PHYSICAL REVIEW B **70**, 174413 (2004)

Magnetic properties and magnetostructural phase transitions in $Ni_{2+x}Mn_{1-x}Ga$ shape memory alloys

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(Received 6 January 2004; revised manuscript received 28 May 2004; published 11 November 2004)

A systematic study of magnetic properties of $Ni_{2+x}Mn_{1-x}Ga$ $(0 \leqslant x \leqslant 0.19)$ Heusler alloys undergoing structural martensite-austenite transformations while in the ferromagnetic state has been performed. From measurements of spontaneous magnetization, $M_s(T)$, jumps ΔM at structural phase transitions were determined. Virtual Curie temperatures of martensite were estimated from the comparison of magnetization in martensitic and austenitic phases. Both saturation magnetic moments in the ferromagnetic state and effective magnetic moments in paramagnetic state of Mn and Ni atoms were estimated and the influence of delocalization effects on magnetism in these alloys was discussed. The experimental results obtained show that the shift of martensitic transition temperature depends weakly on composition. The values of this shift are in good correspondence with the Clapeyron-Clausius formalism taking into account the experimental data on latent heat at martensite-austenite transformations.

DOI: 10.1103/PhysRevB.70.174413 PACS number(s): 75.30.Cr, 75.50.Cc, 64.70.Kb

I. INTRODUCTION

In the Ni_2MnGa Heusler alloy, a structural transformation from cubic austenitic to tetragonal martensitic phase is observed upon cooling. The interest in the study of Ni_2MnGa-based alloys has mainly been conditioned by the fact that the martensitic phase in these alloys is ferromagnetic. The combination of ferromagnetic ordering and martensitic transformation allows the realization of the magnetically driven shape memory effect, which expands the area of technical applications of this effect considerably.

Despite a large number of experimental and theoretical studies, many fundamental aspects of Ni_2MnGa-based alloys are not yet clearly understood. For instance, magnetic properties of the thoroughly studied $Ni_{2+x}Mn_{1-x}Ga$ system were not sufficiently clarified. For these alloys, the compositional dependencies of the Curie temperature T_C and martensite-austenite transformation temperature T_m were determined, but the temperature and compositional dependencies of magnetization have not been investigated in detail. In particular, no systematic study was performed on the jump of magnetization at the martensitic transition, which determines the shift of T_m under external magnetic field. In addition, the exchange interaction parameters have not been estimated for these alloys. All these factors are important for obtaining a better insight into physical mechanisms, underlying the magnetically driven shape memory effect. This paper deals with a systematic study of the magnetic properties of $Ni_{2+x}Mn_{1-x}Ga$ $(0 \leqslant x \leqslant 0.19)$ alloys.

II. CRYSTAL STRUCTURE AND MAGNETIC PROPERTIES OF THE $Ni_{2+x}Mn_{1-x}Ga$ SYSTEM

The high-temperature austenitic phase of $Ni_{2+x}Mn_{1-x}Ga$ Heusler alloys has a cubic structure of $Fm\bar{3}m$ space group. A structural transition to a modulated tetragonal $(c/a<1)$ phase is observed in these alloys on cooling. It is worth noting that the crystal structure and space group of the low-temperature phase is still a subject of controversy (see, for example, Refs. 1,2), which is aggravated by a compositional dependence of the crystal structure of martensite. Thus, for example, recent results of high-resolution neutron diffraction[3] give grounds for concluding that for the stoichiometric Ni_2MnGa composition the martensitic phase, which was considered for a long time as tetragonal, has indeed an orthorhombic symmetry of *Pnnm* space group. The structural martensitic transformation in Ni_2MnGa-based Heusler alloys was described as driven by a band Jahn-Teller effect.[4,5]

The martensitic transformation temperature T_m, which is about 200 K in stoichiometric Ni_2MnGa, linearly increases with x in $Ni_{2+x}Mn_{1-x}Ga$ alloys and reaches about 330 K at $x=0.18-0.19$ (Ref. 6). The alloys with a higher Ni content were not studied so far. Note that different values of T_m are given in the literature, indicating probably the sensitivity of physical properties of these alloys to structural disorder[7,8] and/or deviations from the nominal composition.

