## THE UNIFYING NATURE OF THE CONFIGURATIONAL FORCE BALANCE

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## 1. Introduction: some conditions for nonmaterial interfaces

The past half-century has seen much activity among materials scientists and mechanicians concerning nonmaterial interfaces, a central outcome being the realization that such problems generally result in an interface equation over and above those that follow from the classical balances for forces, moments, and mass. In *two space-dimensions* with the interface a *curve* S, this extra interface condition takes a variety of forms, the most important examples being:

Mullins's equation. This is a geometric equation,

$$bV = \psi K \tag{1}$$

for the respective motions of an isotropic grain-boundary and an isotropic grain-vapor interface, neglecting evaporation-condensation. Here V and K are the (scalar) normal velocity and curvature of the grain-boundary (or interface) S, while  $\psi$ , b,  $\rho$ , and L are strictly positive constants, with  $\psi$  the interfacial free-energy (density), b a kinetic modulus (or, recipro-



Figure 1.1 The interface S. Our convention is that K be positive on concave upward portions of S. The symbols  $(\pm)$  label the phases on the two sides of the interfaces.

cal mobility),  $\rho$  the atomic density of the solid, and L the mobility for Fickean diffusion within S. The argument of Mullins (1956) in support of (1) is physical in nature and based on work of Smoluchowski (1951), Turnbull (1951), and Beck (1952).

Variationally derived chemical potential. Working within a framework that neglects deformation and mass transport and invoking an assumption of local equilibrium, Herring (1951) defines the chemical potential U of a solid-vapor interface as the variational derivative of the total free energy with respect to variations in the configuration of the interface. Following Herring, Wu (1996), Norris (1998), and Freund (1998) generalize earlier work of Asaro and Tiller (1972) and Rice and Chuang (1981) to compute the chemical potential U of a solid-vapor interface in the presence of deformation, allowing for interfacial stress. Their result is given by

$$U = \Psi - \mathbf{Sn} \cdot \mathbf{Fn} - (\psi - \bar{\sigma}\lambda)K - \frac{\partial \tau}{\partial s}.$$
 (2)

Here  $\Psi$  is the bulk free-energy (density); **S** is the bulk Piola stress;  $\mathbf{F} = \nabla \mathbf{y}$  is the deformation gradient (with  $\mathbf{y}$  the deformation);  $\mathbf{n}$  and  $\mathbf{t}$ are the interface normal and tangent (Figure 1.1); K is the curvature; sdenotes arc length;  $\psi$  is the interfacial free-energy (density); and  $\lambda = |\mathbf{Ft}|$ is the interfacial stretch. The result (2) is derived variationally. It hinges on the assumption that the vapor pressure and vapor free-energy vanish, and is based on standard bulk constitutive relations in the solid and a constitutive equation  $\psi = \hat{\psi}(\lambda, \vartheta)$  for the interfacial free-energy, and  $\bar{\sigma}$ and  $\tau$  defined by

$$\bar{\sigma} = \frac{\partial \psi}{\partial \lambda}, \qquad \tau = \frac{\partial \psi}{\partial \vartheta},$$
(3)

with  $\vartheta$  the counterclockwise angle to the interface tangent **t**.

Kinetic Maxwell equation. This is a condition

$$\left[\!\left[\Psi - \sum_{\alpha} \rho^{\alpha} \mu^{\alpha} - \mathbf{Sn} \cdot \mathbf{Fn}\right]\!\right] = b(\vartheta) V.$$
(4)

 $\mathbf{2}$ 

for a propagating coherent phase interface between two phases composed of atomic species  $\alpha = 1, 2, ..., N$ . Here,  $\mu^{\alpha}$  and  $\rho^{\alpha}$  are the chemical potentials and atomic densities in bulk,  $\llbracket \varphi \rrbracket = \varphi^+ - \varphi^-$  is the jump of a bulk field  $\varphi$  across the interface, and, as in  $(1)_1$ ,  $b(\vartheta)$  is a constitutively determined kinetic modulus. The kinetic Maxwell condition was first obtained by Heidug and Lehner (1985), Truskinovsky (1987), and Abeyaratne and Knowles (1990), who ignored atomic diffusion but allowed for inertia. Their derivations are based on determining the local energy dissipation associated with the propagation of the interface and then appealing to the second law. When b = 0, (4) reduces to the classical Maxwell equation

