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Temperature dependence of reversible field-induced strain in Ni–Mn–Ga single crystal

L. Straka [a], O. Heczko [b,*], S.-P. Hannula [b]

[a] *Laboratory of Biomedical Engineering, Helsinki University of Technology, Rakentajanaukio 2C, P.O. Box 2200, FIN-02015 Espoo, Finland*
[b] *Laboratory of Materials Science, Helsinki University of Technology, Vuorimiehentie 2A, P.O. Box 6200, FI-02015 TKK, Espoo, Finland*

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Abstract

Temperature dependence of reversible magnetic shape memory effect and magnetization changes were studied in $Ni_{49.7}Mn_{29.1}Ga_{21.2}$. The twinning stress, equivalent magnetic stress, switching field and maximum strain all increase with decreasing temperature. A 5–6% reversible strain is observed in the temperature range of 307–263 K and it sharply decreases to 1–2% at temperatures below 263 K. The main features of the observed behavior are explained using a simple energy-controlled model. Predictions of the model agree well with the experiment.

Keywords: Ni–Mn–Ga; Shape memory alloys; Magnetic properties; Martensite

1. Introduction

Giant magnetic-field-induced strain or magnetic shape memory effect (MSME) is due to the redistribution of martensitic twin variants or structural reorientation [1,2]. According to a simple model the redistribution is driven by the difference of magnetic energy between differently oriented martensitic variants. The MSME occurs when magnetic energy difference exceeds the energy threshold needed for twin boundary motion [3,4].

The mobility of the martensitic twin boundaries depends on the twinning stress, σ_{tw}, the magnitude of which in turn depends on the microstructure and the character of the obstacles within the material. However, very little is known of the latter. Experimentally the twinning stress, $\sigma_{tw}(\varepsilon)$, where ε is macroscopic strain, can be obtained from the stress–strain curve, i.e. from mechanical testing [4].

If the twinning stress is very low and the external stress, σ_{ext}, exceeds the twinning stress, fully reversible behavior can be obtained, i.e. about 6% reversible strain in a variable magnetic field [5]. The condition for full reversibility of the MSME in the magnetic saturation can be written as

$$K_u/\varepsilon_0 - \sigma_{tw} > \sigma_{ext} > \sigma_{tw}, \qquad (1)$$

where K_u is the magnetic anisotropy constant and ε_0 the tetragonal distortion of the lattice. If the external stress is smaller than the twinning stress we obtain only partial reversibility. Although all internal parameters depend on temperature, the temperature dependence of the MSME and its reversibility is mainly determined by the temperature dependence of the twinning stress [4]. The temperature dependence of the MSME as a one-way effect has been studied previously [4–8]. Much less is known about the behavior of the reversible strain with temperature. Cui et al. [9] studied the reversible strain of an unstressed Ni–Mn–Fe–Ga crystal, concluding there was a negligible temperature dependence of about 1% strain. However, the applicability of this result for unalloyed Ni–Mn–Ga alloys is questionable particularly considering the small magnitude of the reversible strain which is much less than the full MSME. Heczko and Ullakko studied the temperature evolution of the magnetization curves during MSME [10].

* Corresponding author. Tel.: +358 9 4512684; fax: +358 9 4512677.
E-mail address: oleg.heczko@tkk.fi (O. Heczko).

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The main purpose of this report is to present the temperature dependence of reversible MSME of a compressed Ni–Mn–Ga specimen. Furthermore it is shown that the observed complex behavior can be described by a simple energy-controlled model.

