Hysteretic structural changes within five-layered modulated 10M martensites of Ni-Mn-Ga(-Fe)

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Ni-Mn-Ga-based Heusler alloys are broadly studied for their magnetic shape memory (MSM) functionality originating from coupling between ferroelastic and ferromagnetic orders. The ferroelastic order is established after martensitic transformation. Formed ferroelastic domains with different orientation are separated by twin boundaries. In modulated phases, these boundaries are extremely mobile and can be manipulated by magnetic field. Thanks to these, the single crystals of five-layered modulated 10M martensite of Ni-Mn-Ga exhibits magnetically induced reorientation (MIR) of ferroelastic (twin) domains in a moderate field of the order of 0.1 T [1, 2]. This results in 6 % magnetic field induced strain (MFIS) down to liquid helium temperature [3]. Such unique behaviour makes the 10M martensite a perfect candidate for applications in actuators, sensors and energy harvesters.

The ferroelastic microstructure represents a challenge for proper determination of martensite phase structure. Due to the modulated nature together with complex hierarchical twinning (*compound* and *type I* and *II a*/*c* twins; and *non-conventional* twins) [4, 5], the structure of the 10M martensite has not yet been completely solved. There is even an ongoing discussion about the nature of the modulation where two main concepts are considered: *i)* general crystallographic wave modulation approach, and *ii)* nanotwinning. As the structural modulation seems to be the critical factor for the extremely high twin boundary mobility [5], the problem is pressing.

Using the X-ray and neutron diffraction, we investigated on the character and temperature evolution of 10M martensite phase. We found transition from *commensurate* to *incommensurate* 10M modulated structure in $Ni_{50}Mn_{27}Ga_{22}Fe_1$ single crystal [6]. The modulation vector gradually increases upon cooling from commensurate $\mathbf{q} = (2/5)\,\mathbf{g}_{110}$, where $\mathbf{g}_{110}$ is the reciprocal lattice vector, to incommensurate with $\mathbf{q}$ up to pseudo-commensurate $\mathbf{q} = (3/7)\,\mathbf{g}_{110}$. Further cooling results in transition to 14M with $\mathbf{q} = 2/7\,\mathbf{g}_{110}$. Upon heating, reverse changes of the *commensurate-incommensurate* transition are observed with a thermal hysteresis of ≈ 60 K. We detected the same hysteretic behaviour in the electrical resistivity and the effective elastic modulus. Scanning electron microscopy showed that the changes are accompanied by the refinement of the *a*/*b* laminate.

Furthermore, we observed continuous modulation changes within the 10M martensite of wide range of Ni-Mn-Ga(-Fe) compositions that undergo the Austenite → 10M → 14M martensite transition sequence. Based on these observations, we suggest that the commensurate state is a metastable form of 10M martensite. Upon cooling, this phase evolves through nanotwinning into a more irregular and more stable incommensurate structure, further supported by recent high-resolution TEM observation [7].

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Keywords: magnetic shape memory; phase transformations; Ni-Mn-Ga; twinned microstructure; structural modulation

This work was supported by Operational Programme Research, Development and Education financed by the European Structural and Investment Funds and the Czech Ministry of Education, Youth and Sports, project number SOLID21 CZ.02.1.01/0.0/0.0/16_019/0000760. P.V. thanks for the support by the Grant Agency of the Czech Technical University in Prague, grant number SGS19/190/OHK4/3T/14. We acknowledge the Institut Laue-Langevin and the project LTT20014 financed by the Ministry of Education, Youth and Sports, Czech Republic, for the provision of neutron radiation facilities.