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Multiscale approach for the modeling of chemo-magneto-thermo-mechanical couplings – reversible framework

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Abstract. We focus in this paper on a multiscale modeling approach of the materials' reversible behavior involving couplings of the chemo-magneto-thermo-mechanical type. It is shown that it is possible to take into account a large variety of these coupled environments by a unified approach using the springs of the scale change and the build of an appropriate Gibbs free energy function. The approach is well suited to situations where some fields can be considered homogeneous at a relevant scale and where free deformation can be defined.

Introduction

Our time is characterized by an increasingly intensive use of actuators in extremely large fields of applications, from the largest to the smallest scales. The miniaturization of these systems adds new design constraints and requires the development of materials with controlled properties and robust models. One of the solutions being considered is the use of material exhibiting at least one strong multiphysic coupling (one of the physic being mechanics). This includes magnetostrictive materials [1], classical (SMA) or magnetic shape memory alloys (MSMA) [2], piezoelectric materials, multi-ferroic composite media, etc. One of the modeling challenges is to better describe the complex interactions observed experimentally (nonlinearity, non-monotony, irreversibly, dynamic and multiaxial effects etc ...), and to derive constitutive models with sufficient domains of validity for the considered applications. A multiscale model of a representative volume element (RVE) of such materials is proposed. It is relevant for multiaxial stress, thermal and magnetic loadings in a pseudo-reversible framework (heat exchange can be considered for instance). The objective of the paper is to recall the main hypotheses and principles required to develop this modeling. The reader will be able to find some practical examples and applications of this modeling approach in the different references added all along the text.

Gibbs free energy at the magnetic domain family scale

In the following, g denotes a *grain*. It is supposed to be composed of several phases φ (typically austenite, martensite). ϕ denotes a *variant* meaning that a phase φ can develop different variants depending on its symmetry; α denotes a *magnetic domain family* inside the variant ϕ and represents the lower scale. The scale organization is illustrated in Fig. 1, from the RVE to the domain scale, and corresponds to scales where some physical fields can be considered as homogeneous leading to some simplifications in the energy description. Indeed the selection of a phase, a variant or a domain family is based on the principle of minimum energy. A first step consists to build an energy function at the domain scale where anisotropic crystallographic phenomena are significant and some fields can be simplified.

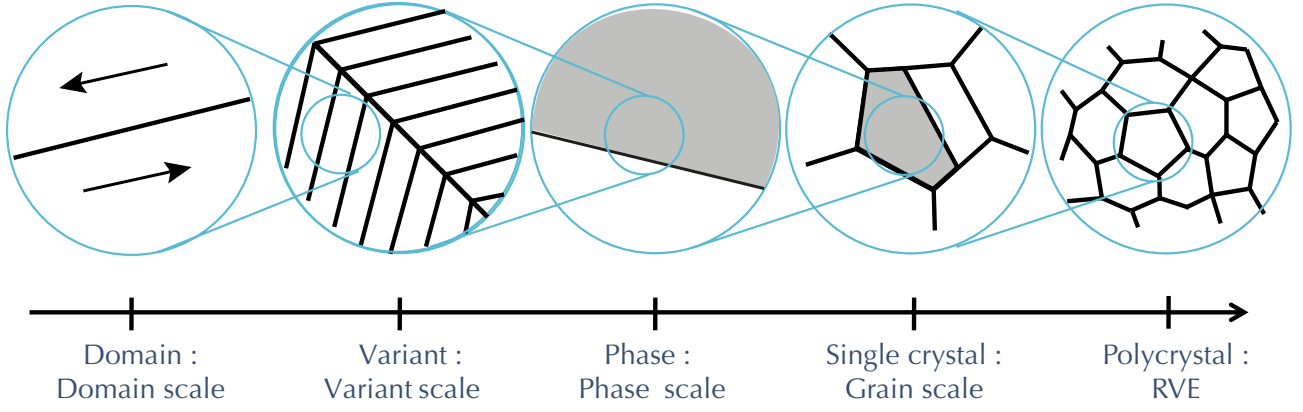


Figure 1: Detail of scales involved in the modeling approach

The first principle given at the local scale (J/m^3) express that the total energy density composed of kinetic energy density and internal energy density can only change by the action of external work and heat power. The internal energy density at the domain scale is written as: u_α . At constant velocity, kinetic energy is constant. There is a direct relationship between the variation of internal energy density and power sources:

$$du_\alpha = dh_\alpha + T_\alpha ds_\alpha + \bar{\sigma}_\alpha : d\bar{\epsilon}_\alpha + \bar{H}_\alpha \cdot d\bar{B}_\alpha \quad (1)$$

