Magnetic field-induced strain in single crystal Ni-Mn-Ga

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ABSTRACT

Field-induced strains up to 10% at room temperature have been observed in magnetic shape memory alloys based on off-stoichiometric compositions of the intermetallic compound Ni_2MnGa. This occurs by the motion of twin boundaries in the ferromagnetic ($T_C \approx 90^oC$) martensitic (T_{mart} typically 30 to 70^oC) state under magnetic fields of a few kOe. Some data illustrating the interdependence of strain, stress, and magnetic field are reviewed. Phenomenological models describe many of these observations by minimization of free energy terms including Zeeman energy, magnetocrystalline anisotropy energy, stored elastic energy and fractional twin-boundary distribution. Two important questions have been raised about field-induced strain in FSMAs. They are 1) the role of body forces (due to action of the field on the sample), and 2) the role of magnetostriction (stress/strain in a single variant under magnetization rotation) in the twin boundary motion. These questions are addressed in light of published data and models.

Keywords: Ferromagnetic shape memory alloy, field-induced strain, Ni-Mn-Ga, magnetostriction, field-induced stress

1. INTRODUCTION

Magnetic shape memory alloys, such as those based on off-stoichiometric compositions of the intermetallic compound Ni_2MnGa, have exhibited large strains under application of a magnetic field [1-3]. A reversible 6% field-induced strain under bias stress was observed in tetragonal marten sites ($1 - c/a \approx 0.94$) at room temperature in 1998 [4]. A 9% field-induced strain has recently been reported in orthorhombic martensites ($a \neq b \neq c$, $1 - c/a \approx 0.90$) [5]. Significant field-induced strain has also been observed in $Fe_{70}Pd_{30}$ [2]. This occurs by the motion of twin boundaries in the ferromagnetic ($T_C \approx 90^oC$) martensitic (T_{mart} typically 30 to 70^oC) state under magnetic fields of a few kOe. Some data illustrating the interdependence of field-induced strain, stress, and field are reviewed in light of two important questions about the physical origin of this phenomenon. The energy driving twin boundary motion cannot exceed the magnetocrystalline anisotropy energy density, K_u, of the martensite, which is of order 0.15 MJ/m^3 [6, 7]. It is useful and generally valid to equate this input magnetic energy density to the output work per unit volume: $\mu_0 M_s H = \sigma\, \varepsilon_0$ (for $H < H_a$) or $K_u = \sigma\, \varepsilon_0$ (for $H \geq H_a$), where $\varepsilon_0 = 1 - c/a = 6\%$ is the maximum output strain for complete change from one twin variant to another in a tetragonal Ni-Mn-Ga alloy.

Stress along the path of the bias spring measured during dynamic actuation varies non-monotonically with field strength and shows anomalies where twin boundary motion occurs [8]. Single crystals constrained at only one end have shown strain correlated with twin-boundary motion for magnetic field pulses of order 0.6 milli-second duration [9]. Fields in excess of the anisotropy field appear not to add to the driving force on the twin boundaries. Phenomenological models describe many of these observations by minimization of free energy terms including Zeeman energy, magnetocrystalline anisotropy energy, stored elastic energy and fractional twin-boundary distribution.

Two important questions have been raised about field-induced strain in FSMAs. They concern 1) the role of body forces (due to action of the field on the sample), and 2) the role of magnetostriction (stress/strain in a single variant under magnetization rotation) in the twin boundary motion. These questions are addressed here in light of published data and models.

2. CRYSTAL RESPONSE TO STRESS AND MAGNETIC FIELD

A typical stress strain curve for any shape memory material, including ferromagnetic ones, is shown schematically in Fig. 1. The relatively flat plateau portions of the loop indicate twin boundary motion, which initiates at a yield stress typically less than 10 MPa. Single crystals cannot be put in tension so stress-strain data typically represent the first quadrant behavior. The usual way of actuating Ni-Mn-Ga crystals to induce cyclic strains is by application of a magnetic

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field under a bias stress are now described. The crystal is trained so that the active twin planes lie at 45° to both the bias stress and the external field (Fig. 2). The bias stress favors growth of twin variants having the crystallographic c–axis ($c < a$) parallel to the bias stress axis (Fig. 2a). When a magnetic field is applied as shown in Fig. 2b and with the bias stress still present, twin boundaries move cyclically as the ratio of the magnetic field energy to the stress energy, $\mu_0 M_s H/\sigma\varepsilon_0$, is greater or less than unity.

