4-1-2009

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Published in *Journal of Applied Physics*, Vol. 105 No. 7 (2009) at doi: 10.1063/1.3067446

Magnetic and electrical properties of $Ni_{50}Mn_{35}In_{15-x}Si_x$ Heusler alloys

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(Presented 13 November 2008; received 16 September 2008; accepted 1 November 2008; published online 13 February 2009)

We have studied the magnetic and electrical properties of the polycrystalline ferromagnetic $Ni_{50}Mn_{35}In_{15-x}Si_x$ $(1 \leq x \leq 5)$ Heusler alloys through magnetization, thermal expansion, and resistivity measurements. It was observed that an increase in Si concentration strongly affects the ground state of the martensitic phase and the magnetic properties of compounds. A magnetic phase diagram has been constructed for these alloys. It was found that both martensitic transition temperature (T_M) and Curie temperature of austenitic phase (T_C) decrease, while ferromagnetic ordering temperature of the martensitic phase increases with increasing Si concentration. The magnetoresistance $(\Delta\rho/\rho)$ associated with martensitic transformation was found to vary from –47% for $x=2$ at $T=261$ K to −26% for $x=5$ at $T=230$ K for a magnetic field change of 5 T. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3067446]

The off-stoichiometric $Ni_2Mn_{1+y}X_{1-y}$ Heusler alloys that undergo structural (martensitic) transformations at T_M, below the ferromagnetic ordering temperature (T_C), are of considerable interest because of their shape memory, magnetocaloric, exchange bias, and magnetoresistance (MR) properties.[1–7] In some composition ranges, these alloys possess at least two temperature-induced phase transitions: (i) a first-order structural martensitic transition at T_M, accompanied by a change in the magnetic state of the compound and (ii) a ferromagnetic-paramagnetic transition at T_C of the austenitic phase,[3,5] and characterized by paramagnetic (PM) or antiferromagnetic (AFM) type of magnetization curves of the martensitic phase in the vicinity of T_M. In this case, the magnetization of the martensitic phase is considerably smaller than that of the austenitic phase.[1] The coupling between Mn magnetic moments in the martensitic phase deviate from ferromagnetic (FM) alignment when the Mn atoms occupy the X sites in off-stoichiometric $Ni_2Mn_{1+y}X_{1-y}$ Heusler alloys. The magnetism in these alloys is basically determined by the Mn magnetic moments and described in terms of an indirect exchange via conduction electrons.[8] Thus, the strong influence of excess Mn on the magnetic behavior of the martensitic phase in $Ni_2Mn_{1+y}X_{1-y}$ compounds and the oscillatory character of the indirect exchange interaction present an opportunity to control the magnetic behavior of the martensitic phase of $Ni_2Mn_{1+y}X_{1-y}$ compounds with constant Mn concentration by controlling internal parameters such as interatomic distance. Recently the effect of the partial substitution of In by Si on the phase transition temperatures and respective magnetic entropy changes in $Ni_{50}Mn_{35}In_{15-x}Si$ with $1 \leq x \leq 5$ has been studied.[9] It was found that a small amount of Si in the In site (about 20%) results in a significant variation in the magnetization process in the vicinity of T_M and also in an increase in the positive magnetic entropy changes of more than 300% (relative for the $x=0$).[9] In the present work, we report the structural, electrical, and magnetic properties of $Ni_{50}Mn_{35}In_{15-x}Si_x$ Heusler alloys studied using thermal expansion, x-ray diffraction, resistivity, and magnetization measurements.

The samples were prepared and the magnetization measurements were performed by the methods described in Ref. [9]. Before the magnetization measurements $[M(T)]$, the samples were heated to 380 K and then cooled down to 5 K at present or absent of magnetic field to perform field cooled (FC), and zero-field cooled (ZFC) $M(T)$ accordingly. Both ZFC and FC measurements were done by heating the samples from 5 to 400 K in an external magnetic H. The phase purity and crystal structures were determined by powder x-ray diffraction using Cu Kα ($\lambda_{K\alpha}=1.543114$ Å) radiation. Thermal expansion measurements were carried out using a capacitance dilatometer [10].

Room temperature XRD measurements revealed that the samples were in the austenitic phase and possess the Heusler $L2_1$ cubic structures belonging to the $Fm\bar{3}m$ space group up to $x=4$, beyond which alloys become mixed with unknown phases.

