Rearrangement of crystallographic domains driven by magnetic field in ferromagnetic Ni$_2$MnGa and antiferromagnetic CoO

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Rearrangement of crystallographic domains driven by magnetic field in ferromagnetic Ni$_2$MnGa and antiferromagnetic CoO

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Abstract. We have investigated the rearrangement of crystallographic domains (martensite variants) in Ni$_2$MnGa ferromagnetic shape memory alloy and CoO antiferromagnetic oxide by applying magnetic field up to 8.0 MA/m. From the result of optical microscope observation of Ni$_2$MnGa single crystal, when a magnetic field is applied along [001]$_p$ (p represents a parent phase), the rearrangement of crystallographic domains occurs and the single domain state is obtained below $T_{Ms} = 202$ K. The same rearrangement occurs but partially when a magnetic field is applied along [110]$_p$. On the other hand, when a magnetic field is applied along [111]$_p$, the rearrangement does not occur. In case of the CoO single crystal, when a magnetic field is applied along [001]$_p$ below $T_{Ms} = 293$ K, the rearrangement occurs at $170 \leq T \leq 293$ K, but does not occur at $T < 170$ K. When a magnetic field is applied along [110]$_p$ and [111]$_p$, the rearrangement does not occur below $T_{Ms}$. In order to explain the rearrangement in the alloy and the oxide, we have evaluated the magnetic shear stress, $\tau_{mag}$, which is derived from the difference in magnetic energy among crystallographic domains and have compared it with the shear stress required for the twinning plane movement, $\tau_{req}$. As a result, we have found that the rearrangement occurs when the value of $\tau_{mag}$ is larger than or equal to the value of $\tau_{req}$ for the present alloy and oxide.

1. Introduction

Recently, it has been found that a large strain of several percent appears in some ferromagnetic shape memory alloys by applying magnetic field$^{1-6}$. This large magnetic field-induced strain in ferromagnetic shape memory alloys is not due to conventional magnetostriction, but due to the rearrangement of crystallographic domain (martensite variant) of the martensite phase under a magnetic field. This phenomenon is of quite interest because magnetic field influences not only its intensive variable of magnetization but also another variable of strain. Thus, in a sense, a ferromagnetic shape memory alloy is a kind of multi-ferroic materials. Concerning the condition for the occurrence of such rearrangement, we have discussed about the energy evaluation and derived the condition for the occurrence of the rearrangement of crystallographic domains driven by magnetic field, as described below. That is, we introduce a shear stress acting across a twinning plane driven by magnetic field (We call it a magnetic shear stress), $\tau_{mag}$, considering the fact that the twinning plane movement certainly occurs by magnetic field$^{3,4}$. Using the value of $\tau_{mag}$, the condition is that the value of $\tau_{mag}$ is equal to or larger than the shear stress required for the twinning plane movement, $\tau_{req}$. The value of $\tau_{mag}$ is expressed as...
\[ \tau_{\text{mag}} = \frac{\Delta U_{\text{mag}}}{s} \geq \tau_{\text{req}}, \] (1)

where \( \Delta U_{\text{mag}} \) is the magnetic energy difference per unit volume between the two crystallographic domains separated by the twinning plane, and \( s \) is the corresponding twinning shear. The condition described above, \( \tau_{\text{mag}} \geq \tau_{\text{req}} \), has been confirmed to be appropriate for some ferromagnetic shape memory alloys.1-6

By the way, considering the condition of the equation (1), we speculate that the rearrangement of crystallographic domains is realized even in antiferromagnetic martensite phases when \( \tau_{\text{mag}} \) is equal to or larger than \( \tau_{\text{req}} \). In order to the validity of this speculation, we select a NaCl-type 3\( d \) transition metal monoxide CoO because it is reported that CoO shows a simultaneous magnetic and structural transition from a paramagnetic cubic phase to an antiferromagnetic pseudo tetragonal phase.7,8 Then, we investigate the rearrangement of crystallographic domain driven by magnetic field in antiferromagnetic CoO and show the validity of this condition in CoO. For comparison, we also investigate the rearrangement in a typical ferromagnetic shape memory alloy of Ni\textsubscript{2}MnGa.

2. Experimental Procedure

Single crystals of Ni\textsubscript{2}MnGa and CoO were prepared by a floating zone method. They were cut into a parallelepiped for optical microscope observations after determining the orientation by a Laue camera. The specimen was mounted on a cooling/heating stage and inserted into the center of a magnetic field. In order to determine the shear stress required for the twinning plane movement, \( \tau_{\text{req}} \), the tensile tests along the [001]\textsubscript{p} (\( \text{p} \) represents a parent phase) direction were performed by using an Instron-type machine.

3. Results

We have observed the rearrangement of crystallographic domains (martensite variants) driven by magnetic field in Ni\textsubscript{2}MnGa ferromagnetic shape memory alloy which transforms to a low temperature phase (martensite phase) at 202K. Optical microscope observations under a magnetic field up to 1.2 MA/m at 77 K \( \leq T \leq 300 \) K have been made and a typical result at 77 K is shown in figure1. As known from the figure, when a magnetic field is applied along [001]\textsubscript{p}, the rearrangement of crystallographic domains occurs and the single domain state is obtained (figure 1(a) and (b)). The same

![Figure 1](image1.png)  
**Figure 1.** A series of optical micrographs of Ni\textsubscript{2}MnGa under a magnetic field at 77 K. The field direction is along [001]\textsubscript{p} (a), (b); [101]\textsubscript{p} (c), (d); and [111]\textsubscript{p} (e), (f).

