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Internal Variables and Dissipative Structures

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Abstract

The formalism of internal state variables is established when *gradients* of these variables are involved, thus allowing for a spatial localization of dissipative effects that give rise to *dissipative structures*. Therefore, evolution-diffusion equations, rather than usual evolution equations, are obtained. This leads to a comparison between dissipative structures and local structures exhibited by conservative systems. Three illustrative cases are briefly sketched out: nematic liquid crystals, localization of damage coupled to elasticity or plasticity, and localization of plastic strains in plasticity with hardening. Thanks to the absorption of some terms in the extra entropy flux, the approach is valid to any order in the gradients and the accompanying heat equation has always the same form, providing in fact a possible basis for the observation of some of the dissipative structures of interest.

Introduction

The use of thermodynamical *internal* variables of state has been advocated as an efficient means of dealing with *relaxation* and *hysteresis* effects in complex thermodynamical systems [1–3]. However, several recent works [4–6] emphasize that such internal variables should be determined by “complete balance laws” including both a rate term and a divergence term and not according to usual “evolution laws” where the divergence term is absent and with which plasticity, viscoplasticity, damage and fracture in solid mechanics [7] and solutions of macromolecules [8–10] have accustomed us. That is, although the origin of thermodynamical internal variables is to be traced back to the description of reacting chemical systems [11] and fluids with internal degrees of freedom [12], where diffusion of the internal variable is of no practical importance, it seems that many physical phenomena exhibiting a *spatial localization* or a *marked diffusion* of dissipative processes are directly amenable by means of *spatially non-uniform* internal variables. It is the purpose of the present paper to show that the contemporary framework of irreversible thermodynamics accommodates directly

this type of approach. Then the thermodynamical force associated with the internal variable is none other than the (spatial) Euler-Lagrange derivative of the free energy with respect to this variable, and the extra entropy flux is shown to include contributions from the flux of internal variables. Of necessity, thermal equations need to be developed simultaneously.

Short illustrations of the general theory are proposed which pertain to nematic liquid crystals, brittle or ductile damage in deformable solids, and plastic strain localization in plasticity with hardening. Together with nonlinearities the spatial gradients thus introduced allow one to exhibit a general structure which is none other than the one of so-called dissipative structures (see, e. g., Ref. [13]). These are markedly different from solitonic structures which rather follow from a description by means of thermodynamical observable state variables relevant to conservative systems. Essential differences between these two types of structures are emphasized. Finally, it is noticed that the heat equation in the presence of dissipative structures offers a direct means to appraise the dissipation carried by these structures through experimental techniques such as infrared thermography.

1. Contents

The general balance laws and basic thermodynamical statements are recalled in Section 2. State laws and the residual dissipation inequality are given in Section 3 when spatial gradients of the internal variables are taken into account. The corresponding general form of the heat equation is obtained in Section 4. In Section 5 we show how to exploit the dissipation inequality by using dissipation potentials. The ensuing general form of dissipative structures is given in Section 6. These structures and solitonic ones are contrasted in Section 7 by emphasizing that these two different types of localized structures follow from two different limit processes.

Three examples pertaining to nematic liquid crystals (the internal variable is a vector field), damage coupled to elasticity or plasticity (the internal variable is a scalar damage parameter) and plasticity with hardening (the internal variable is the cumulated plastic strain) illustrate our formal considerations in Section 8. General concluding comments are given in Section 9.

We shall use the notation, and work in the spirit, of our forthcoming book on the thermomechanics of plasticity [7].

2. General equations

We consider a continuous medium which is a *simple* material in appearance, conducts heat and, to fix ideas more precisely, we specify that its thermodynamically recoverable behaviour will be of the elastic type in small strains (this will be relaxed in one of the examples). Irreversible processes, other than heat conduction, taking place in the medium will be taken care of through a thermodynam-

ical *internal variable* α and its spatial non uniformity (e. g., its first gradient $\nabla \alpha$). Here α denotes an n vector of \mathcal{R}^n , i. e., it is the ordered list of the n independent components of a certain geometrical object, a scalar ($n = 1$) yielding the simplest case. As our medium apparently is a simple material, the following Euler Cauchy equations of motion hold true as the notion of *simple material* is strictly related to that of Cauchy's stress ([14]; here $T =$ transpose):

$$\rho \dot{\mathbf{v}} = \operatorname{div} \boldsymbol{\sigma} + \mathbf{f}, \quad \boldsymbol{\sigma} = \boldsymbol{\sigma}^T \quad \text{in } \Omega, \quad (2.1)$$

$$\boldsymbol{\sigma} \cdot \mathbf{n} = \mathbf{T} \text{ on } \partial\Omega, \quad \boldsymbol{\sigma} \cdot \mathbf{n} = \hat{\mathbf{T}} \quad \text{on } \partial\Omega_T, \quad (2.2)$$

$$\mathbf{u} = \hat{\mathbf{u}} \quad \text{on } \partial\Omega_u, \quad (2.3)$$

wherein ρ is the matter density, \mathbf{v} is the velocity field, $\boldsymbol{\sigma}$ is Cauchy's stress tensor, \mathbf{f} and $\hat{\mathbf{T}}$ represent volume and surface prescribed forces, $\hat{\mathbf{u}}$ is a prescribed displacement field, and \mathbf{n} is the unit outward normal to the regular boundary $\partial\Omega = \partial\Omega_T \cup \partial\Omega_u$ of the open simply connected set Ω of \mathcal{E}^3 occupied by the material at time t . In the small strain approximation $\mathbf{v} \simeq \dot{\mathbf{u}}$, where a superimposed dot denotes the material time derivative hereby reduced to a partial time derivative.

The thermodynamical aspect of the problem is approached through the two laws of thermodynamics here written in global form for Ω as:

First law of thermodynamics:

$$\dot{K} + \dot{E} = \mathcal{P}_{(\text{ext})} + \dot{Q}; \quad (2.4)$$

Second law of thermodynamics:

$$\dot{\mathcal{N}} \geq \dot{N} \quad (2.5)$$

where

$$\left. \begin{aligned} K &= \int_{\Omega} \frac{1}{2} \rho \mathbf{v}^2 dv, & E &= \int_{\Omega} \rho e dv \\ \mathcal{P}_{(\text{ext})} &= \int_{\Omega} \mathbf{f} \cdot \mathbf{v} dv + \int_{\partial\Omega} \mathbf{T} \cdot \mathbf{v} da, & \dot{Q} &= - \int_{\partial\Omega} \mathbf{q} \cdot \mathbf{n} da, \\ \mathcal{N} &= \int_{\Omega} \rho \eta dv, & \dot{N} &= - \int_{\partial\Omega} \mathbf{S} \cdot \mathbf{n} da. \end{aligned} \right\} \quad (2.6)$$

Here K and E are the total kinetic energy and internal energy, $\mathcal{P}_{(\text{ext})}$ is the power developed by prescribed forces, \dot{Q} is the total flux of heat, \mathcal{N} is the total entropy, and \dot{N} is the total flux of entropy. Because of complex dissipative mechanisms occurring in the material we assume that \mathbf{S} and \mathbf{q} are related by the somewhat unconventional relation (cp. Ref. [12])

$$\mathbf{S} = \frac{1}{\Theta} \mathbf{q} + \mathbf{k}, \quad (2.7)$$

where Θ is the thermodynamical temperature ($\Theta > 0$, $\inf \Theta = 0$), and \mathbf{k} is an extra entropy flux density to be determined in the subsequent procedure.

