

Niobium hydride formation in superconducting qubit thin films

Tyler J. Leibengood^{1,*}, Pierre-Clément A. Simon², P. Graham Pritchard,¹
James M. Rondinelli¹, and Peter W. Voorhees^{1,†}

¹Department of Materials Science and Engineering, *Northwestern University*, Evanston, Illinois 60208, USA

²Computational Mechanics and Materials Department, *Idaho National Laboratory*, Idaho Falls, Idaho 83415, USA



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The formation of nonsuperconducting hydrides in 160–170-nm-thick films of niobium is examined. We identify six elastically distinct orientation relationships ϵ -Nb₄H₃ takes within the matrix, solid-solution α -NbH_x phase. We employ a phase field model to assess the impact of elastic energy induced by the strain of phase transformation on the morphology and transformation dynamics of ϵ precipitates within a thin film. We consider the dimensions of the thin film, crystallographic growth direction, and diffusion rates to predict the timescale of hydride evolution. Leveraging the finite element method, we predict two-dimensional and three-dimensional equilibrium shapes of ϵ -hydrides within a bulk sample and in a thin film that has a traction-free surface. Our results suggest that niobium hydrides migrate to the free surface of the film. Precipitates which reach the free surface coarsen, while precipitates within the film dissolve. Precipitates in both two dimensions and three dimensions experience a repulsive interaction force at the free surface, that is attractive in the bulk, shown in experiment and theory of previous studies.

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I. INTRODUCTION

Through various advances in quantum computing technology, elemental niobium and tantalum have been targeted for development and refinement as primary constituent materials in superconducting qubits [1–4]. Niobium has successfully maintained a position as the flagship superconducting material in other quantum technologies, such as superconducting radio-frequency (SRF) cavities. Impurities are often introduced during the fabrication of niobium SRF cavities and must later be removed. One such impurity, which produces nonsuperconducting precipitates at cryogenic operating temperatures, is hydrogen. Various phases of the Nb-H system are known and well studied [5], but the niobium hydride of interest which emerges at residual concentrations of hydrogen and at the critical superconducting temperature of niobium, $T_c = 9.25$ K, is the ϵ -Nb₄H₃ phase.

The pollution of SRF cavities with hydrogen greatly suppresses the superconducting transition temperature and increases the surface resistivity of bulk niobium cavities with a correspondingly massive drop in the cavity quality factor (Q). This condition is colloquially referred to as “Q disease” [6]. Understandably, great efforts have been made to study the sources which introduce hydrogen into the system and methods to remove it [7–9]. The most effective and widely accepted method of hydrogen removal is high-temperature

annealing. Though this procedure is successful in treating Q disease in macroscopic SRF cavities, such a treatment performed on a transmon qubit with nanoscopic features would ruin the circuit. Accordingly, several studies have been performed over the past decade to characterize the effect of hydrogen on the performance of thin-film niobium qubits and microwave resonators and devise suitable treatments to remove hydrogen from the devices [1,7,10–12].

There is a long characterization history of the niobium-hydrogen system [13,14]. Near room temperature, the two primary stable hydride phases are α -NbH_x and β -Nb₄H₃. The α phase is stable up to 1–10 at. % hydrogen depending on the temperature. At higher hydrogen concentrations, a two-phase mixture of the α and β phases is present, up to ≈ 43 at. % hydrogen. In the α phase, hydrogen randomly occupies the tetrahedral sites of the body-centered cubic (bcc) niobium lattice, whereas a predominantly ordered occupation is present in the β phase. At the millikelvin operating temperature of qubits and microwave resonators, the β phase transforms into the ϵ phase which possesses a modified ordering of hydrogen at tetrahedral sites [5,15]. In all phases, the underlying niobium lattice dilates with increasing hydrogen saturation.

Studies have shown that in niobium thin films, there are critical hydrogen concentrations and film thicknesses that dictate the microstructure of β -hydrides [11,12]. These studies are focused on progressive loading of niobium films of constant film thickness with hydrogen until an observable transition can be measured. Each of these studies observes coherent to semicoherent transitions at lower concentrations for larger film thicknesses. Critical radii for cylinders of β -hydride that extend through the height of the film are reported by Nörthemann and Pundt. Wagner *et al.* also report large regions of hydride that create plastic deformation. These are accurate snapshots of the final configuration that hydrides

*Contact author: tjleibengood42@gmail.com

†Contact author: p-voorhees@northwestern.edu

take, but lack insight about the early microstructure. Second, recent work on Nb thin films has shown that the degree of hydrogen loading in qubit thin-film samples is a function of both the choice of etchant and the duration of chemical treatment [7]. Introduction of hydrogen is closely tied to processes that remove lossy niobium oxides which form on the niobium film surface, as recently reported by Bal *et al.* [1].

Niobium hydrides are effectively characterized in bulk systems and at room temperature. However, there is a knowledge gap in early niobium hydride morphology and evolution at the nanoscale. Microscopic dendrites and platelets of β -hydrides have been observed as early as 1976 [14]. The strain accompanying the phase transformation is due to the misfit strain, or the difference in lattice parameters between the hydride and niobium. The magnitude of this misfit strain is significant, causing niobium hydrides to lose coherency at length scales on the order of 10 nm [11,16]. Hence, microscopic precipitates produce many dislocations from this misfit and plastically deform the surrounding Nb lattice. Elastic stress has been shown to cause morphological anisotropy in coherent misfitting precipitates [17–21]. Furthermore, misfitting precipitates interact elastically with structures such as point defects, line defects, other precipitates, or plane defects like the thin-film free surface. Because the surface of a niobium thin film is stress free, and the film can be elastically constrained by the substrate, it is natural to expect the dynamics of early hydride nuclei to be affected by the presence of the film surface.

In this work, we present a phase field model which includes an elastic energy contribution to the free energy functional to investigate the impact of heterogeneous stiffness, misfit strain, and the presence of a traction-free surface on the morphology and evolution of nanoscale ε -Nb₄H₃ precipitates. We consider the various orientation relationships that the ε phase may take in an α single crystal. Our results provide a picture of how hydrides grow and migrate to reduce the total energy of the system before reaching late stages of coarsening.

II. PHASE FIELD MODEL OF THE α -NbH_x/ ε -Nb₄H₃ SYSTEM

In the phase field method, field variables that describe the underlying microstructure evolve in space and time to reduce the total free energy of a system. While models which consider only the chemical free energy and interfacial free energy are well studied, the influence of elastic energy has not been due to the increased complexity of such models [22–24]. As mentioned, misfit strain cannot be neglected in constructing a model for the solid-solution α to ε -Nb₄H₃ phase transition, as it induces considerable amounts of elastic energy. Consequently, the possible orientation relationships of α -NbH_x and ε -Nb₄H₃ are needed to correctly incorporate the elastic energy into the free energy functional. Our treatment of the chemical free energy and interfacial free energy follows that which was performed by Kim, Kim, and Suzuki (KKS) [22], where the interfacial and bulk free energies are decoupled. This decoupling allows one to choose an interface width independently of the bulk and interfacial energies, which saves computational cost. Simon used a multiphase model of

a binary, metal-hydrogen system [17], the foundation from which our model was based upon.

A. Phase field model formulation

The microstructure of our model of the α -NbH_x/ ε -Nb₄H₃ system is described by field variables for the concentration and order parameters. The concentration field, c , represents the atomic fraction of hydrogen, and the order parameters, η_ν , represent the phases present and the variants of the ε phase, ν ($\nu = \alpha, \varepsilon_i$). The order parameters range from 0 to 1, where 0 indicates an absence and 1 indicates the presence of the phase variant associated with that order parameter. The order parameters and concentration field transition continuously between values across different phases through the diffuse interface.

We defined three order parameters: one for the matrix, α phase, η_α , and two for variants of the ε phase, η_{ε_1} and η_{ε_2} . The free energy functional which describes the total free energy of a given system is defined as

$$F_{\text{tot}} = \int_{\Omega} f_{\text{tot}} d\Omega = \int_{\Omega} (f_{\text{chem}}(c, \vec{\eta}) + f_{\text{int}}(\vec{\eta}) + f_{\text{el}}(\vec{\eta})) d\Omega, \quad (1)$$

where F_{tot} is the total free energy, f_{tot} is the local total free energy density, f_{chem} is the local chemical free energy density, f_{int} is the local interfacial free energy contribution, f_{el} is the local elastic energy density, and $\vec{\eta} = [\eta_\alpha, \eta_{\varepsilon_1}, \eta_{\varepsilon_2}]^T$.

The total hydrogen concentration, c , as derived by KKS [22], is a weighted, interpolated average of the concentrations in each respective phase, c_α and c_{η_i} ($i = 1, 2$), given as

$$c = \sum_{\nu} h_{\nu}(\vec{\eta}) c_{\nu}, \quad (2)$$

where h_{ν} is the interpolation function for phase ν . Key properties of the interpolation function include $h_{\nu}(\eta_{\nu} = 0) = 0$, $h_{\nu}(\eta_{\nu} = 1) = 1$, $dh_{\nu}/d\eta_{\nu}(\eta_{\nu} = 0) = 0$, and $dh_{\nu}/d\eta_{\nu}(\eta_{\nu} = 1) = 0$. The generalized formula for the interpolation function of any number of phases as used in our model is presented by Moelans [25] as

$$h_{\nu}(\vec{\eta}) = \frac{\eta_{\nu}^2}{\sum_{\nu'} \eta_{\nu'}^2}. \quad (3)$$

Because $\sum_{\nu} h_{\nu} = 1$, h_{ν} may be thought of as a phase fraction. Consequently, Eq. (2) may be generalized to interpolate any material property, including linear elastic strains, stresses, and stiffnesses as well as chemical energies. The concentrations of each phase at the interface are approximated to be at equilibrium [22], which subjects the solution to the following constraint:

$$\mu = \frac{\delta f_{\text{tot}}}{\delta c_{\nu}} = \frac{\delta f_{\text{tot}}}{\delta c_{\nu'}}, \quad \nu, \nu' \in \{\alpha, \varepsilon_i\}, \quad (4)$$

where μ is the chemical potential.

