






# Supplementary material for "A ternary map of Ni-Mn-Ga Heusler alloys from *ab initio* calculations"

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## 1. Crystalline and magnetic structures

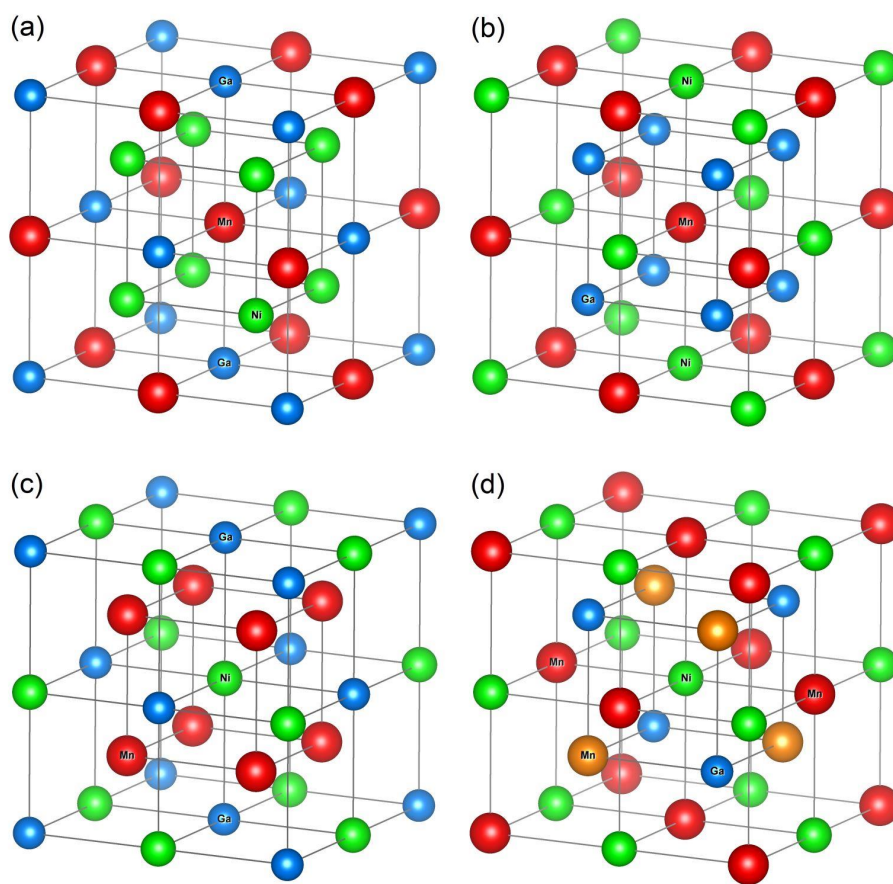
In the present work, the five cases of atomic arrangement into the cubic Cu<sub>2</sub>MnAl- and Hg<sub>2</sub>CuTi-type Heusler structures are considered in dependence on a nominal composition of Ni-Mn-Ga compounds. The Wyckoff positions for corresponding structures are presented in Table S1. In the case of Cu<sub>2</sub>MnAl-type structure (space group no. 225: *Fm* $\bar{3}$ *m*), the main difference between atomic arrangement considered is related to the occupation of 8*c* Wyckoff positions by an excess kind of atoms in a composition. For Ni-, Ga-, and Mn-rich compounds, the atomic arrangement is denoted as #225, #225', and #225'', correspondingly. In addition, the mixed occupation of 8*c* sites by Ni and Ga atoms is taken into consideration and labelled as #225<sup>mix</sup>. The Hg<sub>2</sub>CuTi-type structure (space group no. 216: *F* $\bar{4}$ 3*m*) is considered only for Mn-rich compounds since it is not preferable for Ni- and Ga-rich compounds.

**Table S1.** Positions of the Ni, Mn and Ga atoms considered in the regular (space group #225) and inverse (space group #216) Heusler structures.

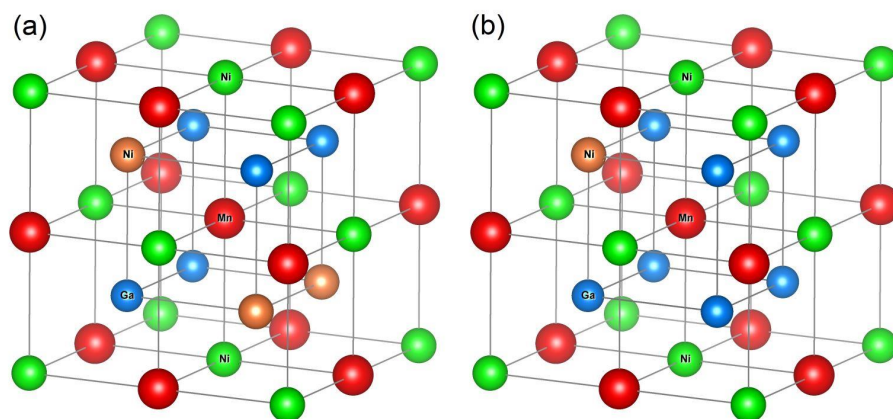
Wyckoff position	#225	#225'	#225''	#216	#225 <sup>mix</sup>
4 <i>a</i> (0, 0, 0)	Ga	Ni	Ga	Mn	Ni
4 <i>b</i> ( $\frac{1}{2}, \frac{1}{2}, \frac{1}{2}$ )	Mn	Mn	Ni	Ni	Mn
8 <i>c</i> * ( $\frac{1}{4}, \frac{1}{4}, \frac{1}{4}$ )	Ni	Ga	Mn	Mn	Ga or Ni
	Ni	Ga	Mn	Ga	Ga or Ni

\* For #216 crystal structure 8*c* Wyckoff position split into two positions: 4*c*(1/4, 1/4, 1/4) and 4*d*(3/4, 3/4, 3/4).

By way of example, Figures S1 and S2 illustrate the 16-atom supercells with mentioned above atomic configurations for stoichiometric Ni<sub>8</sub>Mn<sub>4</sub>Ga<sub>4</sub>, Ga<sub>8</sub>Mn<sub>4</sub>Ni<sub>4</sub>, Mn<sub>8</sub>Ni<sub>4</sub>Ga<sub>4</sub>, and off-stoichiometric Ni<sub>7</sub>Mn<sub>4</sub>Ga<sub>5</sub>, Ni<sub>5</sub>Mn<sub>4</sub>Ga<sub>7</sub>, respectively.



**Figure S1.** (Color online) 16-atom cubic supercells for stoichiometric Ni-Mn-Ga with  $L2_1$  crystal structure. (a)  $\text{Ni}_8\text{Mn}_4\text{Ga}_4$  ( $Fm\bar{3}m$ , #225); (b)  $\text{Ga}_8\text{Mn}_4\text{Ni}_4$  ( $Fm\bar{3}m$ , #225'); (c)  $\text{Mn}_8\text{Ni}_4\text{Ga}_4$  ( $Fm\bar{3}m$ , #225''); (d)  $\text{Mn}_8\text{Ni}_4\text{Ga}_4$  ( $F\bar{4}3m$ , #216). Here, Ni, Mn ( $\text{Mn}_1$  for #216 structure), Ga, and  $\text{Mn}_2$  (for #216 structure) atoms are denoted by green, red, light blue, and orange colors, respectively.

