Large magnetic-field-induced strains in Ni–Mn–Ga nonmodulated martensite

V. A. Chernenko,1,a) M. Chmielus,2,b) and P. Müllner2
1Dpto. Electricidad y Electronica, Universidad del País Vasco, P.O. Box 644, E-48080 Bilbao, Spain and IKERBASQUE, Basque Foundation for Science, 48011 Bilbao, Spain
2Department of Materials Science and Engineering, Boise State University, Boise, Idaho 83725, USA

(Received 8 July 2009; accepted 24 August 2009; published online 11 September 2009; publisher error corrected xx September 2009)

Large magnetic field-induced strains of up to 0.17% for a stress-free Ni53.1Mn26.6Ga20.3 single crystal with nonmodulated martensite phase were generated in a rotating magnetic field. This magnetic-field-induced strain, which is ten times larger than values reported so far for nonmodulated martensites, evidences significant magnetic-field-induced twin boundary motion, which so far was thought to be impossible. This result reinforces the interest in nonmodulated martensites, which are formed as a ground state in the Heusler-type ferromagnetic shape memory alloys. © 2009 American Institute of Physics. [doi:10.1063/1.3227661]

The magnetically weak anisotropic cubic Ni–Mn–Ga Heusler alloys exhibit a martensitic transformation (MT) resulting in martensitic phases with elastically soft crystal lattices and strong magnetocrystalline anisotropies (e.g., Refs. 1 and 2). The magnetic state of these martensites is coupled with a twin microstructure with highly mobile twin boundaries giving rise to the ferromagnetic shape memory effect (FSME). The FSME is the macroscopic effect of the magnetic-field-induced rearrangement of twins, whereby a strain up to the spontaneous lattice distortion, $\varepsilon_{\text{c}} = 1 - c/a$, where $c$ and $a$ are the lattice constants of the martensite phase, is generated depending on the mutual orientation of the magnetic field and the easy magnetic direction of the twin variant. While the theoretically highest values of magnetic-field-induced strains (MFISs) of $\varepsilon_{\text{c}}^{\text{tetra}} \approx 6\%$ (Refs. 3 and 4) and $\varepsilon_{s}^{\text{ortho}} = 10\%$,5,6 have already been achieved in the modulated pseudotetragonal (10M) and pseudoorthorhombic (14M) martensites, respectively, with $c/a < 1$, this has not been accomplished yet for nonmodulated (NM) tetragonal martensite with $c/a > 1$ and $\varepsilon_{s}^{\text{NM}} \approx 20\%$.7,8

Generally, in order to force twin boundaries to move, one needs to apply a stress larger than the twinning stress $\sigma_t$ of martensite. The application of a magnetic field close to the saturation value, $H_s$, produces a maximum magnetoelastic stress (magnetostress), $\sigma(H_s)$, which is equivalent to the applied mechanical stress when

$$\sigma(H_s) > \sigma_t$$

In Eq. (1), $M$ is the magnetization and $\delta = -1.5 \text{ MPa/T}^2$ is the magnetoelastic constant of 10 M martensite (see, e.g., Ref. 9). For a typical value of $M$ of about 0.6 T, Eq. (1) results in a compressive stress, $\sigma(H_s)$, equal to about 3 MPa. While the relationship

$$\sigma(H_s) = 6\delta M^2$$

was shown experimentally to be satisfied and large MFIS obtained for the modulated martensites,7,8,10 these effects have not yet been demonstrated in the case of NM martensite. Experiments with externally stress-free samples showed an MFIS of only about 0.02%.27 Such a small value was proposed to be due to the enhanced twinning stress (>6 MPa) (Refs. 2, 7, and 10) and a reduced value of $\sigma_t$.11 On the other hand, because of the statistical character of $\sigma_t$ (e.g., Refs. 9 and 12) and utilizing the reduction of $\sigma_t$ by training,1,2,6,9,10,12 one can expect to generate samples with conditions where Eq. (2) is satisfied, at least, in a significant volume fraction of the sample. In such a case, the activation of a significant fraction of $\varepsilon_{s}^{\text{NM}}$ is expected. We demonstrated previously for 10M and 14M Ni–Mn–Ga samples that a combination of thermomechanical and subsequent magneto-mechanical training leads to an enormous enhancement of the MFIS.6,13 We furthermore found a training effect in the NM Ni–Fe–Ga(Co) martensite although the magnetostrain effect was only 60 ppm.14

In the present work, thermo-mechanical and magneto-mechanical training was applied to a Ni53.1Mn26.6Ga20.3 (numbers indicate atomic percent) with NM martensite. It was of profound importance to get evidence of a large MFIS in this type of martensite because (i) it is frequently encountered in the Heusler alloys representing their ground state,15 and (ii) it has higher ductility than the modulated martensites.16

The oriented parallelepiped-shaped single crystal with $c/a = 1.18 \pm 0.02$ described previously16–18 was used. The temperature of the forward MT and the Curie temperature are 366 and 373 K, respectively. The martensite in this alloy has an easy-plane type of magnetic anisotropy, which prescribes different domain configurations as compared to the modulated martensites.18 Two samples A and B with identical size of 2.85 mm × 2.35 mm × 4.15 mm were prepared by cutting an initial piece in half parallel to the 100 direction so that this direction has the longest dimension in both samples while the other two directions are kept along (110). The magnetomechanical properties of both samples were tested.

