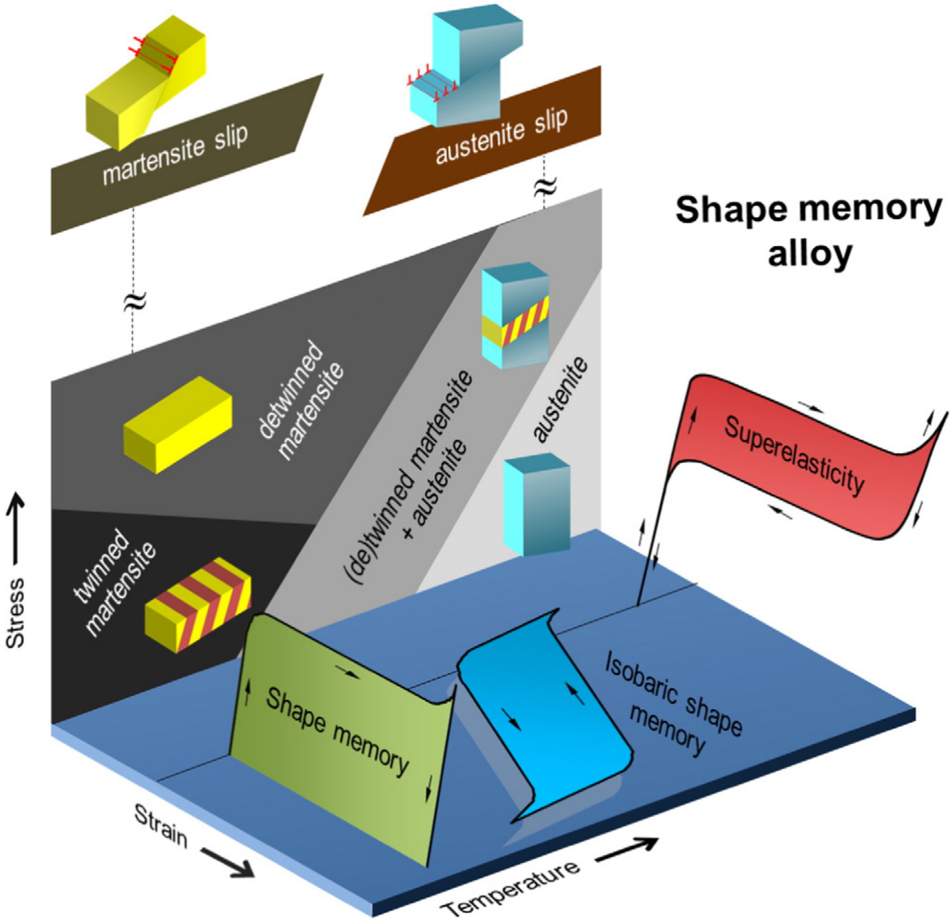

Fig. 1. General mechanical behaviors of SMAs in the stress/strain/temperature space and the associated structural changes.

vulnerability to slipping however becomes a predominant issue considering the fact that the other desired qualities start deteriorating as a result of excessive plastic flow. This essentially sets a high priority on the precise determination of resistance to slip on either experimental or theoretical ground.

Understanding the slip tendencies, for example, in new alloys has been achieved in the past via extensive experimentations based on a make-them-and-test-them philosophy. Such approaches are known to be slow and expensive, requiring a considerable allotment of labor and resources. Computational recipes have not been utilized to that end mainly because of the complexities of representing multiple elements and the challenges in modeling crystallographic nuances stemming from sub-atomic physics. Noting that the SMA attributes are fundamentally founded on the inherent electronic properties of the constituent elements, one needs to account for the complex interactions among various sub-lattice phenomena therein [17] (discussed in next section). One feasible solution is to extract fault energy landscapes, which can reveal the underlying activation barriers to be overcome in order to create a certain defect (e.g. dislocation). The proper framework to address such a problem is the usage of quantum mechanics based crystal descriptions [18,19], which have come of age as a powerful modeling platform to incorporate the sub-atomic effects.

