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Giant magnetic-field-induced strains in Heusler alloy NiMnGa with modified composition

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A giant magnetic-field-induced strain (MFIS) of -3100 ppm has been obtained in Heusler alloy $Ni_{52}Mn_{22.2}Ga_{25.8}$ single crystal in the [001] direction at a temperature from 23 to 31 °C. This MFIS reaches saturation in an applied field about 6 kOe, and exhibits the same amplitude with an opposite sign while the field is perpendicular to the samples. According to a previous model, this MFIS associates with the twin boundary motion. The martensitic self-strain has been found to be 2%, implying a preferential orientation of martensite variants. Results related to the magnetic properties are discussed. © *1999 American Institute of Physics.* [S0003-6951(99)02545-0]

Heusler alloy Ni_2MnGa has been systematically investigated in relation to its structure,[1,2] premartensitic transition,[3–6] and magnetic properties[7,8] for years. In an effort to find a large magnetic-field-induced strain (MFIS) for developing new actuator materials, recent studies focused on traditional ferromagnetic shape-memory (SM) materials.[9] Some materials, such as FePd (Ref. 10) or Ni_2MnGa,[11] exhibited a macrostrain larger than that of the giant magnetostrictive material of $(Tb, Dy)Fe_2$. Experiments and calculations by Ullakko *et al.*, O'Handley, and James indicated that a large MFIS could be expected, if martensite variants were preferentially oriented in these materials.[10–12] Here, we report a giant magnetic-field-induced strain up to -3150 ppm achieved in a magnetic field of about 6 kOe at room temperature. It is obtained from single crystals of Heusler alloy of $Ni_{52}Mn_{22.2}Ga_{25.8}$.

The composition of the material was modified from the stoichiometric Heusler alloy, Ni_2MnGa, in order to obtain a material with its martensitic transformation temperature near room temperature.[13] The single crystals were grown at a rate of 5–30 mm/h by a Czochralski instrument with a cold crucible system. The starting material was prepared from metal elements Ni, Mn, and Ga with purity of 99.95%. Grown crystals were annealed at 850 °C for four days and then were quenched in ice water. It is believed that the anneal and quench are important to the high degree of $L21$ structure order.[13] The single crystals were oriented by back-reflection Laue diffraction and cut into $1\times1\times3$ mm pieces for magnetic measurements, and $2\times9\times12$ mm pieces with the length direction parallel to the [001] direction for strain measurements. Figure 1 shows the orientation of samples and the relative alignment of Ni coil strain gauge with the applied field. The MFIS was measured in the [001] direction, while the field was applied along the axial [001] direction [Fig. 1(a)] and the lateral [001] direction [Fig. 1(b)] of the sample. The thermodynamic parameters of martensitic transformation were determined by measuring the axial strain as a function of temperature in the range of -30–50 °C without applied field. All strain-H loops were measured just after the sample was repeatedly magnetized up to ±20 kOe for 10 cycles at the given temperature. This procedure allows us to have repeatable measured curves.

Figure 2 shows MFIS measured in the [001] direction of the sample as a function of magnetic fields applied in the axial and lateral direction at 27 °C. The giant magnetic-field-induced strain up to -3150 ppm has been obtained with an axial applied field up to 20 kOe [Fig. 2(a)]. It can be seen that the obtained MFIS was reversible and reached saturation at about 6 kOe. Another improved result is that the MFIS value measured in the lateral field was equal to that in the axial field in our crystals, but with an opposite sign, as shown in Fig. 2(b). This is different from that measured in Ni_2MnGa, where the MFIS in the lateral field is much less than that in the axial field.[11] This observation in our material further supports a simple model based on an intermediate anisotropy and the corresponding variant geometry.[12] In this model, the martensite twin variants are orthogonal to each other and the applied field is parallel to the magnetization in one of them. On the other hand, this model has predicted a decrease in strain with an increase of the field at a very high

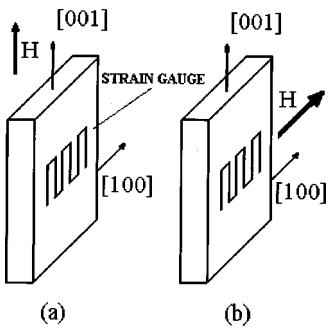

FIG. 1. Orientation of the samples and the relative alignment of Ni coil strain gauge with the applied field.

