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# Magnetic properties and large magnetic-field-induced strains in off-stoichiometric Ni–Mn–Al Heusler alloys

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

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Magnetic properties and magnetic-field-induced strains (MFIS) have been investigated for off-stoichiometric Ni–Mn–Al Heusler alloys with an ordered $L2_1$ structure. A clear martensitic transformation in $Ni_{53}Mn_{25}Al_{22}$ alloy was revealed below the Curie temperature. In the polycrystalline specimen, an irreversible relative change due to the MFIS was confirmed between the martensite start and finish temperatures $M_s$ and $M_f$, and a maximum relative length change $\Delta L/L|_{7\mathrm{T}}$ of about $-100$ ppm was observed at just above $M_f$. On the other hand, a large irreversible relative length change of about 1000 ppm has been demonstrated in the magnetic field of 7 T for a single crystal cut from the polycrystalline specimen. A delay of the response of strains against the magnetic field was also confirmed. © *2000 American Institute of Physics.* [S0003-6951(00)01645-4]

Recently, various magnetic properties have received attention for development of the ferromagnetic shape-memory alloys as magnetomechanical materials.[1,2] In particular, Fe–Pd and $Ni_2MnGa$ alloys have been extensively investigated due to their large strains induced by an external magnetic field. These strains have been discussed in terms of the rearrangement of twin variants.[3–6]

In the Ni–Mn–Al ternary system, the thermoelastic martensitic transformation has been investigated.[7–9] The magnetic state of a quenched B2 phase of the off-stoichiometric $Ni_2MnAl$ Heusler alloys has been reported to be antiferromagnetic and/or spin glass.[8] On the other hand, an ordered $L2_1$ phase, which is very recently observed,[9] is ferromagnetic and also shows a martensitic transformation in analogy with $Ni_2MnGa$. Therefore, the presence of the magnetic-field-induced strains (MFIS) is also expected in the present system.

In the present study, the fundamental magnetic properties and the MFIS were investigated for off-stoichiometric $Ni_2MnAl$ Heusler alloys. We have observed a large strain. Since the martensitic transformation temperature and the Curie temperature are very sensitive to the concentration, the investigation has been restricted to the ferromagnetic specimens of $Ni_{50+x}Mn_{25}Al_{25-x}$ $(0 \leqslant x \leqslant 3.0)$, which show a relatively high martensitic transformation temperature of 200–250 K.[10,11]

Ni–Mn–Al ternary alloys with a $B2$ single phase were prepared in an induction furnace under an argon atmosphere. To obtain an $L2_1$ phase, all of the cast alloys were solution treated at 1000 °C for 72 h and quenched in ice water, followed by aging at 400 °C for 500–600 h. The magnetization was measured with a superconducting quantum interference device magnetometer. The length change of the specimen parallel to the applied magnetic field was measured by a three-terminal capacitance method. Most of the measurements were carried out for the polycrystalline specimens, and a single-crystalline grain block of about $2 \times 2 \times 2$ mm cut from the polycrystalline specimen was also used for the measurement of the relative length change.

Figure 1 shows the temperature dependence of magnetization in the magnetic field $H$ of 5 mT for $Ni_{50+x}Mn_{25}Al_{25-x}$ $(x = 0.0$, 2.5, and 3.0). A magnetic susceptibility divergence

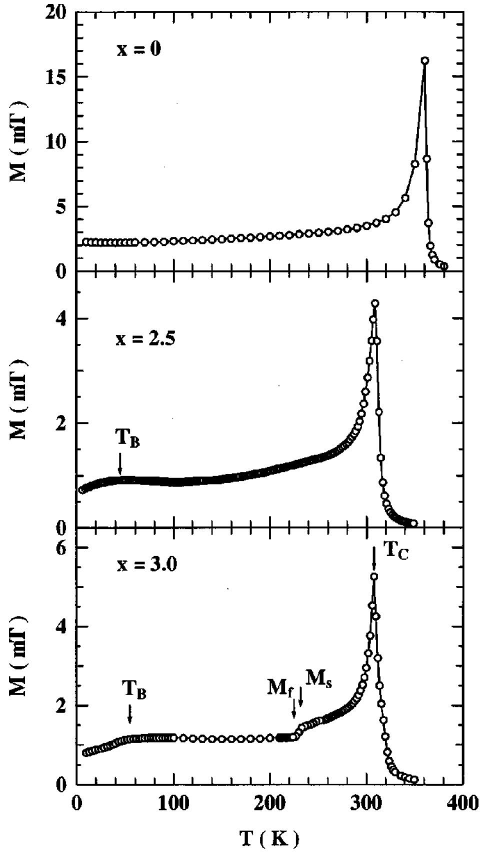

FIG. 1. Temperature dependence of the magnetization for $Ni_{50+x}Mn_{25}Al_{25-x}$ $(x = 0$, 2.5, and 3.0) alloys. The Curie temperature $T_C$, the martensite start and finish temperatures $M_s$ and $M_f$, and the blocking temperature $T_B$ are, respectively, indicated by the arrows.