Accounting for the scalar product of equations (2.1) and (2.2)₁ with \mathbf{v} in equation (2.4), we are led to the classical energy equation

$$\dot{E} + \mathcal{P}_{(i)} = \dot{Q}, \quad \mathcal{P}_{(i)} \equiv - \int_{\Omega} \boldsymbol{\sigma} : \dot{\boldsymbol{\varepsilon}} dv, \quad (2.8)$$

where

$$\boldsymbol{\varepsilon} = (\nabla \mathbf{u})_s = \frac{1}{2} [\nabla \mathbf{u} + (\nabla \mathbf{u})^T]. \quad (2.9)$$

On localization this yields at all regular points in Ω :

$$\dot{\Sigma} = p_{(i)} - \nabla \cdot \mathbf{q}, \quad p_{(i)} \equiv \boldsymbol{\sigma} : \dot{\boldsymbol{\varepsilon}}, \quad (2.10)$$

where $\Sigma = \rho e$ is the internal energy per unit volume (we account for the small-strain hypothesis), and $p_{(i)}$ is the elementary power developed by the Cauchy stress. The localization of equation (2.5) directly yields the local entropy inequality as ($S = \rho \eta$)

$$\dot{S} + \nabla \cdot \mathbf{S} \geq 0 \quad (2.11)$$

or (as $\Theta > 0$)

$$\Theta \dot{S} + \nabla \cdot (\Theta \mathbf{S}) - (\mathbf{S} \cdot \nabla) \Theta \geq 0. \quad (2.12)$$

Now we combine (2.10) and (2.12) in the usual manner by introducing the Helmholtz free energy per unit volume by

$$\Psi = \Sigma - S \Theta. \quad (2.13)$$

The following Clausius Duhem (CD) inequality follows at once:

$$- (\dot{\Psi} + S \dot{\Theta}) + p_{(i)} + \nabla \cdot (\Theta \mathbf{k}) - (\mathbf{S} \cdot \nabla) \Theta \geq 0. \quad (2.14)$$

3. State laws and dissipation inequality

We need to specify the functional dependence of Ψ to proceed with the exploitation of the CD inequality (2.14). It is only at this point that internal variables enter the picture. We assume (small-strain hypothesis) that $\boldsymbol{\varepsilon}$ splits additively in an elastic part $\boldsymbol{\varepsilon}^e$ and an anelastic part $\boldsymbol{\varepsilon}^p$:

$$\boldsymbol{\varepsilon} = (\nabla \mathbf{u})_s = \boldsymbol{\varepsilon}^e + \boldsymbol{\varepsilon}^p. \quad (3.1)$$

In the spirit of plasticity and viscoplasticity, we suppose that Ψ depends on $\boldsymbol{\varepsilon}$ only through $\boldsymbol{\varepsilon}^e$, and it further depends on Θ and α , and possibly on the first gradient of the latter, i. e., $\nabla \alpha$ (see Appendix for the second gradient). Ψ is assumed to be

concave in Θ but it is *not necessarily convex* in ε^e and α , although it may be so in many special cases. Convexity with respect to $\nabla\alpha$ holds good. Thus, symbolically,

$$\Psi = \Psi(\varepsilon^e, \Theta; \alpha, \nabla\alpha). \quad (3.2)$$

Through a straightforward calculation, this allows us to obtain $\dot{\Psi}$ in the form

$$\dot{\Psi} = -\hat{S}\dot{\Theta} + \hat{\sigma} : \dot{\varepsilon}^e - \mathcal{A}\dot{\alpha} + \nabla \cdot (\mathbf{B}\dot{\alpha}), \quad (3.3)$$

where we have *defined* the following free energy derivatives:

$$\begin{aligned} \hat{S} &= -\frac{\partial \Psi}{\partial \Theta}, & \hat{\sigma} &= \frac{\partial \Psi}{\partial \sigma^e} \\ \mathcal{A} &= -\frac{\partial \Psi}{\partial \alpha}, & \mathbf{B} &= -\frac{\partial \Psi}{\partial \nabla\alpha} \end{aligned} \quad (3.4)$$

and

$$\mathcal{A} = A - \nabla \cdot \mathbf{B} = -\delta\Psi/\delta\alpha, \quad (3.5)$$

where

$$\frac{\delta}{\delta\alpha} = \frac{\partial}{\partial\alpha} - \nabla \cdot \frac{\partial}{\partial\nabla\alpha} \quad (3.6)$$

is the ‘‘space’’ Euler-Lagrange derivative with respect to α . Substituting from (3.3) into (2.14) and accounting for (3.1), we obtain

$$(\hat{S} - S)\dot{\Theta} + (\sigma - \hat{\sigma}) : \dot{\varepsilon}^e + \hat{\sigma} : \dot{\varepsilon}^p + \mathcal{A}\dot{\alpha} + \nabla \cdot (\Theta\mathbf{k} - \mathbf{B}\dot{\alpha}) - (\mathbf{S} \cdot \nabla)\Theta \geq 0. \quad (3.7)$$

As S and σ are assumed not to depend on $\dot{\Theta}$ and $\dot{\varepsilon}^e$ while the remaining coefficients in (3.7) may in general depend on their respective factors, and (3.7) has to remain in one sign for any $\dot{\Theta}$ and $\dot{\varepsilon}^e$ (see the argument on p. 112 in Ref. [15]), we obtain that

$$\sigma = \hat{\sigma} = \frac{\partial \Psi}{\partial \varepsilon^e}, \quad S = \hat{S} = -\frac{\partial \Psi}{\partial \Theta}. \quad (3.8)$$

At this point it is astute to select the extra entropy flux \mathbf{k} as

$$\mathbf{k} = \frac{1}{\Theta} \mathbf{B}\dot{\alpha}, \quad (3.9)$$

so that (3.7) reduces to the following *residual dissipation inequality*

$$\Phi \equiv \sigma : \dot{\varepsilon}^p + \mathcal{A}\dot{\alpha} - (\mathbf{S} \cdot \nabla)\Theta \geq 0. \quad (3.10)$$

Very often this is split in two parts (resulting thus in stronger conditions):

$$\Phi_{\text{intr}} \equiv \boldsymbol{\sigma} : \dot{\boldsymbol{\varepsilon}}^p + \mathcal{A} \dot{\alpha} \geq 0, \quad (3.11)$$

$$\Phi_{\text{th}} \equiv -(\mathcal{S} \cdot \nabla) \Theta \geq 0, \quad (3.12)$$

where, in some sense, we recognize the different qualitative nature of the two classes of dissipative processes. Φ_{intr} and Φ_{th} are the intrinsic and thermal dissipations, respectively. The split (3.11)–(3.12) will be assumed in all applications.

