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Magnetically induced reorientation of martensite variants in constrained epitaxial Ni–Mn–Ga films grown on MgO(001)

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Abstract. Magnetically induced reorientation (MIR) is observed in epitaxial orthorhombic Ni–Mn–Ga films. Ni–Mn–Ga films have been grown epitaxially on heated MgO(001) substrates in the cubic austenite state. The unit cell is rotated by 45° relative to the MgO cell. The growth, structure texture and anisotropic magnetic properties of these films are described. The crystallographic analysis of the martensitic transition reveals variant selection dominated by the substrate constraint. The austenite state has low magnetocrystalline anisotropy. In the martensitic state, the magnetization curves reveal an orthorhombic symmetry having three magnetically non-equivalent axes. The existence of MIR is deduced from the typical hysteresis within the first quadrant in magnetization curves and independently by texture measurement without and in the presence of a magnetic field probing microstructural changes. An analytical model is presented, which describes MIR in films with constrained overall extension by the additional degree of freedom of an orthorhombic structure compared to the tetragonal structure used in the standard model.

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1. Introduction

Magnetic shape memory (MSM) alloys [1] are a new class of smart materials, which change their shape and magnetization under the influence of magnetic fields or stress [2, 3]. An external magnetic field can drive a shape change either by a magnetically induced martensitic (MIM) [4] transformation or by a magnetically induced reorientation (MIR) of twin variants in the microstructure of these materials. MIR occurs in the low temperature martensitic phase. This phase consists of differently oriented twinned martensitic variants, which are connected by easily movable twin boundaries. In an external magnetic field, the favourably oriented variants grow at the expense of those variants having their hard polarization axis close to the external field direction. Consequently, a shape change results as the length of the martensitic crystallographic axes are different. This effect was first observed in ferromagnetic Ni–Mn–Ga [1] single crystals and is also called the MSM effect or magnetic field-induced strain (MFIS). The maximum strain in Ni–Mn–Ga single crystals can exceed 10% [5], which triggered intense research on bulk materials. Similar effects have been found in antiferromagnetic [6] and paramagnetic [7] single crystals.

MSM materials open up opportunities for new functionalities in microdevices [8], e.g. micromotors and miniaturized linear actuators. For such applications, the integration of active materials in the shape of films is an absolute requirement. While progress has been achieved with active constrained epitaxial films made from related ferroelectric [9] and multiferroic [10] materials, investigations on MSM films have been mostly restricted to polycrystalline films [11, 12]. Epitaxial films are seen as the most promising approach as a significant strain in bulk MSM materials is only obtained in single crystals. Epitaxial growth of Ni–Mn–Ga has so far only been reported on GaAs(001) substrates using an Sc_{0.3}Er_{0.7}As buffer [13], on Al₂O₃(110) [14] and on MgO(100) [15]. The very recent report [15] showed that epitaxial films are also very helpful to examine the electronic properties at the phase transition by x-ray absorption spectroscopy. In these cases the materialities phase was only observed at low temperatures. As the low blocking stress of less than 3 MPa already inhibits actuation in the Ni–Mn–Ga film is not able to bend a thick rigid substrate

by MIR. Therefore indications of MIR at low temperatures have been so far only reported for partially free-standing films [16]. The fundamental precondition for most technical applications, however, is that films are ferromagnetic and martensitic at room temperature. Since both of these properties strongly depend on composition, combinatorial methods have been used to optimize transition temperatures in thin films [17].

The present paper focuses on a sample with one selected composition. The influence of the substrate–film interface is analysed. It is shown that the coupling of the film with the substrate controls not only the epitaxial growth but also variant orientations and reorientation in a magnetic field.

The simple phenomenology of MIR in bulk single crystals is usually pictured considering only two different twin variants and the redistribution of their volume fractions. The film material investigated here has a pseudo-orthorhombic structure with an important monoclinic distortion. As it will be shown, this low symmetry of the martensite is crucial for the activation of a constrained thin film. It allows for the existence of differently oriented twins, which enable stress release and variant rearrangement in the active layer in magnetic fields without the loss of coherent attachment to the substrate.

2. Experimental

Ni–Mn–Ga films were deposited on single crystal MgO substrates with (001) orientation using dc sputtering at various substrate temperatures from 300 to 600 °C. A film prepared at 400 °C was selected for detailed analysis. The thickness of the film was 0.47 μ m as determined by scanning electron microscopy (SEM) analysis from cross-sectional cuts prepared by a focused ion beam (FIB). The cross-section exhibited homogeneous contrast within the film crosssection, without any distinctive features. The composition of the film was determined to be Ni₅₂Mn₂₃Ga₂₅ with an accuracy of about 0.5 at.% by electron dispersive x-ray (EDX) analysis using a Ni₅₀Mn₂₅Ga₂₅ standard. Surface morphology was studied by polarized light optical microscopy, SEM with backscatter electron contrast and atomic force microscopy (AFM) in tapping mode (DI dimension 3100). The structure was examined by x-ray diffraction (XRD) using θ –2 θ scan with Co K_{α} radiation and texture measurements with Cu K_{α} in Phillips X'pert machines. The same texture measurement set-up was used to probe the microstructure changes in the constant magnetic field of about 0.2 T. For this, the sample was placed between two permanent NdFeB magnets connected by a yoke. Magnetic measurements were performed using a Quantum Design PPMS with a vibration sample magnetometer (VSM) addition.