The typical ZFC and FC $M(T)$ curves in a magnetic field of 0.01 T for $x=2$ are shown in Fig. 1(a). The curves split at temperature $T \approx 200$ K. As the temperature increase from this point, the sample undergoes a transition at the Curie temperature of martensitic phase (T_{CM}) and the magnetization shows a minimum (at $T \approx 250$ K). Further increasing the temperature, the sample passes from a low magnetic moment state (AFM or PM) to a FM state at the martensitic transition (T_M), and, finally, from a FM austenitic phase to PM austenitic phase at T_C. The splitting of the ZFC and FC curves at temperature lower than T_{CM} is attributed to a coexistence of AFM and FM phases.[11,12] The high field ($H=5$ T) ZFC $M(T)$ magnetization is shown in Fig. 1(b). As the temperature increases, the magnetizations of all the samples show steplike changes at T_M, indicating an AFM (or PM) to ferromagnetic transition. Upon further increase in the

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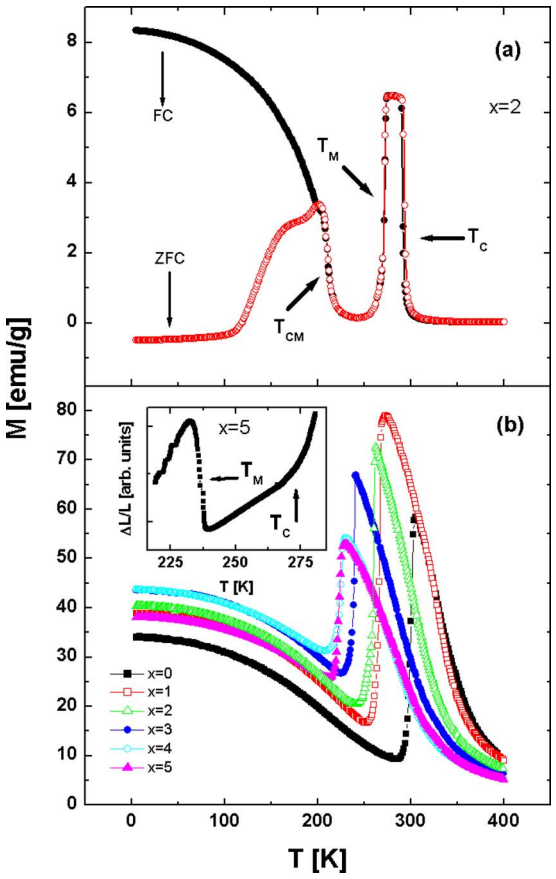

FIG. 1. (Color online) (a) ZFC and FC magnetization (MvT) curves of $Ni_{50}Mn_{35}In_{13}Si_2$ at $H=0.01$ T. (b) ZFC magnetization (MvT) of $Ni_{50}Mn_{35}In_{15-x}Si_x$ at $H=5$ T. [Inset 1(b)] Thermal expansion of $Ni_{50}Mn_{35}In_{10}Si_5$ near the martensitic transition.

temperature, the magnetization decreases and the sample transforms to the PM state. The jumplike variation in the thermal expansion curves at T_M and the slope change in the same at T_C are typical for first order transitions and second order transitions, respectively [see inset in Fig. 1(b)]. Thus the sharp change in magnetization near T_M is due to a first order martensitic transformation from an AFM/PM to a FM austenitic state.

Figure 2 shows the variation in transition temperatures and saturation magnetizations (M_s) at $T=5$ K with conduction electron concentration (e/a), where e/a is proportional to the Si concentration. The increase in the e/a ratio results in an increase in T_{CM} and a decrease in both T_M and T_C for concentrations interval $x \leq 4$, i.e., where solid solution of $Ni_{50}Mn_{35}In_{15-x}Si_x$ exists. The M_s increases with the increasing e/a ratio for $x \leq 4$. The increase in M_s with Si concentration could be attributed to a change in both the electron concentration and lattice parameters or to an increase in the fractional FM components in these compounds.

