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Spin Disordered Resistivity of the Heusler Ni_2MnGa-based Alloys

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Electrical resistivity of the selected Heusler off-stoichiometric $(NiCo)_2Mn(GaIn)$ alloys was studied in a wide range of temperature and magnetic field. A step-like change of resistivity ($\Delta\rho \approx 24$ $\mu\Omega$cm) was detected in the off-stoichiometric $Ni_{1.85}Mn_{1.21}Ga_{0.94}$ alloy at temperature of martensitic structural transition. This $\Delta\rho$ is much more significant than one in the stoichiometric Ni_2MnGa alloy. In the case of the off-stoichiometric $(NiCo)_2Mn(GaIn)$ alloys, an enormous change of resistivity, $\Delta\rho \approx 200$ $\mu\Omega$cm, accompanies the structural transition. Simultaneously, the maximum of the spin disordered resistivity $\rho_{sd}(T)$ of austenite phase of the alloys is slightly dependent on composition of the alloy and vary from ≈ 30 $\mu\Omega$cm up to ≈ 45 $\mu\Omega$cm, in good agreement with theoretical calculations. Due to high sensitivity of the structural transition temperature of the alloys to magnetic field, the very pronounced magnetoresistance effects have been observed in the studied alloys.

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1. Introduction

The ferromagnetic Heusler alloys have drawn attention for a long time due to their pronounced multifunctional properties [1]. The shape-memory effect as well as the magneto- and barocaloric effects [2, 3] and magnetotransport properties [4] of the Ni_2MnGa-based alloys originate from a structural martensitic transition from low-temperature (low structural symmetry) martensite (M) to high-temperature (cubic) austenite (A). To optimize properties of the alloys from the point of view of their potential applications, many types of substitutions and off-stoichiometry were tested. An excess of Mn at the expense of Ga leads to a strong decrease of magnetization of martensite and the appearance of a so-called "paramagnetic gap" in a temperature range below the martensitic M–A transition. Magnetization obeys the Curie–Weiss law with reasonable values of effective paramagnetic moments (≈ 5.2 μ_B/f.u.), but the Curie temperature of martensite T_C^M (≈ 200 K) is almost insensitive to composition of the Mn-rich alloys and to external pressure or magnetic field [3]. An arrangement of magnetic moments below T_C^M has not been unambiguously described up to now. On the other hand, antiferromagnetic interaction between moments of Mn-atoms that are placed at regular Mn-sites and Ga-sites of the crystal lattice is accepted generally [5].

We have selected three Mn-rich Ni_2MnGa-based alloys to study a relation between their magnetotransport and magnetic properties. The temperature dependence of magnetization of the selected alloys, measured in magnetic field of 1 T, is presented in Fig. 1 in relative units with respect to saturated magnetization of the stoichiometric Ni_2MnGa alloy (3.62 μ_B/f.u. at 5 K). As a consequence of a high sensitivity of magnetization of the martensite phase to composition of the alloys, an increasing content of Mn-, Co- and In-atoms leads to a significant increase of magnetization at the transition from martensite to austenite, see Fig. 1.

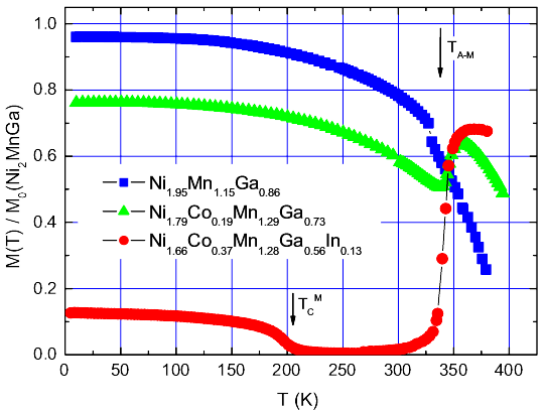

Fig. 1. Temperature dependence of magnetization of the selected alloys with respect to $M(5\ \mathrm{K})$ of the Ni_2MnGa alloy.

