Magnetocrystalline anisotropy and twinning stress in Ni-Mn-Ga ferromagnetic shape memory alloys

N Okamoto, T Fukuda and T Kakeshita
Department of Materials Science and Engineering, Osaka University,
2-1 Yamadaoka, Suita, Osaka 565-0871, Japan
E-mail: fukuda@mat.eng.osaka-u.ac.jp

Abstract. The condition for the rearrangement of martensite variant (RMV) by magnetic field has been studied quantitatively in a wide temperature range for two types of alloys: a Ni$_{2}$MnGa with 10M martensite and a Ni$_{2.02}$Mn$_{1.09}$Ga$_{0.89}$ with 14M martensite. The RMV by magnetic field occurs at any temperature for the 10M martensite but only in a limited temperature range for the 14M martensite. The magnetic field at which the RMV starts to occur, $H_s$, increases with decreasing temperature for both alloys. The condition for the RMV by magnetic field is explained by two shear stresses: the magnetic shear stress acting across twinning plane $\tau_{\text{mag}}$ (which is evaluated as the difference in magnetic energy between variants divided by twinning shear) and the shear stress required for twinning plane movement $\tau_{\text{req}}$. That is, the RMV by magnetic field starts to occur when $\tau_{\text{mag}}$ exceeds $\tau_{\text{req}}$ regardless of temperature and structure of martensite.

1. Introduction
Some Ni-Mn-Ga ferromagnetic shape memory alloys exhibit a large magnetic field-induced strain of several percent due to rearrangement of martensite variant (RMV) [1-3]. When a magnetic field is applied to the martensite of these alloys, the martensite variant which reduces the magnetic energy most grows consuming other variants by twinning plane movement. The magnetic energy difference arises mainly due to a large magnetocrystalline anisotropy [4, 5]. Since the RMV by an external stress is usually discussed on the basis of the shear stress acting across twinning plane, we consider that the RMV by magnetic field is also caused by a magnetic shear stress $\tau_{\text{mag}}$, which acts across twinning plane due to the magnetic energy difference. That is, we consider that the RMV by magnetic field is explained by comparing $\tau_{\text{mag}}$ with the shear stress required for twinning plane movement $\tau_{\text{req}}$. In terms of $\tau_{\text{mag}}$ and $\tau_{\text{req}}$, the RMV by magnetic field will occur when $\tau_{\text{mag}}$ exceeds $\tau_{\text{req}}$.

The value of $\tau_{\text{mag}}$ will be evaluated by $\Delta U_{\text{mag}}/s$, where $\Delta U_{\text{mag}}$ is the magnetic energy difference per unit volume between the variants and $s$ is the twinning shear obtained from lattice parameters. The value of $\Delta U_{\text{mag}}$ will be evaluated by the sum of Zeeman energy and magnetocrystalline anisotropy energy. In the case that the magnetic field is applied along the [001] direction (subscript “P” means a parent phase), $\Delta U_{\text{mag}}$ will be expressed as $\Delta U_{\text{mag}}=M\langle H-(M/4K_u)H\rangle$, where $H$ is the magnetic field strength, $M$ is the spontaneous magnetization and $K_u$ is the uniaxial magnetocrystalline anisotropy constant. Thus, we can evaluate $\tau_{\text{mag}}$ at any field, when $M$ and $K_u$ are obtained. In particular, the value of $\tau_{\text{mag}}$ when the RMV initiates can be obtained by evaluating the field strength at which the RMV

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starts, $H$. As mentioned above, $\tau_{\text{mag}}$ at $H_s$ should agree with $\tau_{\text{req}}$ if the process of the RMV by magnetic field is equivalent to that by mechanical stress.

In the present report, we will demonstrate that $\tau_{\text{mag}}$ at $H_s$ actually agrees with $\tau_{\text{req}}$ for Ni-Mn-Ga alloys. We will first show $H_s$ and $K_u$ as a function of temperature. Then the value of $\tau_{\text{mag}}$ at $H_s$ will be calculated. The calculated value of $\tau_{\text{mag}}$ at $H_s$ is then compared with $\tau_{\text{req}}$, which is obtained by compressive tests.

2. Experimental procedure
Alloys used in the present study are Ni$_{1.5}$MnGa with 10M (five-layered) martensite and Ni$_{1.02}$Mn$_{1.09}$Ga$_{0.89}$ with 14M (seven-layered) martensite. Single crystalline rods were prepared by a floating zone method. The rods are subjected to a homogenization-heat-treatment at 1073 K (Ni$_{1.5}$MnGa) and 1373 K (Ni$_{1.02}$Mn$_{1.09}$Ga$_{0.89}$). All specimens are cut from the rods followed by an ordering-heat-treatment at 923 K. The martensitic transformation behavior of each alloy has been confirmed to be the following: Ni$_{1.5}$MnGa transforms to the 10M martensite ($c/a=0.94$) at 202 K and Ni$_{1.02}$Mn$_{1.09}$Ga$_{0.89}$ transforms to the 14M martensite ($c/a=0.92$) at 309 K. From the lattice parameters, the value of $s$ is evaluated to be about 0.12 for the 10M martensite and about 0.16 for the 14M martensite. The temperature variation of $s$ is small according to a previous report [7].