At this point we have at hand the thermodynamical state laws (3.8) and the definition of \mathcal{A} and the explicit form of \mathcal{S} , and the inequalities governing dissipative processes, (3.10), or (3.11) and (3.12). The latter are in the standard bilinear form in terms of fluxes and associated forces:

$$\sum_{\beta} X_{\beta} Y_{\beta} \geq 0 \quad (3.13)$$

as used in standard irreversible thermodynamics [16]. Before exploiting (3.13) we examine the general expression of the heat equation.

4. Heat equation

The heat equation is none other than equation (2.10)₁ in a disguise. Indeed, on using (2.13), (3.3) and (3.8) in (2.10)₁ or, equivalently, just comparing (2.12) and (3.10), we deduce the first (general) form of the heat equation:

$$\Theta \dot{S} + \nabla \cdot (\Theta \mathcal{S}) = \Phi_{\text{intr}}, \quad (4.1)$$

where the intrinsic dissipation acts like a body source of heat.

In the present state of progress we can also use the fact that we know \mathcal{S} through (3.8)₂. Hence, evaluating \dot{S} and accounting for (3.8) we readily obtain the second form of the heat equations as

$$C \dot{\Theta} + \nabla \cdot (\Theta \mathcal{S}) = \Phi_{\text{intr}} + \Phi_{\text{te}}, \quad (4.2)$$

in which we have defined the “thermoelastic” dissipation Φ_{te} by

$$\Phi_{\text{te}} \equiv \Theta (\boldsymbol{\tau} : \dot{\boldsymbol{\varepsilon}}^e + l \dot{\alpha} + \boldsymbol{m} \cdot \nabla \dot{\alpha}) \quad (4.3)$$

and set

$$C = -\Theta \frac{\partial^2 \Psi}{\partial \Theta^2} \geq 0, \quad (4.4)$$

$$\boldsymbol{\tau} = -\frac{\partial \boldsymbol{\sigma}}{\partial \Theta}, \quad l = \frac{\partial A}{\partial \Theta}, \quad \boldsymbol{m} = \frac{\partial \boldsymbol{B}}{\partial \Theta},$$

where the non-negativity of the specific heat C follows from the concavity of Ψ

with respect to Θ , ($\partial^2 \Psi / \partial \Theta^2 \leq 0$). τ , together with l and m , are thermoelasticity coefficients. Equation (4.2) emphasizes the role of body heat source played by both Φ_{intr} and Φ_{th} once a simple constitutive law (such as Fourier's) is assumed for the flux S (and not for q).

5. Exploiting the dissipation inequality

Leaving aside the case of S for which a simple law

$$S = -\kappa(\varepsilon^e, \Theta; \alpha) \cdot \nabla \Theta \quad (5.1)$$

with

$$A^T \cdot (\kappa \cdot A) \geq \kappa A^2 \geq 0$$

for any vector A , $\kappa \geq 0$, is sufficient, we examine the exploitation of the inequality (3.11) in the light of dissipation potentials in the manner of Rayleigh [17] or Ziegler [18].

Thermodynamical equilibrium is defined by the condition of spatially uniform temperature and the two conditions:

$$\dot{\varepsilon}^p = 0, \quad \mathcal{A} = 0. \quad (5.2)$$

Therefore, it is natural to postulate the existence of a dissipation potential

$$\mathcal{D}(\sigma, \dot{\alpha}; \varepsilon^e, \Theta, \alpha, \nabla \alpha),$$

a positive function which is homogeneous of a certain degree in σ and $\dot{\alpha}$, and is convex in these two variables so that $\dot{\varepsilon}^p$ and \mathcal{A} be given by the derivatives

$$\dot{\varepsilon}^p = \frac{\partial \mathcal{D}}{\partial \sigma}, \quad \mathcal{A} = \frac{\partial \mathcal{D}}{\partial \dot{\alpha}}. \quad (5.3)$$

With Euler's identity for homogeneous functions the dissipation inequality is automatically satisfied and, thus, (5.1) and (5.3) are *thermodynamically admissible*. The degree of homogeneity specifies the type of dissipative process considered. For instance, if \mathcal{D} is homogeneous of degree one in $\dot{\alpha}$, \mathcal{A} will not in fact depend on $\dot{\alpha}$, and the behaviour with respect to the variable α will be of the "plastic" type, exhibiting no time scale. Similarly, for the more traditional behaviour described by (5.3)₁, if the partial Legendre-Fenchel transform \mathcal{D}^* defined by

$$\mathcal{D}^*(\dot{\varepsilon}^p, \dot{\alpha}; \dots) = \text{Sup}_{\sigma} [\sigma : \dot{\varepsilon}^p - \mathcal{D}(\sigma, \dot{\alpha}; \dots)] \quad (5.4)$$

is homogeneous of degree one in $\dot{\varepsilon}^p$, then the dissipative mechanical behaviour will be of the plastic type (no strain-rate dependence; no time scale). The homogeneity property is conserved in the Legendre-Fenchel transform so that

$$\mathcal{D}(\boldsymbol{\sigma}, \dot{\boldsymbol{\alpha}}; \dots) = \text{Sup}_{\boldsymbol{\varepsilon}^p} [\boldsymbol{\sigma} : \boldsymbol{\varepsilon}^p - \mathcal{D}^*(\boldsymbol{\varepsilon}^p, \dot{\boldsymbol{\alpha}}; \dots)] \quad (5.5)$$

is also homogeneous of degree one in $\boldsymbol{\sigma}$.

