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► To cite this version:

Rémi Dingreville, Abdelmalek Hallil, Stéphane Berbenni. From coherent to incoherent mismatched interfaces: A generalized continuum formulation of surface stresses. *Journal of the Mechanics and Physics of Solids*, 2014, 72, pp.40-60. 10.1016/j.jmps.2014.08.003 . hal-01503887

HAL Id: hal-01503887

<https://hal.univ-lorraine.fr/hal-01503887v1>

Submitted on 23 Sep 2024

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From coherent to incoherent mismatched interfaces: a generalized continuum formulation of surface stresses

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Abstract

The equilibrium of coherent and incoherent mismatched interfaces is reformulated in the context of continuum mechanics based on the Gibbs dividing surface concept. Two surface stresses are introduced: a coherent surface stress and an incoherent surface stress, as well as a transverse excess strain. The coherent surface stress and the transverse excess strain represent the thermodynamic driving forces of stretching the interface while the incoherent surface stress represents the driving force of stretching one crystal while holding the other fixed and thereby altering the structure of the interface. These three quantities fully characterize the elastic behavior of coherent and incoherent interfaces as a function of the in-plane strain, the transverse stress and the mismatch strain. The isotropic case is developed in detail and particular attention is paid to the case of interfacial thermo-elasticity. This exercise provides an insight on the physical significance of the interfacial elastic constants introduced in the formulation and illustrates the obvious coupling between the interface structure and its associated thermodynamics quantities. Finally, an example based on

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atomistic simulations of Cu/Cu₂O interfaces is given to demonstrate the relevance of the generalized interfacial formulation and to emphasize the dependence of the interfacial thermodynamic quantities on the incoherency strain with an actual material system.

Keywords: Surface stresses, interface properties, mismatch, grain boundaries.

1. Introduction

The physical and chemical properties of materials are greatly influenced by the presence of surrounding surfaces and interfaces, whether interfaces are in the form of grain boundaries separating two grains or in the case of a bicrystal between two dissimilar materials (phase boundaries). Interfacial stress and interfacial energy are quantities that play key roles in the thermodynamics of solid surfaces by providing a continuum description of the underlying atomistic complexity of interfaces. Through various constitutive interfacial models, the thermodynamics of interfaces has proven to be an important factor governing the behavior of materials systems separated by an interface or for which the surface to volume ratio is significant (??????????).

Indeed, atoms near an interface experience a different local environment than atoms in the bulk of a material. In the direction normal to a surface or an interface, the periodicity of the atomic lattice loses its translational symmetry over several atomic layers as demonstrated for example by atomistic simulations (???). In the case of an interface between two dissimilar materials, at least one phase must be elastically stretched due to the lattice mismatch between both phases resulting in the formation of defects such as dislocations or vacancies. Similarly, as a consequence, the equilibrium position and energy of atoms located in the vicinity of an interface are, in general, somewhat different from their bulk counterparts. Both the translational and in-plane atomic shuffling near the interface contribute to a change in the total

21 energy of the interfacial system. The excess energy associated with atoms near an
22 interface is called *interfacial excess energy*.

23 For incoherent interfaces (e.g those between two adjacent phases), relaxation of
24 lattice mismatches may result in the formation of peculiar interfacial structures fur-
25 thering the “excess thermodynamic state” over the bulk configuration. The state
26 of interfacial coherency depends on the physical and chemical nature between both
27 phases but is also inherently dependent on external factors such as the tempera-
28 ture or stress field. Qualitatively, the contacting phases possess mismatching lattice
29 constants. For a coherent interface, the mismatch is completely accommodated by
30 straining both phases. In the case of a semi-coherent interface, localized misfit dis-
31 locations are assumed to be responsible for compensating uniform far-field elastic
32 fields, while an incoherent interface is the result of two rigid semi-infinite media in
33 rigid contact (??).

34 There are commonly three approaches adopted to study the properties of inter-
35 faces: (i) the diffuse interface model, (ii) the interphase model and (iii) the divid-
36 ing interface/sharp interface model. In the case of the diffuse interface, interfacial
37 properties are described by a smooth, but rapid transition of both fields (e.g., con-
38 centration or elastic fields) describing the interfacial properties (??). The interphase
39 approach treats the interface in a classical thermodynamic fashion, namely, an arbi-
40 trarily defined finite volume is attributed to the interphase region (hence treated as
41 a three-dimensional material system) and is assigned thermodynamic properties in
42 a classical manner (???). Finally, the concepts of dividing surface, interfacial excess
43 energy and interfacial stress are not new and were initially introduced by ?. In this
44 paradigm, the surface contributions to the thermodynamic quantities are defined as
45 the excesses over the values that would be obtained if the bulk phases retained their
46 properties constant up to a two-dimensional imaginary dividing surface embedded

47 in a three-dimensional continuum. In other words, the interface (not interphase) is
 48 a mathematical surface of zero thickness over which the thermodynamic properties
 49 change discontinuously from one bulk phase to the other. The excess amount is
 50 associated only with the dividing surface. This Gibbsian conceptual approach has
 51 been widely studied in the fields of physics and chemistry (????????????????).
 52 Despite minor differences in the formulations, all interfacial models consist of the
 53 definition of an interfacial strain and the definition of an interfacial constitutive be-
 54 havior linking the interfacial strain to the interface excess stress (also commonly
 55 called “*surface stress*”). Apart from a few exceptions, theories based on such two-
 56 dimensional framework cannot account for the flexural stiffness (????) nor can they
 57 describe the transverse behavior of real material interfaces or account for the in-
 58 terfacial mismatch. Fairly recently, for a coherent interface in an elastic solid, the
 59 so-called Shuttleworth or Shuttleworth-Herring relation (??) relating the interfacial
 60 excess energy Γ to the surface stress Σ^S has been generalized by ? to account for
 61 the three-dimensional nature of the interface in a Gibbsian context such that,

$$\Sigma^S = \left. \frac{\partial \Gamma}{\partial \epsilon^S} \right|_{\sigma^\perp} + \sigma^\perp \cdot \underline{\underline{\mathbb{H}}}, \quad (1)$$

62 where the surface stress Σ^S is not only a function of the interface in-plane strain
 63 ϵ^S (which is the case in the original Shuttleworth relation) but also a function of
 64 the transverse stress σ^\perp . The third-order interfacial tensor $\underline{\underline{\mathbb{H}}}$ measures the inherent
 65 Poisson’s effect in the transverse direction of the interface. However, in the case of
 66 a semi-coherent or incoherent interface, as discussed by ?, it is necessary to define
 67 two strain and stress measures to describe the general deformation at the interface,
 68 rendering equation (1) inadequate since both phases can in principle be stretched
 69 independently along the interface.

70 The present work proposes the use of the Gibbs dividing surface thermodynamic

71 framework to develop generalized expressions for interfacial excess stresses and inter-
72 facial excess energy and to account for their variations with respect to the in-plane
73 surface strain, the transverse stress and the interface mismatch strain. Our goal is
74 to unambiguously define interface stresses and the interfacial excess strain in order
75 to understand the connection between the interface thermodynamic quantities and
76 the interface kinetic quantities to arrive at a generalized Shuttleworth relation for
77 mismatched interfaces. In essence, our approach is close to that of ?; in fact, we
78 mostly render their formulation more general and systematic.

79 The manuscript is organized as follows. Section 2 starts with the definitions of
80 the interfacial kinematics. In sections 3 and 4, we propose a generalized formula-
81 tion of the Shuttleworth relationship when a structural mismatch is present at the
82 interface. Explicit expressions of the interfacial tensors are derived in the special
83 case of an incoherent interface between two dissimilar isotropic solids. The connec-
84 tion between the structural mismatch and elastic properties of interfaces is revealed
85 in the singular case of isotropic thermo-elasticity. Finally, numerical examples are
86 provided in section 5 for incoherent interfaces between Cu and its oxide Cu_2O to
87 illustrate the physicality and insight provided by the proposed formulation in view
88 of atomistic simulations. Note that in the following sections of this manuscript, the
89 terms interface and surface are used interchangeably. A similar remark applies to
90 the designation of the Shuttleworth or Shuttleworth-Herring relationship.

91 A list of notation and convention used throughout this manuscript is cataloged
92 in Appendix A.

93 **2. Interfacial kinematics**

94 Let us consider an incoherent planar interface between two dissimilar elastic crys-
95 talline phases subjected to an anisotropic and nonuniform elastic deformation. The

96 two half spaces are assumed to be mutually insoluble so that no diffusion occurs
97 between them and can be ignored. In the interest of a concise analysis, we choose to
98 consider only processes and equilibria at constant and uniform temperature (hence
99 neglecting configurational and thermal entropies). Therefore, and although an ex-
100 plicit temperature dependence is admitted for all free energy functions and materials
101 parameters, we do not display the temperature as a variable (lattice constants and
102 materials properties may be a function of the temperature). It is also assumed that,
103 if more than one interface is present in the material system, these interfaces do not
104 interact with one another. For thin slabs and small particles, it has been demon-
105 strated that interfaces may interact with one another, altering interfacial properties
106 and showing a finite size effect (?). This size effect is more pronounced for ionic sys-
107 tems (long range electrostatic interaction forces) than it is for more classical metallic
108 systems.

109 We begin with the definitions of interfacial strains and incoherency. The inter-
110 face \mathcal{S} is considered as an imaginary, two-dimensional surface with its location chosen
111 arbitrarily following the Gibbs dividing surface concept. For convenience, let the in-
112 terface be located at $z = 0$ in a Cartesian coordinate system as shown in Fig 1. In
113 this work we consider that the bicrystal is subjected to a homogeneous traction and
114 displacement boundary condition at infinity, such that far away from the interface
115 the deformation can be assumed homogeneous, and the transverse (second Piola-
116 Kirchhoff) stress and the in-plane (Lagrangian) strain tensors in the upper ($z > 0$)
117 and lower ($z < 0$) materials are given by $\boldsymbol{\sigma}_{\pm}^{\perp}$ and $\boldsymbol{\epsilon}_{\pm}^S$ respectively. It has been shown
118 that such homogeneous deformation can be easily constructed in a bimaterial (?)
119 by the so-called “T-decomposition”. Although the deformation far away from the
120 interface is homogeneous, the stress and strain fields near the interface are disrupted
121 by the presence of the interface and the associated disturbance of the atomic lattice

122 periodicity in its vicinity. The deformation of the interfacial system manifests itself
 123 by displacements of the atoms i which can be described by individual displacement
 124 vectors $\mathbf{v}^{(i)}$. In general, the deformation field gives rise to local stresses in the in-
 125 terfacial system, which can be related by balance laws to the stress in the interface
 126 surroundings and external forces acting on the atoms near the interface. If we as-
 127 sume that the displacement varies smoothly at the atomic scale, the equations of
 128 balance can be derived based on a description of the interfacial system as a contin-
 129 uum, where the displacement field is represented by $\mathbf{u}(\mathbf{x})$, with the vector \mathbf{x} being
 130 a continuous position variable. The treatment of the displacement fields and strain
 131 fields is measured from a reference state (Lagrangian measure) of one of the two half
 132 crystals chosen arbitrarily. The reference states for medium “+” ($z > 0$) and for
 133 medium “-” ($z < 0$) can be chosen as states of uniform composition.

134 As mentioned by ?, two measures of the Lagrangian interfacial in-plane strain
 135 tensors, $\boldsymbol{\epsilon}^S$ and $\boldsymbol{\epsilon}^{*,S}$ respectively, are necessary since each crystal has its own refer-
 136 ence state. The displacement fields $\mathbf{u}_+(\mathbf{x})$ and $\mathbf{u}_-(\mathbf{x})$ can be used for the standard
 137 definition of the elastic in-plane surface strain tensor in small deformation $\boldsymbol{\epsilon}_+^S$ and
 138 $\boldsymbol{\epsilon}_-^S$ in medium “+” and medium “-” respectively such that,

$$\boldsymbol{\epsilon}_\pm^S = \frac{1}{2} \left(\nabla^S \mathbf{u}_\pm + \nabla^S \mathbf{u}_\pm^T \right) , \quad (2)$$

139 where ∇^S is the surface gradient. The surface nabla operator ∇^S is simply defined
 140 by the projection of the nabla operator ∇ on the tangential plane of the interface
 141 such that,

$$\nabla^S = (\mathbf{I} - \mathbf{n}(\mathbf{x}^S) \otimes \mathbf{n}(\mathbf{x}^S)) \nabla , \quad (3)$$

142 where $\mathbf{n}(\mathbf{x}^S)$ is the interface unit normal upward pointing from the reference phase
 143 (in this case the “+” half space), \mathbf{x}^S is the interfacial position vector, and \mathbf{I} is the
 144 second-order identity tensor. Having the medium “+” chosen as our reference phase

145 and assuming continuity of displacement along the interface leads to $\boldsymbol{\epsilon}^S = \boldsymbol{\epsilon}_+^S = \boldsymbol{\epsilon}_-^S$.
 146 Because medium “+” is chosen as our reference, $\boldsymbol{\epsilon}^S$ is independent of $\mathbf{u}_-(\mathbf{x})$. The
 147 in-plane strain tensor $\boldsymbol{\epsilon}^S$ represents the interface strain measure associated with
 148 deforming both phases by the same amount. Another assumption stemming for the
 149 “T- decomposition” (see Appendix B for more details) leads to an interfacial normal
 150 stress continuity condition (i.e. $\boldsymbol{\sigma}^\perp = \boldsymbol{\sigma}_+^\perp = \boldsymbol{\sigma}_-^\perp$).

