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Influence of intermartensitic transitions on transport properties of $Ni_{2.16}Mn_{0.84}Ga$ alloy

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Abstract

Magnetic, transport and x-ray diffraction measurements of the ferromagnetic shape memory alloy $Ni_{2.16}Mn_{0.84}Ga$ have revealed that this alloy undergoes an intermartensitic transition upon cooling, whereas no such a transition is observed upon subsequent heating. The difference in the modulation of the martensite forming upon cooling from the high-temperature austenitic state (five-layered (5M) martensite), and the martensite forming upon the intermartensitic transition (seven-layered (7M) martensite) strongly affects the magnetic and transport properties of the alloy and results in a large thermal hysteresis of the resistivity ρ and magnetization M. The intermartensitic transition has an especially marked influence on the transport properties, as is evident from a large difference in the resistivity of the 5M and 7M martensites, $(\rho_{5M} - \rho_{7M})/\rho_{5M} \approx 15\%$, which is larger than the jump of resistivity at the martensitic transition from the cubic austenitic phase to the monoclinic 5M martensitic phase. We assume that this significant difference in ρ between the martensitic phases is accounted for by nesting features of the Fermi surface. It is also suggested that the nesting hypothesis can explain the uncommon behaviour of the resistivity at the martensitic transition, observed in stoichiometric and near-stoichiometric Ni–Mn–Ga alloys.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Intermetallic compounds undergoing thermoelastic martensitic transformation when in the ferromagnetic state (ferromagnetic shape memory alloys) have attracted considerable interest

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(see, for a recent review, [1]). This is due to the fact that they exhibit large magnetic-field-induced strains which can be obtained either by re-orientation of martensitic variants [2, 3] or by shifting the martensitic transition temperature [4, 5]. In addition to this effect of practical significance, ferromagnetic shape memory alloys have been the subject of numerous studies due to their rich phase diagrams. In particular, some of these alloys exhibit several phase transitions between different crystallographic modifications of martensite, induced by a change of composition, temperature or stress, or by the combination of these parameters.

A prototype of the ferromagnetic shape memory alloys, Ni_2MnGa, is a representative of Mn-containing Heusler alloys. It orders ferromagnetically at Curie temperature $T_C = 376$ K. Upon cooling down to $T_m = 202$ K it undergoes a reversible thermoelastic martensitic transformation from the Heusler ($L2_1$) cubic structure to a roughly tetragonal crystal structure. Both T_m and T_C are sensitive to stoichiometry. For instance, a partial substitution of Mn for Ni in $Ni_{2+x}Mn_{1-x}Ga$ alloys results in an increase of T_m and a decrease of T_C until they couple in a composition range $x = 0.18$–0.20 [6].

An early neutron diffraction study [7] of the martensitic structure of stoichiometric Ni_2MnGa showed that, together with strong tetragonal reflections, there were several additional peaks on the diffraction pattern. Based on this observation, the authors suggested that the martensitic phase has a modulated crystal structure. Further studies revealed [8][6] that modulation and, therefore, the crystal structure of the martensite forming from the parent austenitic phase, depend on composition (see [9] and references therein). By now, five- and seven-layered martensitic phases modulated along the $(110)[1\overline{1}0]$ system and a non-modulated martensitic phase have been established to exist in Ni–Mn–Ga alloys. In addition, the observation of longer-period modulations of the martensite has been reported [10].

The crystal structure of martensite was found to be very unstable to the application of external stresses [11–14]. It turned out that the sequence of stress-induced martensite–martensite transformations depends on many factors, such as the composition of the sample, temperature of the test, and the crystallographic direction along which the stress was applied. Besides composition- or stress-induced changes in the crystal structure of martensite, some off-stoichiometric Ni–Mn–Ga alloys undergo a sequence of temperature-induced martensite–martensite phase transitions. Apart from Ni–Ti (see [15] and references therein) and $Ni_{50}Mn_{50-x}Al_x$ [16] systems, temperature-induced intermartensitic transitions have not been observed in other shape memory alloys.

