

## Training-Induced Criticality in Martensites

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We propose an explanation for the self-organization towards criticality observed in martensites during the cyclic process known as “training.” The scale-free behavior originates from the interplay between the reversible phase transformation and the concurrent activity of lattice defects. The basis of the model is a continuous dynamical system on a rugged energy landscape, which in the quasistatic limit reduces to a sandpile automaton. We reproduce all the principal observations in thermally driven martensites, including power-law statistics, hysteresis shakedown, asymmetric signal shapes, and correlated disorder.

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Experiments in martensites reveal intermittent behavior with power-law statistics [1–4], showing an intrinsic complexity comparable to that of turbulence, earthquakes, internet networks, and financial markets. Criticality is known to be an issue of great significance in contemporary science, giving a framework for understanding the emergence of complexity in a variety of natural systems [5,6]. Within materials science, criticality has been recognized in the last years as a key factor in crystal plasticity, brittle fracture, and damage [7]. In the present Letter, we develop a model explaining why similar behavior is also observed in martensitic transformations.

Reversible martensitic transformations involve a coordinated distortion of the crystal lattice and belong to the class of ferroelastic first-order phase changes with athermal character [8,9]. In such systems the macroscopic strain discontinuity typically splits into a set of bursts (avalanches) corresponding to transitions between neighboring metastable states. The individual avalanches can be detected through the measurement of the intermittent acoustic or calorimetric signals. The size distribution observed in shape-memory alloys (Cu-Al-Ni, Cu-Zn-Al, Cu-Al-Mn, Ni-Mn-Ga) was shown to be scale free [1–4,9]. Despite the apparent similarity with driven ferromagnetic systems, where the scale-free Barkhausen noise has been known for a long time, the experiments on memory alloys show features not observed in magnets, and which are instead reminiscent of plastic shakedown. In particular, the critical character of the avalanches [2,3] and the smoothing of the hysteresis profile [3,10] emerge only after multiple thermal cycling through the transition.

The mechanism leading to training-induced critical behavior in martensites strongly resembles the phenomenology associated with self-organized criticality (SOC) [5]. The SOC paradigm in the form of a sandpile automaton has been applied to martensitic transformations in [11]; which, however, lacked a connection to the physics of martensitic transformations. A different set of models exploited the similarity between martensites and magnetics by interpret-

ing both in an Ising-type framework, with zero temperature and quenched disorder [12]. In this context the power-law over a few decades of avalanche sizes is viewed as a sign of proximity of the system to a classical critical point. Criticality then emerges only as a result of tuning the disorder. Furthermore, the symmetric avalanche shapes with scaling collapse, which are expected in these models [12], contrast those experimentally recorded [13]. More recent modeling has focused on the direct simulation of martensitic transformations within the framework of elasticity theory [14]. While the corresponding numerical tests show some scaling in avalanche sizes, the system is unable to memorize its state of disorder upon unloading, failing to exhibit the effects of training.

A key experimental observation left aside in the preceding theoretical work is the dislocational activity assisting the development of the phase transformation in these materials [10,15–18]. It has been repeatedly observed that dislocations are indeed introduced in shape memory alloys during training. In particular, under periodic driving the degree of defectiveness first increases monotonically and then saturates [16,17]. Our model shows that this dislocational activity is highly correlated and is ultimately responsible for the scale-free character of the reversible behavior of martensites. More precisely, the attainment of criticality is due to the ability of the crystal to develop an optimal amount of disorder.

To describe in a unified way the processes involving both dislocations and phase boundaries, we consider a prototypical 2D system of kinematically compatible elastic units resulting from a suitable triangulation of a square lattice (see an example of such procedure in [19]). To each unit with index  $i$  we assign a multiwell strain energy function depending on a single scalar order parameter  $e$  which, in turn, is a combination of the components of the discrete strain tensor. The adiabatic elimination of the harmonic nonorder-parameter strain variables by means of the equilibrium equations and the kinematic compatibility constraints leads to a nonlocal elastic energy of the form

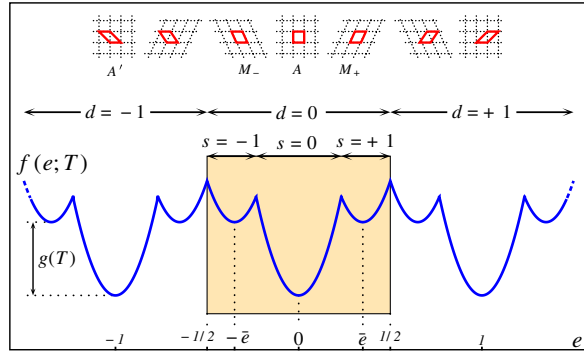


FIG. 1 (color online). Schematic representation of the periodic potential  $f(e; T)$  with the shaded box highlighting a single domain of periodicity. Insets on top show the lattice structures corresponding to the bottoms of the potential wells, with lattice cells marked for the austenite  $A$  and for the martensite variants  $M_{\pm}$ . The austenite  $A'$  corresponds to a different period of the potential.