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

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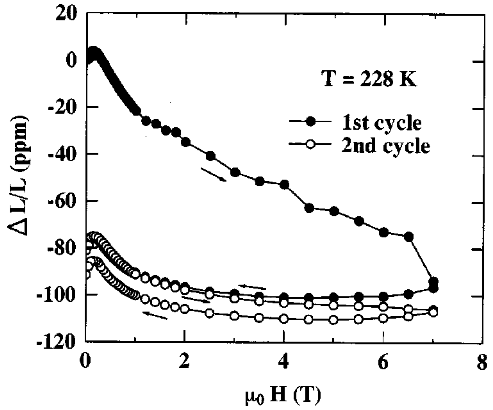

FIG. 2. Relative length change $\Delta L/L$ measured parallel to the direction of magnetic field $H$ at 253 K for the $Ni_{53}Mn_{25}Al_{22}$ polycrystalline alloy.

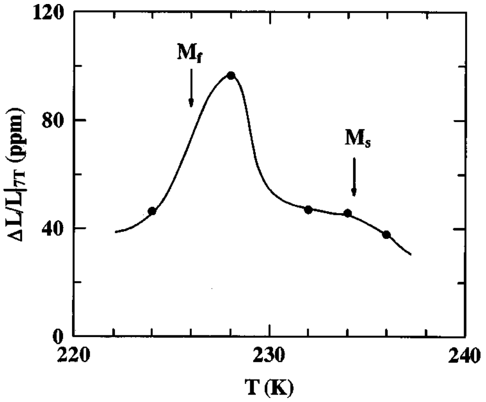

FIG. 3. Temperature dependence of the relative length change in 7 T, $\Delta L/L|_{7\,\mathrm{T}}$ for the $Ni_{53}Mn_{25}Al_{22}$ polycrystalline alloy. The martensite start and finish temperatures $M_s$ and $M_f$ are indicated by the arrows.

at the Curie temperature is observed in the temperature range of 300–400 K for all specimens. A broad shoulder appears for $x=2.5$ and a pronounced terrace-like change occurs for $x=3.0$ between the martensite start temperature $M_s$ and its finish temperature $M_f$ in the $M-T$ curve, revealing the change in the magnetic anisotropy caused by the martensitic transformation.

At low temperatures, the susceptibility for $x=2.5$ and 3.0 decreases with decreasing temperature below $T_B$. Since the B2 phase exhibits an antiferromagnetic behavior,[8,12] compositional inhomogeneity or structural defects can bring about a spin-glass-like phase transition caused by the competition between the ferromagnetic and antiferromagnetic exchange interactions. What has to be noticed is that the divergence of the second nonlinear susceptibility, $\chi_2 = \partial^3 M/\partial^3 H|_{H\sim 0}$, is evidence for the spin-glass transition.[13] However, no divergence around $T_B$ was observed in the temperature dependence of $\chi_2$ obtained from the signal of the $3\omega$ component in an ac magnetic field with a frequency of $\omega$. It has been reported that $L2_1$ Ni–Mn–Al Heusler alloys have nanoscale antiphase domain (APD) structures surrounded by antiphase boundary (APB) regions.[9] It is plausible that the APB regions inhibit the long-range ferromagnetic interaction at low temperatures and, hence, the APDs behave like ferromagnetic clusters. Such a structural characteristic is responsible for the origin of the blocking of the magnetic clusters. Accordingly, the change of susceptibility below $T_B$ does not originate from the spin-glass behavior but should be attributed to the blocking of magnetic clusters, and $T_B$ corresponds to the blocking temperature.

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. 2 is the $\Delta L/L-H$ curve for the polycrystalline sample with $x=3.0$ cooled down from room temperature to 228 K, i.e., just above the $M_f$ point of 226 K. On applying a magnetic field, a shrinkage of the specimen was observed and $\Delta L/L$ becomes about $-100$ ppm at 7 T. An irreversible behavior appears under the decreasing magnetic field and a residual $\Delta L/L$ of about $-80$ ppm is observed after the magnetic field becomes zero. Furthermore, the observed value of $\Delta L/L$ becomes only of about $-20$ ppm in the second cycle in the same procedure.

The temperature dependence of the relative change in 7 T, $\Delta L/L|_{7\mathrm{T}}$, in the first cycle is displayed in Fig. 3. The measurement was carried out in the cooling (the parent to the martensitic transformation) process and $M_s$ and $M_f$ points are indicated by the arrows. As seen in Fig. 3, $\Delta L/L$ becomes larger with decreasing temperature below $M_s$ and exhibits a maximum at just above $M_f$. It should be noted that $\Delta L/L$ is small in the range just below $M_f$. Therefore, the MFIS in the present system should be closely correlated to the phase transition in analogy with the $Ni_{52}Mn_{22.5}Ga_{25.8}$ alloy.[6] Many twin variants are observed at just above $M_f$ and the volume fraction of the parent phase is very small there.[11] In Ni–Mn–Ga alloys, the MFIS have been explained by the rearrangement of twin variants,[2,4,5] and the coexistence of small amounts of the parent phase provides a good environment for the motion of the twin variants.[14] Therefore, the large value of the MFIS in the present system could also be explained in a similar way.