The field variables evolve as described by the Cahn-Hilliard and Allen-Cahn equations [22,23],

$$\frac{\delta c}{\delta t} = \nabla \cdot M \nabla \left(\frac{\delta F_{\text{tot}}}{\delta c} \right), \quad (5)$$

$$\frac{\delta\eta_v}{\delta t} = -L \left(\frac{\delta F_{\text{tot}}}{\delta\eta_v} \right), \quad (6)$$

where M and L are related to the hydrogen diffusion coefficient and the interface mobility, respectively. The definitions of these constants are discussed in Sec. II B.

1. Chemical free energy

The chemical free energy is described as a difference of formation energies between phases. Thermodynamic parameters that might provide approximations of the formation energies of the α and ε phases are unavailable at low temperatures, where the ε phase forms. Without said parameters, we make a series of assumptions to create chemical free energy functions.

The phase diagram of the Nb-H system as compiled by Okamoto shows that the β -to- ε phase transition occurs at $T_i \approx 203$ K [5]. For reasons explained in Sec. II B, the temperature of our simulations is assumed to be constant in space and time at the transition temperature of ε formation, with $T = T_i = 203$ K. Under the assumption that shifts of the equilibrium concentration field will be small, Taylor expansions of order 2 centered on each equilibrium concentration are used:

$$f_{\text{chem}}^v = \frac{1}{2}\lambda_{v,2}(c_v - c_v^{\text{eq}})^2 + \lambda_{v,1}(c_v - c_v^{\text{eq}}) + \lambda_{v,0}, \quad (7)$$

where $\lambda_{v,i}$ ($i = 0, 1, 2$) are the Taylor series coefficients and c_v^{eq} is the equilibrium concentration for phase v .

Because the chemical free energy of the α/ε system is not well characterized, we may simplify the model and nondimensionalize the equilibrium compositions of each phase to arbitrary constants, which is a common practice in phase field simulation. As such, the equilibrium concentrations and common tangents are set to $c_{\alpha}^{\text{eq}} = 0.1$ and $c_{\varepsilon}^{\text{eq}} = 0.9$. Because the common tangent line may be subtracted from both of the chemical free energies without affecting the dynamics of the system, the chemical free energies become

$$\begin{aligned} f_{\text{chem}}^v &= \frac{1}{2}\lambda_{v,2}(c_v - c_v^{\text{eq}})^2 + \lambda_{v,1}(c_v - c_v^{\text{eq}}) + \lambda_{v,0} \\ &\quad - (\lambda_{v,1}(c_v - c_v^{\text{eq}}) + \lambda_{v,0}) \\ &= \frac{1}{2}\lambda_{v,2}(c_v - c_v^{\text{eq}})^2. \end{aligned} \quad (8)$$

Since our study primarily focuses on the effect of size and elastic stress on the calculated hydride morphologies and elastic interactions, we will not focus on an accurate prediction of the final concentration field since this requires more accurate chemical free energy models than those employed herein. The curvatures of the chemical free energies at the common tangents are taken to be equal and of a similar magnitude to the CALPHAD computed curvatures in the Zr-H system [17] with $\lambda_{\alpha,2} = \lambda_{\varepsilon,2} = 200$ eV/ c_H^2 . Should accurate curvatures of the chemical energies become available at a future date, shifts in the concentration field may be rescaled with the following relation:

$$\Delta\tilde{c}_v = \Delta c_v \frac{\lambda_{v,2}}{\tilde{\lambda}_{v,2}}, \quad (9)$$

where Δc_v is the shift away from c_v^{eq} due to Gibbs-Thompson-type effects and $\tilde{\lambda}_{v,2}$ is the hypothetical accurate curvature of

phase v at the common tangent. As the chemical free energy of the α/ε system becomes well characterized, these simplifying assumptions should be challenged, as they potentially affect the quantitative predictions of this study.

The total chemical free energy is then calculated with following sum:

$$f_{\text{chem}}(c, \tilde{\eta}) = \sum_v h_v(\tilde{\eta}) f_{\text{chem}}^v(c_v). \quad (10)$$

As the chemical free energy of the α/ε system becomes well characterized, these simplifying assumptions can be altered.

2. Interfacial free energy

The interfacial free energy contribution to the total energy is

$$f_{\text{int}}(\tilde{\eta}) = wg(\tilde{\eta}) + \frac{\kappa}{2} \sum_v \|\nabla\eta_v\|^2, \quad (11)$$

where κ is the gradient energy coefficient [22] and g is a double-well potential of height w defined as [25]

$$g(\tilde{\eta}) = \sum_v \left(\frac{\eta_v^4}{4} - \frac{\eta_v^2}{2} \right) + \sum_v \left[\sum_{v' \neq v} \frac{\gamma_{vv'}}{2} \eta_v^2 \eta_{v'}^2 \right] + \frac{1}{4} \quad (12)$$

and $\gamma_{vv'} = 1.5$ so the well is symmetric between every phase [24]. The prescribed interface width, l , and interfacial energy density, γ , define the gradient energy coefficient κ , and the double-well potential height, w , is given in the following relations [17,22]:

$$\kappa = \frac{3}{4.4} \gamma l, \quad (13)$$

$$w = 6.6 \frac{\gamma}{l}. \quad (14)$$

To resolve features on the order of 10 nm, the interfacial length was set to 1 nm. The interfacial energy between the α phase and ε phase or between variants of the ε phase is currently unknown. For this reason we assume the interfacial energy is isotropic and the same value as proposed for α -Zr/ δ -ZrH_{1.66} as $\gamma = 1.2483$ eV/nm² (200 mJ/m²) [17]. Generally speaking, metal solid-solid interfaces have interfacial energies in the range 0.1–1 J/m² = 0.6242–6.242 eV/nm².

3. Elastic energy

The total elastic stiffness tensor is defined by an interpolation between each phase using the following equation:

$$C_{ijkl} = \sum_v h_v(\tilde{\eta}) C_{ijkl,v}, \quad (15)$$

where $C_{ijkl,v}$ are the stiffness tensors for each phase and phase variant. The total strain, $\epsilon_{ij}^{\text{tot}}$, is defined to be a sum of the strain due to elastic deformation, $\epsilon_{ij}^{\text{el}}$, and the strain of phase transformation, or misfit strain, ϵ_{ij}^* : $\epsilon_{ij}^{\text{tot}} = \epsilon_{ij}^{\text{el}} + \epsilon_{ij}^*$. Since the misfit strain is a function of the phase, it is interpolated using the switching function, h , as

$$\epsilon_{ij}^* = \sum_v h_v(\tilde{\eta}) \epsilon_{ij,v}^*. \quad (16)$$

TABLE I. Stiffness tensor for α -NbH_x/ ϵ -Nb₄H₃ (GPa).

Stiffness tensor	α -NbH _x (4.2 K)	ϵ -Nb ₄ H ₃ (DFT, 0 K)
C_{11}	252.7	280
C_{22}		274
C_{33}		248
C_{12}	133.2	179
C_{13}		117
C_{23}		125
C_{44}	30.97	53.9
C_{55}		43.2
C_{66}		65.2

The stress-free reference state from which other strains will be measured is the hydrogen-free α phase at 203 K. Though the lattice parameters of the α phase are known to change as functions of temperature and concentration, those effects are neglected, as explained in the Appendix. Consequently, the misfit strain tensor for the α phase must then be identically zero, $\epsilon_{ij,\alpha}^* = \mathbf{0}$, and does not contribute to the sum in Eq. (16). Rearranging the total strain expression using Eq. (16) gives the elastic strain field:

$$\epsilon_{ij}^{el} = \epsilon_{ij}^{\text{tot}} - \sum_v h_v(\vec{\eta}) \epsilon_{ij,v}^*. \quad (17)$$

The misfit strain tensor of the ϵ phase is dependent on its orientation relationship with the α phase. Misfit tensors for each variant may be found in Eqs. (A3) and (A28)–(A32).

The stiffness tensor field and elastic strain fields are calculated before solving the stress field, which is then followed by the elastic energy density. The stress, σ_{ij} , defined in linear elasticity theory is

$$\sigma_{ij} = C_{ijkl} \epsilon_{kl}^{el}, \quad (18)$$

and the elastic energy density is

$$f_{el} = \frac{1}{2} \sigma_{ij} \epsilon_{ij}^{el} = \frac{1}{2} C_{ijkl} \epsilon_{ij}^{el} \epsilon_{kl}^{el}, \quad (19)$$

where sums over repeated indices from 1 to 3 are assumed.

The stiffness tensor of the α phase is available for a wide range of temperatures and hydrogen concentrations [26,27]. To simplify the coupling of the elastic energy component to the evolution equations, we assume that the elastic energy is not a function of concentration, and instead only the order parameter. Elastic stiffness data of the ϵ phase are unknown, so they are calculated using density functional theory (DFT) as described below. The consequence of calculating the stiffness tensor from first principles is that the resulting stiffness tensor is valid at 0 K. To maintain consistency between the same material property between phases, we choose the stiffness tensor for the α phase to be as near to 0 K as is known, 4.2 K. Because the α phase has cubic symmetry and the ϵ phase is orthorhombic, the stiffness tensors have three and nine unique components, respectively, listed in Table I.

B. Kinetic parameters

To capture physical rates of field evolution, the hydrogen mobility constant in the KKS model [17,22] is related to the

isotropic hydrogen diffusion coefficient, D , as

$$M = \frac{D}{f_{cc}}, \quad (20)$$

where $D = 1.69 \times 10^{-6} \text{ cm}^2/\text{s} = 1.69 \times 10^8 \text{ nm}^2/\text{s}$ at $T = 203 \text{ K}$ [28]. And f_{cc} is related to the second derivatives of the free energies of α and ϵ [22]. Using the free energies given in Eq. (8), and since f_{cc} of each of the phases are equal,

$$f_{cc} = \frac{\prod_v \frac{\partial^2 f_{\text{chem}}^v}{\partial c_v^2}}{\sum_v h_v(\vec{\eta}) \frac{\partial^2 f_{\text{chem}}^v}{\partial c_v^2}} = \frac{200}{h_\alpha + h_\epsilon}. \quad (21)$$

Hydride precipitation in niobium has been shown to be diffusion controlled, much like other M-H systems at low temperature [9,17]. Because the system is diffusion controlled, the interface mobility, L , is set equal to the hydrogen mobility constant, M [17].