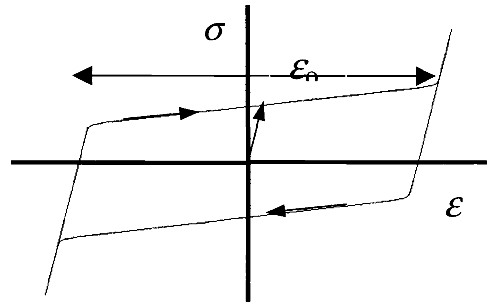

Fig. 1. Schematic stress-strain curve in compression and tension for a typical shape memory material.

Phenomenological models capture the complementary roles of stress and magnetic field in moving twin boundaries in a magnetic shape memory alloy [4, 6, 7]. These models begin with consideration of appropriate free-energy densities for the magnetization (Zeeman and magnetic anisotropy energies) and the state of strain (internal effective elastic and external stress energies) in each of three types of allowed variants in tetragonal martensite:

$$g_i = -\mu_o \boldsymbol{M}_i \cdot \boldsymbol{H} + K_u \sin^2\theta_i + (1/2)C_{eff}\varepsilon^2 + \vec{\sigma}\cdot\vec{\varepsilon}. \tag{1}$$

Minimization of the energy density for all variants leads to expressions for field-induced strain of the form [6]:

$$\varepsilon(H) = \varepsilon_o \delta f = \frac{\mu_o M_s H(1-h) + K_u h^2 - \sigma\varepsilon_o}{C_{eff}\varepsilon_o} = \frac{2K_u h(1-h/2) - \sigma\varepsilon_o}{C_{eff}\varepsilon_o}. \tag{2}$$

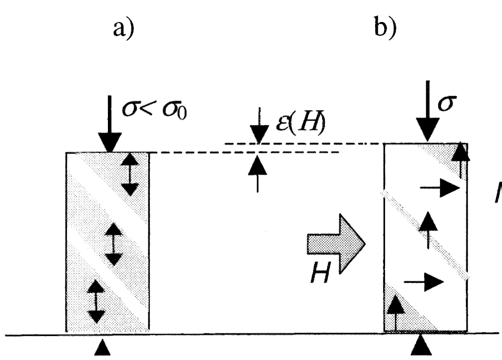

Fig. 2. The shaded and un-shaded regions in panels a) and b) are twin variants with the c axis ($c < a$) vertical and horizontal, respectively.

In Eq. 2, ε_o is the transformation strain, δf is the fractional change in twin variant distribution, h is the reduced magnetic field, $h = \mu_o M_s H/2K_u$, and C_{eff} is an effective internal restoring force (not always significant in single crystals) that describes any elastic quality of twin boundary motion. Expressions similar to Eq. 2 have been derived by a different method [7] and both models are able to fit the field dependence of strain in Ni-Mn-Ga [1, 4, 6, 7].

