Magneto-shape-memory effect in Co–Ni single crystals

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A Co–33 wt % Ni single crystal was grown by an optical floating-zone furnace without crucible in a high-purity argon atmosphere. In the [001] direction, the as-grown single crystal exhibited a saturation magnetization of 124 A m^2/kg, which is nearly twice as high as that of Ni_2MnGa, a typical magneto-shape-memory material. A reversible strain of 3% was induced by an applied pulse magnetic field of 2 T in the [001] direction in the temperature range between M_d and M_s. According to the martensitic transformation theory [T. Y. Hsu (Z. Y. Xu), Sci. China, Ser. E: Technol. Sci. **40**, 561 (1997)] for fcc (γ)→hcp (ε), a possible mechanism based on the reversible motion of the Shockley partial dislocations is proposed in this letter. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1376148]

Among various functional materials, piezoceramics, magnetostrictive materials, and shape-memory alloys have attracted extensive attention in research for their ability to convert between a mechanical quantity (force or displacement) and a signal of another physical form. In comparison, piezoceramics and magnetostrictive materials have an advantage in applications because of their high response frequencies but have the disadvantage of producing only small strains, normally in an order of 10^{-3}. Conventional shape-memory alloys, on the other hand, can yield strains in excess of 6% in tension, but often show a small bandwidth due to the restriction of thermal conduction. The magneto-shape-memory (MSM) alloys, however, are functional materials, which exhibit the characteristics of both large output strain as in conventional shape-memory alloys and rapid response frequencies as in piezoceramics and magnetostrictive materials.

The phenomenon of magnetostriction originates from the rotations of magnetization in magnetic domains in a material under the influence of an external magnetic field. Terfenol (Fe–Dy–Tb),[1] a typical magnetostrictive material, offers a linear strain up to 0.24%. On the other hand, Ni_2MnGa, a typical MSM material, can produce a reversible magnetic-field-induced strain. Ullakko *et al.*[2] first reported a measurement of 0.2% strain along the [001] direction of a Ni_2MnGa single crystal when subjected to a magnetic field of 8 kOe at 265 K. This strain is caused by the motion of twin boundaries induced by the applied magnetic field. Later, James and Wuttig[3] observed in nonstoichiometric single-crystal Ni_2MnGa a magnetic-field-induced strain of 1.3% below 273 K. After that, a single crystal of a tetragonally distorted Heusler alloy in the NiMnGa system has shown a 5% shear strain at room temperature in a field of 4 kOe, reported by O'Handley *et al.*[4] More recently, strains of 6% have been produced at room temperature in NiMnGa by application of fields of the order of 400 kA/m under an opposing stress of the order of 1 MPa.[5] To explain these observations, some mechanisms and theoretical analyses have been reported in the literature.[4–8]

The Ni_2MnGa, however, is an intermetallic compound. Its brittleness hinders its practical application. Therefore, the development of MSM materials with improved mechanical properties is of primary importance for the commercial utilization of the MSM effect. From this point of view, we selected for study some Co–Ni alloys. In this letter, the MSM effect in a Co–33 wt % Ni single crystal is explored and a possible mechanism based on the reversible movement of the Shockley partial dislocations is proposed.

The starting material was prepared by using elemental Co and Ni flakes (⩾99.95% purity). It was melted by using a nonconsumable electrode arc furnace. A cylinder-shaped ingot with a 10 mm diam was obtained for single-crystal growth. A Co–33 wt % Ni single crystal was grown by the crucibleless method in an optical floating-zone furnace in a high-purity argon atmosphere. The as-grown single crystal was annealed at 1173 K for 24 h followed by quenching in ice water. Its orientation was determined by both an x-ray backreflection Laue diffraction camera and an x-ray crystal orientation instrument. The growth direction (the axis of the cylinder) of the single crystal (fcc structure) was found to be parallel to [001]. Samples of $2\times2\times4$ mm in dimension were cut along the [001] and [111] directions for intrinsic magnetic measurements by a JKC-1-type magnetic measuring system, a vibrating sample magnetometer (VSM), and a magnetization balance (MB). Samples of $1\times4\times10$ mm in