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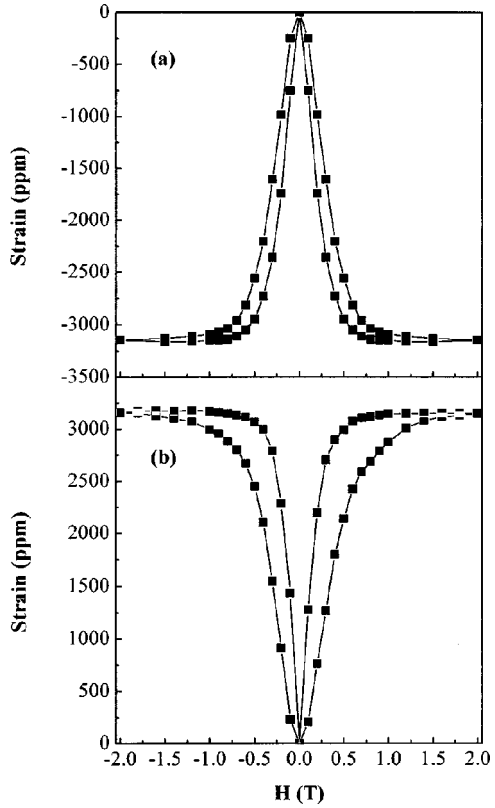

FIG. 2. MFIS measured in the [001] direction as a function of the magnetic field applied along the axial direction (a) and lateral direction (b) of the samples at 27 °C.

field due to the driving force domination from Zeeman energy becoming anisotropy energy. This decrease has not been found in this work; perhaps the field was not large enough to rotate the magnetization into the field direction significantly, but a slight increase of MFIS of about 10–30 ppm has really been observed when the applied field is turned back from the maximum value of 2 T or lower [see Fig. 2(a) and (b)]. It implies that the giant MFIS in this work is dominated by twin boundary motion. The largest magnetostriction of single-crystal $(Tb, Dy)Fe_2$ is 1640 ppm without prestress and 2375 ppm under 24 MPa prestress.[14] The exceptionally large macroscopic strain exhibited in our single crystal of $Ni_{52}Mn_{22.2}Ga_{25.8}$ is almost twice as large as that of $(Tb, Dy)Fe_2$, and this MFIS is obtained at room temperature without prestress and bias field. The large MFIS in the lateral field, which is not in $(Tb, Dy)Fe_2$, would be a superior advantage for a new type of actuator, if a bias field could be properly designed.

The detailed martensitic transformation behavior of the $Ni_{52}Mn_{22.2}Ga_{25.8}$ alloy was examined by measuring the strain in zero field while varying the temperature. The temperature for martensitic transformation and reverse transformation, M_s, M_f, A_s, and A_f, are listed in Table I. A martensitic self-strain of about 2% has been observed in the [001] direction, which is reversible: contracting the sample 2% in axial length at temperature M_s for cooling and elongating the sample in the same amplitude at A_s for heating. This martensitic self-strain is from an intrinsic shape-memory effect, based upon the nucleation and growth up of the variants. It was reported that the martensitic transformation could create about 6% *c*-axis contraction in Ni_2MnGa,[1,9] but only a martensitic self-strain of about 0.02% was observed due to self-accommodation by which the variants tend to have a random orientation for minimizing the free energy of the system.[11] However, the martensitic self-strain is 2% in our material, about a hundred times as large as that of Ni_2MnGa reaching to about one third of the lattice distortion. It strongly suggests that the martensite variants in a relative large fraction are preferentially oriented, and the degree of self-accommodation, therefore, is substantially reduced in our single crystals.