3. Results and discussion

3.1. Structure

The XRD pattern measured in Bragg–Brentano geometry of a sample series deposited at different temperatures is shown in figure 1. The vertical dotted lines mark the positions of peaks corresponding to polycrystalline cubic Ni₂MnGa austenite (a = 0.5821 nm [18]). Two very pronounced and sharp peaks in the measured diffractograms originate from the single crystalline MgO(001) substrate with a lattice constant a = 0.4194 nm. The film deposited at 300 °C exhibits only a single (400) reflection, as expected for the austenite state (c/a = 1). As no reflections of other lattice planes are observed, this film exhibits a high degree of texture,



Figure 1. XRD scans in Bragg–Brentano geometry of Ni–Mn–Ga films deposited at different temperatures (measured at room temperature). The indices show the peak positions for polycrystalline stochiometric Ni₂MnGa austenite.

indicating presumably epitaxial growth. When increasing the deposition temperature to 350 and 400 °C, two distinctive convolutions of peaks close to the $(400)_A$ and $(200)_A$ austenite lines appear.

The convolution of the three peaks corresponds to the three different martensite variants having their crystallographic axes a, b or c aligned approximately perpendicular to the film surface. The highest intensity is observed for the central peak at a position that is slightly shifted from the position of the austenite peak. This central peak might originate from residual austenite and/or variants with the orientation of the b-axis perpendicular to the surface as discussed later. As Bragg–Brentano geometry only probes planes parallel to the substrate, these measurements cannot be used to evaluate volume fractions of the variants.

The three overlapping peaks close to the $(400)_A$ austenite peak can be indexed as a (pseudo-) orthorhombic phase with the lattice constants: a = 0.609 nm, b = 0.578 nm and c = 0.554 nm using the coordinate system related to the parental cubic austenite phase [19]. A similar peak splitting is observed for the $\{200\}_M$ peaks close to the austenite $(200)_A$ line. The presence of $\{200\}_M$ superstructure reflections indicates a high degree of chemical order in the off-stoichiometric film. Apparently, the elevated deposition temperature provides sufficient mobility for the atoms on the surface to achieve ordering in the Heusler alloy.

An increase of the deposition temperature to $450 \,^{\circ}$ C results in a structure with two peaks close to the $(400)_A$ austenite peak with even larger peak splitting giving a c/a ratio of 1.22. This indicates a tetragonal martensite as known also from bulk material. This martensitic structure is known to exhibit no MIR [20]. A further increase of deposition temperature to $600 \,^{\circ}$ C results in several reflections, which could not be indexed unambiguously.

All further analyses are restricted to the orthorhombic film deposited at 400 °C. Lattice parameters and the c/a ratio of 0.91 of this film are comparable to 7M bulk materials



Figure 2. The upper left picture shows the pole figure (Wulff projection) of the $(220)_A$ planes of the film measured at 336 K in the austenite state. Additionally, pole figures of the different $\{220\}_M$ planes of the film are measured at 300 K in the martensitic state. The red marked quarter of $(220)_M$ is intensified by a factor of three in order to show that part of the background consists of discrete reflections with low intensity. A square root intensity scale is used for all pole figures. The direction of the later applied external field *H* is given by the black arrow.

[19, 20]. In fact the orthorhombic cell reported in bulk materials is slightly distorted, resulting in a monoclinic cell [21]. The measured values of lattice constants and the electron density of e/a = 7.56 calculated from the measured composition indicate a modulated 7M structure. For this structure a 10% MIR effect was observed in the bulk [5].

3.2. Texture

To determine the orientation of the Ni–Mn–Ga unit cell, a $(220)_A$ pole figure has been measured in the austenite state at 336 K (figure 2). The four sharp distinct reflections observed in the austenite state show that Ni–Mn–Ga grows epitaxially on MgO and that the unit cell is rotated by 45° relative to the substrate, i.e. the epitaxial relationship is Ni–Mn–Ga (001) [110] || MgO(001)[100]. This orientation gives a reasonably good match between the diagonal of the substrate $d(110)_{MgO} = 0.5931$ nm and the lattice constant of cubic austenitic phase $a_A = 0.583$ nm with a mismatch of 1.9% (measured by a Θ –2 Θ scan at 336 K not shown here).

When cooling below the martensitic transformation (as will be discussed later in the article) to 300 K the film transforms into a (pseudo-)orthorhombic structure. The structural changes result in more reflections, which are observed in all three distinct $(220)_M$, $(202)_M$ and $(022)_M$ poles. In addition to these sharp reflections, a significant increase in background signal is observed, which is more than ten times higher compared to the pole figures of the austenite phase. (For better visibility the background is suppressed in figures 2 and 3.) For the $(220)_M$ reflections, the intensity of the quadrant marked in red is increased by a factor of three

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Figure 3. The $\{400\}_M$ pole figures of the film measured at 300 K in the martensitic state. Circles are shown at $\Delta \psi = 1^\circ$. Slight deviation of the pole figures from the centrosymmetrical position is due to imprecise positioning of the substrate.

to demonstrate that the background indeed contains some fine structure of minor reflections. All three pole figures demonstrate the 4-fold symmetry, which is expected from the (001) cubic substrate symmetry, although the peaks exhibit different splittings. In bulk orthorhombic martensite, all six possible variants are equivalent and should occur with the same frequency due to this symmetry. However, in the investigated epitaxial films the observed peak splitting is different for each of the $\{220\}_M$ poles and thus not all variants possible in bulk occur in the film. Obviously, the symmetrical distribution of variants in thin films is perturbed, which suggests that it is necessary to examine the influence of the rigid substrate in more detail.