The $Ni_{2+x}Mn_{1-x}Ga$ alloys are ferromagnetic at low temperatures. The Curie temperature T_C is about 370 K in stoichiometric composition $(x=0)$. T_C approximately linearly decreases with increasing Ni content, so that for $x=0.18-0.19$ Curie temperature merges with the martensitic transformation temperature T_m. Hence, the alloys with $x=0.18-0.19$ experience a structural (martensitic) transition from paramagnetic austenite to ferromagnetic martensite. At the same time, the magnetic state of the alloys with a lower Ni content does not change during martensitic transformation and both austenitic and martensitic phases are ferromagnetic. The martensitic transformation, however, influences the magnetic parameters of these alloys and reveals itself in a sharp change of magnetic anisotropy and magnetization saturation.[9]

The neutron diffraction measurements of stoichiometric composition[4,10] show that the magnetic moment is localized mainly on Mn atoms. The reported values of the Mn magnetic moment range from $3.8\mu_B$ to $4.2\mu_B$. The magnetic moment of Ni atoms is considerably smaller, about $(0.2-0.4)\mu_B$. The concentration dependence of the magnetic moment in $\mathrm{Ni}_{2+x}\mathrm{Mn}_{1-x}\mathrm{Ga}$ alloys has not been reported. It is known only that magnetization saturation decreases with increasing x.[11,12]

III. SAMPLE PREPARATION AND MEASUREMENTS

Polycrystalline samples of $\mathrm{Ni}_{2+x}\mathrm{Mn}_{1-x}\mathrm{Ga}$ alloys were prepared by a conventional arc-melting method in an atmosphere of spectroscopically pure argon gas. The samples were homogenized at 1050 K for 9 days with subsequent quenching in ice water. For the measurements of physical properties those samples were used whose weight loss during arc-melting was less than 0.2%. The measurements of magnetic properties were performed on samples with $x=0$, 0.04, 0.08, 0.12, 0.16, and 0.19; some measurements were also done on the sample with $x=0.02$.

The magnetization up to 5 T was measured in the temperature range 5-700 K by a Quantum Design SQUID magnetometer; it was also measured by a vibrating sample magnetometer (VSM) in magnetic fields up to 1.8 T. Additionally, measurements in pulsed magnetic fields up to 10 T were performed. Spontaneous magnetization M_s at low temperatures was determined by linear extrapolation of $M(H)$ dependencies from high fields. M_s in the vicinity of the Curie temperature, where $M(H)$ dependencies are nonlinear, was estimated by the Belov-Arrott method for second-order magnetic phase transitions. Using this method, the Curie temperatures were determined for every alloy except the $x=0.19$ sample, where the ferromagnetic-paramagnetic transition is a first-order phase transition. The paramagnetic susceptibility of the alloys was defined from $M(T)$ dependencies measured above T_C up to 700 K in a magnetic field of 0.2 T. The latent heat of the martensitic transition was determined from differential scanning calorimetry, performed by Pyris-1 DSC equipment.

IV. EXPERIMENTAL RESULTS

Temperature dependencies of the spontaneous magnetization M_s of the $\mathrm{Ni}_{2+x}\mathrm{Mn}_{1-x}\mathrm{Ga}$ alloys are shown in Fig. 1. It is seen that M_s gradually decreases with increasing temperature and exhibits a pronounced change (smeared jump) when approaching a certain temperature T_m. This jump in magnetization is caused, as has been shown in numerous studies,[10,13,14] by a structural phase transition from martensite to austenite. As is evident from these measurements, the austenitic phase is ferromagnetic above T_m for $x<0.19$, while in the $x=0.19$ alloy the transformation from martensite to austenite is accompanied by a transition from ferromagnetic to paramagnetic state.

