Final Draft
of the original manuscript:

Chulist, R.; Oertel, C.-G.; Skrotzki, W.; Lippmann, T.:
Direction of modulation during twin boundary motion
In: Scripta Materialia (2009) Elsevier

DOI: 10.1016/j.scriptamat.2009.11.004

Direction of modulation during twin boundary motion

R. Chulist [a,*], C.-G. Oertel [a], W. Skrotzki [a], T. Lippmann [b]

[a] *Institut für Strukturphysik, Technische Universität Dresden, D-01062 Dresden, Germany*
[b] *Institut für Werkstoffforschung, GKSS Forschungszentrum Geesthacht, D-21502 Geesthacht, Germany*

To study the change of modulation direction during deformation, two NiMnGa single crystals with five-layered modulated tetragonal structure (5M) and seven layered modulated orthorhombic structure (7M) were chosen. Synchrotron diffraction experiments show that the modulation takes place on the {110) plane along the <110] directions. After deformation by twinning the *c*-axis orientation changes and with this also the direction of modulation obeying the twin relation.

Keywords: Synchrotron radiation; Diffraction; Martensitic phase transformation; Twinning; Modulation

NiMnGa alloys close to the stoichiometric composition Ni_2MnGa belong to the quite new family of ferromagnetic shape memory alloys These alloys are characterized by the magnetic field induced strain (MFIS) [1,2]. MFIS in NiMnGa alloys is based on the comparably easy motion of twin boundaries under a magnetic field which is especially promising from the point of view of application as actuators and sensors. Depending on the chemical composition and heat treatment at least three martensitic structures can be distinguished in the NiMnGa system. However, the effect mentioned above only exists in two different modulated structures: five-layered modulated tetragonal (5M) and seven-layered modulated orthorhombic (7M) structure. The modulation consists of periodic shuffling along the <110] direction [3,4] and in diffraction patterns is observed as extra spots along this direction. The nature, stability and role the of modulation are described by Zayak et al. [5-7] The twinning stress of the third, non-modulated (NM), martensitic phase is much higher than those measured for the modulated structures [8]. Therefore, and because of the absence of an uniaxial magnetic anisotropy [9], MFIS does not exist in the NM structure. Thus, in order to understand the easy motion of twin boundaries in modulated NiMnGa structures, it is the aim of this work to study the behaviour of modulation during twin boundary motion.

To study the effect of twin boundary motion on modulation, two oriented NiMnGa single crystals with five-layered (5M) and seven-layered (7M) martensitic structure at room temperature were chosen. The single crystals prepared by the Bridgman method were supplied by Adaptamat company. After slight deformation, the orientation of the single crystals was determined by means of Laue patterns in undeformed and deformed areas representing two martensitic variants related by twin relationship. The change of the direction of modulation was measured by diffraction of high-energy synchrotron radiation (100 keV) using beam line HARWI-II at DESY in Hamburg, Germany. To do this, before and after deformation the single crystals were transmitted by X-rays at two specific places. Since the modulation is only visible when the beam direction is parallel to the *c*-axis the samples were X-rayed along directions which are parallel to the main tetragonal and orthorhombic axes (possible c-axis localization) to find optimal diffraction parameters. The modulation in the 5M and 7M structure is visible by the presence of four and six extra spots along the <110] direction between the main reflections, respectively.

All the planes and directions mentioned in this paper are given in cubic coordinate system witch is expressed with respect to cubic axes of the parent $L2_1$ phase. The orthorhombic and tetragonal unit cell dimensions are given in following order a>b>c and a>c, respectively.

Synchrotron diffraction in transmission of a sample volume of 3 mm × 3 mm × 0.5 mm under different directions shows that the structure modulation takes place along <110] directions. Modulation along the [110] and [$1\bar{1}0$] directions has already been shown by Ge et al. with high resolution transmission electron microscopy. The different modulation directions belong to different areas in the single crystal and are separated by a facetted interface [10]. During twin boundary motion the c-axis orientation changes and with this also the direction of modulation with respect to the initial single crystal reference system (Figs. 1 to 3). The modulation is only visible when the beam direction is parallel to the *c*-axis. If the beam direction is along the *a*-axis (or also b-axis in orthorhombic case) the characteristic diffraction features of modulation are absent. Figs. 1, 3 present a diagram with the directions of modulation in two variants. Initially the modulation is on the (110) plane in one variant while after deformation it is still on the (110) plane but in the variant generated by twinning. The modulation direction obeys the twinning relation. Additionally, in Fig. 2 the upper diffraction pattern shows additional spots due to twinning. The non-split spots belong to the common ($\bar{2}02$) plane. The results clearly demonstrate that the direction of modulation in the 5M and 7M structure is crystallographically fixed normal to the *c*-axis. Thus, during twin boundary motion in modulated structures different atom movements have to take place compared to non-modulated ones. This may be the reason for the much lower stresses observed for twin boundary motion in modulated structures.

