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Magnetic domain evolution with applied field in a Ni–Mn–Ga magnetic shape memory alloy

Yanling Ge ^{*}, Oleg Heczko, Outi Söderberg, Simo-Pekka Hannula

Laboratory of Materials Science, Helsinki University of Technology, P.O. Box 6200, FI-02015 TKK, Finland

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Abstract

The evolution of a magnetic domain pattern with an applied field in two perpendicular directions was studied in a Ni–Mn–Ga magnetic shape memory alloy by means of optical and scanning electron microscopy (SEM). The optical contrast of magnetic domains arises from surface undulation. This surface relief is coherent at the (101) twin boundary. However, the surface relief causes the trace of the (011) twin boundary to follow a zigzag pattern. These optical observations were confirmed by SEM Type II magnetic contrast.

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1. Introduction

Some Ni–Mn–Ga alloys are of significant technological interest, as they possess giant magnetic field-induced strain (MFIS) in the martensitic state. The MFIS is a result of structural reorientation under an applied field resulting from twin boundary motion. In the Ni–Mn–Ga alloy with a five-layered modulated martensitic structure, the easy magnetisation axis coincides with the shortest crystal axis and it can display a 6% MFIS [1].

In order to obtain a better understanding of the interaction between the elastic and magnetic properties, the magnetic domain pattern was studied using scanning electron microscopy (SEM) by the present authors [2]. It was found that the dominant martensite variant shows 180° magnetic domains and the second minor martensite variant exhibits a zigzag pattern commensurate with the 180° domains [2,3]. In speculating on the possible mechanisms for this morphology, it was suggested that the morphology should be studied by means of optical microscopy. It turned out that in this optical study not only can the morphology of the

second twin variants be studied but the magnetic domains may also become visible. These observations and the evolution of observed structures in a magnetic field are reported here, and the possible contrast mechanism is discussed.

2. Experimental

Ni_{49.5}Mn_{28.6}Ga_{21.9} alloy was produced by AdaptaMat Ltd.; the annealing procedure was the same as in Ref. [2]. The phase transformation and magnetic transition temperatures of the annealed alloy were determined with a differential scanning calorimeter (DSC) of the Linkam-600 type and the alternating current low-field magnetic susceptibility method. The Curie point (T_C) of the alloy was 370 K, while the start (M_s) and finish temperatures (M_f) of the martensitic reaction were 305.8 K and 299.8 K, respectively, and those (A_s, A_f) for the reverse reaction 314.4 K and 318.5 K, respectively. The martensitic crystal structure is tetragonal, with five-layered modulation.

For the optical microscopy an 8 mm × 5 mm × 4.5 mm specimen was spark-cut from a single crystal. An observation surface with the dimensions 8 mm × 5 mm and corresponding to the crystallographic plane (010) was wet-ground and electropolished in a solution of 25% nitric

^{*} Corresponding author. Tel.: +358 09 4512688; fax: +358 09 4512677.
E-mail address: Yanling.Ge@tkk.fi (Y. Ge).

acid + ethanol at 273 K. The optical observations were carried out with a Leica DM RX microscope using dark field illumination. The same areas were also studied with a LEO-1450 SEM using the backscattered electron image (BEI) mode. The chemical composition of the alloy was determined using an energy dispersive spectroscope (EDS) attached to the LEO-1450. The magnetic field was always applied in two perpendicular directions, i.e. the $\langle 100 \rangle$ and $\langle 001 \rangle$ directions. The directions are defined using the coordinate system of the cubic parent phase.

3. Results and discussion

3.1. Optical observations

Prior to the mechanical polishing the specimen was magnetised in a 1 T field along the short dimension of the observed surface in order to avoid any disturbance of the magnetic contrast by the surface relief in the multivariant martensitic state. This treatment creates a nearly single variant state, with a single crystallographic c -axis along the applied field direction. During mechanical polishing and electropolishing the specimen was held along the crystallographic c -axis. This was necessary because the twinning stress, i.e. the stress needed to rearrange the martensite twins, is less than 2 MPa for the alloy under study, so any other grip would destroy the single variant state and create a new and unwanted multivariant structure.