3. Significance of discrete lattice effects

To comprehend the importance of atomistic contributions, let us first revisit the fundamental definition of a dislocation slip in an otherwise perfect crystal. It is created by means of a non-dilatational shear equal in magnitude to its Burgers vector. Slip would be spontaneous if the energy cost to create the corresponding shear displacement is minimal. The exact topography of the fault energy landscape would essentially be a function of: (a) the crystal lattice structure and (b) the alloy composition. Depending upon the crystallography of a certain alloy lattice, there exist very specific sets of planes and directions that offer the lowest energy barriers for shearing, which are typically the most densely packed ones. The effects of alloying are more complex, in that a multi-element lattice would have inhomogeneous bonding length/strength in three dimensions, giving rise to strong anisotropy.

Moreover, the most influential variables that would dictate the fault energetics are indispensably related with the underlying solid-state effects [20]. The end result of mixing diverse elements would be dictated by [21]: (i) the relative atomic volumes, (ii) the magnetic characteristics, (iii) the valence shell electronic configuration, and (iv) the short/long range atomic ordering. For example, if the solute atoms have significant volumetric mismatch, a strong distortion field would be created in the lattice, which would alter the inherent lattice response to external disturbances (e.g. applied stress or temperature variations). Consequently, the impedance to the slip would be directly proportional to the degree of solid solution effects. Similarly, the magnetic properties (as decided by the electronic spin) of individual elements would have profound contribution to the bonding landscape. Last but not the least is the role of valence electronic configurations of individual solute atoms. It is important to note that the SMAs consist of transition metals whose bonding characteristic is controlled by the condition of partial or full occupation of the outermost *d*-orbitals. In response to the mixing of various chemical species and the nature of applied stimuli (e.g. mechanical, magnetic or thermal driving forces), the quantum states of the valence electrons would be considerably altered, thus contributing to the inter-atomic forces [22]. The combined influences of these sub-atomic nuances would determine the inherent material predilection for

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permanent lattice deformation (manifested in the form of slipping). Evidently, such a problem entails modeling ability beyond the scope of continuum scale phenomenological laws as well as the measurement resolution of existing experimental techniques. The proper solution to such a problem, as has been demonstrated recently, lies at the use of the quantum mechanically based toolset such as the density functional theory (DFT).

DFT simulations are about seeking out the equilibrium lattice configuration by iteratively minimizing the total free energy of a given structure [18]. This is carried out numerically by solving the time-independent Schrodinger's equation within Born-Oppenheimer approximation, where electron densities replace wave functions for computational efficiency. The solutions of the electron densities are obtained by accounting for the electronic/magnetic attributes of individual chemical species (e.g. spin polarization, exchange-correlation effects). This allows for a general framework for accurately modeling defect energetics founded on the quantum forces. The reliability of DFT-extracted energetics is well scrutinized in the context of pure metals as reported earlier [23]. Such approaches can be extended to address more complex interactions underscoring the crystallographic behaviors of SMAs.

4. Establishing atomistic energy barrier

As pointed out earlier, the process of slip amounts to the shearing of a pristine crystal to create a permanent displacement as represented by the Burgers vector. This very fundamental yet physically intuitive definition can prove hugely beneficial in extracting the associated energy barrier from lattice level simulations. The transition period starting from the atomic structure about to be sheared to a fully dislocated one can be re-created both in time and space using DFT computations. Two crystals are rigidly displaced on a pre-defined plane and along a particular direction with angstrom scale resolution. The choice of the slip system in question can be rationalized via experimentally observed prevalence thereof. The associated energy versus shear displacement profile is expected to be of sigmoidal shape with peak(s) and saddle point(s). The peak energy magnitude is termed the *unstable stacking fault energy*, γ_{us}, (Fig. 2(a)) and essentially represents the barrier quantity to be overcome for initiating plastic deformation [24]. The primary challenge in measuring this metric by experiments is that the corresponding crystal structure is metastable and hence indiscernible during the actual deformation. In nature, we only witness the end result i.e. the presence of a stable dislocation, which has already surmounted the requisite γ_{us} level to have come into being. To that end, the ab-initio simulations permit a unique platform to freeze any unstable/metastable atomic structure both spatially and temporally, and conduct in-situ analysis thereof.

In DFT based calculations, one can electronically solve the total free energy of the displaced lattice (E) as well as the bulk (i.e. unsheared) one (E_{bulk}). The differential (E-E_{bulk}) normalized by the adjoining area gives the magnitude of the corresponding fault energies. In this manner, the entire spectrum of fault energy profile can be constructed addressing any transient crystal configurations, which are beyond the reaches of experimental determination. Knowing thus-constructed energetic landscape would enable one to precisely pinpoint the activation barriers, and hence form a basis to compare various slip systems.