Note that in the 7M structure there also occurs another type of twinning system on {110) planes. However, motion of these twins only switches the a- and b-axes while the c-axis remains unchanged. In other words the direction of modulation does not change.

In summary, it is concluded that:
1) Modulation takes place on {110) along <110].
2) During twin boundary motion the *c*-axis orientation changes and with this also the direction of modulation in the sample coordinate system.

The work has been carried out within the priority program SPP 1239 “Magnetic shape Memory: Change of microstructure and shape of solid materials by external magnetic fields” of the German Research Society (DFG) under contract Sk 21/22-1,2.

[1] P. Müllner, V.A. Chernenko, M. Wollgarten, G. Kostorz, Appl. Phys. Lett. 92 (2002) 6708
[2] P. Müllner, V.A. Chernenko, G. Kostorz, J. Magn. Magn. Mater. 267 (2003) 325
[3] J. Pons, V.A. Chernenko, S. Santamarta, E.Cesari, Acta Mater. 48 (2000) 3027
[4] V. V. Martynov, V.V. Kokorin, J. Phys. III 2 (1992) 565
[5] A.T. Zayak, P.Ental, Mater. Sci. Eng. 378 (2004) 419
[6] A.T. Zayak, P.Ental, Hafner, J. Phys. IV 112 (2003) 985
[7] A.T. Zayak, P.Ental, J. Enkovaara, A. Ayuela, R.M. Nieminen, J. Phys.:Condens. Matter 15 (2003) 159
[8] A. Sozinov, A. A. Likhachev, K. Ullakko, IEEE Trans. Magn. 36 (2000) 3266
[9] A. Sozinov, A. A. Likhachev, K. Ullakko, Proc. SPIE 4333 (2001) 189
[10] Y. Ge, H. Jiang, A. Sozinov, O. Söderberg, N. Lanska, J. Keränen, E. I. Kauppinen, V. K. Lindroos, S. P. Hannula, Mater. Sci. Eng. A 438 (2006) 961

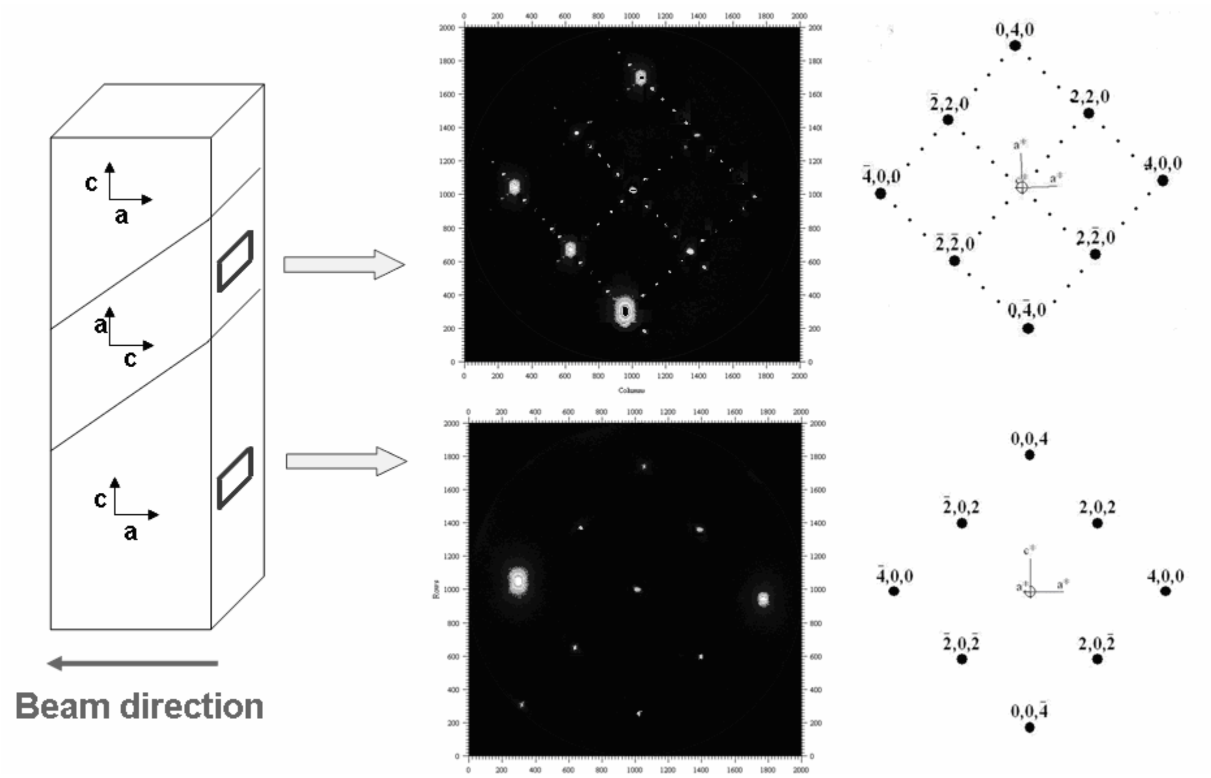

Fig. 1: X-ray diffraction of a 5M single crystal with twin band (*c*-axis differently oriented). The modulation is only visible when the beam is parallel to the *c*-axis.

