Stress-assisted magnetic-field-induced strain in Ni-Fe-Ga-Co ferromagnetic shape memory alloys

<table>
<thead>
<tr>
<th>著者</th>
<th>石田 清仁</th>
</tr>
</thead>
<tbody>
<tr>
<td>作者姓名</td>
<td>石田 清仁</td>
</tr>
<tr>
<td>研究分野</td>
<td>超硬合金</td>
</tr>
<tr>
<td>研究目的</td>
<td>磁場効果を利用した形状記憶合金の研究</td>
</tr>
<tr>
<td>研究内容</td>
<td>鉄磁性形状記憶合金の磁場効果についての研究</td>
</tr>
<tr>
<td>使用材料</td>
<td>Ni-Fe-Ga-Co</td>
</tr>
<tr>
<td>研究方法</td>
<td>電気抵抗法</td>
</tr>
<tr>
<td>研究結果</td>
<td>磁場効果により形状記憶効果が向上する</td>
</tr>
<tr>
<td>研究成果</td>
<td>磁場効果を利用した新形状記憶合金の開発</td>
</tr>
<tr>
<td>研究意義</td>
<td>新材料開発への可能性</td>
</tr>
<tr>
<td>URL</td>
<td><a href="http://hdl.handle.net/10097/34896">http://hdl.handle.net/10097/34896</a></td>
</tr>
</tbody>
</table>
Stress-assisted magnetic-field-induced strain in Ni–Fe–Ga–Co ferromagnetic shape memory alloys

H. Morito, a A. Fujita, K. Oikawa, and K. Ishida
Department of Materials Science, Graduate School of Engineering, Tohoku University, Aoba-yama 6-6-02, Sendai 980-8579, Japan
K. Fukamichi and R. Kainuma
Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, Katahira 2-1-1, Sendai 980-8577, Japan

(Received 9 November 2006; accepted 8 January 2007; published online 5 February 2007)

To obtain a large strain for Ni–Fe–Ga ferromagnetic shape memory alloys, the Curie temperature was increased by adding Co, and the magnetic-field-induced strain (MFIS) has been investigated under static stresses. The magnetocrystalline anisotropy constant $K$ is increased by the addition of Co, and the Ni$_{49}$Fe$_{18}$Ga$_{27}$Co$_{6}$ alloy gives a relatively large value of 1.15 $\times$ 10$^6$ erg/cm$^3$ at 300 K. From the stress-strain curves for this alloy, the twinning stress is estimated to be 8–9 MPa. Consequently, the Ni$_{49}$Fe$_{18}$Ga$_{27}$Co$_{6}$ alloy exhibits a large MFIS of about 8.5% at room temperature under a static compressive stress of about 8 MPa. © 2007 American Institute of Physics. [DOI: 10.1063/1.2450667]

Ferromagnetic shape memory alloys (FSMAs) having a large magnetic-field-induced strain (MFIS) have attracted much attention, because they are promising as magnetic-controlled actuators and sensors. Large MFISs have been observed in martensite phases of Ni$_2$MnGa alloys and other FSMAs. However, intrinsic high brittleness of Ni$_2$MnGa is a disadvantage for practical applications. Therefore, developments of ferromagnetic shape memory alloys with excellent mechanical properties are strongly expected.

The MFIS is explained by the rearrangement of martensite variants due to an external magnetic field. When the magnetocrystalline anisotropy energy is larger than the energy driving variant boundaries, the angle between magnetization and the applied magnetic field directions is lowered by not only the independent rotation of magnetization but also the variant rearrangement in order that the magnetic easy axis is aligned parallel to the magnetic field direction. Since the MFIS is caused by the reorientation of the martensite variants, the behavior of MFIS is affected by magnetocrystalline anisotropy as well as mobility of variant boundaries. The magnetocrystalline anisotropy energy is required to be larger than the energy to drive the variant boundaries.

Recently, Ni–Fe–Ga Heusler-type alloys have drawn attention as practical FSMAs. It has been reported that the ductility of Co–Ni–Al alloy can be increased by introducing a small amount of γ(Al) phase. In a similar way, therefore, the Ni–Fe–Ga system is expected to have excellent mechanical properties. However, in the ternary Ni–Fe–Ga alloys, the magnetocrystalline anisotropy constant in the vicinity of room temperature is not so large because of a low Curie temperature. To overcome such a practical problem, the increase of the Curie temperature has been searched by adding Co. As a result, the Curie temperature is effectively increased and hence the single-variant martensite phase of Ni$_{52}$Fe$_{18}$Ga$_{27}$Co$_{3}$ exhibits a large MFIS of about 0.7% at 300 K. The reason why the magnitude of the present MFIS is smaller than the 10% strain of Ni$_2$MnGa (Ref. 2) is that the magnetocrystalline anisotropy constant is still small in Ni$_{52}$Fe$_{18}$Ga$_{27}$Co$_{3}$.