5. Case studies

To exemplify, let us consider two case studies from literature in details: (a) NiTi austenite of cubic B2 lattice and (b) Ni_2FeGa martensite of tetragonal $L1_o$ structure. Fig. 2(a) demonstrates the most possible slip systems (i.e. a particular set of plane and direction) in NiTi austenite (as previously identified via electron microscopy). By displacing two crystal half-spaces relative to each other by one Burgers vector for any given plane and direction, the corresponding γ_{us} levels are determined [25]. In Fig. 2(a), the arrows indicate the slip direction and are colored according to their γ_{us} magnitudes. The slip systems are found to have the following energetic order: $\gamma_{us}^{(011)[0\bar{1}1]} > \gamma_{us}^{(001)[010]} > \gamma_{us}^{(\bar{2}11)[\bar{1}01]} > \gamma_{us}^{(011)[1\bar{1}1]} > \gamma_{us}^{(011)[001]}$. As it transpires, the (011)[001] system is most probable to occur while the $(011)[0\bar{1}1]$ being the least likely one. This trend reflects the prevalently observed slip systems in experiments. The underlying rationale for such preference can be established upon carefully scrutinizing the unstable lattice structures (i.e. corresponding to the highest energy level) from simulations as paused in time and space. The most favored slip system is discovered to be the one which achieves the shortest nearest neighbor distances by means of shear-induced displacement. That is to say, the favorable slip plane/direction corresponds to the crystal structure with highest degree of atomic packing along the entire pathway. This observation conforms to the well-known fact that the dense atomic arrangements foster slipping as widely evidenced in more conventional metallic lattices (e.g. in fcc pure metals).

Similarly, the preference of slip in SMA martensitic lattice can also be established. Fig. 2(b) presents such a scenario in Ni_2FeGa martensite, which comprises a tetragonal $L1_o$ lattice. Four slip systems are considered on the basis of their experimental observations. Their energy based comparison is as follows [26]: $\gamma_{us}^{(111)[\bar{1}01]} > \gamma_{us}^{(111)[\bar{1}10]} > \gamma_{us}^{(111)[\bar{2}11]} > \gamma_{us}^{(111)[11\bar{2}]}$. Upon shearing along individual directions by one full Burgers vector, various stable defects are created, namely, anti-phase boundary, complex stacking fault and superlattice intrinsic stacking fault. These planar defects as well as their relative prevalence in the $L1_o$ lattices have been verified through electron microscopy in the literature. The minimization of energy barriers is again rationalized in terms of achieving the most closed-packed atomic configurations as generated during incremental displacements. The takeaway message from these predictions is that the proposed computational recipe is essentially generic in nature. They can be employed to address any lattice type irrespective of the alloy constituents and the composition. The predictions serve to pinpoint which slip systems are favored during plastic deformation.

Fig. 3 presents an overview of the most important SMAs being actively researched today with a view to explore a synergy of desired properties. The experimentally determined transformation strain (i.e. the total strain minus the elastic and plastic strain), transformation temperature (i.e. the range of temperature over which phase transformation occurs) and slip resistance are cross-referenced for a better perspective on their mechanical properties. This kind of database is very helpful for a modern design engineer to carry out materials selection slated for certain applications. It should be noted that establishing such a condensed digest of material traits requires exhaustive sets of experimentations over a long period of time. On the other hand, the predicted γ_{us} (normalized by the magnitudes of the Burgers vectors) for the most likely slip systems to be activated are put side-by-side [25–27] with the experimental results. It follows that the predictions (representing the lattice shear resistance) can serve as a similar materials map, which appears to be remarkably in tune with the empirical trends for the case studies hitherto undertaken. Of course, one needs more material simulations in this regard to firmly establish the theoretical trend. Further substantiation as well as population of the theoretical map with additional SMA data remains a promising future endeavor to pursue.

