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Control of microstructure by a magnetic field in iron-based ferromagnetic shape memory alloys

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Abstract

We have investigated control of microstructure, that is, the rearrangement of martensite variants (crystallographic domain), in a disordered Fe–31.2Pd (at.%) single crystal and an ordered Fe_3Pt single crystal, exhibiting cubic to tetragonal martensitic transformation at 230 and 85 K, respectively. When a magnetic field is applied along the [001] direction to the specimen with the multi-variant state, the specimen expands along the field direction for Fe–31.2Pd and contracts for Fe_3Pt, because the specific variant which reduces the magnetocrystalline anisotropy most will be selected to grow by consuming other variants. The fraction of such variants reaches 100% for Fe–31.2Pd but does not for Fe_3Pt. In the field removal process, part of the field-induced strain recovers for Fe_3Pt but not for Fe–31.2Pd. The recoverable strain of Fe_3Pt depends on temperature and shows a maximum value of nearly 1% at 20 K. From the magnetization curve, the energy dissipated due to the rearrangement of variants by a magnetic field is obtained to be about 260 kJ m^{-3} for Fe–31.2Pd and about 180 kJ m^{-3} for Fe_3Pt. This is of the same order of magnitude as that evaluated from the stress–strain curve of Fe–31.2Pd, meaning that the rearrangement of variants by a magnetic field occurs by nearly the same path or process as that by external stress. On the basis of these results and magnetocrystalline anisotropy constants of martensite phases, the mechanism of rearrangement of variants under a magnetic field is discussed.

1. Introduction

Recently, ferromagnetic shape memory alloys have attracted considerable attention because we can control their twinned microstructure by the application of a magnetic field, resulting in a giant magnetic field-induced strain (MFIS) of several per cent. The first report of the control of microstructure, that is, rearrangement of martensite variants, by a magnetic field was made

for a single crystal of an off-stoichiometric Ni–Mn–Ga alloy by Ullakko *et al* [1], and they considered the mechanism of rearrangement of variants by a magnetic field as follows [2]. The martensite of the Ni–Mn–Ga alloy has a tetragonal structure [3] and the easy axis of magnetization is the *c* axis [4]. When an external field is applied, the specific variant which reduces the magnetocrystalline anisotropy energy most will be selected to grow by consuming other variants.

Considering this mechanism, we expected a disordered Fe–Pd alloy and an ordered Fe_3Pt to also be candidates for showing a large field-induced strain because both alloys are ferromagnetic [5, 6] and exhibit thermoelastic martensitic transformations from cubic structures (A1 type for Fe–Pd and $L1_2$ type for Fe_3Pt) to tetragonal structures (so-called fct martensite) [7, 8]. Actually, rearrangement of variants by a magnetic field in Fe–30at% Pd alloy was first reported by James and Wutting [9], and after that by our group [10, 11]. In addition, the same behaviour was found in Fe_3Pt by our group [12]. In this paper, we show our new results on rearrangement of martensite variants by a magnetic field for two iron-based ferromagnetic shape memory alloys, Fe–Pd and Fe_3Pt. We also show a magnetization curve and a stress–strain curve accompanying rearrangement of variants. By evaluating the energy dissipated during these processes and the magnetocrystalline anisotropy constant obtained from those results, we give a quantitative discussion of the mechanism of the rearrangement of variants under a magnetic field.

2. Experimental details

Ingots of Fe–31.2Pd (at.%) and Fe_3Pt alloys were prepared by an arc melting method with a high purity iron rod (99.99%), a palladium plate (99.97%), and a platinum plate (99.95%) as starting materials. Single crystals of Fe–31.2Pd (at.%) and Fe_3Pt were prepared by a floating zone method. Parallelepiped specimens composed of $(001)_P$, $(110)_P$ and $(110)_P$ planes (the symbol P represents the parent phase) were cut from the single crystals. After homogenization at 1373 K, the Fe–31.2Pd alloy was quenched into iced water and the Fe_3Pt was ordered at 923 K for 360 ks. The degree of order of the present Fe_3Pt is about 0.8, referring to a previous report [8].

The martensitic transformation temperature and magnetic properties were examined by using a superconducting quantum interference device (SQUID) magnetometer. The lattice parameters were determined from x-ray diffraction profiles obtained by a diffractometer with copper Kα radiation. The thermal expansion and magnetic field-induced strain (MFIS) were measured by using a three-terminal capacitance method and/or a strain gauge method. Optical microscope observation under a magnetic field was carried out using a Nomalski-type differential interference contrast optics. Tensile testing was carried out under a constant strain rate of 5×10^{-5} s^{-1}.