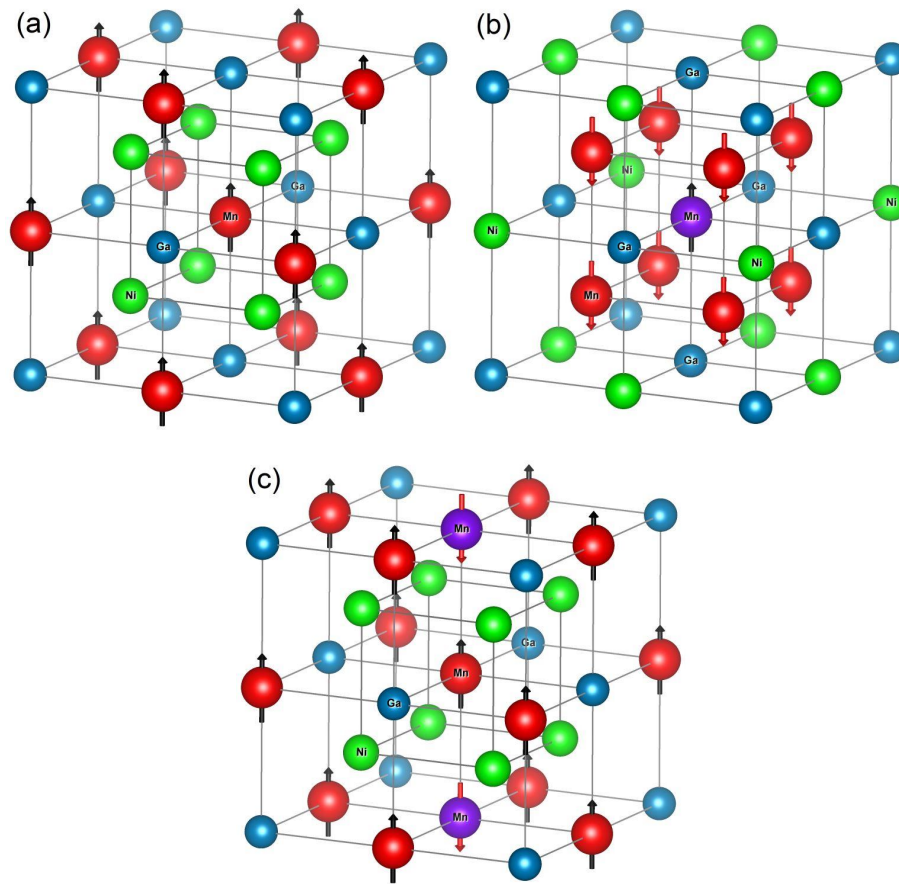


**Figure S2.** (Color online) 16-atom cubic supercells for off-stoichiometric Ni-Mn-Ga with #225<sup>mix</sup> structure: (a)  $\text{Ni}_7\text{Mn}_4\text{Ga}_5$  and (b)  $\text{Ni}_5\text{Mn}_4\text{Ga}_7$ . Here Ni, Mn, Ga, and Ni-excess atoms are denoted by green, red, light blue, and orange colors, respectively.

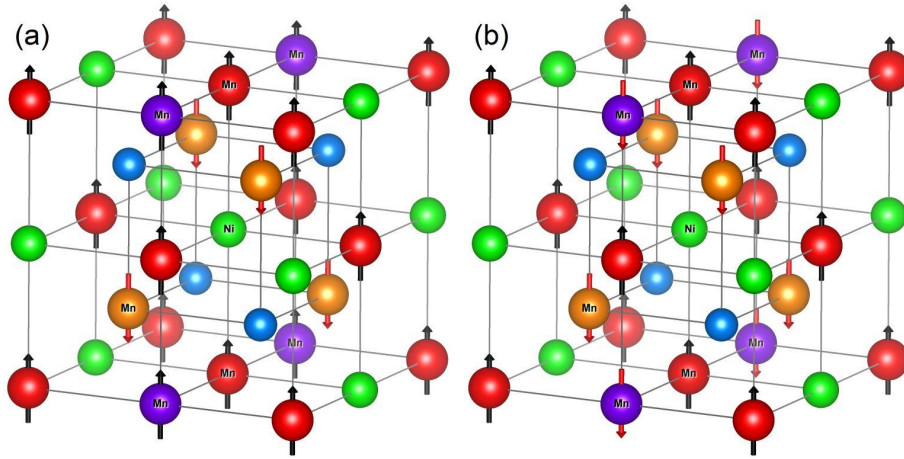
15 The ferro- and ferrimagnetic (FM and FIM) configurations specified by the Mn spin orientation  
 16 in the compounds with regular and inverse Heusler structures are listed in Table S2. The preferable  
 17 magnetic configurations are depicted in Figures S3 and S4

**Table S2.** The considered orientation of spin magnetic moment of Mn atoms located at different Wyckoff positions in the regular (#225) and inverse (#216) Heusler structures. Here the reversed spin orientation is marked in a red color to enhance visibility. Notice that the Ni spin magnetic moment is aligned along the direction  $\uparrow$ .

Symmetry group	Magnetic state	$(0,0,0)$	$\left(\frac{1}{4}, \frac{1}{4}, \frac{1}{4}\right)$	$\left(\frac{1}{2}, \frac{1}{2}, \frac{1}{2}\right)$	$\left(\frac{3}{4}, \frac{3}{4}, \frac{3}{4}\right)$
#225	FM	$\uparrow$	$\uparrow$	$\uparrow$	$\uparrow$
	FIM-1	$\uparrow$	$\downarrow$	$\uparrow$	$\downarrow$
	FIM-2	$\downarrow$	$\uparrow$	$\uparrow$	$\uparrow$
	FIM-1(2)	$\downarrow$	$\uparrow$	$\downarrow$	$\downarrow$
#216	FM	$\uparrow$	$\uparrow$	$\uparrow$	$\uparrow$
	FIM-3	$\downarrow$	$\uparrow$	$\uparrow$	$\uparrow$
	FIM-4	$\uparrow$	$\uparrow$	$\downarrow$	$\uparrow$
	FIM-5	$\uparrow$	$\uparrow$	$\uparrow$	$\downarrow$
	FIM-6	$\uparrow$	$\uparrow$	$\downarrow$	$\downarrow$
	FIM-7	$\downarrow$	$\uparrow$	$\downarrow$	$\uparrow$
	FIM-8	$\downarrow$	$\uparrow$	$\uparrow$	$\downarrow$
	FIM-9	$\downarrow$	$\uparrow$	$\downarrow$	$\downarrow$



**Figure S3.** (Color online) Structures in a cubic space group #225 for Ni-Mn-Ga with (a) FM, (b) FIM-1, and (c) FIM-2 spin configurations. Here, Ni, Mn, Ga, and Mn-excess atoms are denoted by green, red, light blue, and violet colors, respectively.



**Figure S4.** (Color online) Structures in a cubic space group #216 for Ni-Mn-Ga with (a) FIM-3, (b) FIM-7 spin configurations. Here Ni, Mn<sub>1</sub>, Mn<sub>2</sub>, Ga, and Mn-excess atoms are denoted by green, red, orange, light blue, and violet colors, respectively.

## 2. Convex hull diagram

Thermodynamic stability of compounds under study is assessed using an energy convex hull implementation in the formation energy-composition space. Figures S5(a) and (b) display the three-dimensional convex hull construction into the formation energy-composition space of Ni-Mn-Ga in the austenitic phase with a cubic structure and martensitic phase with tetragonal and orthorhombic structures. The most of investigated compositions have a negative formation energy but lie above or closely above the convex hull. This finding indicates that compositions are in thermodynamically metastable austenitic or martensitic phase and they can decompose into a mixture of stable binary phases and pure elements. The decomposition energy can be estimated from the cross section of hull energy convex as a difference between the formation energy of a Ni<sub>x</sub>Mn<sub>y</sub>Ga<sub>z</sub> compound and the formation energies of thermodynamically stable compounds, which lie on the convex hull. The cross-sections of the convex hull for Ni<sub>x</sub>Mn<sub>y</sub>Ga<sub>z</sub> ( $y = 25, 50, \text{ and } 75 \text{ at.}\%$ ) in the austenite and martensite phases are presented in Figures S5(c) and (d). It is seen that potentially stable or nearly stable compositions are located closely above the convex hull.