$$\tilde{\Phi}(\mathbf{e}; T) = \sum_i f(e_i; T) + \frac{1}{2} \sum_{i,j} \mathbf{K}_{ij} e_i e_j, \quad (1)$$

where  $\mathbf{K} = \{K_{ij}\}$  is the kernel of the long-range elastic interactions, and  $f$  is a periodic function as in Fig. 1. In each period we use the three-parabolic approximation  $f(e; T) = \frac{1}{2}(e-w)^2 + g(T)s^2$ , where  $s = 0$  in the high symmetry phase (austenite) and  $s = \pm 1$  in the two variants of the low symmetry phase (martensite). The parameter  $w = d + \bar{e}s$ , where  $\bar{e}$  is the transformation strain, defines the location of the bottoms of the energy wells; the integer-valued parameter  $d$  specifies the period of  $f$ . We emphasize that no randomness has been assumed in the model.

The global periodicity of the energy takes large shearing distortions into account [20], so that both the phase change and dislocation formation can be handled simultaneously. When the transformation strain is small and the energy barriers for slip are much higher than the barriers for the phase transition and twinning (“weak transitions”, as with the rhombohedral  $R$  phase of NiTi [10], or the premartensitic transformation in Ni-Mn-Ga [4]), no lattice-invariant shears occur and modeling can proceed according to the Landau theory. Such phase changes do not generate significant dislocational activity and are largely reversible, as is assumed, for instance, in [14,21]. On the contrary, in “reconstructive transformations” the transformation strain is large, as it lays at the boundary of the periodicity domain (e.g., the ideal Bain transformation from bcc to fcc). In those cases the energy barriers to slip are only as high as the transformation barriers, and the phase change advances “halfway” towards the formation of dislocations [19]. As a result, defects proliferate making the transition irreversible [22]. In between these two extremes, a range of possibilities exists, where defect formation plays an increasing role as the transformation strain gets closer to the boundary of the maximal periodicity domain. In particular, all the martensitic transformations considered in the experiments that we are concerned with [1–4,9] involve a transformation

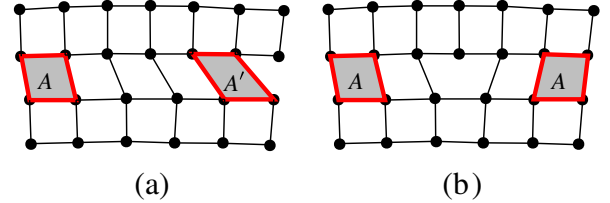


FIG. 2 (color online). Two representations of a dislocation. (a) Continuous lattice deformation involving partial slip, as is interpreted in the text. (b) The same atomic configuration viewed within the classical interpretation involving a discontinuous deformation and a nonzero Burgers vector. The shaded units refer to austenite in the wells  $A$  and  $A'$  in Fig. 1.

strain  $\bar{e}$  which is very close to the ideal Bain strain [23]. When slowly driven, these systems are expected to exhibit cell deformations not confined to one periodicity domain, but rather extending to an unbounded portion of the periodic energy landscape. This involves formation of dislocations, which our kinematic compatibility assumption does not exclude, as is exemplified in Fig. 2, where we compare the standard representation of a dislocation [Fig. 2(b)] with the one adopted in this Letter [Fig. 2(a)].

We drive the system quasistatically through the function  $g(T)$ , by changing the temperature  $T$ . Since the transformations are typically athermal [9], we consider a purely mechanical setting with overdamped dynamics. By making this assumption we treat the acoustic emission as a part of dissipation. In the limit of infinitely slow driving, the dynamics projects on the local minima of the total energy  $\tilde{\Phi}$ , which form a discrete set of branches  $e = e(g)$ , with  $e^- < e < e^+$ , where the extremes  $e^{\pm}$  correspond to marginally stable configurations [24]. For piecewise parabolic  $f$  as in Fig. 1, the limits  $e^{\pm}$  of each branch can be explicitly written as a function of  $g$  and  $\bar{e}$ . When such limits are reached, the instability resolves through a fast event (avalanche) which brings the system to another equilibrium branch. In this way the dynamics becomes piecewise continuous (see [25] for a study of the 1D case).

