

## Magnetic shape memory in Ni<sub>2</sub>MnGa as influenced by applied stress

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The authors report on the stress-strain and strain-temperature behaviors of a Ni<sub>2</sub>MnGa alloy. Depending on the applied stress during cooling from austenite, different levels of transformation strain were observed culminating in the formation of a single variant of martensite and transformation strain of 4%. The results underscore an optimum stress level that maximizes shape memory strains. At higher stress levels, plasticity curtails the level of transformation strain and the concomitant magnetic shape memory strains. Similarly, the results uncover an optimum bias stress to maximize magnetic induced shape memory, producing strains exceeding 5% with reversible strains of 3%. © 2006 American Institute of Physics. [DOI: 10.1063/1.2390669]

We focus on the role of externally applied stress influencing the ferromagnetic shape memory effect in the Ni<sub>2</sub>MnGa class of materials. These alloys attain a martensitic state through a thermoelastic phase transformation.<sup>1,2</sup> Depending on the composition of the material, large strains are realized due to martensite variant reorientation under an applied magnetic field. Magnetic shape memory (MSM) strains of 0.2% were first shown by Ullakko *et al.* in 1996,<sup>3</sup> and later work demonstrated strains approaching 6%.<sup>4</sup> The biasing stress permitted during MSM experiments is typically limited to 1–4 MPa, depending upon the magnetic anisotropy.<sup>5</sup> Particular alloys with high magnetic anisotropy levels produce an increased magnetic shape memory response at higher bias stress levels.<sup>6</sup> The current material is capable of overcoming a higher biasing stress compared to previous works, exceeding 7 MPa. The ability of a particular alloy to achieve magnetic field induced strains under external bias stress can be ascertained from the results of temperature cycling under constant load and isothermal uniaxial loading. We present a clear understanding of the role of stress-strain and strain-temperature responses in regard to the prediction of MSM behavior.

Single crystals of the Ni<sub>48.4</sub>Mn<sub>27.0</sub>Ga<sub>24.6</sub> at. % were grown using the Bridgman technique in an inert environment. Isothermal uniaxial loading, constant stress thermal cycling, and MSM experiments have been conducted. In the MSM experiments, the reported strains are along the [001] direction with the magnetic field applied perpendicular to a (100) plane. The load was held constant during magnetic field application via closed loop control of a MTS Tytron 250 horizontal load frame. The magnetic field was measured using a dc magnetometer manufactured by Alpha Lab, Inc. Strain measurements were attained using a capacitive sensor from Capacitec, Inc. Liquid nitrogen was forced through the specimen grips for temperature control.

To understand the MSM mechanism, consider a tetragonal martensite unit cell with a long axis  $a$  and short axes  $c$ ; the magnetic permeability of the short axes is higher.<sup>4</sup> Magnetic field applied along the long axis provides a driving force for reorientation of the martensite such that the short axis attempts to align with the magnetic field. For a specimen in a saturating magnetic field, Heczko<sup>6</sup> has shown that for twin reorientation to occur the following energy balance must hold:

$$\frac{K_u}{\varepsilon_0} \geq \sigma_{tw} + \sigma_{bias}, \quad (1)$$

where  $K_u$  is the magnetic anisotropy,  $\varepsilon_0$  is the theoretical maximum reorientation strain given by  $1-c/a$ ,  $\sigma_{tw}$  is the twinning stress, and  $\sigma_{bias}$  is the external compressive biasing stress. The values for  $K_u/\varepsilon_0$  and  $\sigma_{tw}$  are each functions of temperature. A higher magnetic anisotropy coupled with lower twinning stress would result in a material capable of overcoming a higher bias stress. Compared to a trained preferred variant structure, a self-accommodating tetragonal martensite structure has a lower effective  $K_u$ , limiting the MSM effect.

Various training methods have been suggested<sup>5,7–9</sup> to produce a martensite with a preferred orientation. A common method used for training utilizes isothermal uniaxial loading below the  $M_s$  temperature to induce a single variant preferentially oriented to the loading direction. However, the complete transition to a preferred variant, if attainable, may require loading beyond the plateau stress and can induce localized plastic deformation.<sup>10</sup> A less utilized method to induce variant orientation is the application of constant axial stress during temperature cycling. Cooling under a uniaxial compressive stress from the austenitic region will achieve a preferentially oriented martensitic state without plastic deformation, but the acquisition of a single variant is highly sensitive to the external load as demonstrated later in this work. As a consequence, this method requires the monitoring of strain-temperature results over a broad range of stress levels to ascertain single variant formation.<sup>11</sup>