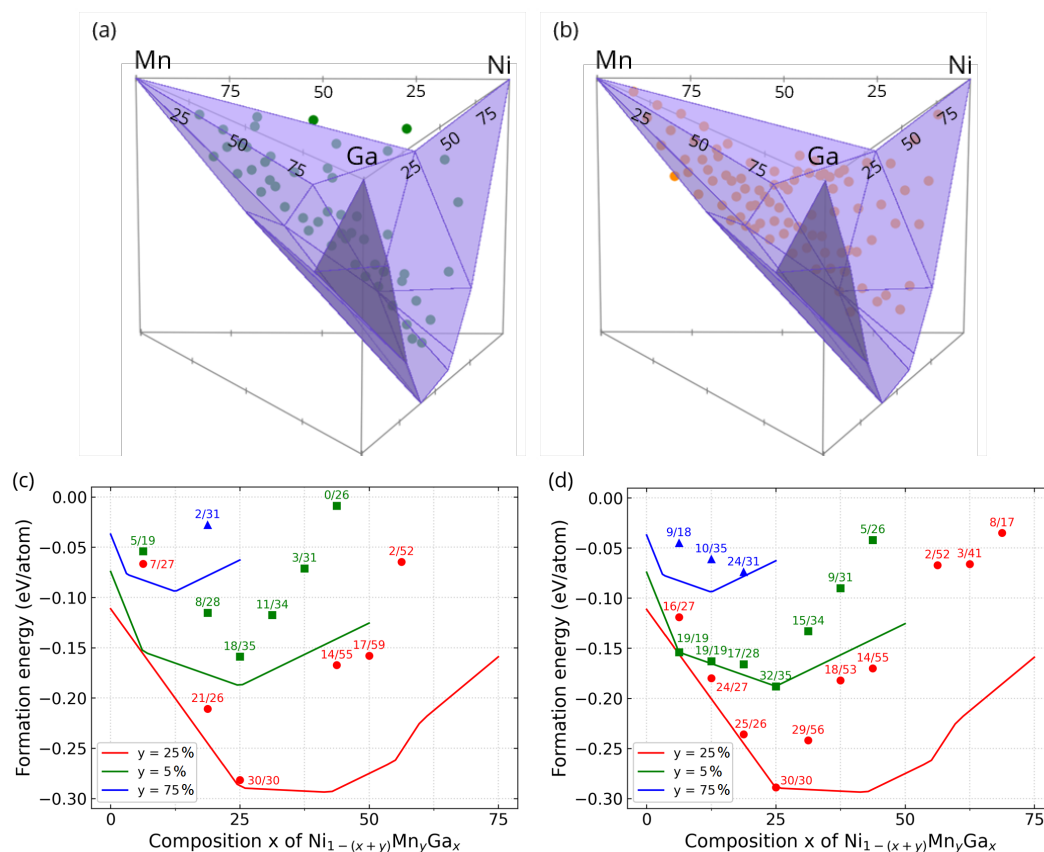
The pivot points (stable compounds) for the convex hull construction are listed in Table S3. As an example, Table S4 contains the possible decomposition reactions for Ni<sub>9</sub>Mn<sub>4</sub>Ga<sub>3</sub>.

**Table S3.** Space group, lattice parameter  $a$  (in Å),  $c/a$  ratio, and total energy  $E_{tot}$  (in eV/atom) per atom for the pivot points of the ternary Ni-Mn-Ga convex hull.

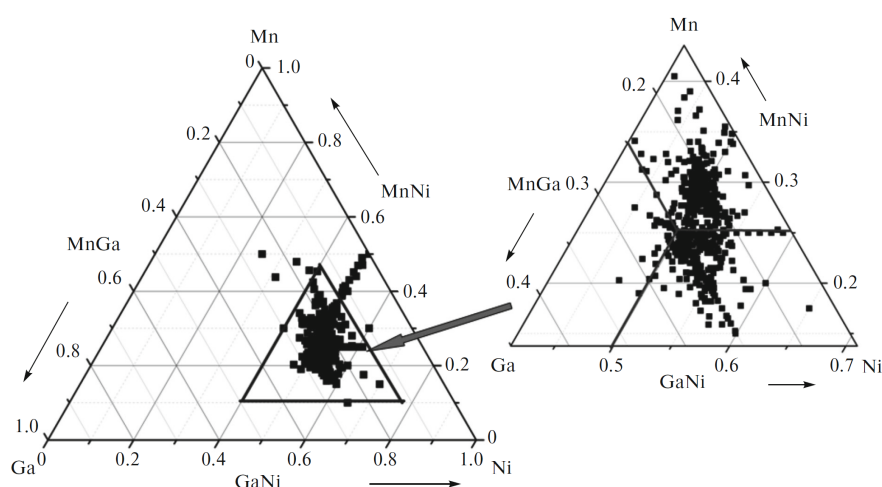
	Space group	$a$	$c/a$	$b/a$	$E_{tot}$
Ga	Cmce	4.613	0.996	1.654	−2.9095
Ga <sub>3</sub> Ni <sub>2</sub>	P3̄m1	4.088	1.193	1.000	−4.4588
Ga <sub>3</sub> Ni <sub>5</sub>	Cmmm	6.800	0.549	1.102	−5.0696
Ga <sub>5</sub> Ni	I4/mcm	6.406	1.506	1.000	−3.5391
Ga <sub>7</sub> Ni <sub>3</sub>	Im3̄m	8.457	1.000	1.000	−4.0925
Ga <sub>9</sub> Ni <sub>13</sub>	C2/m	9.969	0.852	0.791	−4.9915
GaNi <sub>3</sub>	Pm3̄m	3.574	1.000	1.000	−5.3482
Mn	I4̄3m	7.4262	1.000	1.000	−9.1617
MnGa	P4/mmm	2.704	1.358	1.000	−6.1611
MnGa <sub>4</sub>	Im3̄m	5.589	1.000	1.000	−4.3257
MnNi <sub>3</sub>	Pm3̄m	3.558	1.000	1.000	−6.7393
Ni	Fm3̄m	3.507	1.000	1.000	−5.7834

**Table S4.** Possible decomposition reactions and the decomposition energy  $E_{dec}$  (in eV/atom) between the investigated alloy and reaction products for  $\text{Ni}_9\text{Mn}_4\text{Ga}_3$ . The positive value indicates the phase instability against the decomposition process and *vice versa*.

#	Reaction	$E_{dec}$
1	$29 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 4\text{Mn}_{29} + 87\text{Ga}_1\text{Ni}_3$	0.007
2	$2 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 2\text{Mn}_1\text{Ni}_3 + 3\text{Ni}_4\text{Mn}_2\text{Ga}_2$	0.142
3	$232 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 522\text{Ni}_4 + 32\text{Mn}_{29} + 87\text{Ga}_8$	-0.843
4	$116 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 203\text{Ni}_4 + 16\text{Mn}_{29} + 116\text{Ga}_3\text{Ni}_1$	-0.344
5	$58 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 58\text{Ni}_4 + 8\text{Mn}_{29} + 29\text{Ga}_6\text{Ni}_{10}$	-0.116
6	$580 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 1218\text{Ni}_4 + 80\text{Mn}_{29} + 87\text{Ga}_{20}\text{Ni}_4$	-0.708
7	$812 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 1566\text{Ni}_4 + 112\text{Mn}_{29} + 87\text{Ga}_{28}\text{Ni}_{12}$	-0.500
8	$174 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 203\text{Ni}_4 + 24\text{Mn}_{29} + 29\text{Ga}_{18}\text{Ni}_{26}$	-0.140
9	$116 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 261\text{Ni}_4 + 4\text{Mn}_{29} + 348\text{Mn}_1\text{Ga}_1$	-0.655
10	$232 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 522\text{Ni}_4 + 26\text{Mn}_{29} + 87\text{Mn}_2\text{Ga}_8$	-0.688
11	$116 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 87\text{Ni}_4 + 4\text{Mn}_{29} + 174\text{Ni}_4\text{Mn}_2\text{Ga}_2$	0.030
12	$2 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 3\text{Ni}_4 + 6\text{Mn}_1\text{Ga}_1 + 2\text{Mn}_1\text{Ni}_3$	-0.544
13	$232 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 8\text{Mn}_{29} + 87\text{Ga}_8 + 696\text{Mn}_1\text{Ni}_3$	-0.509
14	$87 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 5\text{Mn}_{29} + 87\text{Ga}_3\text{Ni}_1 + 203\text{Mn}_1\text{Ni}_3$	-0.084
15	$174 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 16\text{Mn}_{29} + 87\text{Ga}_6\text{Ni}_{10} + 232\text{Mn}_1\text{Ni}_3$	0.033
16	$580 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 24\text{Mn}_{29} + 87\text{Ga}_{20}\text{Ni}_4 + 1624\text{Mn}_1\text{Ni}_3$	-0.396
17	$812 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 40\text{Mn}_{29} + 87\text{Ga}_{28}\text{Ni}_{12} + 2088\text{Mn}_1\text{Ni}_3$	-0.213
18	$522 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 44\text{Mn}_{29} + 87\text{Ga}_{18}\text{Ni}_{26} + 812\text{Mn}_1\text{Ni}_3$	0.033
19	$232 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 2\text{Mn}_{29} + 87\text{Mn}_2\text{Ga}_8 + 696\text{Mn}_1\text{Ni}_3$	-0.354
20	$4 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 1\text{Ga}_8 + 4\text{Mn}_1\text{Ga}_1 + 12\text{Mn}_1\text{Ni}_3$	-0.447
21	$11 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 6\text{Ga}_3\text{Ni}_1 + 15\text{Mn}_1\text{Ga}_1 + 29\text{Mn}_1\text{Ni}_3$	-0.192
22	$14 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 3\text{Ga}_6\text{Ni}_{10} + 24\text{Mn}_1\text{Ga}_1 + 32\text{Mn}_1\text{Ni}_3$	-0.169
23	$32 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 3\text{Ga}_{20}\text{Ni}_4 + 36\text{Mn}_1\text{Ga}_1 + 92\text{Mn}_1\text{Ni}_3$	-0.368
24	$16 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 1\text{Ga}_{28}\text{Ni}_{12} + 20\text{Mn}_1\text{Ga}_1 + 44\text{Mn}_1\text{Ni}_3$	-0.258
25	$40 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 3\text{Ga}_{18}\text{Ni}_{26} + 66\text{Mn}_1\text{Ga}_1 + 94\text{Mn}_1\text{Ni}_3$	-0.162
26	$1 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 1\text{Ga}_1\text{Ni}_3 + 2\text{Mn}_1\text{Ga}_1 + 2\text{Mn}_1\text{Ni}_3$	-0.212
27	$3 \text{ Ni}_9\text{Mn}_4\text{Ga}_3 \rightarrow 1\text{Mn}_1\text{Ga}_1 + 1\text{Mn}_2\text{Ga}_8 + 9\text{Mn}_1\text{Ni}_3$	-0.350