If both \mathcal{D} and \mathcal{D}^* are homogeneous of degree *two* in $\boldsymbol{\sigma}$ or $\boldsymbol{\varepsilon}^p$, then the *complementary constitutive equation* (5.3)₁ will exhibit a characteristic (relaxation) time while there may exist a convex set limiting the values of $\boldsymbol{\sigma}$ so that we have viscoplasticity (cp. Ref. [7]). For the time being, on account of (3.5) we record the following possible set of constitutive equations:

$$\boldsymbol{\sigma} = \frac{\partial \Psi}{\partial \boldsymbol{\varepsilon}^e}, \quad \boldsymbol{\varepsilon}^p = \frac{\partial \mathcal{D}^*}{\partial \boldsymbol{\sigma}}, \quad \boldsymbol{\varepsilon} = \boldsymbol{\varepsilon}^e + \boldsymbol{\varepsilon}^p = (\nabla \mathbf{u})_s \quad (5.6)$$

and

$$\frac{\delta \Psi}{\delta \alpha} + \frac{\partial \mathcal{D}}{\partial \dot{\alpha}} = 0. \quad (5.7)$$

We shall comment more thoroughly on the second of these.

6. Dissipative structures

Equation (5.7) reads

$$\left. \begin{aligned} \frac{\delta \Psi}{\delta \alpha} + \frac{\partial \mathcal{D}}{\partial \dot{\alpha}} &= 0 \quad \text{with} \\ \Psi &= \Psi(\boldsymbol{\varepsilon}^e, \Theta; \alpha, \nabla \alpha) \\ \mathcal{D} &= \mathcal{D}(\boldsymbol{\sigma}, \dot{\boldsymbol{\alpha}}; \boldsymbol{\varepsilon}^e, \Theta, \alpha, \nabla \alpha). \end{aligned} \right\} \quad (6.1)$$

For instance, if \mathcal{D} is quadratic in $\dot{\alpha}$ and Ψ is highly nonquadratic in α while remaining quadratic and convex in $\nabla \alpha$, equation (6.1)₁ will result in an *evolution-diffusion* equation of the general type

$$\dot{\alpha} = f(\alpha, \boldsymbol{\sigma}, \boldsymbol{\varepsilon}^e, \Theta) + D_f \alpha, \quad (6.2)$$

where D_f is an operator of diffusion (e.g., ∇^2) and f is *highly nonlinear* in α depending, in addition, on the state of stresses and the field of temperature, these two fields being themselves governed by the evolution-diffusion equations (2.1) and (4.2) in the bulk of the material. The system thus obtained for the fields \mathbf{v} , \mathbf{u} (such that $\dot{\mathbf{u}} = \mathbf{v}$, a simple evolution equation by itself), $\boldsymbol{\varepsilon}^p$, α and Θ clearly offers a formidable potential for the existence of so-called *dissipative structures* and *auto-wave phenomena* (cf. Vasilev et al. [13]) which are so fashionable in contemporary nonlinear physics. The essence of the phenomena here is contained in equations (6.1)₁ or (5.3)₁ and (5.6)₂, the former of which deserves greater attention.

7. Dissipative and solitonic structures

The “dissipative” nature of equation (6.1)₁ directly stems from the manner in which it was obtained and, obviously, from the fact that α is considered as an internal variable. As a consequence of this very formulation no boundary condition is a priori associated with (6.1)₁, while its diffusion like nature, when Ψ depends on $\nabla\alpha$, is evident. Thus accompanying boundary conditions will have to be deduced from (6.1)₁ by using a “local” method, e. g., the so-called pill-box method. However, whether α indeed is an *internal* variable or a more classical *observable* variable of state often is a matter of decision at the outset from the part of the scientist, i. e., it depends on the respective scales (most often *time* scales) at which the processes related to internal and observable variables proceed. Had we considered the new variable α as an additional observable variable, then internal powers associated with the generalized velocities $\dot{\alpha}$ and $\nabla\dot{\alpha}$ would have contributed to $p_{(i)}$ and $\mathcal{P}_{(i)}$ – cp. [14] – and a new balance law would govern α with, in general, a kinetic energy, and thus an inertia, associated with α . Then, after complementing this by a simple study of allied dissipative processes the α -field equation would have read

$$\frac{\delta \mathcal{L}}{\delta \alpha} - \frac{\partial \mathcal{D}}{\partial \dot{\alpha}} = 0, \quad (7.1)$$

where

$$\mathcal{L} = \mathcal{K} - \Psi \quad (7.2)$$

is the Lagrangian per unit volume and

$$\frac{\delta}{\delta \alpha} = \frac{\delta}{\delta \alpha} - \frac{\partial}{\partial t} \frac{\partial}{\partial \dot{\alpha}}. \quad (7.3)$$

The latter is nothing but the *total* Euler-Lagrange derivative with respect to α . Equation (7.1) is the celebrated equation of Rayleigh ([17], Vol. 1, p. 78), used by Maxwell in his famous treatise ([19], Vol. 2, p. 396).

In spite of their common appearance, the philosophies underlying the two equations (5.7) and (7.1) are quite different. Obviously, if time scales are such that relaxation effects dominate over inertial ones (this is practically always the case in nematic liquid crystals where α is the director field \mathbf{d} – cp. de Gennes [20], Kats and Lebedev [21]), then we can write (7.1) as

$$\frac{\delta \Psi}{\delta \alpha} + \frac{\partial \mathcal{D}}{\partial \dot{\alpha}} + \varepsilon \frac{\partial}{\partial t} \left(\frac{\partial \mathcal{K}}{\partial \dot{\alpha}} \right) = 0, \quad (7.4)$$

emphasizing with a scaling factor ε the fact that there are inertial effects which will be discarded. But these will generally be second-order derivatives in time, so that (7.4) may present a *singular* perturbation in the time domain. On the condition that nothing dramatic occurs as ε goes to zero, (7.4) will render our equation for dissipative structures obtained from internal-variable theory, i. e.,

$$\frac{\delta \Psi}{\delta \alpha} + \frac{\partial \mathcal{D}}{\partial \dot{\alpha}} = 0. \quad (7.5)$$

On the contrary, in many cases described by equation (7.1) the problem is almost conservative and dissipation produces only a regular perturbation, so that (7.1) may be rewritten as

$$\frac{\partial}{\partial t} \left(\frac{\partial \mathcal{K}}{\partial \dot{\alpha}} \right) + \frac{\delta \Psi}{\delta \alpha} + \varepsilon \frac{\partial \mathcal{D}}{\partial \dot{\alpha}} = 0, \quad (7.6)$$

where the ε has been shifted to the dissipative contribution. As ε goes to zero, the resulting conservative (albeit, in general, nonlinear and dispersive) equation

$$\frac{\partial}{\partial t} \left(\frac{\partial \mathcal{K}}{\partial \dot{\alpha}} \right) + \frac{\delta \Psi}{\delta \alpha} = 0 \quad (7.7)$$

will follow without ambiguity.