To obtain a large MFIS in the vicinity of room temperature, in the present letter, we have tried to increase the magnetocrystalline anisotropy constant by controlling the Co concentration and then investigated the MFIS under static compressive stresses. The single crystals of Ni$_{55−x}$Fe$_{18}$Ga$_{27}$Co$_x$ ($x=3−6$) were grown by an optical floating-zone method under a helium atmosphere. The single crystal specimens were annealed at 1433–1473 K for 2 days to homogenize and followed by quenching in ice water. After the homogenization, they were additionally heat treated at 873 K for 1–3 days to make the antiphase domain large and heat treated at 673 K for 1 day to achieve a high degree of atomic order in the Heusler structure. Although the martensitic transformation starting temperature $T_s$ decreases from 331 to 300 K with increasing Co content $x$ from 3 to 6, the Ni$_{55−x}$Fe$_{18}$Ga$_{27}$Co$_x$ ($x=3−6$) single crystal samples are in a martensite phase at room temperature. The crystallographic orientations were determined from the electron backscattering diffraction patterns. The cubic specimens in the parent phase were trimmed so that the (100)$_P$ (P: parent) directions are parallel to the faces. The (100)$_P$ $a$ axis in the $L_1$ cubic parent phase corresponds to either the (110)$_M$ or the $2 \times (001)_M$ (M: martensite) $c$ axis in the $L_1$ tetragonal martensite phase. In order to obtain the single-variant specimen, uniaxial compressive stresses were applied to the [010]$_P$ and [001]$_P$ directions in the martensite phase of the above-mentioned single crystals. After applying compressive stress, the $c$ axis of the $L_1$ unit cell is oriented to the [100]$_P$ direction. The magnetization was measured up to 20 kOe with a superconducting quantum interference device magnetometer. The MFIS was measured with an extensometer under a compressive stress in magnetic fields.

The magnetocrystalline anisotropy constant $K$ in the single-variant specimen was determined from the magnetization curves measured along the $c$ axis and $p$ plane of the $L_1$ structure. The magnetization curves at $T=300$ K for the Ni$_{55−x}$Fe$_{18}$Ga$_{27}$Co$_x$ alloys ($x=3−6$) are shown in Fig. 1. The
The concentration dependences of magnetization for the single-variant martensite phase in the Ni$_{55-x}$Fe$_{18}$Ga$_{27}$Co$_{x}$ ($x = 3$–$6$) alloys are shown in Fig. 2. The saturation magnetization $M_{\text{sat}}$ and the magnetocrystraline anisotropy constant $K$ at $300 \, \text{K}$ for the Ni$_{55-x}$Fe$_{18}$Ga$_{27}$Co$_{x}$ ($x = 3$–$6$) alloys are presented in Fig. 3. The reverse transformation starting temperature $T_C$ is $303 \, \text{K}$; therefore, the sample is in the martensite phase at room temperature in the martensite phase for the Ni$_{49}$Fe$_{18}$Ga$_{27}$Co$_{6}$ alloy. Although the compressive stresses were applied to the martensite phase, the expression of crystal directions is related to that of the parent phase. In the first cycle, the compressive stress was applied along the [001]$_p$ direction. The variant rearrangement was initiated above the applied twinning stress $\sigma_{\text{tw}}$ of $10$–$14 \, \text{MPa}$, which corresponds to the plateau, and the obtained twinning strain $\epsilon$ is about $4\%$ in the first cycle. In the second cycle, the compressive stress was applied along the [010]$_p$ direction. The variant rearrangement takes place at a much lower stress of $8$–$10 \, \text{MPa}$. Furthermore, the value of the remaining strain is increased up to $10\%$ and the sample becomes in a single-variant state. For the evaluation of the variant boundary mobility, the compressive stress was applied along the [100]$_p$ direction of the single-variant sample. Stress of $8$–$9 \, \text{MPa}$ for the variant rearrangement and strain of $15\%$ are achieved in the third cycle. The value of $K$ is exactly equal to the magnetic driving force applied to the variants; therefore, $K$ has to exceed or be of the same order as the mechanical energy $\epsilon \sigma_{\text{tw}}$ consumed for the variant rearrangement. From the x-ray diffraction, it was confirmed that the $14M$ and nonmodulated $L1_0$ structures coexist in Ni$_{49}$Fe$_{18}$Ga$_{27}$Co$_{6}$ alloy. However, under the stress, the $L1_0$ structure becomes more stable, and hence the high twinning stress cuts off the appearance of the large MFIS.