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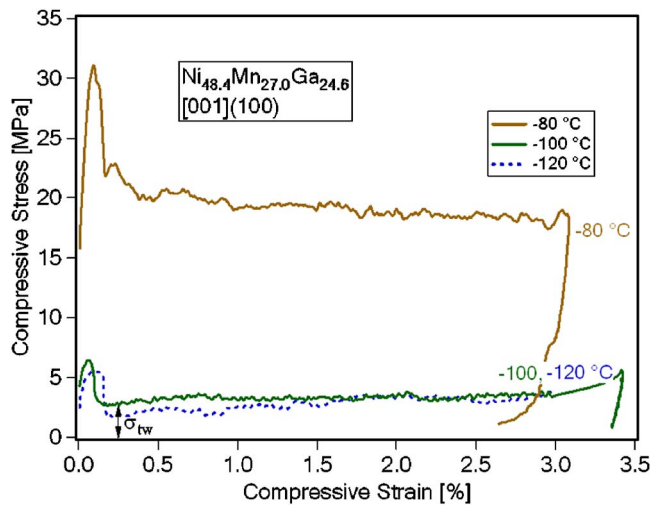


FIG. 1. (Color online) Compressive stress vs strain results for the  $\text{Ni}_{48.4}\text{Mn}_{27.0}\text{Ga}_{24.6}$  at. % alloy.

The operating temperature range for this material was determined from differential scanning calorimetry. The material examined in this work produces a five-layered tetragonal martensite with the stress-free onset of martensite formation at  $-95^\circ\text{C}$  ( $M_s$ ). The Curie temperature is approximately  $109^\circ\text{C}$  based on our differential scanning calorimetry results and is consistent with published values for similar compositions.<sup>12</sup> The large temperature difference between the Curie temperature and the testing temperature results in a high value of  $K_u/\epsilon_0$ ,<sup>13</sup> which is an indicator that the material could achieve MSM under high external bias stress.

The stress-strain response of the material is shown in Fig. 1 at several temperatures. We note that the twinning stress  $\sigma_{\text{tw}}$  is sometimes referred to as the critical stress. The twinning stress, as measured using a 0.25% strain offset, at  $-100$  and  $-120^\circ\text{C}$  are 2.7 and 1.8 MPa, respectively. This low twinning stress coupled with high anisotropy suggests a material capable of achieving MSM with a high bias stress. Notice that an initially higher stress of at least 5 MPa is reached followed by a stress drop prior to the plateau stress. A similar stress drop has been reported in the literature,<sup>14</sup> and it is indicative of a barrier stress to move the twin boundary interface. As the temperature is raised beyond  $M_s$ , the material exhibits a steep increase in stress required for martensite formation curtailing any opportunity for MSM generation at higher temperatures.

We now discuss the strain-temperature behavior prior to the application of a magnetic field. The twinning stress of 1.8 MPa (from the stress-strain curve) is lower than the stress of 6 MPa required to achieve the maximum transformation strain as shown in the strain-temperature curves given as Fig. 2. The discrepancy is attributed to the barrier stress that must be overcome prior to growth of a preferred variant. The transformation proceeds isothermally in Fig. 2, with negligible elastic strain energy storage, which implies the growth of a single interface.<sup>11</sup> The external load facilitates detwinning; hence a single crystal (or single variant) of martensite grows. Furthermore, the experimental transformation strain at saturation, nearly 4%, is very close to the theoretical saturation strain (4.1%) for austenite to single variant martensite.<sup>15</sup>

The selection of the applied stress during cooling to below  $M_f$  plays a role in the subsequent MSM response as

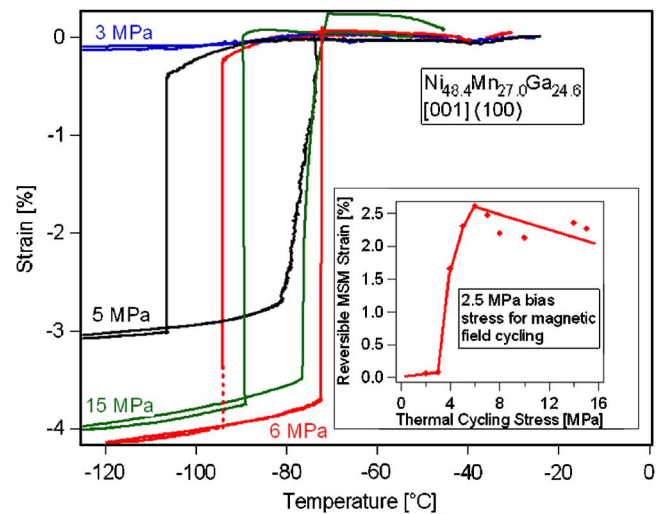


FIG. 2. (Color online) Strain-temperature results for a sample loaded under several compressive stress levels. The inset figure shows the effects of thermal cycling stress on the maximum reversible MSM strains.