In Ni–Mn–Ga intermartensitic transitions are, as evident from calorimetric measurements [17], first-order phase transitions. As compared with the martensitic transformation, the intermartensitic transitions exhibit several distinctive features. They are a large, exceeding 100 K, temperature hysteresis and a considerable difference exists in transport properties between the martensitic phases involved in an intermartensitic transition [18–25]. Transport measurements of Ni–Mn–Ga alloys undergoing intermartensitic transitions [6, 22–24] have indicated that the difference in the resistivity between martensitic phases is comparable or even larger than that observed at the martensitic transformation temperature. This seems to be unusual because martensitic transformation has a stronger influence on the physical characteristics (crystal structure, Fermi surface, magnetic properties) of the materials.

[6] It is necessary to stress that the crystal structure of the martensitic phase in Ni–Mn–Ga is still the subject of controversy. For instance, although the crystal structure of martensite in the stoichiometric Ni_2MnGa has been described as tetragonal for a long time, recent high resolution neutron diffraction measurements [8] imply that the martensitic phase has actually an orthorhombic crystal structure. In off-stoichiometric Ni–Mn–Ga alloys this situation is even more complicated, which is caused by the coexistence of different martensitic phases [8] or the influence of room-temperature ageing on diffraction patterns [8].

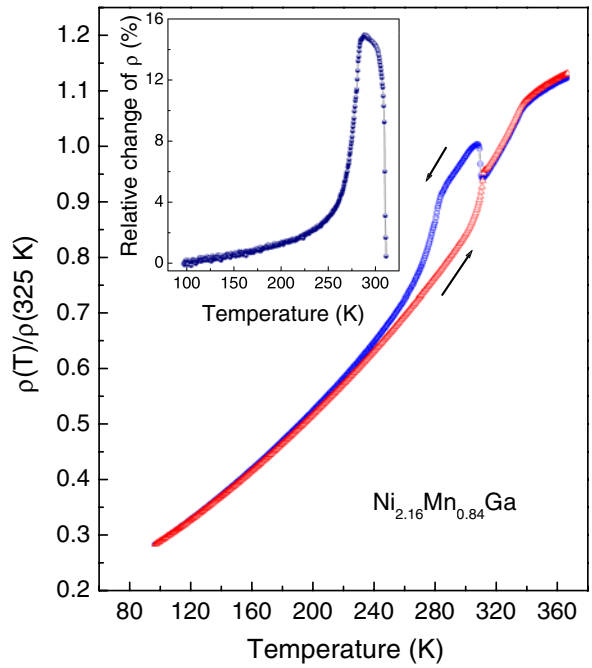

Figure 1. Temperature dependencies of electrical resistivity for $Ni_{2.16}Mn_{0.84}Ga$ measured during cooling and heating. The inset shows the difference between ρ measured upon cooling and heating, $(\rho_{\text{cooling}} - \rho_{\text{heating}})/\rho_{\text{cooling}}$.

Since these features of intermartensitic transitions have not been discussed earlier, we have studied and analysed the transport properties of $Ni_{2.16}Mn_{0.84}Ga$ undergoing an intermartensitic transition. In our study we have also used a stoichiometric Ni_2MnGa sample, prepared by the same method as $Ni_{2.16}Mn_{0.84}Ga$.

2. Experimental details

A polycrystalline ingot of $Ni_{2.16}Mn_{0.84}Ga$ composition was prepared by arc melting high purity constituent elements in argon atmosphere. In order to get a good compositional homogeneity, the ingot was re-melted several times and annealed at 1050 K for nine days with subsequent quenching in ice water. Samples for resistivity and magnetization measurements were cut from the middle part of the ingot. Temperature dependencies of resistivity and magnetization were measured with a heating/cooling rate of 1 K min^{-1} by a standard four-probe technique and by a vibrating sample magnetometer, respectively. The crystal structure of the alloy was examined using a Philips X-Pert system in a wide temperature interval. For the powder x-ray diffraction measurements, part of the ingot was crushed into a fine powder. The powder was sealed in an evacuated quartz tube and annealed at 1050 K for five days in order to remove residual stress and improve the peak shape of diffraction patterns.

