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Hierarchical twin microstructure in modulated 10M Ni-Mn-Ga single crystals. An analysis including shuffling of atomic layers

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Abstract

The trained and self-accommodated martensite microstructures of 10M Ni-Mn-Ga single crystals were studied by back scattered electron imaging and electron backscatter diffraction in a scanning electron microscope. These data were then compared with theoretical calculations using a long periodic commensurate modulated monoclinic lattice and Gautam and Howe (G-H) model in order to establish twin boundary (TB) hierarchy according to their surface energy. The whole spectrum of TBs including type I, type II, and compound twins were detected and theoretically predicted. Using the G-H model and long periodic structure along with a small displacement of atoms connected with lattice modulation allowed to calculate all twin elements including their surface energy. These data are then confronted with theoretical predictions of classical continuum mechanics and minimum shear approaches, which disregard the lattice modulation. As it is shown including the modulation modifies the twin systems (twinning plane and twinning direction) for some TBs. Moreover, it strongly affects the interfacial energy which allows to rank TBs in 10M Ni-Mn-Ga system. Large differences in surface energy between different TBs are associated with atomic interface configurations and de-shuffling of atoms to form a coherent twin plane. Therefore, unlike the classical geometrical concepts, the G-H model allows to perform not only a quantitative but also qualitative analysis of all possible twin boundaries. As a result of new approach, a model that involves interfacial energy, shuffling of atoms and homogenous shear type deformation for TB determination is presented. In addition, irrational or step-like planes not only for type II but also for type I twin boundaries is predicted. Furthermore, a description of twin formation with respect to two different reference systems is completed, i.e. parent-based and monoclinic ones. In this way, a hierarchical twin microstructure of 10M martensite is established.

Keywords: Ni–Mn–Ga; EBSD; Twinning; Modulation

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Introduction

Ni-Mn-Ga alloy is one of the most studied ferromagnetic Heusler systems due to extremely low twinning stress allowing variant reorientation under very small magnetic fields [Morin et al., 2011; Straka et al., 2012; Wang et al., 2013; Haldar et al., 2014; Lester et al. 2015; Ranzieri et al., 2015; Musiienko et al., 2018; Zreihan et al., 2018, Zou et al. 2018]. Extremely low twinning stresses of about 0.5 MPa for type I and even down to 0.05 MPa for type II are an inherent part of the complex modulated crystal structure [Straka et al. 2012; Soroka et al., 2018]. These two types of twin boundaries (TBs) are involved in the so-called variant reorientation resulting in easy exchange of a and c -axes. Consequently, a large magnetic field-induced strain (MFIS) can be produced [Murray et al., 2000; Zaki et al., 2012; Heczko et al., 2013; Sozinov et al., 2013; Pagounis et al., 2014; Zhang et al., 2018a,b]. However, lowering the crystal symmetry by a periodic modulation along the $\langle 110 \rangle$ direction from the basic structures (tetragonal or orthorhombic) provides a monoclinic distortion. This symmetry reduction gives rise to form other boundaries such as compound a/b $\{110\}$, modulation (MB) $\{100\}$ or even non-conventional TBs where neither twinning plane nor shearing vector have rational character (for twin element description see Table 1), [Seiner et al. 2019]. The nomenclature used here is strictly related to a modulated monoclinic Ni-Mn-Ga crystal structure expressed with respect to the main axes of austenite. The so-called parent-based description (p) together with an unmodulated monoclinic cell are often introduced in order to avoid confusion with different nomenclature, coordinate systems and lattice parameters reported in the literature, which causes difficulties when comparing the results. Moreover, the parent-based approach allows to present the complex twin microstructure of modulated martensitic phases and the geometry of magneto-mechanical twinning in the most simple and understandable way.

All twin boundaries or interfaces between martensite variants can be predicted quite well using continuum mechanics and Hadamard compatibility conditions [James and Hane 2000; Bhattacharya, 2003]. However, to envision the twin microstructures, twin types and the austenite/twinned martensite interfaces in most cases the already mentioned parent-based coordinates are used. This coordinates refer directly to austenite cubic symmetry reducing the long periodic modulated symmetry. It is due to the fact that the simple shear deformation or Bain distortion which transforms austenite into individual martensitic variants, operates along the main orthogonal axes of austenite (for orthorhombic, monoclinic, triclinic crystal structures) or the martensite unit lattice is rotated 45 degrees about the $[001]$ direction with respect to austenite (tetragonal case). However, using the parent-based coordinates and an approximate