This expression is composed of:

- dh_α : chemical energy variation= bound energy variation from one phase to another;
- $\bar{\sigma}_\alpha : d\bar{\epsilon}_\alpha$: mechanical energy variation (mechanical power), function of deformation variation $d\bar{\epsilon}_\alpha$ and local stress $\bar{\sigma}_\alpha$.
- $T_\alpha ds_\alpha = dq_\alpha$: heat quantity variation (thermal power), function of entropy variation ds_α (second law of thermodynamics) and local temperature T_α ; this expression applies in a reversible framework.
- $\bar{H}_\alpha \cdot d\bar{B}_\alpha$: magnetic energy variation (magnetic power), function of induction variation $d\bar{B}_\alpha$ and magnetic field $\bar{H}_\alpha$.

The introduction of free (Helmholtz) energy $\psi_\alpha = u_\alpha - T_\alpha s_\alpha$ (Legendre transformation of heat quantity) allows the energy variation to be defined as function of temperature variation instead of entropy variation. The magnetic free enthalpy $k_\alpha = \psi_\alpha - \bar{H}_\alpha \cdot \bar{B}_\alpha$ (Legendre transformation of magnetic quantity) allows the energy variation to be defined as function of magnetic field variation instead of induction variation. The Gibbs free energy (or free enthalpy) $g_\alpha = k_\alpha - \bar{\sigma}_\alpha : \bar{\epsilon}_\alpha$ (Legendre transformation of mechanical quantity) allows finally the energy variation to be defined as function of stress variation instead of deformation variation. Variation of Gibbs free energy is given by:

$$dg_\alpha = dh_\alpha - s_\alpha dT_\alpha - \bar{\epsilon}_\alpha : d\bar{\sigma}_\alpha - \bar{B}_\alpha \cdot d\bar{H}_\alpha \quad (2)$$

Since a domain α is a part of a variant inside a phase, variation of chemical energy is null leading to [3]:

$$dg_\alpha = -s_\alpha dT_\alpha - \bar{\epsilon}_\alpha : d\bar{\sigma}_\alpha - \bar{B}_\alpha \cdot d\bar{H}_\alpha \quad (3)$$

Entropy density, strain and magnetic induction finally derive from the Gibbs free energy function following:

$$s_\alpha = -\frac{dg_\alpha}{dT_\alpha} \quad \bar{\epsilon}_\alpha = -\frac{dg_\alpha}{d\bar{\sigma}_\alpha} \quad \bar{B}_\alpha = -\frac{dg_\alpha}{d\bar{H}_\alpha} \quad (4)$$

A second order Taylor expansion of entropy leads to derive the classical expression [4]:

$$s_\alpha = s_\alpha^0 - \rho_\alpha c_\alpha^p \left[1 - \frac{T_\alpha^0}{T_\alpha} + \ln \left(\frac{T_\alpha^0}{T_\alpha} \right) \right] \quad (5)$$

where s_α^0 is the reference entropy at the reference temperature T_α^0 , ρ_α is the mass density and c_α^p the specific heat capacity. The chemical part of the Gibbs free energy is defined as:

$$g_\alpha^C = h_\alpha - T_\alpha s_\alpha = h_\alpha - T_\alpha s_\alpha^0 - \rho_\alpha c_\alpha^p \left[T_\alpha - T_\alpha^0 + T_\alpha \ln \left(\frac{T_\alpha}{T_\alpha^0} \right) \right] \quad (6)$$

