Magnetic-field-induced strain of Fe-Ni-Ga in single-variant state

著者	及川　勝成
journal or publication title	Applied Physics Letters
volume	83
number	24
page range	4993-4995
year	2003
URL	http://hdl.handle.net/10097/34930

APPLIED PHYSICS LETTERS VOLUME 83, NUMBER 24 15 DECEMBER 2003

Magnetic-field-induced strain of Fe–Ni–Ga in single-variant state

H. Morito,[a] A. Fujita, K. Fukamichi, R. Kainuma, and K. Ishida
Department of Materials Science, Graduate School of Engineering, Tohoku University, Aoba-yama 02, Sendai 980-8579, Japan

K. Oikawa
National Institute of Advanced Industrial Science and Technology, Sendai 983-8551, Japan

(Received 3 July 2003; accepted 14 October 2003)

The magnetic-field-induced strain (MFIS) and the magnetocrystalline anisotropy in $Fe_{19.3}Ni_{54.2}Ga_{26.5}$ ferromagnetic shape memory alloy have been investigated in a single-variant state. From the magnetization curves, the magnetocrystalline anisotropy constant K in the single crystal $Fe_{19.3}Ni_{54.2}Ga_{26.5}$ β' martensite phase is estimated to be 1.8×10^6 erg/cm^3 at 5 K. In the single-variant martensite phase, the reversible MFIS of 0.02% is observed, and the value of K is reduced with increasing temperature. On the other hand, the magnitude of MFIS increases up to 100 K, and then decreases with increasing temperature. Finally, no MFIS is observed above 150 K. From these data, the condition of K for the MFIS can be confirmed at low temperatures. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1632039]

Recently, ferromagnetic shape memory alloys (FSMAs) attracted a great deal of attention as magnetostrictive materials.[1–8] A large magnetic-field-induced strain (MFIS) of the FSMA has been reported to be caused by the variant rearrangement via the twin boundaries motion in the ferromagnetic martensite phase.[1,9–12] The thermoelastic martensite phase consists of an assembly of variants arranged coherently with the parent crystal structure during the martensitic transformation. In each variant, the magnetic moment M is directed to the magnetic easy axis in the magnetocrystalline anisotropy. In the strong magnetocrystalline anisotropy, the angle between M and the applied magnetic field H is lowered by not only the independent rotation of M but also the variant rearrangement so that both the directions of M and the easy axis approach in the parallel direction of H. To be precise, the appearance of the MFIS depends on the Zeeman, magnetocrystalline anisotropy, and twin boundary friction energies.

Recently, Fe–Ni–Ga alloy has been reported to exhibit a martensitic transformation from the $L2_1$-Heusler structure in the parent β phase to a seven-layer modulated (14 M) structure and/or a five-layer modulated (10 M) structure in the martensite β' phase in the ferromagnetic state,[13,14] therefore, this alloy system has drawn attention as a recent FSMA.[15–17] Ni_2MnGa alloy having the modulated layer structures such as 10 and 14 M has low twinning stresses[18] and a large magnetocrystalline anisotropy,[19] meeting the condition for appearance of a large MFIS. The Fe–Ni–Ga alloy also has such seven-layer and/or five-layer modulated layer structures[14] and a large magnetocrystalline anisotropy in the martensite phase.[16] Therefore, the existence of low stresses to move the twin boundaries and a potential to show a large MFIS is expected in the alloy. In the present paper, taking account of the temperature dependence of the magnetocrystalline anisotropy, the MFIS of Fe–Ni–Ga single-variant β' phase has been investigated in a wide temperature range.

A single crystal $Fe_{19.3}Ni_{54.2}Ga_{26.5}$ alloy, which has both the martensitic transformation finishing temperature M_f and the Curie temperature T_C above room temperature, was grown by an optical floating-zone method under a helium gas atmosphere. The single crystal having the B2 phase was annealed at 1453 K for 24 h to homogenize and followed by quenching in ice water. To obtain the $L2_1$ ordered phase, the homogenized specimen was heat-treated at 1073 K for 1 h, and then slowly furnace cooled. Since the martensitic transformation starting temperature M_s of the B2 specimen is below room temperature, its crystallographic orientations were determined by electron backscattering diffraction pattern before heat treatment. The plate-like sample in the parent phase was trimmed so that the $[010]_P$ (P: Parent) and $[\bar{1}01]_P$ directions were parallel and the $[101]_P$ direction was perpendicular to the faces. The magnetization was measured with a superconducting quantum interference device magnetometer in magnetic fields up to 25 kOe.