**Figure S5.** (Color online) The three-dimensional convex hull in the formation energy of Ni-Mn-Ga compositions with the (a) austenite and (b) martensite phases. (c), (d) The cross-sections of convex hull plotted at the fixed Mn content ( $y = 25, 50$ , and  $75$  at.%). Here lines denote the profiles of the convex hull. The degree of stable reactions among the possible ones for a particular compound against the decomposition into a mixture of stable phases is also indicated above symbols.



**Figure S6.** Compositional ternary phase diagram of Ni-Mn-Ga compounds from (Sokolovskaya Yu.A., Sokolovskiy V.V., Zagrebin M.A., Buchelnikov V.D., Zayak A.T. Ab Initio Study of the Composite Phase Diagram of Ni-Mn-Ga Shape Memory Alloys. *J. Exp. Theor. Phys.* **2017**, 152, 125–132.). The points map the most studied compositions (about 900) reported in the literature.

### 3. The ground state structure properties of the austenite and martensite phases

The structural ground-state properties and preferable atomic configurations for Ni-Mn-Ga compositions with the austenite and martensite phases are listed in Tables S5-S10, correspondingly. Note, the empty cells denote the unstable compositions. The martensitic transition temperatures  $T_m$  are summarized in Table S16.

### 4. The magnetic configurations of the austenite and martensite phases

The partial and total magnetic moments and preferable magnetic configurations for Ni-Mn-Ga compositions with the austenite and martensite phases are listed in Tables S11-S15, correspondingly. Note, the empty cells denote the unstable compositions.

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**Table S5.** Optimized lattice constants  $a$ ,  $b$ ,  $c$  (in Å) and their ratios, the total energy  $E_{tot}$  and the formation energy  $E_{form}$  (in eV/atom) as well as the preferable structure for compounds in the austenite phase from the area I.

Area I – austenite phase										
Concentration (at.%)			Lattice parameters			$b/a$	$c/a$	$E_{tot}$	$E_{form}$	Structure
Ni	Mn	Ga	$a$	$b$	$c$					
6.25	6.25	87.5								
6.25	12.5	81.25								
6.25	18.75	75								
6.25	25	68.75								
12.5	6.25	81.25								
12.5	12.5	75								
12.5	18.75	68.75								
12.5	25	62.5								
18.75	6.25	75	6.263			1.000	1.000	-3.838	0.001	225'
18.75	12.5	68.75								
18.75	18.75	62.5								
18.75	25	56.25	6.047		6.047	1.000	1.000	-5.076	-0.065	225'
25	6.25	68.75	6.158		6.156	1.000	1.000	-4.025	-0.006	225'
25	12.5	62.5								225'
25	18.75	56.25	6.002	6.002	6.002	1.000	1.000	-4.923	-0.123	225'
25	25	50	5.948	5.948	5.948	1.000	1.000	-5.349	-0.158	
31.25	6.25	62.5								225''
31.25	12.5	56.25								
31.25	18.75	50	5.907	5.907	5.907	1.000	1.000	-5.203	-0.223	225''
31.25	25	43.75	5.906	5.906	5.906	1.000	1.000	-5.538	-0.167	225''
37.5	6.25	56.25	5.932	5.932	5.935	1.000	1.000	-4.586	-0.208	225''
37.5	12.5	50								
37.5	18.75	43.75	5.866	5.866	5.864	1.000	1.000	-5.375	-0.215	225''
37.5	25	37.5								
43.75	6.25	50	5.846	5.846	5.846	1.000	1.000	-4.875	-0.317	225''
43.75	12.5	43.75								225''
43.75	18.75	37.5	5.907	5.835	5.837	0.988	0.988	-5.576	-0.237	225''
43.75	25	31.25								
50	6.25	43.75	5.821	5.821	5.820	1.000	1.000	-5.073	-0.336	225
50	12.5	37.5	5.783	5.872	5.783	1.015	1.000	-5.446	-0.318	225
50	18.75	31.25	5.806	5.806	5.805	1.000	1.000	-5.819	-0.300	225
50	25	25	5.811	5.811	5.811	1.000	1.000	-6.191	-0.281	225
56.25	6.25	37.5	5.773	5.770	5.814	1.000	1.007	-5.228	-0.311	225
56.25	12.5	31.25	5.773	5.773	5.791	1.000	1.003	-5.6	-0.292	225
56.25	18.75	25	5.769	5.769	5.769	1.000	1.000	-5.971	-0.273	225
56.25	25	18.75	5.775	5.774	5.775	1.000	1.000	-6.2999375	-0.211	225
62.5	6.25	31.25								
62.5	12.5	25								
62.5	18.75	18.75								225
62.5	25	12.5								225
68.75	6.25	25	5.710	5.710	5.709	1.000	1.000	-5.533	-0.257	225
68.75	12.5	18.75								225
68.75	18.75	12.5								225
68.75	25	6.25	5.696	5.696	5.695	1.000	1.000	-6.515	-0.067	225
75	6.25	18.75	5.683	5.683	5.681	1.000	1.000	-5.616	-0.160	225
75	12.5	12.5	5.681	5.683	5.679	1.000	1.000	-5.951	-0.105	225
75	18.75	6.25								
81.25	6.25	12.5								
81.25	12.5	6.25								
87.5	6.25	6.25								



**Table S6.** Optimized lattice constants  $a$ ,  $b$ ,  $c$  (in Å) and their ratios, the total energy  $E_{tot}$  and the formation energy  $E_{form}$  (in eV/atom) as well as the preferable structure for compounds in the austenite phase from the area II.

Area II – austenite phase										
Concentration (at.%)			Lattice parameters			$b/a$	$c/a$	$E_{tot}$	$E_{form}$	Structure
Ni	Mn	Ga	$a$	$b$	$c$					
62.5	31.25	6.25								225
56.25	31.25	12.5								225
56.25	37.5	6.25								
50	31.25	18.75	5.794	5.794	5.790	1.000	0.999	-6.518	-0.218	225
50	37.5	12.5								225
50	43.75	6.25	5.788	5.788	5.786	1.000	1.000	-7.172	-0.090	225
43.75	31.25	25	5.808	5.808	5.807	1.000	1.000	-6.367	-0.246	225
43.75	37.5	18.75	5.811	5.811	5.810	1.000	1.000	-6.692	-0.181	225
43.75	43.75	12.5	5.819	5.819	5.821	1.000	1.000	-7.021	-0.119	225
37.5	31.25	31.25	5.875	5.860	5.858	0.998	0.997	-6.138	-0.197	225
37.5	37.5	25	5.780	5.793	5.793	1.002	1.002	-6.549	-0.217	225
37.5	43.75	18.75								
31.25	31.25	37.5								
31.25	37.5	31.25								
31.25	43.75	25	5.815	5.815	5.755	1.000	0.990	-6.737	-0.194	225
25	31.25	43.75	5.904	5.904	5.902	1.000	1.000	-5.717	-0.135	225'
25	37.5	37.5								
25	43.75	31.25								
18.75	31.25	50	5.961	5.961	5.959	1.000	1.000	-5.543	-0.141	225'
18.75	37.5	43.75	5.913	5.913	5.911	1.000	1.000	-5.938	-0.145	225'
18.75	43.75	37.5								
12.5	31.25	56.25								
12.5	37.5	50								
12.5	43.75	43.75								
6.25	31.25	62.5								
6.25	37.5	56.25								
6.25	43.75	50	5.921	5.921	5.919	1.000	1.000	-5.903	-0.079	225'

**Table S7.** Optimized lattice constants  $a$ ,  $b$ ,  $c$  (in Å) and their ratios, the total energy  $E_{tot}$  and the formation energy  $E_{form}$  (in eV/atom) as well as the preferable structure for compounds in the austenite phase from the area III.

