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Large magnetic-field-induced strains in Ni-Mn-Ga alloys in rotating magnetic field

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Abstract: A single-crystal samples of near-stoichiometric Ni_2MnGa alloys with large magnetic field induced strain (MSM effect) at room temperature have been investigated. It was found that there is a reversible reorientation of easy magnetization direction and magnetic domain structure under applied magnetic field. By means of x-ray Laue diffraction method and difractometric analysis it was ascertained that these phenomena are connected with the growth of martensitic twin variant with the short crystallographic axis (*c*-axis) directed along the applied magnetic field. Furthermore, by means magneto-optical investigations we have found the correlation between martensitic microstructure and magnetic domain structure. The field-induced reorientation of martensite was able to change the dimensional lengths of the samples more than 4.5 %.

1.Introduction

Magnetic field control of the shape of ferromagnetic alloys with martensitic structure was suggested recently [1, 2]. Such materials are highly beneficial for actuators [3, 4]. Field-induced strains, larger than 4 %, have been observed in the single-crystal Ni_2MnGa alloys [5, 6]. There are no detailed studies concerning the martensitic microstructure changes connected with MSM (magnetic shape memory) effect. The aim of this work was to fill up this gap.

2.Experimental procedures

The alloy $Ni_{48.7}Mn_{30.1}Ga_{21.3}$ was melted in an induction furnace in argon atmosphere. The composition of the alloy was measured by wave-length dispersive spectroscopy (WDS). After homogenization at 1000° C during 3 days and aging at 800° C 1 day the alloy was air cooled to room temperature. X-ray diffraction measurements revealed the Heusler type ordered structure ($L2_1$) for the alloy in austenitic state. Martensitic transformation points M_s, M_f, A_s, A_f, and Curie point T_c were measured using low field *ac* magnetic susceptibility technique (M_s=29° C, M_f = 26° C, A_s = 32° C, A_f = 35° C, T_c = 99° C). Samples for magnetic investigation with dimensions of 4×4×4 mm were cut using spark cutting machine and one single $Ni_{48.7}Mn_{30.1}Ga_{21.3}$ grain.

All the *M-H* curves were obtained by using a vibrating sample magnetometer (VSM). The specimen stage was mounted between the pole pieces of a 1 T electromagnet and could be rotated for different field orientations. In all measurements the rotation of the samples was carried out at zero magnetic field. Strains induced by magnetic field were measured by strain gauges. The rather large changes in the sample shape and size gave us the possibility to confirm the strain gauge data by micrometric measurements. Temperatures were carefully controlled with a temperature control unit, *i.e.* a large volume alcohol circulator.

Back-reflection Laue technique was used to determine the change in an orientation of crystals. X-ray tube with W-anode was used. A low voltage level V = 12 kV (I = 30 mA) was applied in order to avoid the characteristic line effect and to decrease the fluorescent radiation from sample. The distance between the film and the sample was 30 mm. Indexing and orientation analyses were carried out by using the OrientExpress v.3.3 program [7].

Philips X'Pert MRD diffractometer equipped with Co-tube, x-ray lens and thin film collimator have been used to study martensitic structures.

Magneto-optical (MO) method based on the Faraday effect have been used for the magnetic microstructure observation [8, 9]. The angle of Faraday rotation of the polarization plane of the light is proportional to the component of local magnetic field in the direction of propagation of the light. Transparent magneto-optically active ferrite-garnet indicator films with in-plane anisotropy [9] was placed on the surface of the sample. The vector of magnetization of the indicator film rotates in the direction of the sample local magnetic field, producing a normal to the sample surface component of magnetization. A Leica DMR polarized light microscope was used for the visualization of magnetic microstructure contrast. MO images were obtained at zero field with near cross orientation of the polarizer and the analyzer.

3.Results and discussion

In the previous article [10] it was shown that the short martensite *c*-axis (<001> direction in cubic coordinates) is the easy axis of magnetization and that the initial slope of *M-H* curves is conditioned by the volume fractions of martensitic variants, in which the easy axis of magnetization is directed along the field.