Fig. 3 shows three selected strain-versus-magnetic-field loops taken at 2 Hz under conditions similar to those in Fig.2 with a *dynamic* bias stress (from a reset spring) instead of a *static* bias stress [8]. What is evident is that there is a threshold field (which can be added to Eq. 2 through the external field, $H \to H \pm H_{th}$) [4] for initiation of twin boundary motion. From Eq. 2, it can be seen that in the large anisotropy limit, $h \ll 1$, the field-induced strain results from a competition between magnetic and stress energies: $\mu_o M_s H - \sigma\varepsilon_o$. These observations suggest an equivalence of magnetic and stress quantities such that the threshold stress for twin boundary motion implies a threshold field for twin boundary motion of order $H_{th} = \sigma\varepsilon_o / \mu_o M_s$ [4]. This relation holds quite well. Further, the magnitude of the field-induced strain can be first enhanced by a weak bias stress (sufficient to reset the sample after magneto-plastic deformation). Larger bias stresses suppress the field-induced strain altogether. The phenomenological models also describe these features. Finally, it is noted from Fig. 3 that for stresses close to the blocking stress, increasing field strength is NOT able to produce additional strain. This is due to the fact that at large fields, the magnetization in the unfavorably oriented variants has rotated into the field direction and the only energy difference between the two types of variants is the constant anisotropy energy. This is reflected in the models in the large h limit where the field-induced strain is now governed by the difference $K_u - \sigma\varepsilon_o$ (see Eq. 2 for $h \to 1$).

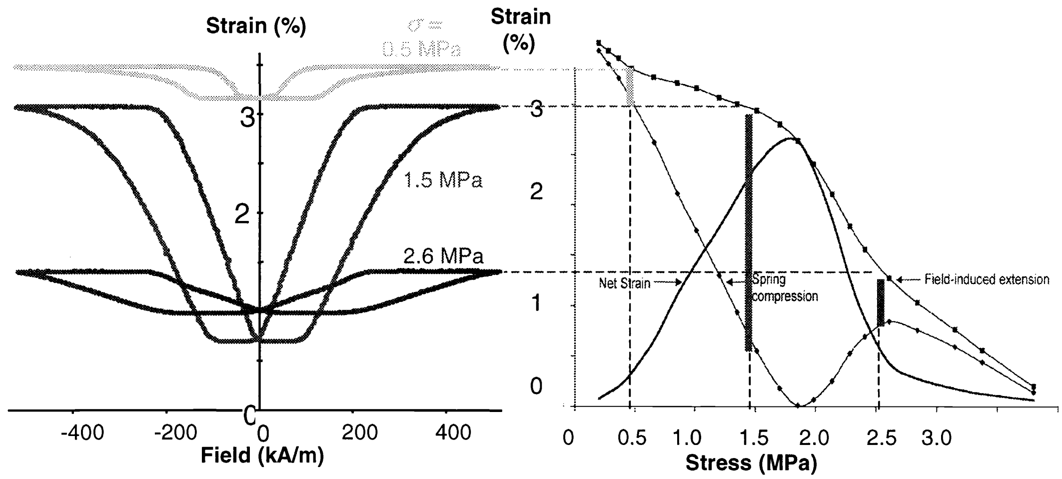

Fig. 3. Left, selected field-induced strain curves for three different initial bias stress levels. The loops are displaced on the vertical scale according to the absolute extension at the moving end of the crystal. The panel at the right shows the locus of strains at maximum and minimum fields for the full set of field-induced strain curves (data points). The solid line is the difference between the two extension curves and so represents the magnitude of the field-induced strain as a function of initial bias stress. [8]

It is tempting, given the relative success of the phenomenological models in describing the major aspects of field induced twin boundary motion, to assume that the phenomenological driving force for twin boundary motion is indeed the magnetic energy difference across the twin boundaries. However, the question has been asked [10], "Could the body force on a constrained sample in a magnetic field actually be driving the twin boundary motion?" That question is considered next.

