MARTENSITIC TRANSFORMATIONS: FROM CONTINUUM MECHANICS TO SPIN MODELS AND AUTOMATA

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Abstract

We present a new methodology for the systematic reduction of a continuum theory of martensitic transformations to a spin system whose dynamics can be described by an automaton. Our prototypical model reproduces most of the experimental observations in martensites associated with criticality and power law acoustic emission. In particular, it explains in a natural way why cyclic training is necessary to reach scale-free behavior.

Introduction

Both acoustic and calorimetric signals which are routinely detected in shape-memory alloys submitted to quasistatic loading have a marked intermittent character [1-4]. Such behavior indicates that the finite strain effect associated with the martensitic transformation splits in a steadily driven system into a set of strain avalanches of different sizes which reflect transitions between neighboring metastable states [1-5]. A statistical analysis shows that after multiple thermal cycling through the transition the amplitudes of the avalanches evolve towards a powerlaw distribution [2,3]. This means that during such loading and reloading process (training), shape-memory alloys self-organize towards criticality. In this sense martensites¹ are essentially different from many other apparently similar systems, say driven ferromagnets displaying Barkhausen noise, because in those systems training is not required to reach a scale-free behavior.

Criticality has attracted a great deal of interest over the last decades due to its ubiquity in nature, as powerlaw distributions have been reported in a wide variety of complex systems [6,7]. At criticality there are no characteristic length scales and microscopic details do not play a crucial role. This generates classes of universal behavior which can be studied by means of relatively simple, prototypical models. Thus, sandpile automata, showing self-organized criticality (SOC) [8], and driven models of the Ising type with quenched disorder, showing classical criticality [9], are known to give different prototypes expected to represent a broad class of avalanche-mediated behaviors observed in physical systems as diverse as magnetics and earthquakes.

Within the context of martensites, considerable recent effort has also been devoted to understanding of the mechanisms leading to criticality. A qualitative link to SOC has been established early on in [1, 10]. Quantitative models implying the existence of continuous disorder-induced phase transitions as in spin systems with quenched disorder have been studied by many groups (see, for instance, the most recent paper [11]). Large scale numerical simulations of cyclically loaded martensites within the framework of classical elasticity have been initiated in [12, 13]. Finally, there was a recent attempt to link martensites to spin glasses [14]. While a scale-free behavior is predicted by some of the above approaches, none of these can in principle explain why criticality in martensites requires cycling and why the power-law acoustic emission is observed only after a plasticity-like shakedown.

In a recent paper [15] we proposed an explanation for the peculiar emergence of criticality in martensites based on the experimental evidence of considerable dislocational activity in cyclically driven shape memory alloys [16–20]. In this paper we conjectured that the disorder needed for criticality in martensites is not quenched but is acquired by the system in the process of training. Here we provide additional details on a method leading to this conclusion which is based on the reduction of a continuum elasticity theory first to a discrete spin model and then to an automaton of a sand pile type. The resulting prototypical system preserves all the essential features of the original continuum model, in particular it captures all the three major phenomena: phase transformation, twinning, and plasticity. Most importantly, the model accounts for

¹Except those with very small spontaneous strain.

training-induced criticality in martensites including the prediction of such fine features of the scale free nonequilibrium steady state as the shape of avalanches and the fractal nature of the emerging spatial complexity.

Model

We start with a classical continuum theory of martensites which is based on the assumption that the elastic energy density is a nonconvex function of the deformation gradient, e.g. [21,22]. The general structure of this function in 2D has been characterized in [23, 24]. The classical description of martensitic transformations based on the Landau theory may be viewed as particular cases of the general framework described in [23, 24]. The main advantage of considering the general theory is that it takes into account large distortions which are responsible for the formation of dislocations. Therefore the interplay between the phase transformation and the evolution of such defects can be studied in a natural way within this framework. We mention that a version of the Landau theory with discontinuous displacements which assumes conventional description of dislocations has been recently proposed in [25].