The magnitude of the MFIS should depend on the crystallographic orientation, because the twin boundary motion is correlated to the magnetic anisotropy in the twin variants.[2] In order to bear out this point, the $\Delta L/L-H$ measurement was carried out for a cubic single-crystalline grain block cut out from the polycrystalline specimen. In the single-crystalline specimen, the difference between $M_s$ and $M_f$ is very small. As shown in Fig. 4, $\Delta L/L$ measured parallel to $H$ increases remarkably with increasing magnetic field, especially above 2 T, and becomes about 1000 ppm at 7 T. The inset shows

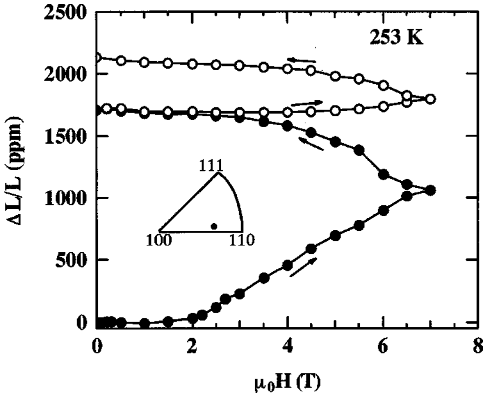

FIG. 4. Relative length change $\Delta L/L$ measured parallel to the direction of magnetic field $H$ at 253 K for the single-crystalline specimen cut from the $Ni_{53}Mn_{25}Al_{22}$ polycrystalline alloy. The inset shows the stereo triangle indicating the direction of applied magnetic field.

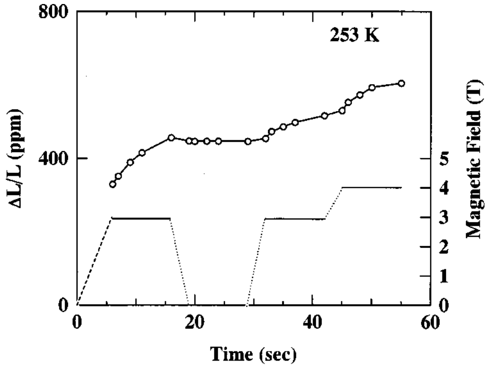

FIG. 5. Time dependence of the relative length change $\Delta L/L$ in the intermittent and steady magnetic fields.

the stereo diagram indicating the direction of applied magnetic field. The important point to note is that the sign of $\Delta L/L$ is positive and the magnitude of $\Delta L/L$ is ten times larger, compared to the data of the polycrystalline specimen given in Fig. 2. When the magnetic field is reduced from 7 T, $\Delta L/L$ still keeps increasing with decreasing magnetic field and the total irreversible strains becomes about 1700 ppm. A similar behavior is observed in the second cycle, but the magnitude of $\Delta L/L$ is much smaller than that in the first cycle.

As seen in Figs. 2 and 4, the increase of $\Delta L/L$ has been observed even in the decreasing of the magnetic field. A feasible explanation for such a behavior is delay of the response of strains against the magnetic field. Shown in Fig. 5 is the time dependence of $\Delta L/L$ for the single crystal at 253 K in the magnetic field of 3 T. The values of $\Delta L/L$ start to increase just after the magnetic field is fixed, and the increment becomes sluggish with increasing time. When the magnetic field is turned off, no change of $\Delta L/L$ against time is observed. Therefore, the time dependence of $\Delta L/L$ is closely related to the magnetic field. By applying a magnetic field of 3 T again, the time-dependent change of $\Delta L/L$ revives, but the initial rate is smaller than that in the first run. Namely, the present time-dependent phenomenon is an irreversible activation process caused by the magnetic field. In the next stage, the initial rate of the time dependence of $\Delta L/L$ becomes larger when the magnetic field of 4 T is applied, although the decay is observed again. Consequently, the present activation process is also a function of the strength of the magnetic field. For a detailed discussion on the irreversibility in the rearrangement of twin variants, further observations of the structure and the magnetic domain are necessary.

In summary, a clear martensitic transition has been observed in ferromagnetic off-stoichiometric Ni–Mn–Al Heusler alloys. In the $Ni_{53}Mn_{25}Al_{22}$ polycrystalline alloy, an irreversible behavior due to the magnetic-field-induced strains was observed in a range between the martensite start and finish temperatures $M_s$ and $M_f$. A maximum value of the MFIS of about $-100$ ppm was observed at just above $M_f$. In a single-crystal cut from the polycrystalline specimen, the magnitude of the MFIS, accompanied by a time-dependent change, attains about 1700 ppm.

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