3. FIELD-INDUCED STRAIN DUE TO MAGNETIC BODY FORCES

From Fig. 2 it is clear that in the stress-favored state (Fig. 2a) the magnetization of the sample may lie largely in one direction along the sample length (parallel to the local c axis). (The possible presence of 180° domain walls in these stress-favored variants, which would reduce the net torque on the sample due to a magnetic field, can be excluded for fields in excess of the weak domain wall coercivity, $H_c \approx 100$ Oe [4]. For fields in excess of H_c, the residual variants having c axis and $\boldsymbol{M}$ parallel to H *without* 180° domain walls will cause significant magnetostatic energy near the twin boundary if the adjacent variant does contain 180° walls.) Thus, the application of a field generally causes a net torque on the sample, $\boldsymbol{T} = \mu_0 \boldsymbol{M} \times \boldsymbol{H} = f_2 \mu_0 M H \sin\theta_2$ as illustrated in Fig. 4a. (Here f_2 is the volume fraction of field-aligned variants.) The sample will rotate if it is unconstrained. The counter torque on the sample due to the constraint is resolved across the twin boundary (Fig. 4b) in a way that promotes the growth of the field-favored variants (Fig. 4c). Carrying out the analysis leads to expressions for torque-induced twin boundary motion identical to Eq. 2. The result is even consistent in the form of approach to saturation, $h(1-h/2)$, because the torque vanishes when the magnetization rotates into the direction of the field, i.e. $h \to 1$.

So the mathematical form that describes actuation driven by magnetic energy difference across the twin boundary is the same as that for actuation driven by a shear stress on the sample that counters the field-induced torque. It is difficult to conceive an experiment that would distinguish whether the mode of actuation is due to the difference in magnetic energy favoring a change in the atomic positions across the twin boundary or action by the mechanical constraint torque on those atomic positions. The reason for this is that the mathematical forms that describe these two mechanisms are identical (at least at the mesoscopic level). However, one piece of evidence arising from pulsed-field actuation experiments does seem to support the purely magnetic mechanism, namely pulse-field actuation.

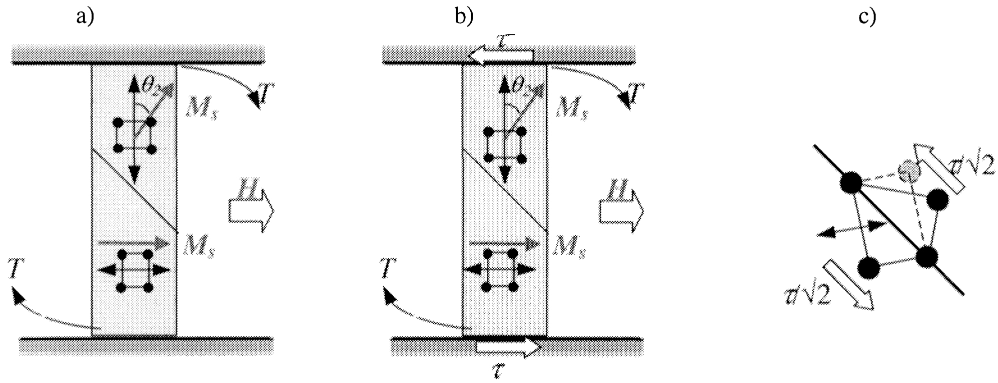

Fig. 4. a) Schematic of twinned Ni-Mn-Ga martensite crystal constrained against the clockwise torque, ***T***, induced by application of a magnetic field. b) The constraint applies a counterclockwise shear couple to the sample to maintain equilibrium. c) Schematic of twinned unit cell straddling the twin boundary. The component of the constraining shear across the twin boundary promotes growth of the field-favored variant.

Pulse field actuation of twin boundary motion helps to reveal the microscopic processes involved in twin boundary pinning and motion [9] as well as demonstrating the extent to which FSMAs can be actuated by low-inductance air coils rather than heavier iron-filled windings. In this experiment, the crystals are *bonded at one end to a fixed plane*, a magnetic field pulse is applied across the width of the crystal, and the displacement of the free end is measured with a reflected laser beam. What is important to note for the question at hand is that the sample is constrained at only one end so the stress distribution in the sample is non-uniform along its length. If the constraining mechanical torque on the sample were responsible for twin boundary motion, then twin boundary motion might be expected to occur primarily at one end and not the other. This is not observed. Photographs of crystals before and after pulse-field actuation show no preference for actuation at one end or the other [11]. This supports the idea that twin boundary motion is directly caused by the magnetic field itself and not by the accompanying mechanical torque and the shear stress it produces in a constrained sample.