The timescale, t , at which processes will take place over a characteristic distance, d , is

$$t = \frac{d^2}{D}. \quad (22)$$

Using the diffusion coefficient at 203 K given previously, for $d = 100 \text{ nm}$ the characteristic timescale is 0.06 ms. Thus, the system will reach equilibrium in milliseconds. In addition, the cooling process of a qubit does not outpace the rate at which hydrides evolve along the thickness of the film. Thus, it is valid to keep the temperature constant in our model.

III. DENSITY FUNCTIONAL THEORY CALCULATIONS

We performed electronic structure calculations based on density function theory (DFT) to obtain the elastic tensor for ϵ -Nb₄H₃ (Table II) using the Vienna Ab initio Simulation Package (VASP) [29,30]. The elastic tensor is obtained by simulating the stress response to six independent strains, including the effects of ionic relaxations. DFT simulations were performed using the Perdew-Burke-Ernzerhof (PBE) generalized gradient approximation (GGA) functional with projector augmented wave (PAW) pseudopotentials to represent core and valence electrons with the following electron configuration: $4s^2 4p^6 5s^1 4d^4$ (Nb) and $1s^1$ (H) [31]. We note that the PBE functional has previously been used to accurately calculate the elastic tensor of bcc Nb [32]. An energy cutoff of 800 eV and a $8 \times 8 \times 24$ Γ -centered k -point mesh were used. Before computing the elastic tensor, the ϵ -Nb₄H₃ structure was relaxed to a force tolerance of $10^{-1} \text{ meV } \text{\AA}^{-1}$. The relaxed lattice constants were $a = 9.755 \text{ \AA}$, $b = 9.626 \text{ \AA}$, and $c = 3.443 \text{ \AA}$. These agree well with the experimentally reported lattice constants: $a = 9.724 \text{ \AA}$, $b = 9.600 \text{ \AA}$, and $c = 3.421 \text{ \AA}$ [15].

IV. NUMERICAL METHOD

The finite element method in the Multiphysics Object-Oriented Simulation Environment (MOOSE) is used to perform the simulations [33–35]. To reduce the computational resources and time, adaptive mesh and adaptive time-stepping schemes were implemented. The size of the elements depends on the simulation and ranges from 10 to 40 nm. The mesh is

TABLE II. The aspect ratios of two-dimensional particles and the area ratio of precipitating ε -Nb₄H₃ may be approximated with the linear regression slopes listed here.

Stimulus	ψ	$d\psi/d\sigma$ (1×10^{-6} MPa ⁻¹)
Tension in $[\bar{1}10]_\alpha$	Phase boundary	-2.313
Compression in $[\bar{1}10]_\alpha$	Phase boundary	2.358
Tension in $[110]_\alpha$	Phase boundary	-2.558
Compression in $[110]_\alpha$	Phase boundary	2.580
Applied axial stress in $[\bar{1}10]_\alpha$	Aspect ratio	-796.9
Applied axial stress in $[110]_\alpha$	Aspect ratio	979.6

initialized with a minimum element size for about six element widths per interface width. A typical calculation involved 10^5 – 10^6 degrees of freedom that can also vary with time during a calculation.

Nuclei are inserted as circles or spheres at a specified position, radius, diffuse interface width, and initial concentration depending on the simulation. Nuclei throughout this work are initialized with $\eta = 1$ on the interior, $\eta = 0$ on the exterior, and with diffuse interfaces of $l = 1$ nm width in the η and c fields. c_H^α is initialized at some supersaturation above equilibrium and c_H^ε is 0.91061 in every simulation.

The boundary conditions were either set to periodic, zero traction, or displacement fixed depending on the desired effect. In most simulations, periodic boundaries were applied in the $\hat{x}$ and $\hat{z}$ directions and zero traction was applied to the boundaries in $\hat{y}$ for the coordinates specified in Eq. (23). In certain cases, symmetry was utilized so the simulation domain size was reduced to either 1/4 or 1/8 of the true domain size to reduce computational costs. In simulations in which these mirror symmetries were exploited, zero displacement in the direction of the mirror plane normal, $\vec{u} \cdot \hat{n}_{\text{mir}} = 0$, was applied. Zero flux conditions are applied on nonperiodic boundaries.

V. RESULTS

A. Two-dimensional precipitates in the bulk

To understand the equilibrium shape of an ε -phase precipitate in the absence of external factors, single precipitates were simulated in two dimensions. We chose the simulated plane to be a cross section of the single-crystal thin film with the build direction $[\bar{1}10]_\alpha$ pointing in $\hat{y}$ and $[001]_\alpha$ pointing in $\hat{z}$. For this choice of orientation, the principal axes of variants 1 and 2 are aligned along the coordinate system of the simulation environment:

$$\begin{aligned}\hat{x} & \parallel [100]_{1,\varepsilon} \parallel [010]_{2,\varepsilon} \parallel [110]_\alpha \\ \hat{y} & \parallel [010]_{1,\varepsilon} \parallel [\bar{1}00]_{2,\varepsilon} \parallel [\bar{1}10]_\alpha \\ \hat{z} & \parallel [001]_{1,\varepsilon} \parallel [001]_{2,\varepsilon} \parallel [001]_\alpha,\end{aligned}\quad (23)$$

where "1, ε " denotes variant 1 of the ε phase, and "2, ε " denotes variant 2 of the ε phase. Note that the axes of variant 1 are interchangeable with the simulation axes and will be used as the reference coordinate system of choice moving forward.

Since variant 1 and variant 2 are identical by a rotation of 90° , variant 2 was chosen to represent the resulting single-precipitate morphology in the $[001]_{1,\varepsilon}$ plane. A length scale of each precipitate is either $L = \sqrt{A_\varepsilon}$ in two dimensions or

$L = \sqrt[3]{V_\varepsilon}$ in three dimensions, where A_ε is the area of a two-dimensional (2D) ε -phase precipitate and V_ε is the volume of a three-dimensional (3D) ε -phase precipitate. Pictured in Fig. 1 are the equilibrium shapes for length scales $L = 10.25$, 19.65, and 32.01 nm. The interfacial energy is isotropic, and thus any anisotropy in the shape of the hydride is due to the anisotropic elastic field. Since the effects of elastic energy scale with the area or volume of the particle, the larger particles exhibit more anisotropic shapes.

The resulting stress field engendered by a single precipitate may be used to predict the evolution of itself and some other precipitate. The elastic interaction energy, $f_{el,v}^{\text{int}}$, is given by [20]

$$f_{el,v}^{\text{int}} = -\sigma_{ij}\epsilon_{ij,v}^*. \quad (24)$$

This interaction energy gives rise to a configurational force on an interface, $\vec{F}_c = f_{el,v}^{\text{int}}\vec{n}$, where $\vec{n}$ is the normal to the interface pointing into the particle. The configurational force acts to move an interface in a material via local changes in interfacial composition that induce diffusion, or through a driving force at an interface. Thus, the elastic interaction energy can be divided into regions of repulsion and attraction, where $f_{el,v}^{\text{int}}$ is positive and negative, respectively, and v is the variant of the second precipitate at this location.

Figure 2 shows interaction fields for single, two-dimensional precipitates of $L = 10.25$ and 32.01 nm of the same variant. Large wells of attraction are apparent for both precipitates on the right and left. Regions of attraction are also seen above and below each precipitate, but more distant. Regions of repulsion extend in the $\langle 100 \rangle_\alpha$ directions, connected on the tops and bottoms of the precipitate. The regions of

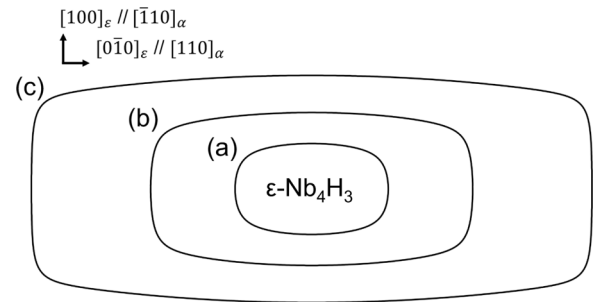


FIG. 1. Equilibrium shapes of two-dimensional, variant 2 precipitates of ε -Nb₄H₃ of length scales (a) $L = 10.25$ nm, (b) $L = 19.65$ nm, and (c) $L = 32.01$ nm.

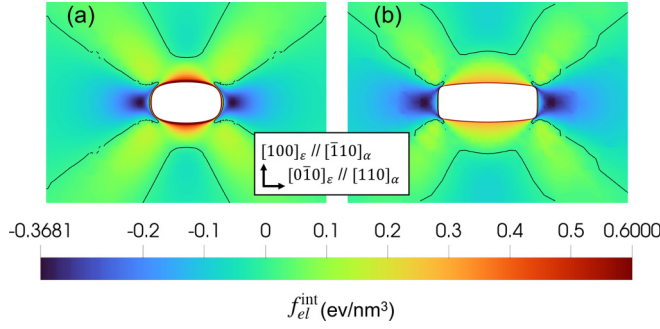


FIG. 2. Calculated interaction energies of ε variant 2 with ε variant 2 of length scales (a) $L = 10.25$ nm and (b) $L = 32.01$ nm. The black line indicates a contour of $f_{el}^{int} = 0$.

attraction and repulsion are separated by a contour of $f_{el}^{int} = 0$, shown in black.

