Multi-Scale Dynamics of Twinning in SMA

Eilon Faran¹ · Doron Shilo¹

Published online: 16 May 2015
© ASM International 2015

Abstract The mechanical response of shape memory alloys (SMA) is determined by the dynamics of discrete twin boundaries, and is quantified through constitutive material laws called kinetic relations. Extracting reliable kinetic relations, as well as revealing the physical characteristics of the energy barriers that dictate these relations, are essential for understanding and modeling the overall twinning phenomena. Here, we present a comprehensive, multi-scale study of discrete twin boundary dynamics in a ferromagnetic SMA, NiMnGa. The combination of dynamic-pulsed magnetic field experiments, in conjunction with low-rate uniaxial compression tests, leads to the identification of the dominant energy barriers for twinning. In particular, we show how different mechanisms of motion for overcoming the atomic-scale lattice potential give rise to several kinetic relations that are valid at different ranges of the driving force. In addition, a unique statistical analysis of the low-rate loading curve allows distinguishing between events at different length scales. This analysis leads to the identification of a characteristic length scale ($\sim 15 \mu\text{m}$) for the distance between barriers that are responsible for the twinning stress property. This characteristic distance is in agreement with the typical thickness of the internal micro-twin structure, which was recently found in these materials.

Keywords Magnetic shape memory · NiMnGa · NiTi · Mechanical behavior · Shape memory · Twinning · Twinning shear

Introduction

Shape memory alloys (SMA) are characterized by a high symmetry (austenite) crystallographic phase and a low symmetry (martensite) crystallographic phase. The low symmetry phase is associated with a spontaneous strain and typically exhibits a twinned microstructure, in which a grain or a single crystal is divided into different twin variants [1, 2]. Each of these variants has a different orientation of the unit cell and in accordance different mechanical and physical properties. When subjected to an external driving force, one twin variant may expand at the cost of the other through nucleation and propagation of twin boundaries and twinning dislocations, in a process collectively known as twinning. The twinning transformation is associated with a significant strain change (i.e., beyond elasticity) and is therefore used in a variety of SMA-based actuation applications [3–9]. In particular, unfolding the dynamics of twinning transformation is essential for the design and implementation of SMA-based applications.

Apart from playing a fundamental role in the activation of SMA, twinning serves as a dominant deformation mechanism in other material systems. For example, twinning governs the mechanical response of hexagonal close-packed (HCP) metals (e.g., Mg and Ti) [10], nano-crystalline and nano-structures of face-centered cubic (FCC) metals (e.g., Cu and Ni) [11–13], and other advanced materials such as ferroelastic minerals [1], and ferroelectric ceramics [14]. Consequently, insights gained by studying twinning phenomena in SMA are valuable for a wide range of research fields and can contribute to the development of new and improved materials.

A quantitative description of a twinning-based macroscopic response requires the knowledge of a kinetic

✉ Eilon Faran
eilonfa@tx.technion.ac.il

¹ Department of Mechanical Engineering, Technion,
3200003 Haifa, Israel

relation. This fundamental material law describes the velocity of an individual twin boundary v_{TB} (or a twinning dislocation v_{TD}) as a function of the external driving force g [15]. For example, following the common ‘discrete twin boundary dynamics’ modeling approach, which is similar to the discrete dislocation dynamics approach but is more simple and straightforward, the macroscopic strain rate $\dot{\epsilon}$ can be described by the relation:

$$\dot{\epsilon} = N_{TB} \cdot \epsilon_T \cdot v_{TB}(g). \quad (1)$$

N_{TB} is the number of mobile twin boundaries, ϵ_T is the strain jump across the twin boundary, i.e., the twinning strain, and the kinetic relation is given by $v_{TB}(g)$. Equation (1) is valid under several assumptions, which are accurate in many cases: the existence of one active twinning system (i.e., reorientation takes place between two types of twin variants), and no interaction between twin boundaries beyond the nm scale. In addition, N_{TB} is usually constant and known for a given crystal (e.g., when active twin boundaries are stabilized [16]).

Despite their important role, experimentally validated kinetic relations are scarce. In the absence of reliable kinetic relations, simulations, which are typically based on the phase field [17–20] and discrete twin boundary approaches [21, 22], usually assume an over-simplified linear kinetic law. Such a kinetic law is valid only in cases where the driving force is much larger than all energy barriers for twin boundary motion. As we show in this paper, this condition holds only for a limited range of the driving force. Generally, different types of kinetic relations may arise for different ranges of the driving force and temperature, reflecting on the different mechanisms of motion that are active under different loading conditions.

Another modeling tool that was recently applied for studying twinning phenomena is molecular dynamics (MD) simulations. MD enables calculation of the kinetic relations (see. for example Ref. [23]) and in addition, can reveal the mechanisms by which the TB propagates [23–26]. The latter information can guide the formulation of analytical kinetic relations, which describe TB propagation in accordance with MD observation. Moreover, MD allows the evaluation of TB properties at the atomistic scale. At the same time, MD is limited in the sense that the modeled material volume is typically at the sub- μm scale. Consequently, this size limitation prohibits from capturing the effects of long-range barriers (which are at the μm scale) on twinning phenomena. Moreover, the effects of surfaces and other interfaces/defects are ignored due to periodic boundary conditions that are typically applied. Consequently, MD provides limited capabilities in capturing phenomena such as nucleation of twin boundaries and twinning dislocations, since these processes are faster near

surfaces, interfaces (e.g., grain boundaries), and in the vicinity of other lattice defects [27–29].

