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Phase Transitions and Magnetoresistance in Ni50Mn50−xInx Heusler Alloys

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Phase transitions and magnetoresistance in $Ni_{50}Mn_{50-x}In_x$ Heusler alloys

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The phase transitions and magnetoresistance in polycrystalline ferromagnetic $Ni_{50}Mn_{50-x}In_x$ ($15 \leqslant x \leqslant 16.2$) Heusler alloys were studied through ac susceptibility, magnetization, thermal expansion, and resistivity measurements in the temperature interval of 5–400 K. The temperatures of the martensitic transformations were found to be strongly dependent on In concentration and on the strength of the applied external magnetic field. We observed large magnetoresistance (MR) $\Delta\rho/\rho_o \approx -80\%$ for $x=16$ at $T \approx 125$ K and $\Delta\rho/\rho_o \approx -56\%$ for $x=15$ at $T \approx 309$ K for $\Delta H = 5$ T. In addition to large MR, the $Ni_{50}Mn_{50-x}In_x$ system exhibits ferromagnetic shape-memory effect and a large magnetic entropy change. Hence this system has potential to be a multifunctional applied material. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2828599]

The Ni–Mn–In based Heusler alloys that undergo martensitic transformations are of considerable interest because of their unique physical properties and multifunctional applications.[1–6] These alloys possess at least two temperature-induced phase transitions: The first order structural martensitic transition (T_M), accompanied by a change in the magnetic state of the compound, and the ferromagnetic-paramagnetic transition at the Curie temperature (T_C) of the austenitic phase.[1,4] The transition temperatures T_M and T_C are strongly dependent on the indium concentration.

In this article, we report on the phase transitions and magnetoresistance (MR) properties of the $Ni_{50}Mn_{50-x}In_x$ ($15 \leqslant x \leqslant 16.2$) Heusler system. The concentration range of In was chosen to cover the interval of the possible existence of a coupled magnetostructural transition ($T_M = T_C$).[7] The MR was found to vary with In concentration and reaches a peak value of about −80% for the sample with $x=16$ at $T \approx 125$ K. This value of MR is comparable to that found recently in the $Ni_{50}Mn_{50-x}Sn_x$ Heusler system.[8,9]

The samples were prepared and the magnetization measurements were performed by the methods described in Ref. 4. Direct current resistivity, using the four-probe method, was measured in the temperature range of 5–400 K. The ac susceptibility [$\chi_{ac}(T)$] was measured in the temperature range of 80–325 K using an ac susceptometer (Lake Shore model 7000) with an ac magnetic field of amplitude of 5 Oe and a frequency of 95.2 Hz. Thermal expansion measurements were carried out using a capacitance dilatometer.[10] The MR of the $Ni_{50}Mn_{50-x}In_x$ in the vicinity of T_M was determined by resistivity (ρ) measurements in external magnetic fields of 0 and 5 T using the relation $MR = [\rho(H) - \rho(0)]/\rho(0) \times 100\%$.

Figure 1 shows the temperature dependence of the dc magnetization $M(T)$ for $15 \leqslant x \leqslant 16.2$ in the temperature range of 5–400 K with an applied magnetic field of 0.1 T. As an example, consider the sample with $x=15.2$. The magnetization of this sample increases sharply near 210 K and passes through a maximum (220–290 K). The magnetization decreases upon further increase in temperature and shows typical paramagnetic behavior when $T > T_C$ (≈ 325 K). The alloys with $15 \leqslant x \leqslant 16$ undergo a martensitic transformation to the ferromagnetic austenitic phase at T_M, accompanied by a jump-like variation in cell volume (see inset of Fig. 1). Such a variation in cell volume near the martensitic transformation confirms the first order nature of the transition at T_M. For the alloy with $x=16.2$, no anomaly of $M(T)$ associated with a martensitic transformation was observed. The magnetization $M(T)$ of the samples with $x=15$ and 15.05 (see Fig. 1) were found to decrease around 175 K and pass through a minimum near $T=250$ K. The temperature at which the magnetization decreases is the T_C of the martensitic phase (for example, see Fig. 1, $x=15$ represented by T_{CM}).

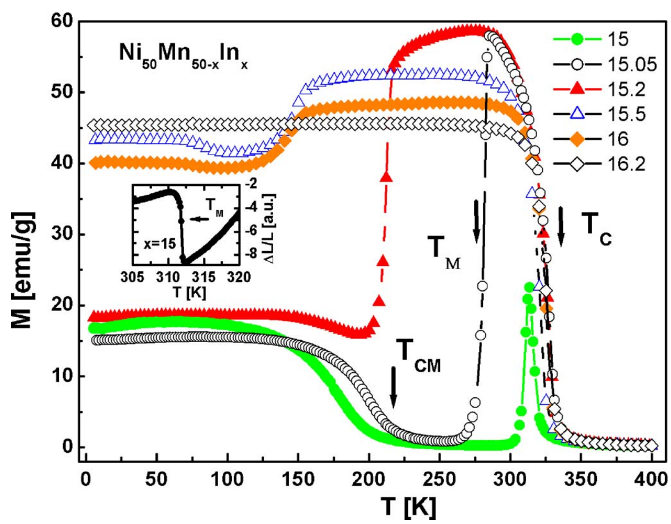

FIG. 1. (Color online) Magnetization (M) vs temperature (T) in a magnetic field (H) of 0.1 T for different In concentrations. Arrows indicate the transition temperature for $x=15.05$. (Inset) Thermal expansion of $Ni_{50}Mn_{35}In_{15}$ near the martensitic transition.