Additional insight is obtained from the $\{400\}_M$ poles (figure 3). The $(400)_M$ and $(004)_M$ exhibit splitting in four distinct reflections, showing that these planes are tilted from the normal by 2.5° and 2.0°, respectively. The $(040)_M$ pole figure is dominated by one reflection close to normal, indicating that this plane remains nearly parallel to the parent austenite. This explains why the $(040)_M$ reflection contributes with the highest intensity in Bragg–Brentano geometry (figure 1). As the lattice spacings of the $(040)_M$ and $(004)_A$ planes (and of the $(020)_M$ and $(020)_A$ planes) are quite similar, the existence of some remaining austenite cannot be excluded by these pole figures.

The orientation of these coherent variants (resulting in sharp reflections) can be understood by the following simple geometrical model. It assumes that a thin austenitic layer at the film–substrate interface exists, which is likely, as a martensitic transformation is hindered by a rigid substrate. Following bulk experiments [21], it is considered that the habit planes, describing the austenite–martensite interfaces on top of this thin layer, is close to a $(101)_M$ plane, which cannot be parallel to the substrate plane. As an example, one of the six possible crystallographic orientations of this plane is sketched in figure 4. Two of the six orientations describe planes perpendicular to the substrate plane. Martensite variants with this interface orientation would require an additional coherent interface with the austenite layer along the rigid substrate, which is not allowed by crystallography. Therefore, these variants cannot exist close to the substrate. The other four orientations can form a microstructure on top of a {101} facetted interface, which remains on average coherent with the (001)-plane. As shown in the following, this model is able to describe the observed major reflections in the x-ray experiments.



Figure 4. Geometry at the austenite–martensite interface close to the substrate describing the connection between the cubic austenitc (A) and orthorombic martensitic (M) unit cells axis. Figure (a) shows a three-dimensional (3D) sketch of the (101) Habit plane and (b) the x-z plane used to calculate the tilt angle γ . Only one exemplarily (101) plane out of six possible crystallographic equivalent planes is shown; thus the coordinate system used here is not the sample coordinate system.

Using the measured lattice constants, the interface sketched in figure 4 has a misfit as low as 0.15% along the $[101]_M$ direction and 0.86% along the $[010]_M$ direction, suggesting that the $(101)_M$ interface is energetically favoured. From the geometry sketched in figure 4, one obtains for the tilt angle of *a*- and *c*-axes:

$$\gamma = 45^{\circ} - \arctan\left(\frac{a_{\rm M}}{c_{\rm M}}\right). \tag{1}$$

Using the measured lattice parameters, γ is calculated to be 2.7°, which is in fair agreement with the tilt measured from the $(400)_M$ and $(004)_M$ pole figures. The deviation between experiment and model as well as the difference between both pole figures very likely originate from the monoclinic distortion of the 7M martensite, which is neglected in this simplified model (this distortion would result in a deformation of the circle in figure 4 into an ellipse, which is beyond the resolution in these thin film XRD measurements).

This geometry of the martensite–austenite interface suggests that b_M remains parallel to a_A , which at first glance could easily explain the single dominant central reflection observed in the $(040)_M$ pole figure. However, these interfaces would be perpendicular to the substrate, a geometry not expected following the previous arguments. Thus it is more likely that the measured central reflection originates from remaining austenite. Present measurements do not allow to distinguish between both origins, but the significantly lower integrated intensity of the $(040)_M$ pole compared to the $(400)_M$ and $(004)_M$ poles suggests that the volume fractions of remaining austenite and of the variants with perpendicular b_M together are low. A consequence of the low fraction of perpendicular interfaces is the unequal distribution of the six possible martensite variants in the constrained film observed within the pole figure measurements.

Further information can be obtained when using the transformation matrix instead of the simplified geometrical model. Along the cubic $\langle 100 \rangle_A$ axes the six orthorhombic variants are stretched by $\alpha = a_M/a_A = 1.048$, $\beta = b_M/a_A = 0.995$ and $\gamma = a_M/a_A = 0.954$. As β is approximately equal to 1 there exist almost exact austenite–martensite interfaces for this system [22]. The orientation of these interfaces is of the type (0.959, 0, 1.006)_M almost parallel

to the $(101)_A$ plane. In agreement with the simplified geometrical model the tilt of the $(400)_M$ and $(004)_M$ planes from the surface normal is calculated to be 2.7°. Additionally, for exact austenite-martensite interfaces, this approach allows us to calculate the rotation angles for the $(220)_M$ and $(022)_M$ directions for the different primary variants, giving 3.9° and 3.7°, respectively. This is in good agreement with the values of $4.2(6)^\circ$ and $3.6(6)^\circ$ for the peak splittings extracted from the $(220)_M$ and $(022)_M$ pole figures (figure 2). This rotation also results in the peak splitting of the $(400)_M$ and $(040)_M$ poles along φ .

The variants with the four possible orientations on {101} facets must form a polytwin microstructure near the substrate that keeps the overall strain in the (001)-plane constant. However, neither these single variants nor the possible twin-pairs composed of these variants can form a compatible austenite-martensite interface plane with an overall {001}-orientation. One can understand this incompatibility by the observations that a martensite variant on a {101} facetted interface can have an exact interface only with one facet. However, the whole martensite film must cover the complete substrate. This frustration causes the appearance of a polytwinned microstructure in the whole film. It is expected that further twin variants or rotated twins appear perpendicularly through the film to resolve this frustration.