The two writings (7.4) and (7.6) of (7.1) highlight the essential differences between nonlinear evolution-diffusion systems that may exhibit dissipative structures and auto-waves as $\varepsilon \rightarrow 0$, and nonlinear conservative systems slightly perturbed by dissipation which may exhibit pure solitonic structures as $\varepsilon \rightarrow 0$. Structures in nonlinear dissipative systems of the first kind and those in nonlinear conservative systems may have some features in common. Solitary excitation fronts (E. g., motion of a phase-transition interface) and pulses propagating with a stable shape (so-called travelling pulses) are examples of structures which may propagate in both schemes. However, the physical nature and the behaviour of these structures are quite different in these two schemes, as one can gather from the two limits obtained in (7.4) and (7.6) as ε goes to zero. An example of this is the well known fact that, in the second scheme, colliding solitons penetrate through each other and then recover their individuality up to a possible alteration in phase, whereas in the case of travelling dissipative structures, collision of two travelling pulses or fronts usually results in their mutual annihilation. Furthermore, while shape and propagation velocity of stationary travelling pulses are practically insensitive to initial conditions for dissipative structures, both shape and propagation velocity of solitons are completely fixed by the initial excitation. This was noticed by Vasilev et al. [13], p.15.

The above lack of distinction between observable and internal natures of some state variables unfortunately exhibits a prevailing ambiguity. For instance, permanent electric dipoles in elastic ferroelectrics are governed by an equation such as (7.6) in which the second term involves strong nonlinearities, dispersion, and coupling with elasticity. Electroelastic solitons propagate in such a structure [22]. In that case the electric polarization is viewed as an observable variable (additional degree of freedom). But to study electromechanical hysteresis in ceramics, an internal variable akin to an electric polarization (residual polariz-

ation vector, cumulated (scalar) electric polarization) can be introduced [23]. However, diffusion of this last variable is not yet envisaged, but it could be. Similar remarks hold for the magnetic spin density in elastic ferromagnets (e. g., [24] and [3]).

It seems that the internal-variable thermodynamical approach is the best suited general framework for the first scheme (dissipative structures). This should come as no surprise if we recall the origin of the concept of internal variables in the kinetics of reactive systems [11]. Altogether, however, specific results can only be obtained once the physical and geometrical (tensor order) nature of the relevant internal variables has been specified (see below).

8. Examples

A. Vectorial internal variable (liquid crystals)

Let $\alpha = \{d_i; i = 1, 2, 3\}$ a vector field \mathbf{d} called director. Mechanically, the continuous medium considered does not present any plasticity-like behaviour. It rather is a fluid of the viscous type, although incompressible. It is easy to see that $\dot{\epsilon}$ is replaced by the exact rate-of-strain tensor $\mathbf{D} = (\nabla \mathbf{v})_S$ and $p_{(i)}$ in (2.10) reads

$$p_{(i)} = \boldsymbol{\sigma}^v : \mathbf{D}, \quad \boldsymbol{\sigma}^v = \boldsymbol{\sigma} + p\mathbf{I}, \quad (8.1)$$

where p is the mechanical pressure. Equation (3.2) is replaced by

$$\Psi = \Psi(\Theta; \mathbf{d}, \nabla \mathbf{d}). \quad (8.2)$$

This free-energy function satisfies a space-isotropy requirement and thus, classically, we must check the invariance requirement (obtained by invariance of (8.2) through an infinitesimal rotation of the spatial frame; square brackets indicate anti-symmetrization):

$$\frac{\partial \Psi}{\partial d_{[i}} d_{j]} + \frac{\partial \Psi}{\partial d_{p,k}} \delta_{p[i} d_{j],k} + \frac{\partial \Psi}{\partial d_{k,[i}} \delta_{j]p} d_{k,p} = 0 \quad (8.3)$$

and this also reads [compare (3.4)_{3,4}]

$$A_{[i} d_{j]} + B_{k[i} d_{j],k} + B_{pk} \delta_{p[i} \delta_{j]q} d_{k,q} = 0. \quad (8.4)$$

Through an elementary computation this allows one to show that equation (3.3) is replaced by

$$\dot{\Psi} = -S\dot{\Theta} - \tilde{\boldsymbol{\sigma}} : \mathbf{D} - \mathcal{A} \cdot \mathbf{N} + \nabla \cdot (\mathbf{B} \cdot \mathbf{N}), \quad (8.5)$$

wherein

$$N_i = (D_J \mathbf{d})_i = \dot{d}_i - \Omega_{ij} d_j, \quad \boldsymbol{\Omega} = (\nabla \mathbf{v})_A \quad (8.6)$$

$$\tilde{\sigma}_{ij} = B_{(i|k|} d_{k,j)}, \quad (8.7)$$

where the subscript A denotes antisymmetrization and parentheses denote symmetrization. Proceeding then as in Section 3 one is led to the following residual dissipation inequality

$$\Phi = \tilde{\sigma}^v : \mathbf{D} + \mathcal{A} \cdot \mathbf{N} - (\mathbf{S} \cdot \nabla) \Theta \geq 0, \quad (8.8)$$

with

$$\tilde{\sigma}_{ij}^v = \sigma_{ij}^v + \tilde{\sigma}_{ij} = \sigma_{ij} + p \delta_{ij} + B_{(ik} d_{k,j)}, \quad (8.9)$$

$$k_i = \frac{1}{\Theta} B_{iq} N_q. \quad (8.10)$$

Here N , the director velocity, is defined as the Jaumann co-rotational time derivative of the director field. This is a so-called *objective* time rate (cf. Ref. [15], p. 83). As a matter of fact, Φ is entirely expressed in terms of objective quantities. The last of (8.9) shows that the fluid considered does not obey Euler's hydrodynamics at rest unless the distribution of \mathbf{d} 's is spatially uniform. In fact, it is not difficult to recognize in (8.1)–(8.10) the equations governing the thermodynamics of *nematic liquid crystals* [12, 20, 21], where Ψ is specified as Frank's energy density, the director inertia is neglected, and N is written as a linear function of \mathcal{A} , thus providing an evolution-diffusion equation for the internal variable \mathbf{d} . The evolution equation for the fluid velocity (equation of motion (2.1)₁) will be highly nonlinear in \mathbf{d} as $\tilde{\sigma}^v$ depends on \mathbf{d} . The extra entropy flux (8.10) agrees with the one obtained by Kats and Lebedev ([21], p. 23, equation (16)) once N is taken linear in \mathcal{A} .