After electropolishing, the specimen had a flat smooth mirror-like surface without any feature contrast except for a few twin traces at the corner of the specimen. The 1 T magnetic field applied along the long dimension of the surface under observation resulted in reorientation of the structure as a result of the magnetic shape memory (MSM) effect. After the removal of the applied field the 180° magnetic domains and some twin traces appeared, as shown in the optical images in Fig. 1(a)–(c). The small fraction of twin traces and magnetisation curve imply that after magnetising the specimen was again in a nearly single variant state, with the c -axis along the field and containing some residual variant bands preserving previous orientations. As the observation surface is the (010) plane, the two kinds of visible twin traces correspond to the (101) and (011) twin planes.

The optical microscopy shows that 180° domains run through the whole observed surface; see Fig. 1(a). This magnetic contrast is also visible in the bright field illumination, but the dark field illumination enhances the contrast considerably. The contrast can be further enhanced to some extent by polarised light. However, this magnetic contrast is believed to truly result from the surface relief, without any contribution of the Kerr effect, as the Kerr rotation in Ni–Mn–Ga alloys is reported to be close to zero [4].

Fig. 1(b) shows the magnetic domain pattern of (101) twins. The structure consists of the major twin variant with the c -axis along the field direction and the minor, residual

variant with c -axis perpendicular to the magnetising field. Since the crystallographic c -axis is in plane for both (101) twins, the magnetisation follows the c -axis in adjacent twins, forming a staircase-like pattern with a 90° domain wall that coincides with the twin boundary. This kind of pattern was observed and discussed previously [5]. The domain width varies from several microns to several dozen microns. The 180° domain walls are not straight, which may be due to the fact that the orientation of the surface deviates slightly from the ideal orientation. Additionally, two (011) twin traces, the nearly vertical lines on the right edge of the figure, are apparent. These are discussed later.

Fig. 1(c) again shows the 180° domains running through the structure horizontally along the direction of the previously applied magnetic field and thin, minor (011) twins, which have their c -axis perpendicular to the surface. These (011) twin traces are seen as the vertical slightly curved black lines in the figure. The (011) twins cross the magnetic domains and the (011) twin traces have a zigzag shape commensurate with the domain width. This pattern was observed for the first time by SEM [2].

Besides these two features, there is another weak contrast present in Fig. 1(c), i.e. the vertical grey strips. We suppose that these strips are the residual surface relief contrast originating from the previous magnetic domains. This can be explained as follows. Before polishing, the sample was magnetised vertically. After the removal of the field both the vertical 180° domains, together with surface relief, appeared on the surface. During polishing this surface relief was removed and the surface was perfectly flat. After the subsequent magnetising of the sample in the horizontal direction new vertical grey bands of contrast appeared as the negative of the previous surface relief. They are still visible in Fig. 1(g) and (i), after magnetisation in different directions, and they cannot be removed unless the surface is mechanically polished again.

Fig. 1(d)–(f) shows the domain structure after the application and removal of the 1 T field in the [001] direction, i.e. along the short dimension of the observed surface (vertically). This is the direction of the field used before polishing. Fig. 1(d) shows that the previous horizontal 180° domains disappeared and new vertical 180° domains appear following the applied field direction. This indicates that structural reorientation occurs and the major variant has its c -axis in the direction of the applied field. It is noted that the crack shown in Fig. 1(a) grows after the application of the field.

Fig. 1(e) shows the same area as Fig. 1(b), but in this case the magnetic domain contrast almost disappears, except for several small domains near the crack. This indicates that the surface is again as smooth as it was after polishing. It does not mean that there are no magnetic domains and we can assume that domains similar to the structure visible in the right area of Fig. 1(d) exist. The non-existence of the contrast only means that there is no surface relief connected with the magnetic domains. A