While a kinetic relation bares fundamental importance in the quantitative description of twinning dynamics, the concept of such a relation assumes that the twin boundary velocity depends only on the driving force, thus ignoring local barriers and the mechanisms by which the twin boundary overcomes these barriers. Consequently, a kinetic relation is meaningful for describing the average twin boundary velocity over length scales much larger than those of relevant barriers for motion. This understanding emphasizes the need for the identification and characterization of the energy landscape under which a twin boundary propagates. This landscape may be generated by barriers of different intensities (amplitude) and widths (length scale), as well as different spatial distributions within a given crystal. Knowledge about the physical characteristics of the different barriers, as well as the mechanisms of motion is required both for modeling twinning transformation and for the development of new and improved materials.

The preceding discussion reveals the complexity involved in the attempt to quantify twin boundary motion in SMA. Moreover, it emphasizes the need for a multi-scale experimental investigation approach, which can capture the dynamics of individual twin boundaries at different length and time scales. In this paper, we describe such a comprehensive analysis in a representative ferromagnetic shape memory alloy (FSMA) NiMnGa.

NiMnGa provides several advantages for the general study of twin boundary dynamics, and in particular for an experimental study under controllable loading conditions. First, single crystals of this alloy usually exhibits just few twin boundaries separated by large distances [30], which facilitates an experimental investigation on the level of individual twin boundaries. Second, in this material system, twin boundary motion can be induced either by a magnetic field and/or a mechanical load, enabling a variety of possible loading conditions. Third, the barriers for twin boundary motion in NiMnGa are much smaller than the barriers for ordinary dislocation motion and therefore twinning is practically the only mechanism for deformation beyond linear elasticity [31, 32]. On the other hand, it is very complicated to simulate NiMnGa by MD since it involves three different atoms, unknown potentials, and is very sensitive to atomic concentrations. Therefore, experimental results have to be analyzed by a comparison with analytical models. Such models simplify the real problem at the atomistic scale, but are general and relevant for many material systems.

Our investigation, which combines analytical analysis and experimental results, starts with a general description

of the main barriers for twin boundary motion, and possible mechanisms for overcoming these barriers. We classify these barriers according to their typical length scale, and in particular distinguish between atomistic-scale lattice barriers to longer range (typically μm scale) barriers. Based on the different barriers and possible mechanisms of motion associated with these barriers, several fundamental kinetic relations are formulated analytically. The experimental part of this study includes different loading methods that allow studying twinning dynamics over a wide range of velocities and driving forces. Insights gained from data analysis of the different experimental methods lead to validation of the formulated kinetic relations and mechanism of motion, as well as reveal the physical origin and characteristics of the different energy barriers.

Energy Barriers for Twin Boundary Motion at Different Length Scales

At the atomistic scale, the periodicity of the crystal induces several energy barriers, which exist even in a crystal free of imperfections (see Fig. 1). First, we consider the lattice barrier that resists the uniform motion of a twin boundary

as a flat plane. This 2D potential represents the change of the twin boundary energy (per unit area) as a function of its position (see, for example, Refs. [26, 33–39], and follows the periodicity of the lattice in the direction normal to the twin boundary (see Fig. 1a, b). In a similar way, we can consider the 1D lattice potential for twinning dislocations, which are line defects on an existing twin boundary and possess the characteristics of ordinary dislocations [40, 41]. Twinning dislocation can also be envisioned as the edges of two dimensional steps that form on the boundary plane [33] (see Fig. 1a that illustrates such a step). The 1D lattice potential, also known as the Peierls potential [42], represents the periodic change of the twinning dislocation energy as a function of its position on the twinning plane (see Fig. 1c). Depending on the driving force and rate limiting step, both the 1D and 2D lattice potentials can give rise to several possible mechanisms of motion, which result in different kinetic relations (see more details in “Twin Boundary Motion Under the Lattice Potential—the Kinetic Relations” section).

At length scales larger than the atomistic scale, several possible barriers for twin boundary motion may play a dominant role. These include interactions with other crystal defects (e.g., dislocations [43], clusters of point defects,

Fig. 1 **a** A schematic representation of the lattice potentials that form barriers for twin boundary motion. The boundary is marked in light green, and contains a step whose edges (marked in red) are twinning dislocations. Black solid and dotted rectangles represent “hills” and “valleys” (respectively) of the periodic 2D potential, which resists the motion of a twin boundary as a flat plane along the e_1 direction at velocity v_{TB} . Red solid and dotted lines represent “hills” and “valleys” (respectively) of the 1D potential, which resists the lateral propagation of twinning dislocations along the e_2 and e_3 directions at velocity v_{TD} . The periodic energy landscapes induced by the 2D and the 1D potentials are shown schematically in (b) and (c), respectively (Color figure online)

and surface defects [44]), strain compatibility along grain boundaries (in a polycrystalline material), as well as barriers imposed by the complex internal twinning microstructure often found in various types of SMA materials. The latter is characteristic to FSMA NiMnGa, where a hierarchical microstructure of internal micro-twins and 180° magnetic domains has been observed [45, 46].

Twin Boundary Motion Under the Lattice Potential—the Kinetic Relations

Analytical Analysis

In cases where twin boundary motion is governed by the lattice potential, either the 1D or 2D barriers shown in Fig. 1, several kinetic relations can be formulated based on averaging over the numerous events that occur at the atomistic scale. Table 1 summarizes these different relations and their occurrences at different ranges of the driving force. Above some transition value g_0 , which can be correlated to the amplitude of the periodic 2D lattice potential, the twin boundary moves uniformly as a flat plane. In this case, motion is modeled based on the Landau–Khalatnikov linear rate law [35, 39], leading to an average twin

boundary velocity that is described by a square root kinetic relation of the type $v_{TB} = \mu \sqrt{g^2 - g_0^2}$, where μ is a mobility coefficient (relation (I) in Table 1). Below this transition value, the 2D lattice potential forces the twin boundary to lie within the potential's wells. In this case, twin boundary motion must involve a thermally activated process, in which two dimensional steps are nucleated on the twin plane and then grow by a motion of the step edges, i.e., motion of twinning dislocations. This mechanism was observed in several computational works of different crystals, see for example Refs. [23, 24, 26].