3. Results

3.1. The magnetic field-induced strain of Fe–31.2Pd (at.%) alloy

The martensitic transformation temperature T_M, for the transformation from the fcc phase to the tetragonal phase, of Fe–31.2Pd was detected as a sudden decrease of the magnetic susceptibility due to the increase of magnetic anisotropy associated with the transformation, and T_M is determined as about 230 K. The transformation was also detected by means of x-ray diffraction from the separation of the 200 reflection. Figure 1 shows the temperature dependence of the lattice parameter determined from the 200 reflection. As seen in the figure,

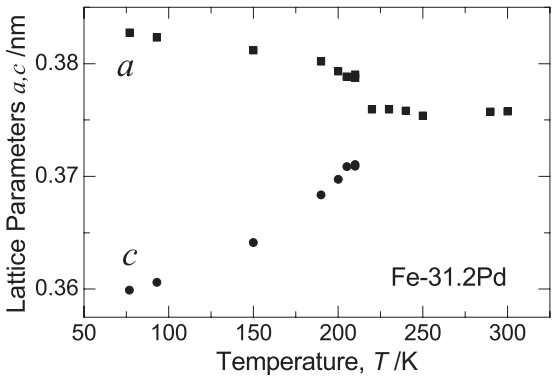

Figure 1. The temperature dependence of the lattice parameters of an Fe–31.2Pd alloy.

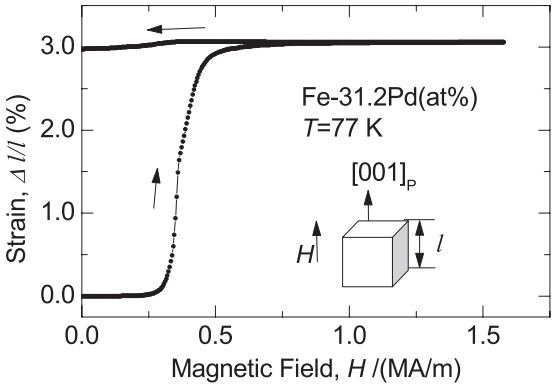

Figure 2. The magnetic field-induced strain of an Fe–31.2Pd alloy single crystal.

the lattice parameters a and c of the tetragonal martensite gradually separate from each other as temperature decreases and the tetragonality c/a is 0.940 at 77 K.

Prior to examining the MFIS, the specimen was cooled down to 77 K under zero magnetic field, and the thermal expansion was measured along the $[001]_P$ direction in the cooling process, where the measurement of the strain was made by a three-terminal capacitance method. Through the cooling process from 300 to 77 K, the specimen contracts by about 1.2%. By using this value and the lattice parameters, the fraction f_a of variants whose a axis (easy axis) is parallel to the $[001]_P$ direction is calculated to be 50% at 77 K.

After cooling to 77 K under zero magnetic field as described above, the MFIS was measured, and the result is shown in figure 2. As seen in the figure, the specimen starts to expand at 0.3 MA m^{-1}, and the strain almost saturates at 1 MA m^{-1}. The saturated strain is about 3.1%. The fraction f_a under the magnetic field of 1 MA m^{-1} is about 100%, meaning that the rearrangement of variants is almost perfect. Since f_a increases when the field is applied, it is speculated that the a axis is the easy axis of magnetization in the martensite phase.

This process was also confirmed by an optical microscope observation under a magnetic field, and the result is shown in figure 3. The surface of the specimen in the parent phase is flat, as seen in figure 3(a). In the cooling process with zero magnetic field, banded surface relief starts to appear below T_M and its contrast increases as temperature decreases. The surface relief at 81 K is shown in figure 3(b). Then, a magnetic field was applied to the specimen with such