3. Experimental results

The temperature dependencies of electrical resistivity of $Ni_{2.16}Mn_{0.84}Ga$, measured upon cooling and heating, are shown in figure 1. Cooling from high temperatures results in the formation of a long-range ferromagnetic ordering at $T_C = 337$ K which is accompanied by a

change in the slope of the resistivity curve due to the decrease in electron–magnon scattering. The jump-like increase of the resistivity at $T_m \approx 309$ K corresponds to the transition from the high-temperature austenitic to a low-temperature martensitic phase.

Besides the change in the slope of the curve at $T_C = 337$ K and the jump-like increase of ρ at $T_m = 309$ K, one more marked change in the slope of the cooling curve is observed at $T_I = 283$ K. Since this anomaly is observed when the sample is in the martensitic state, this means that a martensite–martensite transformation occurs in $Ni_{2.16}Mn_{0.84}Ga$. Based on the results of transmission electron microscopy (TEM) observation of a sample of this composition [26], which revealed that the crystal structure of martensite in $Ni_{2.16}Mn_{0.84}Ga$ is characterized by a five-layered modulation (5M) at room temperature and seven-layered modulation (7M) at $T = 173$ K, we conclude that the anomaly of ρ at $T_I = 283$ K corresponds to the onset of intermartensitic transition from a five- to a seven-layered martensite (5M $\rightarrow$ 7M). Cooling the sample below T_I initially results in a distinct decrease of the resistivity, which becomes less temperature-dependent on further cooling. No anomaly corresponding to the end of the 5M $\rightarrow$ 7M intermartensitic transition was observed on the resistivity curve in the temperature interval of the measurements. The absence of such an anomaly implies that the fraction of the five-layered martensite gradually decreases with decreasing temperature and therefore both the 5M and 7M martensitic phases coexist over a wide temperature interval. Subsequent heating revealed a monotonous increase of the resistivity up to the reverse martensitic transformation temperature.

Since the 5M $\rightarrow$ 7M intermartensitic transition is not completed in the studied temperature interval and because of the absence of the reverse 7M $\rightarrow$ 5M intermartensitic transition upon subsequent heating, the resistivity exhibits very large thermal hysteresis. At temperatures below the martensitic transformation, the heating curve deviates from the curve measured upon cooling, and the difference between ρ measured upon cooling and heating progressively increases as the temperature is increased (the inset in figure 1). Assuming for simplicity that at $T = 100$ K there exists only a tiny fraction of the 5M martensite, we can estimate the difference in the resistivity between the 5M and 7M phases, $\Delta\rho = (\rho_{5M} - \rho_{7M})/\rho_{5M}$. As is seen from the inset in figure 1, $\Delta\rho \approx 15\%$ in a temperature interval from 283 to 300 K.

Due to the absence of the reverse intermartensitic transition upon heating, the behaviour of ρ at the martensitic transformation measured upon cooling and heating is quite different. Whereas ρ shows a jump-like increase during direct martensitic transformation from the parent phase to the 5M martensite, the resistivity steepens when approaching the reverse martensitic transformation from the 7M martensite to the parent phase (figure 1). If the anomaly of ρ at $T_I = 283$ K indeed corresponds to the onset of the intermartensitic transition, below which the fraction of the 5M martensite gradually decreases, the behaviour of ρ at martensitic transformation temperature T_m should depend on the proportion of the 5M and 7M phases. In order to check this, we measured several partial cooling–heating cycles.

