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Magnetization changes in Ni–Mn–Ga magnetic shape memory single crystal during compressive stress reorientation

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Abstract

Magnetization as a function of strain of a $Ni_{48.5}Mn_{30.8}Ga_{20.7}$ single crystal during 9 MPa compressive loading test was obtained by means of vibrating coil magnetometry for different static magnetic fields up to 1.15 T. The dependency is monotonic, nonlinear and shows a small hysteresis. Maximum reversible change of magnetization is 30% for a 0.6 T field, and irreversible change of about 50% is observed for a 0.4 T field. The magnetization dependencies determined are compared with a simple linear model.

Keywords: Ni–Mn–Ga; Shape memory alloys (SMA); Magnetic properties

1. Introduction

Some Ni–Mn–Ga alloys exhibit giant magnetic field-induced strain, also called the magnetic shape memory effect. The effect occurs due to the rearrangement of martensitic variants or structure reorientation in a magnetic field [1]. Interrelationship between magnetization changes and martensitic variant rearrangement was recognized early and it was demonstrated that the peculiar shape of the magnetization curve is a result of the martensitic variant redistribution [2,3].

The suggestion of utilizing this effect in sensor applications was probably first published by Suorsa et al. [4]. The magnetization changes during compressive stress-induced reorientation of Ni–Mn–Ga martensite in a static magnetic field were first studied by Müllner et al. [5]. The magnetization change was monitored by a Hall probe during compressive loading and unloading. They observed an almost 30% local magnetization change at the sample's surface in a magnetic field of 0.7 T for about 10 MPa compressive stress. However, the maximum reversible strain was just 2%, which indicates that the reorientation was far from complete as about 6% strain is expected for full reorientation of the 5 M martensite [6]. Similar measurement of magnetization changes during stress-induced reorientation in static field of 0.5 T was also presented by Li et al.; the maximum reversible strain was small, about 2.5% [7]. A disadvantage of these measurements is that the Hall probe surveys only local magnetic flux, which is related to local magnetization, and thus depends on the local configuration of the variants [8]. The overall magnetization changes during sample compression determined by vibrating coil magnetometer in a static field of 0.8 T were reported in Ref. [9]. The magnetization change was about 14% for a strain of 5.2%. This value of strain indicates the nearly full structural reorientation of the sample. The compressive stress changes the volume ratio of the martensitic variants, which leads to changes in the magnetization curve. This was demonstrated also in Ref. [10] for various strains of compressed samples; however, only minor magnetization curves were measured, as the maximum field used, 0.16 T, is well below saturation.

The measurement of magnetization changes during stress-induced martensitic transformation (superelastic

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regime) were first published in Ref. [11] and monitoring the magnetization changes to detect the martensite fraction was suggested in Ref. [12].

In this article, we study magnetization changes during almost complete stress-induced martensite reorientation for various magnitudes of static magnetic field up to saturation. Using vibrating coil magnetometry (VCM) instead of a Hall probe, we were able to capture the overall changes. The magnetization changes are associated with the redistribution of the martensitic variants and they can be directly interpreted or modeled using magnetization curves of a sample containing a single martensitic variant.

2. Experimental

A single crystal sample for measurement was cut from an ingot produced by AdaptaMat, Finland. The composition of the sample was determined by energy dispersive X-ray spectroscopy as $Ni_{48.5}Mn_{30.8}Ga_{20.7}$ (at.%). Alternating current susceptibility measurement was used to determine the martensitic transformation temperatures $M_s = 306$ K, $M_f = 304$ K, $A_s = 310$ K, $A_f = 312$ K and the Curie point $T_c = 373$ K. Saturation magnetization was $M_s = 65$ Am^2/kg. The room temperature structure of the sample was that of a five-layered modulated tetragonal martensite with $a = 0.5965$ nm, $c = 0.5610$ nm. The faces of the sample were cut parallel to the {100} and {001} set of planes of one martensitic variant. After electropolishing, the final dimensions of the sample were about $6 \times 7 \times 9$ mm^3.