4. ROLE OF MAGNETOSTRICTION IN FSMA ACTUATION

Attention is turned now to the possible role of conventional magnetostriction in the actuation process of magnetic martensites. The large field-induced strain of certain magnetic martensites is due to twin boundary motion. Twin boundary motion may or may not occur upon field-induced rotation of the magnetization in a given variant; it can occur with even negligible rotation of the magnetization. All of the deformation associated with twin boundary motion is localized at the twin boundary. It can be misleading to refer to the strain accompanying field-induced twin-boundary motion as magnetostriction. Anisotropic magnetostriction, which is observed in any ferromagnetic material, is an anisotropic strain that rotates with the direction of magnetization; the magnetostrictive strain is essentially uniform throughout a domain [12].

In an actuation experiment configured as in Fig. 2 but having the external stress provide by a compression spring, one would expect the stress in the load path to be proportional to the strain. That is, as the sample strains under an applied field, the spring is compressed beyond its initial value, increasing the stress on a load cell. In this case, the load cell output would start at a non-zero value and increase with a field dependence similar to the strain-field curves in Fig. 3. This in fact is not observed. Fig. 5 shows the output of a load cell during cyclic actuation of the Ni-Mn-Ga sample whose field-induced strain is shown in Fig.3 [8]. Note that the stress increases quadratically even at fields below the threshold for twin boundary motion. The pressure on the twin boundaries due to application of the magnetic field overtakes the magnetostrictive stress at the threshold field. For intermediate stresses this transition is marked with a plateau, while for low or high average bias stresses, only a discontinuous slope marks the threshold field. The twinning strain saturates when the initial bias stress is near the low or high limit of its range. However, at intermediate stresses where large field-induced strain is observed (cf. Fig. 3), significant stress develops in the spring forcing compressive "reset" of variant 1 to variant 2.

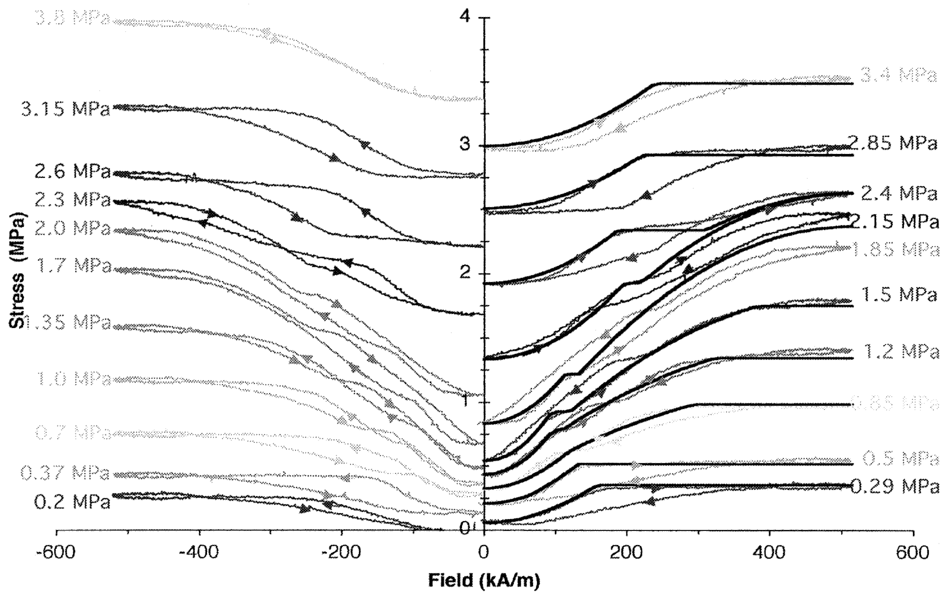

Fig. 5. Magnetic field dependence of stress measured by a load cell during actuation of Ni-Mn-Ga crystal. Theoretical results of Eq. 4 are plotted for the increasing field branch on half of the data set using different values of y_0 that correspond to the initial bias stress as labeled [8].