unit cell does not define the symmetry and atom positions of the entire crystal structure [Straka et al., 2012; Seiner et al., 2014; Zhou et al., 2017; Heczko et al., 2018]. Especially the small atomic displacement associated with lattice modulation is not included. The same applies to another method based upon the minimum shear criterion often used to determine twin elements even though the long periodic lattice parameters are used [Zhang et al. 2010, Li et al. 2011a]. Although this method may be used for calculations of the monoclinic long periodic 10M structure, the twin elements are computed only with respect to the long periodic lattice parameters excluding shuffling of atoms. Thus, the calculations based on the theory of martensite microstructure, continuum mechanics and minimum shear criteria use a monoclinic approximation which does not reflect perfectly the atomic positions of the 10M martensite. In other words it can be stated that these methods do not account for surface energy associated with the wavy or corrugated character of atoms across a twin plane in modulated martensites [Han et al. 2007, 2008a,b].

This is a very important point since in general, the energy related to a twin interface for TBs is assumed to be very low, therefore, it is disregarded in continuum models [Pitteri and Zanzotto 2003]. However, in the case of modulated structures, atoms at the twin boundary have to deviate from their equilibrium positions to form a flat interface or they create a step-like or wavy boundary. This increases the surface energy. This also affects the determination of twin elements (twinning plane and twinning direction) and it provides confusion with different twin nomenclature (type I, type II and compound).

Therefore, in this study the so-called G-H model [Gautam and Howe, 2011] and the lattice parameters of long periodic commensurate 10M crystal structure including shuffling of atoms are used to predict twin elements. The G-H model assumes that the most preferential orientation relationships of crystals are those where the total overlapping intensity of diffraction spots reaches a maximum value. Thus, the energy of the interface between the crystals attains one of the local minima forming a favorable, from the thermodynamic point of view, twin/grain boundary. Applying this model allows not only to provide a clear distinction between type I, type II and compound twin boundaries but also to involve the effect of shuffle of atoms. Apart from its geometric character it permits atom chemistry to be included. Additionally, this model allows to rank TBs according to their total energy and to scan the neighboring areas of particular twin axis variation.

As a result, new boundaries are uncovered and different symmetry operations for already existing ones assigned. Since the lattice parameters of the 10M NiMnGa may vary depending on the twin configuration, this allows to define the hierarchical twin microstructure produced not only upon martensitic transformation but also after different types of magneto-

mechanical training [Purohit et al. 2002; Chmielus et al., 2008; Müllner and Kostorz, 2008; Straka et al., 2011 Chulist et al., 2012a, 2012b, 2016, 2017; Mirzaeifar et al., 2013; Li et al., 2013; Witherspoon et al., 2013; Auricchio et al., 2014; Liu et al., 2014; Pascan, et al., 2015; Quyang et al., 2015; Xin et al., 2016; Yu et al., 2015, 2016; Bruno et al. 2018; Feuchtwanger et al., 2018; Timofeeva, et al., 2018; Dai et al., 2018].

Additionally, the relation between two parallel concepts of the reference systems, i.e. parent-based and monoclinic one is shown. The use of two approaches combine high precision for twin boundary identification and the ease with which the analysis of experimental data can be performed. In this way, a hierarchical twin microstructure of 10M martensite has been established.

Experimental procedure

An ingot produced at AdaptaMat Ltd. with the composition of $\text{Ni}_{50.2}\text{Mn}_{28.3}\text{Ga}_{21.5}$ (± 0.5 at.%) was first oriented in austenite by means of X-ray diffraction at a temperature above A_f and then samples were cut along the $\{100\}$ planes of the parent $L2_1$ cubic phase. Subsequently, the samples were mechanically polished using silicon carbide paper up to 7000 grit and then electropolished with 10 vol.% HClO_4 in ethanol at 0°C and at 40 V. Samples with a standard dimension of $0.9 \times 2.5 \times 20 \text{ mm}^3$ were used for microstructural investigations.

The crystal structure was determined by X-ray diffraction at room temperature to be 10M martensite with the following lattice parameters: $a_m = 4.223 \text{ \AA}$, $b_m = 5.584 \text{ \AA}$, $c_m = 21.015 \text{ \AA}$, $\beta_m = 90.27^\circ$. The given lattice parameters were measured for a bulk single crystal of self-accommodated state using synchrotron radiation. The lowercase character “*m*” denotes the long-periodic monoclinic structure. The lattice parameters recalculated for the parent-based coordinates and the average lattice [De Wolff, 1977; 1981; Janner and Janssen 1977; Janssen et al., 1992] are $a_p = 5.972 \text{ \AA}$, $b_p = 5.944 \text{ \AA}$, $c_p = 5.584$, $\gamma_p = 90.27^\circ$ (lattice parameter corresponds to each other). Subscript “*p*” denotes the parent-based monoclinic basic lattice parameters. For mathematic calculation both unit cells were used while to index the 10M structure in EBSD measurements only the long periodic one was applied. The microstructure and orientation inspections were performed using a FEI Quanta 3D SEM and the TSL system. In order to determine a very small difference between modulated variants and *a* and *b* lattice parameters a dedicated software based on the extremely small changes in Hough space was developed. Such an approach provides a much higher precision when indexing different twin variants [Chulist et al. 2019b]. For the sake of clarity type I and type II TBs are represented by twin plane and

twin axis in the discussion part, respectively (rational indices).