6. Concluding remarks and future research

It should be noted that the afore-discussed energetic considerations are computed in pristine lattice settings. In real-life microstructure, the presence of interfaces, texture, micro-cracks, precipitates etc. would give rise to complex deformation scenario, which would require a more comprehensive modeling framework. Understanding slip propensity can

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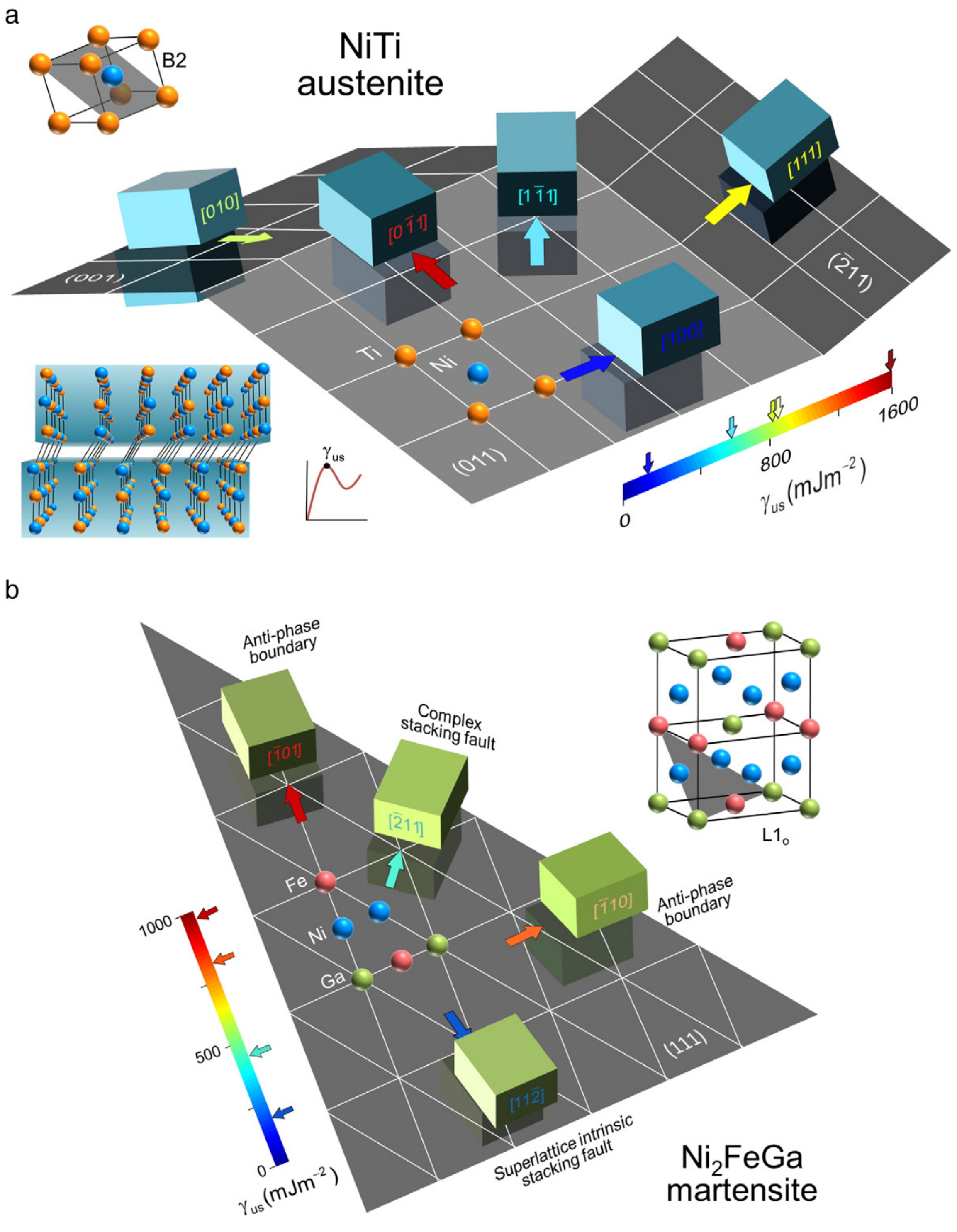

Fig. 2. (a) — Shear simulation of a Burgers vector for various slip systems in NiTi austenite consisting of B2 lattice. The energetically based order of the slip system is found to be $\gamma_{us}^{(011)[0\bar{1}1]} > \gamma_{us}^{(001)[010]} > \gamma_{us}^{(\bar{2}11)[\bar{1}01]} > \gamma_{us}^{(011)[1\bar{1}1]} > \gamma_{us}^{(011)[001]}$. (b) — Slip systems and energy barriers to slip in L1o martensite in Ni_2FeGa lattice. The order of slip systems in terms of the energy barrier is: $\gamma_{us}^{(111)[\bar{1}01]} > \gamma_{us}^{(111)[\bar{1}10]} > \gamma_{us}^{(111)[\bar{2}11]} > \gamma_{us}^{(111)[11\bar{2}]}$.

