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# Stress-induced phase transformations in Ni–Mn–Ga alloys: experiments and modelling

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Ni–Mn–Ga ferromagnetic shape memory alloys (FSMA) appear to be very promising active materials because they exhibit giant and fast strains. In this paper, a study of the martensitic transformation is proposed. The pseudoelastic behaviour under pure compression of a  $\text{Ni}_{49.7}\text{Mn}_{27.7}\text{Ga}_{22.9}$  single crystal previously measured for five different external temperatures (more than  $M_s^0$ ) by Pons et al. [Proceedings of ICOMAT 02, J. Phys. IV, in press]—compression axes of the initial cubic austenitic phase are  $\langle 001 \rangle$  and  $\langle 110 \rangle$ —is compared to a model for ferromagnetic shape memory alloy pseudoelastic behaviour. The identification of model parameters permits to fit well the experimental results. Moreover a prediction of initiation surface of phase transformation (austenite  $\rightarrow$  martensite) is proposed for Ni–Mn–Ga polycrystals under biaxial mechanical loadings.

*Keywords:* Phase transformations; Magnetic shape memory alloys; Experiments; Modelling; Pseudoelasticity

## 1. Introduction

Ferromagnetic shape memory alloys (FSMA) as Ni–Mn–Ga can be the seat of phase transformation (p.t.) between austenite and martensite, and/or rearrangement process of martensite platelets as in classical SMA (Ni–Ti or copper based alloys). FSMA can be actuated, or deformed, by stress field and by temperature changes as all SMA, but also, by magnetic field.

In single crystal, rearrangement process under magnetic field and/or mechanical stresses is well investigated in FSMA [1,2]. Less attention is paid to the phase transformation process between the austenitic mother phase A and the product phase called martensite M [2,3]. Pons et al. [3] obtained different isothermal stress–strain curves for phase transformation on Ni–Mn–Ga single crystal compressed along  $\langle 001 \rangle$  and  $\langle 110 \rangle$  axes. Some models have been developed for the prediction of rearrangement or phase transformation for FSMA [4–6].

The aim of the present paper is to build a model able to represent the experimental observations of Pons et al. [3] and others [7]. In the frame of the thermodynamics of irreversible process, the model can be considered as phenomenological at the macroscopic scale and the volume fraction of martensite is chosen as an internal variable. In the first part, the model equations are rapidly stated. In the second part, a tentative of modelling pseudoelastic behaviour of Ni–Mn–Ga single crystal is done. In the third part, the determination of the yield surfaces of phase transformation is extended to the polycrystals.

## 2. Model for ferromagnetic shape memory alloy pseudoelasticity

Let's consider a piece of single crystal (representative volume element RVE) which is in the austenitic state at the reference state (stress-free state, temperature  $T = T_0$ ).

Under mechanical loading, the mother phase A is transformed into a martensite variant  $M_i$  (the best oriented towards the loading axis). One has to note that the magnetic field action on the phase transformation is not included in this paper because it is nowadays an opened problem. For

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instance, which are the magnetic domains and the easy axis of magnetisation in the austenitic phase? Which are their evolutions associated with the phase transformation?

Consider the following form of the specific free energy  $\Phi$  of the two phases mixture [8]:

$$\Phi = \Phi(T, \underline{\varepsilon}, z) = (1 - z)\Phi_{(1)} + z\Phi_{(2)} + z(1 - z)\Phi_{\text{it}}(T) \quad (1)$$

where  $T$ ,  $\underline{\varepsilon}$ ,  $z$  and  $\Phi_{(\alpha)}$  denotes respectively temperature, strain tensor, volume fraction of martensite and free energy of  $\alpha$  phase with  $\Phi_{\text{it}}(T) = u_0 - Ts_0$  ( $u_0$  and  $s_0$  are constants).

$$\begin{aligned} \alpha = 1, & \quad (1 - z) \text{ volume fraction of A} \\ \alpha = 2, & \quad z \text{ volume fraction of M} \end{aligned} \quad (2)$$

The following assumptions are made:

