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Direct optical observation of magnetic domains in Ni–Mn–Ga martensite

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This letter reports the direct optical observation, i.e., without polarization, of the magnetic domain structure explained by a large surface relief in Ni–Mn–Ga martensite. The authors suggest that the relief is due to the different straining of the surface and the bulk caused by the internal stresses associated with the magnetic shape memory effect. As a result of the relief the projection of the (011) twin traces upon the (010) plane creates the observed zigzag pattern. The surface tilt angle calculated from the zigzag pattern is ~3°. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2335811]

The Ni–Mn–Ga alloys have attracted intensive interest because they exhibit in certain martensite phases a giant magnetic field-induced strain, which is much larger than the observed in the conventional magnetostrictive materials.[1–4] The mechanism for the magnetic field-induced strain or the magnetic shape memory effect (MSME) is the crystal structure reorientation when one martensitic twin variant, which has the short crystallographic *c* axis and also the easy axis of magnetization along the applied magnetic field, grows on the expense of other variants with different orientation.[1–6] This results in large shape changes up to 10%.

The studied alloy has approximately tetragonal structure with a five-layered modulation along [110] direction in the martensitic state. As a result of cubic to tetragonal transformation, there are three deformation martensite variants with different orientations. There is one twinning system with two kinds of twin planes, (101) and (011), when referring to the tetragonal structure of the major martensite variant. The crystal axes of tetragonal martensite originate from cube edges of the parent phase. The magnetic domain structure of the twinned martensite exhibiting the MSME has been interpreted previously.[7,8]

Here, we report the observation of magnetic domain patterns by optical microscopy using nonpolarized light. The optical contrast indicates a large surface relief associated with magnetic domains. Similar optical observation of magnetic domain structure due to the surface relief has been reported in Terfenol D.[9] The observed surface relief in the optical study is identical with the magnetic domain structure detected by scanning electron microscope (SEM). This surprising and unexpected observation suggested, and indeed facilitated, a new explanation for the observed zigzag pattern of minor twin martensitic variants first reported by Ge *et al.*[7]

A single crystal sample of $Ni_{49.5}Mn_{28.6}Ga_{21.9}$ with dimensions of $4\times5\times9$ mm^3 was cut from an ingot manufactured by a modified Bridgman method in AdaptaMat Ltd. The detailed sample preparation process and characterization will be published elsewhere. The sample exhibited about 6% MSME. For observation of the magnetic domains the sample was magnetized to saturation in 1 T field along the longest dimension of the specimen. Now, the specimen was nearly in the single variant state in which the major twin variant had the *c* axis along the field. However, there were also traces of two other minor variants. The studied surface was parallel to the field and could be considered as derived from the (010) plane of the parent cubic phase. The optical images were carried out by a conventional optical microscope with normal incidence. Even though the polarized light source may enhance contrast, the present magnetic contrast was well visible without polarization. All optical images presented in this letter were obtained using dark field illumination without polarization. In addition, the type II magnetic contrast of SEM backscattered electron image (BEI) is used to observe magnetic domains and magnetic domain wall contrast to confirm optical images. This type of magnetic contrast is enabled by the particular setup of SEM, where the electron beam is perpendicular to the sample surface and backscattered electrons are detected with a four quadrant backscattering detector under an objective lens.[7,10]

Figure 1 shows the optical image of the magnetic domain configuration of the major variant when it is crossing the (101) twin boundaries. The two minor (101) twin variant runs from top-left to bottom-right, one with width ~20 μm and one less than 2 μm. The 180° domains with width of 3–30 μm are horizontal in major variant and vertical in minor (101) twin variants. The observed structure is very simi-