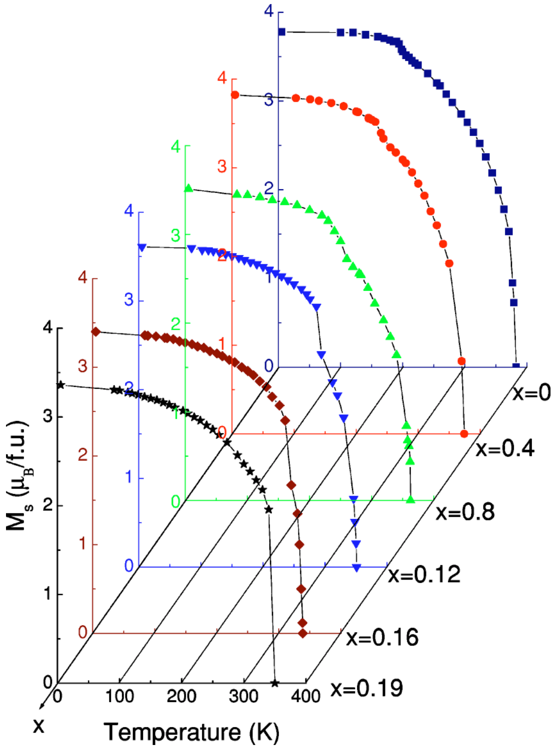

FIG. 1. Temperature dependencies of spontaneous magnetization of the $\mathrm{Ni}_{2+x}\mathrm{Mn}_{1-x}\mathrm{Ga}$ alloys. M_s was determined from field dependencies of magnetization measured up to 10 T.

The compositional dependence of the martensitic transformation temperature T_m is shown in Fig. 2. This figure also shows the dependence of ferromagnetic ordering temperature T_C on Ni content x. It is seen that both these dependencies are practically linear with T_m increasing and T_C decreasing with Ni content. These temperatures merge in a range of $x=0.18-0.19$. The phase diagram of the $\mathrm{Ni}_{2+x}\mathrm{Mn}_{1-x}\mathrm{Ga}$ system obtained in the present study is in good agreement with the previously found diagram.[6,15]

The magnetic moment of these alloys, $M_s(0)$, was obtained by extrapolation of $M_s(T)$ to 0 K. It was found that $M_s(0)$ approximately linearly decreases at substitution of Mn by Ni, as is shown in Fig. 3. The value of magnetic moment in the stoichiometric $\mathrm{Ni}_2\mathrm{MnGa}$ appears to be close to those reported in other studies.[4,10,11]

The $M_s(T)$ dependencies in $\mathrm{Ni}_{2+x}\mathrm{Mn}_{1-x}\mathrm{Ga}$ alloys, shown in Fig. 1, show that the change of spontaneous magnetization at the martensite-austenite transformation increases with Ni content. A jump of magnetization at this transition is also observed in magnetic fields larger than the saturation field, as is shown in Fig. 4. The compositional dependencies of the

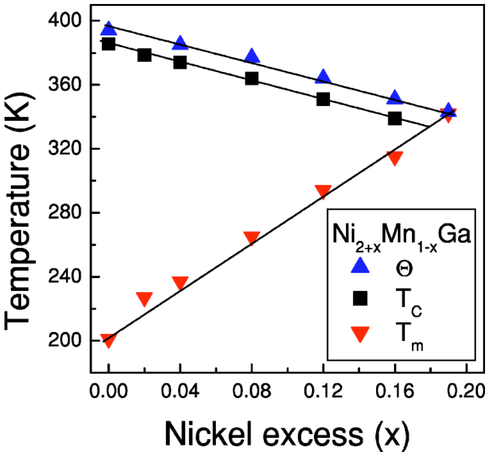

FIG. 2. Compositional dependencies of the martensitic transformation temperature T_m, Curie temperature T_C, and paramagnetic Curie temperature Θ.

magnitudes of magnetization jump ΔM measured in various magnetic fields are shown in Fig. 5.

As can be seen from Fig. 4, with increasing magnetic field the magnetization jumps shift to higher temperatures. This is due to the influence of magnetic field on martensitic transformation temperature. It follows from these measurements that the shift ΔT of T_m under magnetic field increases weakly with Ni content (see Table I).

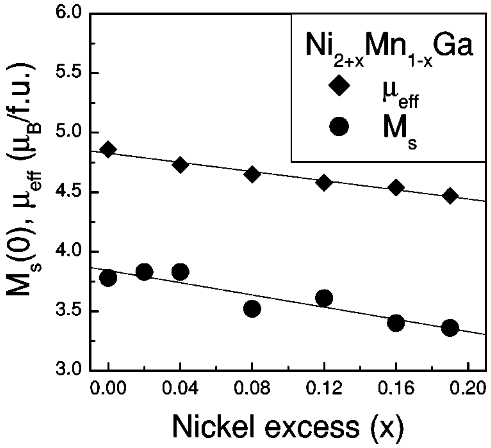

FIG. 3. Compositional dependencies of saturation magnetic moment $M_s(0)$ and effective magnetic moment μ_{eff} of the $\text{Ni}_{2+x}\text{Mn}_{1-x}\text{Ga}$ alloys.

