Supplementary Information

Neutron Diffraction Study of the Martensitic Transformation of $Ni_{2.07}Mn_{0.93}Ga$ Heusler Alloy

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1. Crystal Data from Structural Refinement for Austenite and Modulated Martensite for $Ni_{2.07}Mn_{0.93}Ga$ Compound

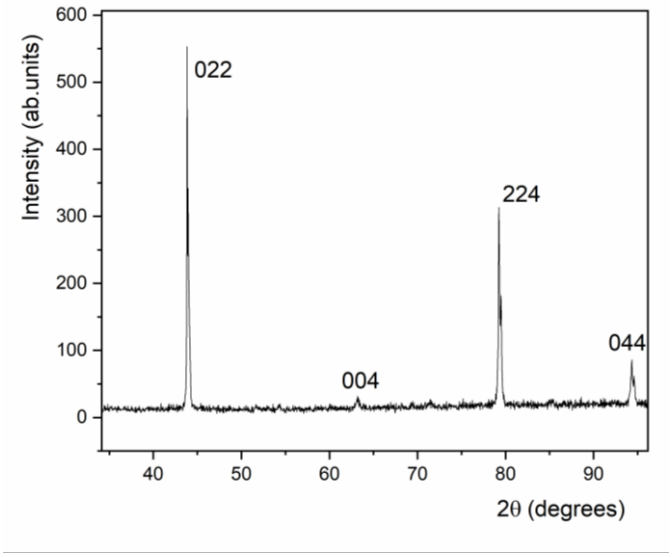

Figure S1. X-ray diffraction pattern of the polycrystalline alloy $Ni_{2.07}Mn_{0.93}Ga$ indicating the presence of cubic austenite as single phase.

Table S1. Crystal data of austenitic phase based on the structural refinement of the neutron diffraction data collected at 350 K (above *T*c)

Composition	**$Ni_{2.07}Mn_{0.93}Ga$**
Space Group	F m -3 m
System	Cubic
Z	4
a (Å)	5.8319(4)
$\alpha\ \beta\ \gamma$ (°)	90 90 90
Density (g/A^3)	0.90
Source	Neutrons (λ = 1.594Å)
R_p, R_{wp} (%)	5.56, 7.64

Table S2. Atomic coordinated, occupancy factors and thermal parameters for the cubic austenite at 350K.

	Wck position	**X**	**y**	**z**	**Occupancy factor**	**Uiso**
Ni1	8c	0.25	0.25	0.25	1.0	0.023(2)
Mn1	2a	0	0	0	0.93(5)	0.033(4)
Ni2	2a	0	0	0	0.07(5)	0.033(4)
Ga1	2b	0.5	0	0	1.0	0.015(2)

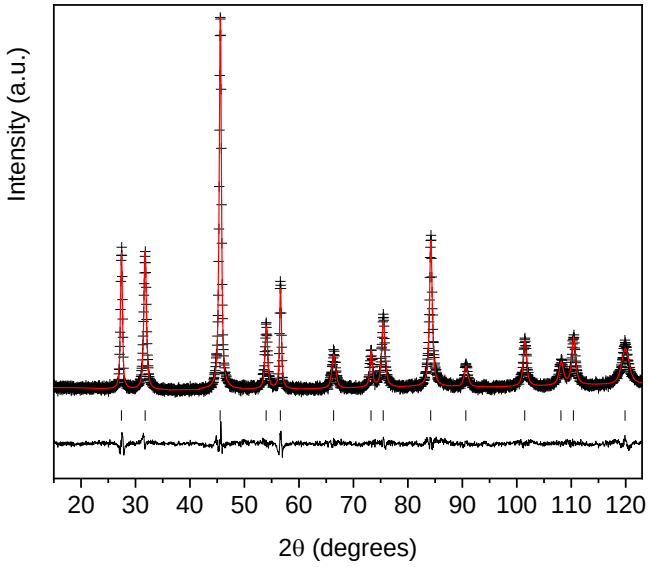

Figure S2. Rietveld plot based on neutron diffraction data for the austenitic phase at 350K.

2. Modulated Martensite at 200K

Table S3. Crystal data of the modulated martensitic phase based on the structural refinement of the neutron diffraction data collected at 220K.

Composition	**$Ni_{2.07}Mn_{0.93}Ga$**
Superpace Group	Immm(00γ)s00
System	Othorhombic
Z	2
a (Å)	4.2171(3)
b (Å)	5.5487(5)
c (Å)	4.1869(2)
$\alpha\ \beta\ \gamma$ (°)	90 90 90
V(Å^3)	
q	0.4226(3)$\boldsymbol{c^*}$
Density (g/A^3)	0.90
source	Neutrons (λ = 1.594Å)
R_p, R_{wp} (%)	6.45 9.23

Table S4. Atomic coordinate, occupancy factors, thermal parameters and modulation parameters A1x for the martensite at 220K.