$$\left[\!\left[\Psi - \sum_{\alpha} \rho^{\alpha} \mu^{\alpha} - \mathbf{Sn} \cdot \mathbf{Fn}\right]\!\right] = 0 \tag{5}$$

first derived variationally by Larché and Cahn (1978).<sup>1</sup>

**Leo–Sekerka relation**. This is a condition for an interface in *equilibrium*. Relying on a variational framework set forth by Larché and Cahn (1978) (cf., also, Alexander and Johnson, 1985; Johnson and Alexander, 1986), Leo and Sekerka (1989) consider coherent and incoherent solidsolid interfaces as well as solid-fluid interfaces. For an interface between a vapor and an alloy composed of N atomic species, neglecting vapor pressure and thermal influences, the Leo–Sekerka relation takes the form

$$\sum_{\alpha} (\rho^{\alpha} - \delta^{\alpha} K) \mu^{\alpha} = \Psi - \mathbf{Sn} \cdot \mathbf{Fn} - (\psi - \bar{\sigma}\lambda) K - \frac{\partial \tau}{\partial s}.$$
 (6)

The relation (6) is based on a constitutive equation  $\psi = \hat{\psi}(\lambda, \vartheta, \vec{\delta})$ , with  $\vec{\delta}$  the list of interfacial atomic densities  $\delta^{\alpha}$ ,  $\alpha = 1, 2, ..., N$ , supplemented by the definitions

$$\bar{\sigma} = \frac{\partial \hat{\psi}}{\partial \lambda}, \qquad \tau = \frac{\partial \hat{\psi}}{\partial \vartheta}, \qquad \mu^{\alpha} = \frac{\partial \hat{\psi}}{\partial \delta^{\alpha}}.$$
 (7)

### 2. The need for a configurational force balance

One cannot deny the applicability of the interface conditions discussed above; nor can one deny the great physical insight underlying their derivation. But in studying these derivations one is left trying to ascertain the status of the resulting equations (1), (2), (4), (5), and (6):

 $<sup>^1\</sup>mathrm{Cf.}$  also Eshelby (1970), Robin (1974), Grinfeld (1981), James (1981), and Gurtin (1983), who neglect compositional effects.

are they balances, or constitutive equations, or neither?<sup>2</sup> This and the disparity between the physical bases underlying their derivations would seem to at least indicate *the absence of a basic unifying principle*.

That additional configurational forces may be needed to describe phenomena associated with the material itself is clear from the seminal work of Eshelby (1951, 1956, 1970, 1975), Peach and Koehler (1950), and Herring (1951) on lattice defects. But these studies are based on variational arguments, arguments that, by their very nature, cannot characterize dissipation. Moreover, the introduction of configurational forces through such formalisms is, in each case, based an underlying constitutive framework and hence restricted to a particular class of materials.<sup>3</sup>

A completely different point of view is taken by Gurtin and Struthers (1990),<sup>4</sup> who — using an argument based on invariance under observer changes — conclude that a configurational force balance should join the standard (Newtonian) force balance as a *basic* law of continuum physics. Here the operative word is "basic". Basic laws are by their very nature independent of constitutive assumptions; when placed within a thermodynamic framework such laws allow one to use the now standard procedures of continuum thermodynamics to develop suitable constitutive theories.

# 3. A framework for the study of evolving nonmaterial interfaces

A complete theory of evolving interfaces in the presence of deformation and atomic transport may be developed using a framework based on: (a) standard (Newtonian) balance laws for forces and moments that account for standard stresses in bulk and within the interface; (b) an independent balance law for configurational forces that accounts for configurational stresses in bulk and within the interface;<sup>5</sup> (c) atomic balances, one for each atomic species, that account for bulk and surface diffusion; a mechanical (isothermal) version of the first two laws of ther-

 $<sup>^{2}</sup>$ Successful theories of continuum mechanics are typically based on a clear separation of balance laws and constitutive equations, the former describing large classes of materials, the latter describing particular materials.