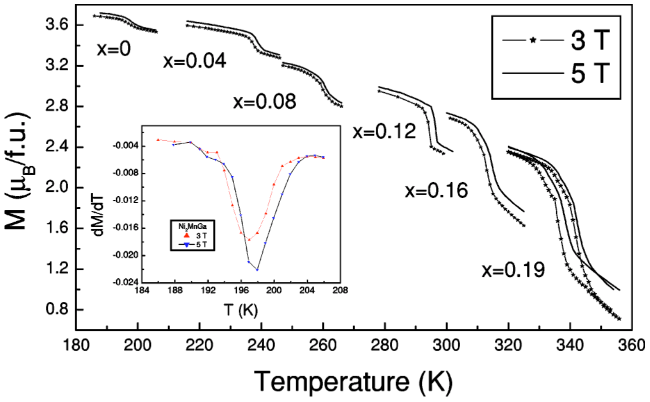

FIG. 4. Magnetization jump at the martensitic transition in magnetic fields of 3 T (dashed line) and 5 T (solid line). For compositions $0 \leqslant x \leqslant 0.16$ $M(T)$ dependencies were measured upon heating. For the composition $x=0.19$ a temperature hysteresis loop of the magnetization observed at martensitic transition is shown. The inset shows temperature derivatives of magnetization for Ni_2MnGa measured in magnetic fields 3 and 5 T.

The influence of a magnetic field on the martensite-austenite transition temperature was studied only for $x=0$ (Ref. 14) and $x=0.18-0.19$ (Refs. 15–17) compositions. For the stoichiometric composition the shift of T_m under magnetic field was estimated as $dT_m/dH \approx 0.2$ K/T.[14] For the $x=0.18$ and $x=0.19$ compositions, $dT_m/dH \approx 1$ K/T was reported in Refs. 15,17, whereas in Ref. 16 this quantity was estimated as 3.5 K/T. It is worth noting that the shift of T_m is determined with a significant error. This is caused mainly by the fact that the jump of magnetization at the martensitic transformation is broad, which makes the correct determination of T_m temperature difficult. Besides, the martensitic transformation is a first-order structural phase transition and is characterized by a temperature hysteresis. Therefore, the T_m temperature can differ from the temperature at which the jump of magnetization occurs. The most correct method for determining T_m is to determine this temperature as the average of the temperatures at which the magnetization jump is observed on cooling and heating, respectively. In the present study, T_m was determined as a temperature of the magnetization jump while heating the sample. The temperature hysteresis loop was measured for the $x=0.19$ sample. It was found (see Fig. 4) that T_m determined at increasing temperature differs from T_m estimated from averaging of measurements in hysteretic regime by 2–3 K. The width of the temperature hysteresis loop is approximately the same in different magnetic fields, so the additional error in the determination of the shift of T_m caused by a magnetic field does not exceed 0.3 K.

Temperature dependencies of reciprocal paramagnetic susceptibility of the $\text{Ni}_{2+x}\text{Mn}_{1-x}\text{Ga}$ system are shown in Fig.

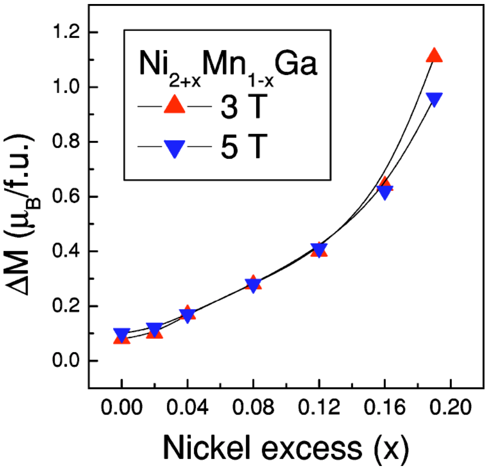

FIG. 5. The magnetization jump at the martensitic transition in various magnetic fields as a function of Ni concentration in the $\text{Ni}_{2+x}\text{Mn}_{1-x}\text{Ga}$ alloys.

