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High-Field Galvanomagnetic Properties and Structure of Ni-Mn-Ga Nanocrystalline Alloys with Shape Memory

E.B. Marchenkova · V.V. Marchenkov · N.I. Kourov · V.G. Pushin · A.V. Korolev · H.W. Weber

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Abstract Ni-Mn-Ga based compounds are of great interest due to their magneto- and temperature operated shape memory. To obtain new results on their electronic structure, we studied the low-temperature kinetic properties and the structure of Ni-Mn-Ga alloys at temperatures $T \ll T_M, T_C$ (T_M is the temperature of the martensitic transition and T_C the temperature of the magnetic (Curie) transition, which are both close to room temperature). Ordered (cast) and disordered samples, having a nanocrystalline substructure, were investigated. The galvanomagnetic and electrical properties were measured in the temperature interval from 2 to 80 K and in magnetic fields of up to 15 T. We find that the electrical and the high-field properties of these alloys strongly change due to the transition into the nanocrystalline state.

Keywords High-field galvanomagnetic properties · Ni-Mn-Ga alloys · Shape memory

1 Introduction

Studies of the physical properties of the stoichiometric Ni_2MnGa compounds are interesting, particularly in view of creating new materials with the best functional properties, i.e. with high reversible magneto-deformation and a large magneto-caloric effect, with a high martensitic transformation temperature and with magnetic phase transitions close to room temperature. A successful search for such materials requires a detailed knowledge of their electronic characteristics. Therefore, we studied the crystalline structure, the galvanomagnetic and the electrical properties of

E.B. Marchenkova (✉) · V.V. Marchenkov · N.I. Kourov · V.G. Pushin · A.V. Korolev
Institute of Metal Physics, 620041 Ekaterinburg, Russia
e-mail: march@imp.uran.ru

H.W. Weber
Atomic Institute of the Austrian Universities, 1020 Vienna, Austria

$Ni_{50+x}Mn_{25-x+y}Ga_{25-y}$ ($x = 0$, $y = 0$; $x = 4$, $y = 0$ and $x = 0$, $y = 3.5$) alloys at low temperatures $T \ll (T_M$ and $T_C)$. Changes in their crystalline structure and peculiarities of their low temperature kinetic properties after a quench treatment and severe plastic deformation were investigated.

2 Experimental Details

The crystalline specimens of the alloys 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 described in [1–4]. Three types of samples were investigated: ordered (cast) samples; nanocrystalline samples, disordered by ultra-rapid melt quenching (RMQ); and nanocrystalline samples, disordered by severe plastic deformation by torsion under Bridgman anvil pressure (SPDT). The crystalline structure was studied by transmission electron microscopy and by X-ray diffraction. The galvanomagnetic and electrical properties were measured by conventional methods.

3 Results

From X-ray diffraction and transmission electron microscopy, we find that these alloys have the austenitic fine-grained structure of the $L2_1$ type at temperatures $T > T_M$. The average grain size is about 500 μm in the cast samples; about 300–500 nm and 10–20 nm in the nanocrystalline samples after the RMQ and SPDT treatments, respectively.

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 $5M - 7M$. At lower temperatures $T \ll (T_M$ and $T_C)$ the alloys have the 7M martensitic substructure [1–4]. In addition, the samples contain untwinned lamellar crystals of the 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 $L2_1 - 5M + UM - 7M + UM$.

Figure 1 shows the temperature dependence of the electro- and magnetoresistivity of the alloys. The transition into the nanocrystalline state strongly changes both the electro- and the magnetoconductivity: the residual resistivity ρ_0 increases and a region with a negative temperature resistance coefficient appears in the SPDT alloys. Such a behavior is typical for high resistivity magnetic alloys. The resistivity $\rho(T)$ can be described by the following expression, which is typical of ferromagnetic metals [5]

$$\rho = \rho_0 + aT + bT^2. \tag{1}$$

Here a and b are coefficients. The experimental data for ρ_0, a and b were obtained at $H = 0$ and in a filed of 10 T. Accordingly, the deviation from the stoichiometric compound composition in the cast alloys at $H = 0$ leads nearly to a doubling of ρ_0,