151 The second strain measure $\boldsymbol{\epsilon}^{*,S}$ associated with the interface represents the inter-
 152 face components of the strain of medium “-” relative to medium “+”. As illustrated
 153 in Fig. 2, following ? and ?, the strain tensor $\boldsymbol{\epsilon}^{*,S}$ is only defined in medium “-”
 154 such that it can be expressed and decomposed into two components such that,

$$\boldsymbol{\epsilon}^{*,S}(\mathbf{x}) = \boldsymbol{\epsilon}_-^{*,S}(\mathbf{x}) = \frac{1}{2} \left[(\nabla^S \mathbf{u}_- - \nabla^S \mathbf{u}_+) + (\nabla^S \mathbf{u}_- - \nabla^S \mathbf{u}_+)^T \right] = \boldsymbol{\epsilon}^{0,S} + \boldsymbol{\epsilon}^{m,S} g(\mathbf{x}) . \quad (4)$$

155 The strain tensor $\boldsymbol{\epsilon}^{0,S}$ is an eigenstrain related to stress free configuration corre-
 156 sponding to the change in molar volume between medium “+” and medium “-”.
 157 For cubic systems, the molar volume V_-^0 of stress free medium “-” is given in terms
 158 of the molar volume V_+^0 of medium “+” in its reference state such that,

$$V_-^0 = \left[1 + \epsilon_{kk}^{0,S} \right]^3 V_+^0 \approx \left[1 + 3\epsilon_{kk}^{0,S} \right] V_+^0 . \quad (5)$$

159 The strain tensor $\boldsymbol{\epsilon}^{m,S}$ represents the in-plane structural mismatch or deviation
 160 from the coherency at the interface (i.e. $z = 0$). The spatial function $g(\mathbf{x})$ defined in
 161 equation (4) corresponds to the variation of this structural mismatch over few atomic
 162 layers and vanishes far from the interface. As such, $g(0) = 1$ by definition of the
 163 in-plane structural mismatch $\boldsymbol{\epsilon}^{m,S}$, and $g(\mathbf{x}) = 0$, $\forall \mathbf{x} \leq -h$, where h is an arbitrary
 164 distance far away from the interface (see illustrations in Fig. 1 and Fig. 2). In the
 165 present manuscript (semi-infinite media), the function $g(\mathbf{x})$ does not vary with the
 166 interfacial material system size (as it would be the case for thin films, for example)

167 since we are not accounting for interfaces interactions. The structural mismatch at
 168 the interface is usually defined as (??):

$$\epsilon_{\alpha\beta}^{m,S} = 2 \frac{\ell_+ - \ell_-}{\ell_+ + \ell_-} \delta_{\alpha\beta}, \quad (6)$$

169 where ℓ_+ and ℓ_- are the crystal lattice parameters in phase “+” and “-” respectively
 170 and $\delta_{\alpha\beta}$ is the Kronecker delta symbol.

171 In light of the definitions of $\epsilon^{0,S}$ and $\epsilon^{m,S}$, the strain tensor $\epsilon^{*,S}$ corresponds to
 172 an in-plane eigenstrain related to the change of the interface structure. For example,
 173 if we consider the interface between a crystal of finite thickness and a flat thick
 174 substrate, changing the in-plane lattice spacing of the crystal while keeping the lattice
 175 spacing of the substrate constant requires the introduction of misfit dislocations at
 176 the interface to relax the interfacial system (?).

177 3. Thermodynamic framework for an incoherent interface

178 3.1. Interfacial thermodynamic framework

179 We now use the Gibbs dividing surface thermodynamic framework to define the
 180 interfacial excess free energy, interfacial excess stress and interfacial excess strain for
 181 a coherent and incoherent interface. In this continuum representation, the total free
 182 energy \mathcal{F} of the interfacial system (i.e. both semi-infinite crystal Ω_+ and Ω_- and the
 183 dividing interface \mathcal{S}) is given by

$$\mathcal{F} = \int_{\Omega_+} \Psi_+(\mathbf{x}) dV + \int_{\Omega_-} \Psi_-(\mathbf{x}) dV + \int_{\mathcal{S}} \Gamma(\mathbf{x}) dA, \quad (7)$$

184 where $\Psi_{\pm}(\mathbf{x})$ is the bulk free-energy density per unit volume in medium “+” and
 185 “-” respectively, while $\Gamma(\mathbf{x})$ is the interfacial excess free energy.

186 Both bulk phases are being strained by the same amount in the plane of the
 187 interface and loaded transversely to the interface along its normal direction (“T-
 188 decomposition”). Additionally each solid half space is subjected to eigenstrains

189 $\boldsymbol{\epsilon}_\pm^*(\mathbf{x})$ ($\boldsymbol{\epsilon}_+^*(\mathbf{x}) = 0$) in medium “+” or medium “-” respectively and the bulk con-
 190 stitutive behavior is treated in a classical manner such that,

$$\Psi_\pm = \hat{\Psi}_\pm(\nabla \mathbf{u}, \boldsymbol{\epsilon}^*, \mathbf{x}) \text{ in } \Omega_\pm, \quad (8)$$

$$d\Psi_\pm = \boldsymbol{\sigma}_\pm : d\boldsymbol{\epsilon}_\pm = \underline{\underline{\mathbb{C}}}_\pm : (\boldsymbol{\epsilon}_\pm - \boldsymbol{\epsilon}_\pm^*) d\boldsymbol{\epsilon}_\pm, \quad (9)$$

191 where $\boldsymbol{\sigma}_\pm$ is the second Piola-Kirchhoff stress tensor in medium “+” or medium “-”
 192 respectively, $\underline{\underline{\mathbb{C}}}_\pm$ is the corresponding elastic stiffness tensor and the elastic strain
 193 tensor in small deformation $\boldsymbol{\epsilon}_\pm$ is defined in a standard fashion. The bulk description
 194 is augmented by the constitutive behavior of the interface in the form,

$$\Gamma = \hat{\Gamma}(\boldsymbol{\epsilon}^S, \boldsymbol{\epsilon}^{m,S}, \boldsymbol{\sigma}^\perp, \mathbf{x}^S), \quad (10)$$

195 where $\boldsymbol{\epsilon}^S$ is the surface strain defined in equation (2), $\boldsymbol{\sigma}^\perp$ denotes the transverse
 196 stress tensor normal to the interface (based on the “T-decomposition”) and $\boldsymbol{\epsilon}^{m,S}$ is
 197 the in-plane structural mismatch strain tensor defined in equation (4).

198 The variables $(\boldsymbol{\epsilon}^S, \boldsymbol{\sigma}^\perp, \boldsymbol{\epsilon}^{m,S})$ being defined, the next task is to identify their appro-
 199 priate interfacial conjugate variables through the generalized thermodynamic frame-
 200 work for the interface free energy density for which the interfacial power density of
 201 internal forces takes the form in the most general case,

$$p^S = \boldsymbol{\Sigma}^S : \dot{\boldsymbol{\epsilon}}^S + \mathbb{D}^\perp \cdot \dot{\boldsymbol{\sigma}}^\perp + \boldsymbol{\Upsilon}^S : \dot{\boldsymbol{\epsilon}}^{m,S}. \quad (11)$$

202 In this context, the surface tensor $\boldsymbol{\Sigma}^S$ can be identified as the interfacial stress, ther-
 203 modynamic conjugate of the in-plane strain field $\boldsymbol{\epsilon}^S$, the surface tensor $\boldsymbol{\Upsilon}^S$ can be
 204 identified as the incoherent interfacial stress, thermodynamic conjugate of the inter-
 205 facial mismatch strain $\boldsymbol{\epsilon}^{m,S}$, and the tensor \mathbb{D}^\perp can be associated with the interfacial
 206 excess strain, thermodynamic conjugate of the transverse stress field $\boldsymbol{\sigma}^\perp$. For coher-
 207 ent interfaces the last term is missing since $\boldsymbol{\epsilon}^{m,S} = 0$. The term in $\boldsymbol{\Upsilon}^S : \dot{\boldsymbol{\epsilon}}^{m,S}$ can

208 be viewed as a correction to the interfacial energy due to deviation from coherency.
 209 Expressions for tensor Σ^S , \mathbb{D}^\perp and Υ^S stated as a function of the interfacial excess
 210 energy and its derivatives with respect to the state variables $(\epsilon^S, \sigma^\perp, \epsilon^{m,S})$ constitute
 211 the generalized Shuttleworth relationship (see section 3.5).

212 3.2. Interfacial excess energy

213 The interfacial excess energy Γ of a mismatched interface is defined using the
 214 standard Gibbs definition of the interfacial excess energy per unit of undeformed
 215 area combined with the ‘‘T-decomposition’’ (see Appendix B) such that,

$$\Gamma = \int_0^\infty (\Psi(\mathbf{x}) - \Psi_+) d\mathbf{x} + \int_{-\infty}^0 (\Psi(\mathbf{x}) - \Psi_-) d\mathbf{x}, \quad (12)$$

216 where

$$\begin{aligned} \Psi(\mathbf{x}) &= \hat{\Psi}_0 + \hat{\boldsymbol{\tau}}^S(\mathbf{x}) : [\boldsymbol{\epsilon}^S - \boldsymbol{\epsilon}^{*,S}(\mathbf{x})] \\ &+ \frac{1}{2} [\boldsymbol{\epsilon}^S - \boldsymbol{\epsilon}^{*,S}(\mathbf{x})] : \underline{\underline{\mathbb{C}}}^S(\mathbf{x}) : [\boldsymbol{\epsilon}^S - \boldsymbol{\epsilon}^{*,S}(\mathbf{x})] + \frac{1}{2} \boldsymbol{\sigma}^\perp \cdot \mathbb{M}^\perp(\mathbf{x}) \cdot \boldsymbol{\sigma}^\perp, \end{aligned} \quad (13)$$

217 and

$$\Psi_+ = \frac{1}{2} \boldsymbol{\epsilon}^S : \underline{\underline{\mathbb{C}}}_+^S : \boldsymbol{\epsilon}^S + \frac{1}{2} \boldsymbol{\sigma}_+^\perp \cdot \mathbb{M}_+^\perp \cdot \boldsymbol{\sigma}_+^\perp, \quad (14)$$

$$\Psi_- = \frac{1}{2} (\boldsymbol{\epsilon}^S - \boldsymbol{\epsilon}^{0,S}) : \underline{\underline{\mathbb{C}}}_-^S : (\boldsymbol{\epsilon}^S - \boldsymbol{\epsilon}^{0,S}) + \frac{1}{2} \boldsymbol{\sigma}^\perp \cdot \mathbb{M}_-^\perp \cdot \boldsymbol{\sigma}^\perp. \quad (15)$$

218 Keep in mind that equations (14) and (15) are the result of having medium ‘‘+’’ as
 219 the reference medium. As such, the eigenstrain $\boldsymbol{\epsilon}^{0,S}$ appears only in medium ‘‘-’’ and
 220 not in medium ‘‘+’’. Making use of equations (13), (14) and (15) in equation (12),
 221 it follows that the interfacial excess energy Γ can be expressed as,

$$\begin{aligned} \Gamma &= \Gamma_0 + \boldsymbol{\Gamma}^{(1)} : \boldsymbol{\epsilon}^S + \frac{1}{2} \boldsymbol{\epsilon}^S : \underline{\underline{\boldsymbol{\Gamma}}}^{(2)} : \boldsymbol{\epsilon}^S + \frac{1}{2} \boldsymbol{\sigma}^\perp \cdot \boldsymbol{\Lambda}^{(2)} \cdot \boldsymbol{\sigma}^\perp \\ &+ \boldsymbol{\Upsilon}^{(1)} : \boldsymbol{\epsilon}^{m,S} + \frac{1}{2} \boldsymbol{\epsilon}^{m,S} : \underline{\underline{\boldsymbol{\Upsilon}}}^{(2)} : \boldsymbol{\epsilon}^{m,S} - \boldsymbol{\epsilon}^{m,S} : \underline{\underline{\boldsymbol{\Phi}}} : \boldsymbol{\epsilon}^S, \end{aligned} \quad (16)$$

222 where,

$$\begin{aligned} \Gamma_0 &= \int_{-\infty}^{\infty} \hat{\Psi}_0(\mathbf{x}) d\mathbf{x} - \int_0^{\infty} \hat{\tau}^S(\mathbf{x}) \epsilon^{0,S} d\mathbf{x} \\ &+ \frac{1}{2} \int_{-\infty}^0 \epsilon^{0,S} : [\underline{\underline{\mathbb{C}}}^S(\mathbf{x}) - \underline{\underline{\mathbb{C}}}_-^S] : \epsilon^{0,S} d\mathbf{x} , \end{aligned} \quad (17)$$

$$\mathbf{\Gamma}^{(1)} = \int_{-\infty}^{\infty} \hat{\tau}^S(\mathbf{x}) d\mathbf{x} - \int_{-\infty}^0 \epsilon^{0,S} : [\underline{\underline{\mathbb{C}}}^S(\mathbf{x}) - \underline{\underline{\mathbb{C}}}_-^S] d\mathbf{x} , \quad (18)$$

$$\underline{\underline{\mathbf{\Gamma}}}^{(2)} = \int_0^{\infty} [\underline{\underline{\mathbb{C}}}^S(\mathbf{x}) - \underline{\underline{\mathbb{C}}}_+^S] d\mathbf{x} + \int_{-\infty}^0 [\underline{\underline{\mathbb{C}}}^S(\mathbf{x}) - \underline{\underline{\mathbb{C}}}_-^S] d\mathbf{x} , \quad (19)$$

$$\mathbf{\Upsilon}^{(1)} = - \int_{-\infty}^0 \hat{\tau}^S(\mathbf{x}) g(\mathbf{x}) d\mathbf{x} + \int_{-\infty}^0 \epsilon^{0,S} : \underline{\underline{\mathbb{C}}}^S(\mathbf{x}) g(\mathbf{x}) d\mathbf{x} , \quad (20)$$

$$\underline{\underline{\mathbf{\Upsilon}}}^{(2)} = \int_{-\infty}^0 \underline{\underline{\mathbb{C}}}^S(\mathbf{x}) g^2(\mathbf{x}) d\mathbf{x} , \quad (21)$$

$$\mathbf{\Lambda}^{(2)} = \int_0^{\infty} [\mathbb{M}^\perp(\mathbf{x}) - \mathbb{M}_+^\perp] d\mathbf{x} + \int_0^{\infty} [\mathbb{M}^\perp(\mathbf{x}) - \mathbb{M}_-^\perp] d\mathbf{x} , \quad (22)$$

$$\underline{\underline{\mathbf{\Phi}}} = \int_{-\infty}^0 \underline{\underline{\mathbb{C}}}^S(\mathbf{x}) g(\mathbf{x}) d\mathbf{x} . \quad (23)$$

223 Note that, even though the range of the integrals defined in equations (17)–(23)
 224 is infinite, the convergence of these integrals is satisfied. Indeed $\hat{\tau}^S(\mathbf{x})$ and $g(\mathbf{x})$
 225 both vanish far away from the interface so that the integrals only need to be car-
 226 ried out over a finite thickness in the interphase region (see Fig. 1(b) and Fig. 2).
 227 Equation (16) expresses the interfacial excess energy as a function of the in-plane
 228 strain ϵ^S , the transverse stress σ^\perp and the interfacial mismatch $\epsilon^{m,S}$. As illustrated
 229 in equation (24), note the explicit dependence and correction of the residual inter-
 230 facial excess energy Γ_0 and some of the interfacial elastic tensors due to the state of
 231 coherency of the interface through the eigenstrain $\epsilon^{0,S}$ and the short range function
 232 $g(\mathbf{x})$:

$$\Gamma|_{\epsilon^S=0, \sigma^\perp=0} = \Gamma^* = \Gamma_0 + \mathbf{\Upsilon}^{(1)} : \epsilon^{m,S} + \frac{1}{2} \epsilon^{m,S} : \underline{\underline{\mathbf{\Upsilon}}}^{(2)} : \epsilon^{m,S} . \quad (24)$$

