Giant magnetic field-induced strain due to rearrangement of variants in an ordered Fe$_3$Pt

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Abstract

Magnetic field-induced strain due to rearrangement of martensite variants in a ferromagnetic Fe$_3$Pt alloy single crystal with degree of order of about 0.8 has been investigated. The alloy exhibits a martensitic transformation from an ordered L1$_2$-type structure to a tetragonal one at 85 K. The tetragonality of the martensite decreases as temperature decreases, and is about 0.945 at 14 K. When a magnetic field is applied along the [001]$_P$ direction (‘$P$’ stands for ‘parent’ phase) at 4.2 K after cooling down to the temperature under zero magnetic field, the specimen contracts more than 1% along this direction due to the rearrangement of variants. In association with this contraction, the fraction of the variant whose easy axis (c axis) is parallel to the [001]$_P$ direction reaches 70%. On the other hand, when the magnetic field is removed, a part of the strain initially induced by the magnetic field recovers and its value is 0.6% at 4.2 K. This recoverable strain repeatedly appears in the subsequent field applying and removing processes. The rearrangement of variants by a magnetic field is also confirmed by an X-ray measurement under the magnetic field. The energy dissipated due to the rearrangement of variants by the magnetic field is obtained from the magnetization curve to be about 180 kJ/m$^3$. Based on these results and the magnetocrystalline anisotropy constant of the martensite phase ($K_u < 500$ kJ/m$^3$ at 4.2 K), the mechanism of rearrangement of variants under the magnetic field is discussed.

1. Introduction

Recently, a giant magnetic field-induced strain (MFIS) due to rearrangement of martensite variants has been found in ferromagnetic shape memory alloys, such as Ni–Mn–Ga [1–4] and Fe–Pd [5–7]. Since the value of MFIS is larger than that of conventional magnetostrictive materials, the ferromagnetic shape memory alloy is expected to be a promising candidate material for using an actuator driven by a magnetic field. Thus, a lot of studies have been made so far.

According to the previous study by Ullakko et al., they proposed a criterion of the appearance of MFIS as the following: rearrangement of variants occurs when magnetic anisotropy energy is larger than the energy required for the rearrangement of variants [8]. Considering this criterion, it is expected that the giant MFIS will appear in other ferromagnetic shape memory alloys. As for such a material, we found an ordered Fe$_3$Pt [9].

An Fe–Pt alloy containing nearly 25 at.% Pt (A1-type) exhibits a non-thermoelastic martensitic transformation when the degree of order is low. However, as the degree of order becomes higher, the martensitic transformation changes from non-thermoelastic-type (burst-type) to thermoelastic-type [10–12]. Especially, if the degree of order is nearly 0.8, the alloys transform thermoelastically from the L1$_2$-type parent phase to the so-called f.c.t. martensite phase [13] (the crystal structure is probably L$6_0$-type with simple tetragonal Bravais lattice). In the martensite phase, three variants, whose c axes are parallel to [100]$_P$, [010]$_P$ and [001]$_P$, respectively (‘$P$’ stands for the ‘parent’ phase). The easy axis of magnetization of the martensite is c axis.

In a previous paper, we only reported that a giant MFIS appears in an ordered Fe$_3$Pt single crystal [9]. In the present

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paper, therefore, we report the detailed features about the MFIS with the new experimental results including an X-ray diffraction under a magnetic field and a magnetization curve associated with the rearrangement of martensite variants. Moreover, considering these results, we will discuss the mechanism of the MFIS.