shown by the inset plot in Fig. 2. If too high stress is selected, plastic deformation occurs and the resulting dislocations obstruct variant motion and reduce the MSM strains. When a small stress is applied during cooling, a volume fraction of the material remains in a self-accommodated arrangement, which lowers the effective magnetic anisotropy ( $K_u$ ) and results in small MSM strains. Therefore, the results point to the selection of an optimum stress to promote single variant formation without the plastic deformation effects.

Upon establishing that a single variant forms, without plastic accommodation, under a 6 MPa compressive stress, the MSM strains were measured (Fig. 3). We present both the peak strain and the reversible strain, which are denoted in the inset, at several biasing stress levels. The presentation of MSM results in this format is useful for benchmarking sample to sample variation, a common problem in these alloys. At low biasing stresses it is apparent from the peak strain attained that the magnetic field is able to reorient the martensite, but the biasing stress is not sufficient to promote the regrowth of the initial variant when the field is removed.

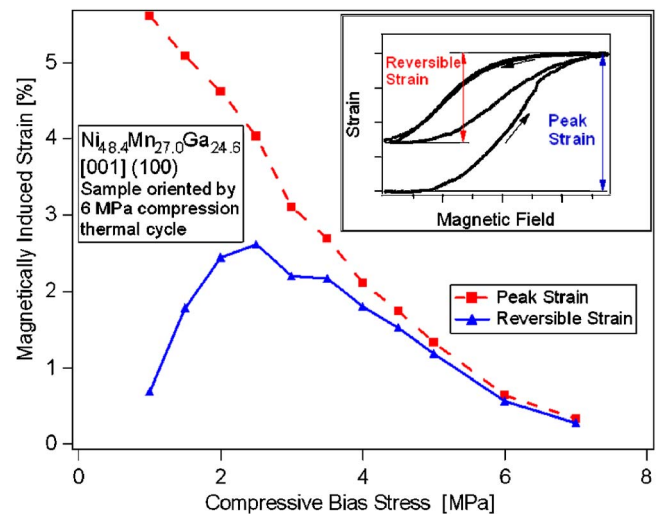


FIG. 3. (Color online) Peak and reversible MSM strain at  $-95^\circ\text{C}$  for various compressive biasing stresses. The sample was trained by 6 MPa compression thermal cycling. The blocking stress exceeds 7 MPa.

The differential between the peak strain and the recoverable strain is indicative of stabilization of a secondary martensite variant. At higher bias stress, the magnetic anisotropy no longer provides enough driving force to ensure complete reorientation of the martensite; Eq. (1) is no longer satisfied. The biasing stress required to achieve the optimum amount of MSM can be deduced from the stress-strain results. Even though the twinning stresses obtained from such results are for the reorientation of multiple variants, some correlation is evident. The twinning stress at  $-100\text{ }^{\circ}\text{C}$ , 2.7 MPa, is close to the stress of 2.5 MPa where peak MSM strains were recorded.

The blocking stress, the bias stress for which MSM strains are essentially zero, for this alloy exceeds 7 MPa at  $-95\text{ }^{\circ}\text{C}$ , still producing MSM strains of nearly 0.3%, which exceeds the highest previously reported stress<sup>5</sup> for notable MSM strains. Based on Eq. (1), the twinning stress results of Fig. 1 and the results from Ref. 13, the maximum biasing stress for MSM strains for this material would be over 8 MPa at temperatures approaching  $M_s$ .

In summary, we have shown MSM strains for a  $\text{Ni}_{48.4}\text{Mn}_{27.0}\text{Ga}_{24.6}$  at. % alloy trained to induce a single variant by the careful selection of uniaxial compressive stress applied during temperature cycling. The stress applied during this training procedure is higher than that required for twin reorientation suggested by stress-strain results. The application of excessive stress during training by constant stress thermal cycling resulted in a degradation of MSM performance. Strain-temperature curves at multiple stresses must be evaluated for proper load selection during training by con-

stant stress thermal cycling. We present conclusive evidence of single variant formation and show that the twinning stress obtained from stress-strain results is useful in determining the optimum bias stress for MSM strain production.

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