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First vs. second gradient of strain theory for capillarity effects in an elastic fluid at small length scales

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ABSTRACT

Mindlin [22] wrote a milestone paper claiming that a second strain gradient theory is required for a continuum description of volume cohesion and surface tension in isotropic elastic media. The objective of the present work is to compare Mindlin's approach to more standard capillarity models based on a first strain gradient theory and Korteweg's equation. A general micromorphic model is then proposed as a numerical method to implement Mindlin's theory in a finite element code.

1. Introduction

Due to their different local environments, atoms at a free surface and atoms in the bulk of a material have a different associated energy and lattice spacing. This excess of energy associated with surface atoms is called surface free energy and gives rise to surface tension. The surface region is only a few atomic layers thin, that is why the surface tension can be neglected when the characteristic length of the microstructure of the considered material is in the micrometer range or larger. But in the case of nano-sized materials, the ratio between the surface and the volume is much higher and the surface region behaviour cannot be neglected anymore. There are several ways to introduce the mechanical properties of the surface. If an interface separating two homogeneous bulk phases is considered, one can define the interfacial properties by using an inter-phase with a finite volume and assign thermodynamic properties in the usual way, as in Capolungo et al. [3]. Three phases are considered in this approach and the boundaries of the inter-phase have to be defined more or less arbitrary.

One can also consider that the two homogeneous phases are separated by a single dividing surface; the thermodynamic properties of the interface are defined as the excess over the values obtained for both bulk phases separated by a zero-thickness surface [25,8]. The continuum mechanical theory of surface/interface behaviour has been settled by Gurtin and Murdoch [18,19]. It introduces a volume stress tensor in the bulk of the material and a surface stress tensor in the surface or interface modelled as a membrane. Both stress tensors fulfill balance of momentum equations. A specific elastic behaviour is attributed to the membrane and kinematic constraints ensure that the bulk part and the surface remain coherent. The most common manifestation of surface behaviour is capillarity effects in elastic fluids. It is described by the Young–Laplace equation which states that the internal pressure, p, in a spherical droplet is proportional to the surface tension, T, multiplied by the surface curvature, $1/r$:

$$p = \frac{2T}{r}. \tag{1}$$

When the size of the considered object is small enough, there is no clear way to define a sharp interface or surface. Instead, a continuum model can be used to describe a transition domain between two bulk regions, or between the bulk and the outer free surface. Such continuum theories for diffuse surface or interfaces have been developed for a refined description of capillarity in elastic fluids and solids. They are based on higher order gradient theories like the Korteweg equation which involves the gradient of density vector, see [30]. A more general strain gradient theory

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has been proposed by Casal [4–6]. In a unsufficiently known paper, Mindlin [22] claims that a second gradient of strain or, equivalently, third gradient of displacement theory is in fact needed to describe, in a continuous manner, capillarity and cohesion effects in isotropic linear elastic solids and fluids. Based on a simple one-dimensional atomic chain model, he identifies the higher order elasticity modulus that is responsible for the variation of lattice spacing from the free surface into the bulk in a semi-infinite crystal. This model has not been discussed in literature, so that there seems to be no general opinion whether a first or second strain gradient theory is needed for capillarity effects in linear elastic media. On the other hand, Mindlin's second gradient of strain theory is challenging from the computational point of view to compute fields of lattice parameters in nano-objects like nano-particles or nanocrystals. Molecular static simulation provide such non-homogeneous distributions of lattice spacing in crystals close to free surfaces or grain boundaries, that could be represented by a suitable continuum model.

The objective of the present work is, firstly, to compare Korteweg's equation with the first and second gradient of strain theories in order to highlight the main differences, and, secondly, to provide a framework for the numerical implementation of Mindlin's second strain gradient theory. Finite element implementations of the first gradient of strain theory exist in literature. They are based on the introduction of additional strain degrees of freedom in the spirit of Eringen's micromorphic approach [28,7,12]. That is why a second order micromorphic model is formulated in the last section of the present work. Then, an internal constraint must be enforced by means of Lagrange multiplier or suitable penalization, so that the general micromorphic model reduces to Mindlin's second gradient of strain model.

The article is organized as follows. The links between Korteweg's equation and the first strain gradient theory are presented in Section 2. Arguments pleading for the necessity of a second strain gradient model are provided in Section 3. A micromorphic generalization of Mindlin's model is finally proposed in Section 4, as the suitable framework for a future finite element implementation of higher order gradient theories.

For the sake of conciseness, the small strain framework is adopted. Volume forces are not considered throughout the work. The analysis is limited to the static case. We follow Mindlin's notation as closely as possible. However, we adopt an intrinsic notation where zeroth, first, second and third order tensors are denoted by $a, \underline{\boldsymbol{a}}, \underset{\sim}{\boldsymbol{a}}, \underset{\approx}{\boldsymbol{a}}$, respectively. The simple, double and triple contractions are written ., :, and $\vdots$, respectively. In index form with respect to an orthonormal Cartesian basis ($\underline{\boldsymbol{e}}_1$, $\underline{\boldsymbol{e}}_2$, $\underline{\boldsymbol{e}}_3$), these notations correspond to