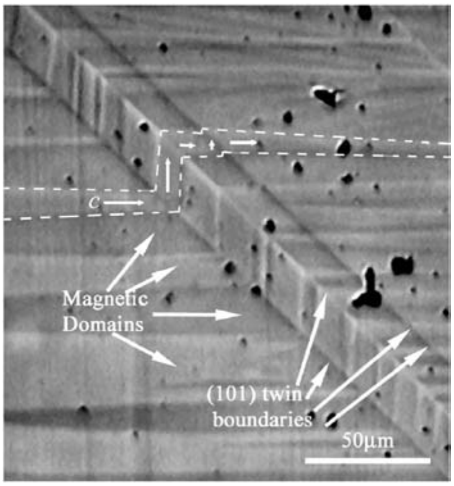

FIG. 1. Optical image shows a staircaselike magnetic domain configuration crossing diagonal (101) twins. One domain is outlined by the dashed line and *c* axis in different twin variants is indicated by the arrow.

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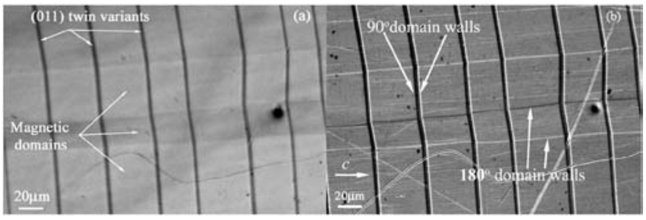

FIG. 2. (a) Optical image of 180° domains (broad horizontal band) and (011) twins (dark vertical line). (b) Backscattered electron image (BEI) shows the same area as in (a); the domain wall contrast is visible as the alternating white and dark lines. The *c* axis of the dominant variant is horizontal and the *c* axis in the minor variant (dark vertical lines) is perpendicular to the observed surface.

lar to that observed by SEM (Ref. 7) or by Bitter pattern.[6] The easy axis is in-plane in both twins and the magnetization follows the easy axis in each twin variant, creating a staircaselike pattern. The detailed interpretation of this kind of magnetic domain pattern has been published previously.[7,8]

Figure 2 shows the magnetic domain structure of the major twin variant observed both by the optical and electron microscopes. Figure 2(b) is the backscattered electron image obtained from the same area as the optical image in Fig. 2(a). Both images show the same 180° magnetic domain pattern with domain width larger than 20 μm and the additional contrast arising from the minor twin variant with width ~ 3 μm. The 180° domain structure is typical for a uniaxial material with the easy magnetization axis in-plane. Magnetic domain wall contrast is also clearly visible as the alternating white and dark lines in the BEI.[7] This indicates that the broad bands are the magnetic domains with antiparallel magnetization vector. The existence of the domain contrast in the optical image [Fig. 2(a)] indicated that there is a surface relief corresponding to the magnetic domains.

In Fig. 2, the 180° domains of the dominant twin variant are more or less along the [001] direction, though some small deviation originates from the orientation deviation of sample surface from (010) plane. The traces of the (011) twinned minor martensitic variant form nearly parallel zigzag patterns. This peculiar zigzag pattern was observed for the first time by BEI.[7] It was then proposed that the zigzag pattern arises from the magnetoelastic interaction between the variants.[7] However, after an intensive research of the subject and considering different possible explanations, we have come to the conclusion that the zigzag pattern is actually a trace of the (011) twin plane in the descending and ascending parts of the surface relief projected upon the (010) plane.

The model of the surface relief associated with one magnetic domain such as in Fig. 2(a) is schematically drawn in Fig. 3. The projection (observation) plane is (010) and θ is the angle between (010) and (011) planes, α is the angle between the projection of (011) on (010) and [100], and β is the angle between the ascending part of the surface relief and (010) plane. The angle β is a measure of the extent of the surface relief. The angle β is given by $\tan \beta = \tan \alpha \tan \theta$. If the ascending and descending parts of surface relief are considered symmetrical, the angle α can be measured as half of the angle between the projections of ascending and descending parts. The measured average value of angle α is about 2.8° from the first four (011) twin traces from left in Fig. 2. The angle θ can be obtained from the lattice parameters of martensite ($a=b=5.95$ Å and $c=5.61$ Å) to be 46.7°. Using these values the angle $\beta=3°$.