Anisotropic magnetostriction does occur and is observed to be about –100 to –150 ppm in Ni-Mn-Ga [3]. In a multivariant martensite as the magnetization is rotated, the domains are not free to fully express their magnetostrictive strain because of the constraints of adjacent variants. In these cases, the magnetic stress (the magnetoelastic coupling coefficient, B_1 in this case) builds up inside the constrained variant. This stress can be as large or larger than the yield stress for twin boundary motion (for Terfenol-D, $B_1 \approx 60$ MPa and in most other magnetic materials it is of order 10 MPa). The question here concerns the role of this stress accumulation on twin boundary motion as a field is applied to a multi-variant magnetic martensite.

The free energy in Eq. 1 must be supplemented with the magnetoelastic anisotropy energy density in cubic systems, which has the form [13]:

$$g_{me} = B_1\left[\varepsilon_{xx}(\alpha_x^2 - \tfrac{1}{3}) + \varepsilon_{yy}(\alpha_y^2 - \tfrac{1}{3}) + \varepsilon_{zz}(\alpha_z^2 - \tfrac{1}{3})\right] \qquad 3)$$

where the ε_{ii} are the principal magnetostrictive strains and α_i are the direction cosines of the magnetization. Because the magnetoelastic energies are small ($B_1\varepsilon \approx 10^3\,\mathrm{J/m^3}$) compared to the magnetic anisotropy, the magnetization direction, determined from $dg/d\theta = 0$, is only weakly affected by magnetoelastic considerations. However, to determine the stress inside the crystal, the derivative of the fee energy with respect to strains is taken. As pointed out above, the magnitude of B_1 is such that it cannot be neglected relative to the other stresses in the problem that affect twin distribution. Because B_1 is positive for Ni-Mn-Ga [3], a negative magnetostrictive strain, ε_{2x} (positive ε_{2y}), decreases the energy as H_x and therefore θ_2 increases. This is the definition of negative magnetostriction. Therefore, when the magnetization in variant 2 rotates in the presence of a field (Fig. 4), it exerts a compressive stress on variant 1 proportional to $-B_1 \sin^2\theta_2$, which in turn causes a strain, ε_{1y}, in variant 1. When the total energy density is written and minimized to find the strain, σ_y, the result is [8]:

$$\sigma_y = \frac{\partial g}{\partial \varepsilon_{2y}} + \sigma_{ext} = \sigma_{2y} + \sigma_{ext}$$
$$= \tfrac{3}{2}[B_1(f_1 - h^2) - \sigma_{ext}(y_0, h)] + \sigma_{ext}(y_0, h) \qquad 4)$$

The independent variables are the reduced magnetic field h, and bias stress from the spring, $\sigma_{ext}(y_0, h)$, which depends on its initial displacement, y_o, as well as additional displacement due to the application of the field. The dependent variables are the orientations of the magnetization vectors in variant 2, θ_2, the magnetostrictive strains, ε_i, and the variant volume fraction $f_1 = 1 - f_2$. The stress predicted by Eq. 4 is plotted as a solid line in Fig. 5. The model results correctly predict the quadratic increase in stress before twin-boundary motion initiates. The plateau of little or no increase in stress while twin-boundary motion occurs (small modulus) is also contained in the model. Finally, the saturation of stress above the anisotropy field is also properly described.

The question that remains is, "How does this stress accumulation affect actuation?". Clearly an increase in compressive stress, σ_y, on the sample means that a larger field is required to reach the threshold for twin boundary motion [4, 12]. On decreasing the field, the enhanced stress causes reset to occur sooner than it would otherwise. Thus, the magnetostrctive stress increases the threshold field but appears to not affect the ε-H hysteresis. This is understandable because the magnetostrctive stress is the result or magnetization rotation, which is a largely reversible process.

4. CONCLUSIONS

It is difficult and possibly only a semantic exercise to distinguish between magnetic-energy driven and reaction-stress driven mechanisms for field-induced twin boundary motion in FSMAs. The existence of a negative magnetostriction in Ni-Mn-Ga martensite appears to increase the threshold magnetic field required for twin boundary motion without enhancing the hysteresis in the ε-H loops.

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