$$\underline{\boldsymbol{a}} . \underline{\boldsymbol{b}} = a_i b_i, \quad \underset{\sim}{\boldsymbol{a}} : \underset{\sim}{\boldsymbol{b}} = a_{ij} b_{ij}, \quad \underset{\approx}{\boldsymbol{a}} \vdots \underset{\approx}{\boldsymbol{b}} = a_{ijk} b_{ijk}, \tag{2}$$

where repeated indices are summed up. The tensor product is denoted by $\otimes$. The nabla operator with respect to the reference configuration is denoted by ∇. For example, the component ijk of $\underset{\sim}{\boldsymbol{a}} \otimes \nabla$ is $a_{ij,k}$. In particular, ∇^2 is the Laplace operator. Index notation is also used at places to avoid any confusion. For instance, we give the chosen intrinsic and index notations for the second gradient of a scalar field and of a second rank tensor:

$$\nabla \otimes \nabla \rho = \rho_{,ij} \underline{\boldsymbol{e}}_i \otimes \underline{\boldsymbol{e}}_j, \quad \underset{\sim}{\boldsymbol{\varepsilon}} \otimes \nabla \otimes \nabla = \varepsilon_{ij,kl} \underline{\boldsymbol{e}}_i \otimes \underline{\boldsymbol{e}}_j \otimes \underline{\boldsymbol{e}}_k \otimes \underline{\boldsymbol{e}}_l \tag{3}$$

2. Korteweg's equation and first strain gradient model

The Van der Waals and Korteweg equations are the first attempts to introduce capillary effects in a continuum mechanical theory. For an elastic medium, they include not only the effect of mass density, ρ, on stress but also that of the density gradient $\nabla\rho$, in the form

$$\underset{\sim}{\boldsymbol{T}} = -p(\rho)\,\underset{\sim}{\mathbf{1}} - \alpha(\nabla\rho)^2\,\underset{\sim}{\mathbf{1}} - \beta\nabla\rho\otimes\nabla\rho + \gamma(\nabla^2\rho)\,\underset{\sim}{\mathbf{1}} + \delta\nabla\otimes\nabla\rho, \tag{4}$$

where $\underset{\sim}{\boldsymbol{T}}$ is the stress tensor and α, β, γ and δ are material parameters, namely higher order elasticity moduli. The divergence of $\underset{\sim}{\boldsymbol{T}}$ is assumed to vanish, in the absence of volume forces. In their account of Korteweg's constitutive theory, Truesdell and Noll [30] show how it can be used to represent a spherical non-uniform field of mass density, $\rho(r)$, thus allowing for the presence of an interface between liquid and vapor in a water droplet. The balance equation,

$$T'_{rr} + \frac{2}{r}(T_{rr} - T_{\theta\theta}) = 0, \tag{5}$$

is combined with the constitutive relation,

$$T_{rr} - T_{\theta\theta} = -\beta\rho'^2 - \delta\frac{\rho'}{r} + \delta\rho'', \tag{6}$$

where the prime denotes derivation with respect to r. Integrating Eq. (5) on the interface zone $[r_1,r_2]$ yields the relation

$$T_{rr}(r_2) - T_{rr}(r_1) = 2\delta\left(\frac{\rho'(r_2)}{r_2} - \frac{\rho'(r_1)}{r_1}\right) - 2\int_{r_1}^{r_2} \frac{\beta\rho'^2 - \delta\rho''}{r} dr. \tag{7}$$

In order to obtain results appropriate to a thin shell of transition, we calculate the limit of the latter relation as r_1, r_2 tend to r_0. The first term on the right-hand side vanishes whereas, under suitable assumptions of smoothness, the second term is proportional to the mean curvature, $1/r_0$. Accordingly, Eq. (7) can be interpreted as the diffuse counterpart of Laplace sharp interface equation.

The compatibility of such higher grade constitutive equations, formulated within the framework of classical continuum mechanics, with continuum thermodynamics has been questioned by Gurtin [17] (see also [27]). Gurtin [17] argued that such higher order constitutive statements can be acceptable only if higher order stress tensors are introduced in addition to the usual Cauchy simple force stress tensor. To see that, let us now rephrase Korteweg's equation within the linear elasticity framework. For that purpose, we define the dilatation as the trace of the small strain tensor,

$$\Delta = \mathrm{trace}\ \underset{\sim}{\boldsymbol{\varepsilon}} = 1 - \frac{\rho_0}{\rho}, \quad \nabla\Delta = \rho_0\frac{\nabla\rho}{\rho^2} \simeq \frac{\nabla\rho}{\rho_0}, \tag{8}$$

within the small strain approximation with respect to a reference mass density ρ_0. The Korteweg Eq. (4) can therefore be written as

$$\underset{\sim}{\boldsymbol{T}} = -p(\Delta)\,\underset{\sim}{\mathbf{1}} - \alpha(\nabla\Delta)^2\,\underset{\sim}{\mathbf{1}} - \beta\nabla\Delta\otimes\nabla\Delta + \gamma(\nabla^2\Delta)\,\underset{\sim}{\mathbf{1}} + \delta\nabla\otimes\nabla\Delta, \tag{9}$$

$$T_{ij} = -p(\Delta)\delta_{ij} - \alpha\Delta_{,k}\Delta_{,k}\delta_{ij} - \beta\Delta_{,i}\Delta_{,j} + \gamma\Delta_{,kk}\delta_{ij} + \delta\Delta_{,ij}. \tag{10}$$