![Figure 2](image2.png)  
**Figure 2.** A series of optical micrographs of CoO under a magnetic field at 225 K. The field direction is along [001]\textsubscript{p} (a), (b); [101]\textsubscript{p} (c), (d); and [111]\textsubscript{p} (e), (f).
rearrangement occurs but partially when a magnetic field is applied along [110]p (figure 1(c) and (d)). On the other hand, when a magnetic field is applied along [111]p, the rearrangement of crystallographic domains does not occur (figure 1(e) and (f)). In case of the CoO single crystal, CoO transforms from the parent phase to the martensite phase at 293K. Optical microscope observations under a magnetic field up to 8.0 MA/m have been made and a typical result at 225 K is shown in figure 2. As known from the figure, when a magnetic field is applied along [001]p, the rearrangement of crystallographic domains occurs (figure 1(a) and (b)). This rearrangement occurs at temperatures in the range of $170 \leq T \leq 293$ K, but does not occur at $T < 170$ K. When a magnetic field is applied along [110]p and [111]p, the rearrangement of crystallographic domains does not occur (figure 1(c) ~ (f)) below $T_{Ms}$. These results show that the rearrangement of crystallographic domains is certainly realized in CoO as same as in Ni$_2$MnGa and has magnetic field direction and temperature dependences.

4. Discussion

We evaluate the magnetic shear stress, $\tau_{mag} = \Delta U_{mag}/s$, in Ni$_2$MnGa as in the previous study. The value of $\Delta U_{mag}$ is obtained from the area enclosed by the two magnetization curves: along hard magnetization axis and along easy magnetization axis. Typical magnetization curves of Ni$_2$MnGa are shown in figure 3. The maximum of $\Delta U_{mag}$ is equal to the uniaxial magnetocrystalline anisotropy constant $|K_u|$ in the case that a magnetic field is applied to [001]p direction. The value of $s$ is given by the lattice parameters as $s = \{(1-(c/a)^2)\}/(c/a)$. Using the values of $\Delta U_{mag}$ and $s$, the values of magnetic shear stress, $\tau_{mag}$, are evaluated and shown in figure 4. The value of $\tau_{req}$ obtained by tensile tests is also shown in the figure. As known from the figure, when $\tau_{mag}$ is nearly equal to or larger than $\tau_{req}$, the rearrangement occurs.

![Figure 3](image1.png) **Figure 3.** Typical magnetization curves of Ni$_2$MnGa along easy and hard magnetization axes at 4.2 K.

![Figure 4](image2.png) **Figure 4.** Temperature dependence of $\tau_{mag}$ and $\tau_{req}$ in Ni$_2$MnGa.

We also evaluate the magnetic shear stress in CoO. Considering that the magnetic susceptibility, $\chi$, of CoO depends on the magnetic field direction, the value of $\Delta U_{mag}$ is evaluated as $(\chi_a-\chi_c)H^2/2$, where $\chi_a$ and $\chi_c$ are the magnetic susceptibility along $a$- and $c$-axes of tetragonal martensite phase in case that magnetic field is applied along [001]p direction. Typical magnetization curves along $a$- and $c$-axes are shown in figure 5. The value of $s$ is given by the lattice parameters as $s = \{(1-(c/a)^2)\}/(c/a)$. Using the values of $\Delta U_{mag}$ and $s$, the values of $\tau_{mag}$ are calculated when the magnetic field of 8.0 MA/m is applied along [001]p, [110]p, and [111]p directions. The result is shown in figure 6. The value of $\tau_{req}$ obtained by tensile tests is also shown in the figure. As known from the figure, when $\tau_{mag}$ is
equal to or larger than $\tau_{\text{req}}$, the rearrangement occurs, as in ferromagnetic Ni$_2$MnGa. As a result, we have confirmed the validity of the condition, $\tau_{\text{mag}} \geq \tau_{\text{req}}$ even in antiferromagnetic CoO.

**Figure 5.** Typical magnetization curves of CoO along $a$- and $c$-axes of tetragonal phase at 200 K.

**Figure 6.** Temperature dependence of $\tau_{\text{mag}}$ and $\tau_{\text{req}}$ in CoO.

5. Conclusions
We have investigated the rearrangement of crystallographic domains (martensite variants) driven by magnetic field in ferromagnetic Ni$_2$MnGa and antiferromagnetic CoO. As a result, we have confirmed that when $\tau_{\text{mag}}$ is larger than or equal to $\tau_{\text{req}}$, the rearrangement occurs in antiferromagnetic CoO, as in Ni$_2$MnGa. This shows that the rearrangement of crystallographic domain is able to be explained quantitatively by $\tau_{\text{mag}}$ and $\tau_{\text{req}}$ irrespective of the magnetism of materials.

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