Temperature dependence of total electrical resistivity of ferromagnetic metals can be described by three terms [6]:

$$\rho(T) = \rho_0 + \rho_{ph} + \rho_{sd}, \tag{1}$$

where $\rho_{ph} \sim AT$ and $\rho_{sd} \sim BT^2$. The terms correspond to residual resistivity at 0 K (ρ_0), resistivity due to the electron–phonon scattering (ρ_{ph}), and magnetic part of resistivity due to spin-disorder scattering (ρ_{sd}), respectively. The last one reaches the maximum at the Curie temperature T_C and remains constant in the paramagnetic state. Resistivity of the stoichiometric T_2MnX alloys (T = transition metal, X = *sp* metal) was reliably fitted by the relation (1) [6] and recently the spin-disorder resistivity of the alloys was calculated from first prin-

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ciples (based on the disordered-local-moment approach) in agreement with the experimental results [7]. In the stoichiometric Ni_2MnGa alloys only a small change of resistivity was observed at temperature of $M-A$ transition, T_{M-A} [4]. But a pronounced change of electric resistivity of the off-stoichiometric and doped alloys was observed in the temperature range around T_{M-A} in the $Ni_2(MnCr)Ga$ alloys [8]. In this paper we describe and discuss a correlation of the resistivity changes with the magnetic behavior of the alloys.

2. Experimental details

Polycrystalline samples of the $Ni_{1.95}Mn_{1.15}Ga_{0.86}$, $Ni_{1.79}Co_{0.19}Mn_{1.29}Ga_{0.73}$ and $Ni_{1.66}Co_{0.37}Mn_{1.28}Ga_{0.56}In_{0.13}$ alloys were prepared by arc melting and described in details elsewhere [3]. A SQUID magnetometer (Quantum Design Co.) and a superconducting magnet up to 4.5 T with a LN_2 cryostat were used for the magnetization and electrical resistivity measurements, respectively. The four point AC resistivity measurement ($f = 311.1$ Hz, $I = 1$ mA) was performed by using a SR830 Lock-in amplifier.

3. Results

In contrast to the stoichiometric alloys, the Mn-rich $Ni_{1.95}Mn_{1.15}Ga_{0.86}$ alloy exhibits significant decrease of $\rho(T)$ at T_{M-A} (24 $\mu\Omega$cm) mainly due to a different temperature dependence of resistivity in martensite and austenite phases, see Fig. 2, and a high value of $\rho_0^M = 60$ $\mu\Omega$cm of martensite compared with $\rho_0^A = 42$ $\mu\Omega$cm of austenite. The linear part of $\rho(T)$ of austenite was roughly fitted from the $\rho(T)$ curve above T_C^A with $A^A = 10.5 \times 10^{-2}$ $\mu\Omega$cm/K. The determined value of $\rho_{sd}^{\max}(T_C^A) = 32$ $\mu\Omega$cm (see Fig. 2) well agrees with theoretical data. A fit of the martensite part of $\rho(T)$ provided ρ_0^M (shown above) and constants $A^M = 10.2 \times 10^{-2}$ $\mu\Omega$cm/K and $B^M = 2.2 \times 10^{-4}$ $\mu\Omega$cm/K^2 that can be well compared with data in literature and with the constant B^A (2.3×10^{-4} $\mu\Omega$cm/K^2) of austenite phase that was determined by a simple relation $\rho_{sd}^{\max}(T_C^A) = B^A(T_C^A)^2$. In the case of this alloy, a decrease of $\Delta\rho$ at T_{M-A} is accompanied by a small decrease of magnetization, see Fig. 1.

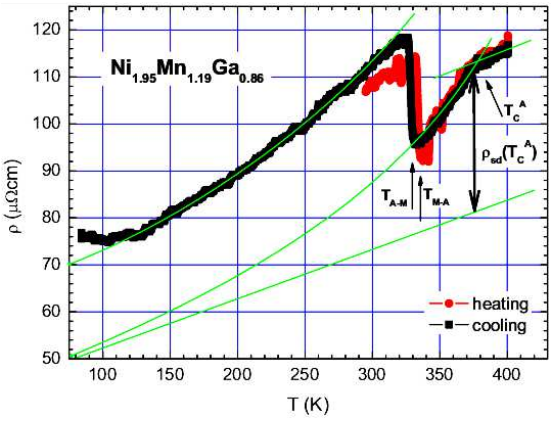

Fig. 2. Temperature dependence of resistivity of the Mn-rich alloy with fits (lines) by linear and quadratic polynomials.