A variant-2 precipitate was subject to various uniaxial external stresses in the directions of $[\bar{1}10]_{\alpha,2}$ and $[110]_{\alpha,2}$. The stresses range from -600 to 600 MPa in both directions. The resulting change in precipitate shape is shown in Fig. 3. The application of external stress causes the free energy of the ε phase to shift, to make hydride precipitation more favorable or less favorable depending on the sign of the stress. This shift can be tracked by calculating the normalized concentration, in atomic percent hydrogen, of the α phase at equilibrium. Thus, we can observe a change in the phase diagram boundary for a constant temperature as a function of applied external stress. The normalized concentration of the α phase is defined as

$$\bar{c}_H^\alpha = \frac{c_\alpha - c_\alpha^{eq}}{c_\varepsilon^{eq} - c_\alpha^{eq}}. \quad (25)$$

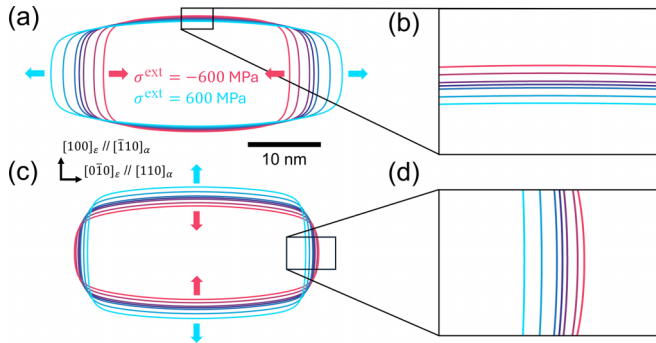


FIG. 3. Stresses of -600 , -350 , -100 , 0 , 100 , 350 , and 600 MPa were applied in the $[\bar{1}10]_{\alpha,2}$ and $[110]_{\alpha,2}$ directions of a variant-2 precipitate of control length $L = 20.12$ nm. The control condition is when the applied stress is 0 MPa. (a) The resulting equilibrium shape under compression (red) and tension (blue) for external applied stress in the $[110]_{\alpha,2}$ direction. (b) Under closer inspection, the side of the precipitate in the $[\bar{1}10]_{\alpha,2}$ direction moves in response to the applied stress, though the stress and side normal are perpendicular. (c) The resulting equilibrium shape under compression (red) and tension (blue) for external applied stress in the $[\bar{1}10]_{\alpha,2}$ direction. (d) The side of the precipitate in the $[110]_{\alpha,2}$ direction moves in response to the applied stress by a greater amount than seen in (b).

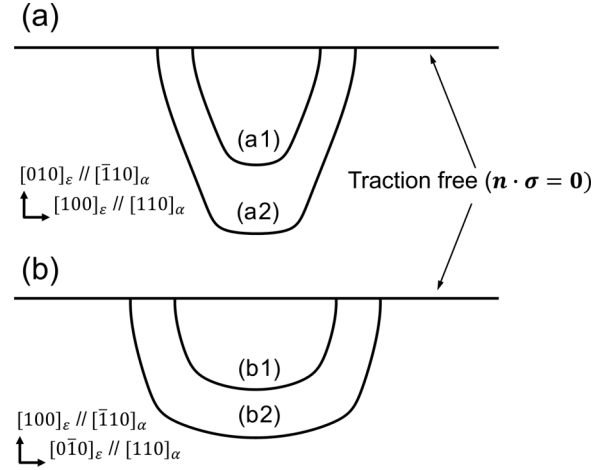


FIG. 4. Equilibrium shapes of precipitates at the traction-free surface with (a1) variant 1, $L = 17.12$ nm; (a2) variant 1, $L = 26.66$ nm; (b1) variant 2, $L = 17.57$ nm; and (b2) variant 2, $L = 27.13$ nm.

The shift in the aspect ratio of the precipitate may also be measured. A collection of the response rates from each linear regression is provided in Table II.

B. Two-dimensional precipitates near a traction-free surface

Precipitates nucleated with radii of 5 nm are initialized with the center 15 nm away from the free surface. The variant-1 precipitate with a final characteristic length of $L = 17.12$ nm reached equilibrium in 0.65 ms [Fig. 4(a1)] and the precipitate with characteristic length of $L = 26.66$ nm reached equilibrium in 1.1 ms [Fig. 4(a2)]. Similarly, the variant-2 precipitate with final characteristic length of $L = 17.57$ nm reached equilibrium in 0.94 ms [Fig. 4(b1)] and the precipitate with characteristic length of $L = 27.13$ nm reached equilibrium in 1.3 ms [Fig. 4(b2)].

Although Figs. 4(a1) and 4(b1) have the same initial condition, the difference in energy between the final states is 4.633×10^{-4} eV/nm² with the variant-2 precipitate lower in energy. Similarly, between Figs. 4(a2) and 4(b2), the difference in energy is 5.992×10^{-4} eV/nm², again favoring variant 2. The reason for this difference is that for precipitates of the same size at the free surface, variant 2 has both a lower total elastic energy per square nanometer of precipitate area and a lower total interfacial energy per nanometer of characteristic precipitate length.

The growth and migration of the precipitate from Fig. 4(b2) and interaction energy with variant 2 acting on variant 2 is shown in Fig. 5. The precipitate is drawn to the free surface by the low interaction energy, and then it adheres to the free surface. The precipitate moves through hydrogen diffusion, dissolving on one side while growing on the opposite side. The character of the interaction field changes as the precipitate approaches the free surface and as it comes to equilibrium. The relative positions of the attractive and repulsive regions are entirely different from the variant-2 precipitate in the bulk.

Four combinations of interaction energies were calculated for the precipitates at the free surface for all combinations of variants 1 and 2 interacting with variants 1 and 2. The

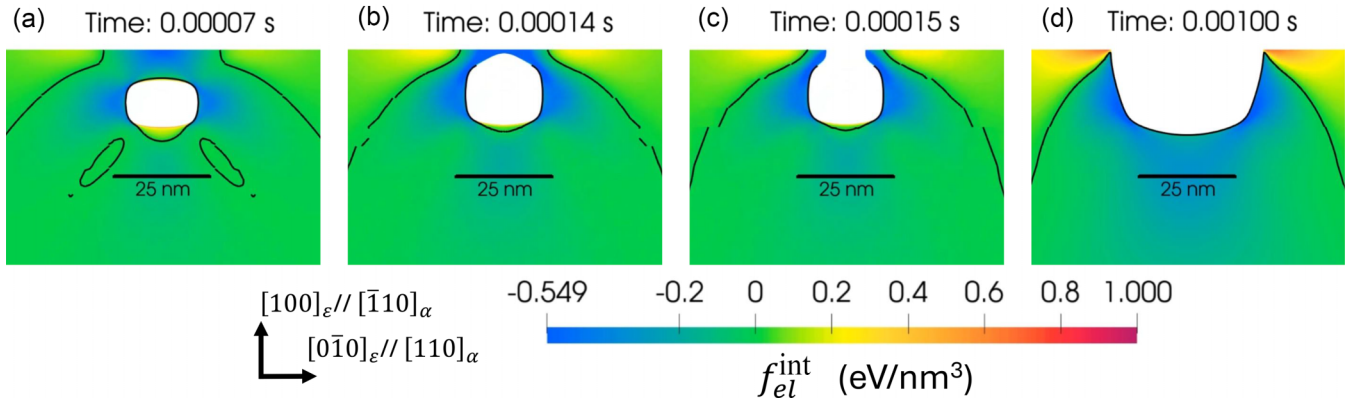


FIG. 5. Growth and migration of the variant-2 precipitate from Fig. 4(b2). The interaction energy field is for variant 2 acting on variant 2 and is separated by a zero contour (black) into regions of attraction (blue) and repulsion (red).

resulting interaction energies are shown in Fig. 6. Though the regions of attraction and repulsion are qualitatively the same between all interaction fields, the magnitude of attraction is greatest between variants of the same type. The regions of repulsion are found across the entirety of the free surface with the depth of the $f_{el}^{int} = 0$ contour increasing roughly linearly with distance from the precipitate edge.

To understand the implications of the interaction energies, two cases of two variant-1 precipitates are initialized as being already adhered to the traction-free surface as semicircles. The first case has nuclei of radius 3 nm [Figs. 7(a)–7(d)] and the second case has nuclei of radius 12 nm [Figs. 7(e)–7(h)]. In both simulations, the precipitate interfaces were separated by 1 nm, as close as possible without the onset of significant interface interaction. The frames on display for each case progress in time from early shape development, to interaction-energy-induced migration, to final equilibrium configuration.

In the case with precipitates of initial radius 3 nm, the precipitates begin by repelling each other symmetrically. Ma-

chine tolerance allows the symmetry to be broken, from which the unstable equilibrium of two equal-sized precipitates descends to the equilibrium state of one precipitate through coarsening. Figure 7(d) shows that the final state contains one precipitate located off center from the stress-induced precipitate interaction. The precipitates are dominated by repulsion because neither reaches deeply enough into the film to enter a significant portion of the other's attractive region as described in Sec. V B.

The precipitates initialized with radii of 12 nm experience attraction and eventually coalesce. These precipitates reach deeply enough into the film to enter a significant portion of the attractive region in the interaction energy field. We see that a larger fraction of the space between the precipitates in Fig. 7(e) contains negative interaction energy than positive interaction energy, where the space between precipitates in Fig. 7(a) contains more positive interaction energy than negative interaction energy. Thus, if the density of hydrides at the surface is sufficiently high, then a continuous film of hydride is expected to form.

C. Three-dimensional precipitates in the bulk

The three-dimensional morphology of the ϵ -Nb₄H₃ precipitate is not easily predictable from the elastic anisotropy of the phases. The Zener anisotropy coefficient, as calculated for our choice of stiffness in pure niobium, is 0.52, while the Zener anisotropy coefficient for ϵ -Nb₄H₃ is 1.2. This would suggest the elastically soft directions of the α phase are of type $\langle 111 \rangle_\alpha$ and the elastically soft directions for variants 1 and 2 of the ϵ phase are of type $\langle 100 \rangle_\epsilon$. Returning to the result from Eq. (23), this means the elastically soft directions of the ϵ phase are $[110]_\alpha$, $[\bar{1}10]_\alpha$, and $[001]_\alpha$. As a result, the elastically soft and hard directions of each phase are unaligned. In the simple two-dimensional case, where the elastically soft directions are aligned, at sufficiently large sizes the precipitates will take on a plate or rodlike shape along these directions. However, in three dimensions, simple consideration of elastic anisotropy ratios cannot reliably predict the outcome of the hydride precipitate shape outright.