The overall sideways velocity of the twin boundary in this mechanism may be limited either by the nucleation rate, or by the velocity of twinning dislocations. In the former case, i.e., where nucleation is the rate limiting process while motion is a fast athermal process, a standard nucleation and growth type analysis leads to an exponential kinetic relation (see relation (II) in Table 1). If, on the other hand, the rate limiting step for the overall motion of the twin boundary is the lateral propagation of individual twinning dislocations under the 1D Peierls potential, we employ a simple pile up model and show that the twin boundary velocity v_{TB} is directly proportional to the velocity of a twinning dislocation v_{TD} , according to the relation $v_{TB} \propto g v_{TD}$ (see Ref. [47]). v_{TD} is evaluated by distinguishing between two modes of motion, that are associated with overcoming the 1D Peierls potential. Above a transition driving force g_p , which is proportional to the amplitude of the 1D Peierls potential, individual twinning dislocations move uniformly in a fast athermal motion [48] and follow a kinetic relation identical to relation (I). Consequently, the overall TB velocity in this case is given by $v_{TB} = \mu' g \sqrt{g^2 - g_p^2}$, where μ' is a mobility coefficient (relation (III) in Table 1). For driving force values smaller than g_p , twinning dislocation motion is typically limited by the nucleation rate of kink pairs, as in classical dislocation dynamics models [49, 50]. In this case, the velocity of individual TDs is an exponential function of the driving force. Consequently, the resulting kinetic relation for TB motion also follows an exponential dependence of the form given by relation (IV) in Table 1 (see, e.g., Refs. [51, 52]). The two scenarios shown in Table 1, which lead to four kinetic behaviors, imply that different mechanisms of motion may give rise to several possible kinetic relations, which may be valid at different ranges of the driving force.

Table 1 Different kinetic relations that describe twin boundary motion under the resistance of the 2D and 1D lattice potentials

	(I) $v_{TB} = \mu \sqrt{g^2 - g_0^2}$
	(III) $v_{TB} = \mu' g \sqrt{g^2 - g_p^2}$
	(II) $v_{TB} = v_0 \exp \left[-A \left(\frac{g_0}{g} + \frac{2}{3\pi} \left(\frac{g_0}{g} \right)^2 \right) \right]$
	(IV) $v_{TB} = g v_0 \exp \left[-B \left(1 - \left(\frac{g}{g_p} \right)^p \right)^q \right]$

Depending on the rate limiting process, different mechanisms of motion leads to different kinetic relations that take place at different ranges of the driving force g . The driving force values g_0 and g_p correspond to the amplitudes of the 2D and 1D potentials and represent the transition values at which twin boundary kinetics switch from thermally activated to athermal motion. In relations (I) and III: μ and μ' are mobility coefficients. In relations (II) and (IV) v_0 is a pre-exponential term. A and B are driving force independent terms that contain the temperature and material properties such as the energy of twinning dislocations. $\hat{g}$ represents the driving force required for twinning dislocation motion at 0 °K and p, q are constants whose values are typically between 0 and 2 (see, e.g., Ref. [51]).

Experimental Investigation

Validation of the different kinetic relations presented in Table 1 is performed using a unique experimental setup which allows evaluating the velocity of individual twin boundaries in a ferromagnetic crystal as a function of the

external driving force. The experimental system applies rectangular-shaped magnetic pulses, with amplitude of up to 2 T, duration in the range 10–120 μs and rise and fall times that are less than 10 % of the overall pulse duration. Under these conditions, the external driving force is kept nearly constant throughout the majority of the pulse duration. Consequently, the average twin boundary velocity is evaluated based on the ratio between the boundary displacement during a single pulse and the pulse duration. The twin boundary displacements are measured by an optical microscope and are therefore at the μm scale, i.e., larger by several orders of magnitude than the periodicity of the lattice potential. Thus, effects caused by the periodic lattice potential are always averaged. On the other hand, the twin boundary displacements are comparable to the distance between the long-range barriers and therefore we can consider two situations: if the twin boundary does not meet a long-range barrier during the pulse then the measured average velocity represents the kinetic relation of a “defect-free” crystal, i.e., kinetics that is dictated by the lattice potential. Otherwise, the measured average velocity will be smaller and will represent an effect of the long-range barriers. Thus, for a large enough number of data points, we expect that the maximal velocities under each value of the driving force will represent the kinetic relation in a defect-free crystal, while smaller velocity values will exhibit scattering governed by the long-range barriers.