Fig. 1. Distribution of total overlapping intensity for 10M Ni-Mn-Ga crystal structure obtained for the constant 180° rotation angle showing all preferred twinning rotation axes. Type II TBs are represented by 180° rotation about η_1 whereas type I by rotation around the normal to K_1 by the same angle. In some regions, two distinct boundaries are located.

Results and Discussion

The application of the G-H method to the case of long-periodic modulated monoclinic crystal lattice with the given lattice parameters: $a_m = 4.223 \text{ \AA}$, $b_m = 5.584 \text{ \AA}$, $c_m = 21.015 \text{ \AA}$, $\beta_m = 90.27^\circ$ (atomic positions and modulation displacement based on the concept of Righi et al. 2007). Figure 1 shows the most preferential orientation relationship between the martensitic variants studied with and the modulated monoclinic crystal structure.

Fig. 2. Overlapping reflections seen in the reciprocal space for type I (a) and type II (b) TBs. Families of planes that are not disturbed by introducing the twin boundary are marked in red. In the case of type II TB, they belong to the zone axis $[5\ 5\ 1]_m$.

The analysed TBs are represented by twin axis η_1 or the normal to the twinning plane (K_1) for type II and type I, respectively. Thus, type II TBs are reproduced by 180° rotation about η_1 whereas type I by rotation around the normal to K_1 by the same angle. The model assumes that the interface energy decreases with increasing the total intensity coming from the overlapping reflections belonging to two different grains/domains/variants. Thus, the higher the intensity,

the greater the overlapping of potential fields of neighboring martensitic variants, and thus the more energy-favorable mutual orientation [Nalepka et al., 2016]. An example of such overlapping reflections can be seen in Fig. 2 for type I and type II *a/c* TBs along with long-periodic monoclinic indexing. The reflections that undergo complete covering are marked with red color, while those with partial overlapping with gray one. In the case of the type *a/c* II TB, much more reflections show a total overlap, not only presented $\pm(1 -2 5)_m$, $\pm(2 0 -10)_m$, $\pm(-1 -2 15)_m$, but also a number of others omitted due to the readability of the figure. A different situation arises in the case of type I *a/c* TB. For this case, reflections of only one plane family $(1 -2 5)_m$ undergo total overlapping. The rest of reflection spots coincidence to a lesser degree which translates into a lower intensity of type I *a/c* boundary. Thus, it turns out that except $(1 -2 5)_m$ there is no crystalline plane, which would maintain continuity crossing the boundary for type I *a/c* TB. It is worth noting that the axis η_1 of the rotational twin is one of the close packed directions of the initial austenitic structure. Hence, it is the zone axis of a large number of planes that preserve continuity crossing the boundary.

Fig. 3. 3D distribution of total overlapping intensity for twin boundaries: *a/c* type I and type II (a), MB (b) *a/b* (c) non-conventional (d).

The intensity distribution of different TBs, plotted in Fig. 1, corresponds very well with the rotation axes revealed experimentally and those calculated using the continuum model. All detected TBs including type I, type II a/c , b/c , modulation, a/b TBs are replicated in the stereographic projection, see Table 1 and Fig. 4. The same holds for two other components entail on the great circle that correspond to higher order twins often referred to $(20-1)_P$ plane [Ezaz et al., 2012]. However, these boundaries change the orientation of martensite variants with respect to austenite and they need to be activated under special stress-stain conditions. For that reason they are not the object of the given study.

Fig. 4. BSE images and EBSD map showing all possible twin boundaries detected in cube-oriented 10M Ni-Mn-Ga single crystals. The figure (c) shows high resolution TEM image of modulation TB which separates two regions with two different modulation directions.