233 *3.3. In-plane interfacial excess stress*

234 We now define the in-plane interfacial excess stress by decomposing the second
 235 Piola-Kirchhoff stress tensor in its in-plane component and transverse component
 236 (see equation (B.9) in Appendix B), so that we obtain

$$\boldsymbol{\sigma}^S(\mathbf{x}) = \hat{\boldsymbol{\tau}}^S(\mathbf{x}) + \underline{\underline{\mathbb{C}}}^S(\mathbf{x}) : [\boldsymbol{\epsilon}^S - \boldsymbol{\epsilon}^{*,S}(\mathbf{x})] + \boldsymbol{\sigma}^\perp \cdot \underline{\underline{\boldsymbol{\gamma}}}(\mathbf{x}) . \quad (25)$$

237 Furthermore in the bulk of medium “+” and medium “-”,

$$\boldsymbol{\sigma}_+^S = \underline{\underline{\mathbb{C}}}_+^S : \boldsymbol{\epsilon}_+^S + \boldsymbol{\sigma}^\perp \cdot \underline{\underline{\boldsymbol{\gamma}}}_+ , \quad (26)$$

$$\boldsymbol{\sigma}_-^S = \underline{\underline{\mathbb{C}}}_-^S : (\boldsymbol{\epsilon}_-^S - \boldsymbol{\epsilon}^{0,S}) + \boldsymbol{\sigma}^\perp \cdot \underline{\underline{\boldsymbol{\gamma}}}_- . \quad (27)$$

238 Again equations (26) and (27) are the result of having medium “+” as the reference
 239 medium. Following ?, we can define the (second Piola-Kirchhoff) interfacial excess
 240 stress by,

$$\boldsymbol{\Sigma}^S = \int_0^\infty (\boldsymbol{\sigma}^S(\mathbf{x}) - \boldsymbol{\sigma}_+^S) d\mathbf{x} + \int_{-\infty}^0 (\boldsymbol{\sigma}^S(\mathbf{x}) - \boldsymbol{\sigma}_-^S) d\mathbf{x} . \quad (28)$$

241 It results from strain compatibility ($\boldsymbol{\epsilon}^S = \boldsymbol{\epsilon}_+^S = \boldsymbol{\epsilon}_-^S$), traction continuity ($\boldsymbol{\sigma}^\perp = \boldsymbol{\sigma}_+^\perp =$
 242 $\boldsymbol{\sigma}_-^\perp$) and equations (25)–(27) that,

$$\boldsymbol{\Sigma}^S = \boldsymbol{\Gamma}^{(1)} - \underline{\underline{\boldsymbol{\Phi}}} : \boldsymbol{\epsilon}^{m,S} + \underline{\underline{\boldsymbol{\Gamma}}}^{(2)} : \boldsymbol{\epsilon}^S + \boldsymbol{\sigma}^\perp \cdot \underline{\underline{\boldsymbol{\mathbb{H}}}} , \quad (29)$$

243 where $\boldsymbol{\Gamma}^{(1)}$, $\underline{\underline{\boldsymbol{\Gamma}}}^{(2)}$, $\underline{\underline{\boldsymbol{\Phi}}}$ are given by equations (18)–(23) and $\underline{\underline{\boldsymbol{\mathbb{H}}}}$ reads

$$\underline{\underline{\boldsymbol{\mathbb{H}}}} = \int_0^\infty [\underline{\underline{\boldsymbol{\gamma}}}(\mathbf{x}) - \underline{\underline{\boldsymbol{\gamma}}}_+] d\mathbf{x} + \int_{-\infty}^0 [\underline{\underline{\boldsymbol{\gamma}}}(\mathbf{x}) - \underline{\underline{\boldsymbol{\gamma}}}_-] d\mathbf{x} . \quad (30)$$

244 The first term $\boldsymbol{\Gamma}^{(1)}$ represents the internal residual excess stress of the interface. This
 245 component of the surface stress exists even when a remote interfacial deformation
 246 is absent. Note that two factors contribute to this residual surface stress (see equa-
 247 tion (18)). One can be attributed to the pure elastic behavior of the interface (see

248 term in $\hat{\boldsymbol{\tau}}^S$ in equation (18)). This contribution has been discussed and derived in a
 249 similar fashion by others (??). The second contribution can be ascribed to the cou-
 250 pling between the state of coherency, the change in molar volume between the two
 251 media and the elastic stiffness of the interface (see term in $\boldsymbol{\epsilon}^{0,S}$ in equation (18)). The
 252 term $\underline{\underline{\Phi}}$ describes the intrinsic coupling between the surface stress and the structural
 253 interfacial mismatch. The third term $\underline{\underline{\Gamma}}^{(2)}$ represents the interface in-plane elastic
 254 stiffness tensor, while the tensor $\underline{\underline{\mathbb{H}}}$ measures the Poisson's effect of the interface in
 255 the direction normal to the interface. Note that the interface coherency does not af-
 256 fect the surface stiffness (surface elastic tensors $\underline{\underline{\Gamma}}^{(2)}$ and $\underline{\underline{\mathbb{H}}}$), but does have an effect
 257 on the residual surface stress $\boldsymbol{\Gamma}^{(1)}$ and the coupling with the structural mismatch $\underline{\underline{\Phi}}$.
 258 As such the first two terms of equation (29) could be viewed as a correction to the
 259 coherent residual surface stress due to the mismatch at the interface, while the last
 260 two terms are solely due to the elastic stiffness and compliance of the interface.

261 3.4. Transverse interfacial excess strain

262 Similarly, if we consider the transverse interfacial excess strain by decompos-
 263 ing the transverse Lagrangian strain according to the ‘‘T-decomposition’’ (see equa-
 264 tion (B.7) in Appendix B), we get,

$$\boldsymbol{\epsilon}^\perp(\mathbf{x}) = \boldsymbol{\epsilon}^{\perp,*}(\mathbf{x}) - \mathbb{M}^\perp(\mathbf{x}) \cdot \boldsymbol{\tau}^\perp(\mathbf{x}) + \mathbb{M}^\perp(\mathbf{x}) \cdot \boldsymbol{\sigma}^\perp - \underline{\underline{\gamma}}(\mathbf{x}) : [\boldsymbol{\epsilon}^S - \boldsymbol{\epsilon}^{*,S}(\mathbf{x})] . \quad (31)$$

265 with the transverse strain in the bulk of each medium given by,

$$\boldsymbol{\epsilon}_+^\perp = \mathbb{M}_+^\perp \cdot \boldsymbol{\sigma}^\perp - \underline{\underline{\gamma}}_+ : \boldsymbol{\epsilon}^S , \quad (32)$$

$$\boldsymbol{\epsilon}_-^\perp = \boldsymbol{\epsilon}^{\perp,0} + \mathbb{M}_-^\perp \cdot \boldsymbol{\sigma}^\perp - \underline{\underline{\gamma}}_- : (\boldsymbol{\epsilon}^S - \boldsymbol{\epsilon}^{0,S}) . \quad (33)$$

266 Here again equations (32) and (33) are the result of having medium ‘‘+’’ as the
 267 reference medium.

268 The excess transverse interfacial strain is defined by,

$$\Delta^\perp = \int_0^\infty (\epsilon^\perp(\mathbf{x}) - \epsilon_+^\perp) d\mathbf{x} + \int_{-\infty}^0 (\epsilon^\perp(\mathbf{x}) - \epsilon_-^\perp) d\mathbf{x}, \quad (34)$$

269 which becomes,

$$\Delta^\perp = \Lambda^{(1)} + \underline{\mathbb{K}} : \epsilon^{m,S} + \Lambda^{(2)} \cdot \sigma^\perp - \underline{\mathbb{H}} : \epsilon^S, \quad (35)$$

270 with the tensors $\Lambda^{(1)}$ and $\underline{\mathbb{K}}$ defined by,

$$\begin{aligned} \Lambda^{(1)} &= \int_{-\infty}^0 (\epsilon^{\perp,*}(\mathbf{x}) - \epsilon^{\perp,0}) + (\underline{\gamma}(\mathbf{x}) - \underline{\gamma}_-) : \epsilon^{0,S} d\mathbf{x} \\ &- \int_{-\infty}^\infty \mathbb{M}^\perp(\mathbf{x}) \cdot \tau^\perp(\mathbf{x}) d\mathbf{x}, \end{aligned} \quad (36)$$

$$\underline{\mathbb{K}} = \int_{-\infty}^0 \underline{\gamma}(\mathbf{x}) g(\mathbf{x}) d\mathbf{x}. \quad (37)$$

271 The transverse interfacial excess strain tensor Δ^\perp measures the interfacial excess
 272 deformation in the direction transverse to the interface. The first term $\Lambda^{(1)}$ repre-
 273 sents the part of transverse interfacial deformation that exists even when a remote
 274 interfacial deformation is absent. Its physical interpretation could be viewed as an
 275 “intrinsic relaxed thickness” of the interface as compared with an interface which
 276 would retain a perfect bulk lattice arrangement up to the dividing surface. Similarly
 277 to the residual surface stress, two elements compose the “intrinsic relaxed thickness”
 278 of the interface. One comes from the elastic transverse compliance across the in-
 279 terface (see term in \mathbb{M}^\perp in equation (36)) and the other stems from the coupling
 280 between the interface elastic behavior and the interface state of coherency (see term
 281 in $\epsilon^{0,S}$ in equation (36)). The tensor $\Lambda^{(2)}$ describes the transverse compliance of the
 282 interface, while the tensor $\underline{\mathbb{K}}$ represents the coupling between the transverse loading
 283 and the structural mismatch at the interface. Analogously to the remarks made for
 284 the in-plane interfacial excess stress, note that the first two terms of equation (35)
 285 could be viewed as a correction to the “intrinsic relaxed thickness” for a coherent

286 interface due to the mismatch at the interface, while the surface coherency does not
 287 affect the interface elastic compliance (tensor $\Lambda^{(2)}$).

288 3.5. Generalized Shuttleworth relationship

289 Based on the thermodynamic derivations presented above, the generalized form
 290 of the Shuttleworth relationship for a mismatched interface can be obtained. By
 291 combining equations (11) with equations (16) and (29), the relation between the
 292 coherent surface stress Σ^S and the interfacial energy Γ is given by,

$$\Sigma^S = \left. \frac{\partial \Gamma}{\partial \epsilon^S} \right|_{\epsilon^{m,S}, \sigma^\perp} + \sigma^\perp \cdot \underline{\mathbb{H}}, \quad (38)$$

293 while the relation between the interfacial strain Δ^\perp and the interfacial energy Γ is
 294 simply obtained by combining equations (11) with equations (16) and (35) such that,

$$\mathbb{D}^\perp = \Delta^\perp - \Lambda^{(1)} = \left. \frac{\partial \Gamma}{\partial \sigma^\perp} \right|_{\epsilon^S, \epsilon^{m,S}} - \underline{\mathbb{H}} : \epsilon^S + \underline{\mathbb{K}} : \epsilon^{m,S}. \quad (39)$$

295 Note that equations (38) and (39) are consistent with that of ? in the above equa-
 296 tion (1) and reduce to the original formulation of Shuttleworth-Herring when $\sigma^\perp = 0$
 297 and $\epsilon^{m,S} = 0$. The term $\Lambda^{(1)} + \underline{\mathbb{K}} : \epsilon^{m,S}$ corresponds to a correction due to the struc-
 298 ture of the interface (through the mismatch strain) to the intrinsic “relaxation width”
 299 of the interface accounting for the relative movement between the top and bottom
 300 “surfaces” of the interfacial region, even when $\sigma^\perp = 0$ and $\epsilon^S = 0$ (i.e., when the
 301 remote traction the in-plane strain vanishes). Additionally, the coherent residual
 302 surface stress comprises a correction due to (i) the change in molar volume between
 303 medium “+” and medium “-” through the eigenstrain $\epsilon^{0,S}$ in the two-dimensional
 304 tensor $\Gamma^{(1)}$ and (ii) the intrinsic misfit structural coupling through the tensor $\underline{\Phi}$.

305 Finally, by integrating the interfacial power density of internal forces in equa-
 306 tion (11) in conjunction with equation (16) and equations (38) and (39) such that,

$$\Gamma = \int [\Sigma^S : d\epsilon^S + (\Delta^\perp - \Lambda^{(1)}) \cdot d\sigma^\perp + \Upsilon^S : d\epsilon^{m,S}] , \quad (40)$$

307 we obtain a complementary Shuttleworth equation relating the incoherent surface
 308 stress Υ^S to the interfacial excess energy Γ such that,

$$\Upsilon^S = \left. \frac{\partial \Gamma}{\partial \epsilon^{m,S}} \right|_{\epsilon^S, \sigma^\perp} - \sigma^\perp \cdot \underline{\mathbb{K}} + \underline{\Phi} : \epsilon^S . \quad (41)$$

309 All together, equations (38), (39) and (41) constitute the generalized Shuttleworth
 310 relationship for incoherent interfaces. The coherent surface stress Σ^S and the trans-
 311 verse excess strain Δ^\perp represent the thermodynamic driving forces deforming the
 312 interface while the incoherent surface stress Υ^S represents the driving force of stretch-
 313 ing one crystal while holding the other fixed and therefore altering the structure of the
 314 interface. This generalized formulation not only accounts for the three-dimensional
 315 nature of the interface in a Gibbsian sense (2D mathematical object as opposed to
 316 an interphase model) but also explicitly considers its interfacial structure. The three
 317 thermodynamics quantities described above fully characterize the elastic behavior of
 318 coherent and incoherent interfaces under general loading conditions. The extension
 319 to curved interfaces will be briefly discussed in section 3.6.

320 Several differences between the present formulation and that of ? are observed.
 321 Equation (41) is consistent with the incoherent stress introduced in ? when $\sigma^\perp = 0$
 322 and $\epsilon^S = 0$. The absence of the in-plane strain in their formulation is essentially due
 323 to the fact that they expanded the excess free energy Γ in terms of ϵ^S and $\epsilon^{m,S}$ only
 324 to the first order (no surface elasticity).

325 From a surface stress point of view, three major differences need to be noted.
 326 First, the coherent and incoherent surface stresses derived in equations (29), (38)
 327 and (41) reduce in the case of ? to the residual surface stresses such that,

$$\Sigma_{CL}^S = \Gamma^{(1)} \quad \text{and} \quad \Upsilon_{CL}^S = \Upsilon^{(1)} , \quad (42)$$

328 where the subscript ‘‘CL’’ denotes the ? formulation. Second, the expression for the
 329 two-dimensional tensor $\Gamma^{(1)}$ in ? does not account for the contribution attributed to

330 the coupling between the state of coherency, the change in molar volume between
 331 the two media and the elastic stiffness of the interface (see second integral in equa-
 332 tion (18)). Finally neither the intrinsic coupling between the surface stress and the
 333 structural interfacial mismatch (tensor $\underline{\Phi}$) nor the transverse interfacial behavior
 334 (tensors \mathbb{D}^\perp , \mathbb{H} and \mathbb{K}) are accounted for.