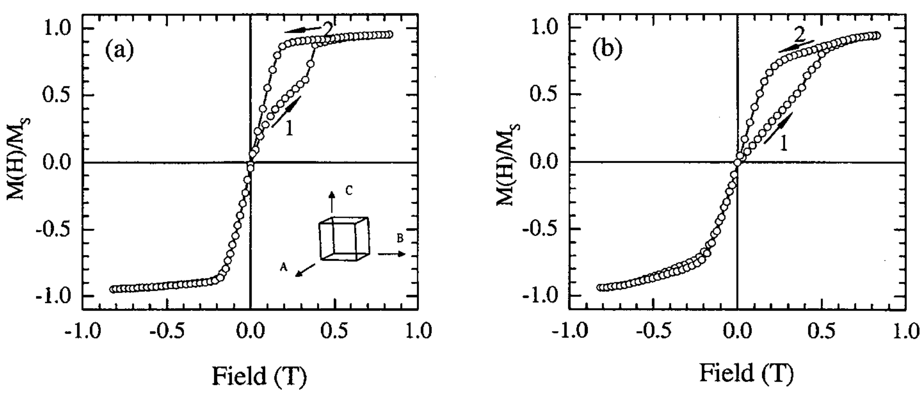

Fig. 1 *M-H* curves at 25 ^{0}C (a) magnetic field is parallel to *A*-direction, (b) – magnetic field is parallel to *B*-direction.

Fig. 1 shows the *M-H* curves for the sample along the two sample cubic directions (*A* and *B*, see insert in the Fig. 1a). The *M-H* curves measured in the *A*-direction are shown in Fig. 1a. The initial part of the magnetization curve (marked as 1 in Fig. 2a) corresponds to the case where the fraction of the martensite variant with the hard axis of magnetization along the *A*-direction is dominant. At the field in the interval 0.3÷0.5 T magnetization abruptly increases and approaches the level of saturation. When the field is decreased the magnetization is staying nearly at the saturation level until it reaches the threshold of about 0.2 T. The subsequent magnetization loops have no particularities and they show saturation at low field, indicating that the easy axis of magnetization now is directed along the field (curve 2 on Fig. 2a). When the sample is rotated by 90° from the *A*- to *B*-direction along the field, the *M-H* curve again shows the same behavior (see Fig. 2b). The effect is fully reversible - 90° rotation of the sample from *B* to *A* position and vice versa does not change the peculiarities of the *M-H* curves. Consequently, under the influence of the magnetic field and within the interval of 0.3÷0.5 T one can observe the easy axis reorientation from the perpendicular to parallel to the field direction. We have to point out that if the magnetic field is applied along the third direction of the sample (*C*-direction), no changes of easy axis direction occurs and *C*-direction always coincides with the hard axis of magnetization.

The Fig. 2 shows the prominent changes in martensitic structure when the magnetic field is rotated. The back-reflection Laue diffraction patterns have been obtain from the same place of plane perpendicular to the *C*-direction. The lattice parameters, $a = 0.5944$ and $c = 0.5617$ nm (in cubic coordinates) [11], were used for indexing of the Laue diffraction patterns and for the angle calculations. In the Fig. 2a the Laue diffraction pattern and the corresponding Key diagram belong to the structure that was formed after the magnetic measurements shown in the Fig. 1a. The Fig. 2b characterizes the structure after the 90° field rotation that is after the magnetic measurements shown in the corresponding Fig. 1b. Only the high intensity spots were used for indexing. The rest can be connected with the fine (twin) structure of the martensite. From the comparison of these two pictures it is evident that the rotation of the magnetic field is associated with the reorientation of the martensitic structure. The Fig. 2c and 2d correspond to the two variant martensitic mixed structure, which was obtained by removing of the magnetic field after partially developed martensite reorientation (interval 0.3÷0.5 T in Fig. 1, curve 1). The simultaneous presence of the two sets of (Laue diffraction) spots, which originate from the different martensite variants, makes it possible to determine the angle ω between the *c*-axes (c_{V1} and c_{V2}, Fig. 2) of such variants. The angle was found to be $\omega = 93.2^\circ \pm 0.5^\circ$, which coincides with the angle for twin orientation of variants ($\mathrm{tg}(\omega/2) = a/c$).

