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Effect of internal stress and bias field on the transformation strain of the Heusler alloy $Ni_{52}Mn_{24.4}Ga_{23.6}$

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Abstract. The effects of internal stress and bias field on transformation strain in the Heusler alloy $Ni_{52}Mn_{24.4}Ga_{23.6}$ were investigated. It was found that both residual internal stress caused by the directional solidification during the growth and additional bias field with a direction consistent with the intrinsic preferential orientation would provide favourable conditions for exhibiting large transformation strain and magnetic-field-induced strain capability. These characteristics can be attributed to either nucleating favourable variants or increasing the volume fraction of favourable variants present through twin boundary motion.

1. Introduction

In the last decade, the Heusler alloy NiMnGa has been attracting investigation as a potential smart material and also as a new functional material. This alloy is interesting for several reasons as follows. First, it is the only known ferromagnetic intermetallic compound undergoing a martensitic transformation from a cubic $L2_1$ structure to a complex tetragonal structure. Second, associated with the martensitic transformation, it exhibits a two-way shape-memory effect [1], superelasticity [2, 3] and magnetic-field-induced strain (MFIS) [4–6]. Various micromagnetic models have been established based on experimental observation [7, 8], which indicated that the preferential orientation of martensitic variants has an important role in obtaining the large transformation strain during martensitic transformation. In the current investigations, the external stress is usually used to promote this preferential orientation and to obtain a large transformation strain for shape memory [9]. In this paper, we report a completely stress-free two-way shape memory behaviour and unusual large MFIS of 0.6% in an $Ni_{52}Mn_{24.4}Ga_{23.6}$ single crystal sample. These results indicate that the residual internal stress caused by the directional solidification during the growth or the additional bias field with a direction consistent with the intrinsic preferential orientation would provide favourable conditions to exhibiting magnetic-field-tuned strain capability.

2. Experiment

The composition of $Ni_{52}Mn_{24.4}Ga_{23.6}$ deviated from a stoichiometric Heusler alloy, Ni_2MnGa, in order to obtain the martensitic transformation temperature at about room temperature [9, 10].

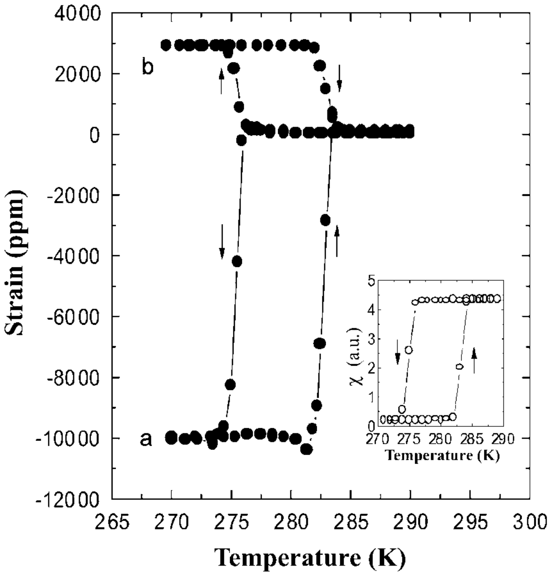

Figure 1. Strain–temperature curves of shape deformation of a single crystal $Ni_{52}Mn_{24.4}Ga_{23.6}$ free sample measured in [001] (a) and [100] (b) directions, respectively. The inset shows the temperature dependence of the ac magnetic susceptibility curve. Measurements have been conducted with an ac field of 1×10^{-3} T and a frequency of 100 Hz in an $Ni_{52}Mn_{24.4}Ga_{23.6}$ single crystal along the [001] direction.

The single crystals were grown in [001] direction according to the cubic parent phase by the Czochralski (CZ) [11, 12] instrument with a cold crucible system [13]. The starting material was prepared from metal elements Ni, Mn and Ga with purity of 99.95%. The single crystals were annealed at 1073 K for 24 hours for high chemical ordering [14]. Then the single crystals were oriented by back-reflection Laue diffraction and cut into $1 \times 1 \times 3$ mm^3 pieces for magnetic measurements by ac susceptibility, and $2 \times 9 \times 12$ mm^3 pieces with the length direction parallel to the [001] direction for strain measurements. The metal strain gauges with maximum measurement of 5% and the highly elastic epoxy resin were utilized to ensure measurement reliability and avoid the gauge debonding.