	Wck position	**X**	**y**	**z**	**Occupancy factor**	**Uiso**
Ni1		0.50	0.25	0	1.0	0.0234
A_{1x}		0.059(3)	0	0		
Mn1		0	0	0	0.93(5)	0.0430
Ni2		0	0	0	0.07(5)	0.0430
A_{1x}		0.067(1)				
Ga1		0	0.5	0	1.0	0.0154
A_{1x}		0.068(5)				

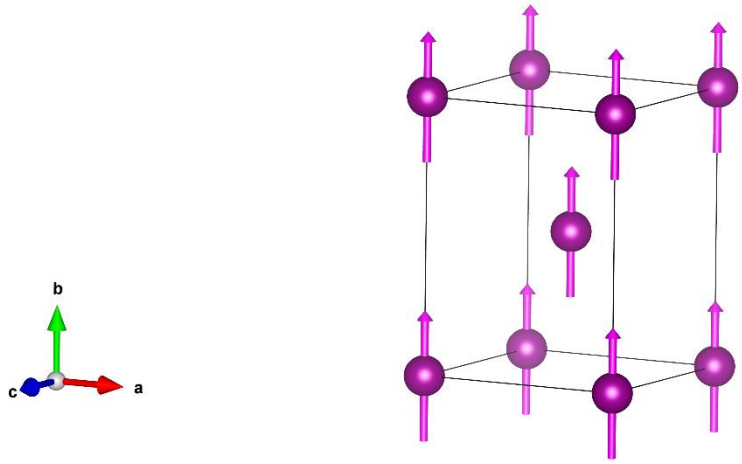

Figure S3. Ferromagnetic orientation of the spins located on the Mn/Ni site of the orthorhombic body centered lattice. The magnetic moment refined on the basis of the neutron diffraction pattern collected at 220K corresponds to 2.86 μ_B

3. Superspace Approach and Related Parameters

Each satellite of the diffraction pattern is associated to four indexes corresponding to the three basic vector $\boldsymbol{a}^*$, $\boldsymbol{b}^*$, $\boldsymbol{c}^*$ and to the modulation vector $\boldsymbol{q} = \boldsymbol{\alpha a}^* + \boldsymbol{\beta b}^* + \boldsymbol{\gamma c}^*$. If the value of on $\boldsymbol{\alpha}$, $\boldsymbol{\beta}$, and $\boldsymbol{\gamma}$ coefficients is a rational number then the modulation is defined commensurate, otherwise, an irrational number determines an incommensurate modulation. The commensurate modulation can always expressed in terms of a three-dimensional superstructure. In order to provide a crystallographic description of an incommensurate structure it is constructed a (3+1)-dimensional virtual space wherein a fourth dimension associated to the modulation vector is added to the classical 3D crystal model.
In the incommensurate modulated crystal the generic atomic position is described as [1]:

$$xi = Xi + ui\ (x\ 4), \tag{1}$$

$$yi = Yi + ui\ (x\ 4), \tag{2}$$

$$zi = Zi + ui\ (x\ 4), \tag{3}$$

where X, Y and Z are the fractional coordinate in the basic undistorted structure (see Fig1 b). Accordingly to incommensurate periodicity of the modulated structure, the function ui (x4) added to the generic position defines the periodic displacement from the basic position of a specific atom. The position of a single atom in the incommensurate structure can be defined by the t parameter. The relationship between $x4$ and t is:

$$t = x4 - \mathrm{q}\ x_i \tag{4}$$

The t parameter can be used, instead of $x4$, for describing the atomic position in the (3+1)-dimensional superspace. The t coordinate corresponds to the fractional coordinates in the conventional 3D symmetry. Generally, the graphical output of the modulation function is reported as a function of the t parameter.

4. The Co-Factor Extrapolation

Among the equivalent 9 possible combinations of crystallographic orientation of the interface between austenite and orthorhombic martensite [2] the following matrix is selected.

$$\begin{bmatrix} \frac{\alpha+\beta}{2} & \frac{\beta-\alpha}{2} & 0 \\ \frac{\beta-\alpha}{2} & \frac{\alpha+\beta}{2} & 0 \\ 0 & 0 & \gamma \end{bmatrix} \tag{5}$$

where $\alpha = \sqrt{2}a_m/a_0$, $\beta = \sqrt{2}b_m/a_0$ and $\gamma = c_m/a_0$ (a_m, b_m and c_m are the unit cell parameters for the martensite and a_0 stands for the unit cell parameter of austenite). The eigenvalue of the matrix so conceived represent the co-factor λ. Having structural data of austenite and martensite for three temperatures (240, 245 and 250K) it is possible to afford the three corresponding co-factors. The values are represented in the Figure 3 c.

References

[1] Van Smaalen S.; Incommensurate crystal structures *Crystallography Reviews* **1995**, *4*, 79–202
[2] James, R.D.; K. F. Hane K.F. Martensitic transformations and shape-memory materials. *Acta Mater*. **2000**, *48*, 197–222.