- (i) The heat capacity  $C_v$ , the elastic modulus  $\underline{L}$ , and the mass density  $\rho$  are supposed to be the same for the two different phases. So, the expression of the free energy of  $\alpha$  phase is:

$$\begin{aligned} \Phi_{(\alpha)}(T, \underline{\varepsilon}_{(\alpha)}, z) = & u_{0(\alpha)}^* - Ts_{0(\alpha)}^* \\ & + \frac{1}{2\rho}(\underline{\varepsilon}_{(\alpha)} - \underline{\varepsilon}_{(\alpha)}^{\text{tr}}) : \underline{L} : (\underline{\varepsilon}_{(\alpha)} - \underline{\varepsilon}_{(\alpha)}^{\text{tr}}) \\ & + C_v \left[ (T - T_0) - \ln\left(\frac{T}{T_0}\right) \right] \end{aligned} \quad (3)$$

where  $u_{0(\alpha)}^*$ ,  $s_{0(\alpha)}^*$ ,  $\underline{\varepsilon}_{(\alpha)}$  and  $\underline{\varepsilon}_{(\alpha)}^{\text{tr}}$  are respectively the specific internal energy, the specific internal entropy, the total strain and the transformation strain of  $\alpha$  phase.

- (ii) The stress state—represented by stress tensor  $\underline{\sigma}$ —is supposed to be uniform or homogeneous in the material. It means that:

$$\begin{aligned} \underline{\sigma} = \rho \frac{\partial \phi_{(1)}}{\partial \underline{\varepsilon}_{(1)}} = & \underline{L} : (\underline{\varepsilon}_{(1)} - \underline{\varepsilon}_{(1)}^{\text{tr}}) \\ = \rho \frac{\partial \phi_{(2)}}{\partial \underline{\varepsilon}_{(2)}} = & \underline{L} : (\underline{\varepsilon}_{(2)} - \underline{\varepsilon}_{(2)}^{\text{tr}}) \end{aligned} \quad (4)$$

By introducing elastic strain  $\underline{\varepsilon}_{(\alpha)}^{\text{el}}$  in  $\alpha$  phase as:

$$\underline{\varepsilon}_{(\alpha)}^{\text{el}} = \underline{\varepsilon}_{(\alpha)} - \underline{\varepsilon}_{(\alpha)}^{\text{tr}}, \quad (5)$$

one obtains uniform elastic strain state—represented by strain tensor  $\underline{\varepsilon}^{\text{el}}$ —in the material as:

$$\underline{\varepsilon}_{(1)}^{\text{el}} = \underline{\varepsilon}_{(2)}^{\text{el}} = \underline{\varepsilon}^{\text{el}} = \underline{L}^{-1} : \underline{\sigma} \quad (6)$$

- (iii) The phase transformation strains associated with each  $\alpha$  phase are:

$$\underline{\varepsilon}_{(1)}^{\text{tr}} = 0, \quad \underline{\varepsilon}_{(2)}^{\text{tr}} = \underline{k} \quad (7)$$

The macroscopic strain  $\underline{\varepsilon}$ —average strain in the representative elementary volume—is also defined as the following mixing rule of strains of  $\alpha$  phase:

$$\underline{\varepsilon} = (1 - z)\underline{\varepsilon}_{(1)} + z\underline{\varepsilon}_{(2)} \quad (8)$$

As consequence (from the following equations), it comes that the macroscopic transformation strain  $\underline{\varepsilon}^{\text{tr}}$  is:

$$\underline{\varepsilon}^{\text{tr}} = \underline{\varepsilon} - \underline{\varepsilon}^{\text{el}} = (1 - z)\underline{\varepsilon}_{(1)}^{\text{tr}} + z\underline{\varepsilon}_{(2)}^{\text{tr}} = z\underline{k} \quad (9)$$

And, the final expression of the free energy becomes:

$$\begin{aligned} \Phi(T, \underline{\varepsilon}, z) = & u_{0(1)}^* - Ts_{0(1)}^* - z\pi_0^f(T) \\ & + \frac{1}{2\rho}(\underline{\varepsilon} - z\underline{k}) : \underline{L} : (\underline{\varepsilon} - z\underline{k}) \\ & + C_v \left[ (T - T_0) - \ln\left(\frac{T}{T_0}\right) \right] + z(1 - z)\Phi_{\text{it}}(T) \end{aligned} \quad (10)$$

where  $\pi_0^f(T) = \Delta u^* - T\Delta s^*$ ,

$$\Delta u^* = u_{0(1)}^* - u_{0(2)}^*, \quad \Delta s^* = s_{0(1)}^* - s_{0(2)}^* \quad (11)$$