3. Results and discussion

Shown in figure 1 are the ε–T curves in the [001] and [100] directions, respectively, without an external applied bias field or a prestress. In the cooling run, one can easy see that the martensitic transformation occurs at about 276 K and causes the sample to shrink about 1% in the longitudinal [001] direction, while the sample expands about 0.3% in its lateral [100] direction. In the heating run, the reverse martensitic transformation occurs at about 282 K and the sample recovers its original shape completely. This means that the Heusler alloy $Ni_{52}Mn_{24.4}Ga_{23.6}$ exhibits a complete two-way shape memory effect with a transformation strain of 1% without the assistance of an external stress or a bias field. The temperature dependence of the low-field ac susceptibility curve is shown in the inset of figure 1. This χ–T curve also shows two sharp kinks at 276 K in the cooling run and at 282 K in the heating run, respectively, which are exact fits to the corresponding branch in the ε–T curve. It is suggested that the magnetic ordering stringently follows the crystal lattice deformation [15].

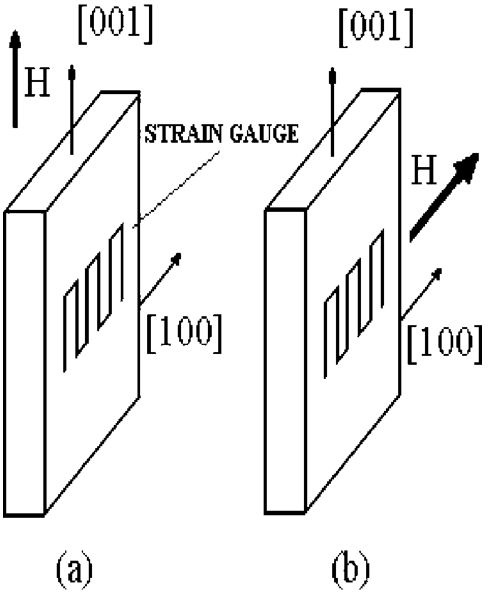

Figure 2. Orientation of the samples and the relative alignment of the metal strain gauge with the applied field.

In general, in view of the crystallography, three directions, [001], [100] and [010], are equivalent to the lattice deformation and variant orientating during the martensitic transformation. In fact, a self-accommodation effect usually occurs in familiar martensitic transformation materials to distribute the variants associated with these directions on a rough average in order to keep the free energy of system at a minimum. The transformation strains in the [001] and [100] directions would be in the same sense (shrinking or expanding mutually) and of the same value, if a complete self-accommodation really occurred in our single crystal sample. Comparing the strain curve a with b in figure 1, however, one can see the apparent difference of strain senses and magnitudes in those two directions. This implies that an extrinsic or technical factor might have been imposed on the sample, which worked as a shape-memory treatment for reducing the intrinsic self-accommodation and establishing a preferential orientation of the variants. Furthermore, it worth noting that the direction in which the shape memory has the largest strain is always the [001], namely the growth direction of the crystals, as shown by curve a in figure 1. Therefore, it is reasonable to believe that the residual internal stress caused by the directional solidification during the growth is the origin of the preferential orientation of the martensitic variants in our crystals. It is similar to some mechanical treatments, such as hot rolling or cold drawing, in order to induce imperfections or internal stress in some shape-memory alloys [16]. Up to now, experimentally, large transformation strain has always occurred in these alloys, which deviate from stoichiometry (in order to increase the imperfections in these alloys) [4, 5].