335 From an interfacial energy point of view, the interfacial power density derived in
 336 equation (11) simplifies in the case of ? to,

$$p_{CL}^S = \mathbf{\Gamma}^{(1)} : \dot{\boldsymbol{\epsilon}}^S + \boldsymbol{\Upsilon}^{(1)} : \boldsymbol{\epsilon}^{m,S} , \quad (43)$$

337 in which neither the surface elasticity ($\underline{\mathbf{\Gamma}}^{(2)}$) nor the couplings both in terms of struc-
 338 tural mismatch and transverse behavior are accounted for (see equation (29)). This
 339 simplification makes the interfacial excess energy Γ_{CL} simply quadratic in terms of
 340 the in-plane surface strain $\boldsymbol{\epsilon}^S$ and mismatch strain $\boldsymbol{\epsilon}^{m,S}$, while it is shown in equa-
 341 tion (16) that the evolution of the interfacial excess energy as a function of the loading
 342 path ($\boldsymbol{\epsilon}^S, \boldsymbol{\sigma}^\perp$) and the structural mismatch strain $\boldsymbol{\epsilon}^{m,S}$ is not that straightforward.

343 3.6. Discussion on the equilibrium condition of incoherent curved interfaces

344 We have intentionally limited ourselves to planar interfaces between two semi-
 345 infinite media when dealing with surface excess stresses and surface excess strain
 346 in the formulation described above. However, in quite a few practical engineering
 347 applications (e.g. composites reinforced with nanoparticles), interfaces can be curved
 348 surfaces, not planar. In that case, and in the limit where the interface characteristic
 349 length scale is much smaller than the interface radius of curvature, the constitutive
 350 relationships describing the interface behavior not only need to account for the in-
 351 plane and transverse behavior (i.e. interfacial Poisson's effect) as well as the interface
 352 structural mismatch (as it is described in this manuscript), but also the intrinsic

353 flexural resistance. As mentioned in section 1, few models have been presented
 354 in the literature to account for the flexural stiffness (????). In this context, the
 355 interfacial excess energy is a function of the local in-plane deformation and also
 356 comprises a dependence on the curvature introducing an intrinsic flexural resistance
 357 of the interface. As described very recently in ?, the interfacial power density of
 358 internal forces for curved interfaces takes the form,

$$p^S = \Sigma^S : \dot{\epsilon}^S + \mathbf{M}^S : \dot{\boldsymbol{\kappa}} , \quad (44)$$

359 where \mathbf{M}^S is the interface bending moment conjugate of the relative curvature $\boldsymbol{\kappa}$.
 360 While accounting for the curvature dependence, these interfacial flexural formulations
 361 do not account for the interfacial mismatch or the interfacial transverse behavior (see
 362 equation (11)). For curved interfaces, the present formulation would need to include
 363 the last term of equation (44) and the interfacial kinematics presented in section 2
 364 would need to be revisited in order to account for the curvature dependence (see for
 365 example section 2.2 in ?). Additionally, as discussed elsewhere (see equation (5.76)
 366 in ?), due to the “T-decomposition” used in the present formulation, supplemental
 367 conditions for the displacement jump across the curved mismatched interface would
 368 need to be considered. In other words, an additional incoherent thermodynamic
 369 driving force (which could be called the incoherent interfacial bending moment) cor-
 370 responding to flexural deformation of the interface would need to be introduced for
 371 curved interfaces. Such an extension of the formulation is beyond the scope of the
 372 present manuscript but will be presented in a subsequent study by the authors.

373 Besides the constitutive relationships describing the interface behavior, the equi-
 374 librium condition of curved coherent interfaces is expressed by the generalized Young-
 375 Laplace equation and describes the equilibrium relationship between the interface
 376 (coherent) stress and the stress in the bulk. For incoherent mismatched curved

377 interfaces, the principle of minimum potential energy can be used to derive the gen-
 378 eralized Young-Laplace equation by accounting for (i) the interfacial excess energy,
 379 which would depend not only on the interface strains (coherent and incoherent) but
 380 also on the interface curvature; (ii) the elastic strain energy of the bulk, which would
 381 include the residual elastic field induced by the interfacial excess energy and eigen-
 382 strains due to the mismatch across the interface; and (iii) the potential of the external
 383 loads. The generalized Young-Laplace equation will also be complemented by the
 384 displacement jump condition described in the above paragraph. Again, such an ex-
 385 tension of the formulation is beyond the scope of the present manuscript but will be
 386 presented in the aforementioned study by the authors. Such a paradigm could be
 387 used to fully describe heterogeneous solids containing incoherent curved interfaces
 388 and inhomogeneities.

389 4. Interfacial isotropic elasticity formulation

390 As an illustration of the general formulation presented above, we consider in
 391 this section a special case in which both semi-infinite crystals are isotropic elastic
 392 materials with the following bulk elastic moduli,

$$393 \quad \mathbb{C}_{ijkl,\pm} = \lambda_{\pm} \delta_{ij} \delta_{kl} + \mu_{\pm} (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}) . \quad (45)$$

394 Using the “T-decomposition” derived in Appendix B through equations (B.8)
 395 and (B.10) along with equation (45), we obtain the following expressions for the
 elastic tensors characterizing the behavior of the bulk phases in the case of elastic

396 isotropic materials,

$$\mathbb{C}_{3k3j,\pm} = (\lambda_{\pm} + \mu_{\pm}) \delta_{3k} \delta_{3j} + \mu_{\pm} \delta_{kj} , \quad (46)$$

$$\mathbb{M}_{kj,\pm} = -\frac{\lambda_{\pm} + \mu_{\pm}}{(\lambda_{\pm} + 2\mu_{\pm}) \mu_{\pm}} \delta_{3k} \delta_{3j} + \frac{1}{\mu_{\pm}} \delta_{kj} , \quad (47)$$

$$\gamma_{i\alpha\beta,\pm} = \frac{\lambda_{\pm}}{\lambda_{\pm} + 2\mu_{\pm}} \delta_{3i} \delta_{\alpha\beta} , \quad (48)$$

$$\mathbb{C}_{\alpha\beta\kappa\lambda,\pm}^S = \frac{2\lambda_{\pm}\mu_{\pm}}{\lambda_{\pm} + 2\mu_{\pm}} \delta_{\alpha\beta} \delta_{\kappa\lambda} + \mu_{\pm} (\delta_{\alpha\kappa} \delta_{\beta\lambda} + \delta_{\alpha\lambda} \delta_{\beta\kappa}) , \quad (49)$$

$$\hat{\tau}_{\alpha\beta,\pm}^S = \tau_{\alpha\beta}^S - \tau_3^{\perp} \frac{\lambda_{\pm}}{\lambda_{\pm} + 2\mu_{\pm}} \delta_{\alpha\beta} . \quad (50)$$

397 4.1. In-plane interfacial excess stress

398 Regarding the in-plane interfacial excess stress Σ^S , by using equations (28)
399 and (46)–(50), we immediately find

$$\Sigma_{\alpha\beta}^S = \Gamma_{\alpha\beta}^{(1)} - \Phi_{\alpha\beta\kappa\lambda} \epsilon_{\kappa\lambda}^{m,S} + \Gamma_{\alpha\beta\kappa\lambda}^{(2)} \epsilon_{\kappa\lambda}^S + \sigma_i^{\perp} \mathbb{H}_{i\alpha\beta} , \quad (51)$$

400 with the elastic surface tensors defined by,

$$\begin{aligned} \Gamma_{\alpha\beta}^{(1)} &= \int_{-\infty}^{\infty} \left[\tau_{\alpha\beta}^S(\mathbf{x}) - \frac{\tau_3^{\perp}(\mathbf{x}) \lambda(\mathbf{x})}{\lambda(\mathbf{x}) + 2\mu(\mathbf{x})} \delta_{\alpha\beta} \right] d\mathbf{x} \\ &- \int_{-\infty}^0 \left[\left(\frac{2\lambda(\mathbf{x}) \mu(\mathbf{x})}{\lambda(\mathbf{x}) + 2\mu(\mathbf{x})} - \frac{2\lambda_- \mu_-}{\lambda_- + 2\mu_-} \right) \delta_{\alpha\beta} \epsilon_{\kappa\kappa}^{0,S} + 2(\mu(\mathbf{x}) - \mu_-) \epsilon_{\alpha\beta}^{0,S} \right] d\mathbf{x} \end{aligned} \quad (52)$$

$$\Gamma_{\alpha\beta\kappa\lambda}^{(2)} = \lambda^S \delta_{\alpha\beta} \delta_{\kappa\lambda} + \mu^S (\delta_{\alpha\kappa} \delta_{\beta\lambda} + \delta_{\alpha\lambda} \delta_{\beta\kappa}) , \quad (53)$$

$$\mathbb{H}_{i\alpha\beta} = \frac{2K^S \nu^S}{E^{\perp}} \delta_{3i} \delta_{\alpha\beta} , \quad (54)$$

$$\Phi_{\alpha\beta\kappa\lambda} = \int_{-\infty}^0 \left[\frac{2\lambda(\mathbf{x}) \mu(\mathbf{x})}{\lambda(\mathbf{x}) + 2\mu(\mathbf{x})} \delta_{\alpha\beta} \delta_{\kappa\lambda} + \mu(\mathbf{x}) (\delta_{\alpha\kappa} \delta_{\beta\lambda} + \delta_{\alpha\lambda} \delta_{\beta\kappa}) \right] g(\mathbf{x}) d\mathbf{x} . \quad (55)$$

401 Taking equations (52)–(55) together with equation (51) gives the in-plane inter-
402 facial excess stress as follows

$$\Sigma_{\alpha\beta}^S = \Gamma_{\alpha\beta}^{(1)} - \lambda^{*,S} \delta_{\alpha\beta} \epsilon_{\kappa\kappa}^{m,S} - 2\mu^{*,S} \epsilon_{\alpha\beta}^{m,S} + \lambda^S \delta_{\alpha\beta} \epsilon_{\kappa\kappa}^S + 2\mu^S \epsilon_{\alpha\beta}^S + \frac{2K^S \nu^S}{E^{\perp}} \sigma_3^{\perp} \delta_{\alpha\beta} , \quad (56)$$

403 where the two new terms compared to ? are the ones containing the in-plane struc-
 404 tural mismatch embodied by $\epsilon_{\alpha\beta}^{m,S}$. In the case of coherent interfaces, i.e. $\epsilon_{\alpha\beta}^{m,S} = 0$,
 405 following ?, the formulation developed by ? would be sufficient only if both mate-
 406 rials have same stress-free molar volumes for which $\epsilon_{\alpha\beta}^{0,S}$ vanishes. In equation (54),
 407 $K^S = \lambda^S + \mu^S$ and the surface elastic constants are defined by,

$$\mu^S = \int_0^\infty (\mu(\mathbf{x}) - \mu_+) d\mathbf{x} + \int_{-\infty}^0 (\mu(\mathbf{x}) - \mu_-) d\mathbf{x} , \quad (57)$$

$$\begin{aligned} \lambda^S &= \int_0^\infty \left(\frac{2\lambda(\mathbf{x})\mu(\mathbf{x})}{\lambda(\mathbf{x}) + 2\mu(\mathbf{x})} - \frac{2\lambda_+\mu_+}{\lambda_+ + 2\mu_+} \right) d\mathbf{x} \\ &+ \int_{-\infty}^0 \left(\frac{2\lambda(\mathbf{x})\mu(\mathbf{x})}{\lambda(\mathbf{x}) + 2\mu(\mathbf{x})} - \frac{2\lambda_-\mu_-}{\lambda_- + 2\mu_-} \right) d\mathbf{x} , \end{aligned} \quad (58)$$

$$\mu^{*,S} = \int_{-\infty}^0 \mu(\mathbf{x}) g(\mathbf{x}) d\mathbf{x} , \quad (59)$$

$$\lambda^{*,S} = \int_{-\infty}^0 \frac{2\lambda(\mathbf{x})\mu(\mathbf{x})}{\lambda(\mathbf{x}) + 2\mu(\mathbf{x})} g(\mathbf{x}) d\mathbf{x} , \quad (60)$$

$$\begin{aligned} \frac{2K^S\nu^S}{E^\perp} &= \int_0^\infty \left(\frac{\lambda(\mathbf{x})}{\lambda(\mathbf{x}) + 2\mu(\mathbf{x})} - \frac{\lambda_+}{\lambda_+ + 2\mu_+} \right) d\mathbf{x} \\ &+ \int_{-\infty}^0 \left(\frac{\lambda(\mathbf{x})}{\lambda(\mathbf{x}) + 2\mu(\mathbf{x})} - \frac{\lambda_-}{\lambda_- + 2\mu_-} \right) d\mathbf{x} . \end{aligned} \quad (61)$$

408 Through this exercise several points are noteworthy:

- 409 • In equation (56), λ^S and μ^S , which could be defined as the surface Lamé
 410 constants, are consistent (in terms of their definition and analytical expressions)
 411 with other classical formulations derived by many authors for surface stress in
 412 solids (??????).
- 413 • The effective transverse surface “Poisson’s ratio” $2K^S\nu^S/E^\perp$ in equation (61)
 414 is consistent with the formulation for an interface containing no mismatch and
 415 stress-free strains (?).

416 • The present formulation introduces two new surface constants $\mu^{*,S}$ and $\lambda^{*,S}$
 417 defined in equations (59)–(60). These constants are solely related to the inter-
 418 facial incoherency and the in-plane structural mismatch strain $\epsilon_{\alpha\beta}^{m,S}$ and corre-
 419 spond to a correction to the interfacial residual stress $\Gamma_{\alpha\beta}^{(1)}$. To our knowledge,
 420 their expressions and physical origins are presented for the first time in the
 421 literature.

422 With this in mind, the in-plane interfacial excess stress components yield

$$\Sigma_{11}^S = \Gamma_{11}^{(1)} - \lambda^{*,S} \epsilon_{\kappa\kappa}^{m,S} - 2\mu^{*,S} \epsilon_{11}^{m,S} + \lambda^S \epsilon_{\kappa\kappa}^S + 2\mu^S \epsilon_{11}^S + \frac{2K^S \nu^S}{E^\perp} \sigma_3^\perp, \quad (62)$$

$$\Sigma_{22}^S = \Gamma_{22}^{(1)} - \lambda^{*,S} \epsilon_{\kappa\kappa}^{m,S} - 2\mu^{*,S} \epsilon_{22}^{m,S} + \lambda^S \epsilon_{\kappa\kappa}^S + 2\mu^S \epsilon_{22}^S + \frac{2K^S \nu^S}{E^\perp} \sigma_3^\perp, \quad (63)$$

$$\Sigma_{12}^S = \Gamma_{12}^{(1)} - 2\mu^{*,S} \epsilon_{12}^{m,S} + 2\mu^S \epsilon_{12}^S. \quad (64)$$

423 Thus, from equations (62)–(64), μ^S and $\mu^{*,S}$ are in-plane interfacial shear moduli as-
 424 sociated with in-plane interfacial shear strain ϵ_{12}^S and in-plane incoherent mismatched
 425 shear strain $\epsilon_{12}^{m,S}$ respectively. The surface elastic moduli λ^S and $\lambda^{*,S}$ are in-plane
 426 interfacial Lamé constants. Furthermore, the in-plane biaxial hydrostatic stress is
 427 given by $\Sigma_{\alpha\alpha}^S/2$ where

$$\Sigma_{\alpha\alpha}^S = \Gamma_{\alpha\alpha}^{(1)} - 2K^{*,S} \epsilon_{\kappa\kappa}^{m,S} + 2K^S \epsilon_{\kappa\kappa}^S + \frac{4K^S \nu^S}{E^\perp} \sigma_3^\perp, \quad (65)$$

428 where $K^{S*} = \mu^{*,S} + \lambda^{*,S}$ can be interpreted as the in-plane bi-axial surface modulus of
 429 the incoherent interface associated with the structural mismatch (or deviation from
 430 coherency) $\epsilon_{\kappa\kappa}^{m,S}$.