The results of these measurements (figure 2) indicate that the behaviour of ρ at T_m upon cooling is always the same (a jump-like increase), but that measured upon heating substantially depends on the temperature down to which the sample was cooled. If the sample is cooled down to $T > T_I$, the resistivity upon subsequent warming follows the cooling path and ρ exhibits a marked jump-like decrease during transformation to the austenitic phase (figure 2(a)). This means that cooling of the sample to $T = 286$ K, which is slightly higher than the T_I temperature, did not result in the formation of a two-phase state and the sample remains in the 5M martensitic state upon subsequent heating.

When the sample is cooled somewhat below T_I, the behaviour of resistivity at T_m upon heating is still similar to that observed upon cooling. This is evident from figure 2(b), where the sample was cooled to $T = 282$ K, i.e. 1 K below the T_I temperature. This means that in

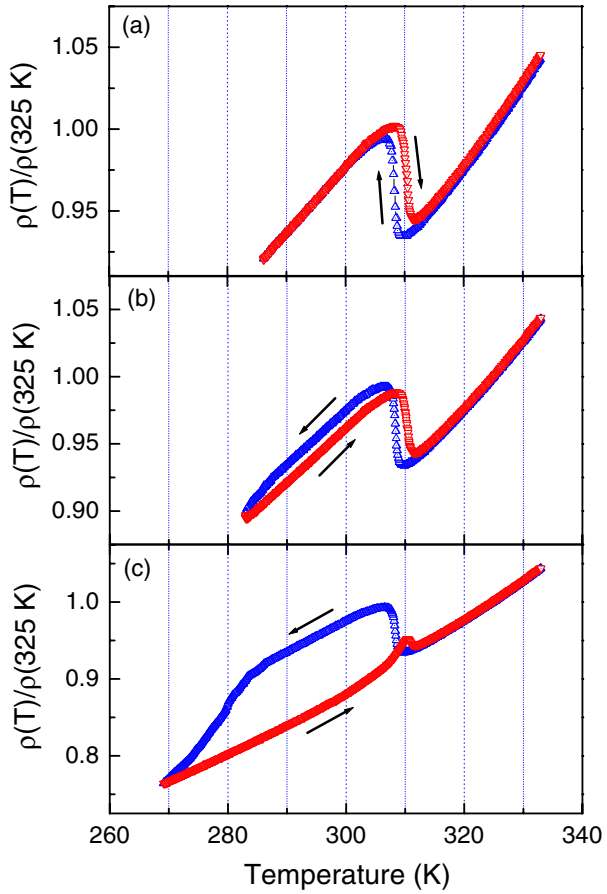

Figure 2. Electrical resistivity of $Ni_{2.16}Mn_{0.84}Ga$ measured upon partial cooling–heating cycles in temperature intervals (a) 286–333 K, (b) 282–333 K and (c) 269–333 K.

the two-phase state of the sample, attained by cooling slightly below T_I, the behaviour of ρ upon heating is governed by a considerably larger fraction of the 5M martensite. Note that in this case the heating curve is parallel to the cooling curve, indicating that the two-phase state existing at $T = 282$ K is preserved up to the reverse martensitic transformation. In other words, the fraction of the 7M martensite does not transform to the 5M martensite upon heating from 282 K. This observation implies that the onset of the reverse intermartensitic transition is above the martensitic transformation temperature T_m.

Finally, when the sample is cooled down to $T = 269$ K, the resistivity upon subsequent heating exhibits behaviour typical of the 7M martensitic phase (figure 2(c)), and ρ shows a small kink at the martensitic transformation temperature. In a temperature interval from 283 to 309 K, the difference in ρ between heating and cooling curves is $\sim$12%, indicating that approximately 80% of the 7M martensite had been formed upon cooling to 269 K. Based on the results presented in figure 2 one can conclude that the 7M martensite appears upon cooling below $T_I = 283$ K and the fraction of this martensitic phase considerably exceeds that of the 5M martensitic phase at $T < 270$ K.