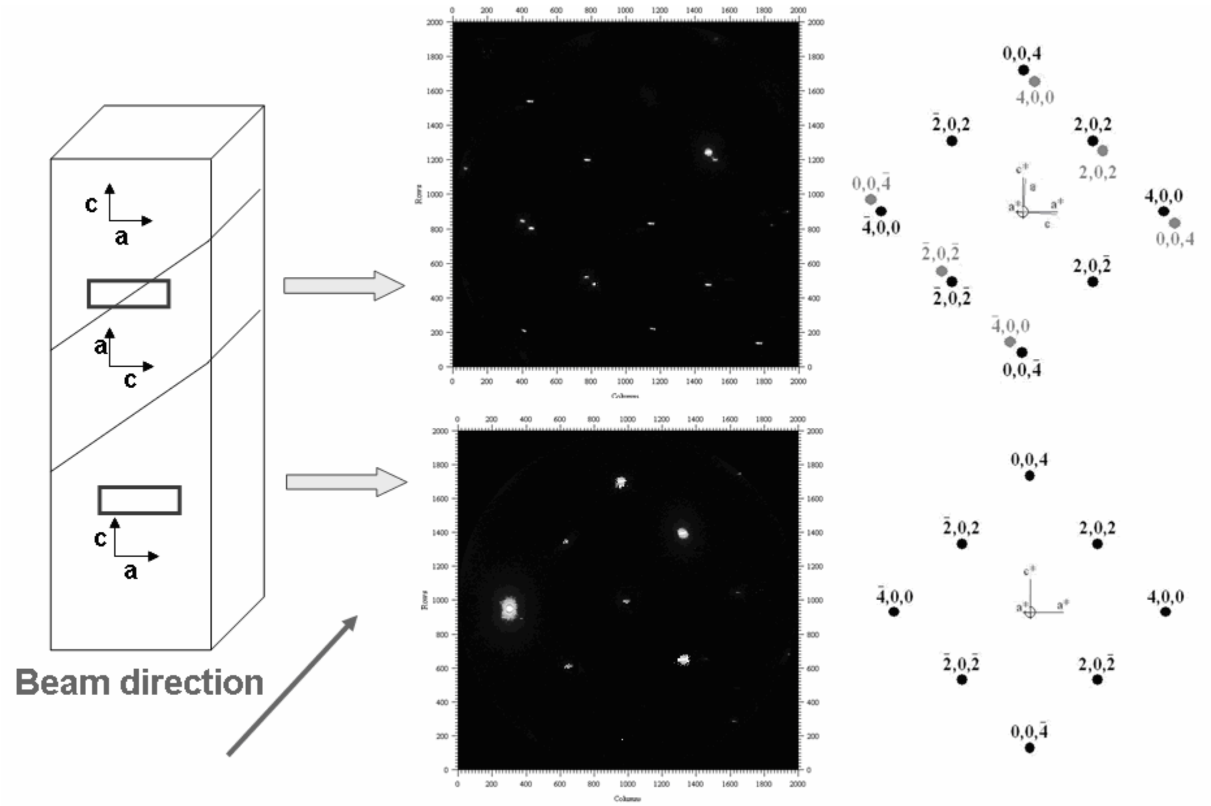

Fig. 2: X-ray diffraction of a 5M single crystal with twin band (*c*-axis differently oriented). Upper diffraction pattern shows a twin relation.