2. Experimental procedure

An ingot of Fe–25Pt (at.%) was prepared by an arc-melting method by using a high purity iron rod (99.99%) and a platinum plate (99.95%) as starting materials. The ingot was grown into a single crystal by a floating zone method at a growth rate of 4 mm/h under a purified argon gas flow of 2 l/min. During the growth, the upper and lower shafts were rotated in the opposite directions at a rate of 26 rpm in order to accelerate the convection of the liquid solution. The diameter of the obtained single crystal is about 11 mm, and the growth direction was nearly parallel to [001]P. After the growth, the single crystal was cooled down to room temperature slowly (about 0.2 K/s) in order to prevent the burst-type martensitic transformation. The single crystal was homogenized at 1373 K, followed by ordering heat treatment at 923 K for 360 ks, leading to about 0.8 of the long range order parameter [14]. Magnetization of the specimen was measured by using a superconducting quantum interference device (SQUID) magnetometer. Lattice parameters were determined by an X-ray diffraction with Cu Kα1 radiation. The specimen used for the X-ray experiment is about 0.5 mm in thick, 3 mm in width and 3 mm in length, and all surfaces are {001}P. Thermal expansion and MFIS were measured by three terminal capacitance method. The specimen used for the thermal expansion and the MFIS measurements is a parallelepiped one with 2.0 mm along [001]P, 2.4 mm along [110]P and 1.8 mm along [110]P. The magnetic field was applied along [001]P and the strain was measured along [001]P. Rearrangement of variants under a magnetic field was also detected by an X-ray diffraction.

3. Results

3.1. Martensitic transformation

Fig. 1 shows temperature dependence of the magnetic susceptibility under a magnetic field of 80 kA/m (1000 Oe) applied along [001]P. In the cooling process, the susceptibility starts to decrease at 85 K, as indicated by an arrow. This temperature is the martensitic transformation temperature $T_M$ (‘M’ stands for the ‘martensite’ phase) of the present Fe$_3$Pt. The gradual decrease is probably due to the gradual increase in the magnetocrystalline anisotropy.

The martensitic transformation was also verified by an X-ray diffraction. That is, 002$_P$ reflection splits into 200$_{M}$ and 002$_M$ ones below $T_M$. Fig. 2 shows temperature dependence of lattice parameters calculated from the peak positions of 002$_P$, 200$_{M}$ and 002$_M$ reflections. As known from the figure, the lattice parameter $a$ of the parent phase increases as temperature decreases. This is due to the Invar effect of this alloy [15–17], and the average volume thermal expansion coefficient between 150 and 300 K is calculated to be about $-3 \times 10^{-5}$/K. In the martensite phase, $a$ increases and $c$ decreases gradually as temperature decreases, and the tetragonality $c/a$ decreases gradually with decreasing temperature, and its value is 0.945 at 4.2 K.

3.2. Rearrangement of variants by magnetic field

Prior to examining MFIS of the martensite phase, the specimen was initially cooled down from room temperature to 4.2 K under zero magnetic field. The thermal expansion in this process is shown in Fig. 3(a). It is noted in the figure that the specimen of the parent phase expands monotonically, and the volume thermal expansion coefficient is about $-3 \times 10^{-5}$/K, which is in good agreement with the result obtained by the X-ray diffraction as described before. In association with martensitic transformation, the specimen contracts slightly at $T_M$ indicated by an arrow, and then expands as temperature decreases. The fraction of the variant, whose $c$ axis (easy axis) lies along [001]P, $f_c$, is...
calculated from the relation as,

\[(1 - f_3) a_T + f_3 c_T = a_{300 \, K}(1 + (\Delta l)_\text{cool}),\]

where \(a_T\) and \(c_T\) are the lattice parameters of the martensite at a temperature \(T\), \(a_{300 \, K}\) is the lattice parameter of the parent phase at 300 K and \((\Delta l)_\text{cool}\) is a strain due to the thermal expansion at a temperature \(T\). By using the thermal expansion (Fig. 3(a)) and the lattice parameters (Fig. 2), the calculated \(f_3\) is shown in Fig. 4 by a broken line as a guide for eyes. The fraction \(f_3\) gradually decreases in the cooling process, and the value at 4.2 K is obtained to be about 30% by using the lattice parameter extrapolated to 4.2 K. This value of 30%, being close to one third, is quite natural because three variants are thermally introduced equivalently.

After cooling down to 4.2 K under zero magnetic field, MFIS was measured by applying magnetic field along [001]_p, and the result is shown in Fig. 5. In the field applying process indicated by 'A', the specimen starts to contract when the magnetic field exceeds 0.3 MA/m, and the contraction almost saturates at 3.2 MA/m. The saturated strain is \(-2.3\%\). At this saturated state, the fraction \(f_3\) is calculated to be about 70%, which is different from the fraction (100%) obtained in Ni–Mn–Ga and Fe–Pd. A characteristic feature is that in the field removing process indicated by 'B', the specimen starts to expand at the field of 0.8 MA/m, and the strain of 0.6% recovers as shown by the curve B in Fig. 5. This recoverable strain of 0.6% always appears in the subsequent field applying and removing processes indicated by 'C', 'D', 'E' and 'F'. Such a recoverable strain is not observed in Ni–Mn–Ga or Fe–Pd ferromagnetic shape memory alloys.