The relative length change $\Delta L/L$ parallel to the applied magnetic field direction was measured by a three-thermal capacitance method. In order to obtain the single-variant state, the uniaxial compressive stress was applied to the $[010]_P$ direction in the martensite phase. To confirm the rearrangement of the variants, the stress-strain curve was measured at room temperature. After applying uniaxial compressive stress, the strain of 6.5% remained. This value corresponds to the difference in the lattice constant between the $[010]_M$ (M: Martensite) in the martensite phase and the $[010]_P$ in the parent phase. Moreover, it was confirmed by optical micrograpy that the surface relief structure in the multivariant state is completely extinguished after applying uniaxial compressive stress. These results indicate that the stress facilitates the growth of the specific oriented variants, resulting in a single-variant state in the martensite phase of the present specimen. The transformation temperatures M_s, M_f, and T_C of the $Fe_{19.3}Ni_{54.2}Ga_{26.5}$ alloy were determined to be $M_s = 320$ K, $M_f = 305$ K, and $T_C = 310$ K, respectively, from both the thermomagnetization curves and differential scanning calorimetric data.

The magnetocrystalline anisotropy at 5 K was determined for the $Fe_{19.3}Ni_{54.2}Ga_{26.5}$ alloy in the single-variant

---

[a] Electronic mail: morito@maglab.material.tohoku.ac.jp

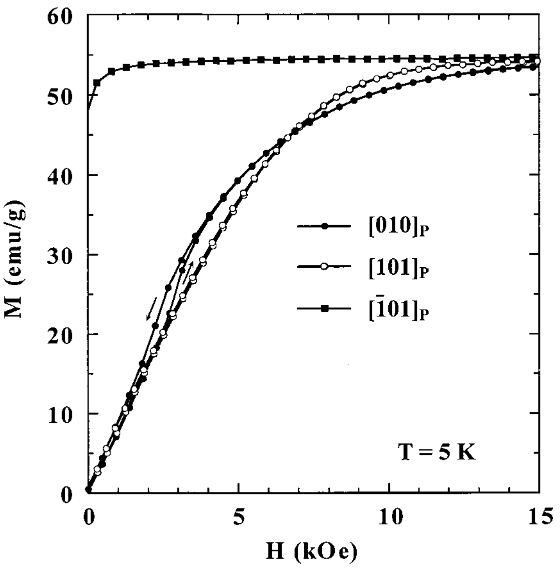

FIG. 1. Magnetization curves along the compressive direction $[010]_P$ and the transverse directions of $[101]_P$ and $[\bar{1}01]_P$ for $Fe_{19.3}Ni_{54.2}Ga_{26.5}$ alloy in the single-variant martensite phase.

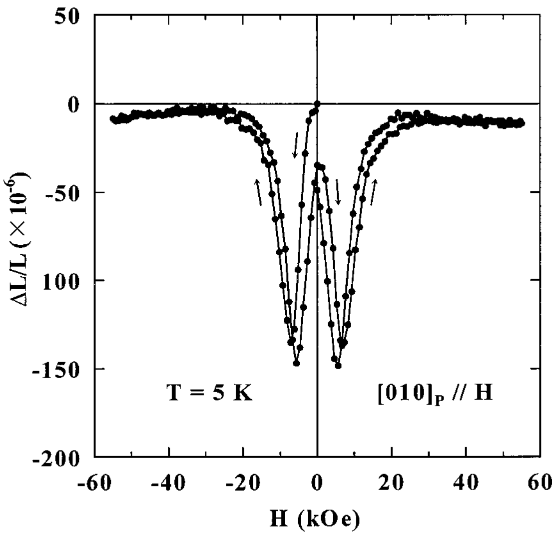

FIG. 2. Relative length change $\Delta L/L$ at 5 K measured parallel to the direction of magnetic field H, applying along the $[010]_P$ direction in the single-variant specimen.