A closer look at the enlarged sections 1 and 2 in Fig. 1 shows a perfect matching between the observed type II a/c TBs and the theoretically predicted (by Bain tensors and G-H model). The situation of the type I a/c TB is more complicated since many high intense areas can be found close to the type I a/c TBs. This can be clearly seen in Fig. 3a where a few maxima of high overlapping potentials are detected in the close vicinity to type I a/c twin axis. It seems that a small rotation of the mirror plane is sufficient to obtain a significant higher overlapping atomic potential fields of adjacent crystallites. This indicates that the type I TBs which are supposed to be rational plane can be comprised of more ledges forming a much more complex atomic structure. This is partially confirmed by experimental studies, which instead of plane $(1\ 2\ 5)_m$ identify another plane inclined by more than 1° [Li et al. 2011].

Fig. 5. The concept of a/b boundaries using inverting stacking fault [Niemann et al., 2017b] or $(001)_m$ mirror plane.

A very high level of consistent is also observed with respect to compound $(105)_m$ and $(-105)_m$ TBs commonly referred to as MBs where the theoretical predictions are indisputably verified by experimental measurements. In Fig. 3b two maxima are clearly visible corresponding to two components $(105)_m$ and $(-105)_m$ (in the parent-based CS $(0 -1 0)_P$ and $(-1 0 0)_P$) which are very often observed in high resolution TEM images, Figs. 4c and e (Ge et al. 2006; Pons et al. 2002; Czaja et al, 2016, 2017). As can be seen in Fig. 3b a very high degree of coincidence is achieved for a broad region suggesting that these boundaries form low-energy interfaces even if the twin plane deviates a few degrees from the perfect twin relation. In fact such a situation is frequently observed in many HRTEM and BSE/SEM images (Figs. 4c,e) when MBs run in a zigzag manner with a faceted shape deviating from straight lines.

When it comes to a/b TBs the G-H approach shows a significant difference between the two boundaries considered as equivalent according to the continuum based method. It turns out that $(0 0 1)[1 0 0]_m$ TBs (see Fig. 5a) are energetically more favorable than $(1 0 0)[0 0 1]_m$, Fig. 3c, Fig. 5b and Table 1. These boundaries can be constructed using two different concepts. The first one is a nanotwinning or periodic shuffling which considers the boundary as a plane which changes the staking fault from the $(3\bar{2})$ to $(\bar{2}3)$ sequence according to Zhdanov notation. In this concept they form the so-called inverting staking fault [Niemann et al., 2017b; Chulist et al., 2019b]. Concurrently, these boundaries can be formed using another concept shown in Fig. 5b where both variants are separated by the $(100)_m$ mirror plane. Comparing both figures, a higher coherency of the first boundary is clearly visible, Fig. 5. These findings are also confirmed by experimental measurements since rather an exchange of a_c and b_c lattice parameters is detected in the EBSD measurements indicating $(001)_m$ than the rotation of monoclinic c -axis, $(100)_m$.

The greatest disagreement between the use of the real modulated structure and unmodulated approximation is revealed for the so-called non-conventional twins. The classical continuum mechanics based model indicates two conjugate TBs with irrational indices. However, using the G-H approach two other boundaries with a relatively high intensities are detected. The first of type I was determined to be $(-1 0 15)_m$ plane and the other of type II has $[15 0 1]_m$ twin axis, Fig. 5. Both twinning planes lie relatively close to the non-conventional TBs predicted by classic theory. Nevertheless, they deviate about 4.4° from the non-conventional ones. For easy comparison the non-conventional TB is marked in Fig. 3d yielding a very low intensity calculated by the G-H model. The observed deviations fall in the range of experimental error when determining the twin planes and twin directions of 10M modulated phases. As presented in [Seiner et al. 2019] the maximum deviation of 4.2° between the experiment and theoretical calculations was detected for non-conventional twins in 10M Ni-

Mn-Ga single crystals. Analyzing the experimentally detected angles it appears that the boundaries computed by G-H model deviate less from the experimental observations than that coming from the classic approach. However, to verify the accuracy of both methods very precise experimental data are of great importance. It requires very accurate geometrical and crystallographic measurements to determine the experimental twin elements with a high precision. The same holds for type I a/c and b/c TBs since very complex energy landscapes close to these planes is calculated using the G-H model. In fact, each of analyzed cases should be treated separately in order to find out the orientation of all discussed TBs.

Fig. 6. Schematic representation of type I and type II twin boundaries along with MB and a/b boundaries with respect to the so-called parent-based (austenite) coordinates.