TABLE I. Theoretical and experimental values of the shift ΔT of the martensitic transition temperature T_m in a magnetic field $\Delta H = 2$ T for $Ni_{2+x}Mn_{1-x}Ga$.

x	Q(J/mol)	T_m(K)	$\Delta M(\mu_B)$	ΔT(K) (ΔH=2 T) Theory	ΔT(K) (ΔH=2 T) Experiment
0	270	201	0.1	0.82±0.2	0.8±0.5
0.04	600	237	0.17	0.75±0.2	0.95±0.5
0.08	910	265	0.28	0.92±0.2	0.95±0.5
0.12	1250	294	0.41	1.07±0.2	1.10±0.5
0.16	1710	315	0.62	1.28±0.2	1.30±0.5
0.19	2260	342	0.96	1.62±0.2	1.60±0.5

6. In the temperature range studied, the susceptibility follows Curie-Weiss law. The compositional dependencies of paramagnetic Curie temperature Θ and effective magnetic moment μ_{eff} are shown in Figs. 2 and 3, respectively. Clearly, both these parameters decrease monotonically with increasing x. The paramagnetic susceptibility was measured earlier in stoichiometric Ni_2MnGa alloy only.[10,18] The values of Θ and μ_{eff} obtained are somewhat larger than those reported previously. This difference can be due to the fact that the present measurements were performed in a wider temperature interval.

The compositional dependence of the latent heat Q of the martensite-austenite phase transition is shown in Fig. 7. Evidently, Q strongly increases with increasing x. These results are in good agreement with recently published results.[19]

V. DISCUSSION

Based on the compositional dependencies of the saturation magnetic moment and effective magnetic moment of $Ni_{2+x}Mn_{1-x}Ga$ alloys (Fig. 3), the magnetic moments and effective magnetic moments of Mn and Ni atoms were calculated from the equations[20]

$$M_s(0) = (1-x)\mu_{\text{Mn}} + (2+x)\mu_{\text{Ni}}, \qquad (1)$$

$$\mu_{\text{eff}} = \sqrt{(1-x)\mu^2_{\text{eff Mn}} + (2+x)\mu^2_{\text{eff Ni}}}. \qquad (2)$$

The results of these calculations are presented in Table II. The obtained values of the magnetic moments of the constituting atoms are in good accordance with the results of neutron diffraction and nuclear magnetic resonance studies for the stoichiometric composition. It was shown[3,4,21] that in Ni_2MnGa the Mn magnetic moment is about $(2.84-3.41)\mu_B$ and the Ni magnetic moment is about $(0.3-0.41)\mu_B$.

Note that these calculations are based on assumptions that the magnetic moments of constituent atoms does not change with deviations from stoichiometry and that the Ni atoms possess similar moments in different crystallographic sites. In general this is not the case, because magnetism of Heusler alloys is described in a band model. It means that the values of magnetic moments depend on the density of states at the Fermi level and on the exchange splitting parameter, therefore being the concentration and structure dependent values. As has been noted in Ref. 22, in Ni_2MnX Heusler alloys the distance between the atoms is sufficiently large so that direct overlap of electron orbitals is negligible and the delocalization effects are of secondary importance. Therefore, in the first approximation a localized moments model is applicable

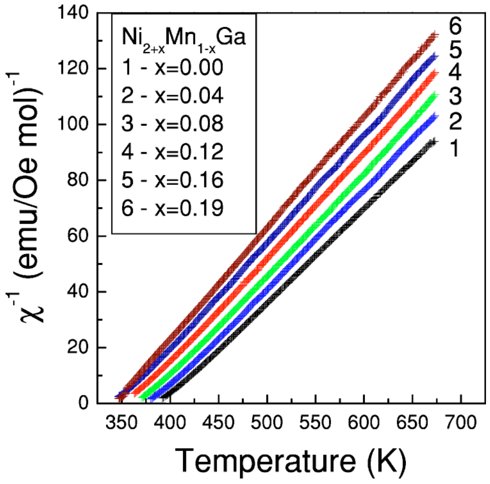

FIG. 6. Temperature dependencies of paramagnetic susceptibility.

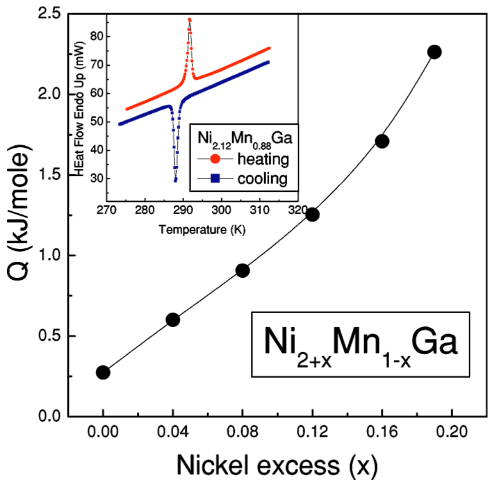

FIG. 7. Compositional dependence of the latent heat of the martensitic transition. The inset shows an example of the differential scanning calorimetry measurements.