The experimental arrangement is shown in an inset in Fig. 2. The magnetic field and compressive stress were applied perpendicular to one another. The sample was loaded and unloaded using compressive stress in a static magnetic field of 1.15, 1, 0.8, 0.6, 0.4 or 0 T, while magnetization, stress and strain were monitored simultaneously. VCM was used for the measurement of magnetization, and a laser dilatometer was used for the strain measurement. Strain ε is defined as $\varepsilon = (l - l_0)/l_0$, where l is the measured length, l_0 is the initial length. A compressed air driven piston was used for loading, i.e. the stress was the controlled parameter rather than the strain. The stress was controlled manually and the maximum stress used was 9 MPa. Before each compression in a static field the sample was magnetized using a 1.2 T magnetic field and nearly zero stress. This, due to the existence of the magnetic shape memory effect, removed from the sample all martensitic variants apart from the one with the *c*-axis along the magnetic field. Thus, the sample was in a defined single variant state before each measurement, with the [100] direction in line with the stress and the [001] direction (easy magnetization axis) in line with the field. The presence of only one variant was confirmed by optical microscopy.

The magnetization curves of the sample containing only one variant (single variant state) were measured using the same arrangement without moving the sample. This guaranteed that the magnetization curves had approximately the same demagnetization factor as during the loading tests. The curve of the soft variant ([001] along the field) was measured at nearly zero stress. The curve of the hard variant ([100] along the field) was measured under a 10 MPa compressive stress. This stress induces the variant with the short *c*-axis along the stress, i.e. with the *c*-axis perpendicular to the magnetic field, and prevents the magnetic field from inducing other variants during the magnetization measurements. Optical observation, however, showed that some thin minor martensitic bands remained after compression. The presence of other variants can also be inferred from a slight deviation of the magnetization curve of the hard variant from linearity in a low field and a slow approach to saturation.

3. Results

Fig. 1 shows the strain and accompanying magnetization changes during compressive loading and unloading at different values of constant magnetic field. Stress-induced deformation with a large plateau is caused by martensitic variants redistribution or structure reorientation. In the initial state the sample contains only one variant with orientation [100] along the stress. The increasing compressive stress induces the nucleation and growth of a variant with [001] along the stress, resulting in large deformation and a plateau on the stress–strain curve. Optical observation shows that this variant appears as broadening parallel bands in the original variant [13]. At 9 MPa the strain reaches −5.2%. This indicates nearly full reorientation when compared with maximum theoretical strain $(c - a)/a = -5.9\%$.

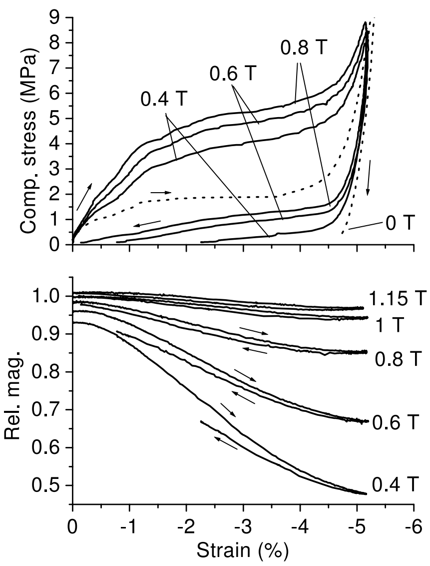

Fig. 1. Simultaneous measurement of strain (stress–strain curve) and magnetization (magnetization–strain curve) for compressive loading and unloading of the sample in various static magnetic fields and in zero field. Stress–strain curves for 1 T and 1.15 T were omitted from the figure for the sake of clarity as they were almost identical to the curve for 0.8 T.