The results of the Laue diffraction studies were re-confirmed by using the diffractometer measurements, presented in Fig. 3. The data in stereographic projection of the two variant martensitic structure (see Fig. 2d), shown in Fig. 3a, were obtained by means the texture type scan at $2\theta=74.0^\circ$ ((400) and (040) planes) and $2\theta=79.1^\circ$ ((004) plane). The angles between [100], [010] and [001] directions for each variant were found to be equal to $90^\circ \pm 0.5^\circ$. The two martensitic variants had a common [100] direction. The angle between [001] directions in two different variants was found to be equal with $93.2^\circ \pm 0.5^\circ$, whereas for the [010] directions it was $86.8^\circ \pm 0.5^\circ$. The Fig. 3b reveals the results of the θ-2θ measurements recorded from the plane perpendicular to the *B*-direction, after magnetization of sample with the field directed along the *B*-direction and along the *A*-direction. The [001] direction and the [010] direction become perpendicular to the plane, accordingly.

The results of the magneto-optical microstructural studies are in the Fig. 4. The Fig. 4a presents the same two variant martensitic structure as in Fig. 2d. The plane of observation is perpendicular to the *C*-direction of the sample. The stripe magnetic domain contrast follows exactly the *c*-axis direction in the martensitic bands. Each martensitic band have also a magnetic domain fine structure. The Fig. 4b and 4c show the MO images of the same plane for single variant martensite sample after the magnetization with the field directed along the *B*-direction and along the *A*-direction, respectively. From the comparison of these two pictures it is evident that the rotation of the magnetic field is highly associated with the about 90° reorientation of stripe magnetic domain contrast. Furthermore, it is largely connected with about 90° reorientation of the easy axis of magnetization. The Fig. 4d shows MO image of the plane which are perpendicular to the direction of magnetization (along *c*-axis or [001] direction). In such case the magnetic domain MO-contrast are largely different from the others and are close to the maze type.

The redistribution of the martensite variant proportions under the applied magnetic field found in this study enable the macroscopic deformation of the sample. The results presented in Fig. 4 confirm this conclusion. Curves 1 and 2 in Fig. 5 show the changes of the strain vs. magnetic field applied across the strain gauge axis. Then the sample was turned by 90° at zero field (*B* sample direction is parallel to the magnetic field), and subsequently the strain was measured as a function of the magnetic field (curves 3 and 4). Increasing the magnetic field after every 90° rotation of the sample arises strain which are found to be larger than 4.5 %. The changes of the sample dimensions due to rotation between the *A*- and *B*-directions are reversible, but applying the magnetic field along the *C*-direction causes no dimensional changes.