After performing a series of MFIS measurements (A–F in Fig. 5), the specimen was heated up to the room temperature under zero magnetic field. The thermal expansion in this process is shown in Fig. 3(b). As seen in the figure, the specimen expands gradually in the heating process. During this process, the calculated fraction \(f_3\) is nearly constant, which is shown in Fig. 4 by a solid line as a guide for eyes. This result means that the thermal expansion in Fig. 3(b) in the martensite phase is mainly due to the temperature dependence of the lattice parameter, not due to the rearrangement of variants.

We also examine the thermal expansion under a magnetic field of 3.2 MA/m, and the result is shown in Fig. 3(c). It is noted in the figure that the specimen starts to contract below its transformation temperature, and the strain gradually decreases as temperature decreases. During this process the calculated fraction \(f_3\) increases as temperature decreases, which is shown in Fig. 4 by a dotted line as a guide for eyes, and its value at 4.2 K is about 75%. This fraction \(f_3\) at 4.2 K is larger than 70%, which is obtained by applying a magnetic field after zero-field cooling as shown in Fig. 5. This difference may be due to the difference between the field-cooling process and the zero-field cooling one on the rearrangement of variants.
3.3. X-ray diffraction under magnetic field

An X-ray experiment under a magnetic field was carried out in order to confirm further that the giant MFIS of Fe$_3$Pt is caused by rearrangement of variants. The magnetic field was applied along [001], and the intensity of the 002$_M$ and 200$_M$ reflections were recorded, where the scattering vector and the direction of the magnetic field are illustrated in the inset of Fig. 6. As seen in the figure, the intensity of 002$_M$ reflection changes by applying the magnetic field at 14 K as follows: under the magnetic field of 0.4 MA/m, the intensity of the 002$_M$ reflection significantly decreases, as shown by the solid line of Fig. 6. When the magnetic field is removed, the intensity of the 002$_M$ reflection slightly recovers. Furthermore, there is no significant peak shift or peak separation corresponding to a magnetic field-induced phase transformation. From these results, it is apparent that the rearrangement of martensite variants occurs by applying a magnetic field. That is, the significant decrease of the intensity of 002$_M$ reflection under the magnetic field means that the fraction $f_c$ of the variant, whose $c$ axis is parallel to the scattering vector, decreases, and the recovery of the intensity by removing the magnetic field means that the recovery of the fraction $f_c$ as shown in Fig. 5. In this way, the results of an X-ray measurement under the magnetic field are in good agreement with those obtained by the MFIS measurement, as described in Section 3.2.

3.4. Magnetization curve

In order to examine the relation between the magnetization process and the MFIS process of A–D in Fig. 5, a magnetization curve in the martensitic state was measured at 4.2 K, and the result is shown in Fig. 7. As known in Fig. 7A, a jump of the magnetization is observed at about 0.5 MA/m. This jump corresponds to the abrupt decrease in the MFIS at about 0.5 MA/m (Fig. 5A). Such a jump does not appear in the field removing process B, and there is a hysteresis between the curves A and B. The area of this hysteresis corresponds to the dissipated energy due to the rearrangement of variants, and is about 100 kJ/m$^3$. Since the fraction of variants rearranged in this field applying and removing process is about 54%, the energy dissipation of 180 kJ/m$^3$ is expected for 100% change in the fraction. Furthermore, the magnetocrystalline anisotropy constant $K_u$ is evaluated from this magnetization curve by using the relation $K_u = M_s H_A / 2$, where $M_s$ of 2.0 T (2.3 $\mu_B$/atom) is the saturation magnetization and $H_A$ of 500 kA/m is the anisotropy field. Thus the obtained $K_u$ is 500 kJ/m$^3$ at 4.2 K. There is also a hysteresis between the curves C and D, which correspond to the recoverable process of Fig. 5C and D. The area of this hysteresis is about 10 kJ/m$^3$. Thus, the energy dissipation corresponding to 100% change in the fraction will be 50 kJ/m$^3$. The magnetization curve for the process of Fig. 5E and F is similar to that for the process of Fig. 5C and D, although they are omitted from Fig. 7 to avoid the complexity of the figure.