In the following, these are called secondary (and ternary etc) variants in order to distinguish them from the primary variants with exact orientation with respect to the austenite or substrate. Additional variants are also needed to fulfil twin boundary conditions between neighbouring variants and keep the volume constant. As the film extension is laterally constrained by the rigid substrate, also the film thickness is constant during a volume-conserving martensitic transformation. Thus, with increasing distance from the substrate, twin boundaries between the primary twin variants and the secondary and further generations of twins are expected. The orientation relation mediated by these twin boundaries is in analogy to figure 4, e.g. $\tau_{(101)} = 2(45^{\circ} - \arctan(a_{\rm M}/c_{\rm M}))$, which differs from the relation created by the exact austenite-martensite interfaces. Thus the orientations of the secondary twins differ from the simple orientation of the primary variants. A consequence of the secondary twinning is e.g. the peak splitting observed in the $(202)_{\rm M}$ pole figure. Besides the intense reflections at $\psi = 45^{\circ}$ resulting from the primary variants (and possibly some remaining austenite), two additional less intense reflections at $\Delta \psi = 5.6(7)^{\circ}$ are observed. These cannot originate from the exact austenite-martensite interface close to the (101) direction, as this plane remains invariant. The measured angular derivation of $5.6(7)^{\circ}$ is in good agreement with the one calculated for a twin boundary $\tau_{(101)} = 5.2^{\circ}$ using the measured lattice constants. With this, all intense reflections observed in the pole figure measurements can be attributed to primary and secondary variants and some remaining austenite.

A refined analysis of the microstructure must also consider whether the formation of a usual habit plane between the remaining austenite and a twinned martensite structure according to the crystallographic theory of martensites is advantageous. This would mean that the transformation of the film takes place according to the particular microstructure derivable from the theory of Wechsler, Lieberman and Read (see [22], chapter 7). Twinned plates of this type may form building blocks from which the full microstructure of the film can be built. The analysis shows that there is only one type of such a habit plane for the transformation from cubic to edge-sharing orthorhombic system with the experimental lattice parameters. The respective twins share their orthorhombic *c*-axis, so that the twin with transformation strains $U_1 = \text{diag}(\alpha, \beta, \gamma)$ and $U_5 = \text{diag}(\alpha, \beta, \gamma)$ forms a habit plane with interface normals $\mathbf{m} = (+/-0.700, +/-0.004, 0.714)$ with length fractions of the two variants $\lambda = 0.099$ and $(1 - \lambda) = 0.901$. All other combinations

of habit plane and twins follow from permutations of the Cartesian coordinates. This result shows that twinning of this type does not exactly fit on to the (001) interface to the substrate. Furthermore, the transformation into a twinned state of this type will always contain one twin variant with a strain β laying almost in the (001)-plane. The construction shows that the full microstructure necessitates to introduce more than two variants and the formation of inexact interfaces between twins, the substrate and/or twinned microstructure plates. Then, details of these structures are determined by microelastic compromises between the best fitting combinations and the magnitude of interface and twin energies. Therefore, stress release of the transformed film on the substrate must again introduce several generations of twins in thick films with multiple variants combined.

Due to the low symmetry of the monoclinic martensitic structure, there are a great number of possible polytwin structures that can occur in several generations perpendicular through the layer. This 3D twinned microstructure occurs in order to release the transformation stress of the half micrometre thick layer. The different variant generations are expected to have an increasing crystallographic derivation from the original austenite. Indeed some minor additional reflections are visible e.g. in the $(220)_{\rm M}$ and $(040)_{\rm M}$ pole figures. The ternary and higher generations of variants lose the simple orientation with respect to the substrate and thus do not contribute to the intense reflections of the 1st and 2nd variants' generations but produce minor reflections at various positions accumulating to the increased background signal (with some fine structure) as observed in the x-ray pole figure measurements. Thus, the microstructure of the polytwinned film is inhomogeneous—a phenomenon that can be understood as a type of branching.

X-ray measurements used here allow probing the complete volume of a thin film, but do not probe interfaces directly. Hence, these indirect measurements do not allow a complete sketch of the twinned microstructure, but compared to preparation of cross-section needed e.g. for TEM studies, they do not introduce additional interfaces, which may alter the existing twinned microstructure. A comparison with a complete theoretical solution is not possible yet, as especially the different interface energies are not known.

3.3. Morphology

The film microstructure was examined by polarized light [23], electron and AFM. The optical image (figure 5(a)) shows that there are macroscopic twin variants on a scale of a few micrometres. By application of a magnetic field (0.4 T), only a change of contrast but no change of the overall pattern was observed. The contrast is totally lost when heating above the martensitic temperature and it returns when cooled. Within these macroscopic twins there are wavy line structures with a line width of about 50-100 nm (figure 5(b)), indicating additional microscopic twinning inside the macroscopic twins. This feature was observed by the backscatter electron contrast in the SEM. The wavy lines are approximately parallel and perpendicular to the large twin boundary of the macroscopic twins. Whereas completely relaxed twins are expected to have parallel twin boundaries, the observed highly disturbed twin pattern indicates that the microstructure is not able to accommodate both the constraints through the twin boundary orientations and through the interface with the substrate. The wavy pattern shows that the twin boundaries in these films are not as perfectly oriented along crystallographic directions and are not as highly coherent as in single crystals. This results in a certain angular variation between neighbouring variants and thus variants close to the free surface do not contribute to sharp reflections but to increased background in the pole figure measurements.



Figure 5. (a) Polarized light microscope image showing different martensitic variants with a size of several ten micrometres. (b) Magnified SEM picture of these variants. Within the variants of the size of several micrometres, a small twinned microstructure on the scale of 50 nm is seen. At the variant boundary, a branching of the twin structure is observed. (c) AFM measurement of the surface morphology originating from the nanoscaled twin structure.