To reduce the computational cost of simulating a complete, single ϵ hydride, we exploit the common symmetries of the stiffness tensors and misfit tensors. In the variant-1 frame,

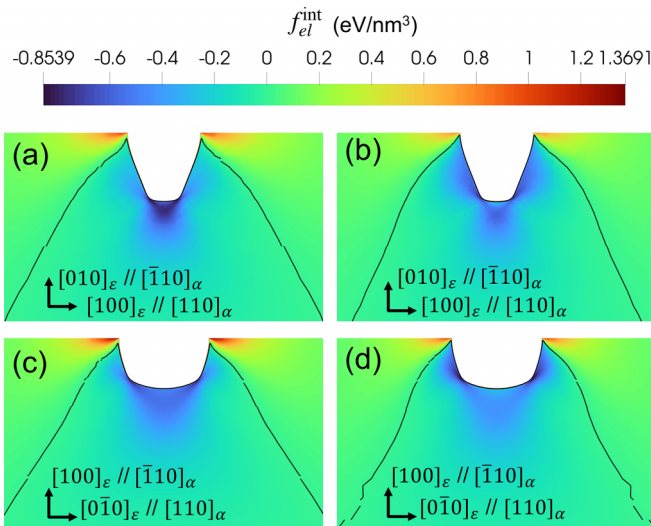


FIG. 6. The interaction energies of precipitates at the traction-free surface of (a) variant 1 acting on variant 1, (b) variant 1 acting on variant 2, (c) variant 2 acting on variant 1, and (d) variant 2 acting on variant 2.

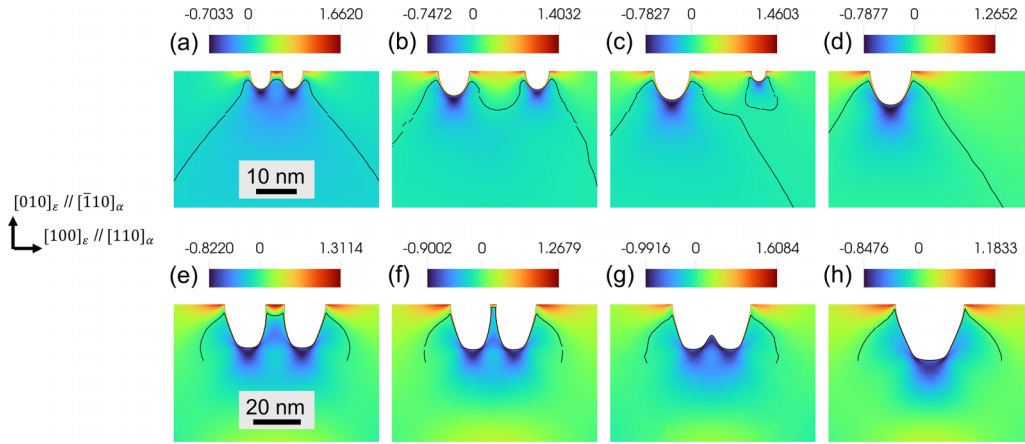


FIG. 7. Interaction energies of two precipitates. [(a)–(d)] Variant-1 precipitates of initial radii 3 nm at times (a) $t = 0.01$ ms, (b) $t = 0.2$ ms, (c) $t = 0.38$ ms, and (d) $t = 0.43$ ms. [(e)–(h)] Variant-2 precipitates of initial radii 12 nm at times (e) $t = 0.16$ ms, (f) $t = 0.45$ ms, (g) $t = 0.50$ ms, and (h) $t = 1.32$ ms.

three mirror symmetries of the α -phase stiffness tensor and ε -phase stiffness tensor and misfit tensor are coincident, the mirror symmetry of each $\langle 100 \rangle_\varepsilon$. By reducing the domain by one-half for each of these directions, we could simulate one octant of the total system and produce the final condition by reflection.

The resulting three-dimensional morphology of an ε -hydride precipitate within the bulk of the α phase as displayed in Fig. 8 is a rectangular bipyramid, or an elongated octahedron. The pyramidal directions are aligned with $[001]_\alpha$ and the sides of the rectangle are aligned with $[\bar{1}10]_\alpha$ and $[110]_\alpha$. The rectangular base of the pyramid lies within the same plane and is oriented in the same direction as the two-dimensional rectangular precipitates observed in Sec. V A. Though the precipitate shown is of variant 1, the morphologies of the other variants are identical up to some rotation as derived in the Appendix.

The interaction energy of the three-dimensional hydride from Fig. 8 is shown in Fig. 9. The $f_{el}^{\text{int}} = 0$ contour surface is pictured in Fig. 9(a). The calculated interaction is for variant 1 acting on variant 1. Also shown is the domain boundary as a frame around the contour surface. The interaction energy is also shown in cross sections of the lattice parameter planes of the ε phase. The majority of the domain is defined by a region of attraction. The region of repulsion is enclosed and extends in the directions of $[100]_\alpha$, $[010]_\alpha$, and $[001]_\alpha$.

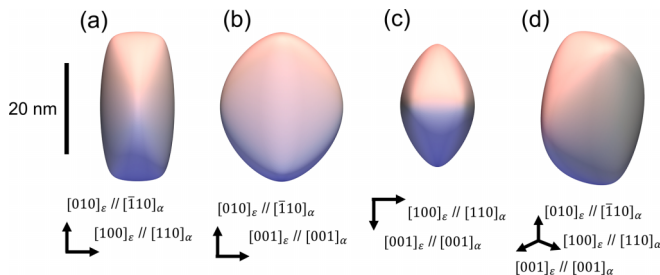


FIG. 8. A three-dimensional variant-1 ε -phase precipitate as observed from different angles: (a) $[001]_\varepsilon$ plane, (b) $[100]_\varepsilon$ plane, and (c) $[010]_\varepsilon$ plane. (d) As viewed from approximately $[111]_\varepsilon$.

D. Three-dimensional precipitates near a traction-free surface

As the introduction of the traction-free surface drastically changed the morphology and dynamics of two-dimensional hydrides, so too we expect the three-dimensional precipitates to experience a transfiguration. In the bulk, all variants were identical up to some rotation, but the addition of the free surface breaks the symmetry. Variants 1 and 2 must be simulated individually. The free surface also breaks the symmetry of our domain reduction method, so instead of simulating one-eighth of the desired domain, we must simulate one-quarter.

The shapes of the three-dimensional hydrides during growth in the presence of the traction-free surface for variants 1 and 2 are featured in Figs. 10 and 11, respectively. As expected, like the 2D precipitates, these hydrides attempt to maximize contact with the surface to reduce interfacial energy, as there is no interfacial energy between the ε phase and the traction-free surface.

Interaction energies are calculated for each variant as it interacts with the same variant, 1 with 1 and 2 with 2. The interaction energies are shown in cross sections in Figs. 12 and 13. The precipitates show repulsive behavior throughout the entire plane associated with the free surface for both precipitates, unlike the bulk.

VI. DISCUSSION

A thorough investigation of the resulting morphologies and dynamics of ε -Nb₄H₃ within environments of varied stress conditions was performed. From the significant magnitude of the misfit strain and clear elastic anisotropy between phases, the elastic energy played a dominant role in the findings, the first being the equilibrium shape of the precipitating nanoscale structures.

Owing to symmetry of the hydrides up to rotation, precipitates both in the appropriate two-dimensional section and in three dimensions take the same morphology within the surrounding α -NbH_x matrix, regardless of variant type. In two dimensions, the precipitates display a rectangular shape, with the shorter direction aligned with $[100]_\varepsilon$ and the longer

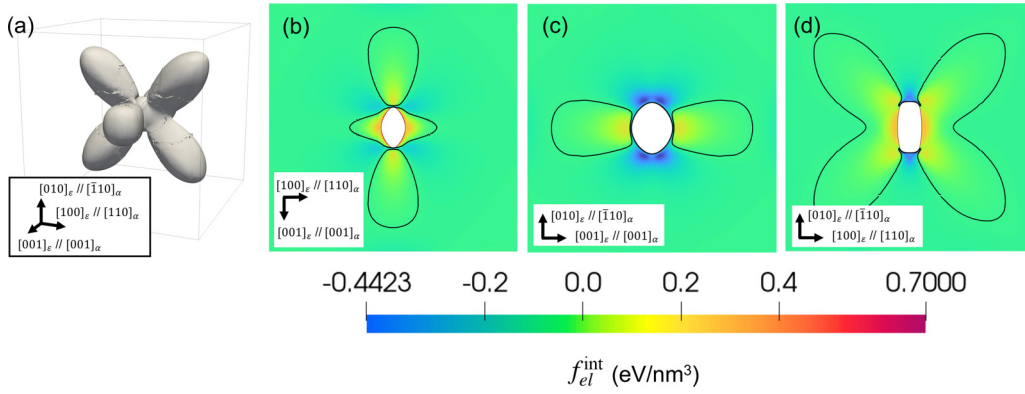


FIG. 9. (a) The $f_{el}^{int} = 0$ contour surface and domain boundary. This contour is colored black in (b)–(d). (b) $[\bar{1}10]_{\alpha}$ cross section through the center of the precipitate. (c) $[110]_{\alpha}$ cross section through the center of the precipitate. (d) $[001]_{\alpha}$ cross section through the center of the precipitate.

direction aligned with $[010]_{\varepsilon}$. This is unsurprising because the misfit strain is greater in $[100]_{\varepsilon}$ than in $[010]_{\varepsilon}$ with $0.04357 > 0.02920$. The dilatational stiffness of the ε phase is also greater in $[100]_{\varepsilon}$ than in $[010]_{\varepsilon}$, or $C_{11,\varepsilon} > C_{22,\varepsilon}$. The flat sides correspond to elastically soft directions of the ε phase and the projected soft directions of the α phase. In three dimensions, the precipitates take on a rectangular bipyramid or elongated octahedron shape. The elastically soft directions of the α phase correspond to the faces of the precipitate. Since the elastic constants of ε are greater in a general sense than α , it is reasonable that the normals to the faces of the 3D precipitate would follow that of the less stiff material, since the less stiff material would be inclined to conform to the more stiff material. The elongation of the rectangular base of the bipyramidal structure agrees with the results from 2D simulations.