---

a)Electronic mail: vladimir.chernenko@gmail.com. On leave from Institute of Magnetism, Kyiv 03142, Ukraine.

b)Also at Helmholtz Centre Berlin for Materials and Energy, Glienicker Str. 100, 14109 Berlin, Germany.
in a magnetic field of 0.97 T, which was rotated with 29 rpm during measurements and with up to 3000 rpm between measurements (see Refs. 6 and 13 for experimental details).

Figure 1 illustrates the geometry of the experiment. The axes of the sample holder coordinate system (index \( s \)) are parallel to the bottom and back surface of the sample holder, while the axes of the sample coordinate system (index \( s \)) are parallel to the edges of the samples. The \( x_s \), \( y_s \), and \( z_s \) surfaces of the sample were cut parallel to the crystallographic \{100\} plane. The \( x_{sh} \), \( y_{sh} \), and \( z_{sh} \) surfaces were cut parallel to crystallographic \{110\} planes. The samples were glued to the sample holder with angles of 45° between \( x_s \) and \( x_{sh} \) and also between \( y_s \) and \( y_{sh} \). Because of the tiling of 45°, the rotation axis of the magnetic field was parallel to the crystallographic direction (100). The magnetic field vector was always parallel to a \{100\} plane. Since prior to testing it was unknown in which plane of the sample twinning would occur, the samples were tested in two orientations to increase the chance that in one of the two orientations a certain twinning system would be activated. Hence, after the first set of experiments, the sample was rotated by 180° around \( y_{sh} \) and then by 90° around \( z_{sh} \).

Samples were glued to the sample holder back plate and to the sliding head (not shown in Fig. 1), which means that both sample ends were constrained by the glue. Sample A was cycled in each orientation for at least 20 000 magnetomechanical cycles (MMC). The integrity of sample and glue was checked after 2000 MMC so that artifacts resulting from a loose sample were excluded. Sample B was cycled for 300 MMC in the initial orientation and for 20 000 MMC in the second orientation, with a check for integrity after 20 000 MMC. A final check was carried out after the rotating field experiments were finished.

Sample A was furthermore tested with the \( y_s \) axis once in horizontal and once in vertical orientation for 20 000 MMC each. After that, sample A was thermomechanically treated (i.e., heated to austenite and then cooled with a stress of 10 MPa applied in \( z_s \) direction) and tested for 20 000 MMC in the orientation with \( y_s \) being horizontal to the sample holder. Then the sample was mounted in the second orientation with \( y_s \) at 45° to the sample holder and magnetomechanically cycled for 500 000 cycles.

The MFIS versus the number of MMC is given in Fig. 2. Sample A showed a larger MFIS than sample B, even though both samples have received identical treatments. Each inspection of the mounting of the samples influenced the measured MFIS. This is attributed to an artifact due to slightly different friction forces related to the condition of the glue. Sample A exhibits MFIS in both directions while sample B exhibits nearly no MFIS in the initial orientation.

Examples for the MFIS during one cycle of the magnetic field are given in Fig. 3. The strain at 10 000 MMC after the inspection is shown for sample A, while the strain at 100 MMC is given for sample B. Both MFIS values are measured during tests with the second sample orientation.

The additional tests of sample A showed MFIS in all four mounting orientations (always with \( z_p \) parallel to \( z_{sh} \), Fig. 4). For the last two tests of sample A, after the thermo-mechanical treatment and mounting with \( y_s \), \( x_{sh} \), the MFIS was at the detection limit of 10 ppm for 500 MMC before it increased to 450 ppm and then decreased again to the detection limit (Fig. 4, open triangles pointing to the right). After removing the sample and remounting it in the second orientation with \( y_s \) at 45° to the sample holder, the MFIS increased to up to 1700 ppm (Fig. 4, half filled squares), which is ten times larger than the MFIS in NM martensite observed so far. Variations of the MFIS are attributed to variations of the stiffness of the glue joint between sample and sample holder. These variations affect the constraints on the sample surface. Any constraints reduce the MFIS. Thus, the largest detected MFIS represents a lower limit of the MFIS, which would be available without constraints. This MFIS is still about two orders of magnitude smaller than the theoretical 1 − \( a/c \), which points to the “ineffective” character of the training as discussed in Ref. 13. Unlike effective training,
which significantly affects the twinning stress, ineffective training hardly impacts the twinning stress. Such an effect would be below the resolution of the instruments used here. Theoretical studies predict that temperature has a stronger effect on the twinning stress. Temperature dependent measurements of the MFIS are anticipated to study these effects.

In conclusion, the large MFIS of 0.17% represents unequivocal evidence of the magnetic-field-induced twin boundary motion in NM martensite, the experimental fact which can evoke a burst of activity similar to the one already observed after discovery of ~0.2% in a modulated Ni–Mn–Ga martensite. This result opens up new opportunities in searching for the training conditions of FSME materials exhibiting NM martensite. These results presented here make NM martensites technologically attractive with large MFIS in an extended temperature range and acceptable ductility.

V.A.C. is grateful to Basque Government, Department of Education for financial support (Project No. IT-347-07). P.M. is grateful to ETH Zürich for the donation of the magneto-mechanical testing device. M.C. appreciates the support of this work by grants of the Deutsche Forschungsgemeinschaft (Grant No. SPP1239 Schn 1106/1). P.M. and M.C. acknowledge financial support of DOE BES Contract No. DEFG-02-07ER46396.