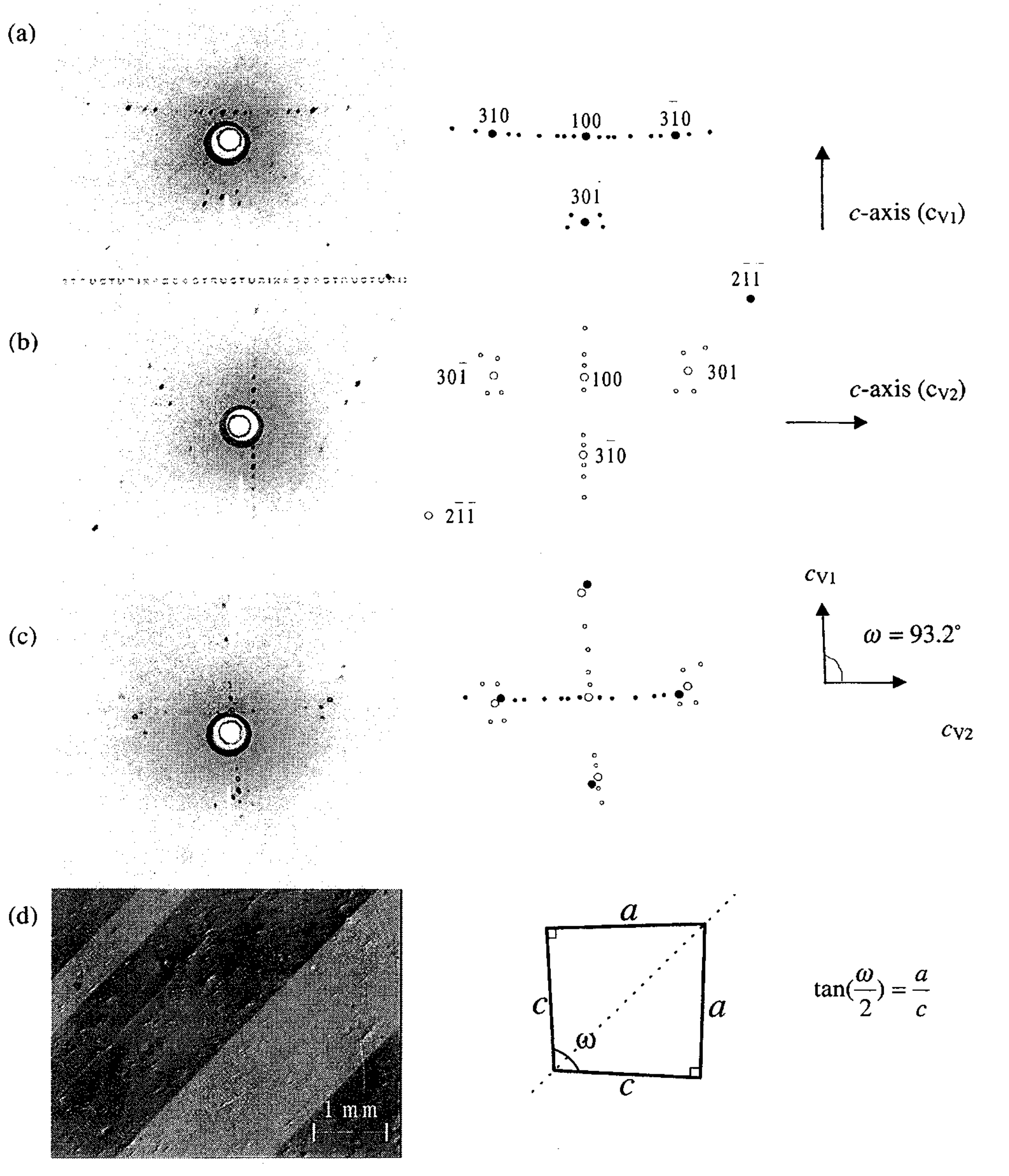

Fig. 2 Laue diffraction patterns and corresponding Key diagrams.
(a)– initial position, (b)–after 90° magnetic field rotation, (c)-intermediate magnetic field rotation position (two variant martensitic structure), (d)–optical image of the two variant martensitic microstructure corresponding to (c)