The ε–T curve in the [001] direction of the sample with the bias field was measured in this work. Figure 2 shows the orientation of samples and the relative alignment of the metal strain gauge with the bias field. The strain measurement was performed in the [001] direction, while the bias field was applied along the axial [001] direction (figure 2(a)) and the lateral [001] direction (figure 2(b)) of the sample. It has been found that the transformation strain could be enhanced up to more than threefold by a bias field of 0.8 T. As shown by curve a in

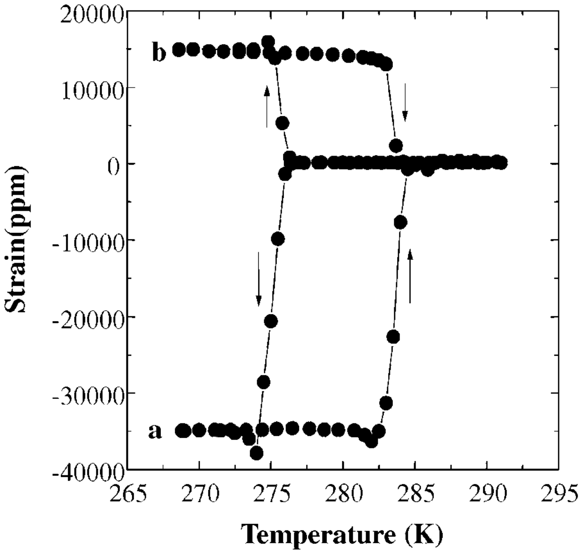

Figure 3. Strain–temperature curves measured with the bias field of 0.8 T applied in the [001] direction (a) and in the [100] direction (b), respectively.

figure 3, the transformation strain in the [001] direction increased from 1% to 3.5%, when the bias field of 0.8 T was applied in the [001] direction, consistent with the measuring direction. The net transformation strain enhanced by the field of 0.8 T is 3.5% − 1% = 2.5%, which is much larger than the transformation strain of 1% from the free sample without the bias field (curve a in figure 1). Moreover, an opposite shape deformation with a transformation strain of 1.5% (curve b in figure 3) can also be obtained by changing the bias-field direction to lateral, the [100] direction of the sample. In this way, a four-stroke shape-memory device may become possible by using a direction-alterable electrical magnet. The phenomenon may be explained as follows. Experimentally, Tickle *et al* [6] have demonstrated that the martensitic variants have strong uniaxial magnetic anisotropy and the easy axis corresponds to the *c*-axis. Therefore, the $\langle 001\rangle$ bias field should energetically favour growth of this direction variant volume fraction over the other two.

Figure 4 shows the MFIS measured in the [001] direction of the sample as a function of magnetic fields applied in the axial and lateral direction at 280 K. The unusual large MFIS, up to 0.6%, was obtained with an axial magnetic field up to 2 T (curve a in figure 4). It can be seen that the obtained MFIS was reversible and reached saturation at about 0.8 T. It is worth noting that this MFIS value measured in a lateral field was equal to that in an axial field in our crystals, but with an opposite sign, as shown by curve b in figure 4. This observation in our material further supports a simple model based on an intermediate anisotropy and the corresponding variant geometry [6]. In this model, the martensitic twin variants are orthogonal to each other and the applied field is parallel to the magnetization in one of them. Then the favourable twin variants under the applied field will grow at the expense of the others through the motion of twin boundaries. However, the reason why the twin boundary becomes reversible is not understood.

In order to find the stability of the MFIS with respect to the temperature variation, the ε–H loops were measured in the temperature range of 250–310 K. Shown in figure 5 is the saturated MFIS as a function of temperature. One can see that the maximum MFIS of 0.6%