As the macroscopic twins observed at the surface are significantly larger than the film thickness of half a micrometre, it is likely that they do not originate from strain compensation but from different martensitic nuclei. Indeed, at a distance of some tens of nanometres from the macroscopic 90-degree boundaries, a change of the twin width, i.e. branching, is visible in the SEM micrograph. These boundaries between macroscopic polytwin structures are typically less mobile compared to highly symmetric twin boundaries. Therefore, twin boundary mobility is only expected for interfaces between the microscopic twins, which are much smaller than



Figure 6. Phase transformations of an epitaxial Ni–Mn–Ga film. Temperaturedependent polarization curves taken at 0.01 and 2 T external magnetic field. From the measurement with an applied external field of 0.01 T, the transition temperatures are extracted, as marked with the lines. In the 2 T field, the film is in the saturated state. The saturation polarization of the film increases about 13% from the austenite to the martensite.

the film thickness. Additional AFM (figure 5(c)) images clearly display the occurrence of a surface relief known from bulk single crystals [24]. The surface relief arises from the twins with different crystallographic orientation and indicates microscopic twinning on the scale of several tens of nanometres.

3.4. Transformation behaviour

The transformation behaviour was studied by measuring the temperature dependence of the magnetic polarization in applied magnetic fields of 0.01 and 2 T (figure 6). It is observed, that 2 T is sufficient to saturate the sample. The curves measured at 2 T exhibit a monotonic increase of the magnetic polarization with decreasing temperature down to 10 K. The sudden increase of polarization around 320 K indicates the occurrence of a martensitic transformation. The increase of the saturation polarization ΔJ_{sat} of the martensitic phase compared to the austenite phase is found to be about 13%, a value which is comparable to bulk samples [25]. In contrast to the measurement at saturation, the low field curve exhibits a decrease of the polarization when transforming into the martensitic phase. A low field of 0.01 T allows alignment of a significant portion of the polarization within the austenite state having low magnetocrystalline anisotropy, but the field is not sufficient to align the polarization in the multivariant, highly anisotropic martensitic phase. This explains the decrease of polarization at low fields, a behaviour also known from bulk single crystals [2, 26]. After transformation, no other abrupt changes in magnetic polarization were detected down to 10 K, indicating that no further intermartensitic transformation [26] occurs.

The transformation temperatures of the system were determined by linear extrapolation in the low field measurement (Curie temperature $T_c = 352$ K, martensite start $M_s = 319$ K and finish temperature $M_f = 314$ K, austenite start $A_s = 321$ K and finish temperature $A_f = 326$ K), see figure 6. The martensitic transformation exhibits a narrow temperature hysteresis of about 10 K. In comparison with the bulk material, the temperature interval of the transformation is slightly wider and the onset and finish of the transformation are rounded. The observed extended tail of the transformation is assumed to originate from the transformation of the fraction of the film for which the martensitic deformation is more difficult to achieve due to constraints from the substrate.

3.5. Magnetic polarization curves

The driving force for MIR is the different total energy of variants in a magnetic field. In magnetic saturation, this difference is given by the magnetocrystalline anisotropy energy [27, 28]. Large magnetocrystalline anisotropy is thus a key requirement for MIR, as in an external magnetic field variants having their easy polarization axis parallel to the field are energetically favoured compared to those having their hard axis in this direction. The magnetocrystalline anisotropy has been determined using the magnetic polarization curves of the austenitic and the martensitic state. The polarization curves of the sample measured in parallel ($H \parallel$ MgO[100] and $H \parallel$ MgO[110]) and normal to the film plane ($H \parallel$ MgO[001]) are shown in figure 7.

In the austenitic state the polarization curve measured in plane saturates at low fields and displays a small coercivity ($\mu_0 H_c = 0.0025 \text{ T}$). The low saturation field indicates a rather low magnetic anisotropy of the austenite phase as for bulk material [29]. The out-of-plane hysteresis exhibits a significant lower slope and saturates at an external magnetic field close to the value of the saturation polarization. This behaviour is caused by the shape anisotropy favouring in-plane magnetization, as the demagnetization factor is equal to 1 for an infinite isotropic film.

The shape of the magnetization loop changes drastically when the material transforms to the martensitic state. Due to the increased magnetocrystalline anisotropy, the coercivity increases as compared to austenite to $\mu_0 H_c = 0.005 \text{ T}$ and, more importantly, the curve exhibits an increased tilt with four distinguishable parts. From the bulk material it is known that the martensite exhibits a relatively strong magnetocrystalline anisotropy and the magnetization curve of an orthorhombic martensite can be analysed by assuming independent contributions from the three differently oriented twin variants with different magnetic anisotropies [27]. The short *c*-axis is the magnetic easy axis, the long *a*-axis is the magnetic hard axis, and the magnetic anisotropy of the *b*-axis is intermediate. All these contributions can be identified in the polarization loops in the martensitic state when the field is parallel to one edge of the unit cell ($H \parallel MgO[110]$). For the variants with the c-axis parallel to the magnetic field, irreversible magnetic domain switching takes place resulting in a steep change of magnetization in the hysteresis loop-part 1. For the variant fraction with the a- and b-axes parallel to the magnetic field, reversible rotation is expected which continuously aligns the polarization vector towards the increasing field—part 2 and 3. These are followed by saturation (part 4). As the anisotropy is different along a and b, two distinct slopes can be observed. From the sharp changes of these polarization curves, 'knees', marked in figure 7, one can directly determine the magnetocrystalline anisotropy for both axes, $K_a = 0.9 \times 10^5 \,\mathrm{J \, m^{-3}}$ and $K_b = 0.2 \times 10^5 \,\mathrm{J \, m^{-3}}$ for the a- and b-axes, respectively. These values for K_a and for K_b are about two and four times lower, respectively, than known anisotropies in bulk materials [27].

