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Magnetocrystalline anisotropy in single-crystal Co–Ni–Al ferromagnetic shape-memory alloy

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The magnetocrystalline anisotropy in a single-crystal $Co_{37}Ni_{34}Al_{29}$ ferromagnetic shape-memory alloy has been investigated. The prestrain was applied to the parent phase in order to nucleate the specific variant in the sample cooled down through the martensitic transformation temperature. The applied magnetic field facilitates the growth of variants parallel to the applied magnetic field in analogy with the prestrain. From these results of selective nucleation of variants, the magnetocrystalline anisotropy energy in the single crystal $Co_{37}Ni_{34}Al_{29}$ β' martensite phase is estimated to be 3.9×10^6 erg/cm^3. In the single crystal, the observed magnitude of the reversible magnetic-field-induced strains is 0.06%.
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The ferromagnetic shape memory alloys (FSMAs) attract a great deal of attention as smart materials. Because FSMAs exhibit both a reversible martensitic transformation and ferromagnetism, we can control the shape memory effect by applying magnetic field in addition to conventional controls by temperature and stress. In the martensite phase of FSMAs, the giant magnetic-field-induced strains (MFIS) have been observed.[1–3] This phenomenon is concerned with the rearrangement of twin variants.

Recently, Co–Ni–Al alloys have been developed as new FSMAs.[4–6] The Co–Ni–Al β phase undergoes a thermoelastic martensitic transformation from a B2 (cubic) to an $L1_0$ (tetragonal; $c/a=0.816$) structure.[7] The $\beta+\gamma$ two phases in Co–Ni–Al alloys excel in workability.[7] Furthermore, the Curie temperature T_C and the martensitic transformation start temperature M_s in Co–Ni–Al alloys can be individually controlled in a wide range of temperature by changing composition.[4,5] These advantages are to be expected that Co–Ni–Al alloys are promising as FSMAs.

The MFIS depend on the crystallographic orientation, because twin boundary motions are closely related with both the magnetocrystalline anisotropy and the Zeeman energies.[1,8] When the martensitic transformation occurs, some variants are introduced in consequence of the minimum elastic energy. Furthermore, microscopic magnetic domains exist in martensite twin variants so as to reduce the magnetic dipole energy. The magnetic domains not parallel to the direction of applied magnetic field H are diminished on applying H, and then magnetization M changes direction to the magnetic field direction. When the magnetocrystalline anisotropy energy K is larger than the driving energy E_t to rearrange the twin variants, the twin variants rearrange and twin boundary is moved in connection with the rotational magnetization, accompanied by macroscopic strains. These energies also play an important role in the nucleation of specific variants during a cooling process in the magnetic field.[9,10] In order to make the behavior of twin variants clear, it is necessary to investigate the relations among the twin variants movement, various energies, crystal structure, and magnetic properties. The study of the relation between the magnetic domain and the twin boundary has been carried out.[6,11,12] In this letter the magnetic properties, in particular magnetocrystalline anisotropy energy, of a single crystal Co–Ni–Al β phase have been investigated.

By using a Co–Ni–Al alloy ingot a single crystal $Co_{37}Ni_{34}Al_{29}$ alloy was grown by an optical floating-zone method under an argon atmosphere. The composition of the Co–Ni–Al β phase was determined by energy dispersion x-ray spectroscopy (EDX). In order to obtain a single β phase, the single crystal was annealed at 1623 K for 24 h followed by quenching in ice water. Its crystallographic orientations were determined by electron backscattering diffraction pattern (EBSP). The disk for the measurement is a diameter $\phi=2.34$ mm, thickness $h=0.54$ mm. The $\langle 100\rangle$ directions were parallel and perpendicular to the circular faces of the sample in the parent phase. The martensitic transformation start temperature M_s, and the Curie temperature T_C, were determined to be $M_s=172$ K, and $T_C=275$ K, respectively, from the thermomagnetization curve. When the martensitic transformation occurs, some variants are induced in order to make the elastic energy minimum. Multivariant states prevent the investigation of the magnetocrystalline anisotropy. For the purpose of selective nucleation of specific variants, the prestrain of the thickness reduction of about 0.1% was applied to the parent phase by rolling along one of the in-plane [100] direction of the sample. Therefore, the rolling direction (RD), the transverse direction (TD), and the normal direction (ND) are almost along $\langle 100\rangle$ of the parent phase. In order to investigate the prestrain effect on the nucleation of variants, the spontaneous ($H=0$) linear thermal expansions (LTE) of $Co_{37}Ni_{34}Al_{29}$ in the RD,