The magnetization M of $Ni_{2.16}Mn_{0.84}Ga$ measured in a 0.1 T magnetic field is shown in figure 3. The Curie temperature, determined from this measurement, is equal to 340 K

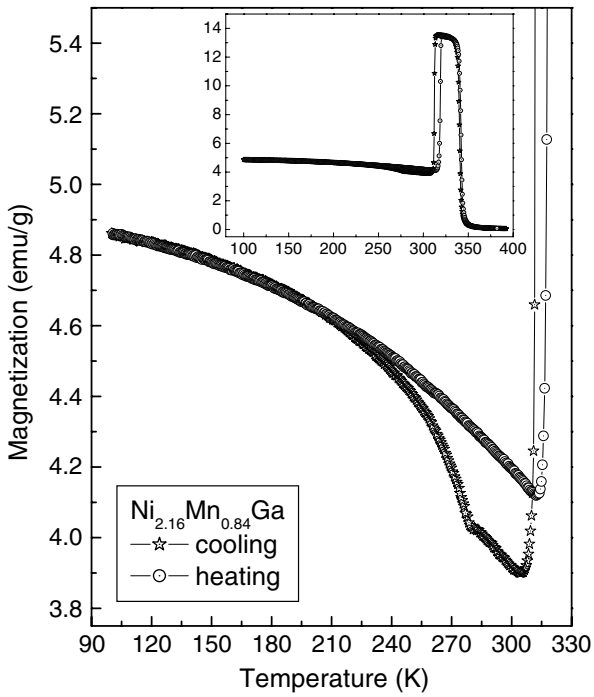

Figure 3. Temperature dependencies of the magnetization of $Ni_{2.16}Mn_{0.84}Ga$ measured in a 0.1 T magnetic field. The inset shows $M(T)$ over the entire temperature interval.

(inset in figure 3). The anomaly at ≈310 K, exhibiting a temperature hysteresis of ∼6 K, corresponds to the martensitic transformation. Like the resistivity, the magnetization of the sample shows a large thermal hysteresis in the martensitic state. A well-defined change in the slope of the $M(T)$ curve measured upon cooling at $T = 279$ K corresponds to the onset of the intermartensitic transition to the 7M phase. This characteristic temperature, determined from the magnetization measurements, is slightly lower than that obtained from the resistivity data. This difference can be accounted for by the fact that $M(T)$ and $\rho(T)$ measurements were performed on different samples. As is seen from figure 3, in the 0.1 T magnetic field magnetization of the 5M martensitic phase is lower than that of the 7M phase and the difference between them gradually diminishes as the temperature is lowered.

The thermal hysteresis of M is observed only in low magnetic fields. Measurements of $M(T)$ in a magnetic field of 1 T showed no thermal hysteresis of M in the martensitic state, which means that both the martensitic phases have the same magnetization in this field. Therefore, it can be concluded that magnetization saturation of these two martensitic phases are the same.

The diffraction pattern of $Ni_{2.16}Mn_{0.84}$, taken at room temperature, is shown in figure 4. To be sure that the measured diffraction pattern corresponds to the 5M martensite, the powder was heated above the martensitic transformation temperature T_m and the measurement was performed on the powder cooled to room temperature from the austenitic state. Preliminary analysis of the room temperature diffraction pattern of $Ni_{2.16}Mn_{0.84}Ga$ showed that the crystal structure of the martensite formed upon cooling from the austenitic phase can be interpreted as a monoclinic one with lattice parameters $a = 0.42$ nm, $b = 0.55$ nm, $c = 2.10$ nm and $\beta = 92°$.