Small perturbation hypothesis allows the total deformation $\bar{\epsilon}_\alpha$ to be considered as a sum of different contributions, allowing some specific couplings between mechanics and another physic to be defined. In the case of the target materials, classical elastic $\bar{\epsilon}_\alpha^e$ and thermal $\bar{\epsilon}_\alpha^T$ deformation are considered first. Free deformation $\bar{\epsilon}_\alpha^f$ associated with magnetostriction on the one hand and transformation deformation on the other hand is the third deformation source leading to:

$$\bar{\epsilon}_\alpha = \bar{\epsilon}_\alpha^e + \bar{\epsilon}_\alpha^T + \bar{\epsilon}_\alpha^f = \mathbf{C}_\alpha^{-1} : \bar{\sigma}_\alpha + \frac{\theta_\alpha}{3} (T_\alpha - T_\alpha^0) \bar{\mathbf{1}} + \bar{\epsilon}_\alpha^f \quad (7)$$

$\mathbf{C}_\alpha$ is the 4th order local stiffness tensor, θ_α is the volumetric dilatation coefficient and $\bar{\mathbf{1}}$ the second order identity tensor. At this step, it is interesting to observe that the free deformation can be considered as stress independent at the local scale, allowing a simple derivation of the mechanical part of Gibbs free energy. Complementary hypotheses can help to simplify the last term.

$$g_\alpha^M = -\frac{1}{2} \bar{\sigma}_\alpha : \mathbf{C}_\alpha^{-1} : \bar{\sigma}_\alpha - \frac{\theta_\alpha}{3} (T_\alpha - T_\alpha^0) \text{tr}(\bar{\sigma}_\alpha) - \int_0^{\bar{\sigma}_\alpha} \bar{\epsilon}_\alpha^f : d\bar{\sigma}_\alpha \quad (8)$$

The magnetic induction is resulting from the local magnetic field and local magnetization of the material $\bar{\mathbf{M}}_\alpha$ following:

$$\bar{\mathbf{B}}_\alpha = \mu_0 (\bar{\mathbf{M}}_\alpha + \bar{\mathbf{H}}_\alpha) \quad (9)$$

where μ_0 is the vacuum magnetic permeability. The magnetic contribution to the internal energy variation can be expressed as:

$$du_\alpha = \mu_0 \bar{\mathbf{H}}_\alpha \cdot d\bar{\mathbf{M}}_\alpha + \mu_0 \bar{\mathbf{H}}_\alpha \cdot d\bar{\mathbf{H}}_\alpha \quad (10)$$

At the local scale (magnetic domain) the magnetization variations can be very high (much higher than magnetic field) so that the second term is often neglected [5] and the Helmholtz magnetic free energy is expressed as function of magnetization. A Taylor expansion is used (3rd order development), taking into account the fact that the magnetic behavior is an odd function. The Helmholtz magnetic free energy is consequently an even function of magnetization [3].

$$\psi_\alpha^\mu = \bar{\mathbf{M}}_\alpha \cdot \bar{\mathbf{P}}_\alpha^0 \cdot \bar{\mathbf{M}}_\alpha + \bar{\mathbf{M}}_\alpha \bar{\mathbf{M}}_\alpha : \mathbf{P}_\alpha^1 : \bar{\mathbf{M}}_\alpha \bar{\mathbf{M}}_\alpha + \bar{\mathbf{M}}_\alpha \cdot \bar{\mathbf{M}}_\alpha \bar{\mathbf{M}}_\alpha : \bar{\mathbf{P}}_\alpha^2 : \bar{\mathbf{M}}_\alpha \bar{\mathbf{M}}_\alpha \cdot \bar{\mathbf{M}}_\alpha \quad (11)$$

Using a second order $\bar{\mathbf{P}}_\alpha^0$, 4th order $\mathbf{P}_\alpha^1$ and 6th order $\bar{\mathbf{P}}_\alpha^2$ tensors as material dependent whose expressions are strongly correlated to material symmetries. The associated expression of Gibbs magnetic free energy (after Legendre transformation) is given by:

$$g_\alpha^\mu = \bar{\mathbf{M}}_\alpha \cdot \bar{\mathbf{P}}_\alpha^0 \cdot \bar{\mathbf{M}}_\alpha + \bar{\mathbf{M}}_\alpha \bar{\mathbf{M}}_\alpha : \mathbf{P}_\alpha^1 : \bar{\mathbf{M}}_\alpha \bar{\mathbf{M}}_\alpha + \bar{\mathbf{M}}_\alpha \cdot \bar{\mathbf{M}}_\alpha \bar{\mathbf{M}}_\alpha : \bar{\mathbf{P}}_\alpha^2 : \bar{\mathbf{M}}_\alpha \bar{\mathbf{M}}_\alpha \cdot \bar{\mathbf{M}}_\alpha - \mu_0 \bar{\mathbf{H}}_\alpha \cdot \bar{\mathbf{M}}_\alpha \quad (12)$$

The sum of g_α^T , g_α^M and g_α^μ gives the total Gibbs free energy of medium that must be minimum with respect to temperature, stress and magnetic field at the equilibrium.