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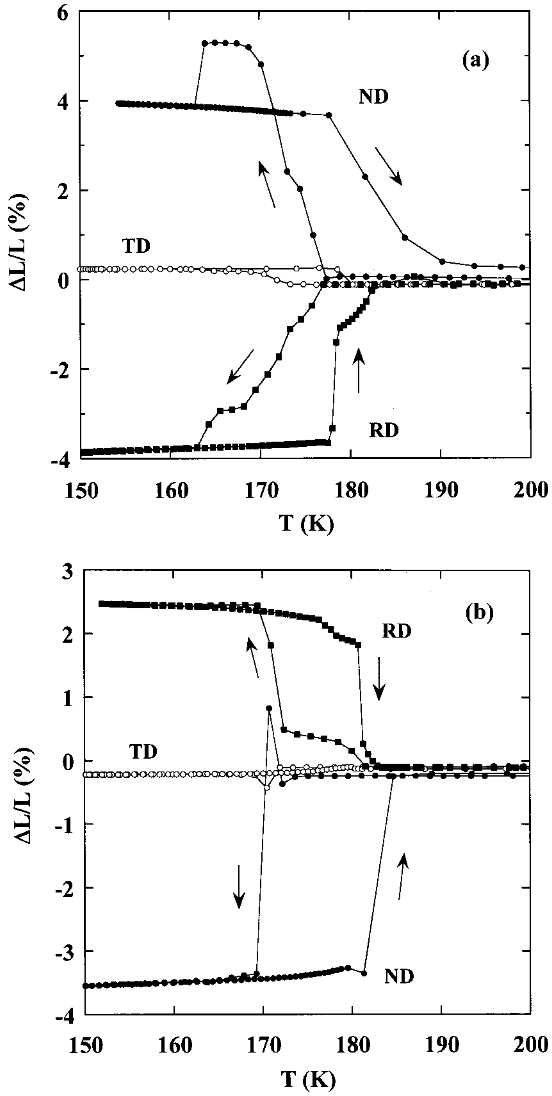

FIG. 1. Spontaneous linear thermal expansion of a single crystal $Co_{37}Ni_{34}Al_{29}$ alloy (a) before applying the prestrain; (b) after applying the prestrain. The arrows indicate the heating and cooling processes.

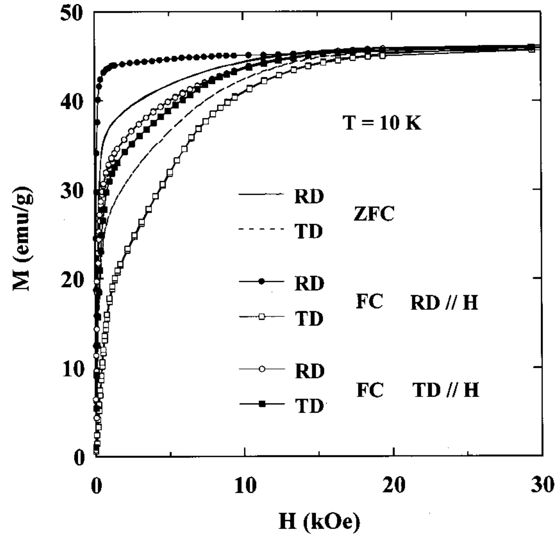

FIG. 2. Magnetization curves along the rolling direction RD and the transverse direction TD for a single crystal $Co_{37}Ni_{34}Al_{29}$ alloy in the martensitic phase.