Calculating the interaction energy within the α phase reveals the configurational force acting on an interface at a given location. Dividing the domain into regions of positive energy contribution and negative interaction energy shows where hydride interfaces would be inclined to move given some stress state. In two dimensions for a precipitate within the bulk, the regions of attraction lie in the $\langle 110 \rangle_{\alpha}$ directions and regions of repulsion lie on $\langle 100 \rangle_{\alpha}$. Though the elastic constant anisotropy of our study is unique, this

result is qualitatively similar to previous work on rectangular precipitates [18–20,36,37]. 3D precipitates within the bulk produce a similar interaction energy field upon inspection of the cross section in the same plane as the 2D results, $[001]_{\alpha}$. The addition of another dimension reveals that precipitates of the same variant aligned in the $[001]_{\alpha}$ direction would be repelled. Precipitates not aligned in the repulsive directions, $[110]_{\alpha}$, $[\bar{1}10]_{\alpha}$, and $[001]_{\alpha}$, are attracted, where the magnitude of attraction is the greatest from the $[\bar{1}10]_{\alpha}$ direction.

Precipitates located near a traction-free surface migrate to, then adhere to, that free surface in a matter of milliseconds. Unlike the stacking behavior of precipitates, where precipitates are attracted from a distance but repelled in close proximity, precipitates experience no repulsion from the traction-free surface. Variants 1 and 2 experience a dramatic decrease in energy when adhered to the free surface as opposed to when they are not. Even if a precipitate cannot migrate quickly enough to a free surface before another precipitate, it will dissolve through coarsening to further reduce the energy of the system. For this reason, we propose that for any system with a limited dimension such that every point of the system is within a characteristic diffusion distance from a free surface, all coherent, misfitting precipitates will either migrate to the free surface or dissolve. The result is that no coherent, misfitting precipitates will exist inside the bulk Nb

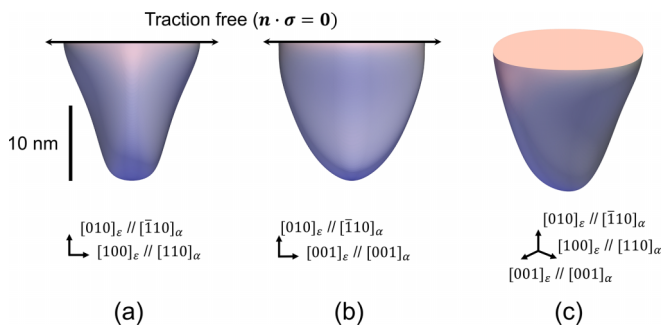


FIG. 10. A growing three-dimensional variant-1 ε -phase precipitate adhered to a traction-free surface as observed from different angles: (a) $[001]_{\alpha}$ plane, (b) $[110]_{\alpha}$ plane. (c) As viewed from approximately $[011]_{\alpha}$.

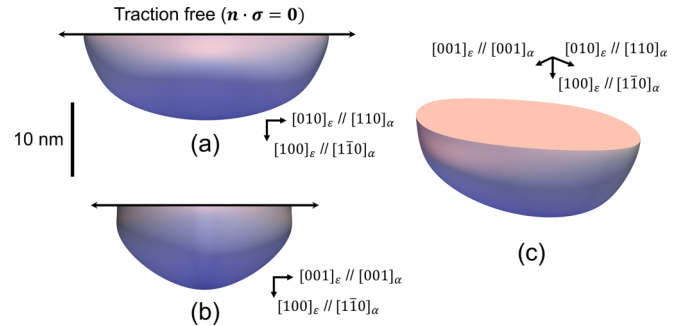


FIG. 11. A growing three-dimensional variant-2 ε -phase precipitate adhered to a traction-free surface as observed from different angles: (a) $[001]_{\alpha}$ plane, (b) $[110]_{\alpha}$ plane. (c) As viewed from approximately $[011]_{\alpha}$.

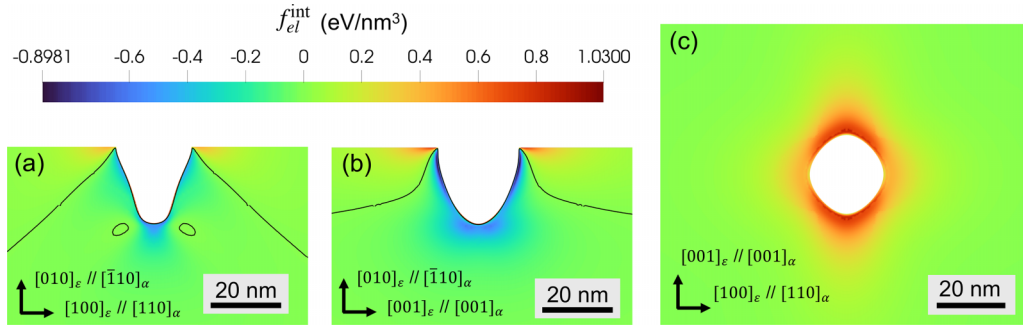


FIG. 12. The interaction energy of variant 1 with variant 1 in three dimensions. The $f_{el}^{int} = 0$ contour is colored black. (a) $[001]_{\alpha}$ plane, (b) $[110]_{\alpha}$ plane, and (c) traction-free surface.

thin film of a qubit and all will be attached to the surface. One significant consequence of this behavior is that secondary-phase volume fraction cannot be accurately approximated from the area fraction at the film surface because this assumes a constant distribution of the secondary phase in depth. So it is not advised to estimate the hydrogen loading of a thin-film sample using the method of measuring area fraction, as it will greatly overestimate the total hydrogen in the system.

The interaction energy of precipitates in two and three dimensions indicates a strong tendency of repulsion at the free surface, as there are no regions of attraction. The only way two precipitates at the free surface might be attracted is if they are both sufficiently large and sufficiently close, which may be unlikely. Precipitates at the free surface grow at the expense of precipitates in the bulk to reduce energy. So precipitates could not grow large relative to those at the free surface, then become attached less than 10 nm away. The much more likely scenario is that all precipitates that adhere to the free surface are at first small and repel one another. Then they grow due to coalescence with precipitates within the bulk or from hydrogen supersaturation. Finally, once the supersaturation has been depleted, coarsening dominates the growth for the remaining distant precipitates. Those at the free surface grow at the expense of distant precipitates in the bulk.

Niobium hydride precipitates have been observed at far greater sizes than considered in our study [9,14,38] and research shows niobium hydrides undergo a transition from precipitates with a coherent to semicoherent interface at sizes similar to or larger than those observed in our work [11].

Because our model does not account for the introduction of dislocations that can partially relax the misfit strains, the results presented should be taken as valid for length scales $L < 30$ nm. This model does not reflect the behavior of larger hydrides and the concomitant plastic relaxation. However, our work captures the early evolution of the precipitates in films as they migrate within the first milliseconds of formation.

Experimentally observed hydrides in bulk materials feature bumpy exteriors and our simulations show precipitates have preferred growth directions, which is indicative of dendrite growth [9]. Hydrides in thin films have been observed to a more limited extent. Surface features have a large impact on the nucleation and growth direction of niobium hydrides in single-crystal, epitaxial, (110) niobium [12]. Lee *et al.* identified hydride “types I and II” which directly correspond to hydride variants 1 and 2 and variants 3–6 [10]. The size distributions of these hydride types are different, with type I having more small, irregularly shaped precipitates and type II having fewer large precipitates with clear boundaries. This might suggest that variants 3–6 have a lower formation energy than variant 2, which was the variant with the lowest formation energy of the 3D simulations in this study. Future models for incoherent, dendritic hydrides would be more accurate for larger precipitates. The introduction of the traction-free surface breaks the reflective symmetries of variants 3–6. Due to the associated computational cost, variants 3–6 were not simulated in three dimensions in the presence of the traction-free surface, but should be considered in future research. For the same reason, multiple precipitates were not simulated in the bulk or near the free surface in three dimensions.

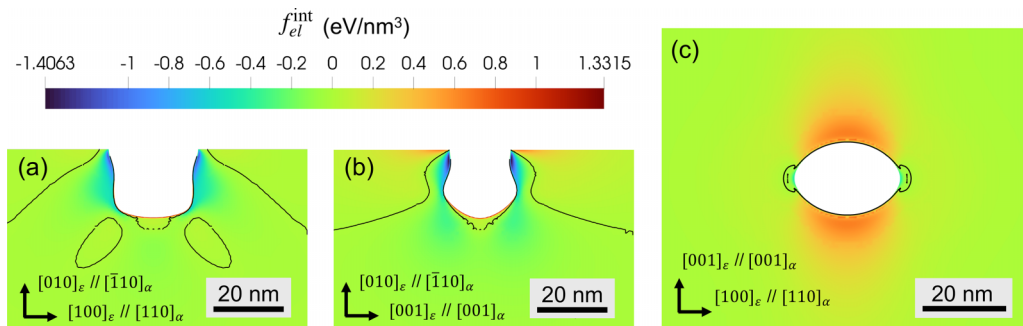


FIG. 13. The interaction energy of variant 2 with variant 2 in three dimensions. The $f_{el}^{int} = 0$ contour is colored black. (a) $[001]_{\epsilon}$ plane, (b) $[110]_{\alpha}$ plane, and (c) traction-free surface.

VII. CONCLUSION

This study examined the combined impact of elastic heterogeneity, misfit strains for various ε -phase variants, multiprecipitate interaction, and finite system size on the morphology and dynamics of ε -Nb₄H₃ nanoparticles in hydrogen-enriched niobium thin films. Interfacial energy effects usually dominate elastic effects at small sizes due to scaling factors, but due to the high misfit strains and comparatively low interfacial energy, elastic effects are shown to be prominent for the nanoscale precipitates studied. As such, the effect of elastic stress is directly responsible for the observed morphology and distribution of ε -Nb₄H₃ precipitates.