Usual simplifications of Gibbs free energy

At this step, it is interesting to observe that the magnetization at the domain scale has a constant norm equal to the saturation magnetization M_s [6]. The magnetization vector can be defined using the direction cosines γ_α^i in the local frame (usually crystal frame) as:

$$\bar{\mathbf{M}}_\alpha = M_s \gamma_\alpha^i \cdot \bar{\mathbf{e}}^i = M_s (\gamma_\alpha^1 \cdot \bar{\mathbf{e}}^1 + \gamma_\alpha^2 \cdot \bar{\mathbf{e}}^2 + \gamma_\alpha^3 \cdot \bar{\mathbf{e}}^3) \quad (13)$$

leading to the so-called dyadic expression of the second order orientation tensor $\bar{\bar{\mathbf{R}}}$:

$$\bar{M}_\alpha \bar{M}_\alpha = M_s^2 \begin{bmatrix} \gamma_\alpha^1 \gamma_\alpha^1 & \gamma_\alpha^1 \gamma_\alpha^2 & \gamma_\alpha^1 \gamma_\alpha^3 \\ \gamma_\alpha^1 \gamma_\alpha^2 & \gamma_\alpha^2 \gamma_\alpha^2 & \gamma_\alpha^2 \gamma_\alpha^3 \\ \gamma_\alpha^1 \gamma_\alpha^3 & \gamma_\alpha^2 \gamma_\alpha^3 & \gamma_\alpha^3 \gamma_\alpha^3 \end{bmatrix} = M_s^2 \bar{R} \quad (14)$$

Using the cubic symmetry and the identity $\gamma_\alpha^1 \gamma_\alpha^1 + \gamma_\alpha^2 \gamma_\alpha^2 + \gamma_\alpha^3 \gamma_\alpha^3 = 1$, the magnetic part of Gibbs free energy simplifies into [3,6]:

$$g_\alpha^\mu = K^0 + K^1(\gamma_\alpha^1 \gamma_\alpha^1 \gamma_\alpha^2 \gamma_\alpha^2 + \gamma_\alpha^1 \gamma_\alpha^1 \gamma_\alpha^3 \gamma_\alpha^3 + \gamma_\alpha^2 \gamma_\alpha^2 \gamma_\alpha^3 \gamma_\alpha^3) + K^2(\gamma_\alpha^1 \gamma_\alpha^1 \gamma_\alpha^2 \gamma_\alpha^2 \gamma_\alpha^3 \gamma_\alpha^3) - \mu_0 \bar{H}_\alpha \cdot \bar{M}_\alpha \quad (15)$$

where K^0 , K^1 and K^2 are the so-called magnetocrystalline constants (defining the magnetocrystalline energy density) and the last term is usually called Zeemann energy density (if $\bar{H}_\alpha$ is considered homogeneous [6]). In case of uniaxial anisotropy of axis $\bar{u}$ (so that $\bar{M}_\alpha \cdot \bar{u} = M_s \gamma_\alpha^u$), a quadratic magnetocrystalline energy is usually enough to express the magnetic part of Gibbs free energy:

$$g_\alpha^\mu = K^0(1 - \gamma_\alpha^u \gamma_\alpha^u) - \mu_0 \bar{H}_\alpha \cdot \bar{M}_\alpha \quad (16)$$

The magnetic part of Gibbs free energy is often used to express the magneto-elastic coupling energy term $g_\alpha^{M\mu}$ considered as linearly dependent to stress and as a quadratic function of magnetization, without excluding some complementary higher order terms [3].