The resistivity of $Ni_{50}Mn_{35}In_{15-x}In_x$ calculated as $R(T)/R$ (400K) as a function of temperature at zero and 5 T magnetic fields are shown in Fig. 3. The resistivity curves of all the samples are relatively constant until the temperature approaches T_M and drops abruptly at T_M (see Fig. 3). As the temperature increases further, the resistivity increases almost linearly until T_C detected from the slope change. This slope

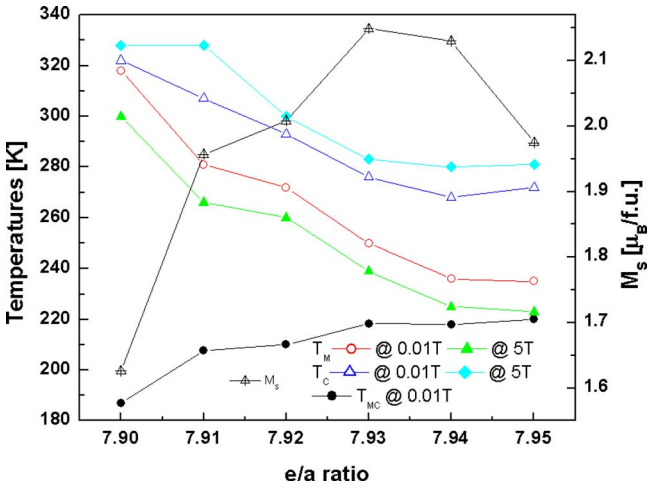

FIG. 2. (Color online) The variation in relevant transition temperatures and saturation magnetization ($T=5$ K) as a function of e/a.

change is the result of the system undergoing the second order transition from the ferromagnetic austenitic state to the paramagnetic austenitic state. The MR of the $Ni_{50}Mn_{35}In_{15-x}Si_x$ in the vicinity of T_M was determined by using the relation $MR=[\rho(H,T)-\rho(0,T)]/\rho(0,T)\times 100\%$. The maximum MR was found to be -47% for $x=2$ at $T \approx 261$ K. It was observed that increasing the e/a ratio decreases the MR (see in inset in Fig. 4). The cause of the decrease in MR due to the substitution of Si for In could be due to a change in the electronic structure as a result of the change in e/a ratio and lattice constant.[13]

We have observed that the small amount of Si substituted in In sites results in a low temperature transition detected from ZFC $R(H)$ at 5 K (shown in Fig. 5) and from $R(T)$ (see inset in Fig. 5, for clear visualization only the low temperature $R(T)$ is shown, where arrows illustrate the hysteresis). Such behavior can be attributed to an AFM-like state observed in ZFC samples [see Fig. 1(a)]; however a detailed study would be required to explain this low temperature transition.

Figure 6(a) shows typical isothermal $\rho(H)$ curves of the $Ni_{50}Mn_{35}In_{15-x}Si_x$ system near T_M (shown for $x=2$ at $T=255$ K). An increase in the external magnetic field de-

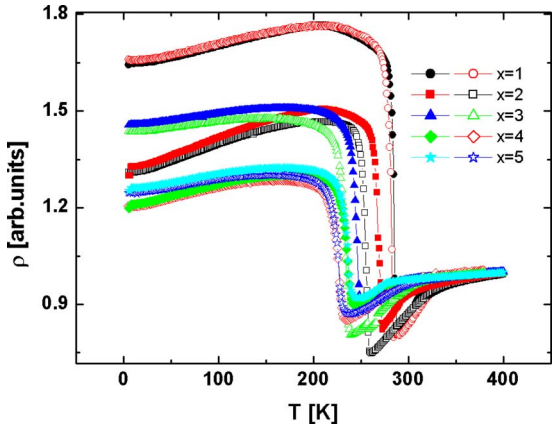

FIG. 3. (Color online) ZFC resistivity of $Ni_{50}Mn_{35}In_{15-x}Si_x$ as a function of temperature at zero (closed symbols) and external magnetic field (opened symbols).

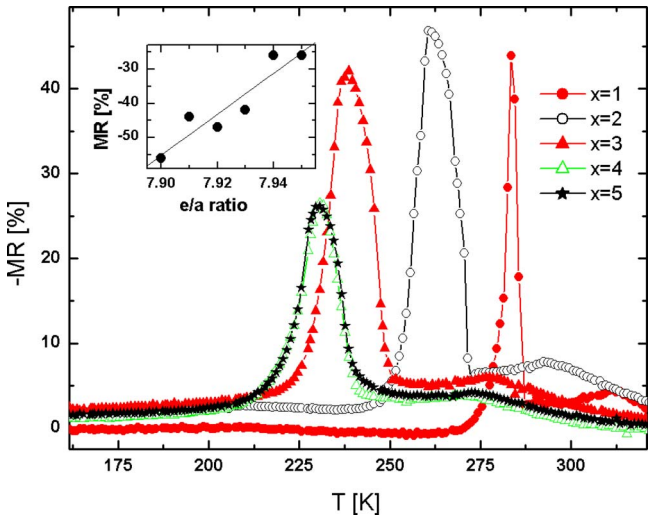

FIG. 4. (Color online) MR as a function of temperature (t). The inset shows the variation in MR with conduction electron concentration.

