RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 61, NUMBER 22 1 JUNE 2000-II

Magnetic-field-induced twin boundary motion in magnetic shape-memory alloys

Harsh Deep Chopra* and Chunhai Ji
Thin Films & Nanosynthesis Laboratory, Materials Program, Mechanical and Aerospace Engineering Department,
State University of New York at Buffalo, Buffalo, New York 14260

V. V. Kokorin
Institute of Magnetism, Kiev, Ukraine

(Received 11 February 2000)

This paper reports direct microscopic evidence of magnetoelastic coupling between ferroelastic twin domains and ferromagnetic Weiss domains in magnetic shape memory alloys, using the Ni-Mn-Ga Heusler alloys. In a martensitically transformed Ni$_{2+x}$Mn$_{1-x}$Ga single crystal, the magnetic domains were found to be superimposed upon the martensite twin domains. Simultaneous observation of magnetic domains and twin domains as a function of applied magnetic field shows that concomitant with the reconfiguration of magnetic domains in an applied magnetic field, ferroelastic twin domains also readjust their relative volume fraction in order to accommodate to the externally applied *magnetic field*. This readjustment is shown to occur by the displacement of twin domain walls. As a result, the volume fraction of one set of favorably oriented twin domains (with respect to magnetoelastic energy) increase at the expense of another set of unfavorably oriented twin domains. These studies would enable subsequent quantitative correlation between the magnetic-field-induced macroscopic strain and the microscopic self-adjustment of twin domains.

Large strains in the shape memory alloys (SMA's) are a direct consequence of a thermoelastic, reversible martensitic phase transition in these systems.[1] As a result of reduction in crystal symmetry upon undergoing a structural phase transition, ferroelastic domains commonly appear in SMA's, which are related to each other in a twinlike manner. Transformation strains in SMA's typically exceed 10–14 %, in contrast to strains of the order of 0.1–0.24 % in magnetostrictive or piezoelectric transducer materials. Towards the goal of combining the advantages of magnetostrictive or piezoelectric materials, viz., short reaction times, high efficiency, etc., along with large displacements and high-energy density associated SMA's recent research has focused on a subset of SMA's, which are also *ferromagnetic* in nature.[2–4] In ferromagnetic SMA's, the ferromagnetic Weiss domains are considered to be magnetoelastically coupled to and superimposed upon ferroelastic twin domains that are formed upon undergoing a martensitic phase transition. Therefore, ferromagnetic SMA's offer the possibility of either inducing a martensitic phase transition or rearrangement of ferroelastic twin domains in the martensite state simply by an applied magnetic field. The magnetic-field-induced shape change in SMA's has the potential to overcome the ''bottleneck'' associated with thermally activated shape memory effect, viz., their slow dynamic response.

Recent attention has primarily focused on the ferromagnetic Ni$_{2+x}$Mn$_{1-x}$Ga Heusler alloys and the Fe-Pd alloy system.[2–4] In particular, the stoichiometric Heusler alloy Ni$_2$MnGa is highly ordered and has a Strukturbericht type $L2_1$ structure.[5] It has a Curie temperature of 376 K, and a magnetic moment of $4.17\mu_B$ largely confined to the Mn sites, and a small moment of $0.3\mu_B$ at the Ni sites.[5] It undergoes a cubic to tetragonal martensitic phase transition at 202 K, and in which state the magnetic easy axis lies along the tetragonal *c* axis.[5] In order to make Ni-Mn-Ga alloys more amenable to practical applications, Chernenko *et al.*[6] have shown that the addition to Ga (at a constant Mn concentration) lowers the martensitic start temperature M_s, whereas the addition of Mn at constant Ni concentration causes the M_s temperature to increase. Furthermore, it was found that the substitution of Ni atoms by Mn atoms at constant Ga results in alloys with lower M_s.[6] Ullakko *et al.* [2] first reported a large strain of 0.2% in an unstressed off-stoichiometric Ni$_{2+x}$Mn$_{1-x}$Ga single crystal at an applied field of 8 kOe; a magnetic field induced strain of 0.5% in Fe-Pd single crystals has been reported by James and Wuttig.[3] More recently, a magnetic field induced strain of 5% has been reported by O'Handley *et al.*[7] in another off-stoichiometric Ni$_{2+x}$Mn$_{1-x}$Ga Heusler alloy at an applied field value of 4 kOe.