431 4.2. Transverse interfacial excess strain

432 Regarding the transverse interfacial excess strain, using equations (35) along
 433 with (46)–(50), we find the interfacial excess strain,

$$\Delta_i^\perp = \Lambda_i^{(1)} + \mathbb{K}_{i\alpha\beta} \epsilon_{\alpha\beta}^{m,S} + \Lambda_{ij}^{(2)} \sigma_j^\perp - \mathbb{H}_{i\alpha\beta} \epsilon_{\alpha\beta}^S, \quad (66)$$

434 with the transverse surface tensors defined by,

$$\begin{aligned}\Lambda_i^{(1)} &= \int_{-\infty}^{\infty} \left[\frac{\lambda(\mathbf{x}) + \mu(\mathbf{x})}{(\lambda(\mathbf{x}) + 2\mu(\mathbf{x}))\mu(\mathbf{x})} \delta_{3i} \tau_3^\perp(\mathbf{x}) - \frac{1}{\mu(\mathbf{x})} \tau_i^\perp(\mathbf{x}) \right] d\mathbf{x} \\ &+ \int_{-\infty}^0 \left[\left(\frac{\lambda(\mathbf{x})}{\lambda(\mathbf{x}) + 2\mu(\mathbf{x})} - \frac{\lambda_-}{\lambda_- + 2\mu_-} \right) \delta_{3i} \epsilon_{\kappa\kappa}^{0,S} \right] d\mathbf{x} \\ &+ \int_{-\infty}^0 [\epsilon_i^{\perp*}(\mathbf{x}) - \epsilon_i^{\perp 0}] d\mathbf{x},\end{aligned}\quad (67)$$

$$\Lambda_{ij}^{(2)} = \left(\frac{1}{E^\perp} - \frac{1}{\mu^\perp} - \frac{4K^S \nu^S}{E^{\perp 2}} \right) \delta_{3i} \delta_{3j} + \frac{1}{\mu^\perp} \delta_{ij}, \quad (68)$$

$$\mathbb{K}_{i\alpha\beta} = \int_{-\infty}^0 \frac{\lambda(\mathbf{x})}{\lambda(\mathbf{x}) + 2\mu(\mathbf{x})} g(\mathbf{x}) \delta_{3i} \delta_{\alpha\beta} d\mathbf{x}. \quad (69)$$

435 Similarly to the formulation for the surface stress, taking equation (66) together
436 with the expressions for the transverse surface tensors yields

$$\Delta_i^\perp = \Lambda_i^{(1)} + \frac{2K^{*,S} \nu^{*,S}}{E^{*,\perp}} \delta_{3i} \epsilon_{\kappa\kappa}^{m,S} + \left(\frac{1}{E^\perp} - \frac{1}{\mu^\perp} - \frac{4K^S \nu^S}{E^{\perp 2}} \right) \delta_{3i} \sigma_3^\perp + \frac{1}{\mu^\perp} \sigma_i^\perp - \frac{2K^S \nu^S}{E^\perp} \delta_{3i} \epsilon_{\kappa\kappa}^S, \quad (70)$$

437 where the transverse elastic constant associated with the structural mismatch can
438 be defined as,

$$\frac{2K^{*,S} \nu^{*,S}}{E^{*,\perp}} = \int_{-\infty}^0 \frac{\lambda(\mathbf{x})}{\lambda(\mathbf{x}) + 2\mu(\mathbf{x})} g(\mathbf{x}) d\mathbf{x}, \quad (71)$$

439 and,

$$\frac{1}{\mu^\perp} = \int_0^\infty \left[\frac{1}{\mu(\mathbf{x})} - \frac{1}{\mu_+} \right] d\mathbf{x} + \int_{-\infty}^0 \left[\frac{1}{\mu(\mathbf{x})} - \frac{1}{\mu_-} \right] d\mathbf{x}, \quad (72)$$

$$\begin{aligned}\frac{1}{E^\perp} - \frac{4K^S \nu^S}{E^{\perp 2}} &= \int_0^\infty \left[\frac{1}{\lambda(\mathbf{x}) + 2\mu(\mathbf{x})} - \frac{1}{\lambda_+ + 2\mu_+} \right] d\mathbf{x} \\ &+ \int_{-\infty}^0 \left[\frac{1}{\lambda(\mathbf{x}) + 2\mu(\mathbf{x})} - \frac{1}{\lambda_- + 2\mu_-} \right] d\mathbf{x}.\end{aligned}\quad (73)$$

440 It follows that the components of the interfacial transverse excess strain can be

441 expressed as,

$$\Delta_1^\perp = \Lambda_1^{(1)} + \frac{1}{\mu^\perp} \sigma_1^\perp, \quad (74)$$

$$\Delta_2^\perp = \Lambda_2^{(1)} + \frac{1}{\mu^\perp} \sigma_2^\perp, \quad (75)$$

$$\Delta_3^\perp = \Lambda_3^{(1)} + 2K^{*,S} \left(\frac{\nu^{*,S}}{E^{*,\perp}} - \frac{\nu^S}{E^\perp} \right) \epsilon_{\kappa\kappa}^{m,S} + \frac{1}{E^\perp} (\sigma_3^\perp - \nu^S (\Sigma_{\alpha\alpha}^S - \Gamma_{\alpha\alpha}^{(1)})). \quad (76)$$

442 It is again noteworthy to point out that $\Lambda_i^{(1)}$ and the second term in Δ_3^\perp are
 443 new when compared to ?. Their origin stems from the (stress-free) eigenstrains and
 444 structural mismatch for incoherent interfaces, i.e. for $\epsilon_{\kappa\kappa}^{m,S} \neq 0$.

445 4.3. Interfacial thermo-elastic properties

446 Of particular interest (see for example the Levin's formula and Hill's connections
 447 problems in ?) is the case for which we assume that the interfacial incoherency is
 448 due to thermal strains associated with an isotropic in-plane interfacial coefficient of
 449 thermal expansion (CTE) denoted α^S , i.e.

$$\epsilon_{\alpha\beta}^{m,S} = \alpha^S \delta_{\alpha\beta} \Delta T, \quad (77)$$

450 where ΔT is the temperature difference. In the bulk of both media, assuming an
 451 isotropic CTE, the eigenstrain $\epsilon^{0,S}$ can simply be expressed by,

$$\epsilon_{\alpha\beta}^{0,S} = (\alpha_- - \alpha_+) \delta_{\alpha\beta} \Delta T, \quad (78)$$

452 where α_- and α_+ are the bulk CTEs for media “-” and “+” respectively while the
 453 reference medium is assumed to be the medium “+”.

454 Thus, the expression for the isotropic surface stress in equation (56) reduces to

$$\Sigma_{\alpha\beta}^S = \Gamma_{\alpha\beta}^{(1)} - d_0^* \delta_{\alpha\beta} \Delta T + \lambda^S \delta_{\alpha\beta} \epsilon_{\kappa\kappa}^S + 2\mu^S \epsilon_{\alpha\beta}^S + \frac{2K^S \nu^S}{E^\perp} \sigma_3^\perp \delta_{\alpha\beta}, \quad (79)$$

455 and,

$$d_0^* = 2 (\lambda^{*,S} + \mu^{*,S}) \alpha^S = 2K^{*,S} \alpha^S . \quad (80)$$

456 The present formulation sheds light on the in-plane thermal-mechanical connection
457 of the interface and its elastic origin assuming a resulting incoherent interface due
458 to temperature difference. This thermal-mechanical relationship is however different
459 from that of ?? and ?. For instance, ? reported the following equation (see equation
460 (3) in their paper):

$$\Sigma_{\alpha\beta}^S = \lambda^S \delta_{\alpha\beta} \epsilon_{\kappa\kappa}^S + 2\mu^S \epsilon_{\alpha\beta}^S - d_0 \delta_{\alpha\beta} \Delta T , \quad (81)$$

461 where d_0 is assumed by the authors to be $d_0 = 2 (\lambda^S + \mu^S) \alpha^S$. Several differences
462 are observed. First, the contribution of the residual surface stress $\Gamma_{\alpha\beta}^{(1)}$ and the
463 contribution of the transverse stress are not taken into account in ?. This is due
464 to the choice of the interfacial constitutive behavior used in their paper. Second,
465 the contribution due to the interfacial mismatch through the term $(\lambda^{*,S} + \mu^{*,S})$ (as
466 defined by our equations (59) and (60)) is arbitrarily taken as $(\lambda^S + \mu^S)$ in ?. In
467 the present formulation, the interpretation of the thermal-mechanical connection
468 d_0^* is unambiguously and explicitly associated with the structural mismatch at the
469 interface coming from temperature change. In turn, this can ultimately be related
470 to interfacial defects and associated deformation fields. This is not the case for d_0
471 in the formulation of ? which appears to be empirically introduced. Note here that
472 the thermo-elastic coupling is spatially defined at every vector position starting from
473 $\mathbf{x} = 0$ to a few atomic distance close to the interface in medium “-”.

474 Additionally, to complete the description of the thermo-elasticity of the interface,
475 the interfacial constitutive behavior is also accompanied by equation (70) which

476 reduces to:

$$\Delta_i^\perp = \Lambda_i^{(1)} + \frac{2d_0^* \nu^{*,S}}{E^{*,\perp}} \delta_{3i} \Delta T + \left(\frac{1}{E^\perp} - \frac{1}{\mu^\perp} - \frac{4K^S \nu^{S^2}}{E^{\perp 2}} \right) \delta_{3i} \sigma_3^\perp + \frac{1}{\mu^\perp} \sigma_i^\perp - \frac{2K^S \nu^S}{E^\perp} \delta_{3i} \epsilon_{\kappa\kappa}^S. \quad (82)$$

477 In particular, the transverse excess strain along the normal to the interface is given
478 by,

$$\Delta_3^\perp = \Lambda_3^{(1)} + \frac{2d_0^* \nu^{*,S}}{E^{*,\perp}} \Delta T + \frac{1}{E^\perp} \left(1 - \frac{4K^S \nu^{S^2}}{E^{\perp 2}} \right) \sigma_3^\perp - \frac{2K^S \nu^S}{E^\perp} \epsilon_{\kappa\kappa}^S. \quad (83)$$

479 Equations (82) and (83) are extensions of ? to thermo-elastic interfaces and were
480 not considered in the works of ?? and ?.

481 4.4. *Insight on interfacial elasticity*

482 An important point is raised in the sections above: deforming a coherent or
483 incoherent interface does not always increase its interfacial excess energy. Indeed,
484 the interfacial excess energy decreases if a deformation path reduces the interface
485 coherent surface, the interfacial excess strain, or the interface incoherent stress. This
486 assertion can have important consequences for problems in which the incorporation
487 of interfacial properties play an important role (???). Indeed, the in-plane strain,
488 transverse stress and mismatch strain can be considered as three process control
489 variables (with the first two being associated with the deformation path and the latter
490 one being associated with the interface structure) that can accentuate or minimize
491 the role of interfacial elasticity.

492 For instance, in the case of thermo-elasticity provided in section 4.3, the loading
493 space for which the coherent surface stress Σ^S and the interfacial excess strain Δ^\perp
494 vanish is of particular interest. Using equation (79) for the biaxial surface stress
495 ($\epsilon_{11}^S = \epsilon_{22}^S = \epsilon^S$) and equation (83) for the transverse interfacial excess strain, this
496 configuration would correspond to the loading path $(\epsilon^S, \sigma^\perp)$ satisfying the following

497 equation:

$$\begin{bmatrix} \frac{\Gamma_{11}^{(1)}}{2} \\ \Lambda_3^{(1)} \end{bmatrix} + \begin{bmatrix} 2K^S & \frac{2K^S\nu^S}{E^\perp} & -d_0^* \\ -\frac{4K^S\nu^S}{E^\perp} & \frac{1}{E^\perp} \left(1 - \frac{4K^S\nu^{S2}}{E^{\perp 2}}\right) & \frac{2d_0^*\nu^{*,S}}{E^{*,\perp}} \end{bmatrix} \cdot \begin{bmatrix} \epsilon^S \\ \sigma^\perp \\ \Delta T \end{bmatrix} = \begin{bmatrix} 0 \\ 0 \end{bmatrix}. \quad (84)$$

498 Equation (84) can be parametrized to obtain the loading path line in the $(\epsilon^S, \sigma^\perp)$
 499 space as a function of the interfacial elastic constants $(K^S, \nu^S, E^\perp, \nu^{*,S}, E^{*,\perp}, d_0^*)$
 500 and the temperature difference ΔT . Clearly, this illustrates the concept that one
 501 can construct a loading path that would minimize the impact of the interface on the
 502 behavior of a material system separated by it or construct a loading path for which
 503 the surface to volume ratio is significant.

504 5. Illustration from atomistic simulations of Cu/Cu₂O interfaces

505 As discussed in the previous sections, the interfacial excess energy is a function of
 506 the in-plane surface strain, the transverse stress and the interface mismatch strain.
 507 In this section we illustrate the formulation developed above with examples based
 508 on atomistic simulations for incoherent interfaces between Cu and its oxide Cu₂O
 509 under various loading configurations.

510 5.1. Atomistic model for Cu(111)/Cu₂O(111) interface and simulation methodology

511 To demonstrate the generalized formulation of a mismatched interface from a
 512 discrete medium point of view, we consider as an example a flat incoherent interface
 513 between Cu and its oxide Cu₂O. As shown in Fig. 3, the crystallographic orientation
 514 used for the present atomistic simulations is chosen as the most probable observed
 515 interface between Cu and its oxide Cu₂O (??????). As such, we consider Cu and
 516 Cu₂O blocks oriented in the (111) direction when the interface is created. Note that
 517 these surfaces are isotropic in the plane of the interface.