Area III – austenite phase										
Concentration (at.%)			Lattice parameters			$b/a$	$c/a$	$E_{tot}$	$E_{form}$	Structure
Ni	Mn	Ga	$a$	$b$	$c$					
43.75	50	6.25	5.761	5.761	5.755	1.000	0.999	-7.347	-0.054	216
37.5	50	12.5								
37.5	56.25	6.25								
31.25	50	18.75	5.844	5.844	5.840	1.000	0.999	-7.049	-0.115	216
31.25	56.25	12.5								
31.25	62.5	6.25								
25	50	25	5.840	5.840	5.840	1.000	1.000	-6.913	-0.159	216
25	56.25	18.75	5.827	5.827	5.827	1.000	1.000	-7.236	-0.091	216
25	62.5	12.5	5.859	5.836	5.836	0.996	0.996	-7.576	-0.040	216
25	68.75	6.25	5.850	5.850	5.850	1.000	1.000	-7.91	0.016	216
18.75	50	31.25	5.917	5.917	5.917	1.000	1.000	-6.692	-0.118	216
18.75	56.25	25	5.825	5.825	5.825	1.000	1.000	-7.101	-0.136	216
18.75	62.5	18.75	5.820	5.820	5.820	1.000	1.000	-7.433	-0.077	216
18.75	68.75	12.5								
18.75	75	6.25	5.804	5.804	5.804	1.000	1.000	-8.095	0.043	216
12.5	50	37.5	5.990	5.990	5.952	1.000	0.994	-6.466	-0.071	216
12.5	56.25	31.25	5.889	5.903	5.903	1.002	1.002	-6.878	-0.092	216
12.5	62.5	25	5.830	5.830	5.796	1.000	0.994	-7.284	-0.108	216
12.5	68.75	18.75	5.828	5.828	5.759	1.000	0.988	-7.618	-0.051	216
12.5	75	12.5								
12.5	81.25	6.25								
6.25	50	43.75	6.042	6.042	6.040	1.000	1.000	-6.224	-0.009	216
6.25	56.25	37.5	5.977	5.977	5.946	1.000	0.995	-6.649	-0.043	216
6.25	62.5	31.25	5.915	5.915	5.829	1.000	0.985	-7.054	-0.057	216
6.25	68.75	25	5.821	5.821	5.821	1.000	1.000	-7.469	-0.081	216
6.25	75	18.75	5.792	5.792	5.792	1.000	1.000	-7.806	-0.028	216
6.25	81.25	12.5								
6.25	87.5	6.25								

**Table S8.** Optimized lattice constants  $a$ ,  $b$ ,  $c$  (in Å) and their ratios, the total energy  $E_{tot}$  and the formation energy  $E_{form}$  (in eV/atom) for compounds in the martensite phase from the area I.

Area I – martensite phase									
Concentration (at.%)			Lattice parameters			$b/a$	$c/a$	$E_{tot}$	$E_{form}$
Ni	Mn	Ga	$a$	$b$	$c$				
6.25	6.25	87.5							
6.25	12.5	81.25	5.993	5.521	7.865	0.921	1.312	-3.859	0.012
6.25	18.75	75	5.756	5.306	8.034	0.922	1.396	-4.24	0.021
6.25	25	68.75	5.312		7.952	1.000	1.497	-4.687	-0.035
12.5	6.25	81.25	5.737		7.893	1.000	1.376	-3.683	-0.023
12.5	12.5	75	5.527	5.734	7.866	1.037	1.423	-4.06	-0.010
12.5	18.75	68.75	5.486		7.813	1.000	1.424	-4.453	-0.012
12.5	25	62.5	5.334		7.853	1.000	1.472	-4.898	-0.066
18.75	6.25	75	5.615		7.718	1.000	1.374	-3.867	-0.028
18.75	12.5	68.75	5.909	5.339	7.330	0.904	1.240	-4.269	-0.039
18.75	18.75	62.5	5.554		7.296		1.314	-4.668	-0.047
18.75	25	56.25	5.412		7.402		1.368	-5.078	-0.067
25	6.25	68.75	5.487		7.661	1.000	1.396	-4.069	-0.050
25	12.5	62.5	5.974	5.998	6.231	1.004	1.043	-4.48	-0.070
25	18.75	56.25							
25	25	50	run						
31.25	6.25	62.5	5.475	5.475	7.752	1.000	1.416	-4.362	-0.164
31.25	12.5	56.25	5.619	5.619	6.687	1.000	1.190	-4.765	-0.176
31.25	18.75	50							
31.25	25	43.75	5.628	5.628	6.506	1.000	1.156	-5.541	-0.170
37.5	6.25	56.25							
37.5	12.5	50	5.742	5.742	6.110	1.000	1.064	-5.046	-0.277
37.5	18.75	43.75							
37.5	25	37.5	6.098	5.813	5.811	0.953	0.953	-5.732	-0.182
43.75	6.25	50							
43.75	12.5	43.75							
43.75	18.75	37.5	5.397	5.397	6.897	1.000	1.278	-5.576	-0.237
43.75	25	31.25	5.373	5.373	6.900	1.000	1.284	-5.972	-0.242
50	6.25	43.75							
50	12.5	37.5							
50	18.75	31.25							
50	25	25	5.380	5.380	6.759	1.000	1.256	-6.199	-0.289
56.25	6.25	37.5							
56.25	12.5	31.25	5.424	5.424	6.552	1.000	1.208	-5.602	-0.294
56.25	18.75	25	5.354	5.354	6.689	1.000	1.249	-5.982	-0.284
56.25	25	18.75	5.301	5.279	6.848	0.996	1.292	-6.325	-0.236
62.5	6.25	31.25	5.324	5.324	6.681	1.000	1.255	-5.402	-0.306
62.5	12.5	25	5.289	5.289	6.727	1.000	1.272	-5.782	-0.295
62.5	18.75	18.75	5.230	5.274	6.814	1.008	1.303	-6.097	-0.219
62.5	25	12.5	5.172	5.189	6.952	1.003	1.344	-6.449	-0.180
68.75	6.25	25	5.238	5.238	6.758	1.000	1.290	-5.556	-0.280
68.75	12.5	18.75	5.188	5.226	6.851	1.007	1.321	-5.861	-0.194
68.75	18.75	12.5	5.128	5.167	6.969	1.008	1.359	-6.232	-0.174
68.75	25	6.25	5.100	5.100	7.075	1.000	1.387	-6.567	-0.119
75	6.25	18.75	5.141	5.141	6.916	1.000	1.345	-5.65	-0.194
75	12.5	12.5	5.108	5.118	6.981	1.002	1.367	-5.998	-0.152
75	18.75	6.25	5.050	5.077	7.086	1.005	1.403	-6.357	-0.120
81.25	6.25	12.5	5.044	5.064	7.051	1.004	1.398	-5.773	-0.138
81.25	12.5	6.25	5.055	5.055	7.051	1.000	1.395	-6.11	-0.084
87.5	6.25	6.25	5.201	6.529	5.154	1.255	0.991	-5.86	-0.045

**Table S9.** Optimized lattice constants  $a$ ,  $b$ ,  $c$  (in Å) and their ratios, the total energy  $E_{tot}$  and the formation energy  $E_{form}$  (in eV/atom) for compounds in the martensite phase from the area II.