Unlike previous work using phase field methods to model precipitate dynamics in an infinite medium, our study intentionally considers the effect of a traction-free boundary to model the physical constraints imposed by the growth of a thin film. We recover rectangular, two-dimensional precipitates at equilibrium within the bulk, which is similar to elongated precipitates from previous studies. In three dimensions, the precipitates take on an elongated octahedron morphology, where the face directions correspond to the elastically soft directions of pure niobium. Interaction energy fields are calculated to determine the relative positions of stability for multiple precipitates within a bulk sample. Using contours of zero interaction energy, we show a clear delineation between regions of attraction and repulsion for each result.

Applied external stress is shown to influence the aspect ratio and final size of precipitates at equilibrium, which shows that external stress has the ability to shift equilibrium compositions of the α phase and, so too, the phase boundary in the phase diagram.

When exposed to a traction-free surface, precipitates are attracted to the surface and are able to grow larger than within the bulk due to a reduction of interfacial and elastic energy, following the Gibbs-Thompson effect. In two dimensions, we observe that variant 2 reduces its energy more than variant 1 does, which implies that variant 2 is more stable at the free surface. Interaction energy fields of precipitates adhered to a free surface differ greatly from those within the bulk. We find that precipitates at the free surface would experience mutual repulsion from all other precipitates at the free surface, a behavior far different than misfitting precipitates experience in bulk environments.

ACKNOWLEDGMENTS

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

APPENDIX: DERIVATION OF VARIANT MECHANICAL PROPERTIES: MISFIT STRAIN AND STIFFNESS TENSORS

The misfit strain tensors for each variant are calculated directly using the lattice parameters and orientation relationship of the ε phase and α phase. The lattice parameters of the α phase and ε phase were obtained at temperatures in the range 13–190 K [39]. To calculate the misfit strain, the lattice parameters were taken to be constant at the prescribed temperature, $T_c = 203$ K = -70°C . Because the lattice parameters of both phases are functions of temperature and hydrogen concentration, so too is the misfit strain. These parameters are known for the α phase but not the ε phase.

The solute dilatation strain in the α phase due to hydrogen is known to be $1/a(\Delta a/\Delta X) \approx 0.05$, where X is the atomic ratio of hydrogen to niobium [15,40]. Because we assume the precipitating hydrides in our system occur at sufficiently low temperatures where the equilibrium concentration of hydrogen in the α phase is less than 1%, the solute dilatation strain is neglected. The thermal expansion coefficient is a strong function of temperature when $80 < T < 200$ K [41]. Since the thermal expansion coefficient is roughly constant at $T < 80$ K, and the thermal expansion coefficient is sufficiently small at $\alpha = 1/a(da/dT) = 6 \times 10^{-6}$, thermal expansion is neglected in our calculation of the misfit strain. The ε phase is orthorhombic with lattice parameters that are geometrically related to the lattice parameters of the cubic α phase:

$$\begin{aligned} [100]_\varepsilon || [110]_\alpha : a_\varepsilon &\approx 2\sqrt{2}a_\alpha \\ [010]_\varepsilon || [\bar{1}10]_\alpha : b_\varepsilon &\approx 2\sqrt{2}a_\alpha \\ [001]_\varepsilon || [001]_\alpha : c_\varepsilon &\approx a_\alpha. \end{aligned} \quad (\text{A1})$$

The lattice transformation matrix which describes the change from the α -NbH_x phase to the ε -Nb₄H₃ phase is the following:

$$\begin{aligned} U &= \begin{bmatrix} \frac{\sqrt{2}}{4} \frac{a_\varepsilon}{a_\alpha} & 0 & 0 \\ 0 & \frac{\sqrt{2}}{4} \frac{b_\varepsilon}{a_\alpha} & 0 \\ 0 & 0 & \frac{c_\varepsilon}{a_\alpha} \end{bmatrix} \\ &= \begin{bmatrix} 1.04257 & 0 & 0 \\ 0 & 1.02920 & 0 \\ 0 & 0 & 1.03754 \end{bmatrix}. \end{aligned} \quad (\text{A2})$$

The linear elasticity small-strain approximation produces the following misfit strain due to phase transformation:

$$\epsilon_{ij,\varepsilon}^{*,\text{mis}} = \frac{1}{2}(U + U^T) - I \quad (\text{A3})$$

$$= U - I \quad (\text{A4})$$

$$= \begin{bmatrix} 0.04257 & 0 & 0 \\ 0 & 0.02920 & 0 \\ 0 & 0 & 0.03754 \end{bmatrix}. \quad (\text{A5})$$

The orientation relationship of $\varepsilon \rightarrow \alpha$ for the example configuration in Eq. (A1) may be readily obtained, where some vector in the ε frame, $\vec{v}_\varepsilon$, may be transformed to a vector in the α frame, $\vec{v}_\alpha$, by the following equation:

$$\vec{v}_\alpha = \frac{\sqrt{2}}{2} \begin{bmatrix} [0.7]1 & -1 & 0 \\ 1 & 1 & 0 \\ 0 & 0 & \sqrt{2} \end{bmatrix} \vec{v}_\varepsilon = L_{\varepsilon,\alpha} \vec{v}_\varepsilon. \quad (\text{A6})$$

Evidenced by Eq. (A1), both unit cell coordinates $\vec{a}_\varepsilon$ and $\vec{b}_\varepsilon$ may be aligned with any of the $\langle 110 \rangle_\alpha$ directions and still satisfy the orientation relationship so long as $\vec{a}_\varepsilon \perp \vec{b}_\varepsilon$.

Theorem A1. Let $\vec{a}_\varepsilon$ be aligned with one of the 12 total $\langle 110 \rangle_\alpha$ directions. For any of the $\langle 110 \rangle_\alpha$ directions, there are exactly two other $\langle 110 \rangle_\alpha$ directions that are perpendicular.

Proof. Without loss of generality, let $\vec{a}_\varepsilon \parallel [110]_\alpha$. Let $\vec{b}_\varepsilon$ be an unknown $\langle 110 \rangle_\alpha$ vector with components $[b_{\varepsilon,1}, b_{\varepsilon,2}, b_{\varepsilon,3}]_\alpha$. For $\vec{a}_\varepsilon \perp \vec{b}_\varepsilon$, we must have

$$\begin{aligned} \vec{a}_\varepsilon \cdot \vec{b}_\varepsilon &= 0 \\ \Rightarrow b_{\varepsilon,1} + b_{\varepsilon,2} &= 0, \quad b_{\varepsilon,3} \text{ unconstrained} \\ \Rightarrow b_{\varepsilon,1} &= -b_{\varepsilon,2}, \quad b_{\varepsilon,3} \text{ unconstrained.} \end{aligned}$$

Case 1: $b_{\varepsilon,1} = 0$. Then $\vec{b}_\varepsilon = [0, 0, b_{\varepsilon,3}]_\alpha$. This is a contradiction because $\vec{b}_\varepsilon$ is a $\langle 110 \rangle_\alpha$ vector and must have at least two nonzero components.

Case 2: $b_{\varepsilon,1} \neq 0$. Then $\vec{b}_\varepsilon = [b_{\varepsilon,1}, -b_{\varepsilon,1}, b_{\varepsilon,3}]_\alpha$. Since $\vec{b}_\varepsilon$ is a $\langle 110 \rangle_\alpha$ vector, one component must be zero. $(b_{\varepsilon,1} \neq 0) \wedge (b_{\varepsilon,1} = 0 \vee b_{\varepsilon,3} = 0) \Rightarrow b_{\varepsilon,3} = 0$. Since $b_{\varepsilon,1}$ may be positive or negative, the result is

$$\vec{b}_\varepsilon \parallel \begin{cases} [1\bar{1}0]_\alpha & \text{if } b_{\varepsilon,1} > 0 \\ [\bar{1}10]_\alpha & \text{if } b_{\varepsilon,1} < 0. \end{cases} \quad (\text{A7})$$

For every choice of an $\vec{a}_\varepsilon$ there are two choices for $\vec{b}_\varepsilon$, and finally $\vec{c}_\varepsilon$ must be constrained. This would suggest that there exist 24 crystallographically distinct variants of $\varepsilon\text{-Nb}_4\text{H}_3$ with respect to the $\alpha\text{-NbH}_x$ crystal coordinate system under rotation. Note that the ε phase belongs to the space group $P2_12_12_1$, which does not include symmetries of reflection. By considering crystal reflections for each variant obtained through rotation, we arrive at a total of 48 crystallographically distinct variants of $\varepsilon\text{-Nb}_4\text{H}_3$ with respect to the $\alpha\text{-NbH}_x$ crystal coordinate system under rotation and reflection operations. Alternatively, one may consider the number of orientation configurations, N_O , within the ε frame, where there is a four-fold axis along $\vec{c}_\varepsilon$, $\vec{c}_\varepsilon$ can take on the same direction as one of the three α -phase coordinate directions in either the positive or negative direction, and the frame-invariant contribution of reflected crystals. The combined multiplicities are

$$N_O = \overbrace{N_{4\text{-fold}}}^{\text{4-fold axis along } \vec{c}_\varepsilon} \times \overbrace{N_{\alpha\text{axis}}}^{\text{choice of axis for } \vec{c}_\varepsilon} \times \overbrace{N_{\pm}}^{\text{choice of direction for } \vec{c}_\varepsilon} \times \overbrace{N_{\text{reflect}}}^{\text{mirror doubling}} = 4 \times 3 \times 2 \times 2 = 48. \quad (\text{A8})$$

To understand the relationship of the mechanical properties between variants, a set of symmetry operators must be defined that generate the group of 48 variants and also operate on the strain and stiffness tensors of the ε phase. We arbitrarily define the transformation matrix from Eq. (A6) as the fundamental relation between the α frame and the reference frame of variant 1. The rotation relationships between variants in the ε frame are not self-evident. However, obtaining crystallographically consistent orientation relationships by rotating in the α frame is simple.

Theorem A2. Any combination of rotations of 90° and reflection operations about the crystal axes of the α frame maintains that $\vec{a}_\varepsilon, \vec{b}_\varepsilon \parallel \langle 110 \rangle_\alpha$ and $\vec{c}_\varepsilon \parallel \langle 100 \rangle_\alpha$.