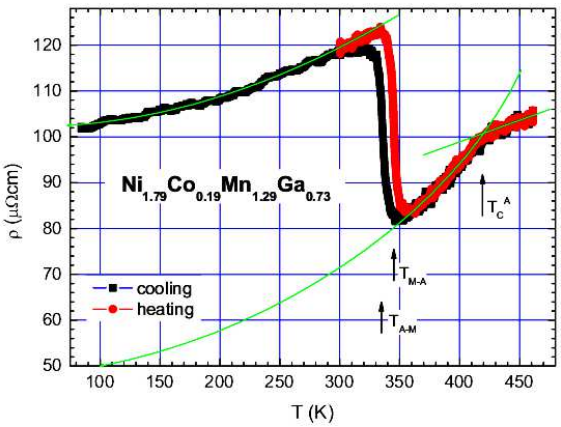

Fig. 3. Temperature dependence of resistivity of Co-doped alloy with fits (lines) by linear and quadratic polynomials.

The decrease of resistivity at T_{M-A} is much more visible in the Mn-rich Co-doped alloy where $\Delta\rho(T_{M-A}) = 40$ $\mu\Omega$cm while magnetization of the alloy increases, see Fig. 1. Figure 3 shows a different behavior of $\rho(T)$ at martensitic phases. In austenite, $A^A = 8.4 \times 10^{-2}$ $\mu\Omega$cm/K was estimated by a linear fit of $\rho(T)$ at $T > T_C^A$ and $B^A = 2.4 \times 10^{-4}$ $\mu\Omega$cm/K^2 is similar to one of the Co-free alloy. While in martensite, the high value of $\rho_0^M = 101$ $\mu\Omega$cm, a very low, moreover negative, value of $A^M = -8.4 \times 10^{-3}$ $\mu\Omega$cm/K and $B^M = 2.2 \times 10^{-4}$ $\mu\Omega$cm/K^2 were estimated by a fit according to relation (1).

The extreme decrease of resistivity $\rho(T)$ at temperature T_{M-A}, $\Delta\rho(T_{M-A}) = 170$ $\mu\Omega$cm, in the Co- and In-doped alloy is accompanied by high increase of magnetization on heating, see Fig. 4 and Fig. 1. However, no significant change of $\rho(T)$ was observed at temperature T_C^M where magnetization of the alloy exhibits a clear decrease into "paramagnetic gap", see Fig. 1. The tan-

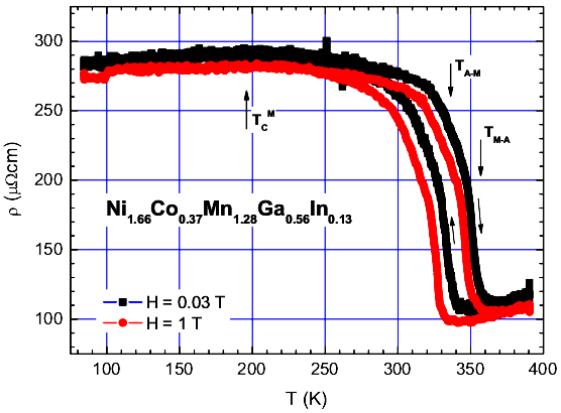

Fig. 4. Temperature dependence of resistivity of the Mn-rich, Co- and In-doped alloy at different magnetic field.

