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Structural Transition and Magnetic Field Induced Strain in Ni₅₀Mn_{26.5}Ga_{23.5} Alloy

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Polycrystalline samples of Ni₅₀Mn_{26.5}Ga_{23.5} were prepared by arc-melting method. The structural transformation from tetragonal to cubic was observed from X-ray diffraction analysis as a function of temperature in the temperature range from 100 K to 370 K. Thermal magnetization curves measured at a low magnetic field of 300 Oe showed a sharp structural transformation at $T_M = 260$ K and a magnetic transition at Curie temperature $T_C = 375$ K. Thermal expansions were investigated in the vicinity of T_M after different cooling procedures: zero-field-cooling (ZFC) and field-cooling (FC). A magnetic-field-induced strain (MFIS) of ~ 0.2 % obtained from the difference between the ZFC and FC thermal expansions curves is remarkable.

Key words: *Shape memory alloy, Martensite, Austenite, Twin variants*

1. Introduction

Ferromagnetic materials constitute an emerging class of active materials because of their possibility to generate strains up to a few percent by the application of a magnetic field along specific crystallographic directions, and to control the martensitic transformation with magnetic field, temperature and stress. The mechanism is based on the magnetic-field-induced rearrangement of the crystallographic domains (twin variants). A great deal of interest has been attracted by NiMnGa alloys. Ullakko *et al.* first reported a magnetic-field-induced extensional strain (0.2%) in an un-stressed stoichiometric single crystal of Ni₂MnGa at 0.8 kOe [1]. Later, a 4.3% extensional strain was observed for stress-biased samples [2]. A shear strain of 6% at an applied field of 4 kOe was reported by Murray *et al.* in 2001 [3]. More recently, Sozinov *et al.* have succeeded in fabricating a single crystal Ni_{48.8}Mn_{29.7}Ga_{21.5} with a giant magnetic-field-induced strain of about 9.5% at an applied magnetic field of 1 T [4]. The strain proved to be caused by magnetic-

field-controlled twin boundary motion. Large magnetic-field-induced strains in single crystals have also been observed for the application of a magnetic field during cooling process across the transition. However, the extension of these results to polycrystalline samples would markedly increase the application capabilities of this class of materials. A detailed study on magnetic-field-induced strain of polycrystalline materials is still lacking. In the present work, the structural transformation in polycrystalline sample Ni₅₀Mn_{26.5}Ga_{23.5} was investigated by X-ray diffraction on changing temperature. The magnetic-field-induced strain was evaluated by investigating self-strain without applied magnetic field (ZFC) and strain when applied field of 4.2 kOe (FC) during the cooling process in the temperature range of 200 K to 300 K. Other interesting magnetic properties of the samples will also be discussed.

2. Experiment

Polycrystalline samples at a nominal composition of Ni₅₀Mn_{26.5}Ga_{23.5} were prepared by arc-melting starting from pure elements of Ni (4N), Mn (4N) and Ga (4N) in a water-cooled crucible under a pure Ar atmosphere. The resulting ingot was turned over and remelted several times to ensure the sample homogeneity.

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The ingot has been high vacuum-sealed in quartz tube and annealed at 1100 °C for ten days. Then it was quenched in ice water. A powder specimen for X-ray analysis was prepared from the same ingot, which was studied using a Phillips X'Pert X-ray diffractometer with Cu $K\alpha$ radiation. Thermal magnetization curves $M(T)$ were measured at a low magnetic field of 300 Oe on a VSM upon heating after different cooling procedures: zero field cooling (ZFC) and field cooling (FC, $H = 300$ Oe). Magnetization measurements in the temperature range of 10 K to 400 K were measured on a SQUID with the field up to 5T. Thermal expansion measurement was carried out by using the strain-gauge technique in the steady magnetic field of up to 4.2 kOe. The magnetic field was applied parallel to the measuring direction.