Interestingly, the obtained intensity or degree of coincidence of two reciprocal lattice is inversely proportional to the amount of shear for the given twinning system. Most favored, from the energetic point of view, are the so-called a/b twins. Lower intensity exhibit MBs followed by type II a/c and type I a/c . The lowest probability is obtained for the non-conventional twins. According to the G-H model, the formation of the so-called non-conventional twins is energetically unfavorable so they are unlikely to occur. This hierarchy shows a reverse tendency to that observed in *in-situ* experiments upon martensitic transformation (the non-conventional TBs are not considered here). At the austenite/twinned martensite interface mostly the a/c TBs are observed. The reverse tendency seems to be due to boundary conditions which are imposed at the austenite/twinned martensite interface (lattice mismatch) and governed by elastic energy which are reduced at most by a/c twins (the highest amount of shear). Therefore, upon martensitic transformation not only the surface energy but also elastic energy has to be considered.

The observed during in-situ heating/cooling experiment sequence is also consistent with EBSD observations for self-accommodated and trained NiMnGa single crystals. For instance, EBSD maps show martensite variants that possess a common c_p -axis (with respect to parent-based CS) but they differ in modulation direction $\langle 110 \rangle_p$. These variants are further internally subdivided by the so-called a/b boundaries $(100)_p$ which alter the modulation sequence ($3\bar{2}$ into $2\bar{3}$ in Zhdanov notation), [Chulist et al. 2019]. In other words, it can be stated that the achieved subdivision indicates the two smallest elements as variants with exactly the same c -axis, the same modulation direction but different modulation sequence. Nevertheless, an situation where the twin order is reversed (e.g. variants which exhibit the same modulation sequence but differ in modulation direction or c -axis) was never observed.

This aforementioned twin hierarchy for a modulated monoclinic structure is schematically shown in Fig. 6. This figure illustrates two scenarios using the parent-based CS for type I and type II a/c twins. The reorientation of variants separated by a/c twins (exchange of a and c -axes) produces the highest strain which can be obtained by twinning, see Table 1. For 10M Ni-Mn-Ga single crystals, it exceeds 6% and can be induced by magnetic or mechanical fields [Sozinov et al., 2013; Pagounis et al., 2014]. As the 10M martensites possesses a monoclinic structure it gives rise to form two different types of a/c TBs i.e. type I and type II. Both types differ in symmetry operation and can be easily distinguished by microstructural, crystallographic and mechanical features [Li et al. 2011; Niemann et al., 2017a, 2017b; Chulist et al. 2019a,b]. Type II TBs exhibit a characteristic misorientation of about 4°

from the type I $\{101\}_P$ plane which is then translated into a deviation of about 5.6° of the twin trace visualized on the $(100)_P$ projection. Such an operation makes the twin plane (K_1) irrational for type II TBs. This morphological distinction between type I and type II TBs can be clearly seen in Fig. 7. Surprisingly, the same 5.6° angle which is found in Fig. 7 spans the five blocks of the modulated structure over one crystal $(100)_P$ plane forming a characteristic inclination of 5.68° (Fig. 7, type II).

Fig. 7. Type I and type II twin boundaries for 10M Ni-Mn-Ga trained single crystal showing the characteristic 5.6° inclination related with five-layered modulation period shown on the $(100)_p$ plane. The irrational plane/direction (red lines) may be represented by two rational (101) and (100) components (black lines).

This theoretical angle was computed employing the measured lattice parameters. It should be stressed that the characteristic deviation can only be observed along the $[100]_p$ direction that is why no mirror symmetry typical for twin relation is revealed in Fig. 7. Thus, it seems that the deviation for type II TBs is strictly linked with modulation order. Type II TBs can be also presented as a step-like interface composed of two rational components i.e. $(101)_p$ and $(001)_p$ suggesting that the twin boundaries in Ni-Mn-Ga modulated structures may be faceted at the atomic level, as visualized in Fig. 7. The same holds for twinning direction for type I TBs where the irrational direction may also be constructed of two rational components $[10\bar{1}]_p$ and $[010]_p$, Fig. 7. In fact, the irrational indices do not represent closed packed planes or directions having much less physical importance when shearing twin variants. It is hence worthwhile to study the atomic structure of type II *a-c* TBs to reveal the real structure that at macro or microscale seems to follow irrational indexes. However, this is a difficult task due to their extremely high mobility under magnetic field in TEM. A very similar concept with faceted twin planes was proposed by Ezaz and Sehitoglu for type II TBs. in NiTi [Ezaz and Sehitoglu, 2011]. Employing the first principle calculations, they showed a energetically favored two-rational component boundary. Furthermore, it was shown that it implies shuffling of atoms in addition to invariant lattice shear which can be critical for twin mechanism in Ni-Mn-Ga as suggested by Ezaz and Sehitoglu [Ezaz and Sehitoglu 2011; Wang and Sehitoglu 2013]. The step like character of type II TBs for modulated phases is also supported by unpublished data where for other modulate structures, such as 4M and 14M, a similar pattern for type II TBs exists.