We proceed by eliminating through minimization the linear elastic strain  $e$  at given  $d$  and  $s$  obtaining  $\mathbf{e} = (\mathbf{1} + \mathbf{K})^{-1} \mathbf{w}$ . The relaxed energy is of the Ising type

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

where the two discrete spin variables describe the phase transformation ( $s$ ) and the plastic slip ( $d$ ), and  $\mathbf{J} = (\mathbf{1} + \mathbf{K})^{-1} - \mathbf{1}$ . Since the energy (2) is supposed to penalize the inhomogeneity of the field  $w_i$  induced by either phase boundaries or dislocations, we assume that the corresponding term has the particular form  $\frac{1}{4} \sum J_{ij} (w_i - w_j)^2$  so that  $J_{ii} = - \sum_{j \neq i} J_{ij}$ . We furthermore assume the kernel  $\mathbf{J}$  to be of the ANNNI type, to account for the competing interactions driving both the coarsening and the refinement of

the microstructure [21,26]. Specifically, we consider  $J_{ij} = J_1 > 0$  for nearest neighbors, and  $J_{ij} = -J_2 < 0$  for next-to-nearest neighbors; the ensuing diagonal-dominated structure of the matrix  $\mathbf{J}$  does not prevent the original matrix  $\mathbf{K}$  in (1) from being dense. Suitable inequalities ensure the metastability of the individual equilibrium branches [25]; in particular, the condition  $J_1 \geq 2J_2$  is sufficient for our automaton to reach a steady state (we use  $J_1 = 0.062$ ,  $J_2 = 0.03$  in the simulations presented below).

Under the above hypotheses, the piecewise continuous dynamics becomes a sandpile automaton, whose main variable is the elastic strain  $\delta_i = e_i - w_i$  representing the “local height.” Once a cell becomes unstable (the condition  $e^- < e_i < e^+$  is violated),  $\delta_i$  is updated as

$$\begin{aligned} \delta_i &\rightarrow \delta_i - 4(J_1 - J_2)r, \\ \delta_j &\rightarrow \delta_j + J_1 r, \quad j \text{ nearest neighbors of } i, \\ \delta_k &\rightarrow \delta_k - J_2 r, \quad k \text{ next-to-nearest neighbors of } i, \end{aligned} \quad (3)$$

where  $r = \pm \bar{e}$  for phase transitions, and  $r = \pm(1 - 2\bar{e})$  for slips. Since each update may make new sites unstable, the updates continue at constant  $g$  until the system is fully equilibrated. For  $J_2 = 0$ ,  $\bar{e} = \frac{1}{3}$ , and  $g = 0$  the automaton (3) reduces to the standard Bak-Tang-Wiesenfeld sandpile [5].

We implement the model numerically on a  $501 \times 501$  grid for an almost reconstructive transformation with  $\bar{e} = 0.47$  and open boundary conditions. The initially homogeneous austenite is chosen to contain only a minimal dislocation loop. The crystal is then thermally cycled through the complete transformation,  $g$  being a periodic triangular function of computational time. The intensity of the acoustic bursts registered experimentally is linked to the size of the avalanches (total number of updates before stabilization). Figure 3(a) shows the development of the phase microstructure during the training period. The level of plastic deformation is monitored through the density  $\rho$  of nearest neighbors with differing values of  $d_i$ . Figure 3(b) shows the formation of dislocations induced by training and marked by the steep initial increase in the variable  $\rho$  [Fig. 4(a)]. The creation of correlated dislocation microstructure quickly saturates, in accordance with the experiments [16,17]. In Fig. 4(b), we observe the smoothing effect of the self-organized defects on the cooling curves (and hence on the hysteresis cycle). The dislocational activity leads to the increase of the martensite starting temperature, similarly to what is reported experimentally [3]. The parallel development of criticality is indicated by the emergence of the power-law statistics for the avalanche sizes [Fig. 4(c)]. At the beginning of the training period the avalanche distribution is supercritical with a peak at large sizes evident from the sharp initial cooling curves in Fig. 4(b). The peak eventually vanishes [Fig. 4(c)], as in the experiments [3]. Two further predictions of the model matching experimental data [13] concern the strong asym-