518 The interatomic potential based on pairwise interactions proposed by ? is used
519 to describe the Cu₂O properties while the embedded-atom method (EAM) potential
520 developed by ? is employed for the Cu metallic phase. The cross atomic interactions
521 between Cu and Cu₂O is modeled through (i) a 9–6 Lennard-Jones (LJ) potential
522 for the interaction between copper metallic atoms and copper atoms from the oxide,
523 and (ii) a Morse potential linking copper metallic atoms with the oxygen atoms.
524 Note that there are two types of metallic bonds in this model: the Cu-Cu non-bond
525 interaction in the copper oxide which is modeled by the 9–6 LJ potential and the
526 Cu-Cu metallic bond modeled by EAM potential. The bond interaction between the
527 oxygen and copper species defined by the Morse potential remains the same whether
528 the Cu atoms belong either to the pure copper phase or the copper oxide compounds
529 (?).

530 Various interfacial structural mismatches are considered by varying the overlap
531 between the surface lattice vectors of each material during the matching process
532 between both phases. Matching Cu and Cu₂O blocks presents some issues in terms
533 of coherency since the Cu₂O (111) unit cell ($l_{\text{Cu}_2\text{O}} = 6.04825 \text{ \AA}$) is larger than the Cu
534 (111) unit cell ($l_{\text{Cu}} = 2.5542 \text{ \AA}$). Following equation (6), in this atomistic context,
535 the structural mismatch is simply defined as,

$$\epsilon_{\alpha\beta}^{m,S} = 2 \frac{n l_{\text{Cu}} - m l_{\text{Cu}_2\text{O}}}{n l_{\text{Cu}} + m l_{\text{Cu}_2\text{O}}} \delta_{\alpha\beta} , \quad (85)$$

536 where l_{Cu} and $l_{\text{Cu}_2\text{O}}$ are the lattice constants of Cu and Cu₂O respectively and (n, m)
537 are scaling factors (taken as integers) used to obtain various mismatch strains. To
538 minimize the structural mismatch, several sets of couple (m, n) have been chosen
539 such that the overall strain is small when the interface is formed.

540 The interface model dimensions and mismatch strains simulated are summarized
541 in Table 1. A typical computation cell contains on average 20,000 atoms composed

542 of N_{Cu} atoms in the Cu phase and $N_{\text{Cu}_2\text{O}}$ atoms in the Cu_2O phase. Periodic bound-
543 ary conditions are used in all directions. Note that the use of periodic boundary
544 conditions in the transverse direction implicitly introduces a second interface at the
545 edge of the calculation box with an identical structure as the interface located at the
546 center of atomic assembly. Dimensions of the simulation box (W, D, L) have been
547 chosen such that atoms far away from the interface have energy levels correspond-
548 ing to the perfect bulk configuration. In other words, the two interfaces present in
549 the atomistic system do not interact, therefore avoiding the size effect mentioned in
550 section 2.

551 Once the initial interfacial assembly is created, a molecular statics calculation,
552 which employs a nonlinear conjugate gradient method, is used to determine the
553 minimum energy interface configuration (i.e., relaxed positions of the atoms) and
554 to find the self equilibrium state of the interface. Due to the in-plane periodicity,
555 relaxation of the structure occurs only in the direction perpendicular to the bound-
556 ary plane. The transverse stress in the relaxed configuration is therefore null. A
557 detailed discussion of interface atomistic structural rearrangement throughout the
558 entire simulation process and resulting interfacial point defects is beyond the scope
559 of the present manuscript.

560 After the minimum interfacial energy configuration is obtained, the interfacial
561 assembly is elastically deformed either by applying a transverse tensile stress σ_3^\perp or
562 by applying biaxial strains $\epsilon_{11}^S = \epsilon_{22}^S = \epsilon^S$ of a couple percents in the interfacial plane.
563 Each of these stresses and biaxial strains is less than the critical load required for
564 defect nucleation (dislocation notably) for the interfaces considered in this work, so
565 that we remain in the elastic domain. In the configuration when the tensile stress
566 is applied, the x - and y -direction boundaries are specified as stress-free throughout
567 deformation.

568 For each deformation process described in the above, MS simulations are per-
 569 formed to compute the interfacial excess energy at the equilibrium state of the sys-
 570 tem. In the case of the discrete atomistic systems used in this work, the integral of
 571 the energy profile defined in equation (12) is discretized into a summation over a set
 572 of atoms within each region, rigorously separating the energy profile normal to the
 573 interface into two regions (?) such that,

$$\Gamma = \frac{1}{2A_0} \left[\sum_{i=1}^{N_{\text{Cu}}} \left(e^{(i)}(z) - e_{\text{Cu}}^{(0)} \right) + \sum_{i=1}^{N_{\text{Cu}_2\text{O}}} \left(e^{(i)}(z) - e_{\text{Cu}_2\text{O}}^{(0)} \right) \right], \quad (86)$$

574 where $e^{(i)}$ is the total energy of atom i enclosed by the area A_0 on the interface, N_{Cu}
 575 and $N_{\text{Cu}_2\text{O}}$ are the number of atoms in the copper region and its oxide respectively,
 576 and $e_{\text{Cu}}^{(0)}$ and $e_{\text{Cu}_2\text{O}}^{(0)}$ are the total energies of an atom in a perfect lattice (bulk config-
 577 uration) far away from the surface in medium Cu and medium Cu_2O , respectively.
 578 The bulk energies, $e_{\text{Cu}}^{(0)}$ and $e_{\text{Cu}_2\text{O}}^{(0)}$, are determined by averaging the total energy $e^{(i)}$
 579 of a group of atoms positioned sufficiently far away from the interface such that the
 580 presence of the boundary can be ignored. Using $e_{\text{Cu}}^{(0)}$ and $e_{\text{Cu}_2\text{O}}^{(0)}$, an excess energy is
 581 computed for every atom within the interface model; the sum of these excess energies
 582 is defined as the total interface energy. Note that the factor 2 in the denominator
 583 of equation (86) accounts for the presence of the two actual interfaces within the
 584 atomic assembly due to the periodic boundary conditions.

585 5.2. Comparisons between the discrete atomistic simulations and the generalized 586 Shuttleworth relationships

587 The above procedure has been used to calculate the variation of the interfacial
 588 excess energy under biaxial straining or transverse loading. The interfacial elastic
 589 properties used for the comparison between the atomistic simulations and the theo-
 590 retical model presented in the above are listed in Table 2. These interfacial elastic

591 properties have been obtained numerically using a least-square method accounting
 592 for the various loading paths used the atomistic system based on equations (87)–
 593 (89), thus limiting the number of interfacial elastic constants that can be calculated
 594 through this method. Note that the semi-analytical methodology developed by ? in
 595 the case of the EAM potential could be used as an alternative to derive the interfa-
 596 cial properties. However this formulation would need to be modified to account for
 597 the complexity of the two interatomic potentials and cross atomic interactions used
 598 in this manuscript for the Cu/Cu₂O interfacial system. Such modifications of the
 599 methodology are beyond the scope of the present manuscript.

600 Looking first at the interfacial excess energy as a function of the mismatch strain
 601 $\epsilon^{m,S}$ when no load is applied ($\epsilon^S = 0$ and $\sigma^\perp = 0$ MPa), equation (16) reduces to

$$\Gamma|_{\epsilon^S=0, \sigma^\perp=0} = \Gamma^* = \Gamma_0 + \Upsilon_{\alpha\alpha}^{(1)} \epsilon_{\alpha\alpha}^{m,S} + \frac{1}{2} \epsilon_{\alpha\alpha}^{m,S} \Upsilon_{\alpha\alpha\beta\beta}^{(2)} \epsilon_{\beta\beta}^{m,S} . \quad (87)$$

602 The MS simulation results for the variation of the interfacial excess energy as a
 603 function of the mismatch strain are shown in Fig. 4 together with the results from
 604 the present continuum model. It is seen that these results agree very well with those
 605 obtained by the generalized interfacial formulation and show a quadratic behavior.

606 Considering now the variation of interfacial excess energy as a function of the
 607 transverse stress σ^\perp for various mismatch strains (no in-plane strain applied), the
 608 interfacial excess energy Γ in equation (16) reduces to

$$\Gamma|_{\epsilon^S=0} = \Gamma^* + \frac{1}{2} \sigma_3^\perp \Lambda_{33}^{(2)} \sigma_3^\perp . \quad (88)$$

609 We again observe a very good agreement between the atomistic simulation results
 610 and the present theoretical formulation as it is plotted in Fig. 5.

611 Similarly, in the case of the variation of interfacial excess energy as a function of

612 the biaxial strain ϵ^S (no transverse stress applied), equation (16) reduces to

$$\Gamma|_{\sigma^\perp=0} = \Gamma^* + \Gamma_{\alpha\alpha}^{(1)}\epsilon_{\alpha\alpha}^S + \frac{1}{2}\epsilon_{\alpha\alpha}^S\Gamma_{\alpha\alpha\beta\beta}^{(2)}\epsilon_{\beta\beta}^S - \epsilon_{\alpha\alpha}^{m,S}\Phi_{\alpha\alpha\beta\beta}\epsilon_{\beta\beta}^S. \quad (89)$$

613 Comparison between the atomistic simulations and the theoretical models are pre-
 614 sented in Fig. 6. The results of the present model are in good agreement with those
 615 obtained by full-blown atomistic simulations.

616 6. Conclusion

617 The main contribution of this manuscript is the derivation of a generalized contin-
 618 uum framework describing the elastic behavior of coherent and incoherent interfaces
 619 under general loading conditions based on the Gibbs dividing surface concept com-
 620 bined with the so-called ‘‘T-decomposition’’. This generalized Shuttleworth-Herring
 621 formulation considers the equilibrium of mismatched interfaces as a function of the
 622 in-plane strain ϵ^S , the transverse stress σ^\perp and the structural mismatch strain $\epsilon^{m,S}$.
 623 This formulation not only accounts for the three-dimensional nature of the interface
 624 in a Gibbsian sense (2D mathematical object as opposed to an interphase model)
 625 but also explicitly considers its interfacial structure. Two surface stresses are thus
 626 introduced: a coherent surface stress Σ^S and an incoherent surface stress Υ^S , along
 627 with a transverse excess strain Δ^\perp . The coherent surface stress Σ^S and the trans-
 628 verse excess strain Δ^\perp represent the thermodynamic driving forces of stretching the
 629 interface while the incoherent surface stress Υ^S represents the driving force of alter-
 630 ing the structure of the interface by stretching one crystal while holding the other
 631 fixed. These three quantities fully characterize the elastic behavior of coherent and

632 incoherent interfaces and read:

$$\begin{aligned}
 \Sigma^S &= \left. \frac{\partial \Gamma}{\partial \epsilon^S} \right|_{\epsilon^{m,S}, \sigma^\perp} + \sigma^\perp \cdot \underline{\mathbb{H}}, \\
 \Delta^\perp - \Lambda^{(1)} &= \left. \frac{\partial \Gamma}{\partial \sigma^\perp} \right|_{\epsilon^S, \epsilon^{m,S}} - \underline{\mathbb{H}} : \epsilon^S + \underline{\mathbb{K}} : \epsilon^{m,S}, \\
 \Upsilon^S &= \left. \frac{\partial \Gamma}{\partial \epsilon^{m,S}} \right|_{\epsilon^S, \sigma^\perp} - \sigma^\perp \cdot \underline{\mathbb{K}} + \underline{\Phi} : \epsilon^S.
 \end{aligned}$$

633 This formulation naturally introduces interfacial elastic tensors, as well as an im-
 634 plicit coupling between the interfacial structure and interfacial stresses. Three sur-
 635 face tensors ($\underline{\Gamma}^{(2)}$, $\Lambda^{(2)}$, $\underline{\mathbb{H}}$) describe the pure elastic behavior of the interface (i.e.,
 636 independent of the structural mismatch) while four surface tensors ($\Gamma^{(1)}$, $\Lambda^{(1)}$, $\underline{\Phi}$,
 637 $\underline{\mathbb{K}}$) describe and/or account for the coupling between the interface and the state of
 638 coherency (i.e. dependent on the structural mismatch).

639 The isotropic case is developed in detail and particular attention is paid to the
 640 case of interfacial thermo-elasticity resulting from the mismatch of coefficient of
 641 thermal expansions between two dissimilar materials. This exercise provides some
 642 insight on the physical significance of the interfacial elastic constants introduced in
 643 the formulation and illustrates the obvious coupling between the interface structure
 644 and its associated thermodynamics quantities.

645 Comparisons between discrete atomistic simulations for incoherent interfaces be-
 646 tween Cu and its oxide Cu_2O and the present continuum formulation demonstrate
 647 that this generalized framework for mismatched interfaces successfully characterizes
 648 the elastic behavior of such interfaces.

649 Acknowledgments

650 A.H. would like to thank the UMI GTL/CNRS (UMI 2958) and Pr. M. Cherkaoui
 651 for their valuable support for part of this work along with the support from the Euro-

652 pean Commission for partial funding of this work under the “NanoInterface” project
653 (NMP-2008-214371). S.B. would also like to thank the support of the French govern-
654 ment through the National Research Agency (ANR) under the program “Investment
655 in the future” (Labex DAMAS referenced as ANR-11-LABX-0008-01).

656 Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed
657 Martin Company, for the United States Department of Energy, under Contract No.
658 DE-AC04-94AL85000.

Accepted Manuscript

Table 1: Geometric parameters used to set up the atomistic simulations.

Interface	$\epsilon^{m,S}$ (%)	n	m	L (Å)	D (Å)	$N_{\text{Cu}} + N_{\text{Cu}_2\text{O}}$
1	-1.476	14	6	218.628	35.923	19696
2	1.34	12	5	217.391	30.479	14088
3	5.42	15	6	215.181	37.645	21204
4	2.524	17	7	216.757	43.017	27964

Table 2: Interfacial elastic properties for an incoherent Cu/Cu₂O interface.