The above-given scheme would also apply to semi-dilute solutions of elongated (uncoiled) macromolecules when some kind of orientational order prevails (cf. Maugin and Drouot [8]). It easily generalizes to the case where the internal variable is a symmetric second-order tensor, the “conformation” of not necessarily elongated macromolecules [10, 25].

B. Scalar internal variable (localization of damage)

We return to a *solid-like* behaviour. Damage is the reduction in elastic properties (decrease in Young's modulus) as a result of the *irreversible* growth of microcavities and microcracks. In a simple modelling, following the original idea of Kachanov [26], this can be described by a scalar variable D , such that $0 \leq D \leq 1$; which measures the decrease in material surface transmitting internal forces (stresses) independently of the orientation of that surface (isotropy hypothesis). It appears that damage is localized and it is subjected to diffusion under the action of stresses. The idea of considering D as an internal variable is not new (cf. Lemaître and Chaboche, [27]).

(a) We first consider *damage coupled to elasticity* for which plasticity effects can be discarded. Hence $\varepsilon^e = \varepsilon$, and we write

$$\Psi = \Psi(\varepsilon, \Theta, D, \nabla D). \quad (8.11)$$

The formulation of Sections 2 through 6 applies directly with $\alpha = D$. A sensible expression for Ψ which steers the whole problem, is given by

$$\Psi = (1 - D) W_e(\varepsilon, \Theta) + \frac{1}{2} v (\nabla D)^2, \quad (8.12)$$

where W_e is the thermoelastic energy of the undamaged (virgin) material. Thus,

$$\left. \begin{aligned} \sigma &= \frac{\partial \Psi}{\partial \varepsilon} = (1 - D) \frac{\partial W_e}{\partial \varepsilon}, \\ \mathcal{A} &= W_e(\varepsilon, \Theta) + v \nabla^2 D, \\ k &= \frac{1}{\Theta} (\nabla D) \dot{D}, \\ \Phi &= \mathcal{A} \dot{D} - (\mathcal{S} \cdot \nabla) \Theta \geq 0. \end{aligned} \right\} \quad (8.13)$$

If there was no diffusion of damage \mathcal{A} would simply be the thermoelastic energy of the virgin sample ($D = 0$). Separating intrinsic and thermal dissipations and assuming linear relations for the dissipative laws, renormalizing the evolution-diffusion equation for D , and writing the time increment form of the stress-strain relations, we finally obtain the following system of evolution-diffusion which summarizes the problem:

$$\left. \begin{aligned} \dot{u} &= v, \quad \dot{\varepsilon} = (\nabla u)_S, \\ \rho \dot{v} &= \operatorname{div} \sigma, \\ \dot{\bar{\sigma}} &= -\bar{\sigma} \dot{D} + (1 - D)(\mathbf{E} : \dot{\varepsilon} - \tau \dot{\Theta}), \\ \tau \dot{D} &= (W_e/W_0) + l_D^2 \nabla^2 D, \\ C \dot{\Theta} + \nabla \cdot (\Theta \mathcal{S}) &= \Phi_{\text{intr}} + \Phi_{\text{te}}, \end{aligned} \right\} \quad (8.14)$$

where we have set

$$\left. \begin{aligned} \bar{\sigma} &= \sigma/(1 - D), \quad \mathbf{E} = \frac{\partial^2 W_e}{\partial \varepsilon \partial \varepsilon} \\ \tau &= -\frac{\partial^2 W_e}{\partial \varepsilon \partial \Theta}, \quad \mathcal{S} = -\chi \nabla \Theta \end{aligned} \right\} \quad (8.15)$$

and W_0 is a reference energy, τ is a characteristic time and l_D is a characteristic length. The stress $\bar{\sigma}$ is the effective stress of damage theory (cp. [27], p. 347).

(b) For *ductile damage*, plasticity with hardening can interfere with damage. Thus we take

$$\Psi = \Psi(\boldsymbol{\varepsilon}^e, \Theta; \alpha = \{\alpha_1 = \beta, \alpha_2 = D\}, \nabla \alpha_2 = \nabla D), \quad (8.16)$$

where β is the internal variable that accounts for strain hardening (cf. Ref. [7]). Only the gradient of $\alpha_2 = D$ is accounted for (compare Paragraph C below). The thermodynamic laws of state are

$$\boldsymbol{\sigma} = \frac{\partial \Psi}{\partial \boldsymbol{\varepsilon}^e}, \quad S = -\frac{\partial \Psi}{\partial \Theta}, \quad \mathcal{B} = -\frac{\partial \Psi}{\partial \beta}, \quad \mathcal{A} = -\frac{\delta \Psi}{\delta D}. \quad (8.17)$$

The intrinsic dissipation reads

$$\Phi_{\text{intr}} = \boldsymbol{\sigma} : \dot{\boldsymbol{\varepsilon}}^p + \mathcal{B} \dot{\beta} + \mathcal{A} \dot{D}. \quad (8.18)$$

Assume that the three dissipative processes involved in (8.18) present no time scale, so that the rates $\{\dot{\boldsymbol{\varepsilon}}^p, \dot{\beta}, \dot{D}\}$ derive from a pseudo-potential of dissipation which is the indicator function of a convex set C whose boundary in $(\boldsymbol{\sigma}, \mathcal{B}, \mathcal{A})$ -space has equation $f(\boldsymbol{\sigma}, \mathcal{B}, \mathcal{A}) = 0$ (cf. Ref. [7]). Then we have the evolution equations

$$\dot{\boldsymbol{\varepsilon}}^p = \dot{\lambda} \frac{\partial f}{\partial \boldsymbol{\sigma}}, \quad \dot{\beta} = \dot{\lambda} \frac{\partial f}{\partial \mathcal{B}}, \quad \dot{D} = \dot{\lambda} \frac{\partial f}{\partial \mathcal{A}} \quad (8.19)$$

where $\dot{\lambda} \geq 0$ if $f = 0$ and $\dot{f} = 0$, and $\dot{\lambda} = 0$ if $f < 0$ or $f = 0$ and $\dot{f} < 0$. For instance, we can have (W_0 , γ and σ_0 are material coefficients)

$$f(\boldsymbol{\sigma}, \mathcal{B}, \mathcal{A}) = (1 - D)^{-1} \left[\|\boldsymbol{\sigma}\| + \mathcal{B} + \frac{W_0}{\gamma + 1} \left(\frac{\mathcal{A}}{W_0} \right)^{\gamma+1} \right] - \sigma_0, \quad (8.20)$$

from which there follows through (8.19) that

$$\beta(t) = \int_0^t (\dot{\boldsymbol{\varepsilon}}^p : \dot{\boldsymbol{\varepsilon}}^p)^{1/2} dt', \quad \dot{\beta} = \frac{\dot{\lambda}}{1 - D} \quad (8.21)$$

and

$$\dot{\boldsymbol{\varepsilon}}^p = \dot{\lambda} \frac{\bar{\boldsymbol{\sigma}}}{\|\boldsymbol{\sigma}\|}, \quad \dot{D} = \frac{\dot{\lambda}}{(1 - D)} \left(\frac{\mathcal{A}}{W_0} \right)^\gamma, \quad (8.22)$$

where $\bar{\boldsymbol{\sigma}}$ is the effective stress (see above).