In the transformed martensite state, the microstructure of Ni$_{2+x}$Mn$_{1-x}$Ga Heusler alloys consist of ferroelastic domains which are in twin orientation with respect to each other across the tetragonal {101} planes,[5] as shown schematically in Fig. 1.[8] Due to the magnetoelastic coupling between the ferroelastic and magnetic domains, the free energy mini-

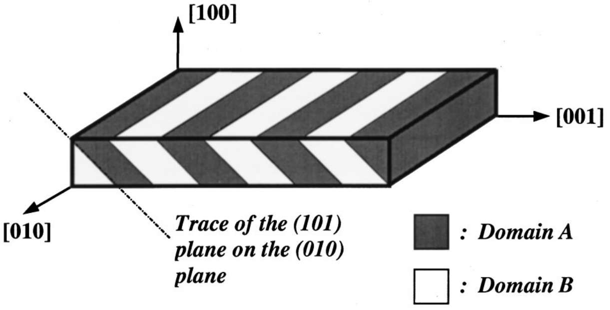

FIG. 1. Schematic of the relative orientation of martensite twins in a Ni-Mn-Ga SMA.

0163-1829/2000/61(22)/14913(3)/$15.00 PRB 61 R14 913

mization during magnetization reversal requires a reconfiguration of the ferroelastic domains when the geometry of the magnetic domains is altered by application of magnetic field. (Conversely, a change in ferroelastic domain configuration by an applied strain should reconfigure the magnetic domain structure). In other words, the observed magnetic field induced *macroscopic* strain reported heretofore in magnetic SMA's (Refs. 2, 3, and 7) is considered to result from a mutual accommodation of ferroelastic twin domains in order to conform to an externally applied magnetic field. Indirect evidence of this magnetoelastic coupling between Weiss domains and structural twins has recently been shown by Qi and James using the magnetic force microscopy.[9] In that study, a $\mathrm{Ni}_{2+x}\mathrm{Mn}_{1-x}\mathrm{Ga}$ single crystal was cooled from the cubic (austenite) state to the tetragonal (martensite) state in the presence of an applied field. Results showed that as the field strength during cooling was changed, the density of twins in the martensite state change,[9] although direct evidence of twin rearrangement in the transformed martensite state itself in an applied magnetic field was not shown. The present paper reports on evidence of microscopic rearrangement of twin domains by magnetic field in the martensitic state of a magnetic SMA, using a $\mathrm{Ni}_{2+x}\mathrm{Mn}_{1-x}\mathrm{Ga}$ single crystal. Detailed studies are currently in progress, which are aimed at establishing a quantitative correlation between the field induced macroscopic strain and microscopic readjustment of twin domains.[10]

In the present study, an off-stoichiometric $\mathrm{Ni}_{2+x}\mathrm{Mn}_{1-x}\mathrm{Ga}$ single crystal with a M_s temperature of $\approx$373 K was investigated. Samples were prepared at the Institute of Magnetism in Kiev, and details of sample preparation are given elsewhere.[6] Based on a previous study by Kokorin and coworkers, this M_s temperature corresponds to a chemical composition of approximately $\mathrm{Ni}_{53.8}\mathrm{Mn}_{23.7}\mathrm{Ga}_{22.5}$.[6] Although the sample was not precisely oriented (and is not critical in the present study), optical observation of transformed martensite on adjacent surfaces indicated the sample to have a [100] orientation for the surface on which domain observations were made. The nature of magnetization reversal and observation of magnetoelastically coupled twin wall motion was made in real time using the high-resolution Interference-Contrast-Colloid (ICC) technique, the details of which have recently been reviewed elsewhere.[11] Briefly, the ICC method employs a colloidal solution to decorate the microfield on a magnetic surface and detects the colloid-decorated microfield using a Nomarski interferometer. This allows simultaneous observation of ferromagnetic domains superimposed upon the ferroelastic twin domains to a high resolution. Samples were polished down to 0.05 μm polishing wheel, followed by a light surface etch to remove the strained layer caused by polishing using a Nital solution (2% HNO_3 in 98% ethanol). Note that it is important not to overetch the sample surface, since etching tends to permanently delineate the twin boundaries in the sample, making the visualization of subsequent twin wall motion difficult.