A static stress aiding the magnetic field was used to achieve a large MFIS, that is, the magnetic field was applied under a compressive stress. Before applying the magnetic field, compressive stress of about $8 \, \text{MPa}$ was applied along the $c$ axis ([100]$_p$) direction which is the hard magnetic direction. As a result, shrinkage of $5\%$ was observed as seen in Fig. 4(a). The magnetic field was then applied parallel to the $c$ axis direction. As demonstrated in Fig. 4(b), during the application of the magnetic field, a steep shrinkage caused by the variant rearrangement due to the magnetization rotation is observed around $H = 4 \, \text{kOe}$, and the maximal strain reaches $8.5\%$ in a field of $10 \, \text{kOe}$. After applying the magnetic field, the easy plane in the variant is aligned parallel to the magnetic field direction and the sample becomes in a near single-variant state with the easy plane along the field direction. By applying a uniaxial stress along the [100]$_p$ direction, the transformation always occurred from the single-variant to the multivariant state because the $c$ axis of the martensite phase orients along the two equivalent directions, i.e., [010]$_p$ and [001]$_p$. In the present alloy system, however,
the transformation occurs from the single-variant to the other single-variant state. The detail mechanism is not clear at the moment, but a preferential nucleation of one variant takes place in the present alloys. The variant rearrangement driven by the magnetic field is discussed quantitatively by introducing the magnetic shear stress \( \tau_{\text{mag}} \) acting across the twinning plane.\(^2\) For the variant rearrangement by applying a magnetic field, the value of \( \tau_{\text{mag}} \) should be larger than the mechanical shear stress \( \tau_{\text{req}} \) required for the variant rearrangement. The value of \( \tau_{\text{mag}} \) is expressed as \( |K|/s \), where \( s \) is the corresponding twinning shear. The twinning plane is \( \{101\}_p \) for the Ni–Fe–Ga–Co alloys; therefore, the value of twinning shear is estimated from \( s = (1 - (c/a)^2)/(c/a) \). Then, the value of \( s \) is calculated from the lattice parameters.\(^7,18\) For the Ni\(_{49}\)Fe\(_{18}\)Ga\(_{27}\)Co\(_6\) alloy, the value of \( \tau_{\text{mag}} \) is evaluated as \( \tau_{\text{mag}} = 0.3 \) MPa. Taking into account the twinning plane and the Schmid factor, \( \tau_{\text{req}} = 0.5\sigma_{\text{tw}} \) is estimated to be 4.0–4.5 MPa from the stress-strain curves. In this case, shear stress of \( \tau = 4 \) MPa was applied before applying a magnetic field. Therefore, \( \tau_{\text{req}} \) is decreased to \( \tau_{\text{req}} < 0.5 \) MPa, which corresponds to the plateau slope in the stress-strain curve. By applying a magnetic field, magnetic shear stress of about 0.3 MPa is added, and the mechanical shear stress, equivalent to the variant rearrangement, is obtained.

In conclusion, the increase of the Curie temperature \( T_C \) by the Co addition effectively increases the magneto-crystalline anisotropy constant \( K \). The value of \( K \) for the present Ni\(_{49}\)Fe\(_{18}\)Ga\(_{27}\)Co\(_6\) alloy at \( T = 300 \) K is evaluated to be \( 1.15 \times 10^6 \) erg/cm\(^3\), being nearly equivalent to that of Ni\(_{51}\)MnGa alloy.\(^8\) From these results, the magneto-crystalline anisotropy energy is enhanced by adjusting the concentration of Co in the Ni–Fe–Ga alloys. In the Ni\(_{49}\)Fe\(_{18}\)Ga\(_{27}\)Co\(_6\) alloy, the variant rearrangement associated with the magnetization rotation was observed, and the maximal strain of 8.5% was confirmed under a compressive stress of about 8 MPa. From these results, it has been revealed that the present Ni–Fe–Ga–Co ferromagnetic shape memory alloys are promising as large MFISs assisted by static compressive stresses.

The authors wish to thank Y. Sutou, T. Ota, and T. Takagi for their experimental support. A part of the present study was supported by the Grant-in-Aids for Core Research for Evolutional Science and Technology (CREST), Scientific Research from the Ministry of Education, Science, Sports and Culture, Japan, and the Research Fellow of the Japan Society.