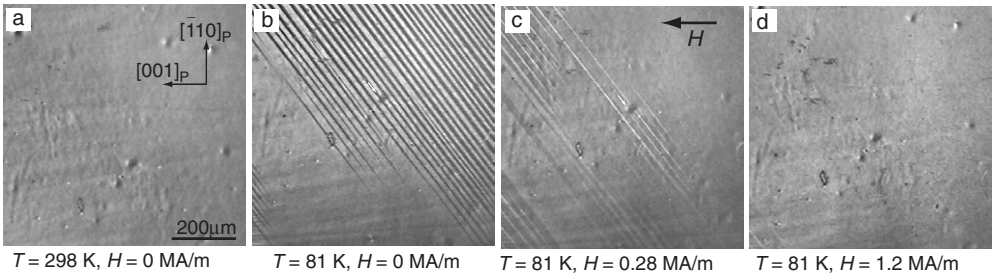

Figure 3. A series of optical micrographs showing rearrangement of variants by a magnetic field in an Fe–31.2Pd alloy single crystal. (a) The parent phase with a flat surface. (b) The martensite phase with zero magnetic field; banded contrast appears due to the self-accommodation of martensite variants. (c) A magnetic field of 0.28 MA m^{-1} is applied along the $[001]_P$ direction. (d) The magnetic field is increased to 1.2 MA m^{-1}; the banded contrast has completely disappeared. The white arrows in (b) and (c) indicate the same place.

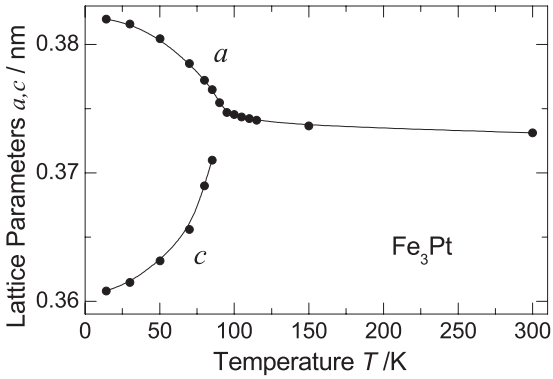

Figure 4. The temperature dependence of the lattice parameters of Fe_3Pt single crystal.

banded surface relief along the $[001]_P$ direction. When the field exceeds about 0.28 MA m^{-1}, the dark bands start to grow, consuming the bright bands. This behaviour is clearly seen if we compare the region indicated by the white arrow in figures 3(b) and (c). The dark bands continue to grow with increasing magnetic field. When the field exceeds about 0.4 MA m^{-1}, the whole area is composed of the dark region as shown in figure 3(d). Comparing the results of optical microscope observation with those for the MFIS shown in figure 2, it is apparent that the MFIS occurs in association with rearrangement of variants by a magnetic field.

3.2. The magnetic field-induced strain of Fe_3Pt alloy

The martensitic transformation temperature T_M, for the transformation from the $L1_2$-type parent phase to the tetragonal martensite phase, of Fe_3Pt was detected as a sudden decrease of the magnetic susceptibility due to the increase of magnetic anisotropy associated with the transformation, and T_M is determined as about 85 K. The transformation was also detected by means of x-ray diffraction from the separation of 200 reflection. Figure 4 shows the temperature dependence of the lattice parameters determined from the 200 reflection. As seen in this figure, the lattice parameters a and c of the tetragonal martensite gradually separate from each other as temperature decreases and the tetragonality c/a is 0.945 at 14 K.

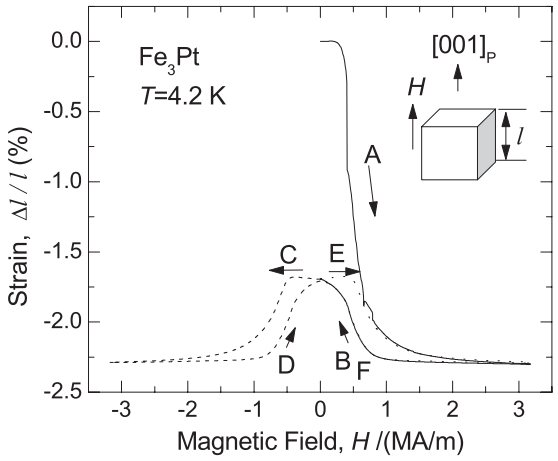

Figure 5. The magnetic field-induced strain of Fe_3Pt single crystal. Measurements were made in the sequence shown by curves A–F.

Prior to examining the MFIS of Fe_3Pt, the specimen was cooled down to 4.2 K under zero magnetic field and the thermal expansion was measured in the cooling process. Through the cooling process from 300 to 4.2 K, the specimen expands by 0.46% along the $[001]_P$ direction. The fraction f_c of the variant, whose c axis (easy axis) is parallel to the $[001]_P$ direction, is calculated to be 30% at 4.2 K, which is a reasonable value considering that three variants are naturally introduced thermally.