Energy

The first step of our reduction procedure is a spatial discretization of the continuum model allowing one to represent a 2D Bravais lattice as a collection of N kinematically compatible units. Each unit has a multi-well energy $\phi(\theta)$ which depends on the temperature θ of the solid. To be specific, we consider a particular case in which the solid undergoes a transformation from square austenite to oblique martensite when lowering θ . The energy of the system along this transformation path can be expressed in terms of a single scalar order parameter variable e as [15]

$$\widetilde{\Phi}(e;\theta) = \sum_{i=1}^{N} f(e_i;\theta) + \frac{1}{2} \sum_{i,j=1}^{N} K_{ij} e_i e_j, \qquad (1)$$

where $\mathbf{K} = \{K_{ij}\}$ is the kernel describing elastic interactions between different units and incorporating the nonlocal interactions due to the compatibility constraints [26]. The renormalized energy of an elastic unit $f(e; \theta)$ inherits the multi-well character of the original energy ϕ . We approximate f by a periodic function with three-parabolic form in each period, where each of the three parabolas is given by the same expression $f(e; \theta) = \frac{1}{2}(e-w)^2 + g(\theta)s^2$ with different parameters w and s [see Fig. 1]. The parameter $w = d + \bar{e}s$ defines the location of the bottoms



Figure 1: Representation of the on-site periodic potential $f(e; \theta)$. The magnitudes $s, d, g(\theta)$, and \bar{e} characterizing $f(e; \theta)$ are explicitly indicated. The shaded box highlights the first domain of periodicity. Insets on top show the lattice structures corresponding to the bottoms of the potential wells, with lattice cells marked for the austenite A and for the two variants of martensite M_{\pm} in the first period. Austenites A_{\pm} are the first two replicas of A outside the first period.

of the energy wells in terms of the two discrete variables: $s = 0, \pm 1$ and $d \in \mathbb{Z}$; \bar{e} is the transformation strain. The parameter s accounts for the phase transition: s = 0 in austenite and $s = \pm 1$ in the two variants of martensite. The value of d specifies the period of f. Finally, the function $g(\theta)$ measures the difference in energy between austenite and martensite. We implement thermal driving by changing periodically the preference between austenite and martensite through cycling $g(\theta)$ at constant rate.

Continuous dynamics

The martensitic transformation has a markedly athermal character at least for the shape-memory alloys we are interested in [28], and we can consider a purely mechanical setting. We assume that our continuous dynamics is overdamped and write the dimensionless evolution equation for the field $\mathbf{e} = \{e_i\}$ in the form

$$\frac{\partial \mathbf{e}}{\partial t} = -\gamma \frac{\partial \Phi(\mathbf{e}; \theta)}{\partial \mathbf{e}},\tag{2}$$

where γ is the ratio of the rate of relaxation to the rate of driving. In the quasi-static limit (i.e., $\gamma \to \infty$), the dynamics projects on the local minima of $\tilde{\Phi}$ which form a discrete set of branches. The system remains on a particular equilibrium branch until it becomes unstable due to the driving. For smooth energies $\tilde{\Phi}$, the marginally stable configurations correspond to points where the Hessian of $\tilde{\Phi}$ becomes degenerate, det $\|\partial^2 \tilde{\Phi} / \partial \mathbf{e}^2\| = 0$, which is in general a global condition [5,29].

