

# Second-gradient theory : application to Cahn-Hilliard fluids

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**Abstract.** The second gradient theory is indispensable for describing a particular class of materials: when the energy depends on the second gradient of the displacement. We show that trying to overcome the difficulty by using extended thermodynamics is similar to trying to describe a linear elastic material without invoking the Cauchy stress tensor. We illustrate the principal features of second gradient theory with the example of Cahn-Hilliard fluids.

**Keywords:** Second-gradient theory, Cahn-Hilliard fluid, Extended thermodynamics

## 1. INTRODUCTION

Force and moment are the basic concepts in the mechanics of rigid bodies, but it is well known that these notions are not sufficient for an accurate description of internal forces in a deformable body. A great improvement in continuum mechanics has been achieved with the introduction of the Cauchy stress tensor. The aim of this paper is to show that, for some particular materials, a comparable improvement is obtained by using the second gradient theory formalized by P. Germain in [Germain, 1973 a], [Germain, 1973 b]. This theory can be formulated only in the framework of the principle of virtual power : one assumes that the power  $\tilde{\mathcal{P}}^{(i)}$  of internal forces in a domain  $\Omega$ , for any possible virtual velocity field  $\tilde{V}$  can be represented by two tensor fields  $\Sigma$ ,  $S$  (of order two and three, respectively) as follows

$$\tilde{\mathcal{P}}^{(i)} = - \int_{\Omega} (\Sigma : \nabla \tilde{V} + S : \nabla \nabla \tilde{V}) dv . \quad (1)$$

If we assume that long range external forces can be represented by a volume density  $f^{(e)}$ , the application of the principle of virtual power, after integration by parts, leads to the local equation:

$$\rho \frac{dV}{dt} = \operatorname{div}(\Sigma - \operatorname{div}(S)) + f^{(e)} \quad (2)$$



where  $\rho$  denotes the mass density. In this momentum balance equation, the tensor  $\sigma := \Sigma - \text{div}(S)$  plays the role of the Cauchy stress tensor. Using  $\sigma$ , we may write the power of internal forces as:

$$\tilde{\mathcal{P}}^{(i)} = - \int_{\Omega} (\sigma : \nabla \tilde{V} + \text{div}(\nabla \tilde{V}^t : S)) \, dv. \quad (3)$$

This power is not determined only by strain and the Cauchy stress tensor : an extra flux  $\nabla \tilde{V}^t : S$  appears. We focus on this main feature of the theory in section 4 while we study boundary conditions in section 6.

The questions which naturally arise are i) what is the connection between this theory and the Cauchy stress theory? ii) is there any material which needs for such a theory? iii) is it possible to reformulate the theory without invoking the virtual power principle, and what are the connections with interstitial working or extended thermodynamics theories? iv) what are the principal mechanical features of this theory? We illustrate the answers to these different questions by the example of Cahn-Hilliard fluids.

## 2. SECOND-GRADIENT THEORY AND CAUCHY CONSTRUCTION OF STRESS TENSOR

To understand how one can get out of the framework of Cauchy continua, we need to reconsider the basic assumptions for Cauchy's construction of the stress tensor : the main postulate states that contact forces can be represented by a surface density of forces which depends only on the normal to the boundary of the considered domain. Noll's theorem establishes this postulate under the implicit assumption that no edge forces are present.

[Noll et al., 1990], [Dell'Isola et al., 1997] have considered the case of continuous media in which edge forces are present : it has been proved that this presence implies a dependence of contact surface forces on the curvature of the boundary of the domain. To be precisely described, the contact forces need two stress tensors  $(\Sigma, S)$ . Moreover, edge forces imply the presence of another type of contact forces which cannot be interpreted in terms of forces or moments, and which correspond to the double forces described by P.Germain in [Germain, 1973 a]. In other words, when edge forces are present, a construction similar to Cauchy's again leads to a second-gradient theory. Moreover, as contact forces contain double forces (i.e. distributions of order one), duality (i.e. virtual power principle) cannot be avoided.

As a matter of fact, edge forces are present in some particular materials, such as Cahn-Hilliard fluids (see section 6).

### 3. SECOND GRADIENT MATERIALS

Let us call a material with a free energy density which depends on the second gradient of the deformation a *second gradient material*. We will see in the next section why such materials need to be described in the framework of the second gradient theory. Such materials do exist. Some have been studied for a long time : the beam model introduced by Euler, Bernoulli and Navier is a one dimensional second gradient material; the first 3D-model was introduced by Cosserat ([Cosserat et al., 1909]). However, in these examples, the way the free energy density depends on the second gradient of the deformation is special : only the gradient of the skew part of the gradient of displacement plays a role. These special cases are called *incomplete second gradient materials* in [Alibert et al., 1999].

