

Low temperature kinetic properties and structure of $\text{Ni}_{50+x}\text{Mn}_{25-x+y}\text{Ga}_{25-y}$ alloys with shape memory

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Abstract. We present results of kinetic coefficient experiments and structure studies in $\text{Ni}_{50+x}\text{Mn}_{25-x+y}\text{Ga}_{25-y}$ alloys at low temperatures. The magnetic, galvanomagnetic and electrical properties were measured in the temperature range from 2 to 80 K and in magnetic fields of up to 15 T. Two types of samples were investigated: cast samples with a grain size of about 100-500 μm and quenched samples with a grain size of about 300-500 nm. We find that the deviation from the stoichiometric composition leads to an increase of the structural and magnetic disorder and, hence, to changes in the low temperature kinetic properties. Similar effects are observed as the result of a quench treatment.

1. Introduction

Studying the physical properties of the stoichiometric Ni_2MnGa compound is of great interest due to the observed magneto-operated shape memory. Such studies are aimed at creating new materials with the best functional properties, i.e., with high reversible magneto-deformation and a big magneto-caloric effect, with a high martensitic transformation temperature T_M and with magnetic phase transitions T_C close to room temperature. However, a purposeful search for such materials requires a detailed knowledge of their electronic characteristics. Therefore, we studied the crystalline structure, the magnetic, the galvanomagnetic and the electrical properties of $\text{Ni}_{50+x}\text{Mn}_{25-x+y}\text{Ga}_{25-y}$ ($x=0, y=0; x=4, y=0$ and $x=0, y=3.5$) alloys at low temperatures $T \ll (T_M \text{ and } T_C)$. The changes in the crystalline structure and the peculiarities of their low temperature kinetic properties after a quench treatment were investigated.

2. Experimental

The crystalline specimens of the alloy were melted in an induction furnace and arc-melted on a water-cooled copper bottom. The ingots were subjected to a long homogenization annealing treatment as in Refs [1-4]. Two types of samples were investigated: ordered (cast) samples and disordered (quenched) samples. Atomic disorder was introduced by rapid quenching (at a rate of about 10^5 K/s). The crystalline structure was studied by transmission electron microscopy and by X-ray diffraction. The magnetic, galvanomagnetic and electrical properties were measured by conventional methods.

3. Results

3.1. Crystalline structure

Using X-ray diffraction and transmission electron microscopy, we find that the alloys have the austenitic fine-grained structure of the $L2_1$ type at temperatures $T > T_M$. The average grain size is about 100-500 μm in the cast samples and about 300-500 nm in the quenched samples.

Cooling the alloys below T_M results in a martensitic transformation. In this case, it turns out that the regions of the fine-lamellar martensite have a so called five-layer modulated substructure [2, 3]. Subsequent cooling to below T_M is accompanied by the martensite-martensitic transformation $5M \rightarrow 7M$. At lower temperatures $T \ll (T_M \text{ and } T_C)$ the alloys have the 7M martensitic substructure [1-4]. Besides, the samples contain untwinned lamellar crystals of unmodulated tetragonal martensite (UM-martensite) at temperatures below T_M .

X-ray diffraction confirmed the development of the sequence of martensitic transformations, both in the cast and in the quenched alloys according to the scheme $L2_1 \rightarrow 5M + UM \rightarrow 7M + UM$.

3.2. Resistivity

Figure 1 shows the temperature dependence of the electro- and magneto-resistivity of the alloys. The

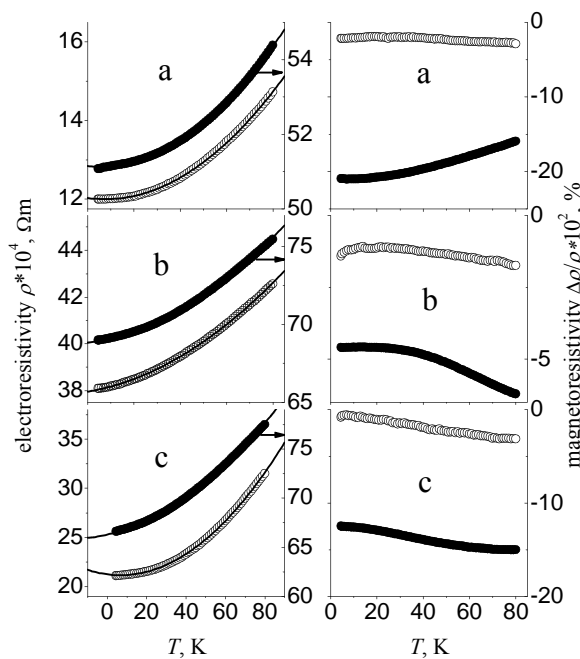


Figure 1. Temperature dependence of the electro- and magneto-resistivity for $\text{Ni}_{50+x}\text{Mn}_{25-x+y}\text{Ga}_{25-y}$ alloys.