In the case of the piece-wise quadratic energy $f(e;\theta)$, the equilibrium equations $\partial \tilde{\Phi} / \partial \mathbf{e} = \mathbf{0}$ give

$$\mathbf{e}(\mathbf{s}, \mathbf{d}) = (\mathbf{I} + \mathbf{K})^{-1} (\mathbf{d} + \bar{e}\mathbf{s}), \qquad (3)$$

where **I** is the identity matrix, $\mathbf{s} = \{s_i\}$, and $\mathbf{d} = \{d_i\}$. This allows us to adiabatically eliminate the remaining continuous degrees of freedom which are in this case harmonic. The stability conditions become local and can be explicitly written as $e^{-}(s_i, d_i; g) < e_i < e^{+}(s_i, d_i; g)$ for every unit *i*, where

$$e^{\pm}(s_i, d_i; g) = \begin{cases} w_i \pm (\bar{e}/2 + g/\bar{e}), & s_i = 0\\ w_i \pm (\bar{e}/2 - g/\bar{e}), & s_i = \pm 1\\ w_i \pm (1 - 2\bar{e})/2, & s_i = \pm 1. \end{cases}$$
(4)

The first two cases correspond to the direct and to the reverse martensitic transformation, respectively. The third limit corresponds to the creation of a slip. When the stability condition is violated by at least one unit, the resulting instability resolves through an avalanche which brings the system to another equilibrium branch characterized by different values of \mathbf{s} and \mathbf{d} . Since these variables are discrete, the continuous dynamics (2) gives rise to a series of jumps [5].

Spin model

By eliminating the continuous variables from Eq. (1), we can rewrite the relaxed energy in terms of the variables s and d. The resulting Hamiltonian is of *Potts* type:

$$\widehat{\Phi} = -\sum_{i,j} \left[\frac{\overline{e}^2}{2} (J_{ij} - \frac{2g}{\overline{e}^2} \delta_{ij}) s_i s_j + \frac{1}{2} J_{ij} d_i d_j + \overline{e} J_{ij} s_i d_j \right],$$
(5)

where $\mathbf{J} = (\mathbf{I} + \mathbf{K})^{-1} - \mathbf{I}$ is the matrix of effective interaction. Here the spin variable \mathbf{s} , describing the martensitic phase transition and twinning, is analogous to similar variables in the conventional spin models of martensitic phase transitions [11, 14]. The new integer-valued variable \mathbf{d} describes the slip. It has the same meaning as the lattice incompatibility measures introduced in classical models of plasticity employing discontinuous displacements (e.g. [25]). Because \mathbf{d} follows a threshold dynamics, this variable can not be adiabatically eliminated and the resulting problem can not be easily formulated in terms of \mathbf{s} only.

Instead of deriving the expression for \mathbf{J} from its explicit representation above through \mathbf{K} , we follow the approach of Ref. [15] and postulate directly the structure of the kernel capturing the main expected features: anisotropy and sign indefiniteness [30]. More specifically, we assume J is of the ANNNI type:

$$J_{ij} = \begin{cases} J_0, & i = j \\ J_1 > 0, & i \text{ n.n. } j \\ -J_2 < 0, & i \text{ n.n.n. } j \\ 0, & \text{otherwise,} \end{cases}$$
(6)

where 'n.n.' and 'n.n.n.' indicate nearest and next-tonearest neighbors, respectively. This simple hypothesis accounts for the competing interactions driving the coarsening and the refinement of the microstructure [31, 32]. In order to penalize the inhomogeneity of the field **w** the self-interaction term is chosen to satisfy $J_0 = J_{ii} =$ $-\sum_{i \neq j} J_{ij}$. Our unpublished work shows that these assumptions on **J** reflect rather faithfully the structure of the matrix obtained from a lattice model with an interaction **K** derived from kinematic compatibility. For instance, despite the short-range structure of the renormalized **J**, the background kernel $\mathbf{K} = (\mathbf{I}+\mathbf{J})^{-1}-\mathbf{I}$ describing elastic interactions in our prototypical lattice has a longrange character.