Area II – martensite phase									
Concentration (at.%)			Lattice parameters			$b/a$	$c/a$	$E_{tot}$	$E_{form}$
Ni	Mn	Ga	$a$	$b$	$c$				
62.5	31.25	6.25	5.130	5.139	7.023	1.002	1.369	-6.791	-0.131
56.25	31.25	12.5	5.202	5.202	6.985	1.000	1.343	-6.665	-0.185
56.25	37.5	6.25	5.155	5.156	7.053	1.000	1.368	-7.01	-0.139
50	31.25	18.75	5.276	5.276	6.916	1.000	1.311	-6.539	-0.239
50	37.5	12.5	5.211	5.220	7.029	1.002	1.349	-6.882	-0.191
50	43.75	6.25	5.209	5.209	6.904	1.000	1.325	-7.215	-0.133
43.75	31.25	25	5.363	5.363	6.767	1.000	1.262	-6.376	-0.255
43.75	37.5	18.75	5.293	5.293	6.914	1.000	1.306	-6.713	-0.202
43.75	43.75	12.5	5.193	5.193	7.105	1.000	1.368	-7.054	-0.152
37.5	31.25	31.25	5.388	5.368	6.940	0.996	1.288	-6.152	-0.211
37.5	37.5	25	5.353	5.374	6.781	1.004	1.267	-6.562	-0.230
37.5	43.75	18.75	5.352	5.350	6.802	1.000	1.271	-6.892	-0.169
31.25	31.25	37.5	5.281	5.281	7.287	1.000	1.380	-5.929	-0.168
31.25	37.5	31.25	5.405	5.368	6.927	0.993	1.282	-6.341	-0.189
31.25	43.75	25	5.347	5.347	6.860	1.000	1.283	-6.751	-0.208
25	31.25	43.75	5.573	5.573	6.650	1.000	1.193	-5.733	-0.151
25	37.5	37.5	5.407	5.407	7.130	1.000	1.319	-6.132	-0.159
25	43.75	31.25	5.369	5.369	6.993	1.000	1.303	-6.536	-0.173
18.75	31.25	50							
18.75	37.5	43.75							
18.75	43.75	37.5	6.107	5.783	5.783	1.000	1.056	-6.321	-0.137
12.5	31.25	56.25	5.211	5.283	8.017	1.014	1.538	-5.323	-0.100
12.5	37.5	50	5.442	5.442	7.107	1.000	1.306	-5.764	-0.151
12.5	43.75	43.75	5.433	5.433	7.028	1.000	1.293	-6.147	-0.143
6.25	31.25	62.5	5.243	5.243	8.108	1.000	1.546	-5.116	-0.073
6.25	37.5	56.25	5.383	5.397	7.534	1.003	1.400	-5.515	-0.081
6.25	43.75	50	5.423	5.423	7.227	1.000	1.333	-5.962	-0.138

**Table S10.** Optimized lattice constants  $a$ ,  $b$ ,  $c$  (in Å) and their ratios, the total energy  $E_{tot}$  and the formation energy  $E_{form}$  (in eV/atom) for compounds in the martensite phase from the area III.

Area III – martensite phase									
Concentration (at.%)			Lattice parameters			$b/a$	$c/a$	$E_{tot}$	$E_{form}$
Ni	Mn	Ga	$a$	$b$	$c$				
43.75	50	6.25	5.105	5.105	7.336	1.000	1.437	-7.447	-0.154
37.5	50	12.5	5.141	5.155	7.330	1.003	1.426	-7.276	-0.163
37.5	56.25	6.25	5.131	5.117	7.232	0.997	1.410	-7.624	-0.120
31.25	50	18.75	5.251	5.251	7.154	1.000	1.362	-7.1	-0.166
31.25	56.25	12.5	5.162	5.169	7.247	1.001	1.404	-7.458	-0.134
31.25	62.5	6.25	5.145	5.142	7.191	0.999	1.398	-7.806	-0.091
25	50	25	5.362	5.362	6.868	1.000	1.281	-6.942	-0.188
25	56.25	18.75	5.267	5.267	7.123	1.000	1.352	-7.273	-0.128
25	62.5	12.5	5.185	5.185	7.185	1.000	1.386	-7.636	-0.100
25	68.75	6.25	5.155	5.155	7.156	1.000	1.388	-7.99	-0.064
18.75	50	31.25	5.416	5.416	6.959	1.000	1.285	-6.707	-0.133
18.75	56.25	25	5.378	5.378	6.894	1.000	1.282	-7.121	-0.156
18.75	62.5	18.75	5.282	5.282	7.009	1.000	1.327	-7.468	-0.112
18.75	68.75	12.5	5.182	5.180	7.161	1.000	1.382	-7.823	-0.076
18.75	75	6.25	5.156	5.156	7.109	1.000	1.379	-8.183	-0.045
12.5	50	37.5	5.403	5.403	7.193	1.000	1.331	-6.485	-0.090
12.5	56.25	31.25	5.392	5.401	7.025	1.002	1.303	-6.894	-0.108
12.5	62.5	25	5.363	5.363	6.975	1.000	1.300	-7.315	-0.139
12.5	68.75	18.75	5.251	5.251	7.102	1.000	1.353	-7.663	-0.096
12.5	75	12.5	5.181	5.189	7.136	1.001	1.377	-8.019	-0.061
12.5	81.25	6.25	5.149	5.149	7.098	1.000	1.379	-8.38	-0.031
6.25	50	43.75	5.330	5.330	7.614	1.000	1.428	-6.257	-0.042
6.25	56.25	37.5	5.370	5.370	7.294	1.000	1.358	-6.673	-0.067
6.25	62.5	31.25	5.365	5.365	7.153	1.000	1.333	-7.093	-0.096
6.25	68.75	25	5.344	5.344	7.091	1.000	1.327	-7.5	-0.112
6.25	75	18.75	5.269	5.269	7.110	1.000	1.349	-7.852	-0.074
6.25	81.25	12.5	5.205	5.205	7.103	1.000	1.365	-8.21	-0.041
6.25	87.5	6.25	5.175	5.175	7.035	1.000	1.359	-8.571	-0.011

**Table S11.** Element resolved magnetic moments (in  $\mu_B$ ), total magnetic moments (in  $\mu_B/\text{f.u.}$ ) and favorable magnetic reference state for compounds in the austenite and martensite phase from the area I.

Area I										
Concentration (at.%)			Austenite				Martensite			
Ni	Mn	Ga	$\mu_{\text{Mn}}$	$\mu_{\text{Ni}}$	$\mu_{\text{tot}}$	Order	$\mu_{\text{Mn}}$	$\mu_{\text{Ni}}$	$\mu_{\text{tot}}$	Order
6.25	6.25	87.5					3.216	0.010	0.838	FM
6.25	12.5	81.25					3.371	0.104	0.120	FM
6.25	18.75	75					2.901	-0.080	2.024	FM
6.25	25	68.75					1.697	-0.245	1.404	FM
12.5	6.25	81.25					2.87	0.026	0.719	FM
12.5	12.5	75					3.119	0.097	1.583	FM
12.5	18.75	68.75					2.885	0.135	2.154	FM
12.5	25	62.5					2.093	0.023	1.902	FM
18.75	6.25	75					2.69	0.030	0.618	FM
18.75	12.5	68.75					2.466	0.109	1.133	FM
18.75	18.75	62.5					2.772	0.115	2.097	FM
18.75	25	56.25	2.969	0.086	3.054	FM	2.561	0.211	2.643	FM
25	6.25	68.75	2.022	0.030	0.412	FM	2.965	0.029	0.746	FM
25	12.5	62.5					2.633	0.057	1.308	FM
25	18.75	56.25	2.849	0.092	2.211	FM				
25	25	50	2.945	0.071	2.998	FM				
31.25	6.25	62.5					3.015	0.063	0.814	FM
31.25	12.5	56.25					2.687	0.087	1.393	FM
31.25	18.75	50	2.826	0.072	2.131	FM				
31.25	25	43.75	2.973	0.154	3.132	FM	2.955	0.134	3.173	FM
37.5	6.25	56.25	2.482	0.065	0.695	FM				
37.5	12.5	50					2.464	0.060	1.170	FM
37.5	18.75	43.75	2.800	0.045	2.169	FM				
37.5	25	37.5					3.34	0.287	3.778	FM
43.75	6.25	50	2.266	0.050	0.525	FM				
43.75	12.5	43.75								
43.75	18.75	37.5	3.344	0.237	2.913	FM	3.298	0.408	3.071	FM
43.75	25	31.25					3.29	0.432	4.028	FM
50	6.25	43.75	3.503	0.086	1.044	FM				
50	12.5	37.5	3.420	0.170	2.046	FM				
50	18.75	31.25	3.390	0.263	3.054	FM				
50	25	25	3.387	0.357	4.090	FM	3.305	0.423	4.108	FM
56.25	6.25	37.5	3.437	0.108	1.102	FM				
56.25	12.5	31.25	3.433	0.235	2.242	FM	3.386	0.326	2.437	FM
56.25	18.75	25	3.416	0.338	3.329	FM	3.347	0.345	3.375	FM
56.25	25	18.75	3.355	0.402	4.261	FM	3.215	0.372	4.008	FM
62.5	6.25	31.25					3.394	0.145	1.205	FM
62.5	12.5	25					3.34	0.285	2.356	FM
62.5	18.75	18.75					3.262	0.353	3.295	FM
62.5	25	12.5					3.213	0.361	4.014	FM
68.75	6.25	25	3.346	0.126	1.178	FM	3.242	0.116	1.112	FM
68.75	12.5	18.75					3.277	0.325	2.489	FM
68.75	18.75	12.5					3.203	0.417	3.495	FM
68.75	25	6.25	3.246	0.345	4.110	FM	3.151	0.507	4.496	FM
75	6.25	18.75	3.250	0.177	0.840	FM	3.159	0.211	1.379	FM
75	12.5	12.5	3.298	0.285	2.362	FM	3.163	0.447	2.858	FM
75	18.75	6.25					3.155	0.502	3.803	FM
81.25	6.25	12.5					3.124	0.377	1.937	FM
81.25	12.5	6.25					3.216	0.571	3.376	FM
87.5	6.25	6.25					3.092	0.501	2.441	FM