More recently, a second gradient material has been introduced by Cahn and Hilliard for describing capillary phenomena [Cahn et al., 1959]. In this model one assumes that the free energy density  $\psi$  depends on the gradient of mass density in the simple form:

$$\psi = W(\rho) + \frac{\lambda}{2}(\nabla\rho)^2. \quad (4)$$

Note that, for sake of simplicity, we assume in this paper that the phenomena occur in isothermal conditions. Clearly, the dependence on  $\rho$  of the gradient of the displacement shows that a Cahn-Hilliard fluid is a complete second gradient material.

The simple form (4) is probably not sufficient for the study of complex fluids. However, it allows one to investigate phenomena which otherwise could not be studied in the framework of continuum mechanics : dependence of surface tension upon the radius for very small droplets [Dell'Isola et al., 1995], stability of wetting films [Seppecher, 1993], interpretation of Young's law for contact angle in terms of forces [Seppecher, 1989], removal of the moving contact line paradox [Seppecher, 1996], line tension phenomenon [Alberti et al., 1998], etc. The model can be used also for numerical studies where it removes the difficulty of tracking interfaces.

In expression (4),  $W$  is a non-convex function. This is an important point. Any energy described by a non convex function leads, in general, to ill-posed equilibrium problems : minimizing  $\int_{\Omega} W(\rho)dx$  under some mass constraint has, in general, no solution. This phenomenon arises in every multiphase problem, in damage or plasticity theories, etc. Adding to the energy a term depending, in a convex way, on higher gradient of deformation has two different interests : from a mathematical point of view, it leads to well-posed "regularized problems"; from a mechanical point of view, it gives a description of the transition zones which divide the phases.

Apart from their mechanical and mathematical interest, there is another reason to require that any general theory of continuum mechanics be able to describe second gradient materials. As continuum mechanics is a homogenized description of materials which are heterogeneous at the microscopic level, it is natural to expect it to be stable under homogenization or asymptotic analysis procedures. Cauchy

continua do not enjoy this stability property : it is well known that the 1-D or 2-D second gradient models of beams or plates are the asymptotic limits of Cauchy models. It has recently been proved that homogenization of heterogeneous Cauchy materials can lead to a 3-D incomplete second gradient material [Pideri et al., 1997]. The fact that a complete second gradient material, and more generally any n-th gradient material, can be obtained in that way is, as far as I know, a conjecture.

In the next section we recall why second gradient materials cannot be considered as classical Cauchy continua.

#### 4. THERMODYNAMICAL PARADOX OF SECOND GRADIENT MATERIALS

Let us write the laws of thermodynamics in a very general way. The first law states that the variation of total energy is due to the mechanical power of the external forces  $\mathcal{P}^{(e)}$ , and a heat supply  $Q_e$  :

$$\dot{E} + \dot{K} = \mathcal{P}^{(e)} + Q_e \quad (5)$$

where  $\dot{E}$  and  $\dot{K}$  denote respectively the variations of internal and kinetic energies. Taking into account the fact that the variation of kinetic energy coincides with the power of internal and external forces

$$\dot{K} = \mathcal{P}^{(i)} + \mathcal{P}^{(e)}, \quad (6)$$

we get for the variation of internal energy:

$$\dot{E} = -\mathcal{P}^{(i)} + Q_e . \quad (7)$$

The second law states that the variation of entropy  $\dot{S}$  is larger than the entropy supply  $Q_s$ :

$$\dot{S} \geq Q_s . \quad (8)$$

If we accept the relatively weak assumptions that  $E$ ,  $S$ ,  $\mathcal{P}^{(i)}$  can be represented by volume densities  $e$ ,  $s$ ,  $p^{(i)}$ ; and the supplies  $Q_e$ ,  $Q_s$  by fluxes  $J_e$ ,  $J_s$ , then inequality (8) provides the Clausius-Duhem inequality :

$$T \operatorname{div}(J_s) - \operatorname{div}(J_e) - p^{(i)} + \rho \left( T \frac{d}{dt} \left( \frac{s}{\rho} \right) - \frac{d}{dt} \left( \frac{e}{\rho} \right) \right) \geq 0 \quad (9)$$

and in isothermal conditions

$$\operatorname{div}(T J_s - J_e) - p^{(i)} - \rho \frac{d}{dt} \left( \frac{\psi}{\rho} \right) \geq 0 \quad (10)$$

where  $\psi$  denotes the volume free energy:  $\psi = e - Ts$  and  $T$  the absolute temperature. The thermodynamical paradox of second gradient materials lies in the

incompatibility of the Clausius-Duhem inequality (10) with the three following assumptions:

( $H_1$ ) The free energy density  $\psi$  depends on the second gradient of the displacement.