Automaton

The jump process described above can be conveniently represented as a sandpile automaton. To this end we assume that the elastic strain $\delta_i = e_i - w_i$ represents the 'local height' of the sand pile. From Eq. (4) and the definition of **J**, we obtain that $\delta_i = \sum_j J_{ij}(s_j + \bar{e}d_j)$. The dependence on temperature is fully contained in the variables $\delta^{\pm}(s_i; g(\theta)) = e^{\pm}(s_i, d_i; g) - w_i$ which enter the stability condition $\delta^-(s_i, g) < \delta_i < \delta^+(s_i, g)$. In contrast to e^{\pm} [Eq. (4)], the limits of stability δ^{\pm} do not depend on **d** (i.e., they are independent of the period of the energy).

Once a cell *i* becomes unstable (the condition $\delta^- < \delta_i < \delta^+$ is violated), the strain variable passes to a neighboring energy well. As a consequence, the location of the bottom of the well w_i updates to $w_i + r$, where $r = \pm \bar{e}$ for phase transitions ($s_i = 0 \leftrightarrow s_i = \pm 1$) and $r = \pm (1 - 2\bar{e})$ for slips ($s_i = \pm 1 \rightarrow s_i = \mp 1, d_i \rightarrow d_i \pm 1$). Such rearrangement produces an update in δ ,

$$\delta_j \to \delta_j + J_{ij}r \tag{7}$$

which, given our assumptions concerning (6), can be rewritten in the form

$$\delta_i \quad \to \quad \delta_i - 4(J_1 - J_2)r \tag{8}$$

 $\delta_j \rightarrow \delta_j + J_1 r$, *j* nearest neighbors of *i*,

 $\delta_k \rightarrow \delta_k - J_2 r$, k next-to-nearest neighbors of i.

Notice that each update may make new sites unstable, and therefore the updates must continue at constant guntil the system is fully equilibrated. The total number of updates before stabilization defines the size A of an avalanche. We assume open boundary conditions so that the update of the total elastic strain $\Delta = \sum_j \delta_j$ (when the cell *i* becomes unstable) is

$$\Delta \to \Delta - \begin{cases} 0, & i \in \text{bulk} \\ (J_1 - 2J_2)r, & i \in \text{edge} \\ (2J_1 - 3J_2)r, & i \in \text{corner.} \end{cases}$$
(9)

The automaton variable is then conserved for all the updates except those originated at the boundaries of the system. The inequality² $J_1 > 2J_2$ guarantees that during the avalanches the stress is released (at a rate which decreases with N as $N^{-1/3}$). This 'dissipation' mechanism together with the quasi-static driving provide the necessary conditions for the system to reach a self-organized stationary state [33].

Numerical results

The automaton introduced in the previous section can be easily implemented numerically. Here we present the numerical results corresponding to an almost reconstructive transformation with $\bar{e} = 0.47$. We work on a square grid with 501 × 501 units and assume that $J_1 = 0.062$ and $J_2 = 0.03$ which ensure that the dissipation inequality is satisfied. We impose a synchronous dynamics such that all the unstable cells are updated simultaneously as the discrete time increases by one unit. The number of simultaneous updates in an avalanche defines its duration T. The initially homogeneous austenite is arbitrarily chosen to contain four dislocation loops placed at randomly chosen locations; assuming instead only one initial loop gives similar results [15].

Fig. 2(a) shows the spatial distribution of **s** in the martensitic phase after cycle 1 and after cycle 1000. The complexity of the phase microstructure clearly increases during the training period. The system develops a certain amount of plastic deformation $(d \neq 0)$ induced by the phase transition as shown in Fig. 2(b). We monitor the level of plastic deformation through the density ρ of nearest neighbors with differing values of d_i (our measure of dislocation density). As shown in Fig. 3(a), the evolution of ρ is marked by a steep initial increase (training period) which after approximately 150 cycles leads to a steady regime (shakedown).



Figure 2: Evolution of the phase and defect microstructures in the lattice during thermal cycling. Four dislocation loops were initially placed in random locations. (a) Configuration of the martensitic phase domains, represented by the field s_i , after cycle 1 and cycle 1000 (turquoise and blue indicate s = 1 and s = -1, respectively). (b) The corresponding configurations of the slip variable d. White and blue colors indicate d = 0 and $d \neq 0$ (mostly |d| = 1), respectively.