<sup>&</sup>lt;sup>3</sup>A vehicle for the discussion of configurational forces within a dynamical, dissipative framework derives configurational force balances by manipulating the standard momentum balance, supplemented by hypereslastic constitutive relations (e.g., Maugin, 1993). But such derived balances, while interesting, are satisfied automatically whenever the momentum balance is satisfied and are hence superfluous.

 $<sup>^4\</sup>mathrm{This}$  work is rather obtuse; better references for the underlying ideas are Gurtin (1995, 2000).

 $<sup>^5</sup>$ As extended by Davi and Gurtin (1990), Gurtin (1991), Gurtin and Voorhees (1995), and Fried and Gurtin (1999, 2003) to account for atomic transport.

modynamics in the form of a *free-energy imbalance* that accounts for temporal changes in free-energy, energy flows due to atomic transport, and *power expended by both standard and configurational forces*; (d) *ther-modynamically consistent constitutive relations* for the interface and for the interface the interface and its environment.

One of the more interesting outcomes of this approach is an explicit relation for the configurational surface tension  $\sigma$  in terms of other interfacial fields; viz.,

$$\sigma = \psi - \sum_{\alpha} \delta^{\alpha} \mu^{\alpha} - \bar{\sigma} \lambda.$$
(8)

This relation, a direct consequence of the free-energy imbalance applied to the interface, is a basic relation valid for all isothermal interfaces, independent of constitutive assumptions and hence of material; it places in perspective the basic difference between the configurational surface tension  $\sigma$  and standard surface tension  $\bar{\sigma}$ . There is much confusion in the literature concerning surface tension  $\sigma$  and its relation to surface free-energy  $\psi$ . By (8), we see that these two notions coincide if and only if standard interfacial stress as well as interfacial atomic densities are negligible.

## 4. The normal configurational force balance and the dissipation inequality

The configurational force balance for the interface takes the simple form

$$\frac{\partial \mathbf{c}}{\partial s} + \mathbf{g} + \llbracket \mathbf{C} \rrbracket \mathbf{n} = \mathbf{0}.$$
(9)

Here **c** is the configurational surface stress, **g** is a dissipative internal force associated with the rearrangement of atoms at the interface, and **C** is the configurational stress in the solid. The tangential and normal components  $\sigma = \mathbf{c} \cdot \mathbf{t}$  and  $\tau = \mathbf{c} \cdot \mathbf{n}$  of **c** are the configurational surface tension and the configurational shear; thus, in contrast to more classical discussions, the surface tension actually represents a *force* tangent to the interface, with no *a priori* relationship to surface energy. The theory in bulk shows **C** to be the Eshelby tensor

$$\mathbf{C} = \left(\Psi - \sum_{\alpha} \rho^{\alpha} \mu^{\alpha}\right) \mathbf{1} - \mathbf{F}^{\mathsf{T}} \mathbf{S},\tag{10}$$

a relation that bears comparison with (8). Of most importance is the component

$$\sigma K + \frac{\partial \tau}{\partial s} + \mathbf{n} \cdot [\![\mathbf{C}]\!]\mathbf{n} + g = 0, \qquad g = \mathbf{g} \cdot \mathbf{n}, \tag{11}$$

of (9) normal to the interface, as this is the component relevant to the motion of the interface.

For an interface in the presence of deformation and atomic transport, the normal configurational force balance (11), when combined with (8), (10), and the standard force, moment, and atomic balances, yields the *normal configurational force balance* (Fried and Gurtin, 2003)

$$\sum_{\alpha} (\llbracket \rho^{\alpha} \rrbracket + \delta^{\alpha} K) \mu^{\alpha} = \llbracket \Psi - \mathbf{Sn} \cdot \mathbf{Fn} \rrbracket + (\psi - \bar{\sigma}\lambda) K + \frac{\partial \tau}{\partial s} + g, \quad (12)$$

with  $\bar{\sigma}$  the standard surface-tension. This balance is basic, as its derivation utilizes only basic laws; as such it is independent of material.