Hence,

$$\underline{\sigma} = \rho \frac{\partial \phi}{\partial \underline{\varepsilon}} = \underline{L} : (\underline{\varepsilon} - z\underline{k}) \quad \text{and} \quad s = -\frac{\partial \phi}{\partial T} \quad (12)$$

$$\pi^f(T, \underline{\sigma}, z) = \frac{\partial \phi}{\partial z} = \pi_0^f(T) + \frac{\underline{\sigma} : \underline{k}}{\rho} - (1 - 2z)\Phi_{\text{it}}(T) \quad (13)$$

In a natural way, the form of the energy dissipated during time  $dt$  is given by Clausius–Duhem inequality as:

$$D dt = \pi^f dz \geq 0 \quad (14)$$

Thus, this inequality precludes the parent-martensite transformation at state where  $\pi^f < 0$  and prevents the reverse transformation when  $\pi^f > 0$ . Note that  $\pi^f = 0$  implies the equilibrium conditions. To specify the kinetic equations, we presume that there exists two functions  $\Psi_{(\alpha)}(\pi^f, z)$  ( $\alpha = 1, 2$ ) such that an active process of parent phase decomposition ( $dz > 0$ —the forward transformation) can proceed only when  $\Psi_{(1)} = \text{constant}$  (or  $d\Psi_{(1)} = 0$ ) and an active process of martensite decomposition ( $dz < 0$ —the reverse transformation) can proceed only when  $\Psi_{(2)} = \text{constant}$  (or  $d\Psi_{(2)} = 0$ ).

$$\Psi_{(1)} = \pi^f - k_{(1)}(z), \quad \Psi_{(2)} = -\pi^f + k_{(2)}(z) \quad (15)$$

with  $k_{(1)}(z) = a_1 \ln(1 - z)$ ,  $k_{(2)}(z) = a_2 \ln(z)$ . The consistency equations  $d\Psi_{(1)} = 0$  (or  $d\Psi_{(2)} = 0$ ) give respectively the kinetic law of forward and reverse phase transformation of internal variable  $z$ .

### 3. Ni–Mn–Ga single crystal FSMA modelling

From Eq. (15), the forward transformation is initiated when:

$$\pi^f(\underline{\sigma} = \underline{\sigma}^{\text{AM}}, z = 0, T) = 0 \quad (16)$$

For a mechanical test with constant external temperature it means that:

$$\left(\frac{\underline{\sigma} : \underline{k}}{\rho}\right)^{\text{AM}} = K(T) \quad (17)$$

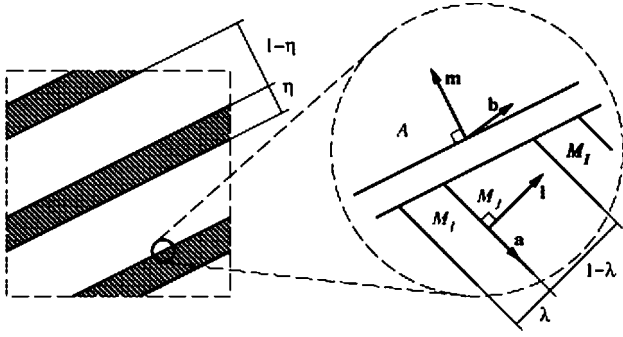


Fig. 1. Schematic representation of an austenite-twinned martensite microstructure.

Concerning the Ni–Mn–Ga, the austenite is transformed into twinned martensite schematically represented in Fig. 1. The compatibility condition at the interface (habit plane) between austenite and a homogenized twinned martensite plates (5M) reads [9,10]:

$$\underline{k} = \lambda \underline{\varepsilon}_i^{\text{tr}} + (1 - \lambda) \underline{\varepsilon}_j^{\text{tr}} = \frac{1}{2} (\vec{b} \otimes \vec{m} + \vec{m} \otimes \vec{b}) \quad (18)$$

with habit plane normal  $\vec{m}$  and shape strain vector  $\vec{b}$ .

Under a given stress condition  $\sigma^0$ , among the  $n$  possible couples of variants, the couple  $(i, j)$  presenting the highest factor  $K$  is selected and  $\underline{k}$  is identified. One can verify which habit plane variant (HPV) is involved as a function of the orientation.

In the one dimensional situation corresponding to a single compressive test Eq. (18) is reduced to:

$$\left( \frac{\sigma^{\text{AM}} \gamma}{\rho} \right) = K(T) \quad (19)$$

where  $\sigma^{\text{AM}}$  and  $\gamma$ , which depend on temperature and on single crystal orientation, can be measured.