- (a) for $\text{Ni}_{50}\text{Mn}_{25}\text{Ga}_{25}$, (b) for $\text{Ni}_{50}\text{Mn}_{28.5}\text{Ga}_{21.5}$ and
- (c) for $\text{Ni}_{54}\text{Mn}_{21}\text{Ga}_{25}$.

Data for the cast samples are marked by the full circles and for the quenched samples by the open circles. The solid lines represent expression (1).

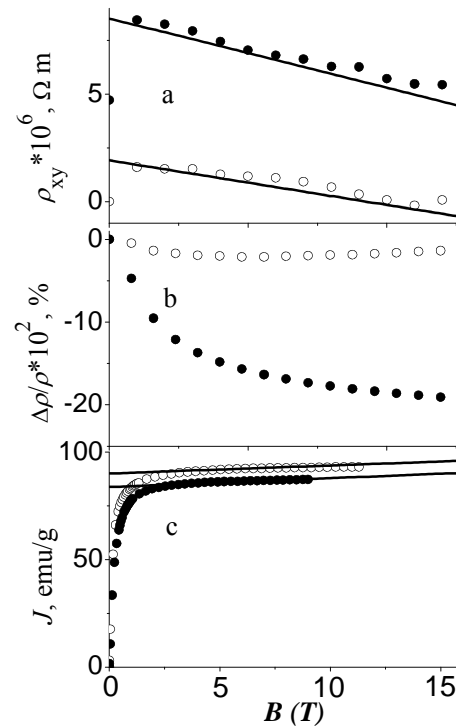


Figure 2. Field dependence of the Hall resistivity (a), the magneto-resistivity (b) and the magnetization (c) for the $\text{Ni}_{50}\text{Mn}_{25}\text{Ga}_{25}$ alloy at $T=4.2$ K. Data for the cast samples are marked by the full circles and for the quenched samples by the open circles. The solid lines represent expressions (2) and (3).

resistivity $\rho(T)$ can be described by the following expression, which is typical of ferromagnetic metals [5]

$$\rho = \rho_0 + a \cdot T + b \cdot T^2 \quad (1)$$

Here ρ_0 is residual resistivity, a and b are coefficients. Table 1 presents the experimental values of ρ_0 , a and b , obtained at $H=0$ and in a field of 10 T. According to Tab.1, the deviation from the stoichiometric compound composition in the cast alloys at $H=0$ leads to almost a doubling of ρ_0 , when the “strong” magnetic Mn atoms are replaced by “weak” magnetic Ni atoms. The residual resistivity ρ_0 increases by a factor of three, when the non-magnetic Ga atoms are replaced by the magnetic Mn atoms. Approximately the same changes take place in the quenched samples (Tab.1). The temperature dependence of $\rho(T)$ in a field of 10T demonstrates a stronger change of ρ_0 in the quenched samples in a magnetic field.

Table 1. Summary of ρ_0 , a and b .

Alloy		B , T	$\rho_0 \cdot 10^4$, Ωm	$a \cdot 10^6$, $\Omega\text{m} / \text{K}$	$b \cdot 10^8$, $\Omega\text{m} / \text{K}^2$
$\text{Ni}_{50}\text{Mn}_{25}\text{Ga}_{25}$	cast	0	12.03	-0.786	5.2
		10	11.77	-0.65	4.764
	quenched	0	51.15	-0.639	5.8
		10	42.23	-0.473	7.9
$\text{Ni}_{54}\text{Mn}_{21}\text{Ga}_{25}$	cast	0	21.26	-2.594	19.38
		10	20.74	-3.205	18.74
	quenched	0	66.3	4.637	11.9
		10	59.2	1.27	11.24
$\text{Ni}_{50}\text{Mn}_{28.5}\text{Ga}_{21.5}$	cast	0	37.95	1.68	5.18
		10	37.48	1.945	4.403
	quenched	0	68.83	1.892	8.185
		10	65.78	2.471	5.218

The analysis of the experimental data (Fig.1 and Tab.1) shows the following. Firstly, the residual resistivity ρ_0 strongly depends on the heterogeneity of the magnetic subsystem in the alloy. Secondly, the coefficient a is mainly caused by conduction electron scattering by the spin waves and the sign of the coefficient a depends on the type of the electron dispersion law. Thirdly, the coefficient b is determined both by electron scattering by the static and dynamic alloy heterogeneities and by the electron-electron transfer from the s - to the d -band.