$$g_\alpha^{M\mu} = -\bar{M}_\alpha \bar{M}_\alpha : \bar{\bar{E}}_\alpha : \bar{\bar{\sigma}}_\alpha = - \int_0^{\bar{\bar{\sigma}}_\alpha} \bar{\bar{E}}_\alpha^f : d\bar{\bar{\sigma}}_\alpha = -\bar{\bar{E}}_\alpha^f : \bar{\bar{\sigma}}_\alpha \quad (17)$$

$\bar{\bar{E}}_\alpha$ is the piezomagnetic 4th order tensor defined by 3 constants in the cubic crystallographic framework. Looking at equation (8), it is possible to identify $\bar{M}_\alpha \bar{M}_\alpha : \bar{\bar{E}}_\alpha$ as the so-called magnetostriction 2nd order tensor $\bar{\bar{\epsilon}}_\alpha^\mu$ reducing to 2 independent constants in case of incompressible deformation [6,7]:

$$\bar{\bar{\epsilon}}_\alpha^\mu = \frac{3}{2} \begin{bmatrix} \lambda_{100}(\gamma_\alpha^1 \gamma_\alpha^1 - \frac{1}{3}) & \lambda_{111} \gamma_\alpha^1 \gamma_\alpha^2 & \lambda_{111} \gamma_\alpha^1 \gamma_\alpha^3 \\ \lambda_{111} \gamma_\alpha^1 \gamma_\alpha^2 & \lambda_{100}(\gamma_\alpha^2 \gamma_\alpha^2 - \frac{1}{3}) & \lambda_{111} \gamma_\alpha^2 \gamma_\alpha^3 \\ \lambda_{111} \gamma_\alpha^1 \gamma_\alpha^3 & \lambda_{111} \gamma_\alpha^2 \gamma_\alpha^3 & \lambda_{100}(\gamma_\alpha^3 \gamma_\alpha^3 - \frac{1}{3}) \end{bmatrix} \quad (18)$$

where λ_{100} and λ_{111} are the magnetostriction constants that correspond to the deformation of a perfect single crystal along $\langle 100 \rangle$ and $\langle 111 \rangle$ axis respectively at the magnetic saturation.

Simplifications can concern the loading itself and some homogeneity hypotheses that can be applied:

- homogeneous stress at the grain scale: $\bar{\bar{\sigma}}_\alpha = \bar{\bar{\sigma}}_\phi = \bar{\bar{\sigma}}_g$
- homogeneous temperature over the volume: $T_\alpha = T_\phi = T_g = T$
- homogeneous magnetic field at the grain scale: $\bar{H}_\alpha = \bar{H}_\phi = \bar{H}_g$

Only stress and magnetic field have usually to be localized (to be defined as function of loading at the upper scale).

Constitutive law, localization and homogenization procedures

Gibbs energy is defined at the domain scale, inside a variant, inside a phase, inside a grain, inside the representative volume element. A stochastic approach is relevant for a description “at equilibrium”. The internal variables of the problem are the volume fraction of domain families denoted as f_α and the orientation of the magnetization inside each domain family (giving the direction cosines γ_α^i). The total number of internal variables strongly depends on the number of phases (n), variants (m) and number of domain families (p) inside each variant. Following [8] we use a probabilistic Boltzmann like function to calculate the volume fraction and implement an

energy minimization for the determination of the magnetization direction. Equation (19) gives the simplest expression of volume fraction f_α , where A_s is an adjusting parameter.

$$f_\alpha = \frac{\exp(-A_s g_\alpha)}{\sum_\varphi \sum_\phi \sum_\alpha \exp(-A_s g_\alpha)} \quad (19)$$

This equation is complemented by the minimization operation reported in equation (20).

$$\gamma_\alpha^i = \min(g_\alpha) \quad (20)$$

This formulation is well suited for a single variant and monophasic ferromagnetic material [8-13] (ferromagnetic ferrite or austenite). A more complex formulation has been proposed recently. The new volume fraction is using up to three different parameters (A_s, B_s, C_s) driving three different kinetics (domain, variant and phase kinetics).

$$f_\alpha = \frac{\sum_\phi \sum_\alpha \exp(-C_s g_\alpha)}{\sum_\varphi \sum_\phi \sum_\alpha \exp(-C_s g_\alpha)} \frac{\sum_\alpha \exp(-B_s g_\alpha)}{\sum_\phi \sum_\alpha \exp(-B_s g_\alpha)} \frac{\exp(-A_s g_\alpha)}{\sum_\alpha \exp(-A_s g_\alpha)} \quad (21)$$