After cooling to 4.2 K under zero magnetic field as described above, the MFIS of Fe_3Pt was measured, and the result is shown in figure 5. As shown by curve A of figure 5, the specimen starts to contract at 0.25 MA m^{-1}, and the strain almost saturates at 3.2 MA m^{-1}. The saturated strain is about 2.3%. The fraction f_c under the magnetic field of 3.2 MA m^{-1} is estimated to be about 70%, i.e., the increase in f_c is 40%. A characteristic feature is that the specimen expands by about 0.6% in the field removal process, i.e., part of the MFIS recovers, as seen in curve B of figure 5. In this process, the ratio f_c decreases from 70% to 60%. Then, the MFIS was measured by changing the polarity as shown by curves C and D of figure 5. The contraction of about 0.6% appears and disappears in the field application and removal processes, respectively. In the subsequent field application and removal processes, the same strain appears and disappears as seen in curves E and F. This recoverable strain depends on the temperature and exhibits a maximum value of about 1% at 20 K. The recoverable strain of Fe_3Pt is more than three or times that of Terfenol-D [13], which is well known as a material exhibiting a giant magnetostriction.

3.3. The magnetization curve and the stress–strain curve

We observed that both Fe–31.2Pd and Fe_3Pt exhibit a giant MFIS in association with rearrangement of variants by a magnetic field. In order to obtain information on this mechanism, we evaluate the energy dissipated due to the rearrangement of variants by a magnetic field from the magnetization curves of Fe–31.2Pd and Fe_3Pt. The magnetization curve along the $[001]_P$ direction of Fe–31.2Pd at 77 K is shown in figure 6 and that of Fe_3Pt at 4.2 K is shown in figure 7.

In figures 6 and 7, the field application process is shown by mark A and the removal process by B. The area of the hysteresis between A and B corresponds to the energy dissipated

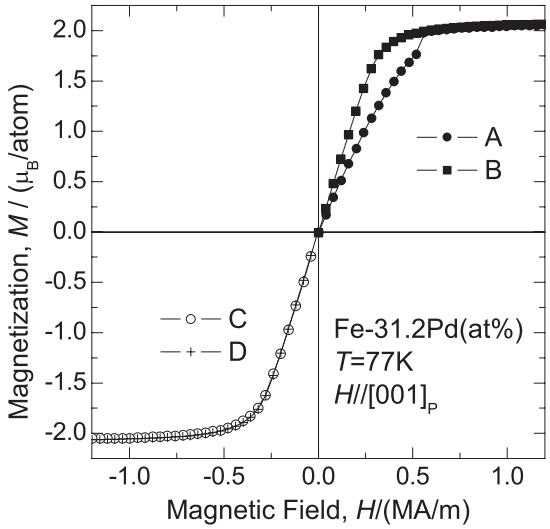

Figure 6. The magnetization curve of Fe–31.2Pd alloy single crystal at 77 K. Measurements were made in the sequence shown by curves A–D.

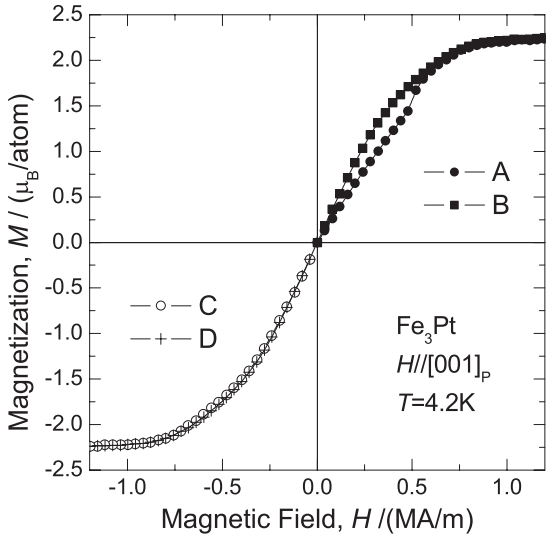

Figure 7. The magnetization curve of Fe_3Pt single crystal at 4.2 K. Measurements were made in the sequence shown by curves A–D.

due to the rearrangement of variants, and the area is about 150 kJ m^{-3} for Fe–31.2Pd and about 100 kJ m^{-3} for Fe_3Pt. During this process, the fraction f_a of Fe–31.2Pd changes by about 57% for Fe–31.2Pd and f_c of Fe_3Pt changes by about 54%. Consequently, the energy dissipation for obtaining a 100% change in the fraction of variants will be about 260 kJ m^{-3} for Fe–31.2Pd and about 180 kJ m^{-3} for Fe_3Pt.