The thermodynamical parameters  $\Delta u^*$ ,  $\Delta s^*$ ,  $u_0$ ,  $s_0$  are identified from each different isothermal compressive curve using the following equations: expression of stresses  $\sigma^{\text{AM}}$  and  $\sigma^{\text{MA}}$  when phase transformation starts and the Clausius Clapeyron (Eqs. (22) and (23)):

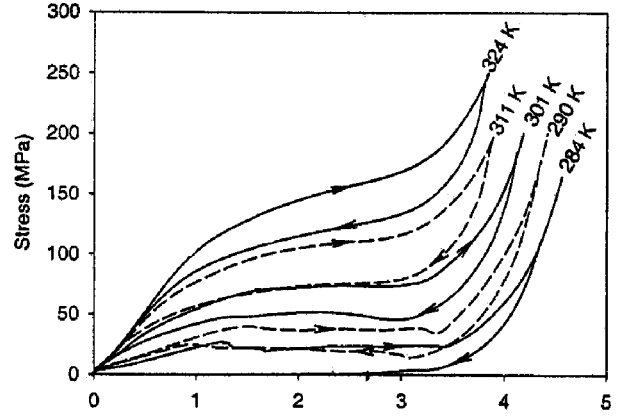
$$\pi^f(\underline{\sigma} = \underline{\sigma}^{\text{AM}}, z = 0, T) = 0 \Leftrightarrow \sigma^{\text{AM}} = \frac{\rho}{\gamma} (-\pi_0^f(T) + \Phi_{it}) \quad (20)$$

$$\pi^f(\underline{\sigma} = \underline{\sigma}^{\text{MA}}, z = 1, T) = 0 \Leftrightarrow \sigma^{\text{MA}} = \frac{\rho}{\gamma} (-\pi_0^f(T) - \Phi_{it}) \quad (21)$$

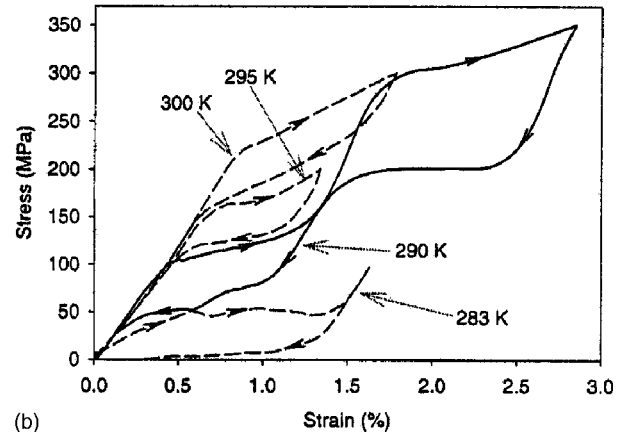
$$\frac{d\sigma^{\text{AM}}}{dT} = \frac{\rho}{\gamma} (\Delta s^* - s_0) \quad (22)$$

$$\frac{d\sigma^{\text{MA}}}{dT} = \frac{\rho}{\gamma} (\Delta s^* + s_0) \quad (23)$$

The experimental curves,  $\sigma$  versus  $\varepsilon$ , for forward and reverse phase transformation permit to determine parameters



(a)



(b)

Fig. 2. Stress–strain single compression curves by Pons et al. [3]: (a) compression in (001) direction; (b) compression in (110) direction.

of kinetic law  $a_1$  and  $a_2$  and hence the curve fitting for two single crystal orientations  $\langle 001 \rangle$  and  $\langle 110 \rangle$  [3] (Figs. 2 and 3).

Moreover, for the  $\langle 110 \rangle$  orientation, the Ni–Mn–Ga single crystal exhibits two consecutive phase transformations: one from austenite to 5M martensite which was the subject of the precedent modelling and another one from the 5M martensite to another 7M martensite. For this second hysteresis loop, the end of the first loop is considered as the origin of the second one and new thermodynamic and kinetic parameters are identified.