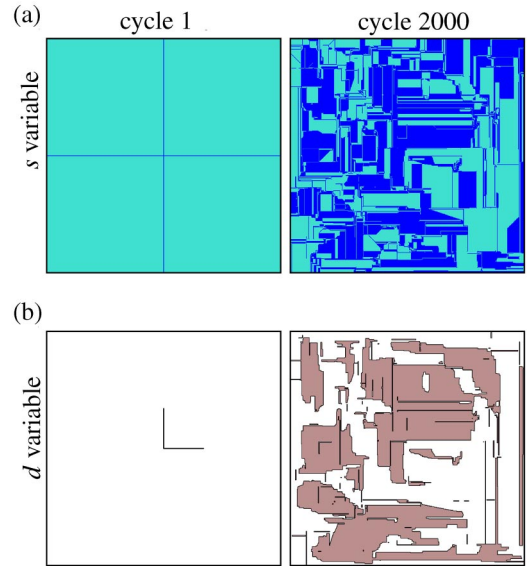


FIG. 3 (color online). Evolution of the phase and defect microstructures in the lattice during thermal cycling. A minimal dislocation loop was placed at the middle of the system. (a) Configuration of the martensitic phase domains, represented by the field  $s_i$ , after cycle 1 and cycle 2000 (turquoise and blue indicate  $s = 1$  and  $s = -1$ , respectively). (b) The corresponding configurations of the slip variable  $d$ . White and brown colors indicate  $d = 0$  and  $d \neq 0$  (mostly  $|d| = 1$ ), respectively. The black contours separate elements with different  $d$ .

metry of the avalanche shapes and the absence of their scaling collapse [see Fig. 4(d)]. Similar effects are also observed in Barkhausen noise, earthquakes, and dynamic fracture (see the discussion in [27]). Figure 4(c) shows the distribution of avalanche durations (number of simultaneous updates in an avalanche) predicted by the model, which deviates from a power law. This indicates that in the present framework a scale-free size distribution does not always imply scaling in time. While this prediction for an idealized system (with neither inertia nor fixed pinning sites) disagrees with the scaling for durations reported experimentally [1], our model does generate an almost power-law distribution of durations after the introduction of a small amount of quenched disorder represented by a Gaussian distribution of  $\delta_i$  with zero average in the initial configuration. Such modification, however, does not influence the power-law distribution of avalanches, nor does it affect the pulse asymmetry.

When in the original setting with two variables  $s$  and  $d$  the phase transformation is suppressed (no variable  $s$ ); the model describes the micromechanics of stress-driven intermittent plastic flow in crystals [28]. In this case the system is defined by the single integer-valued order parameter  $d$  as in the phase-field description of plasticity [29], to which our model offers an analytically accessible automaton alternative [30]. What is more, the present scheme of plasticity does not require *ad hoc* procedures for the nucleation and annihilation of dislocations, as in

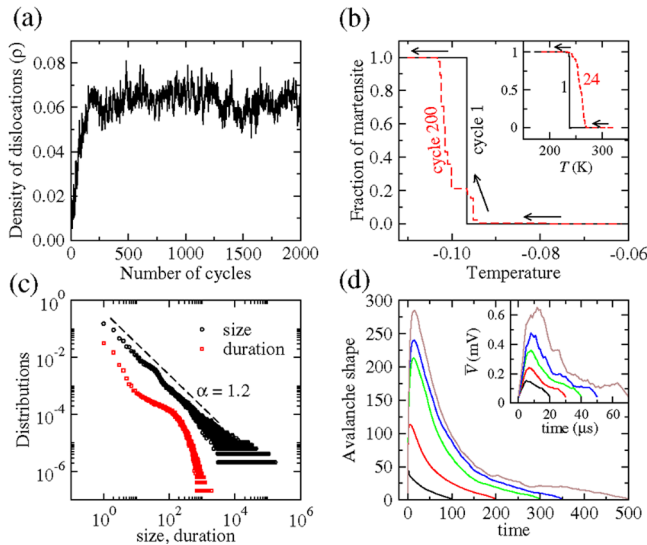


FIG. 4 (color online). Evolution to criticality and features of the critical state. (a) The dislocation density  $\rho$  during the first 2000 cycles. (b) Cooling curves representing phase fraction vs temperature in arbitrary units (cycle 1, solid line; cycle 200, dashed line). The corresponding experimental data [3] are shown in the inset for comparison (1st and 24th cycles). (c) Steady-state power-law distribution of the avalanche sizes and nonpower-law distribution of durations (displaced one decade lower for clarity). (d) Asymmetric avalanche shapes. The inset shows the corresponding experimental data [13].

discrete dislocation dynamics [31]. When, in contrast, the variable  $d$  can be neglected in the original setting, as in weak martensitic transformations (or in magnetics), the model still generates several decades of power-law avalanche distribution in accordance with the experiments in Ni-Mn-Ga [4], but only in the presence of some quenched disorder represented again by a Gaussian distribution of  $\delta_i$  in the initial configuration. In this case the emergence of limited scaling does not involve training and can be explained by the proximity of the system to a classical critical point [12].

In summary, the proposed self-organizing spin model accounts for all the main observed phenomena accompanying the training process in martensites leading to criticality. The agreement with experiment clearly indicates that SOC originates in these systems as a result of the interplay between the reversible phase change and the irreversible development of an optimal amount of plastic deformation.

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