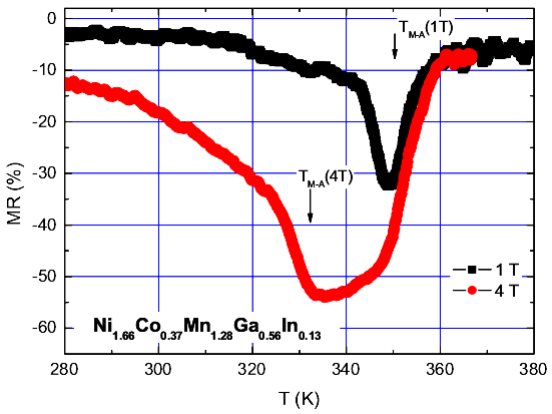

Fig. 5. Temperature dependence of MR effect in the Mn-rich Co- and In-doped alloy at field of 1 T and 4 T.

gent of $\rho(T)$ changes only its sign at T_C^M and resistivity of martensite decreases at "paramagnetic gap" with increase of temperature. It is clear that $\rho(T)$ cannot be fitted by the simple relation (1) in this case.

The structural transition of the Co- and In-doped alloy is strongly sensitive to external magnetic field, in the case of the studied alloy $\mathrm{d}T_{A-M}/\mathrm{d}H = -7.4$ K/T. In consequence of this shift of T_{M-A} or T_{A-M} with field, very pronounced magnetoresistance (MR) effect takes place in a wide temperature range across T_{M-A}. Figure 5 presents temperature dependence of MR at fields of 1 T and 4 T derived from $\rho(T)|_H$ curves and verified by $\rho(H)|_T$ measurements.

4. Discussion and conclusions

The measurement of resistivity clearly confirms the very weak dependence of the electric (as well as magnetic) properties to the composition of the austenite phase of alloys with the cubic crystal structure. Temperature dependence of resistivity of austenite obeys relation (1) satisfactorily albeit this could be verified in a narrow temperature range. The constants A and B do not reflect any significant dependence of electron–phonon or spin-disorder scattering in the cubic phase on composition of the alloys and the spin-disordered resistivity $\rho_{sd}^{\max}(T_C^A)$ increases proportionally to an increase of $(T_C^A)^2$ only.

Martensitic transition leads to a lower symmetry of crystal structure of the alloys (tetragonal, orthorhombic, monoclinic), but the outstanding changes in $\rho(T)$ of martensite cannot be ascribed exclusively to the decrease of crystal symmetry. One of the arguments supporting this statement can be $\rho(T)$ of the stoichiometric Ni_2MnGa alloy that shows just a very small change of resistivity while crystal symmetry is changing from cubic to orthorhombic.

The dominant feature of the resistivity of martensite is the increase of ρ_0^M with increasing content of Mn, Co and In in the alloys. However, ρ_0^M as determined by a fit according to relation (1) cannot be considered to be "residual resistivity" due to its strong composition, magnetic field and temperature dependence. Assuming one residual resistivity for both the phases, ρ_0^M could be more precisely described as $\rho_0^M(T,H) \approx \rho_0^A + \rho^M(T,H)$, where the last term overcomes the spin-disordered scattering in the Mn-rich and doped alloys. It has also to be considered that in the case of the Mn-rich Co-free alloy the constant A is almost identical in both phases.

A very similar problem of a significant resistivity difference has been solved in the case of two magnetic phases (without a crystal lattice change) of the UNiGa intermetallics [9]. The most efficient model of this phenomenon was based on a change in the electronic structure that is induced by a presence of antiferromagnetic stacking of ferromagnetic layers in UNiGa at low temperatures. In contrast to ferromagnetic state, the changed electronic structure restricts a participation of a part of d-electrons on conductivity at low temperatures [10].

The weak sensitivity of the scattering mechanisms to a composition of the alloys in the austenite phase suggests that the huge changes of $\rho(T)$ observed in martensite cannot be ascribed explicitly to compositional variations. The decrease of resistivity of martensite at temperatures above T_C^M and its strong dependence on magnetic field in the Mn-rich Co- and In-doped alloy point to a scenario with a temperature or field excitation of electrons over a restrictive gap in a complex electronic structure of martensite phase of the Mn-rich Co- and In-doped Ni_2MnGa alloy. The electronic structure of the austenite phase of the alloy with the cubic crystal structure is probably so rigid that such scenario cannot play a role.

Acknowledgments

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