The magnetic field applied perpendicular to the stress direction induces the nucleation and growth of the variant with orientation [0 0 1] along the field, i.e. [1 0 0] along the stress, which is identical with the initial orientation. Therefore, reversible deformation may occur during unloading in the magnetic field. Full reversibility is achieved for fields larger than 0.6 T. For 0.4 T and 0.6 T, there is only partial reversibility. The reversibility of the deformation or superelastic behavior in the field depends on twin boundary mobility and was discussed before [6] and the observed increase in the stress needed for reorientation with increasing field was interpreted using a simple energy-controlled model. Similar observations of superelastic behavior affected by a constant field were also presented in Ref. [14] and this was interpreted using a statistical model.

Concurrent magnetization changes during compressive deformation, i.e. magnetization–strain curves, are shown in Fig. 1. In the beginning, at zero stress the magnetization is maximum and it gradually decreases during loading and increases during unloading. These magnetization changes are due to the transformation of the initial variant with [0 0 1] along the field to the variant with [1 0 0] along the field during loading and reverse transformation during unloading. This transformation is also indicated by strain changes as discussed above. The magnetization changes with strain are the largest for the 0.4 T and 0.6 T fields and gradually decrease with increasing field. The initial value of magnetization is smaller for smaller fields.

Narrow hysteresis is observed in the magnetization–strain curves in agreement with previous reports [5,7]. However, these measurements sampled only local changes of magnetization in contrast with overall changes measured here. The hysteresis in the magnetization–strain curves suggests that the configuration and distribution of the variants for the same strain are different and thus structure evolution follows different paths during loading and unloading. This is partly discussed by Müllner et al. for the case of a stress–strain curve [5].

The magnetization curves of the sample with two different orientations, with [0 0 1] and [1 0 0] along the field, are shown in Fig. 2. These two orientations represent the initial state of the sample and the state under maximum compression. In the constant field, the path of magnetization during stress-induced reorientation is represented by a vertical line between the magnetization curves of these two variants, as demonstrated in Fig. 2. Using the superposition assumption, the total magnetization M is given as the sum of the magnetization of individual variants

$$M(H,\varepsilon) = M_{001}(H) - \chi(M_{001}(H) - M_{100}(H)), \quad (1)$$

where M_{001}, M_{100} are magnetizations along the [0 0 1] and [1 0 0] directions, respectively, $\chi = \varepsilon/\varepsilon_{\mathrm{MAX}}$ is a fraction of stress-induced variant where ε is measured strain and $\varepsilon_{\mathrm{MAX}}$ is the maximum strain due to reorientation. However, the observed magnetization dependencies, shown in Fig. 1, are not linear, in contrast with the nearly linear dependencies observed in experiments monitoring local changes of magnetization [5,7]. The departure of the experimental curves from the predicted linearity is due to the magnetic interaction between variants in a lamellar structure being neglected in Eq. (1). Additionally, the internal magnetic field is modified due to the varying demagnetization field in the changing geometry and morphology of lamellar structure during the reorientation.

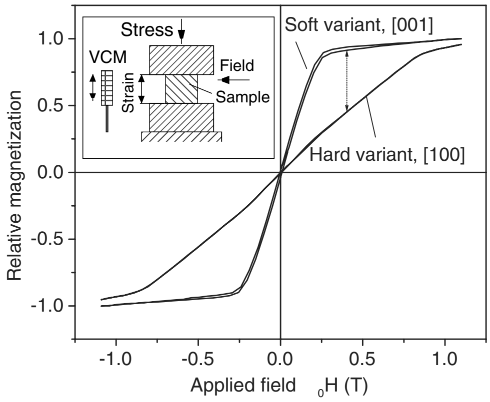

Fig. 2. Magnetization curves of hard (along [1 0 0]) and soft (along [0 0 1]) martensitic variants. Both curves were measured for exactly the same position of the sample. The saturation is not fully reached due to thin bands of residual variants in the sample and by effects of sharp corners of the sample. The path of magnetization during loading in static field is demonstrated by the vertical line between the two curves. The inset shows the experimental arrangement.