The energy dissipated due to the rearrangement of variants by uniaxial stress is also examined by means of a tensile test at 80 K for Fe–31.2Pd alloy. A typical stress–strain curve is shown in figure 8, where the fraction f_a increases by 57% in this process as in figure 6. The

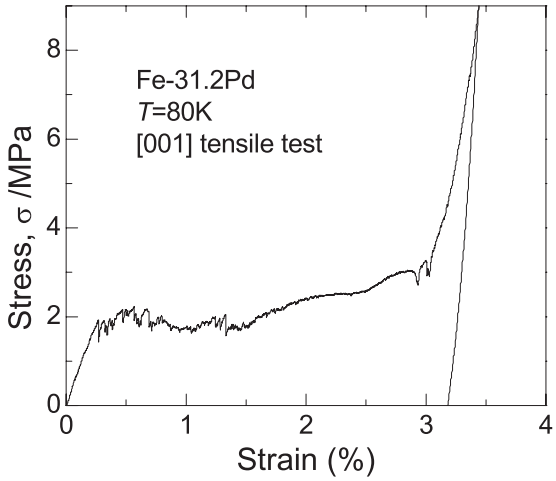

Figure 8. The stress–strain curve obtained from a tensile test along the $[001]_P$ direction of Fe–31.2Pd alloy single crystal at 80 K.

area of the hysteresis of figure 8 is about 80 kJ m^{-3}, and the work required to obtain a 100% change in the fraction of variants will be about 140 kJ m^{-3}. When the same experiment is repeated, an appreciable distribution in the energy dissipation (140–220 kJ m^{-3}) is observed. Consequently, the energy dissipated due to the rearrangement of variants by a magnetic field is roughly the same as that found by mechanical tests.

4. Discussion

In the following, we discuss the conditions for the rearrangement of variants by considering the shear stress acting across the twinning plane. Since rearrangement of variants occurs under a magnetic field, the magnetic field should generate a shear stress τ_{mag} on the twinning plane, and τ_{mag} should be larger than the stress required for the rearrangement of variants τ_{req}.

In evaluating τ_{mag}, we must know the magnetic energy difference ΔU_{mag} between the two states, before (multi-variant state) and after the rearrangement of variants. Since the energy evaluation of the multi-variant state is complex, we consider a simple case in which rearrangement of variants occurs from a single-variant state to another single-variant state although it is a rough approximation. Under this simplification, ΔU_{mag} can be evaluated as the magnetic energy difference between the two variants. Then, the shear stress caused by a magnetic field acting on the twinning plane between the variants will be expressed as $\tau_{mag} = \Delta U_{mag}/s$, where s is the amount of shear. Since the martensite phase is tetragonal, the magnetocrystalline anisotropy energy should be uniaxial. Therefore the magnetic anisotropy energy can be expressed as $K_u \sin^2\theta$, where the angle θ is the angle between the direction of magnetization and the c axis. Thus the maximum value of ΔU_{mag} is the magnetocrystalline anisotropy constant $|K_u|$ in the case where the magnetic field is applied along $[001]_P$. The value of s is obtained, by using the equation $s = \{1-(c/a)^2\}/(c/a)$, as 0.124 for Fe–31.2Pd at 77 K and 0.114 for Fe–31.2Pd at 4.2 K. $|K_u|$ is obtained, by using the equation $|K_u| = (M_s H_A)/2$, as about 350 kJ m^{-3} for Fe–31.2Pd at 77 K (figure 6) and 500 kJ m^{-3} for Fe_3Pt at 4.2 K (figure 7), where M_s is the saturated magnetization and H_A is the anisotropy field. These values are of the same order as reported for Ni–Mn–Ga alloys [14, 15]. Using these values, the maximum of τ_{mag} is obtained as about 2.8 MPa for Fe–31.2Pd at 77 K and 4.3 MPa for Fe_3Pt at 4.2 K.

On the other hand, as we know from figure 7, τ_{req} is about 1 MPa for Fe–31.2Pd at 77 K. In this way, the condition described above ($\tau_{mag} > \tau_{req}$) is actually satisfied for Fe–31.2Pd alloy.

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