Furthermore, we need to specify Ψ . For instance,

$$\Psi = \frac{1}{2} (1 - D) W_e(\boldsymbol{\varepsilon}^e) - \frac{KM}{M+1} \beta^{(1+M)/M} + \frac{1}{2} \nu (\nabla D)^2, \quad (8.23)$$

where K , M and ν are material coefficients. From the consistency condition $\dot{f} = 0$ (the only case where $\dot{\lambda} \neq 0$ possibly) it is found that

$$\dot{\lambda} = \frac{\langle \|\dot{\sigma}\| \rangle (1 - D)}{K\beta^{1/M} + \sigma_0 (\mathcal{A}/W_0)^\gamma}, \quad (8.24)$$

where $\langle \dots \rangle$ denotes the positive part of its enclosure. Then the ‘‘evolution’’ equation for damage will read

$$\dot{D} = \frac{\langle \|\dot{\sigma}\| \rangle}{K\beta^{1/M} (W_0/\mathcal{A})^\gamma + \sigma_0} \quad (8.25)$$

with

$$\mathcal{A} = W_e(\varepsilon^e) + \nu \nabla^2 D.$$

This is strongly coupled to σ and ε^p through equations (8.21) and (8.22)₁. Equation (8.25) is not, *per se*, an evolution equation as it contains no time scale. Furthermore, the dynamical behaviour described by (8.21), (8.22)₁ and (8.25) is highly singular unless unloading never occurs (for unloading, $\dot{\lambda} = 0$). But if we are only in a loading phase so that $\|\sigma\|$ varies monotonically in time, $\sigma = \|\sigma\|$ can be used as a plastic time and for $\gamma \simeq 1$, (8.25) will render the following equation:

$$\frac{dD}{d\sigma} [K\beta^{1/2} W_0 + \sigma_0 (W_e + \nu \nabla^2 D)] = W_e + \nu \nabla^2 D, \quad (8.26)$$

where (linear elasticity)

$$W_e = \frac{1}{2} \sigma : [(\nabla \mathbf{u})_S - \varepsilon^p], \quad \dot{\varepsilon}^p = \dot{\beta} \frac{\sigma}{\sigma}. \quad (8.27)$$

Such a system should be capable of describing the localization of damage ($D \leq 1$) during the monotone loading of an elastic-plastic material with hardening.

C. Localization of plastic strains and shear-band evolution

We consider the free-energy density

$$\Psi = \Psi(\varepsilon^e, \Theta, \alpha; \nabla \alpha, \nabla \nabla \alpha) \quad (8.28)$$

in elastoplasticity with hardening. Spatial gradients of the scalar internal variable, up to the second order, are considered (See Appendix). The intrinsic dissipation will read

$$\Phi_{\text{intr}} = \sigma : \dot{\varepsilon}^p + \mathcal{A} \dot{\alpha}, \quad (8.29)$$

with

$$\begin{aligned}\boldsymbol{\sigma} &= \frac{\partial \Psi}{\partial \boldsymbol{\varepsilon}^e} \\ \mathcal{A} &= - \left(\frac{\partial \Psi}{\partial \alpha} - \nabla \cdot \frac{\partial \Psi}{\partial \nabla \alpha} + \nabla \nabla : \frac{\partial \Psi}{\partial \nabla \nabla \alpha} \right).\end{aligned}\tag{8.30}$$

We envisage a rate-independent behaviour. Therefore (compare Paragraph B or Ref. [7], Chapters 2 and 3) we have the “plastic” evolution equations

$$\dot{\boldsymbol{\varepsilon}}^p = \dot{\lambda} \frac{\partial f}{\partial \boldsymbol{\sigma}}, \quad \dot{\alpha} = \dot{\lambda} \frac{\partial f}{\partial \mathcal{A}},\tag{8.31}$$

where $\dot{\lambda}$ is the plastic multiplier and f is the loading function. For the latter we take a direct generalization of the Huber-Mises yield criterion by writing (this is an experimental datum)

$$f(\boldsymbol{\sigma}, \mathcal{A}) = \|\boldsymbol{\sigma}\| + \mathcal{A} - \sigma_0.\tag{8.32}$$

We have $\dot{\lambda} \geq 0$ if $f = 0$ and $\dot{f} = 0$, and $\dot{\lambda} = 0$ if $f < 0$ or $f = 0$ and $\dot{f} < 0$. From (8.31) there follows then that

$$\dot{\boldsymbol{\varepsilon}}^p = \dot{\lambda} \frac{\boldsymbol{\sigma}}{\|\boldsymbol{\sigma}\|}, \quad \dot{\alpha} = \dot{\lambda}\tag{8.33}$$

and thus, by eliminating $\dot{\lambda}$ between these two equations

$$\alpha(t) = \int_0^t (\dot{\boldsymbol{\varepsilon}}^p : \dot{\boldsymbol{\varepsilon}}^p)^{1/2} dt' \equiv \overline{\boldsymbol{\varepsilon}}^p,\tag{8.34}$$

which is the so-called *cumulated plastic strain* (in fact already introduced as β in (8.21)). We may say that the choice of f has granted a physical meaning to the scalar internal variable α . If Ψ is further specified as

$$\Psi = W_e(\boldsymbol{\varepsilon}^e, \Theta) + \Psi_\alpha(\Theta, \alpha, \nabla \alpha, \nabla \nabla \alpha),\tag{8.35}$$

we may, for instance, obtain \mathcal{A} in a form such as

$$\mathcal{A}(\alpha = \overline{\boldsymbol{\varepsilon}}^p) = -[\kappa(\overline{\boldsymbol{\varepsilon}}^p) - c_1 \nabla^2 \overline{\boldsymbol{\varepsilon}}^p + c_2 \nabla^4 \overline{\boldsymbol{\varepsilon}}^p],\tag{8.36}$$

where c_1 and c_2 are *constant* coefficients [otherwise we should keep

$$- \nabla \cdot (c_1 \nabla \overline{\boldsymbol{\varepsilon}}^p) + \nabla^2 (c_2 \nabla^2 \overline{\boldsymbol{\varepsilon}}^p)$$

if

$$\Psi_\alpha = \Psi_1(\alpha, \Theta) + \frac{1}{2} c_1(\alpha) (\nabla \alpha)^2 + \frac{1}{2} c_2(\alpha) (\nabla^2 \alpha)^2.$$