On the second level the so-called $\{100\}_p$ modulation boundaries are formed. These interfaces separate variants which possess the common *c*-axis, however, they differ in modulation direction, Figs. 4c, e. By comparing these boundaries to other observed in 10M Ni-Mn-Ga, it can be stated that these boundaries to a large extent do not follow a straight line being often composed of two components, $(100)_p$ and $(010)_p$ Figs. 4b, c, e or Ge et al. 2006). Due to different symmetry operation, i.e. reflection or rotation, they entail crossing or parallel configuration for type I and type II TBs, respectively [Chulist et al. 2013].

In the next step, the given microstructure can be subdivided by another element named a/b boundaries. These boundaries separate regions with common c -axes and modulation direction, however, they differ in orientation of a and b -axes, Fig. 6. Using the concept of nanotwinning or periodic shuffling these elements can be portrayed as change of staking fault from $(3\bar{2})$ to $(2\bar{3})$ sequence according to Zhdanov notation creating an inverting staking fault [Niemann et al., 2017b; Chulist et al., 2019a].

Using the so-called parent based CS, one more twin boundary can be expected in NiMnGa 10M martensite. A boundary, termed as a non-conventional twin, forms a configuration where across the interface both modulation direction and modulation sequence (a/b) are changing. This type of boundary has been experimentally found and reported recently. They provide an additional degree of freedom that makes the system even more adaptive [Seiner et al. 2019]. Such boundaries can be also seen in Fig. 4d. Using the so-called parent-based coordinates the boundary is attributed to non-conventional twins since neither the twinning plane nor shear direction possess rational indices (see Table 1).

It is seen that subsequent elements are added in the next steps creating strongly hierarchical twin microstructure of the 10M Ni-Mn-Ga martensite. Thus, the presented in Fig. 6 twin hierarchy is validated experimentally and can be only seen when using the so-called parent based CS. The parent-based coordinates and the related nomenclature allow to show the real hierarchy and the complex geometry in the most simply and accessible way, Fig. 6. However, as already showed with G-H model this approximation does not define perfectly the atomic position, symmetry and twin elements of 10M martensite. On the other hand only with the help of the so-called presented based CS the a/b or non-conventional twins were first theoretically predicted and then experimentally verified. Thus, as can be seen both approaches show strengths and weaknesses which should be used to advantage. Therefore, for convenience and easy comparison all twinning systems are computed both using the Bain tensors (orthogonal coordinates indicated by p), and G-H model (monoclinic coordinates denoted by m). The results are summarized in Table 1. Depending on the method used they may be recalculated using both CSs. The crystallographic relation between them is sketched in Fig. 8 while particular directions or planes can be transformed between both systems with the given equations where the unit plane normal is referred to $\hat{\mathbf{n}}$ whereas the unit vector to $\hat{\mathbf{t}}$:

$$\mathbf{n}_m = \mathbf{M} \mathbf{T} \mathbf{U}_1^{-1} \hat{\mathbf{n}}_p, \quad \hat{\mathbf{n}}_m = \mathbf{n}_m / |\mathbf{n}_m| \quad (1)$$

$$\mathbf{t}_m = \mathbf{M} \mathbf{T} \mathbf{U}_1 \hat{\mathbf{t}}_p, \quad \hat{\mathbf{t}}_m = \mathbf{t}_m / |\mathbf{t}_m| \quad (2)$$

where $\mathbf{U}_1$ is the right stretch tensor for the first martensite variant, $\mathbf{T}$ rotation matrix and $\mathbf{M}$ is the transformation matrix from austenite system (x_p, y_p, z_p) to monoclinic one (x_m, y_m, z_m). The first operator takes the following form:

$$\mathbf{U}_1 = a^{-1} \begin{pmatrix} a_p \cos[(\gamma - 90^\circ)/2 - \theta] & -b_p \sin[(\gamma - 90^\circ)/2 + \theta] & 0 \\ -a_p \sin[(\gamma - 90^\circ)/2 - \theta] & b_p \cos[(\gamma - 90^\circ)/2 + \theta] & 0 \\ 0 & 0 & c_p \end{pmatrix} \quad (3)$$

with austenite lattice constant $a = 5.825 \text{ \AA}$ and $\theta = \tan^{-1} \left(\frac{a_p - b_p}{a_p + b_p} \tan \left(\frac{\gamma - 90^\circ}{2} \right) \right)$, while the second one :

$$\mathbf{T} = \begin{pmatrix} \cos[(\gamma - 90^\circ)/2 - \theta] & -\sin[(\gamma - 90^\circ)/2 - \theta] & 0 \\ \sin[(\gamma - 90^\circ)/2 - \theta] & \cos[(\gamma - 90^\circ)/2 - \theta] & 0 \\ 0 & 0 & 1 \end{pmatrix} \quad (4)$$