To compare our model with experiments where the acoustic emission during the transformation was detected, we link the size of the avalanches in the model with the acoustic bursts in experiments. As shown in Fig. 3(b), the distribution of avalanche sizes p(A) evolves from a supercritical behavior (peak at large values of A) during the first cycles towards a power law in the steady state regime.

We observe that in the present framework, the scalefree size distribution does not necessarily imply scale-free distribution of durations, p(T), in Fig. 4(a). This is the consequence of our simplifying assumptions; the model starts to generate a power-law structure of durations as soon as a small amount of quenched disorder is introduced (in the form of a Gaussian distribution of the initial δ_i with zero mean and standard deviation r). When the initial disorder is small such modification does not influence significantly the power-law character of p(A) [Fig. 4(b)]; big amounts of disorder prevent large avalanches from occurring and the probability distributions develops a subcritical cut-off [9].

Besides the power-law statistics and the microstructure complexity, the proposed model accounts for other experimental observations in martensites such as hysteresis

²In the language of sandpile automata, the condition $J_1 > 2J_2$ ensures that grains of sand are lost at the boundaries of the system.





Figure 3: Evolution towards a critical steady state. (a) Dislocation density ρ during the first 1000 cycles. (c) Distribution of the avalanche sizes after cycle 1 (triangles) and in the stationary state after 1000 cycles (squares). The distribution for the first cycle (displaced vertically for clarity) has been obtained by averaging over 800 realizations of the system with different location of the initial dislocations. The distribution in the stationary state has been obtained by averaging over the last 800 cycles in the series with 1000 transformations.

shakedown and asymmetric signal shapes (see Ref. [15] for more detail).

Conclusions

Our main conclusion is that SOC originates in martensites from the interplay between the reversible phase change and the irreversible development of an optimal amount of plastic deformation. We emphasize that the disorder responsible for criticality is not imposed through initial inhomogeneity but is created by the system itself. This suggests that the emergence of scale-free behavior in the proposed model is an example of a jamming transition.

The phase transition in our picture occurs through intermittent dynamics with large fluctuations which lead to permanent changes in the system. Most of these changes take place during the training period. This period is finite because the dislocational activity eventually saturates: despite periodicity of the 'homogeneous' energy landscape the convex interaction term in the energy makes succes-

Figure 4: Distributions of avalanche (a) durations and (b) sizes in the steady state attained after 1000 cycles with initial quenched disorder (r = 0.01) and without (r = 0).

sive slips progressively more and more expensive.

One of the most unexpected outcomes of the model is that the whole wealth of phenomena accompanying selforganization to criticality in martensites can be captured, at least qualitatively, in a relatively simple 2D spin model with a standard threshold dynamics. The main advantage of this minimal model comparing to its more detailed analogs, say [12, 13, 25], is its full mathematical transparency allowing one to pursue the origin of scalefree behavior analytically. It comes at a cost because due to the schematic treatment of the long-range interactions, of the inertial dynamics, and of the relative energetic cost of twin and phase interfaces, the model does not capture the process of self-organization in full quantitative detail. Among the omitted effects we particularly mention the elastic incompatibility of the energy wells which is known to be important for the level of reversibility achieved by a martensitic transformation [34]. In order to ensure an engineering level of agreement of the theory with experiments, more realistic models of the computational nature must be pursued in parallel to the prototypical treatment presented here.

In conclusion we mention that while we have limited ourselves in this paper to temperature-driven martensitic transformations, our theoretical framework offers a tool for studying self-organization in systems subjected to mechanical driving as well. An example of such a study, exploring the effect of the stiffness of the loading device on the critical exponents, and investigating the possibility of driving-induced crossover between different universality classes, can be found in [35].

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