If we neglect the quadratic terms in the previous expression, we obtain the linearized Korteweg equation

$$\underset{\sim}{\boldsymbol{T}} = -p(\Delta)\,\underset{\sim}{\mathbf{1}} + \gamma(\nabla^2\Delta)\,\underset{\sim}{\mathbf{1}} + \delta\nabla\otimes\nabla\Delta. \tag{11}$$

It involves higher order gradients of the dilatation. This suggests that it could be derived from a strain gradient theory as proposed by Toupin [29], Casal [4] and Mindlin and Eshel [23] in the early sixties. As shown by Toupin, the second gradient of displacement theory is equivalent to the first strain gradient theory. Such theories introduce higher order stresses, as advocated by Gurtin [17]. The link between strain gradient theory and capillarity or surface tension in fluids or solids has been recognized by Casal [5]. The strain gradient theory can be limited to the gradient of density effects and therefore compared to Korteweg's equation, as it was done by Casal and Gouin [6]. The relation between the stress tensor $\underset{\sim}{\boldsymbol{T}}$ in Eq. (4) and the higher order stress measures of strain gradient theories is discussed in the next section. More recent works have developed the concept of a fluid with internal wettability based on modified Korteweg equations within the strain gradient framework, also called Cahn–Hilliard fluid in [20,2].

This idea of considering higher gradients of density to describe the material behaviour close to free surfaces or interfaces has been implemented for phase transformations like in water droplets in vapor [9]. This represents a diffuse interface model for liquid–gas interfaces.

3. Second strain gradient theory

Mindlin [22] claims that a second strain gradient theory is necessary to account for capillarity and cohesion effects in elastic media, instead of the first strain gradient theory reported in the previous section. We recall here his arguments in order to show the fundamental difference between Mindlin's approach and the previous one. The second strain gradient theory is presented and contains the first strain gradient model as a special case. It is based on the assumption that the stress state at a material point depends on the values of the strain, the first and the second strain gradients at that point. Following the method of virtual power [15,1], the virtual work density of internal forces is a linear form with respect to all strain measures:

$$w^{(i)} = \underset{\sim}{\boldsymbol{\sigma}} : \underset{\sim}{\boldsymbol{\varepsilon}} + \underset{\simeq}{\boldsymbol{S}} \,\vdots\, (\underset{\sim}{\boldsymbol{\varepsilon}} \otimes \boldsymbol{\nabla}) + \underset{\approx}{\boldsymbol{S}} :: (\underset{\sim}{\boldsymbol{\varepsilon}} \otimes \boldsymbol{\nabla} \otimes \boldsymbol{\nabla}), \tag{12}$$

where $\underset{\sim}{\boldsymbol{\sigma}}$ is the simple force stress tensor and $\underset{\simeq}{\boldsymbol{S}}, \underset{\approx}{\boldsymbol{S}}$ are the so-called hyperstress tensors as required by the mentioned thermodynamical consistency. Mindlin [22] shows that the stresses must fulfill the following balance equation

$$\underset{\sim}{\boldsymbol{T}} . \nabla = 0, \quad \text{with} \quad \underset{\sim}{\boldsymbol{T}} = \underset{\sim}{\boldsymbol{\sigma}} - (\underset{\simeq}{\boldsymbol{S}} - \underset{\approx}{\boldsymbol{S}} . \nabla) . \nabla. \tag{13}$$

The stress tensor $\underset{\sim}{\boldsymbol{T}}$ is an effective stress tensor whose divergence vanishes. It is expressed in terms of the stress tensors at all orders. This partial differential equation is accompanied by three sets of complex boundary conditions that involve the surface curvature and the normal and tangent derivatives of stress quantities. For the sake of conciseness, they are not recalled here and the author is referred to Eqs. (18a–c) in Mindlin [22]. Corresponding expressions of the boundary conditions for the first gradient of strain theory can be found in [23,15]. The free energy density function, $\Psi(\underset{\sim}{\boldsymbol{\varepsilon}}, \underset{\sim}{\boldsymbol{\varepsilon}} \otimes \boldsymbol{\nabla}, \underset{\sim}{\boldsymbol{\varepsilon}} \otimes \boldsymbol{\nabla} \otimes \boldsymbol{\nabla})$, is then a potential from which stresses are computed:

$$\underset{\sim}{\boldsymbol{\sigma}} = \rho \frac{\partial \Psi}{\partial \underset{\sim}{\boldsymbol{\varepsilon}}}, \quad \underset{\simeq}{\boldsymbol{S}} = \rho \frac{\partial \Psi}{\partial (\underset{\sim}{\boldsymbol{\varepsilon}} \otimes \boldsymbol{\nabla})}, \quad \underset{\approx}{\boldsymbol{S}} = \rho \frac{\partial \Psi}{\partial (\underset{\sim}{\boldsymbol{\varepsilon}} \otimes \boldsymbol{\nabla} \otimes \boldsymbol{\nabla})}. \tag{14}$$

When the terms $\underset{\approx}{\boldsymbol{S}}$ and $\underset{\sim}{\boldsymbol{\varepsilon}} \otimes \boldsymbol{\nabla} \otimes \boldsymbol{\nabla}$ are dropped, the first strain gradient theory is recovered. Korteweg's stress $\underset{\sim}{\boldsymbol{T}}$ can then be interpreted as the effective stress of the first gradient of strain theory, as done in Casal and Gouin [6].