**Table S12.** Element resolved magnetic moments (in  $\mu_B$ ), total magnetic moments (in  $\mu_B$ /f.u.) and favorable magnetic reference state for compounds in the austenite phase from the area II. Here  $\mu_{\text{Mn(Ga)}}$  and  $\mu_{\text{Mn(Ni)}}$  are the magnetic moments of Mn atoms placed at the Ga- and Ni sublattice, respectively.

			Area II – austenite phase					
Concentration (at.%)			$\mu_{\text{Mn}}$	$\mu_{\text{Mn(Ga)}}$	$\mu_{\text{Mn(Ni)}}$	$\mu_{\text{Ni}}$	$\mu_{\text{tot}}$	Order
Ni	Mn	Ga						
6.25	31.25	62.5						
6.25	37.5	56.25						
6.25	43.75	50	1.915		2.321	0.075	3.589	FM
12.5	31.25	56.25						
12.5	37.5	50						
12.5	43.75	43.75						
18.75	31.25	50	2.807		3.011	0.082	3.535	FM
18.75	37.5	43.75	2.308		-3.174	0.030	2.065	FIM-1
18.75	43.75	37.5						
25	31.25	43.75	2.652		2.699	0.136	3.416	FM
25	37.5	37.5						
25	43.75	31.25						
31.25	31.25	37.5						
31.25	37.5	31.25						
31.25	43.75	25						
37.5	31.25	31.25	3.271		-2.792	0.306	3.051	FIM-1
37.5	37.5	25	3.184		-1.225	0.371	3.162	FIM-1
37.5	43.75	18.75						
43.75	31.25	25	3.238		-2.841	0.266	2.983	FIM-1
43.75	37.5	18.75	3.305		-2.720	0.429	4.232	FIM-1
43.75	43.75	12.5	3.364		-2.699	0.563	5.426	FIM-1
50	31.25	18.75	3.368	-3.515		0.304	3.078	FIM-2
50	37.5	12.5						
50	43.75	6.25	3.442	-3.465		0.094	0.899	FIM-2
56.25	31.25	12.5						
56.25	37.5	6.25						
62.5	31.25	6.25						

**Table S13.** Element resolved magnetic moments (in  $\mu_B$ ), total magnetic moments (in  $\mu_B/\text{f.u.}$ ) and favorable magnetic reference state for compounds in the martensite phase from the area II. Here  $\mu_{\text{Mn}(\text{Ga})}$  and  $\mu_{\text{Mn}(\text{Ni})}$  are the magnetic moments of Mn atoms placed at the Ga- and Ni sublattice, respectively.

			Area II – martensite phase					Order
Concentration (at.%)			$\mu_{\text{Mn}}$	$\mu_{\text{Mn}(\text{Ga})}$	$\mu_{\text{Mn}(\text{Ni})}$	$\mu_{\text{Ni}}$	$\mu_{\text{tot}}$	
Ni	Mn	Ga						
6.25	31.25	62.5	2.336		2.851	0.069	2.861	FM
6.25	37.5	56.25	2.503		2.621	0.127	3.675	FM
6.25	43.75	50	2.595		2.471	0.100	4.354	FM
12.5	31.25	56.25	2.682		3.057	0.330	3.449	FM
12.5	37.5	50	2.504		2.388	0.040	3.598	FM
12.5	43.75	43.75	2.474	-3.081	2.228	0.077	2.699	FIM-2
18.75	31.25	50						
18.75	37.5	43.75						
18.75	43.75	37.5	2.630	-3.033	0.366	0.036	1.128	FIM-2
25	31.25	43.75	2.713		-3.082	0.072	1.981	FIM-1
25	37.5	37.5	3.232		-2.573	0.445	2.456	FIM-1
25	43.75	31.25	3.105		-2.184	0.437	1.932	FIM-1
31.25	31.25	37.5	3.011		1.887	0.069	3.821	FM
31.25	37.5	31.25	3.156		-2.153	0.426	2.622	FIM-1
31.25	43.75	25	3.045		-2.271	0.378	1.810	FIM-1
37.5	31.25	31.25	3.244		-2.339	0.442	3.337	FIM-1
37.5	37.5	25	3.097		-2.192	0.392	2.572	FIM-1
37.5	43.75	18.75	3.061	3.038	-2.267	0.469	3.396	FIM-1
43.75	31.25	25	3.176		-2.464	0.399	3.245	FIM-1
43.75	37.5	18.75	3.114	-3.281	-2.633		2.103	FIM-1(2)
43.75	43.75	12.5	3.053	-3.169	-2.693	0.135	1.005	FIM-1(2)
50	31.25	18.75	3.226	-3.364		0.283	2.902	FIM-2
50	37.5	12.5	3.190	-3.247		0.203	1.936	FIM-2
50	43.75	6.25	3.137	-3.156		0.108	0.965	FIM-2
56.25	31.25	12.5	3.177	-3.291		0.264	2.911	FIM-2
56.25	37.5	6.25	3.122	-3.201		0.162	1.863	FIM-2
62.5	31.25	6.25	3.090	-3.211		0.226	2.823	FIM-2



**Table S14.** Element resolved magnetic moments (in  $\mu_B$ ), total magnetic moments (in  $\mu_B/\text{f.u.}$ ) and favorable magnetic reference state for compounds in the austenite phase from the area III. Here  $\mu_{\text{Mn}(4a)}$  and  $\mu_{\text{Mn}(4b)}$  are the magnetic moments of Mn atoms, which occupy 4a and 4b Wyckoff sites while  $\mu_{\text{Mn}(\text{Ga})}$  and  $\mu_{\text{Mn}(\text{Ni})}$  are the Mn magnetic moments at the Ga- and Ni sites, respectively.