4. Discussion

We observed that Fe$_3$Pt exhibits a giant MFIS in association with the rearrangement of variants by a magnetic field. In order to obtain information of this mechanism from a macroscopic point of view, we evaluated a kind of shear stress $\tau_{mag}$ acting on the twinning plane under a magnetic field. In evaluating $\tau_{mag}$, we must know the magnetic energy $\Delta U_{mag}$ between the state before and after the rearrangement of variants. Since the magnetic energy evaluation of the multi-variant state is complex, we consider a simple case in which rearrangement of variants occurs from the least
preferable single variant to the most preferable single one under a magnetic field: the former variant is the one whose c axis (easy axis) is perpendicular to the magnetic field and the latter is the one whose c axis is parallel to it. Under this simplification, $\Delta U_{\text{mag}}$ can be obtained as the magnetic energy difference between the two variants. Then, the shear stress $\tau_{\text{mag}}$ will be evaluated as $\Delta U_{\text{mag}}/S$, where $S$ is the amount of shear. Under the situation in the present Fe$_3$Pt, the magnetocrystalline anisotropy should be expressed as $K_u \sin^2 \theta$, where the angle $\theta$ is the one between the direction of magnetization and the c axis (easy axis). Therefore, the maximum value of $\Delta U_{\text{mag}}$ comes to be the magnetocrystalline anisotropy constant $K_u$. Thus, the maximum value of $\tau_{\text{mag}}$ is $K_u/S$ in the present Fe$_3$Pt, and is about 4.3 MPa at 4.2 K, where the amount of shear is calculated by the relation $S = \{1 - (c/a)^2\}t(c/a)$, and its value is 0.114 at 4.2 K in the present Fe$_3$Pt.

In spite of the existence of $\tau_{\text{mag}}$ of 4.3 MPa, the fraction $f_t$ does not reach 100%, as shown in Fig. 5. This suggests that the shear stress required for the rearrangement of variants increases, and exceeds 4.3 MPa as the rearrangement of variants proceeds. The reason for such an increase during the rearrangement of variants is not known yet, but may be due to an increase in an internal shear stress $\tau_{\text{int}}$ (elastic stress), which may arise from elastic interaction associated with the movement of the interfaces between variants. The existence of $\tau_{\text{int}}$ may be related to the recoverable rearrangement of variants as shown in Fig. 5C and D. In order to obtain more information on $\tau_{\text{int}}$, we need to make tensile and compressive tests associated with rearrangement of variants in the present Fe$_3$Pt.

5. Conclusions

Rearrangement of martensite variants by applying a magnetic field in a ferromagnetic Fe$_3$Pt alloy single crystal, whose martensitic transformation temperature is 85 K, has been investigated. The tetragonality of the martensite decreases as temperature decreases, and is about 0.945 at 14 K. When a magnetic field is applied along [001]$_p$ (P stands for the parent phase) at 4.2 K, the specimen contracts by more than 1%, and the fraction $f_t$ of the variant, whose easy axis (c axis) is parallel to the field direction, reaches 70%. Subsequently, when the magnetic field is removed, a part of the strain (0.6%) recovers at 4.2 K. This recoverable shear strain repeatedly appears in the subsequent field applying and removing processes. The energy dissipations due to the irreversible and recoverable rearrangement of variant are evaluated from the hystereses in the initial magnetization process and the subsequent one to be 180 and 50 kJ/m$^3$, respectively. The rearrangement of variants by a magnetic field is also confirmed by an X-ray measurement under a magnetic field. From the uniaxial magnetocrystalline anisotropy constant of the martensite phase ($K_u \approx 500$ kJ/m$^3$ at 4.2 K), the maximum shear stress $\tau_{\text{mag}}$ by a magnetic field acting on the twinning plane is evaluated to be about 4.3 MPa.

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