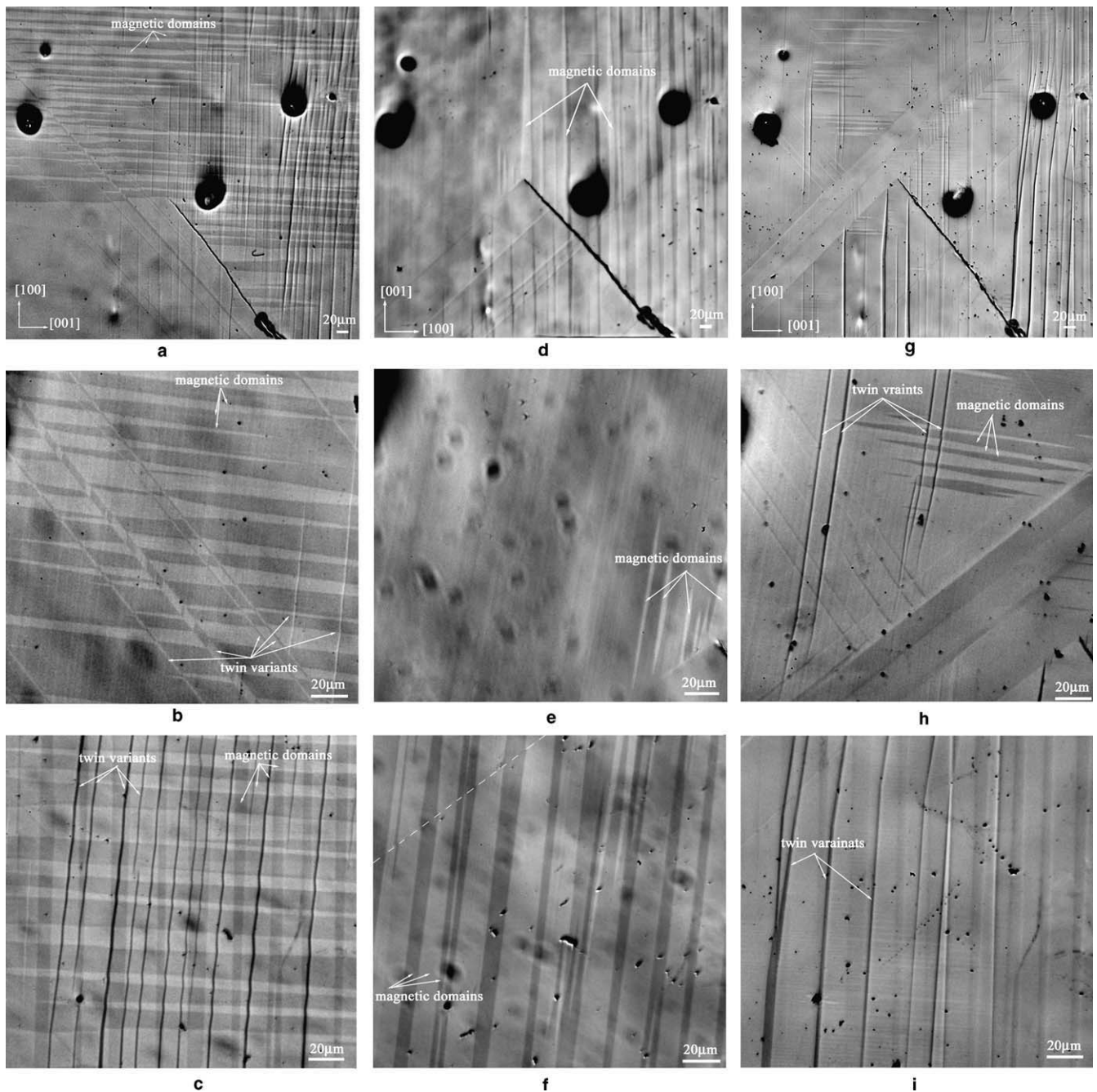


Fig. 1. The evolution of the magnetic domain structure with the changing magnetic field directions that are in-plane with the specimen surface. Prior to polishing, the magnetic field of 1 T was applied to the short dimension of the surface being observed (vertically in the images). Then, after polishing, the field of 1 T was applied along the long dimension of the surface (horizontally). The resulting images are shown on the left (a)–(c). In the images in the middle (d)–(f), the previous field was applied along the short dimension of the surface (vertically), and in the images on the right (g)–(i), again horizontally. The images in the second and third rows are magnifications of the upper-left and bottom-right areas of the images in the first row, respectively.

similar situation occurs in the bottom left of Fig. 1(a), where no contrast is visible.