Let us specify the constitutive equations in the case of an elastic fluid. Mindlin considers that, in an elastic fluid, all stress tensors are spherical so that

$$\underset{\sim}{\boldsymbol{\sigma}} = -p \underset{\sim}{\boldsymbol{1}}, \quad \underset{\simeq}{\boldsymbol{S}} = -\underset{\sim}{\boldsymbol{1}} \otimes \underline{\boldsymbol{p}}, \quad \underset{\approx}{\boldsymbol{S}} = -\underset{\sim}{\boldsymbol{1}} \otimes \underset{\sim}{\boldsymbol{\pi}}, \tag{15}$$

where Mindlin's notations for the stress tensors p, $\underline{\boldsymbol{p}}$, and $\underset{\sim}{\boldsymbol{\pi}}$ have been kept. The notation $\underset{\approx}{\boldsymbol{S}} = -\underset{\sim}{\boldsymbol{1}} \otimes \underset{\sim}{\boldsymbol{\pi}}$ stands for $S_{ijkl} = -\delta_{ij}\pi_{kl}$. As a result, the effective stress is spherical and reads

$$\underset{\sim}{\boldsymbol{T}} = -(p - \underline{\boldsymbol{p}}.\boldsymbol{\nabla} + \underset{\sim}{\boldsymbol{\pi}} : (\boldsymbol{\nabla} \otimes \boldsymbol{\nabla}))\, \underset{\sim}{\boldsymbol{1}}, \tag{16}$$

the divergence of which vanishes. It is in contrast to Korteweg's medium, see Eq. (11), which can transmit shear stresses. The concept of an elastic fluid has been clearly defined by Noll for simple materials [30] and leads to a spherical Cauchy stress tensor. The question of the definition of a first strain gradient elastic fluid has been only tackled very recently by Fried and Gurtin [14] and Podio-Guidugli and Vianello [26]. Clearly, Korteweg and Mindlin's definitions of an elastic first strain gradient fluid differ. Mindlin's formulation is less general than that given by Eq. (11). Following Mindlin's definition, the free energy potential of an isotropic second strain gradient linear elastic fluid takes the form

$$\begin{aligned}\rho\Psi(\Delta, \boldsymbol{\nabla}\Delta, \boldsymbol{\nabla}\otimes\boldsymbol{\nabla}\Delta) = {} & \frac{\lambda}{2}\Delta^2 + a_1(\boldsymbol{\nabla}\Delta)^2 + b_0\boldsymbol{\nabla}^2\Delta + b_1(\boldsymbol{\nabla}^2\Delta)^2 \\ & + b_2(\boldsymbol{\nabla}\otimes\boldsymbol{\nabla}\Delta) : (\boldsymbol{\nabla}\otimes\boldsymbol{\nabla}\Delta) + c_1\Delta\boldsymbol{\nabla}^2\Delta. \end{aligned} \tag{17}$$

All terms are quadratic except the linear term that involves the material parameter b_0. The stress–strain relations follow

$$p = -\rho\frac{\partial\Psi}{\partial\Delta} = -\lambda\Delta - c_1\boldsymbol{\nabla}^2\Delta, \quad \underline{\boldsymbol{p}} = -\rho\frac{\partial\Psi}{\partial\boldsymbol{\nabla}\Delta} = -2a_1\boldsymbol{\nabla}\Delta, \tag{18}$$

$$\underset{\sim}{\boldsymbol{\pi}} = -\rho\frac{\partial\Psi}{\partial(\boldsymbol{\nabla}\otimes\boldsymbol{\nabla}\Delta)} = -(b_0 + c_1\Delta + 2b_1\boldsymbol{\nabla}^2\Delta)\,\underset{\sim}{\boldsymbol{1}} - 2b_2\boldsymbol{\nabla}\otimes\boldsymbol{\nabla}\Delta. \tag{19}$$

It contains, in general, a self-equilibrated higher order stress $\underset{\sim}{\boldsymbol{\pi}} = -b_0\,\underset{\sim}{\boldsymbol{1}}$, in the absence of any external load. The effective stress then becomes

$$\underset{\sim}{\boldsymbol{T}} = -(-\lambda\Delta + 2(a_1 - c_1)\boldsymbol{\nabla}^2\Delta - 2(b_1 + b_2)\boldsymbol{\nabla}^4\Delta)\,\underset{\sim}{\boldsymbol{1}} \tag{20}$$

$$T_{ij} = -(-\lambda\Delta + 2(a_1 - c_1)\Delta_{,kk} - 2(b_1 + b_2)\Delta_{,kkll})\delta_{ij}. \tag{21}$$

The corresponding effective stress for the first strain gradient model is obtained by setting $b_1 = b_2 = 0$. It can be directly compared to Korteweg's Eq. (11). Both relations have the Laplacian term, $\boldsymbol{\nabla}^2\Delta$, in common. Korteweg's equation contains an additional non-spherical term, as already mentioned. Mindlin's second gradient of strain theory introduces an additional contribution of fourth order.