### 3.1. Modelling of experimental curves

From the DSC curves, the following values of transformation temperatures and latent heat ( $q$ ) from A  $\rightarrow$  5M are obtained:  $M_{s0} = 274$  K,  $A_{s0} = 284$  K and  $q = -3.6$  J/g. Magnetic permeability measurements permit to determine the Curie temperature  $T_c = 383$  K. The lattice parameters  $a = 0.594$  nm and  $c = 0.562$  nm for five-layered tetragonal phase (5M) and  $a_0 = 0.582$  nm are considered from reference [11]. As expected for SMA, the curve  $\sigma$  versus  $\varepsilon$  for compression along  $\langle 100 \rangle$  consists on one plateau

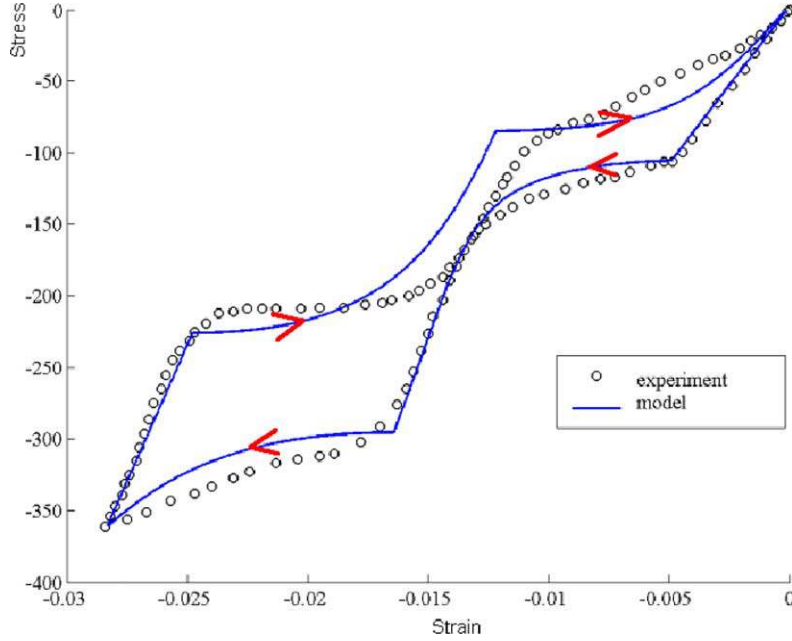


Fig. 3. Stress–strain curves: simulations with RL model and experiments from Pons et al. [3].

corresponding to the formation of five-layered martensite (5M). In contrast, the curve  $\sigma$  versus  $\varepsilon$  for high enough compression strain along (1 1 0) axis consists on two stages corresponding to the sequential formation of five-layered (5M) and seven-layered (7M) orthorhombic martensite, respectively [12]. The lattice parameters  $a = 0.619$  nm,  $b = 0.580$  nm and  $c = 0.553$  nm for the seven-layered orthorhombic phase are considered from reference [13]. The Clausius Clapeyron diagram, i.e.  $\sigma^{\text{AM}}$  versus  $T$ , delivers  $d\sigma^{\text{AM}}/dT = -2.5$  and  $-9.4$  MPa/K in  $\langle 001 \rangle$  and  $\langle 110 \rangle$  axes respectively for  $A \rightarrow 5M$  [3]. One has to note that these values are very useful for modelling (Eq. (22)).

#### 4. Prediction of initiation surfaces of phase transformation (A $\rightarrow$ M)

Let's have a biaxial mechanical loading in Ni–Mn–Ga polycrystals:

$$\underline{\sigma} = \sigma_1 \vec{e}_1 \otimes \vec{e}_1 + \sigma_2 \vec{e}_2 \otimes \vec{e}_2 \quad (24)$$

The predicted phase transformation surface must be at least convex in the stress space ( $\sigma_1, \sigma_2$ ); it must also account for the general asymmetry between tension and compression observed in SMA and at last must fit the experimental yield points obtained for proportional biaxial loading in tension, or (compression)–torsion, or bi-compression.