Fig. 1(f) shows the 180° domains of the major variant following the vertical direction of the previous magnetic field. These new 180° domains occupy the position of the previous residual surface relief, which is visible in Fig. 1(c) as weak vertical grey strips. One thin (101) twin trace in the top left area (indicated by a dashed line) forms

a 90° domain wall, which is visible as these vertical domains become dislocated to a certain extent when there is intersection with this thin (101) twin trace. This additionally confirms that the magnetic domain contrast is due to surface relief. Unlike the domain contrast, the minor (011) twins fully disappear.

In the third step the 1 T field was applied along the [001] direction and then removed again, i.e. along the long

dimension of the observed surface. The resulting domain structure is shown in Fig. 1(g)–(i). Surprisingly, the domain pattern and twinning morphology are much more complex and totally different from those in Fig. 1(a)–(c). Generally, the 180° domains still more or less follow the applied field direction. Additionally, the (101) $(10\bar{1})$ and (011) twins are all visible on the observed surface. The residual surface relief can still be seen in Fig. 1(i). The fine horizontal domain contrast is faintly visible in the centre of Fig. 1(i), while the magnetic contrast is lacking in the rest of the area. Notably, the crack in Fig. 1(g) has grown further. The (011) twins are formed even though there is no apparent external (magnetic) driving force for their formation. This may be a result of the accommodation of internal stresses developed during magnetic field application.

From Fig. 1 it is clear that both magnetic patterns and twin morphology become more complex when the field is switched and alternated three times only without any notable external stress. The observed crack grows considerably during the process, too. This may suggest that the imperfections accumulate when one variant transforms to another. The magnetic domain and martensitic twin nucleation are mutually dependent and a dynamic process. The twin boundary motion will take a unique path determined by the interaction with the local imperfections. Although the field-induced strain is reproducible, the surface morphology is not reversible. This observation illustrates the difficulties involved in studying the morphology and magnetic domain structure in MSM materials. Although the specimen exhibits the same macroscopic behaviour, the path taken to reach the final state is different and magnetic domains and martensitic variants attain different metastable configurations.

Fig. 2 shows the domain pattern near a $(10\bar{1})$ twin boundary observed by optical and electron microscopes. The optical image, Fig. 2(a), suggests a fir tree-like domain across the twin boundary, while the BEI in Fig. 2(b) shows that the fir tree-like domain is a part of a large 180° domain structure. This confirms that the optical magnetic contrast is due to the surface relief. If there is no contrast in the optical image, such as the big area in e.g. Fig. 1(h) or (a), either the magnetic domains are not accompanied by

surface relief or the region may consist of very fine domains. The observed pattern of surface relief close to the twin boundary may be promoted by greater internal stress in the vicinity of the twin boundary.

Fig. 3 shows a detail of the same area as shown in Fig. 1(i) using back-scattered electrons. Although the optical micrograph is nearly featureless, the backscattered electron image reveals fine vertical magnetic domains. The whole area consists of very fine domains with a width of less than $2\ \mu\text{m}$. Additionally, the residual surface relief of previous magnetic domains and newly formed (011) twins is clearly visible in Fig. 3.

3.2. Model and discussion

Direct optical magnetic domain observation has previously been reported for a Terfenol-D crystal, where the magnetostriction effect caused the surface tilting [6]. In our case, with a Ni–Mn–Ga alloy, it is believed that the optical magnetic contrast is also due to the surface undulation; however, its origin is different from that of Terfenol-D. When the saturation magnetic field is applied, the specimen adopts a single domain structure and contracts by 6% in the magnetisation direction. However, some residual variants with different orientation can remain in the material and, therefore, the material is strained. Other reasons for the strain might be inclusions, point defects or other imperfections of the lattice.

We assume that the strain is caused by residual variants, which are not changed during magnetisation, i.e. no MSM effect takes place in these variants. These variants have a short crystallographic axis (c -axis) roughly perpendicular to the c -axis of the surrounding major variant. This different behaviour in a magnetic field can generate additional strain, which results in compressive stress.