Additional to the small hysteresis around H = 0, a pronounced hysteretic behaviour is visible in the first quadrant of the polarization loop of the martensitic phase at higher fields



Figure 7. Hysteresis curves within the austenitic and martensitic states. (a) The polarization curves in the austenitic state at 340 K. The black curve was measured parallel to the MgO[100] direction in the film plane. The blue curve is measured perpendicular to the film plane. (b) Hysteresis curves for the martensite at room temperature. Two measurements in the film plane are shown for an applied field parallel to the MgO[110] direction (red, line) and parallel to the MgO[100] direction (black, dotted line). Additionally, the polarization curve measured perpendicular to the film plane is shown (blue, dots).

(figure 8). When increasing the field to about 0.08 T, a steep increase in polarization occurs. When the field is decreased from saturation the magnetization does not follow the previous path but a jump-like decrease of magnetization occurs at lower fields, resulting in an additional hysteresis in the magnetization loop. The same process occurs in negative fields. The observed jumps are fully reversible and repeatable even after applying high fields up to 8 T. The properties of the film do not change after a heating/cooling cycle through the martensitic transition. Measurements have also been performed at temperatures down to 10 K and similar curves were observed. For MIR, a broad temperature range had been reported [28], but not for the MIM effect, which occurs only in the vicinity of the martensitic transformation. Similar behaviour was found when applying a field perpendicular to the substrate. In this case, the jump occurs at a higher field and is less apparent due to the demagnetization effect.

The observed kind of hysteresis is characteristic for polarization loops observed during MIR in the bulk martensite [2, 30]. In this case, a step in polarization occurs at a field sufficient to move twin boundaries, which results in the reorientation of the martensite. When reducing the field under stress, the original variant state is restored once the internal friction for twin



Figure 8. The 1st quadrant of the polarization curve at 297 K in the martensitic state of the film is shown along both non-equivalent in-plane directions (MgO[110] (red, solid line) and MgO[100] (black)). Jumps in the polarization curve occur in the loops around 0.06 T and around 0.08 T, respectively. The relative change of polarization where the field is applied parallel to the MgO [110] direction is $\Delta J_{\parallel}/J_s = 0.26$, and where the field is parallel to the MgO[110] direction, the observed change is $\Delta J_{45^\circ}/J_s = 0.39$.

boundary motion can be overcome. It is suggested that in the films investigated here the restoring stress originates from film stress relative to the substrate and twin branching at the substrate interface. The fraction of reoriented variants can be estimated, as indicated in figure 8, to $\Delta J_{\parallel}/J_{\rm s} = 0.26 \,(H \parallel \text{MgO}[110])$ by extrapolation of the magnetization curve at high field values to zero. With the field $H \parallel \text{MgO}[100]$ a similar behaviour is observed occurring at slightly higher fields with an even higher jump ($\Delta J_{45^\circ}/J_{\rm s} = 0.39$).

In the following, the situation when the field is applied parallel to the edges of the original cubic austenitic lattice cells is considered first ($H \parallel MgO[110]$). In a tetragonal system with a polytwin structure composed of two variants, MIR increases the fraction of c-variants at the cost of *a*-variants in applied field direction, which must be accompanied by a change of length. Since the length of the film is fixed by the substrate, no reorientation is expected. Indeed a film with a different composition and with a tetragonal structure has also been examined. For this tetragonal film, no sign of field-driven reorientation processes was found, such as characteristic polarization jumps. In films with a (pseudo)orthorhombic structure, a polytwin structure can be composed of three variants with three different lattice parameters in a given direction. This gives an additional parameter of freedom for the rearrangement of variants when compared with the case of a tetragonal unit cell. As a magnetic field disfavours *a*-variants having the highest magnetocrystalline anisotropy, the fraction of *a*-variants having the longest lattice constant can be reduced when the fraction of *b*-variants with a medium lattice constant increases and the fraction of c-variants with the shortest lattice constant is reduced at the same time. Here, the overall length as well as the number of unit cells does not change. However, the overall energy in an applied magnetic field is reduced by this process, as the measured anisotropy energy along the *a*-axis is more than four times as high compared to the *b*-axis.

Below, an analytical model is presented, which predicts an upper limit of 0.5 for the increase of volume fraction of *b*-variants from the value 1/3 in a statistically random distribution

of variants. This limit is higher compared to the experimentally observed increase of $\Delta J_{\parallel}/J_{\rm s} = 0.26$, indicating that the reorientation is incomplete. The incomplete MIR transformation may be imposed by the constraint on the primary polytwin structure due to the coherency at the film–substrate interface.

The model describes the jump in polarization when applying a field along the Ni–Mn–Ga unit cell axis ($H \parallel MgO[110]$). The higher jump observed for $H \parallel MgO[100]$ occurs because reorientation along both equal in-plane directions can contribute. Although this doubles the number of reoriented variants, they only contribute by their projection on to field direction. This suggests an increase by a factor of $2 \cos 45^{\circ} \approx 1.4$. This is in reasonable agreement with the experiments though this simple model ignores any coupling between the variants when MIR occurs simultaneously in both in-plane directions. In this geometry, the 45° angle between easy axis and magnetic field gives an unfavourable lever for MIR. Thus a higher field is required for MIR, which is in agreement with the obtained experimental results.