In order to obtain $H_s$, the magnetization measurements have been made along the [001]$_p$ direction without applying external stress. The specimens used for the measurements are parallelepiped ones with 2 mm cubic and all faces being parallel to the [001]$_p$.

Magnetization measurements along the easy and the hard axes are made to evaluate $K_u$. In the measurement, a compressive stress of about 15 MPa was applied along the [001]$_p$ direction to keep a single variant state.

Compressive tests were made to evaluate $\tau_{\text{req}}$ by using parallelepiped specimens. The longest edge of each specimen is parallel to the [001]$_p$ direction and the dimensions are $2.3 \times 2.3 \times 7.3$ mm$^3$ for Ni$_{1.5}$MnGa and $1.8 \times 2.0 \times 7.8$ mm$^3$ for Ni$_{1.02}$Mn$_{1.09}$Ga$_{0.89}$. The specimens are compressed along the [001]$_p$ direction with a constant cross head speed of 0.04 mm/min.

3. Results and discussion
Figure 1(a) shows a magnetization curve at 4.2 K for the 10M martensite. The measurement was made without applying external stress. In the field applying process, a sharp increase in magnetization appears at the field shown by an arrow. This sharp increase in magnetization is due to the RMV, and the magnetization has a large hysteresis due to the RMV. Almost the same magnetization curve is obtained for the 14M martensite at 300 K as shown in Figure 1(b). We repeated the same measurements in a wide temperature range for both martensites. As a result, similar magnetization curves (showing the RMV) are obtained at any temperature for the 10M martensite but only in a limited temperature range of above 280 K for the 14M martensite. That is, the RMV by magnetic field occurs at any temperature for the 10M martensite but only in the limited temperature range for the 14M martensite. We have evaluated the effective magnetic field strength at which the RMV starts to occur, $H_s$, from Figure 1 by subtracting demagnetizing field. The evaluated value of $H_s$ increases almost linearly with decreasing temperature for both martensites, being in good agreement with a previous report [8].

In order to evaluate $K_u$, the magnetization measurements along the easy and the hard axes have been made. In the measurement, the specimen is compressed to keep a single variant. An example of the measurements for the 10M martensite is shown in the inset of Figure 2(a). The value of $K_u$ is evaluated by the area enclosed by the two curves, and its temperature dependence is shown in Figure 2(a). The value is 430 kJ/m$^3$ at 4.2 K, and it decreases monotonically with increasing temperature. Similar results are obtained for the 14M martensite as shown in Figure 2(b). It should be noted that the value of $K_u$ for the 10M martensite in the present stoichiometric Ni$_{1.5}$MnGa alloy is larger than that for the 10M and 14M martensites in off-stoichiometric Ni-Mn-Ga alloys reported so far.
By substituting the value of $H_s$ and $K_u$ into the formula shown before, we have calculated $\tau_{mag}$ at $H_s$ and its temperature dependence for the 10M martensite is shown in Figure 3(a) by open circles. As known from the figure, the value of $\tau_{mag}$ at $H_s$ decreases with increasing temperature. Similar result is obtained for the 14M martensite as shown in Figure 3(b). In order to compare $\tau_{mag}$ at $H_s$ with $\tau_{req}$, the value of $\tau_{req}$ is evaluated by compressive tests for the 10M and 14M martensites. In the stress-strain curves obtained by the tests, a clear stage due to the twinning plane movement has been observed at a stress level of several MPa. Considering Schmidt factor of about 1/2, the value of $\tau_{req}$ has been evaluated. Temperature dependences of $\tau_{req}$ for the 10M and 14M martensites thus obtained are shown by closed squares in Figure 3(a) and (b), respectively. We notice in the figure that the value of $\tau_{req}$ increases almost linearly with decreasing temperature for both martensites. This behavior is different.
from the previous report for 10M martensite in off-stoichiometric Ni-Mn-Ga [9]. Comparing \( \tau_{\text{mag}} \) at \( H_s \) with \( \tau_{\text{req}} \), we know that the value of \( \tau_{\text{mag}} \) at \( H_s \) is almost equivalent to \( \tau_{\text{req}} \) at any temperature at which the RMV by magnetic field occurs. Therefore, we confirmed quantitatively that the RMV by magnetic field occurs when \( \tau_{\text{mag}} \) exceeds \( \tau_{\text{req}} \) regardless of temperature and the structure of martensite.

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