After removal of the magnetic field, the magnetic vector of residual variants rotates back towards their easy axis and the compressive stress is gradually reduced by reverse movement of the twin boundaries. This results in reverse elongation in the direction of the previous magnetisation – strain up to 0.41% has been observed [7]. Elongation in

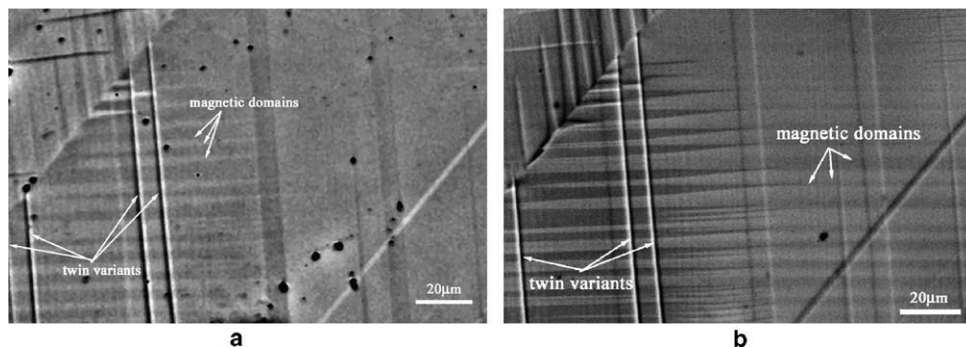


Fig. 2. (a) An optical image with a fir tree-like magnetic structure close to the twin boundary; (b) back-scattered electron image from the same area, revealing again the fir tree-like structure extended with 180° domain by utilising Type II magnetic contrast. This demonstrates that the magnetic contrast visible in the optical image is due to surface relief and that the surface relief is non-homogeneous.

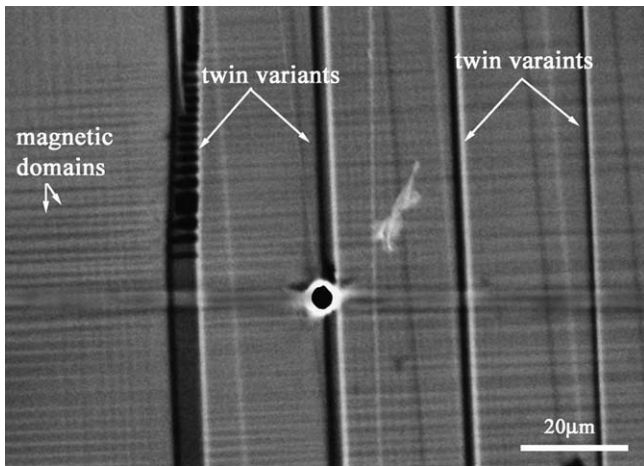


Fig. 3. Back-scattered electron image reveals the fine magnetic domains by utilising Type II magnetic contrast, which are nearly invisible in the optical image, Fig. 1 (i).

one direction will result in a contraction in perpendicular directions as a result of volume conservation.

In the saturation magnetic field the whole sample is a single magnetic domain. When the field is reduced, the single domain breaks into a multidomain structure by the demagnetisation field. The magnetic domains start to nucleate at the surface. The free surface may undergo less contraction than the bulk material under the surface as a result of its unconstrained condition. This produces the roofed or corrugated shape of the surface, as illustrated in Fig. 4(a). Simultaneously, the surface undulation may assist the nucleation of the domain wall. Finally, the domain walls are pinned at the ridges and the valleys of the surface.

On the other hand, it might also be that the nucleated domain walls serve as a place where surface corrugation or ridges and valleys occur initially. Consequently, the magnetic domains accompanied by surface relief are the result of the accommodation of the internal strain and

magnetic interactions between differently oriented twins in order to reduce the internal energy. This mechanism can be quite non-homogeneous and random and, thus, it may lead to different metastable configurations during repeatable magnetisation.