The matrix $\mathbf{M}$ is obtained under the assumption that lattice constants of the long monoclinic and unmodulated monoclinic cell are related in the following way: $\mathbf{a}_m = (\mathbf{a}_p - \mathbf{b}_p)/2$, $\mathbf{b}_m = \mathbf{c}_p$, $\mathbf{c}_m = -5(\mathbf{a}_p + \mathbf{b}_p)/2$. If we use austenite system (x_p, y_p, z_p) (Fig. 4) the monoclinic parameters take the form:

$$\mathbf{a}_m = [a_p + b_p \sin(\gamma - 90^\circ) \quad -b_p \cos(\gamma - 90^\circ) \quad 0]/2 \quad (5a)$$

$$\mathbf{b}_m = [0 \quad 0 \quad 1]c_p \quad (5b)$$

$$\mathbf{c}_m = -2.5[a_p - b_p \sin(\gamma - 90^\circ) \quad b_p \cos(\gamma - 90^\circ) \quad 0] \quad (5c)$$

where:

$$\gamma = \cos^{-1} \left(\frac{a_p^2 + b_p^2 - (2a_m)^2}{2a_p b_p} \right)$$

It allows to express the unit vectors of Cartesian system of the monoclinic long cell in the orthogonal austenite system (see Fig. 4) as follows:

$$\hat{\mathbf{x}}_m = \mathbf{x}_m/|\mathbf{x}_m|, \quad \mathbf{x}_m = \mathbf{a}_m - |\mathbf{c}_m|^{-2}(\mathbf{a}_m \cdot \mathbf{c}_m)\mathbf{c}_m \quad (6a)$$

$$\hat{\mathbf{y}}_m = \mathbf{b}_m/|\mathbf{b}_m| \quad (6b)$$

$$\hat{\mathbf{z}}_m = \mathbf{c}_m/|\mathbf{c}_m| \quad (6c)$$

The above unit vectors constitute successive rows of transformation matrix $\mathbf{M}$.

Table 1. Twinning systems in 10M Ni-Mn-Ga martensite

<i>Variants</i>	K_I	η_I	K_I	η_I	<i>Description</i>	<i>Twinning</i>	<i>Intensity</i>	<i>Shear</i>
Model	parent - cubic		modulated monoclinic			<i>type</i>	$\times 10^6$	
1:2	(0 -1 1)	[-0.04978	(1 2 5)	[-5.35224	b/c	Type I	13.084	0.1252
		0.70623		5				
		0.70623]		-0.92955]				

	(0.05690 - 0.70596 - 0.70596)	[0 -1 1]	(-1.08061 2 -4.59698)	[5 5 1]	b/c	Type II	18.718	0.1252
1:3	(0 -1 -1)	[0.04978 -0.70623 0.70623]	(1 -2 5)	[5.35224 5 0.92955]	b/c	Type I	13.084	0.1252
	(-0.05690 0.70596 -0.70596)	[1.08061 [0 1 1]	2	[-5 5 -1]	b/c	Type II	18.718	0.1252
1:4	(-1 0 1)	[0.70634 -0.04651 0.70634]	(-1 2 5)	[5.3292 5 -0.93416]	a/c	Type I	12.052	0.1346
	(-0.70613 0.05267 -0.70613)	(1.07463 [-1 0 1]	2	[-5 5 1]	a/c	Type II	17.784	0.1346
	(-1 0 -1)	[-0.70634 0.04651 0.70634]	(-1 -2 5)	[5.3292 5 0.93416]	a/c	Type I	12.052	0.1346
1:5	(0.70613 -0.05267 -0.70613)	[1 0 1]	2	[5 5 -1]	a/c	Type II	17.784	0.1346
1:6	(0 -1 0)	[-1 0 0]	(1 0 5)	[-5 0 1]	modulation twin	compound	38.156	0.0094
	(-1 0 0)	[0 1 0]	(-1 0 5)	[-5 0 -1]	modulation twin	compound	38.166	0.0094
1:7	(1 -1 0)	[-1 -1 0]	(1 0 0)	[0 0 1]	a/b	compound	59.506	0.0094
	(-1 -1 0)	[-1 1 0]	(0 0 1)	[-1 0 0]	a/b	compound	82.614	0.0094
1:8	(-0.38268 0.92388 0)	[-0.92388 -0.38268 0]	(-14 0 -29)	[-29 0 14]		Non- conven- tional	2.866	0.0133
	(0.92388 0.38268 0)	[0.38268 -0.92388 0]	(0.08257 0 -0.99659)	[0.99659 0 0.08257]		Non- conven- tional	2.866	0.0133

Fig. 8. Unit cell description for two concepts of the coordinate system: so-called parent-based and monoclinic. The purple color represents the tetragonal approximation of Bain unit cell. The θ angle is defined in order to obtain symmetric distortion of a martensitic variant with respect to austenite.