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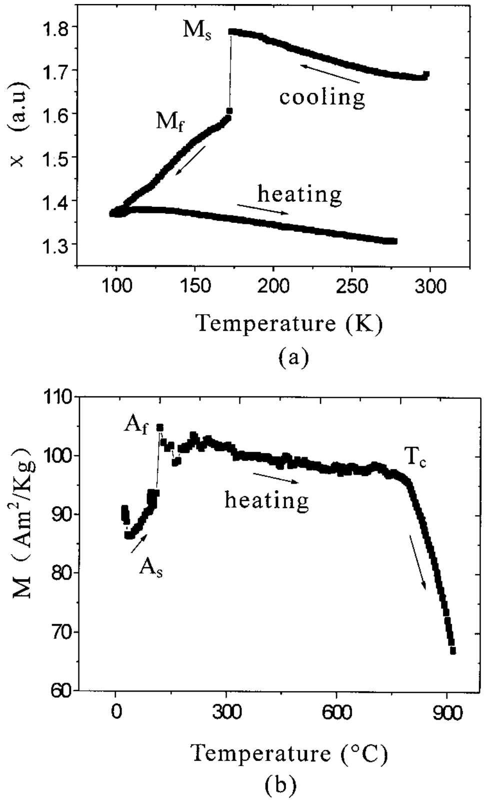

FIG. 1. (a) $\chi-T$ curve with an ac magnetic field of 5 Oe and 70 Hz and (b) $M-T$ curve with a dc magnetic field of 1.2 T in the [001] direction.

[001] and [111] orientations along the length were used for the strain measurements by using strain gauges. The strain gauges made by metal foil were attached to the broad surface of the specimens. The critical temperatures, such as M_s, M_f, A_s, A_f, and Curie temperature T_c were determined by ac susceptibility and magnetization.

Figure 1(a) shows the ac susceptibility as a function of temperature for a specimen oriented in the [001] direction. The ac susceptibility increased gradually with decreasing temperature. At the onset of the martensitic transformation, the ac susceptibility decreased sharply with an obvious kink. From such measurements, M_s and M_f of the martensitic transformation were determined to be 171.5 and 160 K. The reverse transformation, however, occurs above the maximum temperature available on this device, 373 K. Therefore, the reverse transformation was measured by a MB. A M–T curve measured in the [001] direction in the temperature from 10 to 900 °C is shown in Fig. 1(b). From this curve, the austenitic transformation temperatures (A_s, A_f) and the Curie temperature (T_c) were determined to be 382.8, 391, and 1069 K, respectively. The ac susceptibility and the magnetization were also measured for the specimen oriented in the [111] direction. Comparison of the measurements for the two specimens indicates that transformation temperatures are not orientation dependent for single crystals.

Figure 2 shows the magnetization behavior in the [001] direction measured by the VSM. The parent phase with a fcc structure after cooling from 1173 K to the room temperature exhibits a high-saturation magnetization of 124 A m^2/kg, which is twice the magnitude of that of Ni_2MnGa (66 A m^2/kg).[9] The magnetization of the martensitic phase with a hcp structure after cooling down to the liquid nitrogen, followed by heating to the room temperature, is also relatively higher (110 A m^2/kg). A high-saturation magnetization is desirable in applications for magnetic-field-induced martensitic transformations due to the high driving energy, which may supply the energy required for stacking fault expansion and partial dislocation movement.

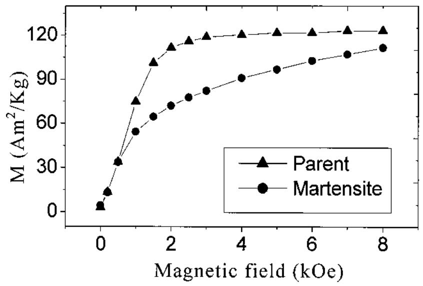

FIG. 2. $M-H$ curves of the parent and martensitic phases in the [001] direction, both at room temperature.

The strain was measured with a pulse magnetic field of 2 T applied along the [001] direction during cooling and heating at different temperatures, as indicated in Fig. 3. It has been defined that the M_d temperature is the highest temperature above M_s, below which the martensitic transformation can be induced by stress. The M_d of Co–33 wt% Ni has been estimated to be between 190 and 200 K.[10] While applying a pulse magnetic field at a temperature above M_d, for example, at 228 K [between (B) and (C) in Fig. 3], a large irreversible strain is induced due to the plastic deformation possibly due to the slip of perfect dislocations of $a/2\langle 110\rangle$. This is suggested by the fact that many of the slip lines are observed on the surface of the specimen based on a test by an optical microscope, a scanning electron microscope, and an atomic-force microscope. It is known that the direction of easy magnetization in Co–Ni single crystals is the $\langle 111\rangle$ direction. When an external magnetic field **H** is applied along the [001] direction, parts of the specimen will tend to turn from the easy-magnetization direction [111] to be aligned with the external magnetic field, which is in the [001] direction. This results in the application of a couple of shear stresses (τ) on the (111) plane just as the external stresses applied on the specimen, as shown in Fig. 4.