Classically the first variant appears when a thermodynamical force associated to the phase transformation is equal to zero. A simple choice can be (see Eqs. (16)–(18)):

$$\left( \frac{\underline{\sigma} : \underline{\varepsilon}^t}{\rho} \right) - k(T) = 0 \quad (25)$$

where  $\underline{\sigma}$  and  $\underline{\varepsilon}^t$  are naturally written in the geometrical frame of the sample.  $\underline{\sigma} : \underline{\varepsilon}^t$  represent the mechanical energy required to transform completely a unit volume of austenite into martensite. If  $\underline{R}$  is the rotation matrix from the austenite cell frame to the geometrical sample one, then

$$\underline{\varepsilon}^t = {}^T \underline{R} \underline{k} \underline{R} \quad (26)$$

The procedure used to calculate the yield surface of polycrystal is purely phenomenological and is adapted from a paper by Huang [14]:

- (i) A polycrystalline material is represented by  $n$  grains (here 1000 grains are chosen) defined by their crystallographic orientation. Obviously, an isotropic texture, which represents a random distribution of the grain crystallographic lattice orientation in the space of the Euler angles, is chosen. Interactions between the grains are not taken into account. As the yield surface represents the apparition of the first activated martensite platelets, interaction stresses are not playing an important part yet.
- (ii) For a given grain  $k$ ,  $m$  variants could be created. Under a given stress condition  $\underline{\sigma}^0$ , the variant presenting the highest factor  $K$  (from Eq. (25)) is selected. It should be noted that we use here a Sachs type model. Indeed, this variant is the one with the largest transformation strain along the stress direction, which is equivalent to the lowest transformation stress; it is similar to a Schmid law. For the polycrystal composed by  $n$  grains, a set of  $n$  factors  $K_k^{\text{max}}$  is determined by this method.
- (iii) A new set of  $K_k^{\text{max}}$  can be calculated under a different stress condition.  $K_{\text{tension},k}^{\text{max}}$  stands for the results

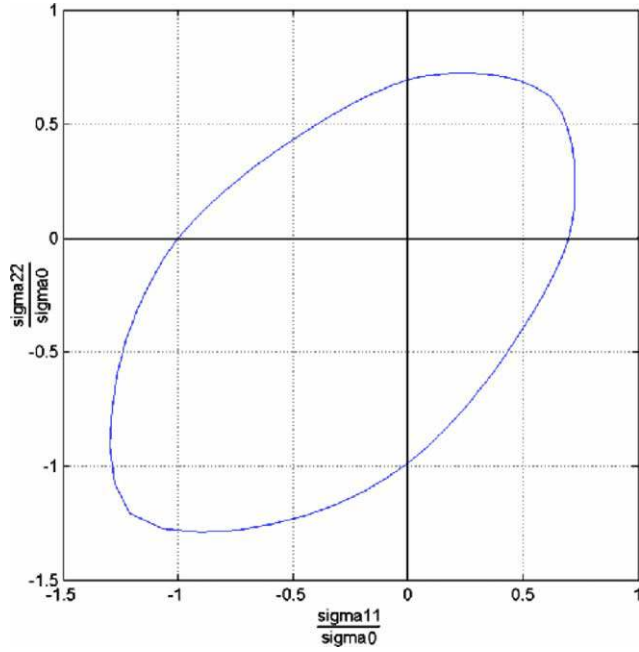


Fig. 4. Initiation surface of a A → 5M phase transformation.

under uniaxial tension. The ratio called  $\beta$  and the phase transformation start stress  $\sigma^t$  are obtained as (see [14]):

$$\beta = \frac{\text{average} \left( \sum_{k=1, \dots, n} K_k^{\max} \right)}{\text{average} \sum_{k=1, \dots, n} K_{\text{tension}, k}^{\max}} \quad \text{and} \quad \sigma^t = \frac{1}{\beta} \sigma^0 \quad (27)$$

(iii) A new stress condition is applied and the corresponding phase transformation stress is determined, and so on.

Hence, a convex yield surface in the biaxial stress space  $(\sigma_1, \sigma_2)$  is determined for the polycrystalline alloys (Fig. 4).

## 5. Conclusion

A mechanical study of Ni–Mn–Ga SMA was carried out in this paper: a driving force  $\pi^f$  was introduced and

used as a yield criterion to preclude the austenite-twinned martensite transformation, and model the ferromagnetic SMA pseudoelasticity. An extension to a Ni–Mn–Ga single crystal modelling permitted to well simulate some experimental curves with two plateaus in a stress–strain space.

The crystallographic theory of martensitic transformation permitted us to show the existence of a clearly defined interface between austenite and twinned martensite. The determination of this interface (habit plane normal  $\vec{m}$  and shape strain vector  $\vec{b}$ ) allowed us to obtain the initiation surface of stress-induced martensitic phase transformation.

A magnetic study and a magneto-mechanical coupling will be done later in order to explain the giant magneto-mechanical strain effect obtained in these FSMA.

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