The free-energy imbalance localized to the interface using the basic balances yields the *interfacial dissipation inequality* 

$$\overset{\Box}{\psi} - \bar{\sigma}\overset{\Box}{\lambda} - \tau\overset{\Box}{\vartheta} - \sum_{\alpha} \mu^{\alpha}\overset{\Box}{\delta}^{\alpha} + \sum_{\alpha} h^{\alpha} \frac{\partial \mu^{\alpha}}{\partial s} + gV \le 0, \tag{13}$$

with a superposed box denoting the time derivative following the normal trajectories of S; this inequality, which is also basic, is used as a starting point for the discussion of constitutive relations.

## 5. Relation of the normal configurational force balance to the classical equations

Each of the interface conditions in  $\S1$  may be derived — without assumptions of local equilibrium — within the framework set out in  $\S3$ .

Leo–Sekerka relation. If we take  $g \equiv 0$ , then the normal configurational force balance (12) reduces to the Leo–Sekerka relation (6). The relation (6) follows rigorously as an Euler–Lagrange equation associated with the variational problem of minimizing the total free-energy of a solid particle surrounded by a vapor. Thus, for solid-vapor interfaces in equilibrium, the format adopted here is completely consistent with results derived variationally. The Leo–Sekerka relation (6) (or similar relations for other types of phase interfaces) is typically applied, as is, to dynamical problems, often with an accompanying appeal to an hypothesis of "local equilibrium", although the precise meaning of this assumption is never spelled out. The more general framework leading to the normal configurational force balance (12) would allow for a nonequilibrium term -g, with  $g = -b(\vartheta)V$ , on the right side of (6). The question as to when the Leo-Sekerka relation is applicable in dynamical situations is equivalent to the question as to when the internal force g is negligible. Our more general framework provides an answer to this question: for sufficiently small length scales g cannot be neglected, because the term emanating from g in the evolution equations for the interface is of the same order of magnitude as the other kinetic term in these equations, which results from accretion (Fried and Gurtin, 2004, §26.3).

Variationally derived chemical potential. We restrict attention to a single atomic species, take g = 0, and neglect the adatom density. Further, consistent with the variational treatment we assume that the vapor pressure and vapor free-energy (and hence the standard and configurational forces) vanish in the vapor. This allows us to replace the jumps by negative interfacial limits from the solid (so that  $\llbracket \Psi \rrbracket$  becomes  $-\Psi^- = -\Psi$ , and so forth). Then the normal configurational force balance reduces to (2) with  $U = \rho \mu$ . The chemical potential U is, by its very definition, a potential associated with the addition of material at the solid-vapor interface, without regard to the specific composition of that material. As such, U cannot be used to discuss alloys. As with the Leo-Sekerka relation, the more general framework discussed here allows for a kinetic term  $b(\vartheta)V$  on the right side, and hence for a nonequilibrium chemical potential.

Mullins's equation and the kinetic Maxwell equation. If in (12) we neglect deformation, adatoms, and all fields related to the bulk material, and consider constitutive relations of the form  $\psi = \hat{\psi}(\vartheta)$  and

$$\tau = \frac{\partial \hat{\psi}}{\partial \vartheta}, \qquad g = -b(\vartheta)V,$$
(14)

with  $b(\vartheta) \ge 0$  a kinetic modulus, then the dissipation inequality (13) is satisfied and the normal configurational force balance (11) reduces to the *curvature-flow equation* 

$$b(\vartheta)V = \left[\psi(\vartheta) + \psi''(\vartheta)\right]K,\tag{15}$$

proposed by Uwaha (1987) and independently, using configurational forces, by Gurtin (1988); for an isotropic material, (15) reduces to Mullins's equation (1).

Similarly, the kinetic Maxwell equation (4) follows from (11) upon neglecting atomic transport as well as interfacial structure and taking  $g = -b(\vartheta)V$ .

#### **6**. A final remark

Interface conditions that in other theories play the role of the normal configurational force balance are typically based on an assumption of local equilibrium or on a chemical potential derived as a variational derivative of the total free-energy with respect to variations in the configuration of the interface. By their very nature, such variational paradigms *cannot* involve the normal velocity V. To the contrary, a framework based on a configurational force balance allows for nonequilibrium terms of this form.