Typical pulsed magnetic field data for an individual type I twin boundary in an FSMA NiMnGa single crystal are shown in Fig. 2a. The scattering in the measured velocity values indicates on interactions with long-range barriers, which lead to temporal slowing down of the moving twin boundary. At the same time, we note that the maximal velocity values (that represent the kinetics under the lattice potential) follow two different trends that display a clear transition between thermally activated mechanism of motion and athermal mechanism of motion. The specific value of the driving force, which we assign as g_{trans} can be correlated with overcoming the 2D lattice potential implying that $g_{\text{trans}} = g_0$. In this case, the two different kinetic behaviors can be described by relations (I) and (II) in Table 1. Alternatively, depending on the actual rate limiting step for twin boundary motion, g_{trans} can be associated with g_p , i.e., the 1D Peierls potential. In this case, kinetic relations (III) and (IV) in Table 1 describe the two different trends of the boundary velocity shown in Fig. 2a. For the data shown in Fig. 2a (as well as for data obtained for type II twin boundaries in the same crystal), the scenario that describes a transition between kinetic relations (I) and (II) provides a slightly better fit. We note, however, that the “real” scenario, which is dictated by the actual rate limiting step at the atomistic level, cannot be conclusively resolved based on our data. A possible way to clarify this issue is by expanding the

measurements of twin boundary velocity to higher values of the driving force than those shown in Fig. 2a. However, pulsed magnetic field method cannot be expanded due to the saturation of the magnetic driving force for twin boundary motion in NiMnGa at field values larger than ~ 0.75 T (see also Ref. [47]). Alternatively, higher driving force values can be obtained through an external mechanical load. Nonetheless, the application of a well-controlled mechanical pulse at the μs time scale is a challenging experimental task.

Fitting the formulated kinetic relations to the experimental data enables the extraction of several material properties which govern twin boundary motion. For example, considering the scenario described by relations (I) and (II) in Table 1 (which are represented by the trend lines shown in Fig. 2a), we are able to extract the amplitude of the 2D lattice potential, the line energy of a twinning dislocation, and the mobility for twin boundary viscous motion as a flat plane [33, 39]. We emphasize that these nano scale material properties typically cannot be evaluated using other existing experimental methods.

Beyond the fundamental understanding of twinning kinetics obtained from the results shown in Fig. 2a, several practical insights arise as well. The most important one is the realization of the required driving force values that are necessary for fast actuation of NiMnGa-based devices. An in-depth discussion of this aspect is presented in Ref. [53], in which we show that fast frequency response, which necessitates fast twin boundary motion, requires the external driving force to exceed the threshold value of g_{trans} (which represent either g_0 or g_p). The latter conclusion indicates that in most of the applications the important material property is g_{trans} rather than other more common properties that are measured under quasistatic conditions, e.g., the twinning stress.

Long-range Energy Barriers: The Origin and Characteristics of the Twinning Stress

Pulsed magnetic field experiments clearly reveal substantial scattering of twin boundary velocity data, over the entire measured range (see Fig. 2a). This observation indicates that in the majority of cases, twinning kinetics is dominated not only by the atomistic-scale lattice potential, but also by other longer range energy barriers. In order to elucidate the characteristics of these long-range barriers, we combine insights gained from pulsed magnetic field experiments, which focuses on twinning kinetics at relatively high twin boundary velocities, together with information obtained from low-rate mechanical experiments that characterize twin boundary motion at lower velocities.

The most common and simple method to study low velocity twin boundary motion is through displacement-

Fig. 2 A complete description of twin boundary kinetics in NiMnGa obtained from different experimental methods. **a** Velocities of an individual twin boundary as a function of the driving force, obtained from high rate pulsed field measurements. The maximal velocity values are marked by *empty squares* and follow two trends that are fitted by the kinetic relations given in Table 1. The transition value, at which slow thermally activated motion switches to fast athermal motion is marked as g_{trans} . **b** Low rate, displacement-controlled uniaxial stress–strain curve showing a plateau-type response at a nominal value of σ_{ts} . The *inset* reveals a saw tooth pattern of the load

profile. **c** Data obtained from the two experimental methods shown in **a** and **b** are combined on a single kinetic chart for the lower driving force range $g < g_{trans}$. *Blue markers* are velocities obtained from pulsed field experiments (data take from **a**), while black markers are nominal twinning stress values obtained from low-rate uniaxial loading tests at different average twin boundary velocities. The data point marked with an *empty red square* corresponds to the experiment shown in **b**. The *shaded red rectangle* represents the average velocity values of individual load drops within the load profile shown in **b** (see text for details) (Color figure online)

controlled uniaxial loading. In this method, the prescribed displacement rate dictates the twin boundary velocity, and the driving force required for obtaining this velocity is measured. Traditionally, such a configuration enables the measurement of a fundamental property of SMA, which is termed the twinning stress. This value, which is taken as the average value of the stress plateau in the loading curve of the martensite phase, represents the threshold stress required to initiate twin boundary motion at low velocities. We note that the twinning stress property cannot be determined from our pulsed magnetic field experiments, since this technique is limited to velocities above 2×10^{-3} m/s, due to the limited spatial resolution in the detection of twin boundary displacement and the short pulse durations.

Figure 2b is a typical uniaxial loading curve of a type I twin boundary in a NiMnGa single crystal, which was measured at a loading rate that corresponds to an average boundary velocity of $\bar{v}_{TB} = 2.8 \times 10^{-6}$ m/s. The load profile shows a typical plateau-type response, which, upon magnification reveals a sawtooth pattern (see inset in Fig. 2b). The nominal value of the plateau stress represents the twinning stress σ_{ts} (marked by the dashed black line in Fig. 2b), which, for this boundary type equals about 0.85 MPa. This value corresponds to a driving force of $g_{ts} \approx 50 \frac{\text{kJ}}{\text{m}^3}$ that is smaller than the transition force associated with the lattice potential (i.e., $g_{trans} \approx 110 \frac{\text{kJ}}{\text{m}^3}$, see the pulsed field data in Fig. 2a). In addition, quasistatic

experiments conducted at different loading rates, which correspond to different average twin boundary velocities in the range of $10^{-7} - 10^{-2}$ m/s (all slower than the minimal values measured in the pulsed field method), reveal no significant change in the value of the twinning stress. Plotting these values on a velocity versus driving force chart (black markers in Fig. 2c), together with the values obtained in pulsed field experiments at $g < g_{\text{trans}}$ (blue markers in Fig. 2c) indicates that the twinning stress property represents a threshold driving force value below which twin boundaries are immobile. This observation implies that while the barrier associated with the lattice potential can be overcome by thermally activated processes (this mechanism of motion takes place at $<g_{\text{trans}}$, as discussed in the previous section), the twinning stress barrier cannot. Consequently, the length scale of the barrier responsible for the twinning stress is expected to be much larger than that of the lattice potential, and as such may be responsible for the scattering observed in our pulsed magnetic field experimental data.