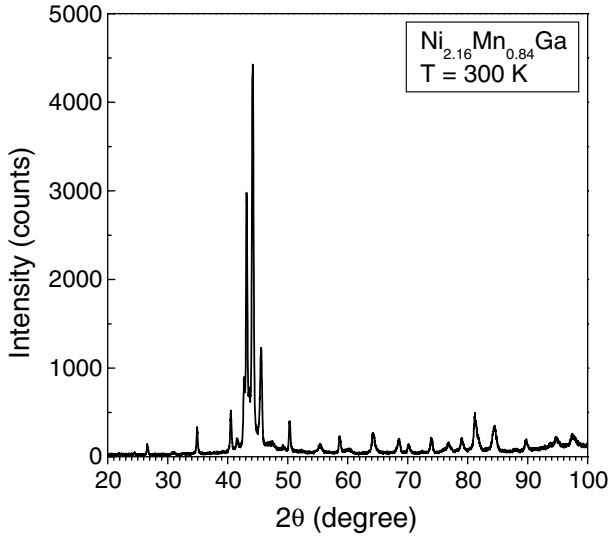

Figure 4. X-ray diffraction pattern of the 5M martensitic phase of $Ni_{2.16}Mn_{0.84}Ga$ forming upon cooling from the high-temperature austenitic phase.

X-ray diffraction measurement at a lower temperature, $T = 77$ K, confirmed the occurrence of the intermartensitic transition, seen on the $\rho(T)$ and $M(T)$ curves. The crystal structure of the 7M martensitic phase was interpreted as monoclinic with lattice parameters $a = 0.426$ nm, $b = 0.543$ nm, $c = 2.954$ nm and $\beta = 94.3°$. Further cooling down to $T = 10$ K did not result in a change of the diffraction pattern observed at $T = 77$ K. The results of these measurements are shown on an enlarged scale in figure 5.

4. Discussion

The results of our resistivity measurements (figure 1) indicate that different martensitic phases are considerably distinguished by their transport properties, namely ρ_{5M} is larger than ρ_{7M} by 15%. Generally, this difference can be caused by two factors: by changes in the scattering probability and/or by changes in the electronic structure. Since both the phases exhibit similar plate-like morphology [26] we suggest that the 15% difference in the resistivity of these phases cannot be accounted for by an increase in the scattering centres. Therefore, the origin of this difference has to be looked for in the Fermi surface features. Indeed, it is generally acknowledged that the formation of a long-range ordering observed in a large number of compounds is associated with the nesting properties of the Fermi surface. This is as true for the case of long-range structural ordering [27, 28] as for the case of long-range magnetic ordering, such as spin- or charge-density waves [29–32]. The periodicity of long-range ordering is determined by the nesting vector on the Fermi surface.

It is conceivable that the various martensitic phases forming in Ni–Mn–Ga alloys are driven by the geometry of the Fermi surface that has a nesting vector corresponding to the modulation of martensite, as was suggested in [33]. This suggestion implies that martensitic phases with different nesting vectors have different fractions of nested Fermi surface. On the other hand, it is well known that the nesting considerably affects the transport properties of a metal due to the condensation of electrons in the nesting parts of the Fermi surface. Therefore, change in the modulation can affect the number of conduction electrons n_{eff} due to the change of the Fermi surface available for conduction.