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1014	Brown, E. N., M. R. Kessler, N. R. Sottos, and S. R. White	<i>In situ</i> poly(urea-formaldehyde) microencapsulation of dicyclopentadiene – <i>Journal of Microencapsulation</i> (submitted)	Feb. 2003
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## List of Recent TAM Reports (cont'd)

No.	Authors	Title	Date
1018	Costello, G. A.	Mechanics of wire rope – Mordica Lecture, Interwire 2003, Wire Association International, Atlanta, Georgia, May 12, 2003	Mar. 2003
1019	Wang, J., N. R. Sottos, and R. L. Weaver	Thin film adhesion measurement by laser induced stress waves – Journal of the Mechanics and Physics of Solids (submitted)	Apr. 2003
1020	Bhattacharjee, P., and D. N. Riahi	Effect of rotation on surface tension driven flow during protein crystallization – <i>Microgravity Science and Technology</i> , in press (2003)	Apr. 2003
1021	Fried, E.	The configurational and standard force balances are not always statements of a single law – <i>Proceedings of the Royal Society</i> (submitted)	Apr. 2003
1022	Panat, R. P., and K. J. Hsia	Experimental investigation of the bond coat rumpling instability under isothermal and cyclic thermal histories in thermal barrier systems – <i>Proceedings of the Royal Society of London A</i> , in press (2003)	May 2003
1023	Fried, E., and M. E. Gurtin	A unified treatment of evolving interfaces accounting for small deformations and atomic transport: grain-boundaries, phase transitions, epitaxy – <i>Advances in Applied Mechanics</i> , in press (2003)	May 2003
1024	Dong, F., D. N. Riahi, and A. T. Hsui	On similarity waves in compacting media – <i>Horizons in Physics,</i> in press (2003)	May 2003
1025	Liu, M., and K. J. Hsia	Locking of electric field induced non-180° domain switching and phase transition in ferroelectric materials upon cyclic electric fatigue – <i>Applied Physics Letters</i> , in press (2003)	May 2003
1026	Liu, M., K. J. Hsia, and M. Sardela Jr.	In situ X-ray diffraction study of electric field induced domain switching and phase transition in PZT-5H— <i>Journal of the American</i> <i>Ceramics Society</i> (submitted)	May 2003
1027	Riahi, D. N.	On flow of binary alloys during crystal growth – <i>Recent Research Development in Crystal Growth</i> , in press (2003)	May 2003
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1029	Fried, E., V. Korchagin, and R. E. Todres	Biaxial disclinated states in nematic elastomers – <i>Journal of Chemical Physics</i> <b>119</b> , 13170–13179 (2003)	July 2003
1030	Sharp, K. V., and R. J. Adrian	Transition from laminar to turbulent flow in liquid filled microtubes – <i>Physics of Fluids</i> (submitted)	July 2003
1031	Yoon, H. S., D. F. Hill, S. Balachandar, R. J. Adrian, and M. Y. Ha	Reynolds number scaling of flow in a Rushton turbine stirred tank: Part I – Mean flow, circular jet and tip vortex scaling – <i>Chemical</i> <i>Engineering Science</i> (submitted)	Aug. 2003
1032	Raju, R., S. Balachandar, D. F. Hill, and R. J. Adrian	Reynolds number scaling of flow in a Rushton turbine stirred tank: Part II – Eigen-decomposition of fluctuation – <i>Chemical Engineering</i> <i>Science</i> (submitted)	Aug. 2003
1033	Hill, K. M., G. Gioia, and V. V. Tota	Structure and kinematics in dense free-surface granular flow – <i>Physical Review Letters,</i> in press (2003)	Aug. 2003
1034	Fried, E., and S. Sellers	Free-energy density functions for nematic elastomers – <i>Journal of the Mechanics and Physics of Solids</i> (submitted)	Sept. 2003
1035	Kasimov, A. R., and D. S. Stewart	On the dynamics of self-sustained one-dimensional detonations: A numerical study in the shock-attached frame – <i>Physics of Fluids</i> (submitted)	Nov. 2003
1036	Fried, E., and B. C. Roy	Disclinations in a homogeneously deformed nematic elastomer – <i>Nature Materials</i> (submitted)	Nov. 2003
1037	Fried, E., and M. E. Gurtin	The unifying nature of the configurational force balance – <i>Mechanics of Material Forces</i> (P. Steinmann and G. A. Maugin, eds.), in press (2003)	Dec. 2003