Proof. A rotation matrix of 90° about a coordinate axis of the α phase may at most reorder and flip the signs of components in a vector as expressed in the α frame. The classification of a $\langle 110 \rangle$ vector is invariant upon flipping signs and switching of components. Similarly, the classification of a $\langle 100 \rangle$ vector is also invariant upon the same operations. A reflection matrix about the coordinate axis of the α phase will flip the sign of one component in the vector it operates on. Per the previous argument, $\langle 110 \rangle$ and $\langle 100 \rangle$ vectors maintain their classification.

Rotations of 90° and any reflection operation about the crystal axes of the α phase generate the 48 crystallographically distinct ε -phase variants. These operations are part of the group known as the full symmetry group of the cube. Applying the obtained symmetry operators reveals degeneracy of the mechanical properties between some variants. Though the crystals may be unique, the rotated or reflected stiffness and strain tensors are identical; thus, they are elastically indistinct from each other. Transforming a stiffness tensor by some arbitrary orthogonal matrix, such as a rotation or reflection, in Einstein index notation is defined as

$$C'_{ijkl} = \Omega_{ip}\Omega_{jq}\Omega_{kr}\Omega_{ls}C_{pqrs}. \quad (\text{A9})$$

Using the ε -phase coordinate system, rotations of 180° and reflections about the coordinate axes are diagonal. So for these operations, Ω_{nm} is nonzero only when $n = m$. It then follows that

$$\Omega_{nm} = \alpha_m \delta_{nm}, \quad (\text{A10})$$

$$C'_{ijkl,\varepsilon} = \alpha_p \alpha_q \alpha_r \alpha_s C_{pqrs,\varepsilon} \delta_{ip} \delta_{jq} \delta_{kr} \delta_{ls}, \quad (\text{A11})$$

$$C'_{ijkl,\varepsilon} = \alpha_i \alpha_j \alpha_k \alpha_l C_{ijkl,\varepsilon}, \quad (\text{A12})$$

where δ_{nm} is the Kronecker delta function and α_m is the m^{th} diagonal element of Ω_{nm} . Since we are in the ε coordinate system, the stiffness tensor has orthorhombic symmetry. Nonzero entries of orthorhombic stiffness tensors have an even number of indices labeled 1, 2, and 3. Let f , g , and h be integers which count the index pairs of 1s, 2s, and 3s, respectively. Because $\alpha_1, \alpha_2, \alpha_3 \in \{-1, 1\}$, Eq. (A12) becomes

$$C'_{ijkl,\varepsilon} = (\pm 1)^{2f} (\pm 1)^{2g} (\pm 1)^{2h} C_{ijkl,\varepsilon} \quad (\text{A13})$$

$$= (1)^{f+g+h} C_{ijkl,\varepsilon} \quad (\text{A14})$$

$$= C_{ijkl,\varepsilon}. \quad (\text{A15})$$

Equation (A15) shows the stiffness tensor is invariant under rotations of 180° and reflections. Removing these degenerate variants leaves the elastically unique variants. Accounting for this modification in Eq. (A8) yields

$$N_{O,el} = \underbrace{N_{4\text{-fold}}}_{\substack{\text{4-fold axis along } \vec{c}_\varepsilon \\ \text{choice of direction for } \vec{c}_\varepsilon}} / 2 \times \underbrace{N_{\alpha\text{axis}}}_{\substack{\text{choice of axis for } \vec{c}_\varepsilon \\ \text{mirror doubling}}} = 2 \times 3 = 6. \quad (\text{A16})$$

We can be certain that all the elastically degenerate variants have been obtained by observing that either a 90° rotation about $\vec{c}_\varepsilon$ or 90° rotation about $\vec{a}_\varepsilon$ does not produce an identical stiffness tensor. The final count is that there are two elastically distinct variants related by a 90° rotation per $\langle 100 \rangle_\alpha$ direction. Proof that six variants is sufficient to describe all distinct misfit tensors follows a similar argument. Since the misfit tensor and transformation matrices of 180° rotations and reflections are diagonal in the ε coordinate system, the same analysis holds:

$$\tilde{\epsilon}_{ij,\varepsilon}^{*,\text{mis}} = \Omega_{ik} \epsilon_{kl,\varepsilon}^{*,\text{mis}} \Omega_{jl} \quad (\text{A17})$$

$$= \alpha_k \alpha_l \epsilon_{kl,\varepsilon}^{*,\text{mis}} \delta_{ik} \delta_{jl} \quad (\text{A18})$$

$$= \alpha_i \alpha_j \epsilon_{ij,\varepsilon}^{*,\text{mis}}, \quad i = j \quad (\text{A19})$$

$$= (\pm 1)^2 \epsilon_{ij,\varepsilon}^{*,\text{mis}} \quad (\text{A20})$$

$$= \epsilon_{ij,\varepsilon}^{*,\text{mis}}. \quad (\text{A21})$$

As a shorthand, let ϵ_a^* , ϵ_b^* , and ϵ_c^* be the misfit strains of variant 1 in the ε reference frame; the misfit tensors for the remaining variants can be obtained through a rotation of 45° into the α reference frame, a 90° rotation about one of the $\langle 100 \rangle_\alpha$ axes, and reverse rotation of 45° back into the ε frame. The composite rotation matrices, $\mathbf{R}_i$, act upon the misfit strain tensor of variant 1, $\epsilon_{1,\varepsilon}^{*,\text{mis}}$, to produce the misfit tensors of the remaining variants. The misfit strain tensors are symmetric, so the lower triangular, off-diagonal terms are not entered for

conciseness:

$$\epsilon_{i,\varepsilon}^{*,\text{mis}} = \mathbf{R}_i \epsilon_{1,\varepsilon}^{*,\text{mis}} \mathbf{R}_i^T, \quad (\text{A22})$$

$$\mathbf{R}_2 = \begin{bmatrix} 0 & -1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad (\text{A23})$$

$$\mathbf{R}_3 = \frac{1}{2} \begin{bmatrix} 1 & -1 & -\sqrt{2} \\ -1 & 1 & -\sqrt{2} \\ \sqrt{2} & \sqrt{2} & 0 \end{bmatrix}, \quad (\text{A24})$$

$$\mathbf{R}_4 = \frac{1}{2} \begin{bmatrix} 1 & 1 & -\sqrt{2} \\ -1 & -1 & -\sqrt{2} \\ -\sqrt{2} & \sqrt{2} & 0 \end{bmatrix}, \quad (\text{A25})$$

$$\mathbf{R}_5 = \frac{1}{2} \begin{bmatrix} 1 & 1 & \sqrt{2} \\ 1 & 1 & -\sqrt{2} \\ -\sqrt{2} & \sqrt{2} & 0 \end{bmatrix}, \quad (\text{A26})$$

$$\mathbf{R}_6 = \frac{1}{2} \begin{bmatrix} -1 & 1 & \sqrt{2} \\ -1 & 1 & -\sqrt{2} \\ -\sqrt{2} & -\sqrt{2} & 0 \end{bmatrix}, \quad (\text{A27})$$

$$\epsilon_{2,\varepsilon}^{*,\text{mis}} = \begin{bmatrix} \epsilon_b^* & 0 & 0 \\ 0 & \epsilon_a^* & 0 \\ 0 & 0 & \epsilon_c^* \end{bmatrix}, \quad (\text{A28})$$

$$\epsilon_{3,\varepsilon}^{*,\text{mis}} = \frac{1}{2} \begin{bmatrix} [1]\epsilon_c^* + \frac{\epsilon_a^* + \epsilon_b^*}{2} & \epsilon_c^* - \frac{\epsilon_a^* + \epsilon_b^*}{2} & \sqrt{2}(\epsilon_a^* - \epsilon_b^*) \\ \bullet & \epsilon_c^* + \frac{\epsilon_a^* + \epsilon_b^*}{2} & -\sqrt{2}(\epsilon_a^* - \epsilon_b^*) \\ \bullet & \bullet & \epsilon_a^* + \epsilon_b^* \end{bmatrix}, \quad (\text{A29})$$

$$\epsilon_{4,\varepsilon}^{*,\text{mis}} = \frac{1}{2} \begin{bmatrix} [1]\epsilon_c^* + \frac{\epsilon_a^* + \epsilon_b^*}{2} & \epsilon_c^* - \frac{\epsilon_a^* + \epsilon_b^*}{2} & -\sqrt{2}(\epsilon_a^* - \epsilon_b^*) \\ \bullet & \epsilon_c^* + \frac{\epsilon_a^* + \epsilon_b^*}{2} & \sqrt{2}(\epsilon_a^* - \epsilon_b^*) \\ \bullet & \bullet & \epsilon_a^* + \epsilon_b^* \end{bmatrix}, \quad (\text{A30})$$

$$\epsilon_{5,\varepsilon}^{*,\text{mis}} = \frac{1}{2} \begin{bmatrix} [1]\epsilon_c^* + \frac{\epsilon_a^* + \epsilon_b^*}{2} & -(\epsilon_c^* - \frac{\epsilon_a^* + \epsilon_b^*}{2}) & -\sqrt{2}(\epsilon_a^* - \epsilon_b^*) \\ \bullet & \epsilon_c^* + \frac{\epsilon_a^* + \epsilon_b^*}{2} & -\sqrt{2}(\epsilon_a^* - \epsilon_b^*) \\ \bullet & \bullet & \epsilon_a^* + \epsilon_b^* \end{bmatrix}, \quad (\text{A31})$$

$$\epsilon_{6,\varepsilon}^{*,\text{mis}} = \frac{1}{2} \begin{bmatrix} [1]\epsilon_c^* + \frac{\epsilon_a^* + \epsilon_b^*}{2} & -(\epsilon_c^* - \frac{\epsilon_a^* + \epsilon_b^*}{2}) & \sqrt{2}(\epsilon_a^* - \epsilon_b^*) \\ \bullet & \epsilon_c^* + \frac{\epsilon_a^* + \epsilon_b^*}{2} & \sqrt{2}(\epsilon_a^* - \epsilon_b^*) \\ \bullet & \bullet & \epsilon_a^* + \epsilon_b^* \end{bmatrix}. \quad (\text{A32})$$

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