( $H_2$ )  $p^{(i)} = -\sigma : \nabla V$  (where  $\sigma$  denotes the Cauchy stress tensor).

( $H_3$ )  $TJ_s = J_e$ , which classically results from two assumptions:  $J_e$  coincides with the heat flux ( $J_e = q$ ) and  $J_s$  coincides with the ratio of heat flux and absolute temperature.

Indeed, under assumption ( $H_3$ ), the term  $div(TJ_s - J_e)$  vanishes. Due to assumption ( $H_1$ ),  $\frac{d}{dt}(\frac{\psi}{\rho})$  contains a term depending linearly on  $\nabla\nabla V$  which cannot be balanced by  $p^{(i)}$  under assumption ( $H_2$ ).

This becomes clear when considering the example of Cahn-Hilliard fluids : when the dependence of  $\psi$  on the displacement is given by (4),  $\frac{d}{dt}(\frac{\psi}{\rho})$  can easily be computed and, under assumptions ( $H_2$ ) and ( $H_3$ ), inequality (10) becomes

$$\begin{aligned} \left( \sigma - (W(\rho) - \rho \frac{dW}{d\rho} - \frac{\lambda}{2} \nabla \rho^2) \text{Id} + \lambda \nabla \rho \otimes \nabla \rho \right) : \nabla V \\ + \lambda \rho (\text{Id} \otimes \nabla \rho) : \nabla \nabla V \geq 0 \end{aligned} \quad (11)$$

which cannot be assured by any constitutive equation for  $\sigma$ .

As, for a second gradient material, ( $H_1$ ) is a basic assumption, the only possibilities for solving the paradox involve revising ( $H_2$ ) or ( $H_3$ ). One can revise ( $H_2$ ) by using the second gradient theory, assuming that  $\mathcal{P}^{(i)}$  has the form (1), i.e., in a local form:

$$p^{(i)} = -\Sigma : \nabla V - S : \nabla \nabla V \quad (12)$$

or, in the equivalent form (3),

$$p^{(i)} = -\sigma : \nabla V - div(\nabla V^t : S) . \quad (13)$$

One can revise ( $H_3$ ) in two ways: either, following [Dunn, 1986], by introducing an “interstitial working” flux  $J_i$ , writing

$$J_e = q + J_i \quad (14)$$

or, as in [Müller, 1985], by writing

$$TJ_s = J_e - J_i . \quad (15)$$

In these two ways, which we can call extended-thermodynamics methods, one has to search for constitutive equations for  $\sigma$  and  $J_i$  which assure inequality (10) while, in the second gradient theory, one has to look for constitutive equations for  $\Sigma$  and  $S$ . Clearly, the two extended-thermodynamics methods are equivalent. The only difference is a question of nomenclature : what is called “heat flux”.

At first glance, the second gradient method seems also to be equivalent to the extended-thermodynamics methods. The term  $\nabla V^t : S$  plays the role of  $J_i$ , and the difference is a question of nomenclature: what is called “power of internal forces”. This is not true. Constitutive equations for  $\Sigma$  and  $S$  concern all admissible velocity fields, and not only the real one; the second gradient theory is stronger. To make this point clear, we show in the next section the consequences of an application of extended thermodynamics to classical Cauchy continua.