The ridges and valleys, which coincide with the domain walls, follow the domain walls across a twin boundary, without a change in height between the ridge and the valley. Thus, the (101) twin boundary will keep straight, and roofed surface morphology will continue in the direction of new magnetisation (and crystallographic) nearly perpendicular to the previous direction, creating the staircase pattern [2,5]. This surface undulation is totally compatible when domains cross the (101) twin boundary.

The (011) twin trace is a different case. When the surface undulations meet at a (011) twin boundary, the lifted surface causes the (011) twin boundary trace to deviate from a straight line, because the (011) twin boundary intersects with the (010) surface at 46.7° (for $a = 5.95 \text{ \AA}$ and $c = 5.59$ of martensite lattice parameter of the alloy under study). As the twin trace crosses the ascending and descending parts of the corrugated surface, the (011) twin trace forms a zigzag pattern on the projection to the surface, i.e. the (010) plane. The angle derived from the (011) twin boundary's zigzag shape can be used to calculate the extent of surface undulation.

Let θ be the intersection angle between the (011) plane and (010) plane in martensite, i.e. $\theta = 46.7^\circ$ for the alloy under study, α be the angle between the projection of the (011) twin trace on the (010) plane and the [100] direction and β be the angle of the tilted surface; see Fig. 4(b). Then by a geometrical relationship the angle β is given by

$$\tan \beta = \tan \alpha \tan \theta, \tag{1}$$

where α is the half of the angle between two adjacent lines of the zigzag twin boundary trace. The average angle measured from Fig. 1(c) is 1.85°; the calculated tilt angle β is 1.96°. The contraction strain needed for the observed tilting can be calculated from the β angle. The relationship is

$$\frac{\Delta l}{l} = 1 - \cos \beta, \tag{2}$$

where l is the original length on the surface, and Δl is the contraction of bulk material under the surface, as shown in Fig. 4. The average strain that causes the observed tilted surface is 0.059%, corresponding to an elongation of 0.11% in the direction of magnetisation. This is smaller than the previously reported reversible strain of 0.41%, indicating that the assumed mechanism is feasible and also that the compressive stress might not be completely released by surface relief.

4. Conclusions

The evolution of the magnetic domain structure in $\text{Ni}_{49.5}\text{Mn}_{28.6}\text{Ga}_{21.9}$ exhibiting a 6% MSM effect using direct optical observation has been reported. By applying a 1 T

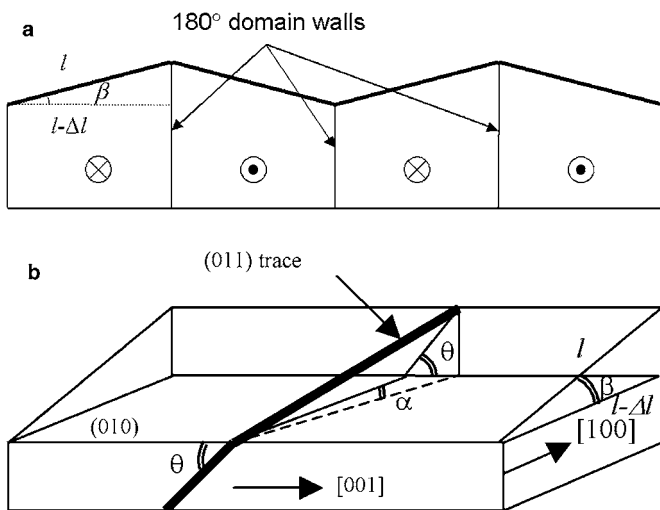


Fig. 4. (a) Illustration of the surface relief and magnetic domain configuration; (b) the model of projection of (011) twin configuration on a tilted surface.

field in two directions alternately, a unique magnetic domain and twin morphology configuration were observed, even though the field-induced strain or MSM effect was reproducible. After the field had been switched off three times and each time turned in a perpendicular direction, the magnetic and twin configuration changed to a more complex one. The observed magnetic domain pattern was due to the surface relief, caused by the less strained surface compared to the bulk material under the surface. The zigzag pattern of (011) twin traces in the (010) plane was explained by surface relief. The extent of the surface tilt to produce the observed zigzag pattern was about 2° .

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