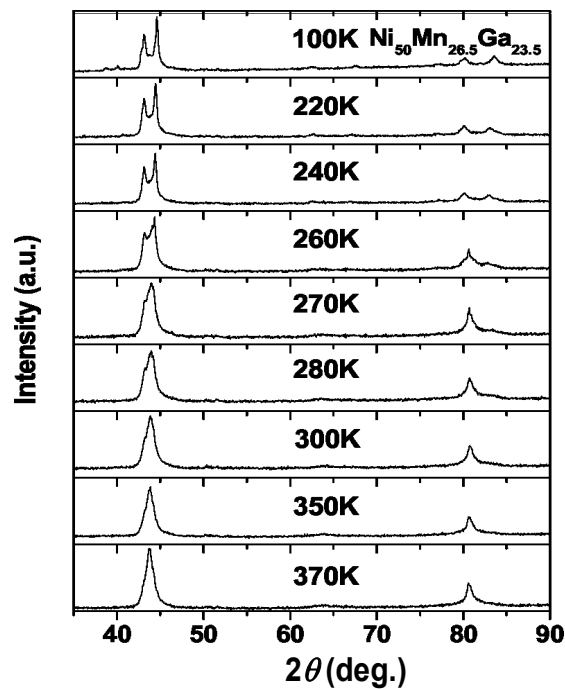


Fig. 1 X-ray diffraction patterns at different temperatures from 100 K to 370 K.

3. Results and discussion

Crystal structure of the sample at different temperature from 100 K to 370 K was analyzed by X-ray diffraction. It is clearly shown in Fig. 1 that at temperatures above 280 K, two peaks appear near $2\theta = 44^\circ$ and 81° which reveal the single phase of cubic austenite structure. At temperatures below 260 K, four peaks near $2\theta = 43^\circ, 45^\circ, 80^\circ$ and 83° were observed, indicating that a pure tetragonal martensite phase crystallizes. The cubic and the tetragonal phase co-exist in the temperature range

from 260 K to 280 K. It is clear in X-ray diffraction patterns that the structure transforms from cubic to tetragonal with decreasing temperature.

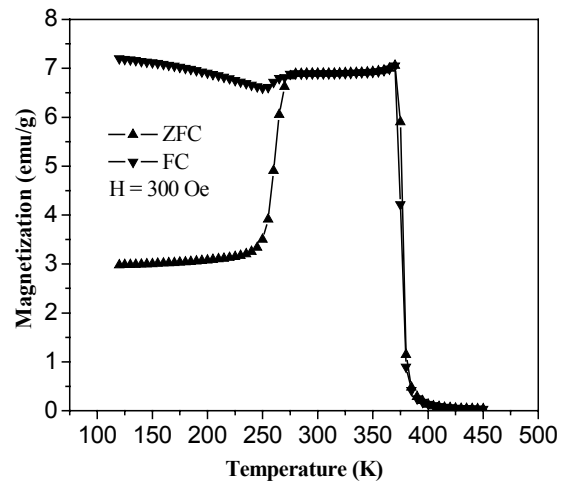


Fig. 2 The magnetization as a function of temperature measured upon heating after different cooling ZFC and FC procedures.

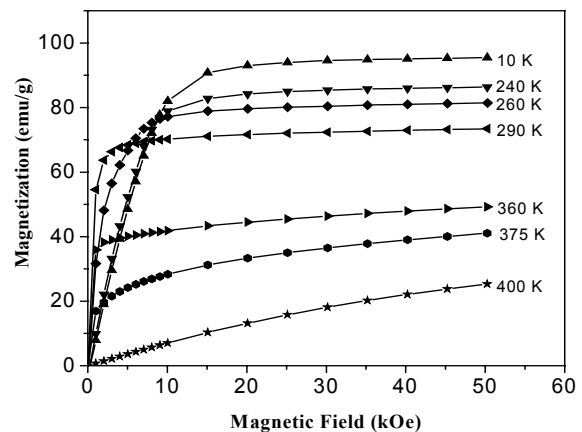


Fig. 3 The magnetization isotherms measured at different temperatures in the vicinity of transition points.

Thermal magnetization curves measured upon heating after different cooling procedures (ZFC and FC, $H = 300$ Oe) are shown in Fig. 2. Two sharp transitions from martensitic tetragonal to austenitic cubic was observed at $T_M = 260$ K and from ferromagnetic to paramagnetic at Curie temperature $T_C = 375$ K. The value of T_M is in good agreement with the X-ray diffraction analyses. The very clear separation between ZFC and FC magnetization curves below 260 K revealed that the martensitic phase has a high anisotropy. Shown in Fig. 3 is a series of magnetization isotherms at different temperatures in the

range from 10 K to 400 K. These curves manifest the whole transformation process. It is clear that at low temperature below the transition point, for example at $T = 240$ K, the magnetization is hard to saturate, which is a character of the martensitic phase, while at high temperature above the transition point, for example at $T = 290$ K, the magnetization is easy to saturate, indicating the austenitic phase. The magnetization curve at $T = 400$ K clearly shows the character of paramagnetic state.