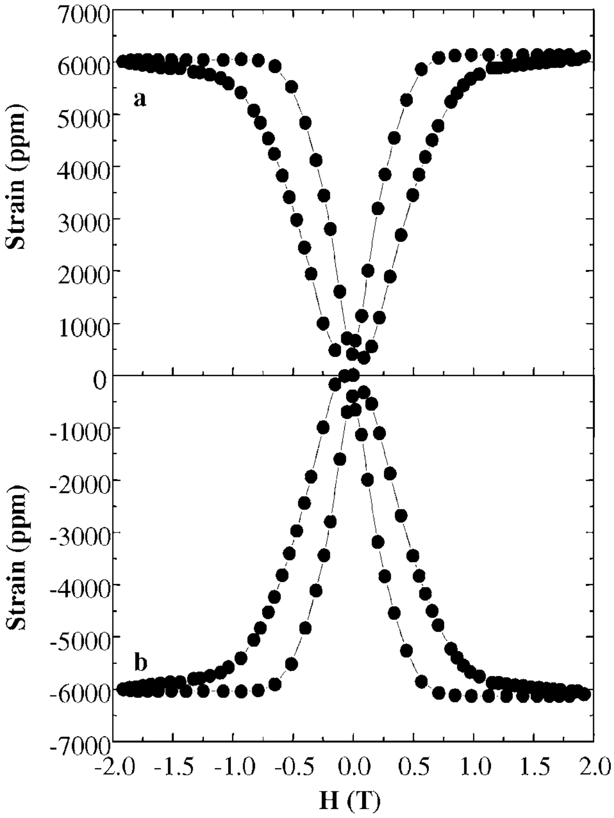

Figure 4. The MFIS measured in the [001] direction as a function of the magnetic field applied along the axial direction (a) and lateral direction (b) of the samples at 280 K.

was stabilized within a range of about 12 K from 270 to 282 K. For different transformation processes (heating or cooling), the maximum MFIS occurs in different temperature ranges and strongly correlate to the phase transition. The large MFIS temperature range is less pronounced on cooling than on heating from deep in the martensitic phase because in the latter case the material is more fully transformed and more martensitic variants would have occurred than occurred on the first cooling through the martensitic transformation temperature.

According to previous work [3], an initial prestress of about 40 MPa was needed to initiate the twin boundary motion and then 6.5 MPa more external stress was required to create a strain of 2.3%. The latter corresponds to a mechanical energy of the order of 230 kJ m^{-3}. However, we found that only 0.8 T bias field, which will provide a magnetic anisotropy energy density [4] $M_sH_a/2 \approx 117$ kJ m^{-3}, is just enough to obtain the same net strain in NiMnGa alloy. It is apparent that the magnetic field is more effective to achieve a large strain shape-memory function than the external stress in this material, due to the large difference of magnetic anisotropy between martensite and austensite phases in this material [4, 17, 18]. However, it is worth noting that both the transformation strain and the phase transformation temperature are less influenced by the bias field up to 2 T. This indicates that the mechanism of field-enhanced strain and the large MFIS in this material are the redistribution of martensitic variants through the twin boundary motion [8], and the magnetic field energy is not involved in the phase transformation.

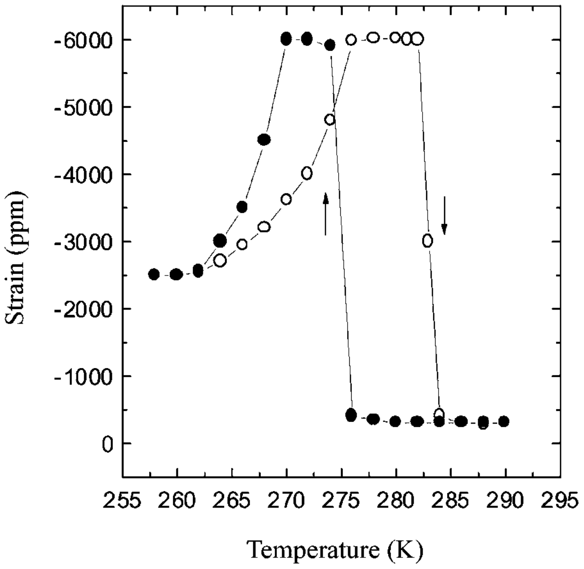

Figure 5. Saturated MFIS values as a function of temperature.

4. Conclusion

The effects of internal stress and bias field on transformation strain in the Heusler alloy $Ni_{52}Mn_{24.4}Ga_{23.6}$ were investigated. A large transformation strain of 1% was obtained in single crystal $Ni_{52}Mn_{24.4}Ga_{23.6}$ along the growth direction. The deformation can be enhanced more than threefold, up to 3.5%, with a bias field of 0.8 T applied along the growth direction. In addition, an unusual large MFIS of about 0.6% during cyclic application of the magnetic field below the martensitic transformation temperature was also obtained in this material. It is suggested that, to achieve a large deformation, the magnetic field exhibits a more evident contribution than the external stress. These characteristics can be attributed to either nucleating favourable variants or increasing the volume fraction of favourable variants present through twin boundary motion.

Acknowledgment

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