martensite phase from the magnetization curves (M-H) along the $[010]_P$, $[101]_P$, and $[\bar{1}01]_P$ directions. The measured M-H curves are shown in Fig. 1. The M-H curve for the $[\bar{1}01]_P$ direction is easily saturated, while the curves for the compressed $[010]_P$ and $[101]_P$ directions are hardly saturated below 15 kOe. These results indicate that the short-axis of the unit cell in the martensite phase is the hard-axis, while the long-axis is the easy-axis. After correcting the demagnetizing field, the magnetocrystalline anisotropy constant K is evaluated from the M-H curves in Fig. 1 as the area cross section between the easiest curve along $[\bar{1}01]_P$ and the hardest curve along $[101]_P$ directions. The value of K is evaluated to be 1.8×10^6 erg/cm^3 at 5 K. In comparison with the value $K=1.6\times10^6$ (erg/cm^3) of $Fe_{22.0}Ni_{51.5}Ga_{26.5}$ alloy,[16] the present value of the $Fe_{19.3}Ni_{54.2}Ga_{26.5}$ martensite phase is relatively large. As shown in Fig. 1, the M-H curve along the $[010]_P$ direction is accompanied by a small hysteresis which is not observed along the other two directions, suggesting the possibility of the rearrangement of variants.

Shown in Fig. 2 is the $\Delta L/L$-H curves in the $[010]_P$ direction at 5 K, where the hysteresis is observed in the M-H curve in Fig. 1. On applying magnetic field, a shrinkage of the specimen is first observed and the value of $\Delta L/L$ reaches about -150×10^{-6} at $H=6$ kOe, whereas the $\Delta L/L$ in the measured direction increases above 6 kOe. This two-step behavior is also observed in the Co–Ni–Al alloy,[7] which would be related to the magnetic domain wall displacement and the magnetization rotation.

To obtain the well-defined relation between the magnetic domain and the twin variant, the vector measurement of magnetization was carried out. The longitudinal magnetization $M_\parallel$ and the transverse magnetization $M_\perp$ were measured by applying magnetic field H along the $[010]_P$ direction. As shown in Fig. 3, $M_\perp$ rises to make a peak and then falls downright, while $M_\parallel$ is gradually increased and saturated. From the magnetization curves, the value of $\sqrt{M_\parallel^2+M_\perp^2}/M_{\mathrm{sat}}$, where M_{sat} is the saturation magnetization, becomes constant above 6 kOe. With increasing magnetic field, the magnetic domain walls displace and the volume of the domain magnetized parallel to H increases, and then an equal volume of the domain magnetized opposite to H decreases, resulting in the increase of magnetization. Finally, the change in M above $H=6$ kOe is accomplished by the coherent rotation. The first shrinkage is associated with the linear magnetostriction due to the magnetic domain wall displacement, and the expansion is correlated with the rearrangement of the variants due to the rotation of magnetizations.

Figure 4 shows the temperature dependence for the magnitude of the maximum MFIS $|\Delta L/L|_{\mathrm{max}}$ in a magnetic field of 55 kOe. With increasing temperature, $|\Delta L/L|_{\mathrm{max}}$ increases and shows the largest value of 180×10^{-6} at 100 K, and then steeply decreases above 150 K. The inset in Fig. 4 shows the temperature dependence of the magnetocrystalline anisotropy constant K and the saturation magnetization M_{sat}. As the temperature increases, both K and M_{sat} decrease near

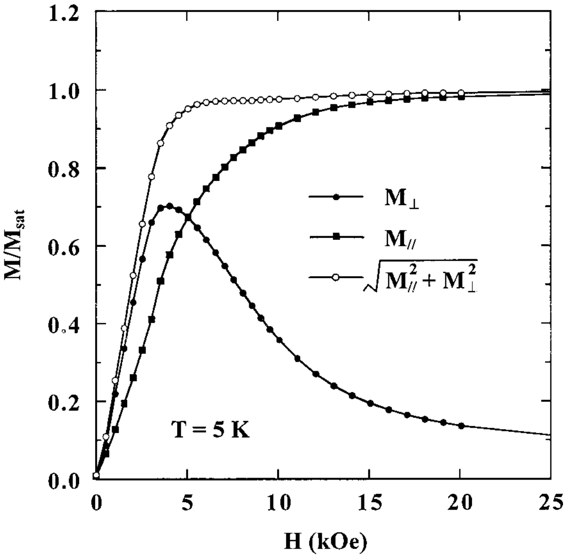

FIG. 3. Vector magnetization curves along the $[010]_P$ at 5 K for $Fe_{19.3}Ni_{54.2}Ga_{26.5}$ alloy in the single-variant martensite phase.