Combining the balance and constitutive equations, the following fourth order partial differential equation is derived

$$(1 - l_1^2\boldsymbol{\nabla}^2)(1 - l_2^2\boldsymbol{\nabla}^2)\Delta = 0, \tag{22}$$

that involves two characteristic lengths such that

$$\lambda l_i^2 = a_1 - c_1 \pm \sqrt{(a_1 - c_1)^2 - 2\lambda(b_1 + b_2)}, \quad i = 1, 2. \tag{23}$$

Note that Eq. (22) follows from a first integration of the fifth order balance Eq. (13). Accordingly, a constant term should be added in the right-hand side of the equation, leading to a homogeneous pressure field to be superimposed to the solution. The Eq. (22) can be referred as a bi-Helmholtz equation. It can also be derived from a non-local elasticity law as done by Lazar et al. [21].

Let us now consider capillary effects in a spherical homogeneous material made of a second strain gradient isotropic linear elastic fluid. We look for a dilatation field $\Delta(r)$ and equations are solved in spherical coordinates. In that specific case, Mindlin shows that the dilatation field is then of the form

$$\Delta = \frac{C_1 l_1}{r_1}\sinh\frac{r}{l_1} + \frac{C_2 l_2}{r_2}\sinh\frac{r}{l_2}. \tag{24}$$

The integration constants C_1 and C_2 must be determined from the boundary conditions. A key point of the analysis is that the outer surface of the droplet of initial radius r_0 is assumed to be free of traction forces. Mindlin shows that these traction-free boundary conditions lead to two equations for the unknowns C_1 and C_2:

$$\sum_{i=1}^{2} C_i r_i \Big(2(b_1 + b_2) - (2a_1 - c_1)l_i^2\Big)(r_i\cosh r_i - \sinh r_i) = 0, \tag{25}$$

$$\begin{aligned}\sum_{i=1}^{2} C_i\left(r_i\left(2b_2\left(1 + \frac{2}{r_i}\right) + 2b_1 + c_1 l_i^2\right)\sinh r_i - 4b_2\cosh r_i\right) \\ = -b_0 r_0^2, \end{aligned} \tag{26}$$

where $r_i = r_0/l_i$. When the radius of curvature is large in comparison with the characteristic lengths l_i (i.e., $r_i \gg 1$), Mindlin derives a relation between the mean dilatation of the droplet and its radius of curvature

$$\bar{\Delta} = \frac{3}{4\pi r_0^3}\int_V \Delta\, dV = -\frac{6c_1}{b_0\lambda}\frac{T}{r_0}, \tag{27}$$

where T can be identified with the surface tension in the Laplace–Young Eq. (1) and takes the value

$$T = \frac{T_0}{1 + (8b_2T_0/b_0^2 r_0)},\quad \text{with}\quad T_0 = \frac{b_0(l_1^2 - l_2^2)}{2\lambda(l_1(l_2^2 + c_1^2/\lambda^2)^2 - l_2(l_1^2 + c_1^2/\lambda^2)^2)}. \tag{28}$$

The identification between the diffuse and sharp interface models for surface tension shows that the relevant material parameters that intervene in the relation are c_1, b_2 and b_0 which are all related to the contributions of the second strain gradient in the elastic potential (17). One should note the key rôle of the initial higher order stress b_0. Indeed, if $b_0 = 0$, the system of Eqs. (25) and (26) is homogeneous and leads to the trivial solution $C_1 = C_2 = 0$ and to a homogeneous mass density field. As claimed by Mindlin, the internal hyperstresses account for material cohesion which is destroyed at a free surface.

The corresponding balance equations and solution of the spherical droplet can also be obtained for the first gradient of strain theory, in the same way. For, the first strain gradient theory can be formally obtained as a special case of the second strain gradient model by setting that the fourth order stress tensor $\underset{\approx}{\boldsymbol{S}}$ vanishes in Eq. (12), or, equivalently, that the second characteristic length l_2 vanishes in (22). The Eq. (22) then reduces to

$$(1 - l^2\nabla^2)\Delta = 0,\quad \text{with}\quad l^2 = \frac{2a_1}{\lambda}, \tag{29}$$

which involves only one characteristic length l. There is still a solution of the form

$$\Delta = \frac{C}{r}\sinh\frac{r}{l_1} \tag{30}$$

for the problem of the spherical droplet made of an linear elastic isotropic medium. However the condition of vanishing simple and double traction at the free outer surface leads to a homogeneous equation so that the integration constant is identified as $C = 0$. This leads to the trivial solution $\Delta = 0$ and homogeneous mass density within the droplet. Accordingly, the description of capillarity effects in a linear elastic medium requires the introduction of the second derivatives of the strain in the free energy density. This fact was not mentioned in the subsequent works on the links between capillarity and strain gradient theories.

4. Micromorphic approach

Higher order strain gradient theories can be viewed as special cases of the general micromorphic medium introduced by Germain [16]. The material point is treated as a material volume with a small, but finite, size. Additional degrees of freedom are introduced to describe more accurately the relative motion of this particle with respect to its center of mass. Following the method of virtual work, the work density of internal forces is introduced as a linear form with respect to the degrees of freedom and their first gradient:

$$w^{(i)}(\underline{\boldsymbol{u}}, \underset{\sim}{\boldsymbol{\chi}}, \underset{\sim}{\underline{\boldsymbol{\chi}}}, \underset{\sim}{\underline{\boldsymbol{K}}}, \underset{\approx}{\boldsymbol{K}}) = (\underset{\sim}{\boldsymbol{\sigma}} + \underset{\sim}{\boldsymbol{s}}) : (\underline{\boldsymbol{u}} \otimes \nabla) - (\underset{\sim}{\boldsymbol{s}} : \underset{\sim}{\boldsymbol{\chi}} + \underset{\sim}{\underline{\boldsymbol{s}}} \,\vdots\, \underset{\sim}{\underline{\boldsymbol{\chi}}}) + \underset{\sim}{\underline{\boldsymbol{S}}} \,\vdots\, \underset{\sim}{\underline{\boldsymbol{K}}} + \underset{\approx}{\boldsymbol{S}} :: \underset{\approx}{\boldsymbol{K}}, \tag{31}$$