TD, and ND of the single crystal before and after applying the prestrain were measured in the temperature range of 150–300 K. The magnetization was measured with a superconducting quantum interference device (SQUID) magnetometer in fields up to 50 kOe. The MFIS parallel to the applied magnetic field was measured by a three-thermal capacitance method.

Figure 1 shows the spontaneous LTE of $Co_{37}Ni_{34}Al_{29}$, and the arrows indicate the cooling and heating processes. Before applying the prestrain, the linear relative length change ($\Delta L/L$) associated with the martensitic transformation in the ND is about +5.2%, and in the RD is −3.8% as shown in Fig. 1(a). After applying the prestrain, on the other hand, the change in the ND is −3.5%, in the RD is +2.5% as given in Fig. 1(b). Note that the signs of $\Delta L/L$ in the RD and the ND are reversed. Furthermore, both the transformation temperature intervals between the martensitic transformation start temperature M_s and the martensitic transformation finish temperature M_f, and between the reverse transformation start temperature A_s and the reverse transformation finish temperature A_f are reduced. These results suggest that the prestrain facilitates the growth of specific oriented variants, and reduces the number of twin boundaries of the martensitic phase. The ⟨100⟩ in the parent cubic phase corresponds to the [110] or [001] in the martensitic tetragonal phase. By considering the change in the lattice constants, the *c* axis is increased and the *a* axis is decreased through the martensite transformation. After applying the prestrain, as shown in Fig. 1(b), only the RD is increased and other directions are decreased through the martensite transformation. Therefore, the *c* axis is preferentially developed in the RD. A sudden increase at M_s in the ND in Fig. 1(b) suggests the formation of a few variants.

Figure 2 shows the magnetization curves for the single-crystal $Co_{37}Ni_{34}Al_{29}$ alloy. The coolings were carried out by the following three kinds of processes.

Zero field cooling (ZFC): The sample was first cooled down from the paramagnetic-parent phase ($T>T_C=300$ K) to the ferromagnetic-martensite phase ($T=10$ K) in zero field, which is the so-called ZFC. The magnetization in the RD and the TD of the sample after ZFC was measured in the maretensite state at $T=10$ K. The RD and TD are, respectively, near ⟨100⟩ in the parent cubic phase at $T=300$ K, therefore, [001] or [110] in the martensitic tetragonal phase appear in the RD and TD at $T=10$ K.

Field cooling (FC) in the RD//H: In order to investigate the influence of magnetic field on the nucleation of the martensitic variants, the sample was cooled down from the paramagnetic-parent phase ($T>T_C=300$ K) to the ferromagnetic-martensite phase ($T=10$ K) in a field of 30 kOe, which is the so-called field cooling (FC) process. To begin with, the applied field in cooling process was parallel to the RD. After FC cooling the magnetization in the RD was measured at $T=10$ K in the decreasing applied field. Next, the sample was rotated by 90° in zero field and subsequently the magnetization in the TD was measured at $T=10$ K as a function of the magnetic field up to 50 kOe.

Field cooling (FC) in the TD//H: In a similar way as the FC-RD//H process, the FC-TD//H process has been carried out. After FC cooling, the magnetization in the TD was measured by decreasing applied field, and then the sample was