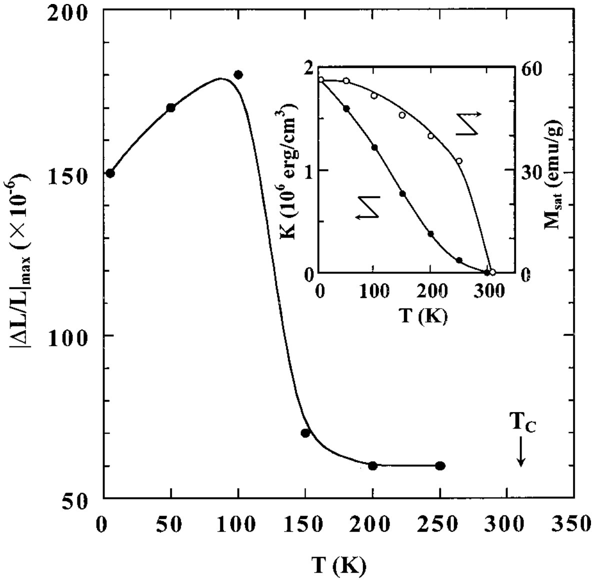

FIG. 4. Temperature dependence of the maximum of the magnetic-field-induced strain (MFIS) for $Fe_{19.3}Ni_{54.2}Ga_{26.5}$ alloy in the single-variant martensite phase. The arrow indicates the Curie temperature T_C. The inset shows the temperature dependence of the magnetocrystalline anisotropy constant K and the saturation magnetization M_{sat}.

T_C, and the decrease of K is more remarkable compared with that of M_{sat}. The temperature dependence of the anisotropy constant K at low temperatures can be described by

$$K(T)/K(0)=[M(T)/M(0)]^3.$$

The validity of this equation has been reported for the Ni_2MnGa martensite phase,[20] but the present $Fe_{19.3}Ni_{54.2}Ga_{26.5}$ martensite phase does not follow this expression. The value of K in the martensite is influenced by the crystal structure, in particular, the lattice constant ratio,[21] and hence we should pay much attention to the lattice constant ratio for the study of the crystal structure in the martensite phase.

The values of $|\Delta L/L|_{max}$ and K are different from each other in the temperature dependence. With increasing temperature, K continuously decreases, while $|\Delta L/L|_{max}$ increases in the temperature range up to 100 K. At low temperatures, K is so sufficiently strong that M in the unfavorably oriented variants cannot rotate to the H direction, the Zeeman energy is reduced by twin boundaries motion, accompanied by large strains. For the martensite phase with a weak anisotropy, the variants remain but H rotates the M directions without moving twin boundaries with large strains.[12] These results indicate that the MFIS caused by the rearrangement of variants in the single-variant martensite phase of $Fe_{19.3}Ni_{54.2}Ga_{26.5}$ is observed above the critical value of K of about 10^6 erg/cm^3 even at low temperatures, following the model proposed by O'Handley.[12] The value of $|\Delta L/L|_{max}$ is effected by the value of K as well as the mobility of the twin boundaries. A large MFIS is observed below M_f in Ni_2MnGa alloys due to balance between K and the mobility of the twin boundaries.[22] In the Fe–Ni–Ga alloy system, however, a sufficiently large K should be kept up to high temperatures to obtain a large value of MFIS, because the mobility of twin boundaries becomes high with increasing temperature, in particular, near M_f. Consequently, the increase of T_C would preserve a high value of K up to high temperatures. Furthermore, the increase of the lattice constant ratio in the martensite phase would also bring about a high value of K.

In conclusion, the magnetocrystalline anisotropy energy in the single crystal $Fe_{19.3}Ni_{54.2}Ga_{26.5}$ β' martensite phase is estimated to be 1.8×10^6 erg/cm^3. In the single-variant state, the observed magnitude of the reversible MFIS is 0.02% at 5 K. With increasing temperature, the value of the magnetocrystalline anisotropy constant K is reduced, but the magnitude of MFIS slightly increases, and yet no MFIS is observed above 150 K. From these data, it is confirmed that the MFIS is observed above the critical value of K, consistent with the model proposed by O'Handley.[12] Consequently, in order to obtain a high critical value of K at high temperatures, say room temperature, we should increase the Curie temperature and the lattice constant ratio in the Fe–Ni–Ga alloy system.