Area III – austenite phase									
Concentration (at.%)			$\mu_{\text{Mn}^{(4a)}}$	$\mu_{\text{Mn}^{(4b)}}$	$\mu_{\text{Mn}^{(\text{Ga})}}$	$\mu_{\text{Mn}^{(\text{Ni})}}$	$\mu_{\text{Ni}}$	$\mu_{\text{tot}}$	Order
Ni	Mn	Ga							
6.25	50	43.75	-2.927	3.015			0.060	0.124	FIM-3
6.25	56.25	37.5	-2.812	2.945		0.471	0.039	0.269	FIM-3
6.25	62.5	31.25	-2.293	2.982		-0.097	0.236	0.760	FIM-7
6.25	68.75	25	-1.748	2.990		-1.233	0.229	0.456	FIM-7
6.25	75	18.75	-1.460	2.885	2.636	-1.137	0.345	1.391	FIM-7
6.25	81.25	12.5							
6.25	87.5	6.25							
12.5	50	37.5	-2.899	3.003		0.087	0.034	0.150	FIM-3
12.5	56.25	31.25	-2.495	3.044		0.490	0.236	0.847	FIM-3
12.5	62.5	25	-1.959	3.021		-0.911	0.294	0.832	FIM-7
12.5	68.75	18.75	-1.830	2.979	2.798	-0.682	0.384	1.778	FIM-7
12.5	75	12.5							
12.5	81.25	6.25							
18.75	50	31.25	-2.710	3.102			0.209	0.587	FIM-3
18.75	56.25	25	-2.153	3.065		-0.503	0.326	1.109	FIM-7
18.75	62.5	18.75	-2.107	3.055	2.893	-0.173	0.376	1.997	FIM-7
18.75	68.75	12.5							
18.75	75	6.25							
25	50	25	-2.345	3.131			0.329	1.191	FIM-3
25	56.25	18.75	-2.397	3.155	3.004		0.329	1.930	FIM-3
25	62.5	12.5	-2.394	3.147	3.095		0.413	2.827	FIM-3
25	68.75	6.25							
31.25	50	18.75	-2.755	3.148			0.259	0.566	FIM-3
31.25	56.25	12.5							
31.25	62.5	6.25							
37.5	50	12.5							
37.5	56.25	6.25							
43.75	50	6.25	-2.791	2.942			0.436	0.213	FIM-3

**Table S15.** Element resolved magnetic moments (in  $\mu_B$ ), total magnetic moments (in  $\mu_B/\text{f.u.}$ ) and favorable magnetic reference state for compounds in the martensite phase from the area III. Here  $\mu_{\text{Mn}(4a)}$  and  $\mu_{\text{Mn}(4b)}$  are the magnetic moments of Mn atoms, which occupy 4a and 4b Wyckoff sites while  $\mu_{\text{Mn}(\text{Ga})}$  and  $\mu_{\text{Mn}(\text{Ni})}$  are the Mn magnetic moments at the Ga- and Ni sites, respectively.

Area III – martensite phase									
Concentration (at.%)			$\mu_{\text{Mn}}^{(4a)}$	$\mu_{\text{Mn}}^{(4b)}$	$\mu_{\text{Mn}}^{(\text{Ga})}$	$\mu_{\text{Mn}}^{(\text{Ni})}$	$\mu_{\text{Ni}}$	$\mu_{\text{tot}}$	Order
Ni	Mn	Ga							
6.25	50	43.75	-2.835	2.718			0.200	0.191	FIM-3
6.25	56.25	37.5	-2.522	2.933		-0.832	0.232	0.310	FIM-7
6.25	62.5	31.25	-2.391	2.945		-1.879	0.109	0.292	FIM-7
6.25	68.75	25	-2.373	2.885		-2.213	0.067	1.110	FIM-7
6.25	75	18.75	-2.251	2.622	2.127	-2.113	0.061	0.647	FIM-7
6.25	81.25	12.5	-2.154	2.381	2.039	-2.066	0.074	0.270	FIM-7
6.25	87.5	6.25	-2.048	2.121	2.005	-2.072	0.124	0.056	FIM-7
12.5	50	37.5	-2.693	2.814			0.029	0.117	FIM-3
12.5	56.25	31.25	-2.346	2.969		-1.718	0.245	0.365	FIM-7
12.5	62.5	25	-2.332	2.918		-2.144	0.171	0.352	FIM-7
12.5	68.75	18.75	-2.248	2.641	2.048	-2.073	0.105	0.046	FIM-7
12.5	75	12.5	-2.216	2.409	2.003	-2.114	0.101	0.212	FIM-7
12.5	81.25	6.25	-2.146	2.221	2.007	-2.167	0.094	0.562	FIM-7
18.75	50	31.25	-2.483	2.962			0.237	0.694	FIM-3
18.75	56.25	25	-2.321	2.959		-2.039	0.236	0.352	FIM-7
18.75	62.5	18.75	-2.290	2.754	2.229	-1.933	0.232	0.754	FIM-7
18.75	68.75	12.5	-2.449	2.524	2.1315	-2.027	0.176	0.787	FIM-7
18.75	75	6.25	-2.354	2.356	2.199	-2.124	0.223	1.310	FIM-7
25	50	25	-2.294	2.997			0.262	1.021	FIM-3
25	56.25	18.75	-2.370	2.858	2.333		0.262	1.398	FIM-3
25	62.5	12.5	-2.618	2.640	2.344		0.239	1.467	FIM-3
25	68.75	6.25	-2.527	2.495	2.398		0.279	2.079	FIM-3
31.25	50	18.75	-2.744	2.996			0.168	0.351	FIM-3
31.25	56.25	12.5	-2.789	2.770	2.455		0.168	0.731	FIM-3
31.25	62.5	6.25	-2.710	2.683	2.5235		0.231	1.480	FIM-3
37.5	50	12.5	-2.969	2.960			0.081	0.030	FIM-3
37.5	56.25	6.25	-2.822	2.820	2.549		0.159	0.778	FIM-3
43.75	50	6.25	-3.052	2.997			0.031	0.127	FIM-3

**Table S16.** Martensitic transition temperature  $T_m$  (in K) for compositions from the areas I, II, and III.

Area I				Area II				Area III			
Concentration (at.%)			$T_m$	Concentration (at.%)			$T_m$	Concentration (at.%)			$T_m$
Ni	Mn	Ga		Ni	Mn	Ga		Ni	Mn	Ga	
6.25	6.25	87.5	23	62.5	31.25	6.25	244	43.75	50	6.25	1160
6.25	12.5	81.25		56.25	31.25	12.5		37.5	50	12.5	
6.25	18.75	75		56.25	37.5	6.25		37.5	56.25	6.25	
6.25	25	68.75		50	31.25	18.75		31.25	50	18.75	
12.5	6.25	81.25		50	37.5	12.5		31.25	56.25	12.5	
12.5	12.5	75		50	43.75	6.25		31.25	62.5	6.25	
12.5	18.75	68.75		43.75	31.25	25		25	50	25	
12.5	25	62.5		43.75	37.5	18.75		25	56.25	18.75	
18.75	6.25	75		43.75	43.75	12.5		25	62.5	12.5	
18.75	12.5	68.75		37.5	31.25	31.25		25	68.75	6.25	
18.75	18.75	62.5	510	37.5	37.5	25	151	18.75	50	31.25	174
18.75	25	56.25		37.5	43.75	18.75		18.75	56.25	25	
25	6.25	68.75		31.25	31.25	37.5		18.75	62.5	18.75	
25	12.5	62.5		31.25	37.5	31.25		18.75	68.75	12.5	
25	18.75	56.25		31.25	43.75	25		18.75	75	6.25	
25	25	50		25	31.25	43.75		12.5	50	37.5	
31.25	6.25	62.5		25	37.5	37.5		12.5	56.25	31.25	
31.25	12.5	56.25		25	43.75	31.25		12.5	62.5	25	
31.25	18.75	50		18.75	31.25	50		12.5	68.75	18.75	
31.25	25	43.75		18.75	37.5	43.75		12.5	75	12.5	
37.5	6.25	56.25	35	18.75	43.75	37.5	162	12.5	81.25	6.25	383
37.5	12.5	50		12.5	31.25	56.25		6.25	50	43.75	
37.5	18.75	43.75		12.5	37.5	50		6.25	56.25	37.5	
37.5	25	37.5		12.5	43.75	43.75		6.25	62.5	31.25	
43.75	6.25	50		6.25	31.25	62.5		6.25	68.75	25	
43.75	12.5	43.75		6.25	37.5	56.25		6.25	75	18.75	
43.75	18.75	37.5		6.25	43.75	50		6.25	81.25	12.5	
43.75	25	31.25						6.25	87.5	6.25	
50	6.25	43.75									
50	12.5	37.5									
50	18.75	31.25	93				684				534
50	25	25									
56.25	6.25	37.5									
56.25	12.5	31.25									
56.25	18.75	25									
56.25	25	18.75									
62.5	6.25	31.25									
62.5	12.5	25									
62.5	18.75	18.75									
62.5	25	12.5									
68.75	6.25	25	266								545
68.75	12.5	18.75									
68.75	18.75	12.5									
68.75	25	6.25									
75	6.25	18.75									
75	12.5	12.5									
75	18.75	6.25									
81.25	6.25	12.5									
81.25	12.5	6.25									
87.5	6.25	6.25									