Before showing this, let us conclude this section by writing possible constitutive equations for a Cahn-Hilliard fluid. For the sake of simplicity we only recall the non dissipative constitutive equations (for the dissipative case refer to [Seppecher, 1996]). Then inequality (10) is actually an equality,

$$\operatorname{div}(TJ_s - J_e) - p^{(i)} - \rho \frac{d}{dt} \left( \frac{\psi}{\rho} \right) = 0 \quad (16)$$

which is satisfied if we set :

either, in the second-gradient theory,

$$\begin{aligned} TJ_s - J_e &= 0, \quad p^{(i)} = -\Sigma : \nabla V - S : \nabla \nabla V, \\ \Sigma &= (W(\rho) - \rho \frac{dW}{d\rho} - \frac{\lambda}{2} \nabla \rho^2) \operatorname{Id} - \lambda \nabla \rho \otimes \nabla \rho, \quad S = -\lambda \rho (\operatorname{Id} \otimes \nabla \rho), \end{aligned} \quad (17)$$

or, in the extended-thermodynamics framework,

$$\begin{aligned} TJ_s - J_e &= J_i, \quad J_i = -\lambda \rho \operatorname{div}(V) \nabla \rho, \quad p^{(i)} = -\sigma : \nabla V, \\ \sigma &= (W(\rho) - \rho \frac{dW}{d\rho} + \frac{\lambda}{2} \nabla \rho^2 + \lambda \rho \Delta \rho) \operatorname{Id} - \lambda \nabla \rho \otimes \nabla \rho. \end{aligned} \quad (18)$$

## 5. A WEAKNESS OF EXTENDED THERMODYNAMICS

In this section we show that applying the methods of extended thermodynamics proposed in (14) or in (15) to Cauchy materials is a vain attempt to describe these materials without invoking the Cauchy stress tensor. For this purpose, let us imagine, for a moment, that we are beginners in continuum mechanics; we try to describe a classical material, for instance a linear elastic one, and we make the following (frequent) error : the divergence of the Cauchy stress tensor  $\operatorname{div}(\sigma)$  corresponds to the volume density of internal forces; denoting it by  $f^{(i)}$ , we write that the power of internal forces is

$$\mathcal{P}^{(i)} = \int_{\Omega} f^{(i)} \cdot V \, dv. \quad (19)$$

Is that really an error? If we use extended-thermodynamics methods (14) or (15), we may introduce an extra flux  $J_i$  (considering it, following the different points of

view, as an interstitial working or an extra entropy flux) and get the following set of equations

$$\rho \frac{d}{dt} V = f^{(i)} \quad (20)$$

$$\rho \frac{d}{dt} \left( \frac{e}{\rho} \right) = f^{(i)} \cdot V - \operatorname{div}(J_i) - \operatorname{div}(q) \quad (21)$$

where  $f^{(i)}$  and  $J_i$  are given by adequate constitutive equations.

It is remarkable that i) this set of equations is (with the right constitutive equations) totally equivalent to the classical set of equations (we let the reader compute the right constitutive equations which give a set of equations totally equivalent to the equations of linear elasticity), ii) in such a presentation the notion of Cauchy stress tensor is not needed!

Actually, the only but important weakness of the formulation is that boundary conditions *cannot* be written. Extended-thermodynamics is not able to detect the original mechanical error (19). We claim that the same phenomenon occurs when extended-thermodynamics are used to describe a second gradient material, i.e. when  $\sigma$  and  $J_i$  are used instead of  $\Sigma$  and  $S$  for the representation of the mechanical state. The set of local equations is the same in both cases, but only the second gradient theory gives the right boundary conditions. We recall these conditions in the next section.

## 6. BOUNDARY CONDITIONS IN SECOND-GRADIENT THEORY

Let us recall the boundary conditions given by the second-gradient theory (details of computation can be found in [Germain, 1973 a] or [Seppecher, 1989]). Integration by parts of the virtual power of internal forces given by (3) yields

$$\tilde{\mathcal{P}}^{(i)} = \int_{\Omega} \operatorname{div}(\sigma) \cdot \tilde{V} \, dv - \int_{\partial\Omega} (\tilde{V} \cdot \sigma + \nabla \tilde{V}^t : S) \cdot n \, ds \quad (22)$$

where  $n$  denotes the external normal to the domain  $\Omega$ . A second integration by parts on the boundary (assumed to be sufficiently regular) gives

$$\begin{aligned} \tilde{\mathcal{P}}^{(i)} = \int_{\Omega} \operatorname{div}(\sigma) \cdot \tilde{V} \, dv - \int_{\partial\Omega} \tilde{V} \cdot (\sigma \cdot n - \operatorname{div}^s((\operatorname{Id} - n \otimes n) \cdot S \cdot n)) \, ds \\ - \int_{\partial\Omega} (n \cdot \nabla \tilde{V}^t) \cdot (S : (n \otimes n)) \, ds \end{aligned} \quad (23)$$