TABLE II. Magnetic moments μ and effective magnetic moments μ_{eff} of Mn and Ni atoms.

	$\mu(\mu_B)$	$\mu_{\mathrm{eff}}(\mu_B)$	$\mu_{\mathrm{eff}}^{\mathrm{loc}}(\mu_B)$	$\mu_{\mathrm{eff}}^{\mathrm{loc}}/\mu_{\mathrm{eff}}$
Mn	2.99±0.32	4.43±0.13	3.86±0.14	0.87±0.11
Ni	0.43±0.14	1.35±0.18	1.05±0.21	0.77±0.10

for the description of magnetic properties of these alloys. However, from the results of magnetic and nuclear magnetic resonance measurements of Ni_2MnGa,[21] it was concluded that in this alloy the Mn magnetic moments are mainly localized, while Ni magnetic moments are essentially delocalized. The character of magnetism can be judged from the comparison of the magnetic moments of the constituting atoms and their effective magnetic moments (see Table II).

In the model of localized magnetic moments for the spin-only state (orbital moment is quenched) the interrelation between effective magnetic moment and the moment in the magnetically ordered state is given by Wohlfarth-Rhodes equation,

$$\mu_{\mathrm{eff}}^{\mathrm{loc}} = \sqrt{\mu(\mu + 2)}. \qquad (3)$$

In the band model the value of $\mu_{\mathrm{eff}}^{\mathrm{loc}}$ calculated from Eq. (3) should be smaller than the experimental value of the effective moment due to the influence of delocalization effects. The values of $\mu_{\mathrm{eff}}^{\mathrm{loc}}$ for the Mn and Ni subsystems are given in Table II. As evident from these data, for both Mn and Ni subsystems μ_{eff} and $\mu_{\mathrm{eff}}^{\mathrm{loc}}$ are close to each other, although in both cases $\mu_{\mathrm{eff}}^{\mathrm{loc}}$ is slightly smaller than μ_{eff}. Within the experimental error of the measurements the $\mu_{\mathrm{eff}}^{\mathrm{loc}}/\mu_{\mathrm{eff}}$ ratio is the same for both Mn and Ni subsystems. Thus, the present experimental data do not suggest that the Ni subsystem is more delocalized that the Mn one.

It should be understood, however, that the magnetic moments in the magnetically ordered state were determined in the martensitic phase, whereas the effective magnetic moments were calculated from the paramagnetic susceptibility measured in the austenitic state. It makes no difference if magnetism is described in the localized model, because in this case the magnitude of the magnetic moment depends weakly on the crystallographic environment. In the band model, magnetic moments depend on the degree of overlap of electron orbitals, which changes at structural transformation. Because of this, the possibility that the magnetic moments will change at structural transition must not be ruled out. The qualitative arguments given above are supported by the experimental data reported in Ref. 21, which indicate that the magnetic moment of Mn is the same in austenitic and martensitic phases, whereas the magnetic moment of Ni in the austenitic phase is larger than that in the martensitic phase. The latter observation is conditioned by a higher density of states of Ni at the Fermi level in austenitic state than that in the martensitic state, as electronic structure calculations have revealed.[5]

As evident from Fig. 2, Curie temperature of the austenitic phase decreases at substitution of Mn for Ni. This is due to the fact that this substitution leads to an increase in the number of atoms with smaller magnetic moments. Similar tendency takes place presumably for a virtual Curie temperature of the martensitic phase. This follows from the observation that in the low-temperature martensitic phase magnetization of the $Ni_{2+x}Mn_{1-x}Ga$ alloys with a higher x decreases more rapidly with increasing temperature.