serve as an important first step towards theorizing real-life damage behaviors [28,29]. Dislocation emission would be facilitated given any presence of locally stressed sites. For instance, in a polycrystalline environment, the material boundaries may act as either obstacles to plastic flow or the sources thereof depending upon the local stress state and the geometry. As a result of interactions among independent plastic systems, residual strain fields will be generated. The energetic descriptions thereof can be performed in terms of the extrinsic values of γ_{us} (i.e. representing restricted glide due to the presence of, say, boundaries) [30]. More promisingly, there are ongoing researches directed at extracting the corresponding lattice frictional stresses using the fault energies to be used in continuum frameworks, feasibility of which has been demonstrated in conventional alloys. Such predictions can directly be compared with experimental damage metrics as well [31]. Due to

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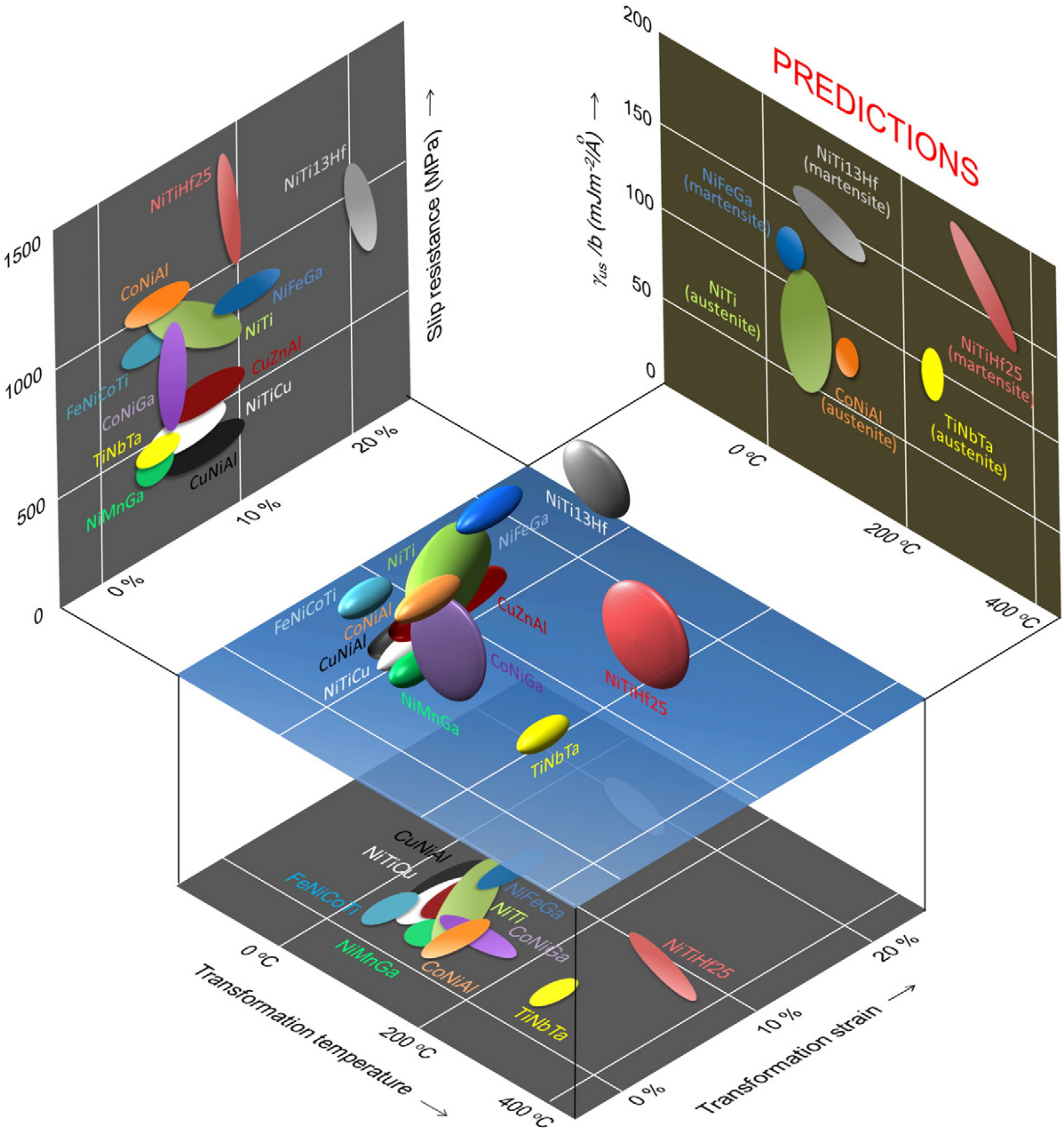