The quantity $h = d\kappa/d\bar{\varepsilon}^p = \partial^2 \Psi_\alpha / \partial (\bar{\varepsilon}^p)^2$ is called the *hardening modulus* (cf. Ref. [7], Chapter 5). Writing the consistency condition $\dot{f} = 0$ to determine $\dot{\lambda}$ allows one to obtain the following differential equation for $\bar{\varepsilon}^p$:

$$\dot{\bar{\varepsilon}}^p = \frac{1}{h} (\boldsymbol{\sigma} : \dot{\boldsymbol{\sigma}} + c_1 \nabla^2 \dot{\bar{\varepsilon}}^p - c_2 \nabla^4 \dot{\bar{\varepsilon}}^p). \quad (8.37)$$

Equations (8.37) and (8.33)₁ and the balance laws of continuum mechanics recalled in (2.1)–(2.3) are none other than the equations that govern the *localization of plastic strains* and *shear-band evolution* in metals according to Zbib and Aifantis [28]. Here these equations are given a fully *thermodynamical basis* which justifies why only even-order gradients occur in (8.37) – this comes from the definition of $\delta/\delta\alpha$ – while the flow rule (8.32) keeps a priori a very usual form. That is, this approach does not suppose an a priori gradient-dependent flow rule – it is the free energy which is gradient dependent (like in phase transition theory à la Ginzburg-Devonshire). Furthermore, it shows that contrary to what is assumed by Zbib and Aifantis [28], it is h which is involved in (8.37) and not a more complex expression h_{eff} involving both h and $dc_1/d\bar{\varepsilon}^p$ and $dc_2/d\bar{\varepsilon}^p$. Had we considered $\bar{\varepsilon}^p$ -dependent c_1 and c_2 , expression (8.37) would have been much more involved, not only through the denominator.

9. Concluding remarks

In spite of their relative complexity and somewhat awkward formulation, the examples briefly treated in Section 8 highlight some of the essential features of our “thermodynamically admissible” approach to dissipative structures. In particular, apart from the unconventional form of the entropy flux, it appears that the remainder of the usual framework of irreversible thermodynamics is left unchanged. That is, the standard bilinear form (3.13) using only classical fluxes (e. g., $\dot{\alpha}$) is still valid. Simultaneously, the accompanying heat equation keeps its standard form (4.2). Not unlike the case of conservative structures, most of the relevant physics still is in the definition of the *force* \mathcal{A} whose vanishing value – cp. (5.2)₂ – defines static structures in a way that recalls elementary equilibrium solutions for the order parameter in Landau’s theory of phase transition.

The second important point is that evolution-diffusion equations seem, with some reason, to take over simple evolution equations and this goes in the direction indicated by Grmela [5] of a general analytical-mechanics formulation of both reversible *and* irreversible processes. In the course of this development, what made the essence of the notion of internal variables [2], seems to be alienated. That is, while internal variables are generally conceived, for the least, as *not easily measurable*, they were viewed in all cases as *non-controllable*. However, with the introduction of diffusion, the notion of accompanying *boundary conditions* enters the picture and thus control of the said “internal” variables through boundaries appears, contrary to the very notion of being “internal”. Hence it seems preferable to consider equations such as (7.5) as resulting from a simplification of

balance equations (7.4), although there remains the troublesome question of singular perturbation in the latter as ε goes to zero.

Finally, in all practical cases of interest in Section 8, the heat equation (4.2) provides, like in the elastic-plastic case (Ref. [7], Chapter 12), the theoretical support for the observation and measurement of temperature distribution due to *localized dissipative structures* by means of *infrared thermography* as internal-variable dissipation and the allied “thermoelastic” process provide the heat source in this equation. This is to be further elaborated upon.

A last remark is in order concerning *nonlocality*. The introduction of gradients to describe damage effects in Paragraph 8B corresponds to a so-called *weak nonlocality*. Other approaches [29]–[31] envisage simple *functionals over space* to describe these effects, although not in the proper thermodynamical framework. Such a framework for *strong nonlocality* can be found in Eringen and Maugin [32], following many works by A. C. Eringen, a strong advocate of nonlocal continuum mechanics. Whether strong or weak nonlocality is needed to account for localization still is a debated matter (in this regard, see Ref. [33]).

Appendix

In most of Sections 3 through 8, Ψ is assumed to depend on the first gradient of α only. The question may be raised about what happens when higher-order gradients, e. g., the second ones $\nabla\nabla\alpha$, are considered? It is a straightforward matter to show that accounting for the second gradient yields

$$\mathcal{A} = -\frac{\delta\Psi}{\delta\alpha} = A - \nabla \cdot \mathbf{B} + (\nabla \otimes \nabla) : \mathbf{C}, \quad \mathbf{C} \equiv -\frac{\partial\Psi}{\partial\nabla\nabla\alpha}, \quad (\text{A.1})$$

while (3.9) will be replaced by

$$\mathbf{k} = \frac{1}{\Theta} \{ [\mathbf{B} - \nabla \cdot \mathbf{C}] \dot{\alpha} + \mathbf{C} \cdot \nabla \dot{\alpha} \} \quad (\text{A.2})$$

as

$$\dot{\Psi} = -\hat{\mathbf{S}}\dot{\Theta} + \hat{\boldsymbol{\sigma}} : \dot{\boldsymbol{\varepsilon}}^e - \mathcal{A}\dot{\alpha} + \nabla \cdot [(\mathbf{B} - (\nabla\mathbf{C}))\dot{\alpha} + \mathbf{C} \cdot \nabla\dot{\alpha}]. \quad (\text{A.3})$$

With this it is shown that (4.1) is left unchanged as Φ_{intr} is formally left unaltered, \mathcal{A} being still the spatial functional derivative. Equation (4.2) is also formally unchanged but for Φ_{te} that will contain a second-gradient of $\dot{\alpha}$, i. e., an additional term $\mathbf{n} \cdot (\nabla \otimes \nabla)\dot{\alpha}$ within parentheses with $\mathbf{n} \equiv -\partial\mathbf{C}/\partial\Theta$.

All conclusions concerning the use and interpretation of (4.2) apply also to this more general case. The same proof will apply to gradients of order higher than two as (A.1) will generalize automatically while other terms appearing in the computation will enter the divergence term in (A.3), and this may always be accommodated in \mathbf{k} .

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