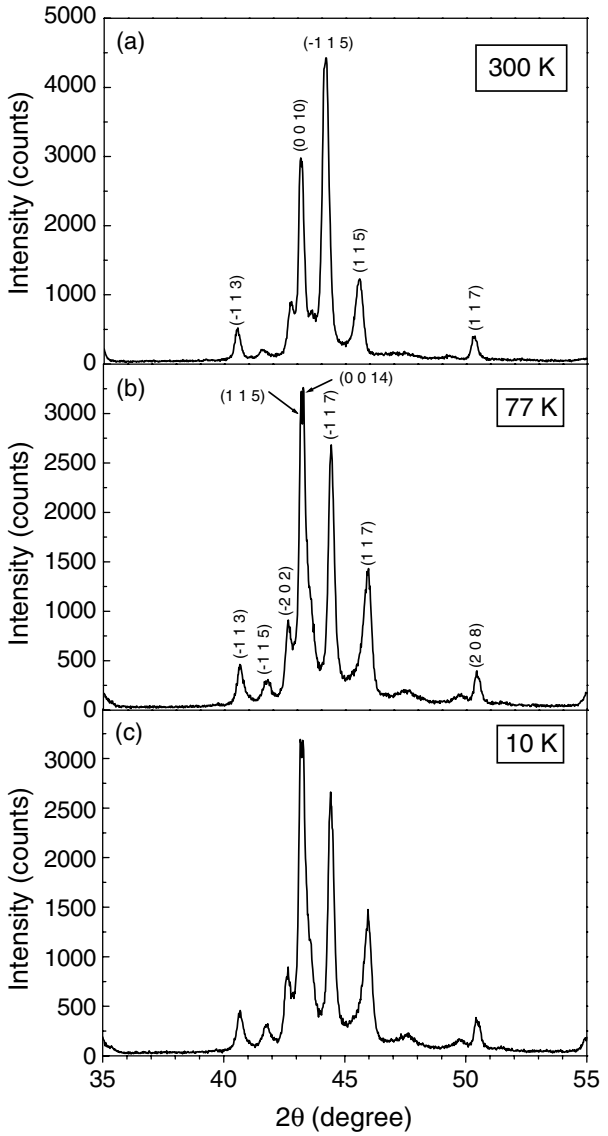

Figure 5. Results of x-ray diffraction measurements of $Ni_{2.16}Mn_{0.84}Ga$ performed at different temperatures upon cooling. The diffraction patterns were collected at (a) 300 K, (b) 77 K and (c) 10 K.

In the simple relaxation time approximation

$$\rho = m^*/n_{\text{eff}}e^2\tau,$$

where m^* is the effective mass, e is the electronic charge and τ is the relaxation time. Assuming that the relaxation time τ is the same for both 5M and 7M martensitic phases, the experimental observation that $\rho_{5\text{M}} > \rho_{7\text{M}}$ indicates that the 5M phase has a fewer number of conduction electrons n_{eff} than the 7M phase. Note that if the proposed explanation is valid, one can expect to observe an anisotropic behaviour of ρ in a Ni–Mn–Ga single crystalline sample, as is the case in Cr [29] and heavy-fermion compounds [34–36].

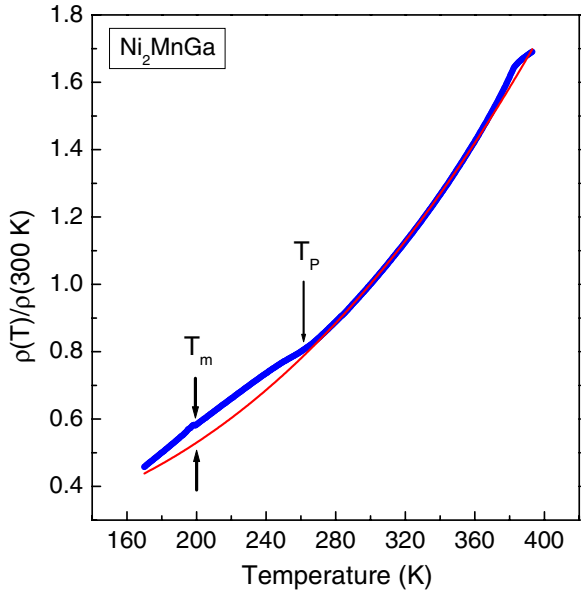

Figure 6. Temperature dependence of electrical resistivity for the stoichiometric Ni_2MnGa. The solid curve is a fit to the experimental curve.

In the above discussion we did not consider the possibility that electron–magnon scattering can be different in the 5M and 7M martensites. However, according to Friedel and de Genner [37], the temperature dependence of the magnetic resistivity ρ_{mag} can be described as

$$\rho_{mag} = \rho_{s\text{-}d}[1 - \sigma^2(T)],$$

where $\rho_{s\text{-}d}$ is the temperature independent spin-disorder resistivity and $\sigma^2(T) = M_s(T)/M_s(0)$, $M_s(T)$ and $M_s(0)$ are magnetization saturations at a finite temperature T and at $T = 0$ K. Since our magnetic measurements have shown that the magnetization saturation M_s of the 5M and 7M phases is the same, it can be concluded that both the phases are characterized by the same electron–magnon scattering.