When the pulse magnetic field is applied at 180 K (between M_s and M_d), a reversible linear strain of 3% [between (D) and (E) in Fig. 3] is induced. The reason may be related

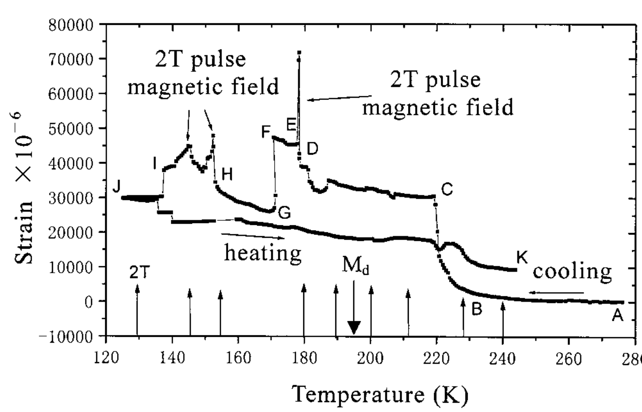

FIG. 3. Strain vs temperature with a pulse (width 5–10 s) magnetic field of 2 T along the [001] direction.

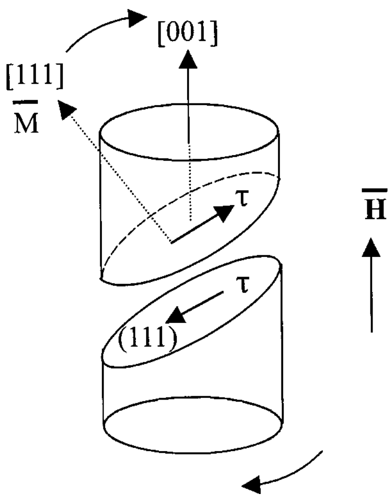

FIG. 4. Schematic presentation of shape change under the applied magnetic field based on the motion of the Shockley partial dislocations of $a/6\langle 112\rangle$ in Co–Ni alloys.

to the generation of strain-induced ε-martensite by the movement of the $a/6\langle 112\rangle$ partial dislocations. The existence of partial dislocations can be proved by the fact that large numbers of stacking faults were observed by a transmission electron microscope. Since the stacking fault energy is relatively low in Co-based alloys, the γ/ε interface will move with less difficulty through the expanding of stacking faults than the motion of twin boundaries in Ni_2MnGa under the influence of an external magnetic field.

The martensitic transformation occurs at 174.8 K, resulting in a shrinkage strain of 2% [from (F) to (G) in Fig. 3]. At temperatures below M_s (such as 155 and 145 K), a recoverable field-induced strain of 1% [between (H) and (I) in Fig. 3] is obtained, which may result from the reorientation of the martensitic variants through the movement of the boundaries between the different variants. At last, about 3% total strain is retained [the height between (J) and (A) in Fig. 3]. After heating to room temperature, $\sim$60% [the height between (J) and (K) in Fig. 3] of the total strain was recovered. From this curve, the M_s and M_f temperatures were determined to be 170.6 and 162 K. These measurements agree with those measured by VSM and MB.

A measurement similar to that in Fig. 3 was also done on the [111] sample, but no detectable strain was measured. This is because the [111] direction is already the easy-magnetization direction.

In summary, from our experiments, several primary results are obtained as follows:

(1) A recoverable strain of 3% is induced by a pulsed magnetic field of 2 T along the [001] direction at 180 K.
(2) While applying the pulsed magnetic field at temperatures below M_s, a recoverable strain of 1% is obtained.
(3) Upon heating to room temperature, $\sim$60% of the total 3% strain is recovered.
(4) When the magnetic field is applied along the [111] direction of the single crystal, no measurable strain can be induced.

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