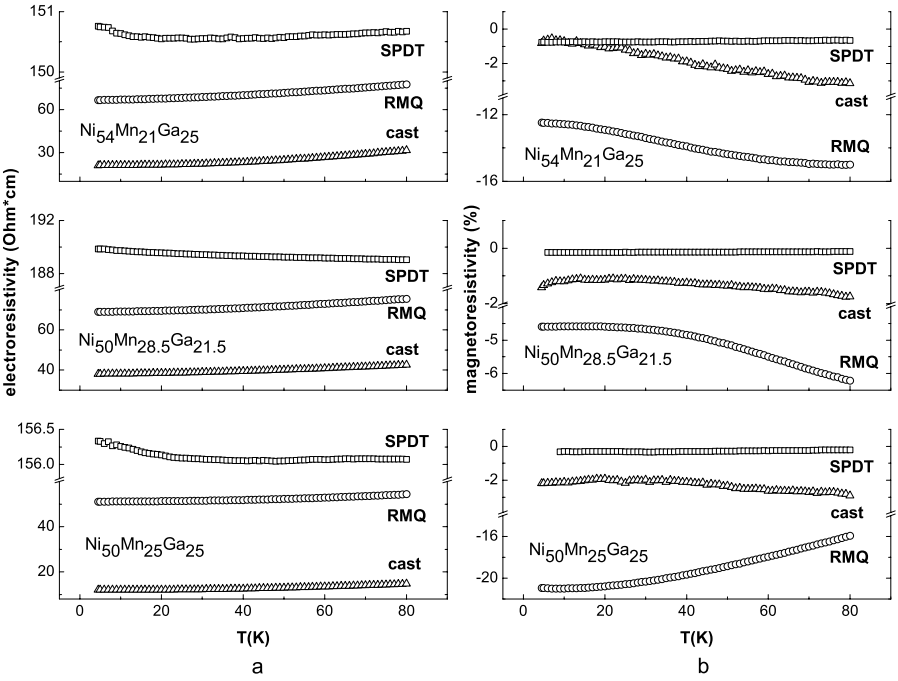

Fig. 1 Temperature dependence of the electro- and magnetoresistivity in a field of 10 T for Ni-Mn-Ga alloys

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 following the RMQ treatment. The transition into the nanocrystalline state under the SPDT treatment leads to a stronger increase of ρ_0 (up to 150–200 μOhm cm).

The relative changes of ρ_0 in a field of 10 T are approximately (0.2–20)%, both in the ordered and disordered alloys. The strongest changes are observed in the a and b coefficients. The analysis of the experimental data shows that the temperature dependence part of the resistivity is apparently determined by two main scattering mechanism: by electron-electron interaction [5] and by conduction electron scattering by the spin waves [6]. The field dependence of the transverse magnetoresistivity $\frac{\rho-\rho_0}{\rho_0}$ was measured at $T = 4.2$ K. The analysis of the experimental curves shows that $\frac{\rho-\rho_0}{\rho_0}$ can be described by the following expression

$$\frac{\rho - \rho_0}{\rho_0} = A + BH + CH^2. \tag{2}$$

Here A, B and C are coefficients. It turns out that $C > 0$ for ordered (cast) alloys, having relatively low resistivity, and $C < 0$ for disordered (RMQ) samples, having higher resistivity. The magnetoresistivity has two main contributions, namely a positive component due to the Lorentz force causing conduction electrons to spiral, and

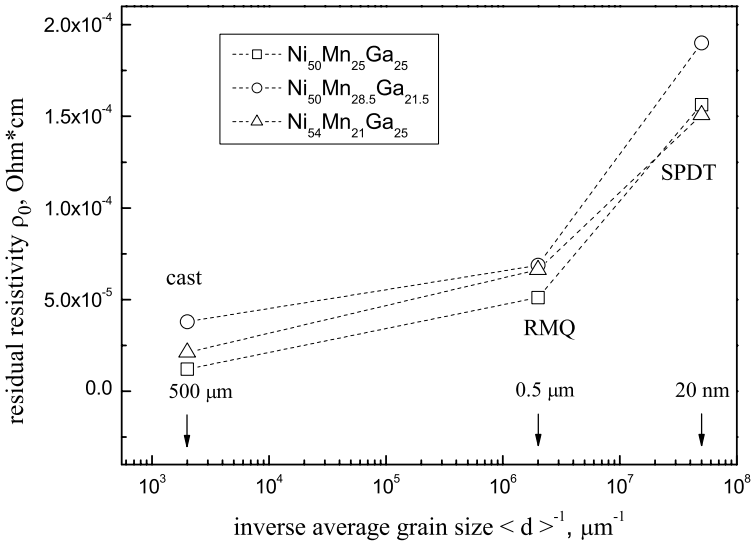

Fig. 2 "Size" dependence of the residual resistivity for Ni-Mn-Ga alloys

a negative component due to the ordering of spin heterogeneities in a magnetic field. The last contribution is dominant.

Figure 2 demonstrates the "size" dependence of the residual resistivity at $T = 4.2$ K, where the abscissa is the inverse average grain size plotted on a logarithmic scale. The decrease of the grain size leads to a transition into the nanocrystalline state and the electroresistivity increases significantly.

4 Conclusions

We have shown that the transition into the nanocrystalline state under RMQ and SPDT treatments leads to essential changes of the electrical and high-field galvanomagnetic properties of Ni-Mn-Ga alloys with shape memory.

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