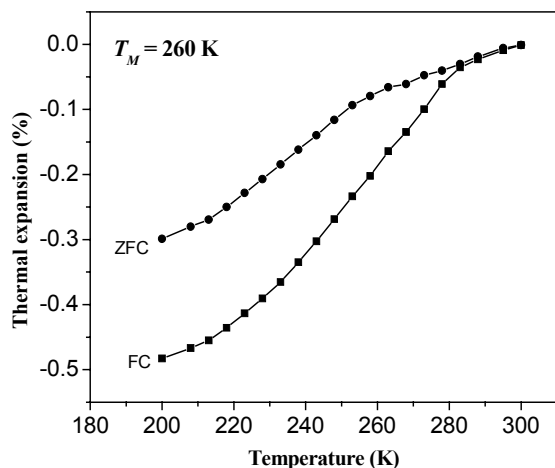


Fig. 4 Linear thermal expansion measured upon heating after different cooling procedures: ZFC ($H = 0$) and FC ($H = 4.2$ kOe).

The linear thermal expansion of the sample measured upon heating from 195 K to 300 K after different cooling procedures is shown in Fig. 4. The ZFC ($H = 0$) curve represents the spontaneous linear thermal expansion after the sample was zero-field cooled. The change in $\Delta l/l$ is associated with the martensitic transformation. The application of a magnetic field when cooling the sample through T_M gives a dramatic effect on the thermal expansion, as observed in the FC curve (FC at 4.2 kOe, field applied parallel to the measuring direction). In this case, the change in $\Delta l/l$ is two times larger. These results suggest that the applied magnetic field facilitates the growth of specific orientation variants along the field direction as the sample is cooled down through the martensitic transition. This preferential orientation takes place to minimize the Zeeman energy, favoring the growth of variants with the easy magnetization direction parallel to the applied magnetic field. The value of $\Delta l/l$ is negative. The sample contracted corresponds to the applied field direction, indicating that in the tetragonal martensitic phase the preferential

orientation, which is parallel to the applied magnetic field, is the c axis (shorter than the cubic axis).

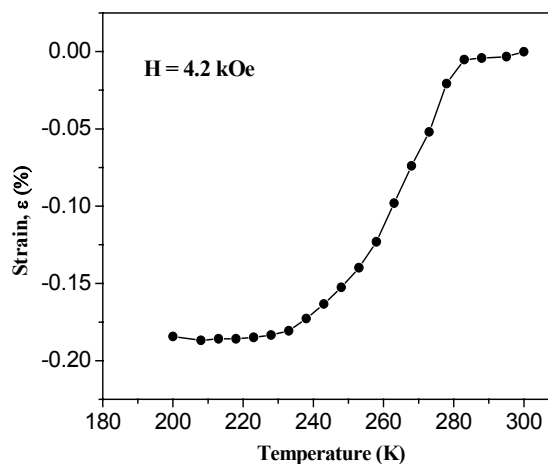


Fig. 5 Magnetic-field-induced strain parallel to the applied magnetic field of $\text{Ni}_{50}\text{Mn}_{26.5}\text{Ga}_{23.5}$ alloy obtained upon heating after FC in $H = 4.2$ kOe.

The magnetic-field-induced strain (MFIS) was obtained from the difference between the linear thermal expansion at an applied magnetic field of 4.2 kOe and zero fields. The result is shown in Fig. 5. The MFIS of about 0.2 % is remarkable for polycrystalline samples. The temperature behavior observed in the MFIS indicates that the major effect of the magnetic field occurred upon cooling through the martensitic transformation.

In conclusion, the very clear picture of structural martensitic transformation process was observed by the X-ray diffraction at different temperatures through the transition point. Field cooling of 4.2 kOe for the sample across the transformation temperature brought about a MFIS of ~ 0.2 %, which value is significantly large for polycrystalline samples.

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