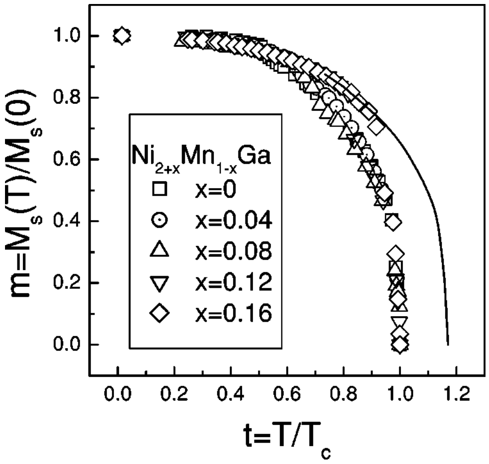

FIG. 8. Reduced magnetization $m=M_s(T)/M_s(0)$ as a function of reduced temperature $t=T/T_C$ for the $Ni_{2+x}Mn_{1-x}Ga$ alloys. The solid line is the reduced magnetization of virtual martensitic phase.

The magnetization data shown in Fig. 1 allow estimation of virtual Curie temperature of the martensitic phase. Figure 8 shows temperature dependencies of reduced spontaneous magnetization $m=M_s(T)/M_s(0)$ of the alloys as a function of reduced temperature $t=T/T_C$. It is seen that the magnetization of the austenitic phase and the magnetization of the martensitic phase change with temperature in a different way, whereas the reduced magnetizations of these phases are similar for different compositions. Since in Ni-Mn-Ga the orbital moment is quenched, these dependencies can be expressed by a Brillouin function with a quantum spin number S. Assuming that $m(t)$ for austenite and martensite is expressed by the same Brillouin function, it can be suggested that the difference in $m(t)$ of martensite and austenite is due to the difference in their Curie temperatures. Comparing $m(t)$ dependencies of martensitic and austenitic phases, it is possible to reconstruct the virtual Curie temperature of the martensitic phase, which is shown by the solid line in Fig. 8. It appears to be 17% higher than the Curie temperature of the austenitic state. This value is twice as large as that obtained from phenomenological Landau theory.[23]

The larger value of the Curie temperature of martensite as compared to the Curie temperature of corresponding austenite is due to changes in interatomic distances and in the overlap of electronic orbitals. As is evident from the analysis of experimental data, this effect cannot be attributed solely to a change in the unit cell volume at the martensitic transformation. Indeed, a study of the influence of hydrostatic pressure on the Curie temperature T_C and martensitic transformation temperature T_m of stoichiometric Ni_2MnGa (Ref. 18) has shown that the exchange interaction of the austenite increases with decreasing unit cell volume. At the same time, it is known[24] that the unit cell volume of martensite is larger than that of austenite. Therefore, it seems likely that the primary role in martensitic transformation in Ni_2MnGa Heusler alloys belongs to the crystal lattice distortions. Such a mechanism of the influence of a structural transition on exchange interaction in intermetallic compounds $Gd_5(Si_xGe_{1-x})_4$ was discussed recently in Ref. 25.

As is evident from Fig. 5, the magnitude of the magnetization jump ΔM at the structural transition strongly increases with Ni content. This is caused by the fact that the increase of x leads to the increase of T_m. Under these circumstances, the difference between magnetizations of martensite and austenite at T_m increases as well. It is also seen from Fig. 5 that the magnetization jumps ΔM at T_m diminishes at increasing magnetic field, which is most pronounced at high x. The behavior of ΔM in the alloys with a small x results from the fact that the martensitic transformation in these alloys occurs at temperatures far below the Curie temperature T_C and therefore the influence of a magnetic field on magnetization is weak. In the alloys with a large x, T_m is close to T_C of the austenitic phase and the external field strongly affects the magnetization of this phase, whereas magnetization of the martensitic phase depends weakly on the magnetic field.

It has been already mentioned that the temperature of structural transition shifts to higher temperatures upon application of a magnetic field. Such behavior is governed by the influence of Zeeman energy, which stabilizes the martensitic phase with a larger magnetization. Experimental data on the shift of T_m are presented in Table I. These results indicate that for the alloys studied the shift is rather small (1–2 K as the magnetic field changes for 2 T) and slightly enhances with increasing Ni content. The table also contains theoretical estimation of the shift of T_m in magnetic field, derived from a thermodynamical Clapeyron-Clausius relation for first-order phase transitions:

$$\Delta T = \Delta M H T_m / Q.$$

The agreement between experimental and theoretical values can be considered as satisfactory.

ACKNOWLEDGMENTS

This study was supported by RFBR Grants No. 02-02-16636 and No. 03-02-17443. Work at Argonne was supported by U.S. Department of Energy, BES Materials Sciences under Contract No. W-31-109-ENG-38. One of the authors (V.V.K.) gratefully acknowledges the Japan Society for the Promotion of Science for Financial support.

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