creases the resistivity of the sample. When H is reduced, the reverse transition takes place, and a hysteresis is observed. Such a hysteresis in $\rho(H)$ indicates a field-induced first order transition. The magnetization isotherms also indicate the existence of metamagnetic transitions in the same temperature intervals [see Fig. 6(b)]. Therefore these observations suggest that large drop in the resistivity in the vicinity of T_M is due to metamagnetic behavior, where the system transforms from a state that contains antiferromagnetically coupled regions to a purely ferromagnetic state.

In conclusion, we studied the magnetic and electrical properties of $Ni_{50}Mn_{35}In_{15-x}Si_x$ $(1 \leq x \leq 5)$ Heusler alloys. It was observed that the substitution of a small amount of Si in In sites increases the saturation magnetization at 5 K and the magnetization at T_M. The transition temperatures T_M and T_C were found to decrease with an increase in Si concentration. It was observed that the MR value decreases with increasing

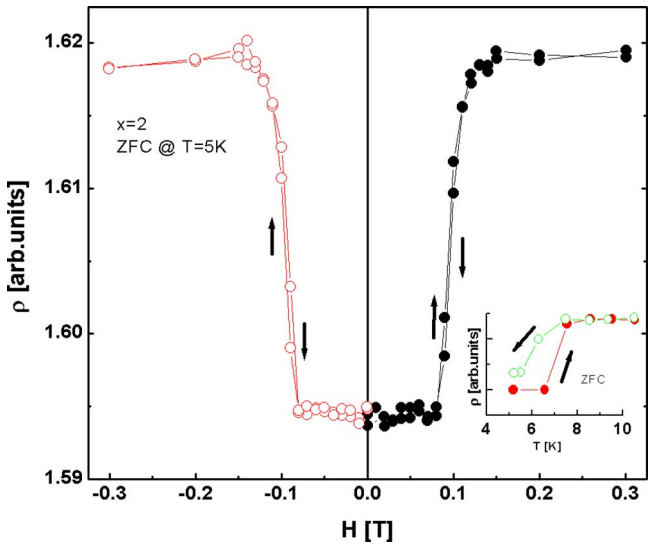

FIG. 5. (Color online) ZFC resistivity as a function of external magnetic field at $T=5$ K. The inset shows the ZFC resistivity as a function of temperature at zero magnetic fields.

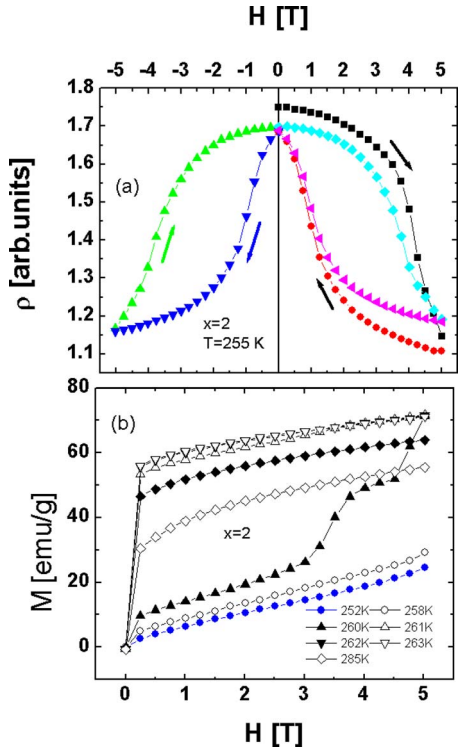

FIG. 6. (Color online) (a) Resistivity as a function of external magnetic field in the vicinity of T_M, and (b) $M(h)$ in the vicinity of T_M and T_C.

e/a ratio. The MR value was found to vary from -47% for $x=2$ at $T=261$ K to -26% for $x=5$ at $T=230$ K for a magnetic field change of 5 T.

This research was supported by a Research Opportunity Award from Research Corporation (Grant No. RA-0357), and by the Office of Basic Energy Sciences, Material Sciences Division of the U.S. Department of Energy (Contract No. DE-FG02-06ER46291).

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