where Germain's development has been truncated after the second order micromorphic terms. In addition to the displacement $\underline{\boldsymbol{u}}$, the microdeformation $\underset{\sim}{\boldsymbol{\chi}}$ and the second order microdeformation $\underset{\sim}{\underline{\boldsymbol{\chi}}}$ are introduced as independent degrees of freedom, together with their first gradients,

$$\underset{\sim}{\underline{\boldsymbol{K}}} = \underset{\sim}{\boldsymbol{\chi}} \otimes \nabla,\quad \underset{\approx}{\boldsymbol{K}} = \underset{\sim}{\underline{\boldsymbol{\chi}}} \otimes \nabla. \tag{32}$$

The simple force stress tensor, $\underset{\sim}{\boldsymbol{\sigma}}$, the relative stress tensors, $\underset{\sim}{\boldsymbol{s}}$ and $\underset{\sim}{\underline{\boldsymbol{s}}}$, the double and triple stress tensors, $\underset{\sim}{\underline{\boldsymbol{S}}}$ and $\underset{\approx}{\boldsymbol{S}}$, are dual quantities of the strain measures in the virtual work form. They must fulfill the balance equations,

$$(\underset{\sim}{\boldsymbol{\sigma}} + \underset{\sim}{\boldsymbol{s}}).\nabla = 0,\quad \underset{\sim}{\underline{\boldsymbol{S}}}.\nabla + \underset{\sim}{\boldsymbol{s}} = 0,\quad \underset{\approx}{\boldsymbol{S}}.\nabla + \underset{\sim}{\underline{\boldsymbol{s}}} = 0, \tag{33}$$

in the absence of volume forces. The corresponding three sets of boundary conditions are

$$(\underset{\sim}{\boldsymbol{\sigma}} + \underset{\sim}{\boldsymbol{s}}).\underline{\boldsymbol{n}} = \underline{\boldsymbol{t}},\quad \underset{\sim}{\underline{\boldsymbol{S}}}.\underline{\boldsymbol{n}} = \underset{\sim}{\boldsymbol{t}},\quad \underset{\approx}{\boldsymbol{S}}.\underline{\boldsymbol{n}} = \underset{\sim}{\underline{\boldsymbol{t}}}, \tag{34}$$

where $\underline{\boldsymbol{t}}$, $\underset{\sim}{\boldsymbol{t}}$, and $\underset{\sim}{\underline{\boldsymbol{t}}}$ are, respectively, contact simple, double and triple tractions. When the microdeformation $\underset{\sim}{\boldsymbol{\chi}}$ is forced to coincide with the macrodeformation gradient, $\underset{\sim}{\boldsymbol{1}} + \underline{\boldsymbol{u}} \otimes \nabla$, by an internal constraint, and when the second order terms are neglected, the micromorphic theory is known to reduce to the strain gradient theory [10,12]. Similarly, when $\underset{\sim}{\boldsymbol{\chi}}$ and $\underset{\sim}{\underline{\boldsymbol{\chi}}}$ are constrained to coincide with the deformation gradient, $\underset{\sim}{\boldsymbol{1}} + \underline{\boldsymbol{u}} \otimes \nabla$, and its gradient, $\underline{\boldsymbol{u}} \otimes \nabla \otimes \nabla$, respectively, Germain's second order micromorphic medium degenerates into Mindlin's second strain gradient continuum. In contrast to strain gradient theories, the general micromorphic models, especially the boundary conditions (34), are quite straightforward to implement in a finite element code. Penalty factors or Lagrange multipliers can then be introduced to obtain a numerical model for strain gradient media.

We illustrate the second order micromorphic model and make the link with Mindlin's theory in the case of an elastic fluid at small deformation. The degrees of freedom are limited to the displacement, $\underline{\boldsymbol{u}}$, a microdilatation, ${}^\chi\Delta$, and second order microdilatation, ${}^\chi\underline{\boldsymbol{K}}$. The associated strains are the dilatation, Δ, the microdilatation, ${}^\chi\Delta$, its gradient, $\nabla{}^\chi\Delta$, the second order microdilatation ${}^\chi\underline{\boldsymbol{K}}$ and its gradient ${}^\chi\underline{\boldsymbol{K}} \otimes \nabla$. The work of internal forces (31) then reduces to

$$w^{(i)} = -p\Delta - {}^\chi p\,{}^\chi\Delta + {}^\chi\underline{\boldsymbol{p}}.\nabla{}^\chi\Delta - {}^\chi\underline{\boldsymbol{\pi}}.{}^\chi\underline{\boldsymbol{K}} + {}^\chi\underset{\sim}{\boldsymbol{\pi}} : ({}^\chi\underline{\boldsymbol{K}} \otimes \nabla). \tag{35}$$