A part of the present study was supported by the Grant-in-Aids for Scientific Research from the Ministry of Education, Science, Sports and Culture, Japan.

[1] K. Ullakko, J. K. Huang, C. Kantner, R. C. O'Handley, and V. V. Kokorin, Appl. Phys. Lett. **69**, 1966 (1996).
[2] R. D. James and M. Wuttig, Philos. Mag. A **77**, 1273 (1998).
[3] T. Kakeshita, T. Takeuchi, T. Fukuda, M. Tsujiguchi, T. Saburi, R. Oshima, and S. Muto, Appl. Phys. Lett. **77**, 1502 (2000).
[4] A. Fujita, K. Fukamichi, F. Gejima, R. Kainuma, and K. Ishida, Appl. Phys. Lett. **77**, 3054 (2000).
[5] K. Oikawa, L. Wulff, T. Iijima, F. Gejima, T. Ohmori, A. Fujita, K. Fukamichi, R. Kainuma, and K. Ishida, Appl. Phys. Lett. **79**, 3290 (2001).
[6] K. Oikawa, T. Ota, F. Gejima, T. Ohmori, R. Kainuma, and K. Ishida, Mater. Trans., JIM **42**, 2472 (2001).
[7] H. Morito, A. Fujita, K. Fukamichi, R. Kainuma, K. Ishida, and K. Oikawa, Appl. Phys. Lett. **81**, 1657 (2002).
[8] M. Wuttig, J. Li, and C. Craciunescu, Scr. Mater. **44**, 2393 (2001).
[9] O. Heczko, A. Sozinov, and K. Ullakko, IEEE Trans. Magn. **36**, 3266 (2000).
[10] S. J. Murray, M. Marioni, S. M. Allen, R. C. O'Handley, and T. A. Lograsso, Appl. Phys. Lett. **77**, 886 (2000).
[11] A. Sozinov, A. A. Likhachev, N. Lanska, and K. Ullakko, Appl. Phys. Lett. **80**, 1746 (2002).
[12] R. C. O'Handley, J. Appl. Phys. **83**, 3263 (1998).
[13] K. Oikawa, T. Ota, Y. Sutou, T. Ohmori, R. Kainuma, and K. Ishida, Mater. Trans., JIM **43**, 2360 (2002).
[14] K. Oikawa, T. Ota, T. Ohmori, Y. Tanaka, H. Morito, A. Fujita, R. Kainuma, K. Fukamichi, and K. Ishida, Appl. Phys. Lett. **81**, 5201 (2002).
[15] Z. H. Liu, M. Zhang, Y. T. Cui, Y. Q. Zhou, W. H. Wang, G. H. Wu, X. X. Zhang, and Gang Xiao, Appl. Phys. Lett. **82**, 424 (2003).
[16] H. Morito, A. Fujita, K. Fukamichi, T. Ota, R. Kainuma, K. Ishida, and K. Oikawa, Mater. Trans., JIM **44**, 661 (2003).
[17] Y. Murakami, D. Shindo, K. Oikawa, R. Kainuma, and K. Ishida, Appl. Phys. Lett. **82**, 3695 (2003).
[18] A. Sozinov, A. A. Likhachev, and K. Ullakko, IEEE Trans. Magn. **38**, 2814 (2002).
[19] R. Tickle and R. D. James, J. Magn. Magn. Mater. **195**, 627 (1999).
[20] L. Straka and O. Heczko, J. Appl. Phys. **93**, 8636 (2003).
[21] J. Enkovaara, A. Ayuela, L. Nordström, and R. M. Nieminen, Phys. Rev. B **65**, 134 422 (2002).
[22] G. H. Wu, C. H. Yu, L. Q. Meng, J. L. Chen, F. M. Yang, S. R. Qi, W. S. Zhan, Z. Wang, Y. F. Zheng, and L. C. Zhao, Appl. Phys. Lett. **75**, 2990 (1999).