TABLE I. Thermodynamic parameters of $Ni_{52}Mn_{22.2}Ga_{25.8}$: martensite transformation start temperature M_s; martensite transformation finish temperature M_f; reverse transformation start temperature A_s; and reverse transformation start temperature A_f. ϵ stands for the martensitic self-strain (strain associated with the martensitic).

Parameter	Value
M_s	16 °C
M_f	13 °C
A_s	28 °C
A_f	31 °C
ϵ	2%

In order to know the stability of the MFIS with respect to the temperature variation, the strain-*H* loops were measured in the temperature range of −15–48 °C. Figure 3 shows the saturated MFIS as a function of temperature. One can see that the largest MFIS of −3150 ppm was stabilized within a range of about 8 °C, from 23 to 31 °C, in the reverse martensitic transformation. For different transformation processes (heating or cooling), the maximum MFIS occurs in different temperate ranges and strongly correlates to the phase transition. This result is consistent with the observation in $Ni_{50}Mn_{28}Ga_{22}$.[15]

Figure 4 shows the initial $M-H$ curves measured by a vibrating sample magnetometer with applied field along the [001] direction at 27 and 35 °C. This measured result is consistent with that of Ni_2MnGa reported by Ullakko *et al.*[11] in the curve shape, and the anisotropy for the two phases. The saturated magnetization σ_s of martensite of about 66 emu/g in our material is higher than that of Ni_2MnGa at 58 emu/g.[11] A high σ_s is very important to obtain high Zeeman energy for twin boundary motion. Therefore, the goal for our further work is an optimized composition which contains more Mn

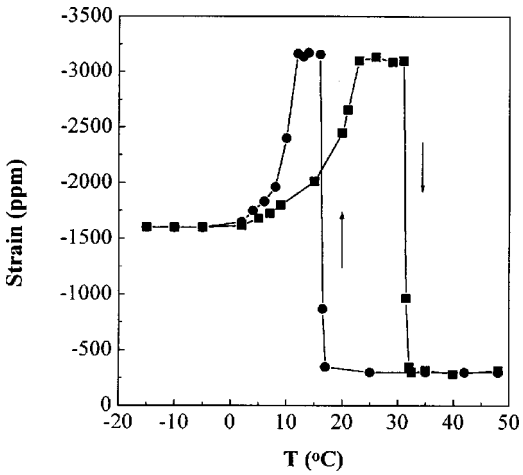

FIG. 3. Saturated MFIS as a function of temperature.

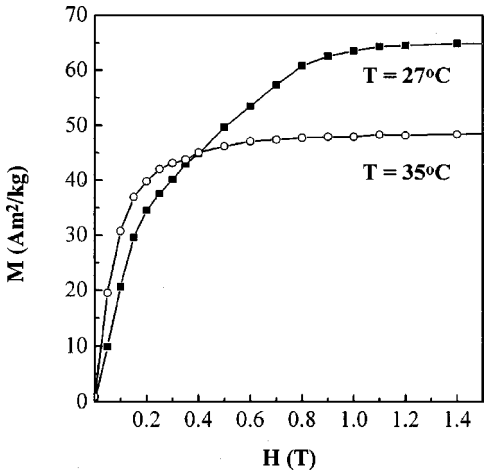

FIG. 4. Initial $M-H$ curves measured by vibrating sample magnetometer with applied field along the [001] direction at temperatures of 27 and 35 °C (heating up).

to exhibit larger σ_s, allows a greater fraction of variants to be preferentially oriented, and has better temperature stability at room temperature. It seems to be possible, because the solid-solution characteristic of the Heusler alloy NiMnGa allows the various physical properties to be modified in quite a wide composition range without its typical $L21$ structure being broken down.[9]

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