where  $\operatorname{div}^s$  denotes the intrinsic surface divergence operator. As  $\tilde{V}$  and  $n \cdot \nabla \tilde{V}^t$  are independent fields on  $\partial\Omega$ , the dual quantities  $\sigma \cdot n - \operatorname{div}^s((\operatorname{Id} - n \otimes n) \cdot S \cdot n)$  and  $S : (n \otimes n)$  should be given there. If the exterior is again a second-gradient material,

these quantities should be continuous across  $\partial\Omega$ . In all generality they are fixed by external contact interaction:

$$\sigma \cdot n - \operatorname{div}^s((\operatorname{Id} - n \otimes n) \cdot S \cdot n) = F^{(e)} \quad (24)$$

$$S : (n \otimes n) = G^{(e)} \quad (25)$$

The quantity  $-F^{(e)}$  corresponds to the surface density of forces exerted by the material on the boundary. It is remarkable that, as  $F^{(e)}$  depends on the curvature of  $\partial\Omega$  (i.e. on  $\operatorname{div}^s(n)$ ), Cauchy's postulate is not verified. When the curvature of the boundary is very large,  $F^{(e)}$  becomes singular; indeed when  $\partial\Omega$  is piecewise regular, a line density of forces appears on the edges  $\partial\partial\Omega$ : in equation (23) one must add the term

$$\int_{\partial\partial\Omega} \tilde{V}^t \cdot S : [[n \otimes \nu]] \, d\ell. \quad (26)$$

Here,  $n$  and the vector  $\nu$ , which is tangent to the boundary and normal to the edge, are discontinuous across the edge and  $[[n \otimes \nu]]$  denotes their jump. On the edges the line density of forces

$$S : [[n \otimes \nu]] = \mathcal{F}^{(e)} \quad (27)$$

is exerted. If the velocity vanishes on the boundary, the boundary conditions (24) and (27) are replaced by the dual condition  $\tilde{V} = 0$  on  $\partial\Omega$  but the condition (25) remains valid.

The quantity  $G^{(e)}$  has the dimension of a moment. Its tangent part actually corresponds to a surface density of moment but the normal part is a surface density of double forces.

For a non dissipative Cahn-Hilliard fluid, equation (17) leads to the boundary conditions :

$$\sigma \cdot n - \operatorname{div}^s((\operatorname{Id} - n \otimes n) \cdot S \cdot n) = F^{(e)} \quad (28)$$

$$- \lambda\rho (n \cdot \nabla\rho) n = G^{(e)} \quad (29)$$

$$- \lambda\rho [[(n \cdot \nabla\rho) \nu]] = \mathcal{F}^{(e)} \quad (30)$$

Note that a non-dissipative Cahn-Hilliard fluid can be subjected only to normal double forces  $G^{(e)}$  (just like a non-viscous fluid can be subjected only to normal forces). Hence let us set  $G^{(e)} = g^{(e)} n$ . The equilibrium state of a Cahn-Hilliard fluid in a rigid container, in micro-gravity conditions, is then given by the following equations:

$$\operatorname{div}(\sigma) = \rho \nabla \left( -\frac{dW}{d\rho} + \lambda\Delta\rho \right) = 0 \quad (31)$$

$$- \lambda\rho (n \cdot \nabla\rho) = g^{(e)} \quad (32)$$



Let us compare these boundary conditions with the natural boundary conditions which are obtained using the extended-thermodynamics method (18):

$$\sigma \cdot n = \bar{F}^{(e)} \quad (33)$$

$$J_i \cdot n = \bar{g}^{(e)} \quad (34)$$

The first condition disappears when the velocity vanishes on the boundary, and the second condition reads

$$-\lambda\rho \operatorname{div}(V)(n \cdot \nabla\rho) = \bar{g}^{(e)} \quad (35)$$

This condition is close to (32), but becomes trivial in case of equilibrium. Equation (32) is stronger as it implies an equation of type (35) for all admissible velocity fields and not only the real one.

Clearly the equilibrium problem is well-posed only when the boundary condition (32) is used. The effect of the value of  $g^{(e)}$  on equilibrium is quite important: it tunes the value of Young's contact angle. This parameter is characteristic of the wetting properties of the wall of the container; this has been shown numerically in [Seppecher, 1989] [Seppecher, 1996] and rigourously in [Modica, 1985].

The mechanical concepts of double forces and hyperstress are not purely theoretical concepts. The Cahn-Hilliard fluid provides an example of material for which these concepts correspond to experimental evidence and are as useful as the classical concepts of surface forces and stress tensor.

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