The generalized pressure tensors satisfy the balance of generalized momentum in the form

$$\nabla p = 0,\quad {}^\chi\underline{\boldsymbol{p}}.\nabla + {}^\chi p = 0,\quad {}^\chi\underset{\sim}{\boldsymbol{\pi}}.\nabla + {}^\chi\underline{\boldsymbol{\pi}} = 0. \tag{36}$$

The corresponding Neumann boundary conditions are

$$p = p_0,\quad {}^\chi\underline{\boldsymbol{p}}.\underline{\boldsymbol{n}} = {}^\chi p_0,\quad {}^\chi\underset{\sim}{\boldsymbol{\pi}}.\underline{\boldsymbol{n}} = {}^\chi\underline{\boldsymbol{\pi}}_0 \tag{37}$$

where p_0, ${}^\chi p_0$ and ${}^\chi\underline{\boldsymbol{\pi}}_0$ are given generalized pressures at the boundary. Dirichlet conditions can also be stated and correspond to the prescription of Δ, ${}^\chi\Delta$ and ${}^\chi\underline{\boldsymbol{K}}$ at the boundary.

The free energy potential, $\Psi(\Delta, {}^\chi\Delta, {}^\chi\underline{\boldsymbol{K}}, {}^\chi\underline{\boldsymbol{K}} \otimes \nabla)$, is used to formulate the state laws

$$-p = \rho\frac{\partial\Psi}{\partial\Delta},\quad -{}^\chi p = \rho\frac{\partial\Psi}{\partial{}^\chi\Delta},\quad {}^\chi\underline{\boldsymbol{p}} = \rho\frac{\partial\Psi}{\partial\nabla{}^\chi\Delta} \tag{38}$$

$$-{}^\chi\underline{\boldsymbol{\pi}} = \rho\frac{\partial\Psi}{\partial{}^\chi\underline{\boldsymbol{K}}},\quad {}^\chi\underset{\sim}{\boldsymbol{\pi}} = \rho\frac{\partial\Psi}{\partial({}^\chi\underline{\boldsymbol{K}} \otimes \nabla)}. \tag{39}$$

The free energy density is taken as an isotropic quadratic function of its arguments:

$$\rho\Psi = \frac{\lambda}{2}\Delta^2 + \frac{{}^\chi\lambda}{2}(\Delta - {}^\chi\Delta)^2 + \frac{a_1}{2}\nabla{}^\chi\Delta.\nabla{}^\chi\Delta + \frac{{}^\chi b}{2}(\nabla{}^\chi\Delta - {}^\chi\underline{\boldsymbol{K}})^2 + \frac{\alpha}{2}(\mathrm{trace}\,({}^\chi\underline{\boldsymbol{K}}\otimes\nabla))^2 + \frac{\beta}{2}({}^\chi\underline{\boldsymbol{K}}\otimes\nabla):({}^\chi\underline{\boldsymbol{K}}\otimes\nabla) + \frac{\gamma}{2}({}^\chi\underline{\boldsymbol{K}}\otimes\nabla):(\nabla\otimes{}^\chi\underline{\boldsymbol{K}}) + (b_0 + c_1\Delta + c_2{}^\chi\Delta)\mathrm{trace}\,({}^\chi\underline{\boldsymbol{K}}\otimes\nabla) \quad (40)$$

In the previous expression, the parameters ${}^\chi\lambda$ and ${}^\chi b$ can be regarded as penalty terms ensuring that the microdilatations ${}^\chi\Delta$ and ${}^\chi\underline{\boldsymbol{K}}$ are sufficiently close to the macrodilatations Δ and $\nabla{}^\chi\Delta$, respectively. The constitutive equations follow,

$$-p = \lambda\Delta + {}^\chi\lambda(\Delta - {}^\chi\Delta) + c_1\mathrm{trace}\,({}^\chi\underline{\boldsymbol{K}}\otimes\nabla),$$

$$-{}^\chi p = -{}^\chi\lambda(\Delta - {}^\chi\Delta) + c_2\mathrm{trace}\,({}^\chi\underline{\boldsymbol{K}}\otimes\nabla), \quad (41)$$

$${}^\chi\underline{\boldsymbol{p}} = a_1\nabla{}^\chi\Delta + {}^\chi b(\nabla{}^\chi\Delta - {}^\chi\underline{\boldsymbol{K}}), \quad -{}^\chi\underline{\boldsymbol{\pi}} = -{}^\chi b(\nabla{}^\chi\Delta - {}^\chi\underline{\boldsymbol{K}}), \quad (42)$$

$${}^\chi\underset{\sim}{\boldsymbol{\pi}} = (b_0 + c_1\Delta + c_2{}^\chi\Delta + \alpha\mathrm{trace}\,({}^\chi\underline{\boldsymbol{K}}\otimes\nabla))\,\underset{\sim}{\boldsymbol{1}} + \beta{}^\chi\underline{\boldsymbol{K}}\otimes\nabla + \gamma\nabla\otimes{}^\chi\underline{\boldsymbol{K}} \quad (43)$$

The stress tensors ${}^\chi p$, ${}^\chi\underline{\boldsymbol{p}}$, ${}^\chi\underline{\boldsymbol{\pi}}$ and ${}^\chi\underset{\sim}{\boldsymbol{\pi}}$ can be eliminated from the previous relations by means of the balance Eqs. (36) in order to obtain the expression of the effective pressure:

$$-p = \lambda\Delta + c_1\nabla^2\Delta - (a_1 - c_2)\nabla^2{}^\chi\Delta + (c_1 + c_2)\ \mathrm{trace}\,({}^\chi\underline{\boldsymbol{K}}\otimes\nabla) + (\alpha + \beta + \gamma)\nabla^2\ \mathrm{trace}\,({}^\chi\underline{\boldsymbol{K}}\otimes\nabla) \quad (44)$$