Quantitative information regarding the actual length scale of the twinning stress barrier, as well as its physical origin, is obtained by a closer inspection of the uniaxial low-rate loading curve. Following an initial linear elastic regime, the curve displays a sawtooth pattern at a nearly constant stress value (see inset in Fig. 2c, as well as Fig. 3a). This behavior is typically attributed to “jerky” motion, which means that the twin boundary moves in a non-uniform manner from one barrier to the next. Such ‘stick-slip’ motion was actually observed experimentally, by in situ monitoring of the twin boundary displacement [53]. Jerky motion, such as that displayed by a twin boundary in NiMnGa under low-rate uniaxial loading, is characteristic to crackling noise phenomena, which arises when a system responds to changing external conditions through discrete, impulsive events spanning a broad range of sizes [54].

Typically, crackling noise dynamics is studied by analyzing the probability distribution of jerky events. In the case of twin boundary motion, jerky events can be correlated to local displacements of the propagating interface. The statistical distribution of the magnitudes and velocities of these local displacements may shed light on the underlying long-range energy barrier responsible for the twinning stress property, as well as on the mechanism of motion.

Evaluation of local twin boundary displacements and velocities are obtained by formulating the kinematics of uniaxial loading of a twinned crystal. In particular, the total strain ε , which is dictated by the constant displacement rate c , is expressed as the sum of two contributions

$$\varepsilon = \frac{c \cdot t}{L_0} = \frac{\sigma}{E} + x_{\text{TB}} \cdot \varepsilon_{\text{T}}. \quad (2)$$

Here, t represents the elapsed time and L_0 is the sample's initial length along the loading direction. The first term on the right hand side of Eq. (2) is the uniform elastic strain, where σ and E are the instantaneous stress and elastic module of the loaded sample, respectively. The term $x_{\text{TB}} \cdot \varepsilon_{\text{T}}$ represents the strain due to the motion of a single twin boundary, where x_{TB} is the boundary displacement, and ε_{T} is the transformation strain associated with such motion (i.e., the twinning strain). Equation (2) leads to explicit relations for the boundary's displacement and velocity:

$$x_{\text{TB}} = \frac{ct}{\varepsilon_{\text{T}}} - \frac{L_0\sigma}{\varepsilon_{\text{T}}E}; \quad \dot{x}_{\text{TB}} = v_{\text{TB}} = \frac{c}{\varepsilon_{\text{T}}} - \frac{L_0\dot{\sigma}}{\varepsilon_{\text{T}}E}. \quad (3)$$

We note that the jerky load profile (a representative part of which is shown in Fig. 3a) is composed of alternating linear rising segments and rapid load drop segments. During the linear rising segments the sample responds as a linear elastic material (i.e., deforms uniformly) and the boundary velocity is practically zero. Complementary, when the time derivative of the load is negative, i.e., during the load drop segments where $\dot{\sigma} < 0$, the velocity v_{TB} exceeds the average value imposed by the external displacement rate, c/ε_{T} .

In order to evaluate the values of boundary displacements x_{TB} during individual load drops, we identify the local maxima $\sigma_{\text{max},i}$ and minima $\sigma_{\text{min},i}$ points, at which the stress is maximal or minimal (see blue and green markers, respectively, in Fig. 3a). The difference $\Delta x_{\text{TB},i} = x_{\text{TB}}(\sigma_{\text{max},i}) - x_{\text{TB}}(\sigma_{\text{min},i})$ between adjacent extrema points defines the irreversible twin boundary displacement during each individual load drop event i . The statistical distribution of all boundary displacement values $\Delta x_{\text{TB},i}$ (shown in Fig. 3b) clearly indicates that the majority of events are centered around a characteristic value of about 15 μm , which is obtained by fitting a log-normal density function to the measured data (represented by the green line in Fig. 3b). This value of distance between barriers (i.e., $\sim 15 \mu\text{m}$) is in very good agreement with the typical thickness of internal micro-twins that were recently observed in NiMnGa [46, 55–57], and thus reveals the physical origin of the twinning stress property in this material system. We note that at the low range of $\Delta x_{\text{TB},i}$, the experimental data deviate from a single log-normal distribution. This observation can be explained by fitting our data to a combination of two log-normal density functions; the main function is narrow and centered around 15 μm (green curve) and the other is broader and centered around 5 μm (gray curve). The two density functions may represent different types of barriers or the same type of barrier (e.g., interaction with the internal micro-twin structure) but different characteristic distances between barriers.