2. Experimental

Nearly single crystalline Ni–Mn–Ga ingot was prepared by a directional solidification method. The ingot was heat treated at 1273 K for 46 h and at 1073 K for 72 h in an evacuated quartz ampoule. Using energy dispersive X-ray spectroscopy, the composition of the alloy was determined as $Ni_{49.7}Mn_{29.1}Ga_{21.2}$ (at.%). The parent phase of the alloy is $L2_1$ cubic with a lattice constant of $a_A = 0.584$ nm measured at 323 K. It transforms during cooling to martensite. The transformation is completed at $T_{MF} = 308$ K. The structure of the martensite is approximately tetragonal with a five-layer modulation (5 M). The lattice constants $a_M = b_M = 0.595$ nm, $c_M = 0.561$ nm were determined by X-ray diffraction at 293 K. The reverse transformation from martensite to austenite starts at about $T_{AS} = 317$ K. No other transformation was detected down to 10 K. The tetragonal distortion or ratio is defined as $\varepsilon_0 = (1 - c_M/a_M) \times 100\%$.

A single crystalline, parallelepiped specimen approximately $5 \times 5 \times 9\ mm^3$ in size, was cut along {100} planes of the parental $L2_1$ cubic structure. A contactless laser dilatometer was used for the measurement of strain and a vibrating coil magnetometer for the measurement of magnetization. The external compressive stress and the magnetic field were applied perpendicularly to each other, both along one of the principal ⟨100⟩ axes of the crystal. The experimental arrangement is described in detail in Ref. [4]. To compare the magnetization curves in different temperatures the relative magnetization dependence was used (Fig. 1). The relative magnetization was determined as a ratio of measured magnetization $M(T)$ and the saturation magnetization M_s at 1.15 T for given temperature. The saturation magnetization M_s at room temperature is 70 Am^2/kg.

The specimen was compressed by using a 14 MPa stress at a particular temperature in order to obtain a well defined, single variant state prior to each measurement. To obtain reversible behavior in the slowly changing magnetic field a constant 1 MPa compressive stress was applied to the specimen during the test. The field-induced strain and magnetization were measured simultaneously in the temperature range of 223–307 K up to a field of 1.15 T. In this paper the switching field, H_{SW}, is defined as the applied magnetic field inducing 1% strain in the first magnetizing cycle and the maximum strain, ε_{max}, as the strain obtained at the maximum field 1.15 T in the first magnetizing cycle. The reversible strain, ε_r, is defined as the difference between the strain at the maximum field 1.15 T and the strain measured at the end of the magnetizing cycle at 0 T.

The twinning stress can be determined from the position of the plateau on the compressive stress–strain curves. To prevent ambiguity in determination of the position of the plateau, or the value of twinning stress, the average twinning stress is taken as the stress needed for 3% deformation (about half of the maximum possible strain) during the compression. This value is taken directly from the stress–strain test as measured value without considering Schmidt factor. The dependence of twinning stress on temperature was presented in Ref. [4] and the data for twinning stress used in this article are taken directly from there, since we use the same specimen as therein.

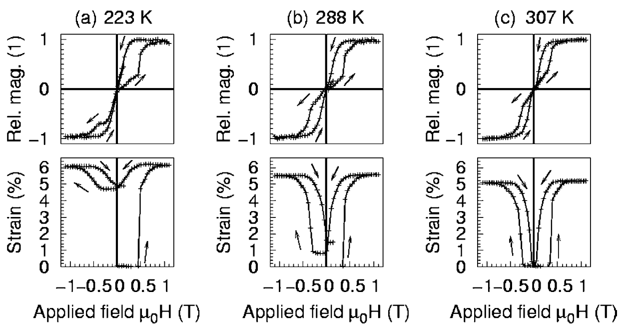

Fig. 1. Selected simultaneous measurements of strain and magnetization under external 1 MPa compressive stress in magnetic field at a temperature of (a) 223 K, (b) 288 K and (c) 307 K. The direction of the magnetic field is perpendicular to the direction of the compressive stress. Rel. mag. denotes the relative magnetization obtained as the ratio of measured magnetization and value of magnetization at 1.15 T.

3. Results and discussion

Fig. 1 shows three selected simultaneous measurements of the strain and magnetization demonstrating different behavior of the specimen at 223, 288 and 307 K. The maximum strain slightly decreases with increasing temperature, while the reversible strain increases considerably, from 1.4% at 223 K to 5% at 307 K, i.e. reaching almost fully reversible behavior.