When the following constraints are enforced:

$${}^\chi\Delta \equiv \Delta, \quad {}^\chi\underline{\boldsymbol{K}} \equiv \nabla\Delta, \quad (45)$$

the effective pressure (44) becomes

$$-p = \lambda\Delta - (a_1 - 2(c_1 + c_2))\nabla^2\Delta + (\alpha + \beta + \gamma)\nabla^4\Delta. \quad (46)$$

This expression can then be identified with the effective pressure (20) in Mindlin's second gradient of strain theory. Therefore, the constrained second order micromorphic model coincides with Mindlin's theory.

The model can now be applied to the problem of the elastic spherical droplet with free outer boundary at $r = r_0$. For the sake of conciseness, the form of the solution is given here only for the first order microdilatation model, i.e. dropping the terms involving ${}^\chi\underline{\boldsymbol{K}}$ (see [13]). The balance and constitutive equations reduce to the following system of equations

$$\lambda\Delta + {}^\chi\lambda(\Delta - {}^\chi\Delta) = 0, \quad {}^\chi\lambda(\Delta - {}^\chi\Delta) + a_1\nabla^2{}^\chi\Delta = 0. \quad (47)$$

Elimination of dilatation leads to the following partial differential equation for the microdilatation:

$${}^\chi\Delta - \frac{a_1(\lambda + {}^\chi\lambda)}{\lambda{}^\chi\lambda}\nabla^2{}^\chi\Delta = 0. \quad (48)$$

For the elastic droplet, a solution of the form

$${}^\chi\Delta = \frac{C}{r}\sinh\frac{r}{{}^\chi l} \quad (49)$$

can be worked out. The characteristic length is found to be

$${}^\chi l^2 = \frac{a_1}{\lambda}\,\frac{\lambda + {}^\chi\lambda}{{}^\chi\lambda}. \quad (50)$$

The macrodilatation turns out to be proportional to the microdilatation

$$\Delta = \frac{{}^\chi\lambda}{\lambda + {}^\chi\lambda}{}^\chi\Delta. \quad (51)$$

When the penalty coefficient ${}^\chi\lambda$ tends to infinity, the macro and microdilatation coincide and the previous length becomes $l^2 = A/\lambda$. Mindlin's result (23), specified for $c_i = b_i = 0$, is retrieved in that way. The outer boundary being free of forces, the constant C is found to vanish so that the material density remains constant and uniform. Only the consideration of the second order microdilatation ${}^\chi\underline{\boldsymbol{K}}$ can lead to a non trivial solution due to the initial cohesion stresses associated with the material parameter b_0, in the same way as in Mindlin's original theory.

5. Conclusion

The continuum description of capillarity effects in elastic bodies is possible based on the introduction of higher order gradients of the strain tensor. The Korteweg equation can be incorporated into the framework of a first strain gradient theory. It can be used to model capillarity effects at the interface between two phases of different densities like droplets in vapor, particularly when it is embedded in a phase field simulation. However we have shown, following Mindlin's arguments, that the linear elastic isotropic first strain gradient theory is not sufficient to describe internal strains and stresses that develop close to free surfaces. Their existence requires initial third order stresses accounting for cohesion forces. The cohesion material property in an isotropic elastic second gradient of strain medium is fully characterized by a single parameter b_0 that can be linked to the surface tension when going to the sharp interface limit. Although the explicit example treated in this work was dedicated to elastic fluids, Mindlin's second strain gradient theory exists for solids at small strains. Also, the balance equations of the second order micromorphic model where given for elastic solids at small strain.

Such a situation is encountered for instance in single crystalline nano-particles, as computed from molecular statics simulations [11]. Indeed, in such small atomic aggregates, the lattice parameter field is strongly inhomogeneous due to the small distance between the particle core and its free surface. A surface tension model would be inappropriate since, at that size, the capillary effect is not confined to an infinitesimal surface. That is why Mindlin's second strain gradient theory seems to be suitable. Corresponding finite element computations of nano-particles based on the proposed second order micromorphic model could then be performed. The obtained strain distribution could be directly compared to atomistic computations. In particular the higher order elasticity moduli could be identified in that way from the discrete computations, as Mindlin did for the one-dimensional atomic chain. Mindlin's theory and the general micromorphic simulations could also be used to represent grain boundary stresses in nanocrystals [24], or in nano-objects like wires and layers [31]. This will require extension of the theory to anisotropic cases.

Finite element simulations based on strain gradient theories are quite challenging, especially because of the complex boundary conditions. In contrast, Eringen's micromorphic theory is based on the introduction of independent deformation degrees of freedom and their gradients [10]. It has been shown that the corresponding boundary conditions are significantly less involved. The numerical implementation merely relies on the introduction of additional degrees of freedom and the computation of their first gradient. Lagrange multipliers or penalization method can then be used to retrieve the strain gradient formulation. Such an implementation has already been done for the first order micromorphic model in [28,7]. It remains to be done for the second order micromorphic theory presented in this work.

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