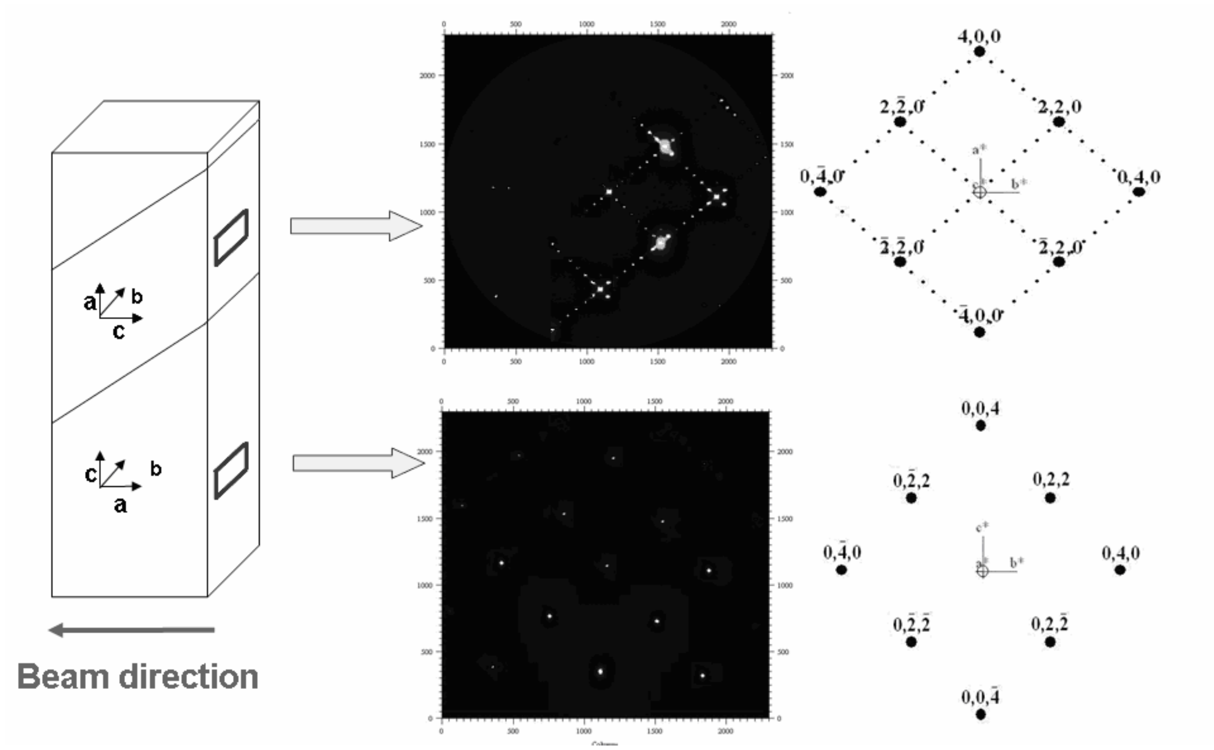

Fig. 3: X-ray diffraction of a 7M single crystal with twin band (*c*-axis differently oriented). The modulation is only visible when the beam is parallel to the *c*-axis.

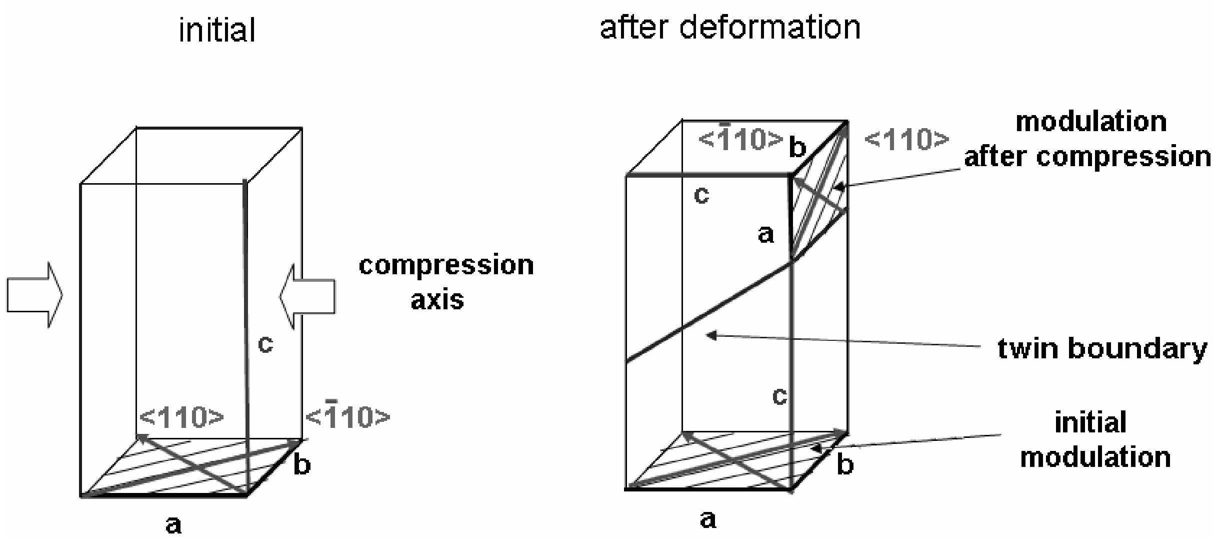

Fig. 4: Change of direction of modulation in a 7M structure during twin boundary motion induced by compression in a-direction.