Fig. 3 A statistical analysis of low-rate uniaxial loading curve leads to evaluation of local displacements and velocities of the propagating twin boundary. **a** A representative zoom-in of several load drops reveals the jerky nature of boundary motion. Extrema points, which define the magnitude and duration of individual load drop events, are marked by *blue* and *green squares*, respectively. **b** Distribution of overall twin boundary displacements Δx_{TB_i} in each individual load drop i . Experimental values of Δx_{TB_i} are calculated according to

The observed load drops events (Fig. 3a) are associated with twin boundary having an area of few mm^2 , which moves a distance of several microns. Consequently, the transformed volume during an individual load drop is larger by about 14 orders of magnitude than the typical volume associated with atomistic-scale barriers and mechanisms of motion, i.e., the nucleation and motion of twinning dislocations [33, 47]. Naturally, the effect of atomistic-scale events, which typically occur at the picosecond time scale [23, 24] is averaged and cannot be resolved in our experiments. Yet, we are able to resolve several smaller, ‘meso-scale’ events, within individual ‘macro-scale’ load drops. A closer inspection of our data reveals that the temporary velocity of the twin boundary v_{TB} varies within an individual ‘macro-scale’ event, as well as between separate events. Thus, load drops are not accompanied by a single determined “path” in terms of v_{TB} ,

Eq. (3) and divided by 1 m to obtain unit-less variables. The distribution is centered around characteristic values at the μm scale, as indicated by the green and black lines that represent two log-normal distributions. **c** Complementary cumulative distribution function (CCDF) of temporary velocity values v_{TB} during all load drop events. The scale-invariant power law distribution indicated by the red line reveals the nature of meso-scale events by which the twin boundary moves (Color figure online)

but rather exhibit noticeable variations, which can be attributed to the existence of many ‘meso-scale’ events [58]. In addition, we notice that the average velocities during all ‘macro-scale’ load drop events, i.e., $\bar{v}_{TB_i} = \frac{\Delta x_{TB_i}}{\Delta t_i}$ are smaller by at least an order of magnitude than the velocity measured under the same driving force but higher loading rate [53]. This is shown by the shaded red rectangle in Fig. 2c that marks the range of $\bar{v}_{TB_i}$ values, calculated from the experimental data shown in Fig. 2b. This observation means that each measured value of $\bar{v}_{TB_i}$ (which characterizes a ‘macro-scale’ event) originates from the contribution of several ‘meso-scale’ events separated by waiting times which are much longer than the propagation times.

The temporary velocities, which are determined by the ‘meso-scale’ events, are calculated by applying the expression for v_{TB} in Eq. (3) to the instantaneous time

derivatives of stress profile during individual ‘macro-scale’ load drops. The calculated temporary velocities exhibit a power law probability density function of the form $p(v_{TB}) \propto v_{TB}^{-\alpha}$, with $\alpha = 2.02$, over the majority of the measured range, which spans two orders of magnitude (see Fig. 3c). A power law distribution has also been observed in analyses of crackling noise during the phase transformation in NiMnGa [59–62].

A power law density function exhibits the same behavior if plotted on different scales and is hence scale-invariant. Interestingly, such behavior is not unique to NiMnGa. In fact, numerous physical systems, e.g., phase transitions, magnetic domain switching, and the motion of tectonic plates, respond to changing external conditions through discrete impulsive events which exhibit a power law distribution [54, 63, 64]. As a result, the power law distribution has been interpreted as a universal law which is valid for many different physical processes regardless of the specific characteristics of each physical system.

While the preceding statistical analysis provides important insights regarding twinning phenomena in NiMnGa, it also demonstrates a new and unique approach for studying crackling noise phenomena. Typically, crackling noise dynamics are described by scale-invariant statistical laws (e.g., the power law distribution) that do not provide information about the mechanisms by which crackling noise occur. Here, we show that while one variable (v_{TB}) shows a scale-invariant type behavior, we are able to identify a different variable (Δx_{TB_i}) that exhibit a clear distribution centered around a characteristic value. The characteristic value of Δx_{TB_i} reveals the details of the underlying energy barrier responsible for the twinning stress property in NiMnGa. The power law distribution of v_{TB} reveals the nature of meso-scale events by which the twin boundary moves. This behavior may originate from local curvatures of the twin boundary (at the sub- μm scale) that induce ‘stochastic’ variations in its overall propagation velocity.

Summary

In this paper, we present a unique investigation of twin boundary dynamics at the level of individual interfaces in a representative ferromagnetic SMA NiMnGa. Analytical kinetic relations are formulated based on the different lattice potentials that resist boundary motion, and the possible mechanisms of motion for overcoming these barriers. The formulated relations are validated experimentally by direct measurements of twin boundary velocity under μs -scale pulsed field loading. Our results reveal a clear transition from thermally activated motion at low velocities to

athermal motion at higher velocities, and the transition occurs at a driving force that represents the amplitude of the lattice potential. In addition, twin boundary motion at low rates, which exhibits a jerky behavior, is investigated by a combined analytical and statistical analysis. By quantifying the magnitudes and velocities of twin boundary displacements during individual stress drops, we show that boundary motion proceeds through events at various scales. At the atomic-scale, twin boundary propagates through nucleation and motion of twinning dislocations. At the meso-scale, boundary velocity displays a scale-invariant distribution, presumably due to local curvatures of the twin boundary. Finally, at the μm scale, twin boundary exhibits stick-slip motion between barriers. The characteristic length scale of the distance between consecutive barriers matches the characteristic thickness of the internal microstructure in NiMnGa. This correlation points on the physical origin of the twinning stress property, below which twin boundaries are immobile.

Finally, we believe that the innovative experimental and analytical methodologies used in this study, as well as the physical insights gained for the particular NiMnGa system, provide an important baseline for future investigations of twinning phenomena in other SMA.

Acknowledgments This research was supported by the ISRAEL SCIENCE FOUNDATION (Grants Nos. 1341/10 and 1268/14).