The magnetization shows hysteresis in both the first and the third quadrants. The sharp change of the shape of magnetization curve is a typical sign of structure reorientation, i.e. the MSME [2], and the hysteresis denotes the energy consumed during the motion of the twin boundaries [5]. The strain and magnetization changes with the field occur simultaneously at all measured temperatures.

Thermal dependence of all parameters of the MSME determined from the experiment, illustrated in Fig. 1, are summarized in Fig. 2. The maximum strain at the saturating field decreases with increasing temperature. The maximum strain depends on the variant distribution and the upper limit is determined by the tetragonal distortion ε_0 of the lattice once the sample reaches the single variant state [3].

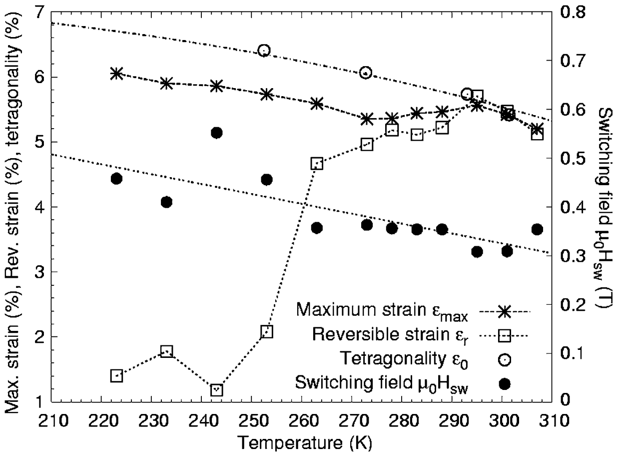

Fig. 2. Temperature dependence of tetragonal distortion ε_0, experimentally determined maximum strain ε_{max} (stars), reversible strain ε_r (empty squares) and switching field H_{SW} (filled circles) under external 1 MPa compressive stress. The dashed lines are just guides for the eye.

A very steep increase of the reversible strain, from about 2% to about 5%, occurs between 253 K and 263 K. This increase is caused by the decrease of the twinning stress close to 1 MPa (Fig. 3). In this case, when magnetic field decreases the applied compressive stress of 1 MPa is strong enough to induce the martensitic variant with the short *c*-axis along the stress and to reorient the martensitic structure. On the other hand, the variant with short *c*-axis (easy magnetization axis) along the field is induced back when the field increases again. This gives the maximum reversible strain up to the tetragonal distortion ε_0. The switching field increases with decreasing temperature (Fig. 2). At least two factors affect the switching field: (i) change of magnetic energy and (ii) change of twinning stress with temperature, which is apparently a dominant factor in the present case.

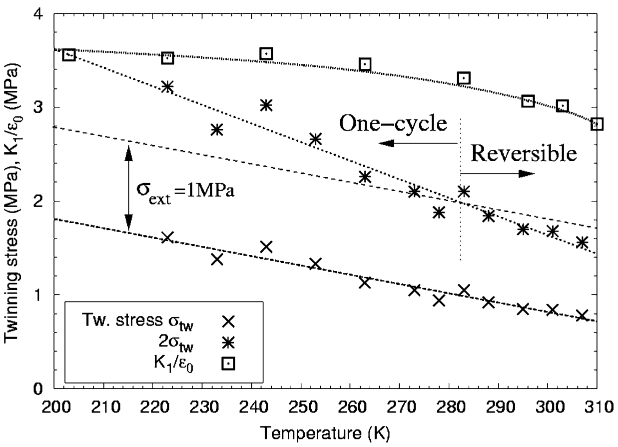

Fig. 3. Twinning stress σ_{tw} (crosses), $2\sigma_{tw}$ (stars) and equivalent magnetic stress K_u/ε_0 (empty squares) as a function of temperature. Linear fits of σ_{tw} and $2\sigma_{tw}$ are represented by dashed and dotted lines. Plot of linear fit of $\sigma_{ext} + \sigma_{tw}$ dependency (dotted line, no points) allows quick graphical analysis of reversibility of MSME (see text). The dashed curve along K_u/ε_0 is just a guide for the eyes.