Property		Value
Γ_0	[J.m ⁻²]	1.102
$\Gamma_{11}^{(1)}$	[J.m ⁻²]	3.00
$\Gamma_{1111}^{(2)} + \Gamma_{1122}^{(2)}$	[J.m ⁻²]	2455.51
$\Lambda_{33}^{(2)}$	[nm.GPa ⁻¹]	0.0210
$\Upsilon_{11}^{(1)}$	[J.m ⁻²]	0.213
$\Upsilon_{1111}^{(2)} + \Upsilon_{1122}^{(2)}$	[J.m ⁻²]	526.71
$\Phi_{1111} + \Phi_{1122}$	[J.m ⁻²]	-9.24

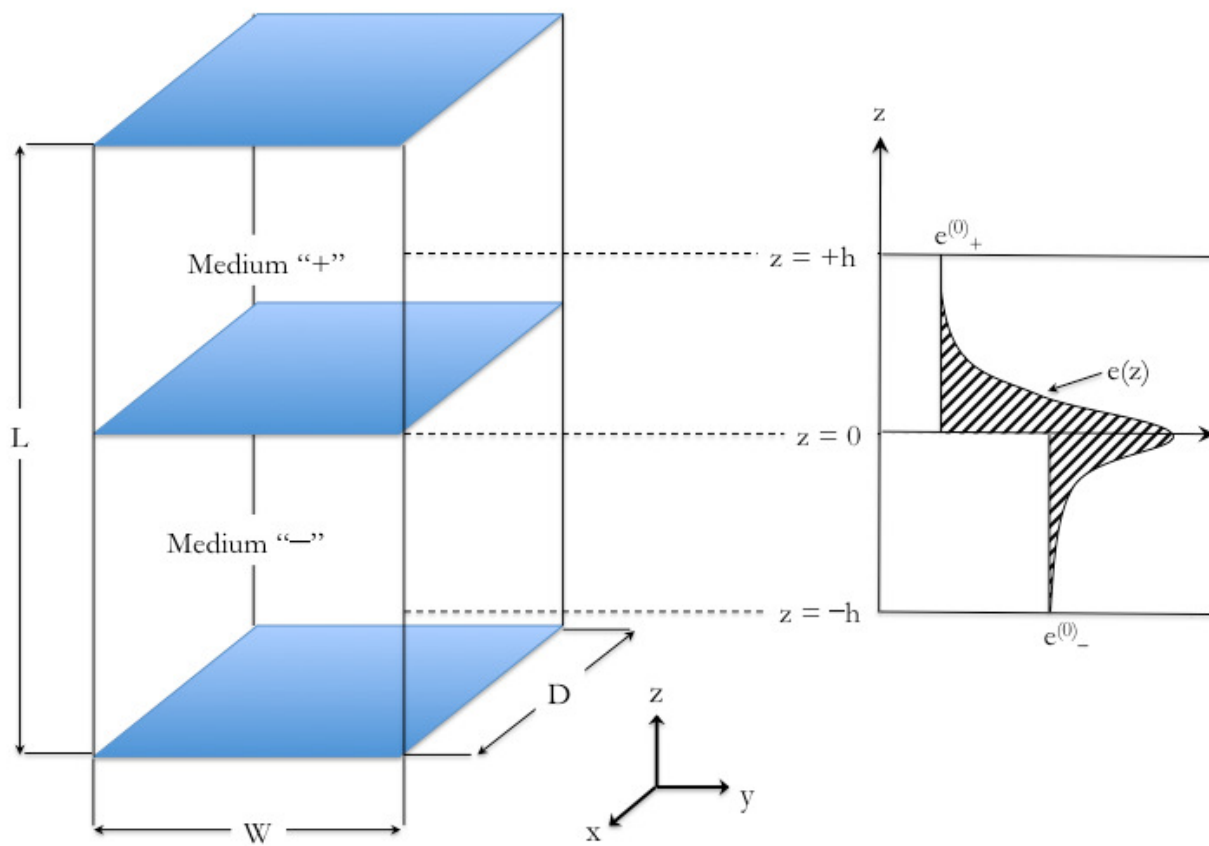


Figure 1: Schematic of a bicrystal interface and the distribution of a given excess thermodynamic quantity across the interface structure along the transverse direction z .

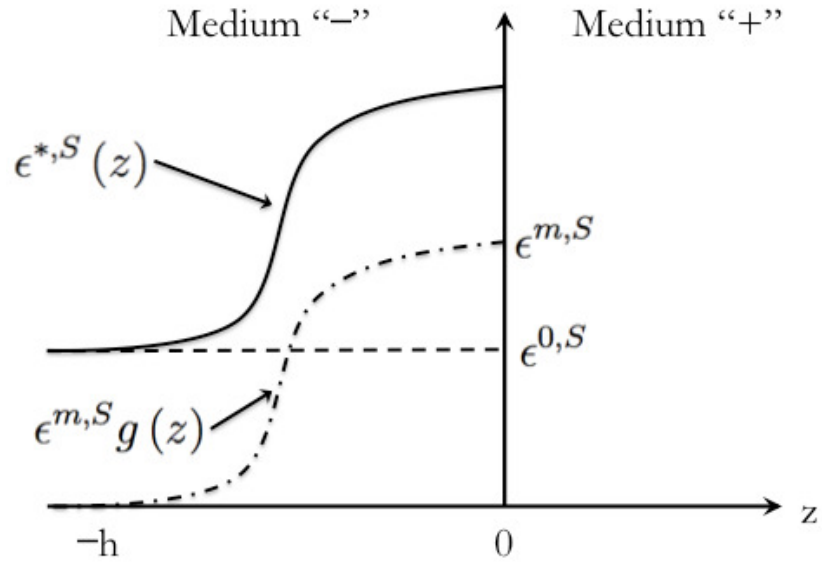


Figure 2: Decomposition of the in-plane eigenstrain $\epsilon^{*,S}$ related to the change of the interface structure.

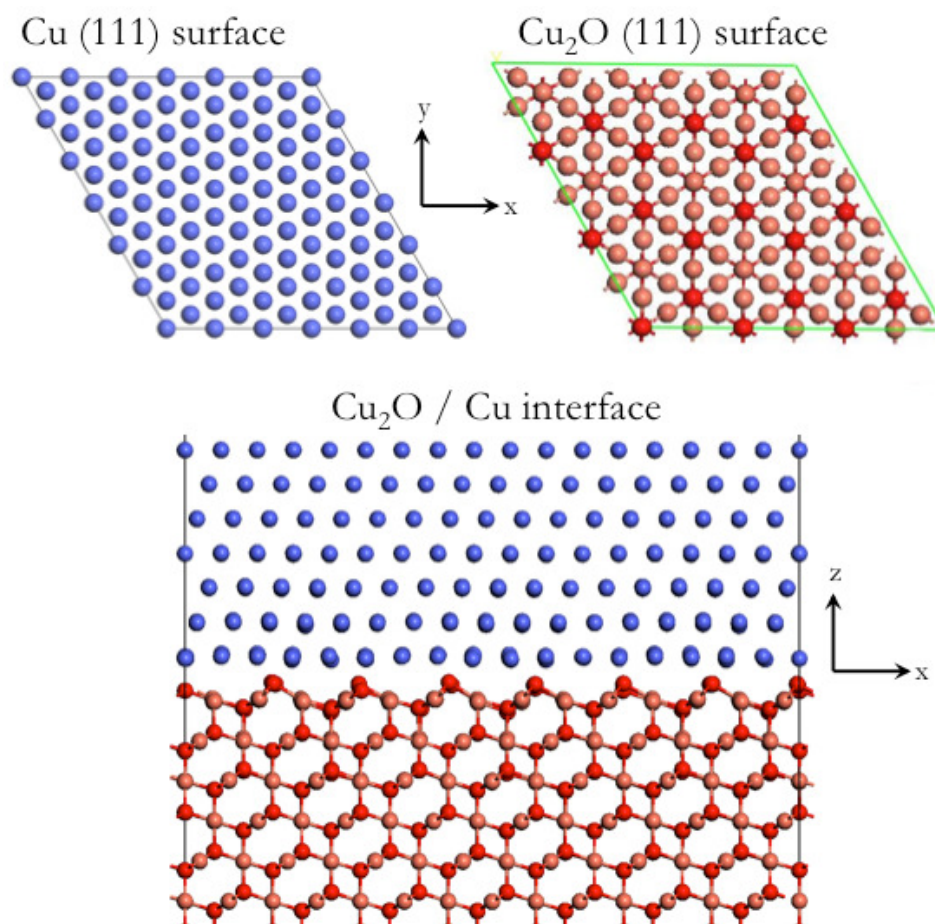


Figure 3: Illustration of the atomic surfaces composing the Cu/Cu₂O interface. Details of the setup of atomistic simulations are summarized in Table 1.

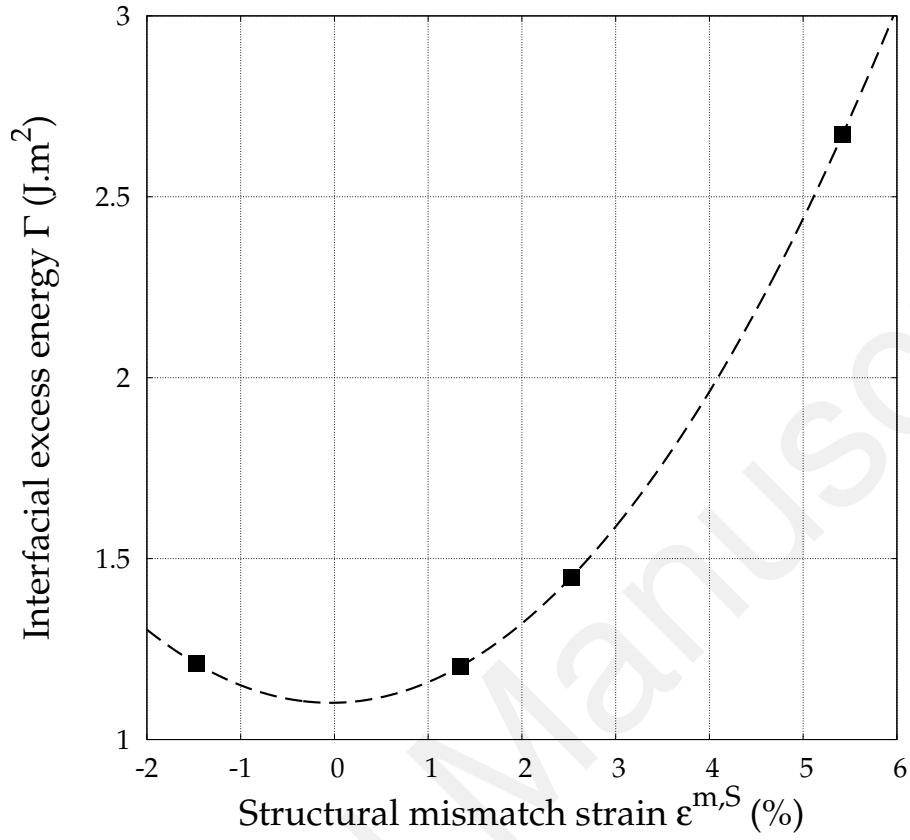
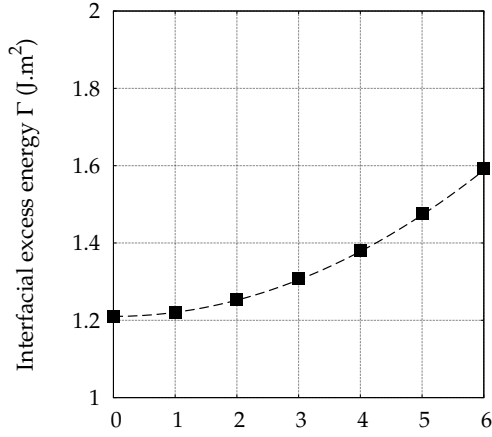
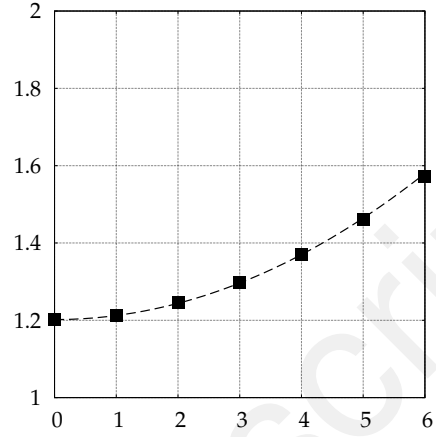


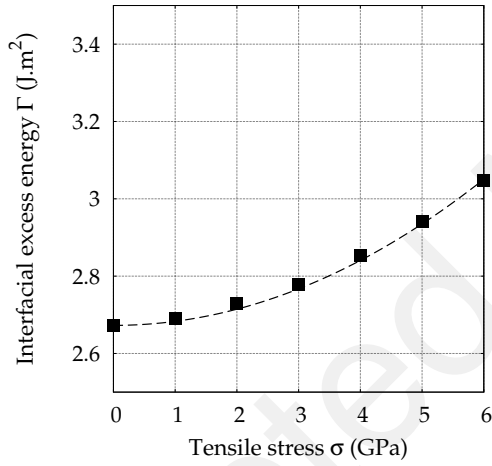
Figure 4: Variation of the interfacial excess energy $\Gamma|_{\epsilon^S=0, \sigma^\perp=0}$ as a function of the mismatch strain $\epsilon^{m,S}$ for an incoherent Cu/Cu₂O (for $\epsilon^S = 0$ and $\sigma^\perp = 0$). Dashed line (---) is based on the theoretical framework presented in equation (16), square symbols (■) are from atomistic simulations. Interfacial elasticity properties used in equation (16) are presented in Table 2.



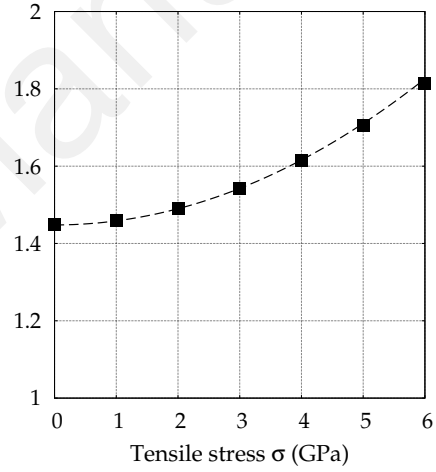
(a) Mismatch strain: $\epsilon^{m,S} = -1.476\%$



(b) Mismatch strain: $\epsilon^{m,S} = 1.34\%$

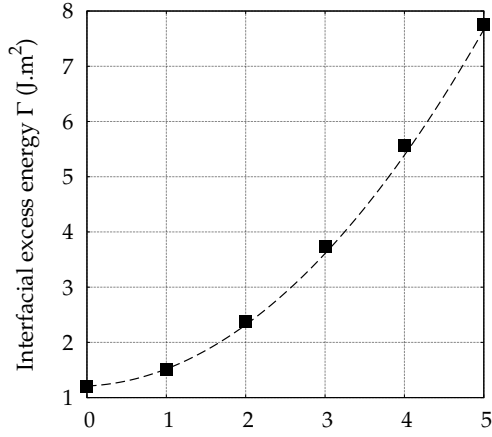


(c) Mismatch strain: $\epsilon^{m,S} = 5.421\%$

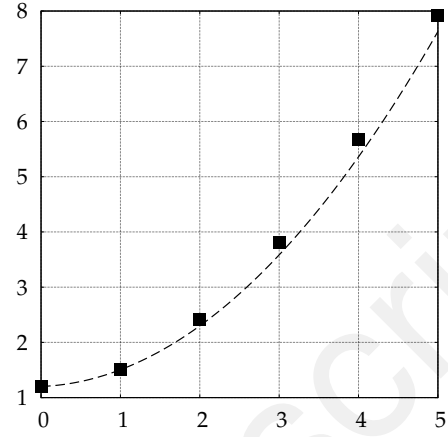


(d) Mismatch strain: $\epsilon^{m,S} = 2.524\%$

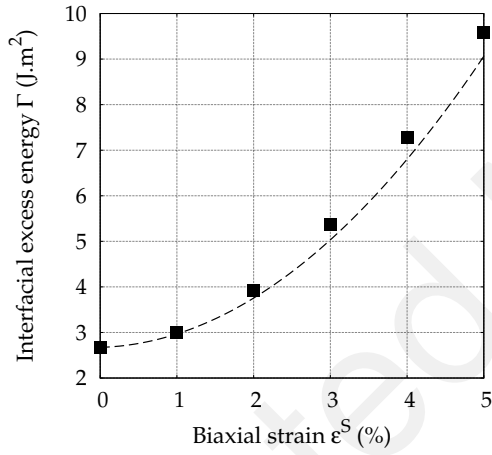
Figure 5: Variation of the interfacial excess energy $\Gamma|_{\epsilon^S=0}$ as a function of the transverse tensile stress σ^\perp (for $\epsilon^S = 0$) for an incoherent Cu/Cu₂O interface with various mismatch strains. Dashed line (---) is based on the theoretical framework presented in equation (16), square symbols (■) are from atomistic simulations. Interfacial elasticity properties used in equation (16) are presented in Table 2.