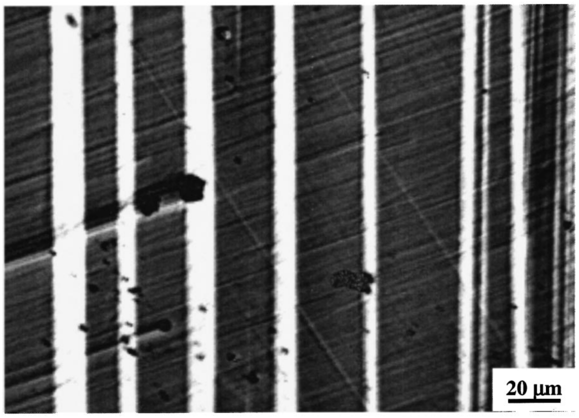

FIG. 2. Optical micrograph of martensite twins in a $\mathrm{Ni}_{2+x}\mathrm{Mn}_{1-x}\mathrm{Ga}$ SMA single crystal.

Figure 2 shows an optical micrograph of a transformed $\mathrm{Ni}_{2+x}\mathrm{Mn}_{1-x}\mathrm{Ga}$ single crystal viewed approximately along its [100] surface. The image was recorded in the differential interference contrast mode of the optical microscope in order to highlight the surface relief associated with a martensite transition. In Fig. 2, the dark and bright bands are structural domains in twin orientation with respect to each other across a $\{101\}$ twin plane. In Fig. 2, the domain periodicity (width of a dark band and a bright band) is approximately 25–40 μm. Figures 3(a) and 3(b) show an ICC image of the $\mathrm{Ni}_{2+x}\mathrm{Mn}_{1-x}\mathrm{Ga}$ sample, which reveals the magnetic domain structure in the sample (fine ripples) superimposed upon the dark and white vertical twin domains. In order to allow the sample to expand in an applied magnetic field, a small adhesive band was used to hold the sample at one of its end. This also prevents the sample from moving and adhering itself to

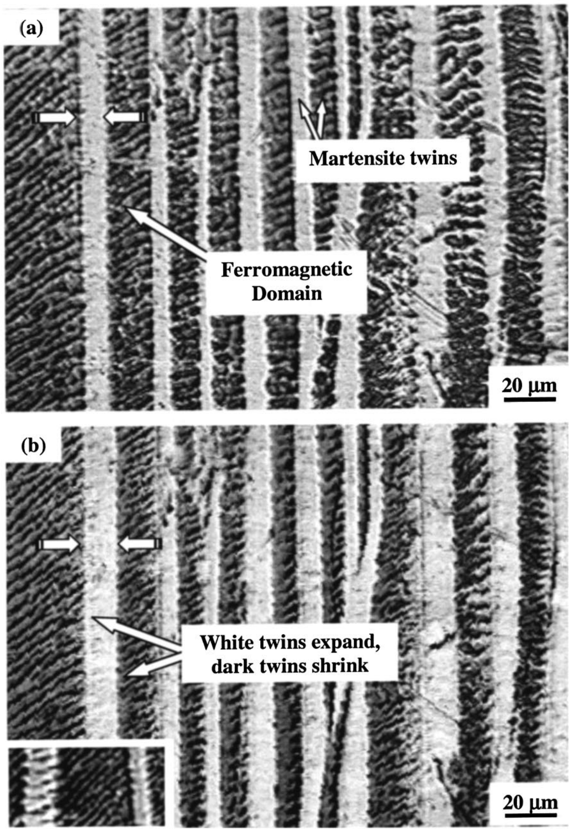

FIG. 3. Magnetic domains superimposed on martensite twins in a $\mathrm{Ni}_{2+x}\mathrm{Mn}_{1-x}\mathrm{Ga}$ SMA single crystal. (a) Applied field strength $H \approx 400$ Oe, (b) applied field strength $H \approx 1800$ Oe. The field direction points from left to right in the micrographs.