At the start of compression the deformation is elastic up to approximately 0.1% strain. In the elastic region no reorientation takes place and thus the magnetization is constant. The beginning of the decrease in magnetization signifies the start of the reorientation. The same situation occurs when the sample is fully reoriented, i.e. it contains only a single stress-induced variant. In this case, only elastic deformation occurs and the magnetization is constant. This was observed by Müllner et al. [5], but note again that they measured only local changes. However, in our case the magnetization is not constant at the maximum load. This indicates that reorientation continues and the sample is not in a single variant state (the full reorientation was not reached) even at maximum stress. This was already concluded from the value of the measured maximum strain during compression. For the annihilation of persisting bands in the whole sample volume, a much higher stress may be needed [13].

The maximum magnetization change during the loading test is determined from Fig. 1 as the difference between the initial magnetization and the magnetization of the fully loaded specimen. Fig. 3 shows a comparison of the maximum change determined in this way and the maximum change predicted from the difference of the magnetization curves of the sample containing a single martensitic variant with [0 0 1] and [1 0 0] along the field (Fig. 2). Both dependencies are in perfect agreement. The maximum magnetization change decreases when approaching saturation and must be zero for full magnetic saturation as the

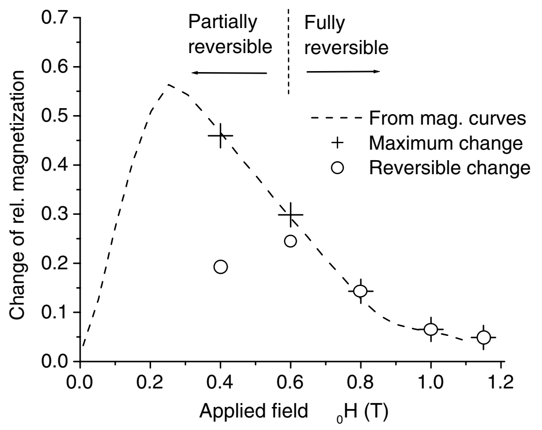

Fig. 3. Maximum changes of relative magnetization determined from loading tests presented in Fig. 1. (Crosses) compared with the prediction from difference of magnetization curves presented in Fig. 2 (dashed line). The reversible magnetization change is shown by empty circles.

magnetization is equal to the saturation value independent of the martensitic variant configuration. Therefore, the observed nonzero magnetization change at the maximum field of 1.15 T indicates that the saturation was not fully reached.

Fig. 3 shows also the reversible magnetization change determined as a difference between magnetization after unloading and magnetization of fully stressed sample. Due to partial reversibility at low fields, the reversible change is lower than the maximum change for the fields lower than 0.6 T. This field is not enough to transform the sample to initial configuration during unloading.

With regard to using the studied effect in sensor applications [4,5], one could utilize the material as a low stiffness strain sensor with high yield. From Fig. 3, it is apparent that the optimum applied field to obtain the maximum reversible magnetization change, about 30%, is well below saturation, in our case about 0.6 T. Increasing the field actually leads to a decrease in the magnetization change. In a smaller field, however, the reversibility will be lost and therefore the reversible magnetization change will also decrease. The reversible behavior depends on the twin mobility as discussed in Ref. [6,9]. Additionally, the magnetization changes in an external static field are strongly influenced by the demagnetization field and thus will depend on the sample shape and the arrangement of the magnetic circuit.

Measured functional dependence of the magnetization versus strain during variant reorientation and origin of the hysteresis in magnetization–strain curves deserves more investigation. However, the analysis is far from trivial due to the magnetic interaction between variants and demagnetization effects in the lamellar structure of variants. In addition to the presented work, we conducted several preliminary experiments monitoring magnetization during partial loading cycles, which showed similar behavior to full cycles.

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