Moreover the volume fraction of a variant ϕ and a phase φ are simply given by:

$$f_\phi = \sum_\alpha f_\alpha = \frac{\sum_\alpha \sum_\alpha \exp(-C_s g_\alpha)}{\sum_\varphi \sum_\phi \sum_\alpha \exp(-C_s g_\alpha)} \frac{\sum_\alpha \exp(-B_s g_\alpha)}{\sum_\phi \sum_\alpha \exp(-B_s g_\alpha)} \quad (22)$$

and

$$f_\varphi = \sum_\phi f_\phi = \frac{\sum_\phi \sum_\alpha \exp(-C_s g_\alpha)}{\sum_\varphi \sum_\phi \sum_\alpha \exp(-C_s g_\alpha)} \quad (23)$$

It can be noticed that, for ($A_s = B_s = C_s$), equation (19) is recovered.

Different materials can be considered: ferromagnetic materials without phase transformation (electric steels, iron, nickel – equation (19)), paramagnetic materials with phase transformation and a single variant (austenite/ferrite allotropic transformation – equation (23) [14]), or with multiple variants (SMA – equation (22) [15]), ferromagnetic materials with phase transformation and multivariants (MSMA – equation (21) [16]), or magneto-caloric materials where the phase transformation temperature coincides with the Curie temperature (equation (21) [17]).

Some localization rules are required to define the stress and magnetic fields at the grain scale (they are supposed homogeneous over the variants and magnetic domains) [8]. An Eshelby approach is applied for the determination of both fields. The stress at the grain scale $\bar{\sigma}_g$ is function of the macroscopic stress $\bar{\sigma}$, a stress concentration tensor $\mathbf{B}_g$, an accommodation stiffness tensor $\mathbf{C}_g^{acc}$ and the contrast between macroscopic $\bar{\epsilon}^f$ and microscopic $\bar{\epsilon}_g^f$ free strain tensor:

$$\bar{\sigma}_g = \mathbf{B}_g : \bar{\sigma} + \mathbf{C}_g^{acc} : (\bar{\epsilon}^f - \bar{\epsilon}_g^f) \quad (24)$$

This relation is complemented by classical averaging operations:

$$\bar{\sigma} = \langle \bar{\sigma}_g \rangle \quad \bar{\epsilon}^f = \langle {}^t \mathbf{B}_g : \bar{\epsilon}_g^f \rangle \quad (25)$$

The magnetic field at the grain scale $\bar{H}_g$ is function of the macroscopic magnetic field $\bar{H}$, an average secant susceptibility $\chi = \|\bar{M}/\bar{H}\|$ and the contrast between macroscopic to microscopic magnetization:

$$\bar{H}_g = \bar{H} + \frac{1}{3+2\chi} (\bar{M} - \bar{M}_g) \quad (26)$$

This relation is complemented by classical averaging operations:

$$\bar{H} = \langle \bar{H}_g \rangle \quad \bar{M} = \langle \bar{M}_g \rangle \quad (27)$$

Conclusion

The principles of multiscale modeling with thermodynamic foundations were presented. The stochastic constitutive law is well adapted to the existence of fine microstructures for which it is possible to consider some fields as homogeneous. It is very well adapted to the description of many couplings involving a mechanical free deformation. The localization procedures from the RVE to the grain scale makes it possible to better account for demagnetizing fields and residual stresses effects [13]. Such a modeling requires physical parameters usually coming from literature and adjusting parameters identified using a differential scanning calorimetry measurement for the phase kinetic [16], a mechanical measurement for variant kinetic [15], and an anhysteretic magnetic measurement for the magnetic domain kinetic [8]. Validation is usually obtained from coupled experiments (involving the concomitant variation of several fields).

This approach must usually be complemented by a numerical modeling, leading *a minima* to the resolution of the heat equation [18]. Indeed chemical enthalpy variation acts as a heat source in the heat equation, making each phase transformation problem a strongly coupled themomechanical situation. Another important issue is the introduction of irreversible phenomena associated with germination and friction effects.

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