the pole pieces of the energized electromagnet. Figures 3(a) and 3(b) were recorded at approximately 400 Oe and 1800 Oe, respectively. The direction of applied field in Figs. 3(a) and 3(b) is in the horizontal direction, and points from left to right in the micrographs. Note that although the magnetic domains in Figs. 3(a) and 3(b) are clearly visible within the dark twin domains, their presence is less obvious in the white twin domains. Well-delineated magnetic domains in the white twin domains are shown more clearly in the inset at the bottom-left in Fig. 3(b). Figures 3(a) and 3(b) show that as the field strength is increased from ≈400 Oe to ≈1800 Oe, the magnetization of the sample is accompanied by a rearrangement of the magnetic domains, as is expected on the grounds of magnetic energy minimization. More significantly, Figs. 3(a) and 3(b) show direct evidence of magnetoelastic coupling between the twin domains and the magnetic domains. With respect to the relative volume fraction of these dark/white twin domains in Fig. 3(a), this manifests itself in Fig. 3(b) by an increase in the volume fraction of the white twin domains by twin wall motion, and at the expense of the dark twin domains. Closer observation of twin wall motion revealed that their displacement is accompanied by a complex dynamic reconfiguration of the magnetoelastically coupled magnetic domains at the twin boundaries. Detailed description of this mechanism is beyond the scope of this preliminary article. However, experiments are currently in progress aimed at understanding the micromagnetic structure of these alloys and its relationship to the crystallographic orientation of the twin domains. Nonetheless, this study would enable subsequent quantitative correlation between the magnetic field induced macroscopic strain and the microscopic self-adjustment of twin domains.

In conclusion, this article presents direct evidence of microscopic rearrangement of twin domains by twin wall motion, which leads to the observed macroscopic strain in magnetic SMA's in an applied magnetic field.

This work was supported by the National Science Foundation, Grant No. DMR-97-31733, and this grant is gratefully acknowledged. It also benefited from the National Science Foundation Division of International Programs (Eastern Division), and the authors gratefully acknowledge this support.

---

*Author to whom correspondence should be addressed. Email address: hchopra@eng.buffalo.edu

[1] See, for example, C. M. Wayman, in *Shape Memory Effects in Alloys, Toronto, Ontario, Canada, 1975*, edited by J. Perkins (Naval Postgraduate School, Monterey, California, 1975), p. 1.

[2] K. Ullakko, J. K. Huang, C. Kantner, R. C. O'Handley, and V. V. Kokorin, Appl. Phys. Lett. **69**, 1966 (1996).

[3] R. D. James and M. Wuttig, Philos. Mag. A **77**, 1273 (1998).

[4] A. N. Vasil'ev, A. D. Bozhko, V. V. Khovailo, I. E. Dikshtein, V. G. Shavron, V. D. Buchelnikov, M. Matsumoto, S. Suzuki, T. Takagi, and J. Tani, Phys. Rev. B **59**, 1113 (1999); F. Zuo, X. Su, and K. H. Wu, *ibid.* **58**, 11 127 (1998); E. Obradó, A. Gonzàlez-Comas, Lluis Mañosa, and A. Planes, J. Appl. Phys. **83**, 7300 (1998).

[5] P. J. Webster, K. R. A. Ziebeck, S. L. Town, and M. S. Peak, Philos. Mag. B **49**, 295 (1984).

[6] V. A. Cherenko, E. Cesari, V. V. Kokorin, and I. N. Vitenko, Scr. Metall. Mater. **33**, 1239 (1995).

[7] R. C. O'Handley, S. J. Murray, M. Marioni, H. Nembach, and S. M. Allen, J. Appl. Phys. **87**, 4712 (2000).

[8] H. D. Chopra, C. Bailly, and M. Wuttig, Acta Metall. Mater. **44**, 747 (1996); H. D. Chopra, A. Roytburd, and M. Wuttig, Metall. Trans. A **27**, 1695 (1996).

[9] Q. Pan and R. D. James, J. Appl. Phys. **87**, 4702 (2000).

[10] H. D. Chopra (unpublished).

[11] H. D. Chopra, S. Z. Hua, D. S. Lashmore, R. D. Shull, W. F. Egelhoff, Jr., and L. J. Swartzendruber, Microscopy and Analysis **28**, 15 (1998).