The experimental observations of the polarization curves strongly point to the existence of MIR in the constrained (pseudo)orthorhombic films. However, other purely magnetic reasons for the peculiar shape of the magnetization curve cannot be excluded. Therefore independent measurements, which directly probe the proposed microstructural changes in an applied magnetic field, have been employed.

3.6. Microstructural changes in the magnetic field—pole figure measurements

In order to analyse the influence of the magnetic field on microstructure, the pole figure measurements with and without an external magnetic field of about 0.2 T parallel to MgO[110] have been performed. This field is larger than the field at which, according to the polarization curve, the proposed reorientation occurs. The field was applied in a diagonal direction as indicated in figure 2. In the first run the texture of the film was measured with the field. The second measurement was performed after removal of the magnet without moving the sample or changing the measurement set-up. Partial $\{220\}_M$ pole figures were taken at each quadrant at $\varphi = 45^\circ$, 135° , 225° and 315° for a region of $\Delta \varphi = \pm 10^\circ$. The tilt angle was varied in the interval $\psi = [35^\circ, 52^\circ]$. A step size of 0.5° was used for both angles. To quantify the influence of a magnetic field, the intensities of the pole figures were integrated over ψ for the (220) and (022) poles and over φ for the (202) pole. The results are presented in figure 9. The overall sum of the intensities from all poles with and without field differs only by 0.03%—confirming that indeed an identical ensemble of variants is probed.

In the quadrants perpendicular to the magnetic field no obvious change of intensity was observed, but along the direction of the applied field, significant differences are seen. Projections of the 1st and 3rd quadrants of the $(022)_M$ pole demonstrate the change in intensities and its distribution (shown in figure 9). The intensities of the reflections themselves do not change significantly, but the background in field direction increases significantly, whereas the background in the antiparallel direction decreases. In order to summarize the asymmetry observed for all $\{220\}_M$ reflections, the changes of the integrated intensities, compared to the measurement without field (100%), are shown as bar graphs in figure 9(b).

The observed changes in the intensity of the pole figure can be explained as follows. The orientation of the primary generation of variants near the substrate is directly imposed by the boundary conditions at the substrate interface, which results in sharp reflections in the pole figures. This rigid interface, however, hinders reorientation in this primary polytwin

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Figure 9. (a) Intensities of the $(022)_{\rm M}$ poles (integrated over ψ) in the direction of the applied magnetic field (quadrant 1, $\varphi = 45^{\circ} \pm 10$) and antiparallel to the field (quadrant 3, $225^{\circ} \pm 10^{\circ}$). Red squares represent measurements with applied field and black circles represent measurements without magnetic field. (b) Bar graphs of the change of the integrated intensities within each quadrant by a magnetic field of 0.2 T relative to the measurement without field (=100%). The intensities are displayed separately for all four quadrants and all {220}_M poles.

system; hence the intensity of the sharp reflections cannot change. As the magnetic field affects only the background intensities, apparently only secondary (or ternary etc) twins at a larger distance to the substrate can be reoriented. For an orthorhombic crystal, several equivalent planes exist, typically resulting in several reflections of one single variant observed in pole figure measurements. In contrast, for monoclinic crystals, only one dominant reflection per index occurs. Thus a monoclinic distortion is required to explain the observed asymmetry in the change of the background intensity by a magnetic field. On the other hand, a bulk sample generally has a hierarchical twinned microstructure at several length scales and, consequently, this complete ensemble averages out any asymmetry. Due to the finite dimension of the films, only few generations of twinning are expected. This means that MIR becomes observable by texture measurements.

Together with the magnetic measurements the changes of the integral intensities in the pole figure measurements prove that the MIR or MSM effect takes place in the constrained film.

3.7. Model for MIR in constrained orthorhombic films

In the following, a linear model is proposed that explains the basic mechanism for the MIR in constrained orthorhombic films. Since the film is constrained, reorientation must take place without changing the film dimension. To simplify the discussion, it is assumed that the field is applied in a direction x along the edge of the cubic lattice cell of the parent austenite structure. For an orthorhombic system, there are three different axes and thus variants can possess three different extensions along the direction of the magnetic field. The variants with the *a*-axis along the field are called *a*-variants. Their fraction n_a^x is given by the fraction of unit cells with their *a*-axis aligned parallel to the x-axis, and likewise the fraction of *b*- and *c*-variants are defined as n_b^x and n_c^x (equivalent for the y and z-directions).

As the processes of martensitic transformation and reorientation are diffusionless, the total number of unit cells and thus the summed fractions of all different variants in one direction must remain constant:

$$n_a^x + n_b^x + n_c^x = 1. (2)$$

By reorienting, the crystal keeps its identity, so the sum of fractions of all orthorhombic unit cells irrespective of their orientation is also constant:

$$n_b^x + n_b^y + n_b^z = 1. (3)$$

Moreover, since the film is constrained by the substrate, its overall extension does not change and is given by the lattice constant of the austenite a_A :