Fig. 3 presents the temperature dependencies of twinning stress σ_{tw}, $2\sigma_{tw}$, equivalent magnetic stress K_u/ε_0, and the sum of external and twinning stresses $\sigma_{ext} + \sigma_{tw}$. It can be used as a graphical presentation of the relationship (1) and the limits for reversible strain can be directly predicted from the figure. If the sum $\sigma_{ext} + \sigma_{tw}$ is less than $2\sigma_{tw}$, then only one-cycle strain (irreversible) can take place. On the other hand when the sum is larger than $2\sigma_{tw}$, then reversible behavior can occur. However, if $\sigma_{ext} + \sigma_{tw}$ exceeds K_u/ε_0, then according to this model, the MSME is fully suppressed and neither reversible nor irreversible strain can happen. Using this graphical method, the low temperature limit for reversible stress is about 280 K (Fig. 3). Experimentally, the reversibility persists to around 263 K (Fig. 2). The difference between the prediction and observation can be explained by a relatively large distribution of twinning stress values around average value used in model. The distribution makes the determination of exact position of the limit inexact. Also note that points $2\sigma_{tw}$ (263 K) and $2\sigma_{tw}$ (273 K) are only very slightly above the $\sigma_{ext} + \sigma_{tw}$ line (Fig. 3).

The relatively large spread of values of twinning stress and also switching field and maximum strain can be a consequence of the different initial distribution of martensitic variants for each measurement or it might be due to some structural intermartensitic changes. More detailed investigation is needed to explain this unambiguously, but this is out of the scope of this short article.

The range of external stress in which the reversible behavior can be expected is easily determined from relationship (1) or Fig. 3. At 294 K $K_u/\varepsilon_0 = 3.1$ MPa, $\sigma_{tw} = 0.8$ MPa, and $K_u/\varepsilon_0 - \sigma_{tw} = 2.2$ MPa. Therefore, the reversible strain should be observed at 294 K when the external compressive stress is in the range 0.8–2.2 MPa. The MSME measurement under different external stresses at 294 K gives 4.3–5.6% reversible strain within the range 0.8–1.8 MPa and less than 0.7% reversible strain for any compressive stress outside of this region in agreement with the model.

Below 200 K the $2\sigma_{tw}$ is always larger than K_u/ε_0 and the reversible strain should not occur at all according to the simple model we use. Therefore 200 K is the lowest temperature for the reversible strain to occur in our specimen whatever the value of applied external compressive stress is. In fact, some small value of the reversible strain might be observed, similarly to the 1–2% observed strain for the measurement under 1 MPa and $T < 263$ K, shown in Fig. 2. The presence of this small strain where the model predicts zero strain is a consequence of our simplification, particularly the use of a single value of average twinning stress, σ_{tw}, instead of the real distribution of twinning stresses as indicated by the stress–strain curve. This assumption is acceptable for large strains as the plateau of the stress–strain curve is very flat.

The condition $K_u/\varepsilon_0 > \sigma_{tw}$ is satisfied down to 165 K, which is the limit for one-way (irreversible) MSME. It was shown in Ref. [4] that the increase of the twinning stress is exponential-like rather than linear below 200 K.

4. Conclusion

- Experimentally determined twinning stress, K_u/ε_0 and switching field and maximum strain increase with decreasing temperature approximately monotonically. The experimentally observed distribution of twinning stresses and switching fields from linear dependence need further investigation.
- The 5–6% reversible strain close to one-way MSME is observed in the range 307–263 K and it suddenly decreases to 1–2% below 263 K.
- The existence and behavior of reversible stress with temperature is reasonably well explained using a simple energy-controlled model. The agreement of the model predictions with the experiments is good.

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