References

1. Salje EK (1993) Phase transitions in ferroelastic and co-elastic crystals. Cambridge University Press, Cambridge
2. Bhattacharya K (2003) Microstructure of martensite. Oxford University Press, Oxford
3. Kanner OY, Shilo D, Sheng J, James RD, Ganor Y (2013) Ferromagnetic shape memory flapper for remotely actuated propulsion systems. *Smart Mater Struct* 22(8):085030
4. Nespoli A, Besseghini S, Pittaccio S, Villa E, Viscuso S (2010) The high potential of shape memory alloys in developing miniature mechanical devices: a review on shape memory alloy mini-actuators. *Sens Actuators A* 158(1):149–160
5. Mohd Jani J, Leary M, Subic A, Gibson MA (2014) A review of shape memory alloy research, applications and opportunities. *Mater Des* 56:1078–1113
6. Ganor Y, Shilo D, Zarrouati N, James RD (2009) Ferromagnetic shape memory flapper. *Sens Actuators A* 150(2):277–279
7. Pagounis E, Laptev A, Jungwirth J, Laufenberg M, Fonin M (2014) Magnetomechanical properties of a high-temperature Ni–Mn–Ga magnetic shape memory actuator material. *Scr Mater* 88:17–20
8. Stephan JM, Pagounis E, Laufenberg M, Paul O, Ruther P (2011) A novel concept for strain sensing based on the ferromagnetic shape memory alloy NiMnGa. *IEEE Sens J* 11(11):2683–2689
9. Pagounis E, Chulist R, Szczerba MJ, Laufenberg M (2014) High-temperature magnetic shape memory actuation in a Ni–Mn–Ga single crystal. *Scr Mater* 83:29–32
10. Christian JW, Mahajan S (1995) Deformation twinning. *Prog Mater Sci* 39(1–2):1–157