$$n_a^x a + n_b^x b + n_c^x c = a_A. ag{4}$$

No direction is favoured without a magnetic field, and by symmetry considerations it is assumed that equation (3) gives $n_b^x = n_b^y = n_b^z = 1/3$ and correspondingly for other variants. This equidistribution, however, cannot hold exactly as this simplification ignores differences between variant population, e.g. due to their anisotropic elastic properties and different twin boundary energies, which are not known for this system. When applying a magnetic field along the x-axis, the symmetry is broken and an alignment of the hard axes in this direction, i.e. *a*- and *b*-variants, is unfavourable. To decrease the overall energy, these variants should transform to variants with lower magnetic energy. In particular, this should reduce the fraction of the *a*-variants n_a^x having the highest anisotropy and the longest lattice parameter. In order to keep the overall extension constant, contraction by reorientation can be compensated by increasing the fraction of *b*-variants n_b^x with the medium lattice parameter and reducing the fraction of *c*-variants n_c^x with the shortest lattice parameter at the same time. This means that *b*-variants are growing at the cost of both *c*- and *a*-variants due to mechanical and magnetic driving forces, respectively. An upper limit of $n_{b,H}^x$ is reached, when the fraction of either $n_{c,H}^x = 0$ or $n_{a,H}^x = 0$. With this, equation (2) together with (4) gives

$$n_{b,H}^{x} = \frac{a_{A} - a}{b - a}$$
 (for $n_{c,H}^{x} = 0$) or $n_{b,H}^{x} = \frac{a_{A} - c}{b - c}$ (for $n_{a,H}^{x} = 0$). (5)

With the measured lattice parameters, one obtains that the maximum volume fraction of *b*-variants is reached when no more *c*-variants are available $(n_{c,H}^x = 0)$. This gives an estimate for the achievable increase of the fraction for the *b*-variant $\Delta n_{b,H}^x = n_{b,H}^x - n_{b,H=0}^x = 0.5$.

The model shows that it is possible to reorient a- and c-variants to b-variants. However, also the reverse process is possible, i.e. the change of b-variants into a- and c-variants. Which reorientation actually occurs depends on the energy balance of the system. The driving force for MIR is the minimization of energy in a magnetic field. Using the principle that the magnetic anisotropy energy of the system is reduced by MIR [30], the following condition for total energy of the system must be fulfilled:

$$n_{a,0}^{x}K_{a} + n_{b,0}^{x}K_{b} + n_{c,0}^{x}K_{c} > n_{a,H}^{x}K_{a} + n_{b,H}^{x}K_{b} + n_{c,H}^{x}K_{c}.$$
(6)

Assuming full reorientation, one derives:

$$\frac{1}{3}K_{a} + \frac{1}{3}K_{b} + \frac{1}{3} \cdot 0 > (1 - n_{b,H}^{x})K_{a} + n_{b,H}^{x}K_{b} + 0 \times 0,$$

$$\frac{1}{3}K_{a} + \frac{1}{3}K_{b} > \left(1 - \frac{a_{A} - a}{b - a}\right)K_{a} + \frac{a_{A} - a}{b - a}K_{b}.$$
(7)

With the anisotropy energies obtained from the experiment $(0.37 \times 10^5 \text{ J m}^{-3} > 0.31 \times 10^5 \text{ J m}^{-3})$, an increase of *b*-variants in a magnetic field is favoured.

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In the experiment a lower fraction of reoriented variants $\Delta J_{\parallel}/J_{\rm s} = 0.26$ compared to the upper limit of 0.5 is observed, indicating that the reorientation is partial and possibly hindered by the film–substrate interface. Additionally, the zero-field ground state of variant volume fractions will, in general, not obey a random statistical distribution as assumed in the simple model.

Despite the simplified assumptions used in the model, it explains well why reorientation in constrained films is possible and allows a quantitative estimation of the possible variant redistribution. The crucial point is embodied in equation (4), which states that the three possible arrangements of an orthorhombic unit cells give an additional free parameter (n_b^x) to fulfil the constraint of a rigid substrate, as compared to other arrangements, e.g. in a tetragonal martensite, with two variants only.

4. Summary

Epitaxial Ni–Mn–Ga films were prepared on heated MgO substrates by sputtering. The films are martensitic and ferromagnetic at room temperature. The substrate determines the key properties of these films. The film orientation is controlled by epitaxy within the austenite state. When transforming to the martensitic state, the boundary condition on a rigid substrate reduces the number of observed variant orientations compared to bulk and appears to slightly broaden the thermal hysteresis.

Independent magnetic polarization and pole figure measurements show that MIR or MSM effects take place in constrained thin epitaxial Ni–Mn–Ga films. The existence of the additional free parameter of an orthorhombic structure compared with a tetragonal structure allows a MIR of variants within a film even though its overall extension remains constant as imposed by the substrate. Experiments are the easiest to interpret in epitaxial films, but this effect is not restricted to epitaxial thin films. Indeed e.g. a similar hysteresis can be found in polycrystalline orthorhombic films [31].

MIR in epitaxial films is observed in significantly lower applied fields compared to bulk single crystals. This may be due to two key differences: first, thin film preparation differs significantly compared to bulk. Films are deposited atom by atom at medium temperatures; hence no high-temperature homogenization and ordering treatment is necessary. This may result in a significantly lower defect density, hindering twin boundary movement. The other difference is the high aspect ratio of thin films favouring an in-plane alignment of magnetization due to demagnetization effects. This gives a demagnetization factor close to 0, compared to 1/3 for a cubic bulk sample. For comparison, hence, the bulk measurements should be corrected for demagnetization, which reduces the switching field by about 1/3 of the spontaneous polarization. For the best single crystals, a corrected switching field would be in the same order of magnitude as in our films, but an accurate comparison would require a detailed knowledge of the sample size. The demonstration of the MIR effect in thin films opens up the possibility of integrating magneto-active thin films into microsystems.

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