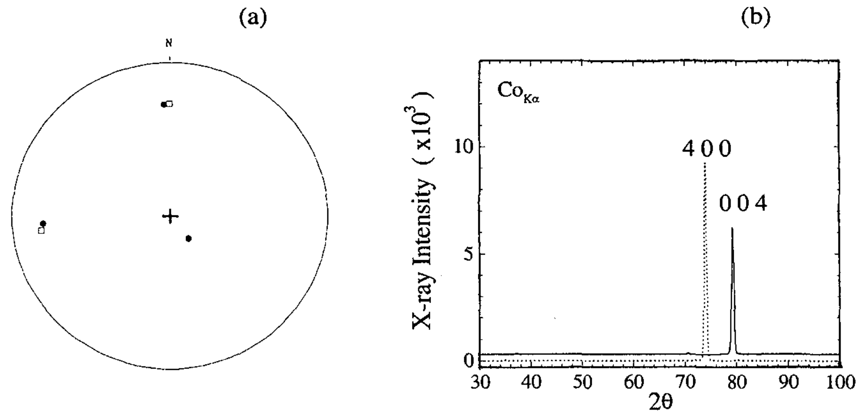

Fig. 3 (a) - Stereographic projection for the two variant martensitic structure; • - [100] and [010], -[001]; (b) - θ-2θ diffraction patterns from the plane perpendicular to the B-direction of the sample, magnetized along the B-direction (solid line) and along the A-direction (dot line).

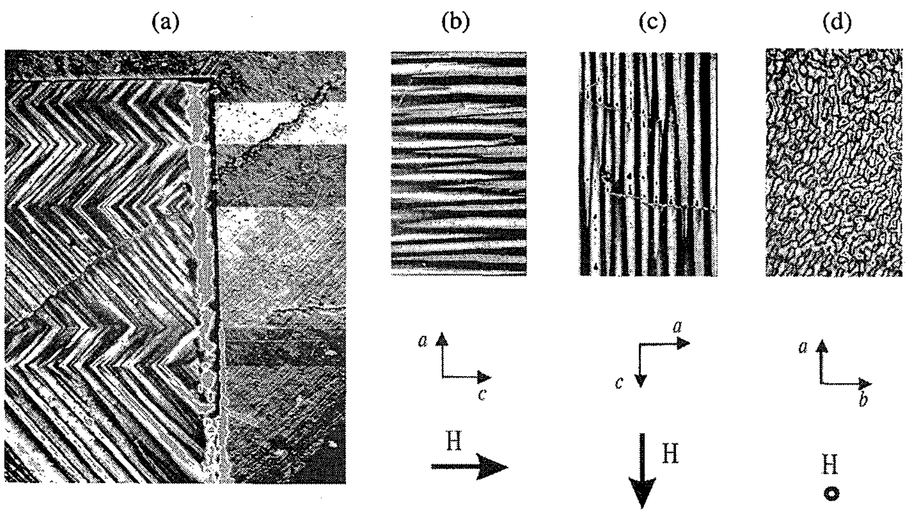

Fig. 4 Magneto-optical images of martensite at zero magnetic fild; a, b and c – crystallographic axes of martensite lattice, H - direction of magnetization.
(a) – two variant martensitic microstructure corresponding to Fig. 2d (right – optic, left – MO images); (b), (c) – single variant martensitic structure, H and c-axis are in the plane of observation; (d) - single variant martensitic structure corresponding to (a) and (b), the plane of observation is perpendicular to H and c-axis.

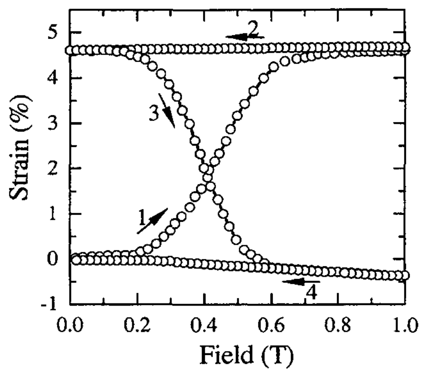

Fig.5 Strain vs. magnetic field at 25 ^{0}C for the sample preliminary magnetized along strain gauge axis; curves 1, 2 - magnetic field across strain gauge axis, curves 3, 4 – after 90^o rotation of sample, magnetic field along the strain gauge axis.

4.Conclusions

4.1 The reversible redistribution of two martensite variants was found under an applied rotating magnetic field. The rearrangement of the martensite variants is accompanied with the reversible reorientation of about 90^o the easy axis of magnetization and the short martensitic *c*-axis. The present results indicate that the reorientation occurs by the movement of the twin boudaries.

4.2 The rearrangement of the martensite variants under the influence of the magnetic field is accompanied with the reversible macroscopic changes of the sample dimensions are exceeding the amount of 4.5 %.

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