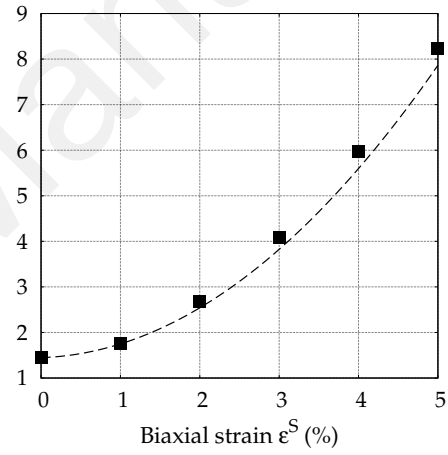
(a) Mismatch strain: $\epsilon^{m,S} = -1.476\%$



(b) Mismatch strain: $\epsilon^{m,S} = 1.34\%$



(c) Mismatch strain: $\epsilon^{m,S} = 5.421\%$



(d) Mismatch strain: $\epsilon^{m,S} = 2.524\%$

Figure 6: Variation of the interfacial excess energy $\Gamma|_{\sigma^\perp=0}$ as a function of the biaxial strain ϵ^S (for $\sigma^\perp = 0$) for an incoherent Cu/Cu₂O interface with various mismatch strains. Dashed line (---) is based on the theoretical framework presented in equation (16), square symbols (■) are from atomistic simulations. Interfacial elasticity properties used in equation (16) are presented in Table 2.

661 **Appendix A. Nomenclature and list of symbols**

662 We denote in boldface a vector \mathbf{a} of the Euclidian space \mathbb{E} only by its element a_i
663 (i.e. $\mathbf{a} = a_i \hat{\mathbf{e}}_i$) where we employ the use of Einstein's convention of summation over
664 repeated indices. The component $\hat{\mathbf{e}}_i$ is an element of $(\hat{\mathbf{e}}_1, \hat{\mathbf{e}}_2, \hat{\mathbf{e}}_3)$, a positive oriented
665 orthonormal basis of \mathbb{E} with dimension 3. Similarly a second-rank Euclidian tensor
666 \mathbf{A} designated in a boldface character is simply written as A_{ij} ($\mathbf{A} = A_{ij} \hat{\mathbf{e}}_i \otimes \hat{\mathbf{e}}_j$). A
667 third-rank Euclidian tensor $\underline{\mathbf{A}}$ designated by an underlined character is simply writ-
668 ten as A_{ijk} ($\underline{\mathbf{A}} = A_{ijk} \hat{\mathbf{e}}_i \otimes \hat{\mathbf{e}}_j \otimes \hat{\mathbf{e}}_k$). A fourth-rank Euclidian tensor $\underline{\underline{\mathbf{A}}}$ designated by a
669 doubly underlined character is simply written as A_{ijkl} ($\underline{\underline{\mathbf{A}}} = A_{ijkl} \hat{\mathbf{e}}_i \otimes \hat{\mathbf{e}}_j \otimes \hat{\mathbf{e}}_k \otimes \hat{\mathbf{e}}_l$).
670 A character not in boldface or not underlined indicates a scalar. The symbol “.”
671 between two tensors designates the product contracted once, while the symbol “:”
672 designates the product contracted twice. Roman indices used in tensorial notations
673 range from 1 to 3 (three-dimensional) and Greek indices range from 1 to 2 (two-
674 dimensional), unless otherwise indicated.

675

676 **Notation indexing:**

- $[\cdot]_{\pm}$ – denotes a variable defined in medium “+” or “-” respectively.
- $[\cdot]^S$ – denotes a variable defined in the plane of the interface.
- $[\cdot]^{\perp}$ – denotes a variable defined transverse to the interface.

677 **Strain and stress definitions:**

- $\boldsymbol{\epsilon}^S$ – interfacial in-plane strain tensor.
- $\boldsymbol{\epsilon}^{*,S}$ – interfacial structure eigenstrain tensor.

$\epsilon^{0,S}$	– in-plane molar volume change eigenstrain tensor.
$\epsilon^{m,S}$	– in-plane structural mismatch strain tensor.
ϵ^*	– eigenstrain tensor.
ϵ	– strain tensor.
σ	– stress tensor.
τ	– residual stress tensor.
κ	– relative interface curvature.

678 **Interfacial thermodynamic quantities:**

\mathcal{F}	– total free energy.
Ψ	– bulk free-energy density per unit volume.
p^S	– interfacial power density of internal forces.
Γ	– interfacial excess energy.
Γ_0	– residual interfacial excess energy.
Σ^S	– coherent surface stress.
Υ^S	– incoherent surface stress.
Δ^\perp	– transverse excess strain.
\mathbb{D}^\perp	– thermodynamic conjugate of the transverse stress field σ^\perp .
$\Gamma^{(1)}$	– residual surface stress.
$\Lambda^{(1)}$	– residual transverse interfacial deformation.
$\Upsilon^{(1)}$	– residual incoherent surface stress.
M^S	– interface bending moment.

679 **Bulk constants:**

λ	– Lamé constant.
μ	– shear modulus.
α	– coefficient of thermal expansion.
$\underline{\underline{\mathbb{C}}}$	– stiffness tensor.
\mathbb{M}	– compliance tensor.
$\underline{\underline{\gamma}}$	– transverse coupling tensor.

680 **Interfacial constants:**

$\underline{\underline{\Gamma}}^{(2)}$	– interface in-plane elastic stiffness tensor.
$\underline{\underline{\Lambda}}^{(2)}$	– interface transverse compliance tensor.
$\underline{\underline{\Upsilon}}^{(2)}$	– interface in-plane incoherency stiffness tensor.
$\underline{\underline{\Phi}}$	– interface in-plane structural interfacial mismatch coupling tensor.
$\underline{\underline{\mathbb{H}}}$	– interfacial Poisson's effect tensor.
$\underline{\underline{\mathbb{K}}}$	– interface transverse structural interfacial mismatch tensor.
$\lambda^S, \lambda^{*,S}$	– surface Lamé constants.
$K^S, K^{*,S}$	– biaxial surface moduli.
$\nu^S, \nu^{*,S}$	– interfacial Poisson's ratios.
$E^\perp, E^{*,\perp}$	– interfacial transverse Young's moduli.
α^S	– interfacial coefficient of thermal expansion.
d_0, d_0^*	– thermal-mechanical coupling coefficient.

681 **Variables used in the atomistic model:**

$e^{(i)}$	– total energy of atom i .
$e^{(0)}$	– atomic bulk energy.
N	– number of atoms.
n, m	– scaling factors to obtain various mismatch strains in the atomistic model.

- ℓ – crystal lattice parameter.
 W, L, D – dimensions of the atomistic simulation box.
 A_0 – surface area.

682 **Miscellaneous symbols:**

- \mathcal{S} – imaginary, two-dimensional surface.
 Ω – semi-infinite crystal.
 z – position across the interface.
 h – arbitrary distance away from the interface.
 \mathbf{x} – position vector.
 \mathbf{x}^S – interfacial position vector.
 \mathbf{u} – displacement field.
 ∇^S – surface gradient.
 $\mathbf{n}(\mathbf{x}^S)$ – interface unit normal upward pointing from the reference phase.
 V^0 – molar volume.
 $g(\mathbf{x})$ – variation of the structural mismatch.

683 **Appendix B. “T-decomposition”: General anisotropic elasticity with eigen-**
684 **strains due to lattice mismatch**

685 In the following “T-decomposition” (??), subscripts $\alpha, \beta, \kappa, \lambda$ vary from 1 to 2,
686 and Roman subscripts j vary from 1 to 3 with the following conventions:

$$\begin{aligned} \sigma_{\alpha\beta}^S &= \sigma_{\alpha\beta}, \quad \tau_{\alpha\beta}^S = \tau_{\alpha\beta}, \quad \tau_{\alpha\beta}^{\perp} = \tau_{\alpha\beta}, \quad \epsilon_{\kappa\lambda}^S = \epsilon_{\kappa\lambda}, \\ \sigma_j^{\perp} &= \sigma_{3j}, \quad \tau_j^{\perp} = \tau_{3j}, \quad \epsilon_{\alpha}^{\perp} = 2\epsilon_{\alpha 3}, \quad \epsilon_3^{\perp} = \epsilon_{33}, \end{aligned} \tag{B.1}$$

687 where superscripts S and \perp respectively denote the in-plane and transverse parts of
688 stresses and strains.

689 Consider an inhomogeneous linear elastic solid subjected to eigenstrains ϵ_{ij}^* , the
 690 strain energy density per unit undeformed volume (Lagrangian approach) is given
 691 by:

$$\Psi = \Psi_0 + \tau_{ij} (\epsilon_{ij} - \epsilon_{ij}^*) + \frac{1}{2} \mathbb{C}_{ijkl} (\epsilon_{ij} - \epsilon_{ij}^*) (\epsilon_{kl} - \epsilon_{kl}^*) , \quad (\text{B.2})$$

692 where ϵ_{ij} and ϵ_{ij}^* are respectively the Lagrangian total strains and eigenstrains.

693

694 The elastic (Lagrangian) strains are given by $\epsilon_{ij}^e = \epsilon_{ij} - \epsilon_{ij}^*$. The Piola-Kirchhoff
 695 stresses associated to the elastic (Lagrangian) strains are given by:

$$\sigma_{ij} = \frac{\partial \Psi}{\partial \epsilon_{ij}^e} = \tau_{ij} + \mathbb{C}_{ijkl} (\epsilon_{kl} - \epsilon_{kl}^*) . \quad (\text{B.3})$$

696 As such, the energy density Ψ , the in-plane stresses $\sigma_{\alpha\beta}^S$ and the transverse stresses
 697 σ_j^\perp can be decomposed in terms of their in-plane and transverse parts:

$$\begin{aligned} \Psi &= \Psi_0 + \frac{1}{2} \tau_{\alpha\beta}^S (\epsilon_{\alpha\beta}^S - \epsilon_{\alpha\beta}^{*,S}) + \frac{1}{2} \sigma_{\alpha\beta}^S (\epsilon_{\alpha\beta}^S - \epsilon_{\alpha\beta}^{*,S}) \\ &\quad + \frac{1}{2} \tau_j^\perp (\epsilon_j^\perp - \epsilon_j^{*,\perp}) + \frac{1}{2} \sigma_j^\perp (\epsilon_j^\perp - \epsilon_j^{*,\perp}) , \end{aligned} \quad (\text{B.4})$$

$$\sigma_{\alpha\beta}^S = \tau_{\alpha\beta}^S + \mathbb{C}_{\alpha\beta\kappa\lambda} (\epsilon_{\kappa\lambda}^S - \epsilon_{\kappa\lambda}^{*,S}) + \mathbb{C}_{\alpha\beta 3k} (\epsilon_k^\perp - \epsilon_k^{*,\perp}) , \quad (\text{B.5})$$

$$\sigma_j^\perp = \tau_j^\perp + \mathbb{C}_{3j\kappa\lambda} (\epsilon_{\kappa\lambda}^S - \epsilon_{\kappa\lambda}^{*,S}) + \mathbb{C}_{3j 3k} (\epsilon_k^\perp - \epsilon_k^{*,\perp}) . \quad (\text{B.6})$$

698 From equation (B.6), the transverse strains can be expressed as,

$$\epsilon_k^\perp = \epsilon_k^{*,\perp} - \mathbb{M}_{kj}^\perp \tau_j^\perp + \mathbb{M}_{kj}^\perp \sigma_j^\perp - \gamma_{k\alpha\beta} (\epsilon_{\alpha\beta}^S - \epsilon_{\alpha\beta}^{*,S}) , \quad (\text{B.7})$$

699 where \mathbb{M}_{kj}^\perp and $\gamma_{k\alpha\beta}$ are given by:

$$\mathbb{M}_{kj}^\perp = \mathbb{C}_{3k 3j}^{-1} , \quad \gamma_{k\alpha\beta} = \mathbb{M}_{kj}^\perp \mathbb{C}_{3j\alpha\beta} . \quad (\text{B.8})$$

700 Introducing equation (B.7) into equation (B.5), the surface stress tensor $\sigma_{\alpha\beta}^S$ is given
 701 by:

$$\sigma_{\alpha\beta}^S = \hat{\tau}_{\alpha\beta}^S + \mathbb{C}_{\alpha\beta\kappa\lambda}^S (\epsilon_{\kappa\lambda}^S - \epsilon_{\kappa\lambda}^{*,S}) + \gamma_{j\alpha\beta} \sigma_j^\perp , \quad (\text{B.9})$$

702 where

$$\hat{\tau}_{\alpha\beta}^S = \tau_{\alpha\beta}^S - \tau_j^\perp \gamma_{j\alpha\beta} \quad , \quad \mathbb{C}_{\alpha\beta\kappa\lambda}^S = \mathbb{C}_{\alpha\beta\kappa\lambda} - \mathbb{C}_{\alpha\beta 3k} \gamma_{k\kappa\lambda} . \quad (\text{B.10})$$

703 Introducing both equations (B.7) and (B.9) into equation (B.4), the energy density

704 Ψ finally becomes:

$$\Psi = \hat{\Psi}_0 + \hat{\tau}_{\alpha\beta}^S \left(\epsilon_{\alpha\beta}^S - \epsilon_{\alpha\beta}^{*,S} \right) + \frac{1}{2} \mathbb{C}_{\alpha\beta\kappa\lambda}^S \left(\epsilon_{\alpha\beta}^S - \epsilon_{\alpha\beta}^{*,S} \right) \left(\epsilon_{\kappa\lambda}^S - \epsilon_{\kappa\lambda}^{*,S} \right) + \frac{1}{2} \sigma_j^\perp \mathbb{M}_{jk}^\perp \sigma_k^\perp , \quad (\text{B.11})$$

705 where

$$\hat{\Psi}_0 = \Psi_0 - \frac{1}{2} \mathbb{M}_{jk} \tau_j^\perp \tau_k^\perp . \quad (\text{B.12})$$