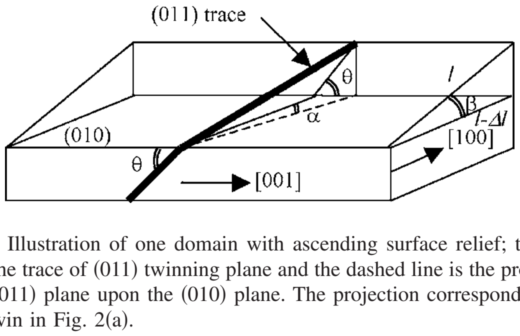

FIG. 3. Illustration of one domain with ascending surface relief; the bold line is the trace of (011) twinning plane and the dashed line is the projection of the (011) plane upon the (010) plane. The projection corresponds to the (011) twin in Fig. 2(a).

It should be noted that this angle varies locally. If the angle α is measured from the last two (011) traces from the left, the angle β can be as large as 5°. If the domain width $w=10$ μm and β is in the range of 3°–5°, the height of the relief, $w \tan \beta$, is between 0.52 and 0.87 μm. Observed local variation of the angle α can be explained as follows: Since the zigzag pattern arises as a two-dimensional projection of a three-dimensional surface on the (010) plane, any additional surface undulation will cause the projected trace to change its direction slightly. As a result the (011) twin trace within one domain is not parallel with its neighbor traces but is tilted; the extent of deviation of this trace depends on the domain width and the degree of the undulation.

The optical contrast of magnetic domains indicated in Fig. 1 suggests the alternating tilt of the surface due to the surface relief. The magnetic domain associated surface relief is continuous and compatible at the (101) twin boundary. The top of hills and the bottom of valleys of the surface relief follow the magnetic domain walls; thus, when the domain wall changes its direction while crossing the twin boundary, so do the hills and valleys of the surface relief. The height between the hills and valleys is unchanged; only their direction changes. The compatibility condition and the fact that the plane (101) is perpendicular to the (010) plane maintain the projection of the (101) plane into the (010) plane as a straight line. Having a fresh look on the previously published observations, the surface relief of the magnetic domain structure is quite nicely demonstrated also by BEI in topographic mode in Fig. 3(b) in Ref. 7.

The observed surface relief associated with the magnetic domains may be due to the constraint imposed by the strained bulk crystal on the unstrained surface. When the sample is magnetized to saturation, the MSME occurs and the sample contracts in the field direction. In saturation the whole specimen is in the single magnetic domain state, but not necessarily in the pure single twin variant state. When the applied magnetic field is decreased from saturation, the internal stresses developed during MSME will cause a partial recovery and the specimen elongates along the earlier magnetization direction. The recovery elongation, $\sim 0.41\%$ in the field direction, has been observed previously.[11] This elongation is accompanied by corresponding perpendicular contraction to keep the volume constant. However, the free surface may not contract by the same amount as the bulk material beneath it, resulting in the observed surface relief or undulation. The contraction $\delta = \Delta l / l$ creating the observed relief can be calculated from the observed angle as $\delta = 1 - \cos \beta$. Thus, the observed surface relief requires only a quite small amount of elongation, less than 0.27%, if compared with the MSME effect (6%). The detailed explanation and account will be published elsewhere.

Magnetic domain structure is formed when the magnetic field decreases from saturation and the demagnetization breaks the magnetic single domain to multidomain structure. At the same time the surface relief or surface undulation is formed. The magnetic domain walls follow the top of hills or the bottom of valleys of the undulation. We suggest that both processes, domain wall nucleation and surface undulation, occur simultaneously in order to reduce magnetostatic and magnetoelastic energies. However, to clarify which process controls the formation of the observed morphology—the surface relief or the magnetic domain structure formation—a detailed study of the dynamic process of demagnetization is required.

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