Conclusions

In the given study a complex twin microstructure of 10M Ni-Mn-Ga single crystals was studied by two theoretical models using two different crystal structures. Both approaches show very good agreement with experimental observations. Especially a good agreement is achieved for type II $a/c; b/c$ boundaries and modulation twins. The so-called classic approach which uses the parent-based CS and the average monoclinic lattice without modulation predicts the twin elements with a good precision and allows to establish a twin hierarchy in 10M martensite. However, using the G-H model and long periodic structure along with a small displacement of

atoms connected with lattice modulation allowed to calculate all twin elements including surface energy. This is of particular importance since the twin interface energy which is typically neglected in continuum mechanics calculations for modulated phases contributes to the total energy and it varies depending on the boundary type. Large differences in surface energy between different TBs are associated with atomic interface configurations and de-shuffling of atoms to form a coherent twin plane. The shuffled atoms that lie in the twinning plane experience elastic distortion (de-shuffling) to preserve a coherent boundary. Hence, this model allows to perform not only quantitative but also qualitative analysis ranking the twin boundary according to their total energy. This study also shows that type II TBs can also be represented by two rational components, which may reduce the total interfacial strain energy. The same holds for irrational twinning direction of type I TBs which suggests that twinning shear may be not sufficient for variant reorientation and some shuffle mechanism proposed by Ezaz and Sehitoglu is required. The intrinsic position of the type I a/c TB is even more complicated since many high intense areas can be found close to the type I a/c TBs. Two kinds of a/b compound TBs are revealed by the G-H approach showing a significant difference between the $(0\ 0\ 1)[1\ 0\ 0]_m$ and $(1\ 0\ 0)[0\ 0\ 1]_m$ twin boundaries. The same applies to the so-called non-conventional twins, which in the parent-based coordinates possess both irrational twin plane and irrational twin direction. Using the long periodic structure and G-H model two new twin boundaries i.e. type I $(-1\ 0\ 15)$ and type II $[15\ 0\ 1]$ TB are predicted. Thus, the analysis beyond the continuum mechanics permits to uncover new boundaries and different symmetry operations for already existing ones. The presented approach opens the possibility of further research, which will also cover precise twin structure determination, modulation character and austenite/twinned martensite interphase in alloys of different chemical composition and modulation order.

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Figure captions

Fig. 1. Distribution of total overlapping intensity for 10M Ni-Mn-Ga crystal structure obtained for the constant 180° rotation angle showing all preferred twinning rotation axes. Type II TBs are represented by 180° rotation about η_1 whereas type I by rotation around the normal to K_1 by the same angle. In some regions, two distinct boundaries are located.

Fig. 2. Overlapping reflections seen in the reciprocal space for type I (a) and type II (b) TBs. Families of planes that are not disturbed by introducing the twin boundary are marked in red. In the case of type II TB, they belong to the zone axis $[5\ 5\ 1]_m$.

Fig. 3. 3D distribution of total overlapping intensity for twin boundaries: a/c type I and type II (a), MB (b) a/b (c) non-conventional (d).

Fig. 4. BSE images and EBSD map showing all possible twin boundaries detected in cube-oriented 10M Ni-Mn-Ga single crystals. The figure (c) shows high resolution TEM image of modulation TB which separates two regions with two different modulation directions.

Fig. 5. The concept of a/b boundaries using inverting stacking fault [Niemann et al., 2017b] or $(001)_m$ mirror plane.

Fig. 6. Schematic representation of type I and type II twin boundaries along with MB and a/b boundaries with respect to the so-called parent-based (austenite) coordinates.

Fig. 7. Type I and type II twin boundaries for 10M Ni-Mn-Ga trained single crystal showing the characteristic 5.6° inclination related with five-layered modulation period shown on the $(100)_p$ plane. The irrational plane/direction (red lines) may be represented by two rational (101) and (100) components (black lines).

Fig. 8. Unit cell description for two concepts of the coordinate system: so-called parent-based and monoclinic. The purple color represents the tetragonal approximation of Bain unit lattice. The θ angle is defined in order to obtain symmetric distortion of a martensitic variant with respect to austenite.