It is apparent that, together with the unusual transport properties of $Ni_{2.16}Mn_{0.84}Ga$ discussed above, the proposed nesting hypothesis can satisfactory explain the uncommon behaviour of ρ at martensitic transformation temperature T_m, observed in stoichiometric and near-stoichiometric Ni_2MnGa alloys. Indeed, since the martensitic transformation results in a drastic change of crystal structure, Fermi surface, mean free path and so on, one can expect to detect a well-defined anomaly at T_m, which is indeed generally observed in shape memory alloys [38]. In contrast to this, $\rho(T)$ measurements for stoichiometric and near-stoichiometric Ni_2MnGa revealed only a change in the slope of the curve at the martensitic transformation temperature [6, 39–41].

We argue that such a peculiar behaviour of ρ at T_m in Ni_2MnGa is due to the premartensitic transition, occurring above T_m, despite the fact that the cubic symmetry of the crystal structure is preserved upon this transition [42]. As is shown in figure 6, for the austenitic phase of stoichiometric Ni_2MnGa, $\rho(T)$ can be fitted by a T^n dependence ($n \approx 3$). The experimental curve deviates from the fit at Curie temperature $T_C = 376$ K and at $T = 266$ K which matches well the premartensitic transition temperature $T_P = 260$ K for the stoichiometric composition [42]. The driving force for the premartensitic transition is believed to be Fermi

surface nesting, as was suggested by Zheludev *et al* [42], and recent theoretical calculation [43] supports this hypothesis. The deviation of the resistivity from the T^n dependence below T_P could be caused, for instance, by an increase in the relaxation time τ due to the modulation of the cubic structure in the premartensitic phase. In our opinion, however, the primary role in this process is played by the condensation of the conduction electrons in the nesting part of the Fermi surface. Assuming that without the premartensitic transition $\rho(T)$ would follow the T^n dependence down to the martensitic transformation temperature $T_m = 202$ K, it can be concluded from figure 6 that in this case the difference in ρ between the high temperature austenitic and low temperature martensitic phases is significant and the resistivity should exhibit a jump-like behaviour, as in other shape memory alloys or in off-stoichiometric Ni–Mn–Ga.

With deviation from the stoichiometry, the martensitic transition temperature increases or decreases, depending on substitution, whereas the T_P temperature is less composition dependent [44–46]. In the case of increasing T_m this leads to the disappearance of the premartensitic transition in a critical composition and, as a result, in off-stoichiometric Ni–Mn–Ga alloys a marked jump-like behaviour of ρ is observed.

5. Conclusion

Temperature-induced intermartensitic transitions observed in certain Ni–Mn–Ga alloys give rise to an anomalously large thermal hysteresis of magnetic and transport properties, which is not observed in other compounds. This thermal hysteresis is accounted for by the coexistence of both martensitic phases in a wide temperature interval. As is evident from the resistivity measurements of $Ni_{2.16}Mn_{0.84}Ga$, the difference in ρ between 5M and 7M martensite is about 15%, which is even larger than that observed upon the martensitic transformation. We have suggested that such a significant difference is accounted for by the geometry of the Fermi surface that has a different nesting vector in 5M and 7M martensitic phases. If this assumption is valid, an anisotropic behaviour of ρ in a Ni–Mn–Ga single crystal of the same or similar composition can be reasonably expected. Therefore, further studies of single crystalline samples are required for a better understanding of the structural instability of various martensitic phases in Ni–Mn–Ga alloys.

In the framework of the nesting hypothesis we have also discussed the peculiar behaviour of ρ at the martensitic transformation temperature T_m in stoichiometric Ni_2MnGa. We have argued that this behaviour of ρ is caused by the condensation of conduction electrons in the nesting part of the Fermi surface occurring upon the premartensitic transition.

Acknowledgments

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