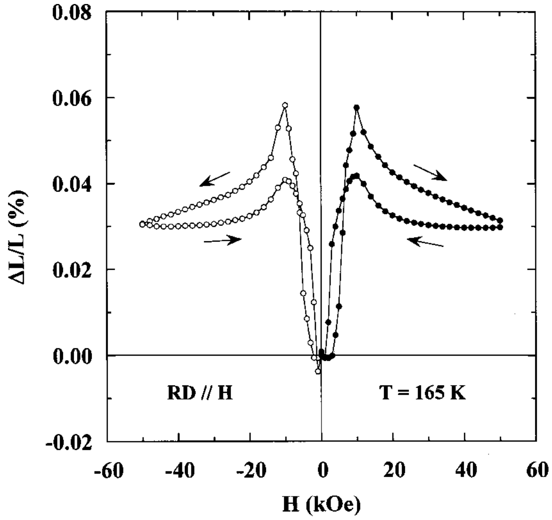

FIG. 3. Relative length change $\Delta L/L$ measured parallel to the direction of magnetic field H at 165 K for the single-crystalline specimen. The magnetic field is applied along the RD. The arrows indicate increasing and decreasing magnetic fields.

rotated by 90°, and its magnetization in the RD was measured.

The measured $M-H$ curve for the RD (FC-RD//H) is situated above the curve for the RD (ZFC), and the $M-H$ curve for the RD (FC-TD//H) lies under the curve for TD (ZFC). On the other hand, the measured $M-H$ curves for the TD (FC-RD//H) and TD (FC-TD//H) are located between the curves for the RD (ZFC) and the TD (ZFC). These results strongly imply that the applied magnetic field stimulates the growth of specific oriented variants along the field direction as cooling down through the martensitic transformation temperature. This preferential orientation takes place to minimize the Zeeman and the magnetocrystalline anisotropy energy, resulting in the growth of variants with the easy magnetization direction parallel to the applied magnetic field. The $M-H$ curves are linked with those of LTE curves. After applying the prestrain, [001] c axis of the tetragonal lattice is preferentially developed in the RD. In this case, [001] grows preferentially parallel to the applied magnetic field, and [110] of the tetragonal lattice grows preferentially perpendicular to the applied magnetic field. Therefore, the $M-H$ curves tell us that the easy axis is the c axis. After correcting the demagnetizing fields, the value of anisotropy energy was evaluated from the magnetic field at which the curve of the hard axis rides on the curve of the easy axis. The anisotropy energy is estimated to be $K_u = 3.9\times10^6$ (erg/cm^3) from the present data. In comparison with the isotropic parent cubic phase, the tetragonal martensitic phase exhibits a strong magnetocrystalline anisotropy, being the same order of that of Ni_2MnGa.[13]

To evaluate the magnitude of the MFIS, the relative length change $\Delta L/L$ parallel to the external magnetic field H was measured. Shown in Fig. 3 is the $\Delta L/L-H$ the curve in the RD of the sample below M_f. The magnetic field was swept from -50 to 50 kOe as shown by the arrows in Fig. 3. On applying the magnetic field, an expansion of the specimen is first observed and $\Delta L/L$ is about 0.06%. Further increasing the magnetic field above ~10 kOe, the sample gradually contracts. Both phenomena are concerned with the twin variant movements, though their mechanism is not clear. To make the relation between the magnetic domain and the twin variant clear would help us understand the mechanism of the twin variant movements. Although the magnetic anisotropy energy is large, the magnitude of the MFIS is small compared with that of Ni_2MnGa (Ref. 1) because the crystal structure and the friction energy of the twin boundary movement influence the twin variant movements, and the martensite phase is incomplete in the single-variant state. By making the complete single-variant state, and estimating the relation of the twin movement and the energies including the elasticity, anisotropy, and boundary friction, the magnitude of the MFIS is expected to be enhanced.

In conclusion, the magnetic anisotropy energy in a single crystal $Co_{37}Ni_{34}Al_{29}$ β' martensite phase plays an important role in the nucleation of specific variants during a cooling process. The applied magnetic field facilitates the growth of specific variants with the c axis parallel to the applied magnetic field as cooling down through the martensitic transformation temperature. The magnetocrystalline anisotropy in the single crystal β' martensite phase is estimated to be 3.9 $\times10^6$ erg/cm^3. The magnitude of the reversible magnetic-field-induced strains (MFIS) is 0.06% for the present single crystal.

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