11. Meyers MA, Mishra A, Benson DJ (2006) Mechanical properties of nanocrystalline materials. *Prog Mater Sci* 51(4):427–556
12. Zhu YT, Liao XZ, Wu XL (2012) Deformation twinning in nanocrystalline materials. *Prog Mater Sci* 57(1):1–62
13. Asaro RJ, Krysl P, Kad B (2003) Deformation mechanism transitions in nanoscale fcc metals. *Philos Mag Lett* 83(12):733–743
14. Bhattacharya K, Ravichandran G (2003) Ferroelectric perovskites for electromechanical actuation. *Acta Mater* 51(19):5941–5960
15. Abeyaratne R, Knowles JK (2006) Evolution of phase transitions: a continuum theory. Cambridge University Press, Cambridge
16. Ezer Y, Sozinov O, Straka L, Soroka A, Lanska N ‘Magnetic Shape Memory Alloys And Specimens Thereof’, *European Patent Number EP2710161*, 2012, Filing Date: 21.05.2012, Publication Date: 2029.2011.2012
17. Dayal K, Bhattacharya K (2007) A real-space non-local phase-field model of ferroelectric domain patterns in complex geometries. *Acta Mater* 55(6):1907–1917
18. Li LJ, Lei CH, Shu YC, Li JY (2011) Phase-field simulation of magnetoelastic couplings in ferromagnetic shape memory alloys. *Acta Mater* 59(7):2648–2655
19. Hildebrand FE, Miehe C (2012) A phase field model for the formation and evolution of martensitic laminate microstructure at finite strains. *Philos Mag* 92(34):4250–4290
20. Hlinka J, Ondrejčovic P, Marton P (2009) The piezoelectric response of nanotwinned BaTiO₃. *Nanotechnology* 20(10):105709
21. Tsou NT, Huber JE, Cocks ACF (2013) Evolution of compatible laminate domain structures in ferroelectric single crystals. *Acta Mater* 61(2):670–682
22. Shilo D, Burcsu E, Ravichandran G, Bhattacharya K (2007) A model for large electrostrictive actuation in ferroelectric single crystals. *Int J Solids Struct* 44(6):2053–2065
23. Daphalapurkar NP, Wilkerson JW, Wright TW, Ramesh KT (2014) Kinetics of a fast moving twin boundary in nickel. *Acta Mater* 68:82–92
24. Salje EKH, Ding X, Zhao Z, Lookman T, Saxena A (2011) Thermally activated avalanches: jamming and the progression of needle domains. *Phys Rev B* 83(10):104109
25. Lu C-T, Dayal K (2011) Linear instability signals the initiation of motion of a twin plane under load. *Philos Mag Lett* 91(4):264–271
26. Hildebrand FE, Abeyaratne R (2008) An atomistic investigation of the kinetics of detwinning. *J Mech Phys Solids* 56(4):1296–1319
27. Rice JR (1992) Dislocation nucleation from a crack tip: an analysis based on the Peierls concept. *J Mech Phys Solids* 40(2):239–271
28. Van Swygenhoven H, Weertman JR (2006) Deformation in nanocrystalline metals. *Mater Today* 9(5):24–31
29. Kuchеров L, Tadmor EB (2007) Twin nucleation mechanisms at a crack tip in an hcp material: molecular simulation. *Acta Mater* 55(6):2065–2074
30. Straka L, Lanska N, Ullakko K, Sozinov A (2010) Twin microstructure dependent mechanical response in Ni–Mn–Ga single crystals. *Appl Phys Lett* 96(13):131903
31. Straka L, Hänninen H, Soroka A, Sozinov A (2011) Ni–Mn–Ga single crystals with very low twinning stress. *J Phys Conf Ser* 303(1):012079
32. Aaltio I, Söderberg O, Ge Y, Hannula S-P (2010) Twin boundary nucleation and motion in Ni–Mn–Ga magnetic shape memory material with a low twinning stress. *Scr Mater* 62(1):9–12
33. Faran E, Shilo D (2013) The kinetic relation for twin wall motion in NiMnGa—part 2. *J Mech Phys Solids* 61(3):726–741
34. Kibey SA, Wang LL, Liu JB, Johnson HT, Sehitoglu H, Johnson DD (2009) Quantitative prediction of twinning stress in fcc alloys: application to Cu–Al. *Phys Rev B* 79(21):214202
35. Abeyaratne R, Chu C, James RD (1996) Kinetics of materials with wiggly energies: theory and application to the evolution of twinning microstructures in a Cu–Al–Ni shape memory alloy. *Philos Mag A* 73(2):457–497
36. Kibey S, Sehitoglu H, Johnson DD (2009) Energy landscape for martensitic phase transformation in shape memory NiTi. *Acta Mater* 57(5):1624–1629
37. Lee WT, Salje EKH, Goncalves-Ferreira L, Daraktchiev M, Bismayer U (2006) Intrinsic activation energy for twin-wall motion in the ferroelastic perovskite CaTiO₃. *Phys Rev B* 73(21):214110
38. Truskinovsky L, Vainchtein A (2003) Peierls–Nabarro landscape for martensitic phase transitions. *Phys Rev B* 67(17):172103
39. Faran E, Shilo D (2011) The kinetic relation for twin wall motion in NiMnGa. *J Mech Phys Solids* 59(5):975–987
40. Pond RC, Celotto S (2003) Special interfaces: military transformations. *Int Mater Rev* 48(4):225–245
41. Howe JM, Pond RC, Hirth JP (2009) The role of disconnections in phase transformations. *Prog Mater Sci* 54(6):792–838
42. Peierls R (1940) The size of a dislocation. *Proc Phys Soc* 52(1):34
43. Paul DI, McGehee W, O’Handley RC, Richard M (2007) Ferromagnetic shape memory alloys: a theoretical approach. *J Appl Phys* 101(12):123917
44. Chmielus M, Glavatsky I, Hoffmann J-U, Chernenko VA, Schneider R, Müllner P (2011) Influence of constraints and twinning stress on magnetic field-induced strain of magnetic shape-memory alloys. *Scr Mater* 64(9):888–891
45. Heczko O (2014) Magnetic shape memory effect and highly mobile twin boundaries. *Mater Sci Technol* 30(13a):1559–1578
46. Seiner H, Straka L, Heczko O (2014) A microstructural model of motion of macro-twin interfaces in Ni–Mn–Ga 10 M martensite. *J Mech Phys Solids* 64:198–211
47. Faran E, Shilo D (2014) Dynamics of twin boundaries in ferromagnetic shape memory alloys. *Mater Sci Technol* 30(13a):1545–1558
48. Faran E, Shilo D (2010) Twin motion faster than the speed of sound. *Phys Rev Lett* 104(15):155501
49. Hirth J, Lothe J (1982) Theory of dislocations. John Wiley & Sons, New York
50. Nadgorny E (1988) Dislocation dynamics and mechanical properties of crystals. Pergamon Press, Oxford
51. Frost HJ, Ashby F (1982) Deformation-mechanism maps: the plasticity and creep of metals and ceramics. Pergamon Press, Oxford
52. Hiratani M, Nadgorny EM (2001) Combined model of dislocation motion with thermally activated and drag-dependent stages. *Acta Mater* 49(20):4337–4346
53. Faran E, Shilo D (2012) Implications of twinning kinetics on the frequency response in NiMnGa actuators. *Appl Phys Lett* 100(15):151901–151904
54. Sethna JP, Dahmen KA, Myers CR (2001) Crackling noise. *Nature* 410(6825):242–250
55. Chulist R, Straka L, Lanska N, Soroka A, Sozinov A, Skrotzki W (2013) Characterization of mobile type I and type II twin boundaries in 10 M modulated Ni–Mn–Ga martensite by electron backscatter diffraction. *Acta Mater* 61(6):1913–1920
56. Heczko O, Kopeček J, Straka L, Seiner H (2013) Differently mobile twin boundaries and magnetic shape memory effect in 10 M martensite of Ni–Mn–Ga. *Mater Res Bull* 48(12):5105–5109
57. Heczko O, Straka L, Seiner H (2013) Different microstructures of mobile twin boundaries in 10 M modulated Ni–Mn–Ga martensite. *Acta Mater* 61(2):622–631
58. Faran E, Salje EKH, Shilo D (under review) The exploration of the effect of microstructure on crackling noise systems. *Phys Rev Lett*

59. Niemann R, Baró J, Heczko O, Schultz L, Fähler S, Vives E, Mañosa L, Planes A (2012) Tuning avalanche criticality: acoustic emission during the martensitic transformation of a compressed Ni-Mn-Ga single crystal. *Phys Rev B* 86(21):214101
60. Balogh Z, Daróczy L, Harasztosi L, Beke DL, Lograsso TA, Schlagel DL (2006) Magnetic emission during austenite-martensite transformation in Ni₂MnGa shape memory alloy. *Mater Trans* 47(3):631–634
61. Baró J, Dixon S, Edwards RS, Fan Y, Keeble DS, Mañosa L, Planes A, Vives E (2013) Simultaneous detection of acoustic emission and Barkhausen noise during the martensitic transition of a Ni-Mn-Ga magnetic shape-memory alloy. *Phys Rev B* 88(17):174108
62. Niemann R, Kopeček J, Heczko O, Romberg J, Schultz L, Fähler S, Vives E, Mañosa L, Planes A (2014) Localizing sources of acoustic emission during the martensitic transformation. *Phys Rev B* 89(21):214118
63. Sethna JP (2007) Statistical mechanics: crackling crossover. *Nat Phys* 3(8):518–519
64. Salje EKH, Dahmen KA (2014) Crackling noise in disordered materials. *Annu Rev Condens Matter Phys* 5(1):233–254