3.3. Magnetic and galvanomagnetic properties

The field dependence of the magnetic and galvanomagnetic properties was measured at $T=4.2\text{K}$. As an example, results for $\text{Ni}_{50}\text{Mn}_{25}\text{Ga}_{25}$ are shown in Figure 2. The magnetization J becomes an almost linear function of field at $B>2.5\text{T}$, i.e., in the region of a “pseudoparaprocess”, when the following expression holds

$$J(H) = J_S + \chi_P \cdot H \quad (2)$$

Subsequently, the spontaneous magnetization J_S and the magnetic susceptibility χ_P can be obtained. To describe the Hall effect in magnetic materials, the following equation is usually employed

$$\rho_{xy} = R_0 B + 4\pi R_S J, \quad (3)$$

where ρ_{xy} is the Hall resistivity, R_0 is the normal and R_S the anomalous Hall coefficient, $B=H+(4\pi N)J$ is the induction in the sample, and N is the demagnetization factor.

Table 2 lists the values of J_S , χ_P , R_0 and R_S for the alloys. Accordingly, the deviation of the alloy composition from the stoichiometric $\text{Ni}_{50}\text{Mn}_{25}\text{Ga}_{25}$ composition leads to a decrease of J_S . This can be understood by stipulating that “weak” magnetic Ni atoms replace “strong” magnetic Mn atoms. However, it is difficult to explain the J_S decrease, when the magnetic Mn atoms are replaced by the non-magnetic Ga atoms. This could be explained by the appearance of frustrated changed coupling,

which leads to antiferromagnetic ordering of the magnetic moments of individual Mn atoms, situated at Ga positions. Most likely the same reason causes the J_S decrease after the rapid quenching. Accordingly, the susceptibility χ_P increases while J_S decreases (Tab.2).

Table 2. Summary of J_S , χ_P , R_0 and R_S .

Alloy		J_S , Am ² /kg	$\chi_P \cdot 10^6$, m ³ /kg	$R_0 \cdot 10^{14}$, m ³ /A·s	$R_S \cdot 10^{13}$, m ³ /A·s
Ni ₅₀ Mn ₂₅ Ga ₂₅	cast	90.1	0.466	-2.9	2.67
	quenched	83.8	0.518	-9.16	12.7
Ni ₅₄ Mn ₂₁ Ga ₂₅	cast	81.6	0.351	0.31	-1.56
	quenched	55.9	0.779	-3.49	3.52
Ni ₅₀ Mn _{28.5} Ga _{21.5}	cast	86	0.26	-3.49	8.35
	quenched	75.3	0.704	-0.054	-1.3

According to Table 2, the magnitude of the normal Hall coefficient R_0 is typical for transition metals. R_0 is negative for almost every sample (Tab.2). $R_0 > 0$ occurs only for the cast Ni₅₄Mn₂₁Ga₂₅ alloy and R_0 is almost zero in quenched Ni₅₄Mn₂₁Ga₂₅ sample. Apparently, the *s*-electrons are the main current carriers in the investigated alloys, in a good agreement with other data [1].

According to Table 2, the value of the anomalous Hall coefficient R_S is higher by two orders of magnitude than that of the normal Hall coefficient R_0 . Hence, the Hall effect in these ferromagnetic compounds is mainly determined by its anomalous contribution. The signs of R_0 and R_S are opposite for almost every sample (Tab.2). This behavior of the Hall effect may be caused by a rearrangement of the electron energy-band structure, when deviations from the stoichiometric Ni₅₀Mn₂₅Ga₂₅ composition occur and under the quenching [1-3].

4. Conclusions

1. We have shown that increasing the structural and magnetic disorder in Ni_{50+x}Mn_{25-x+y}Ga_{25-y} alloys leads to an increase of the residual resistance. The temperature dependent contribution to the resistivity is determined by electron-electron scattering and by electron scattering by the spin waves.
2. We found that the deviation from the stoichiometric Ni₅₀Mn₂₅Ga₂₅ composition and rapid quenching lead to a decrease of the spontaneous magnetization and to an increase of the magnetic susceptibility.
3. We demonstrated that the magnitude and the signs of the Hall coefficients are also changed. These changes are apparently caused by a rearrangement of the electron energy-band structure.

5. Announcements

This work was supported by the Russian Foundation for Basic Research (projects 06-02-16695, 07-03-96062, 07-02-97606) and by the Austrian Academy of Sciences.

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