Fig. 3. Comparison among SMAs in terms of transformation strain, temperature and slip resistance (experimental). The theoretical calculation of shear energy barriers (normalized by the Burgers vector magnitude, b) is plotted against the experimental transformation temperature. (Note that the 3D balloon-like representations are constructed by projecting the known experimental data; as a result one can make a direct visual comparison with predictions).

the generality of these approaches, the understanding of plastic micromechanism(s) in the ordered crystals of SMAs can also benefit considerably from there.

Currently, one of the outstanding issues remains in terms of characterizing fatigue and fracture tolerances of SMAs [32]. In particular, we are still in need of comprehensive literature both on experimental and theoretical grounds to shed further light on the damage mechanisms of SMAs as only few works exist on the topic [33–35]. Understanding fatigue/fracture in SMAs require special modeling platform due to the fact that the near-tip damaging phenomena would encompass a complex blend of slip and residual transformed material both ahead and behind the crack. Unlike the conventional material, where plasticity defines the cracking process [36, 37], the SMAs need a precise demarcation of processes. For example, the relative contributions of slip, twinning/de-twinning etc. and their mutual interactions would control the dynamics of damage propagation at the microscopic length scale. Establishing the predominant deformation mode can be particularly beneficial especially from an energetic perspective. It is well known today that the cracking occurs as a direct consequence of widespread slip irreversibilities [38,39], which is a strong function of alloy chemistry, microstructure and lattice shear resistance. The use of material-specific energy barriers may facilitate the developments of mechanistic models, leading to the quantification of microscale cracking processes. To that end, the energetic concepts are already being pursued and tested in conventional materials, which has shown promising results [40]. It remains to be seen how these progresses in the computational materials science frontiers would further a better understanding of the SMA behaviors. Similarly, the presence of precipitates (coherent, semi-coherent or incoherent) is known to drastically affect the SMA thermomechanical attributes as local triggering sites of phase transformation as well as slip. Utilizing DFT, one can predict the precipitate lattice properties (e.g. lattice constants, elastic moduli) [41], thus identifying the sources of incompatibility with the surrounding matrix. These discoveries essentially pave the way for developing larger-scale mesoscale models of SMAs, using curve-fitted pair-potentials. For example, molecular dynamics based simulations have recently been employed [42–44] to study the phase transformation and the associated constitutive behaviors in defect-free as well as precipitate lattices. It was found that the precipitates indeed influenced the local transformation behavior. Associated with oriented precipitates, favorable martensite variants were activated. It remains to be seen how successful use of mesoscale simulation tools would also uncover the of role of precipitates on the slip phenomena.

Another potential use of the atomistic theories may transpire, as we envision, in the form of suggesting novel SMA crystals with superior mechanical attributes. We have evidenced how the close packing of atoms has been unequivocally related with the minimization of slip energy barriers via the simulations. On a hypothetical ground, one can concoct various lattice structures to examine the likely augmentation of plastic deformation resistance. Such endeavors would allow one to explore the ideal territories of property enhancement. On the basis of hypothetical structure and composition, the next logical undertaking would be to check the feasibility of manufacturing. Instead of solely relying on experimental trial-and-error based approaches to accomplish the next breakthrough, one can utilize the theoretical mixing as an educated basis for fine-tuning the empirical optimization trials. Given the fact the SMAs possess ordered lattice, configuring an existing lattice structure (e.g. $L1_2$, $L1_o$) with an assortment of transition metals may reveal invaluable insights. As has already been witnessed through the combined atomistic-continuum modeling [30], the effect of apparently minute modifications at the lattice level can reverberate across the physical length scales, amplify in extent, and eventually alter the macroscopic mechanical attributes significantly. To conclude, the quantum mechanically based theoretical procedure can be an important first step in the grand scheme of advancing new improved shape memory materials.

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