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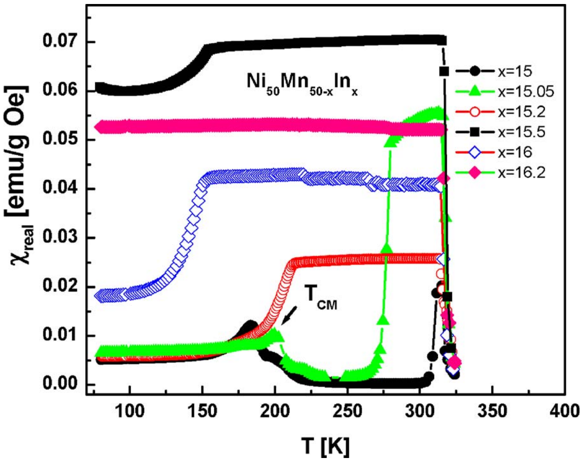

FIG. 2. (Color online) Temperature dependence of the ac susceptibility for different In concentrations.

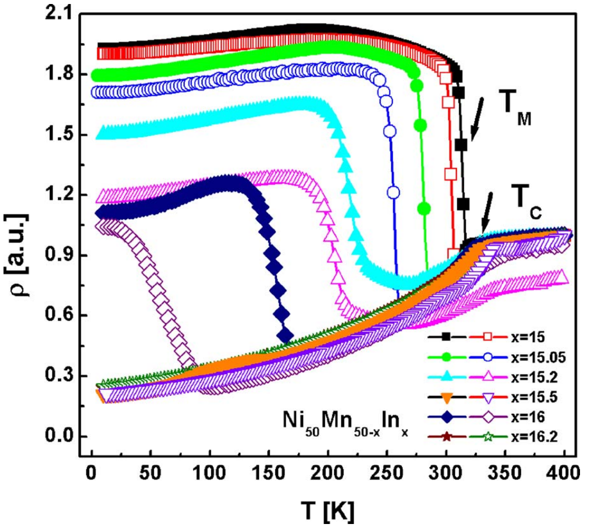

FIG. 3. (Color online) Resistivity (ρ) as a function of temperature (T) for different In concentrations in 0 T (closed symbol) and 5 T (open symbol) applied magnetic fields. Arrows indicate T_C and T_M for $x=15$ sample at $H=0$ T.

In order to confirm the transition temperatures found in the dc magnetization measurements, ac susceptibility measurements were performed. The alloys with $15 \leqslant x \leqslant 16$ undergo a transformation to the ferromagnetic austenitic phase at T_M, which is in accordance with the dc magnetization measurements (see Fig. 2). The sharp peaks in the $\chi_{ac}(T)$ for the samples with $x=15$ and 15.05 confirm the phase transition temperatures of these alloys in the low temperature region (see T_{CM} in Fig. 2). The T_M and T_C, determined from the $M(T)$ curves (Fig. 1), agree with those determined by $\chi_{ac}(T)$ measurements.

We found (although not explicitly shown) that the magnetization of the samples with $15 \leqslant x \leqslant 15.05$ is a linear function of the applied field in the interval $T_{CM} < T < T_M$ and is characterized by a magnetic susceptibility $\chi_{dc}(H)$ that is comparable to the $\chi_{dc}(H)$ in a paramagnetic state in the vicinity of T_C. Therefore, the magnetic state of the samples with $15 \leqslant x \leqslant 15.05$ in the interval $T_{CM} < T < T_M$ is either in the paramagnetic or the antiferromagnetic state. The antiferromagnetic type of magnetization $[M(H)]$ of $Ni_{50}Mn_{50-x}In_x$ at 5 K suggests a long-range antiferromagnetically ordered state below T_{CM}.[11]

The resistivity of all the samples is relatively constant until the temperature approaches their respective T_M and drops abruptly at T_M (see Fig. 3). As the temperature increases further, the resistivity increases almost linearly until they reach their respective T_C where the slope changes. This slope change is the result of the system undergoing the second order transition from the ferromagnetic austenitic state to the paramagnetic austenitic state. The temperature at which the resistivity drop occurs depends on the applied field, and the maximum value of the temperature shift ($\Delta T \approx 100$ K) was found for the sample with $x=16$ at a magnetic field of 5 T.

The resistivity at 0 and 5 T and magnetoresistance for $x=15$ are plotted in Fig. 4(a). A large value of magnetoresistance (-56%) was found for $x=15$ near room temperature ($T \approx 309$ K). The MR value for this polycrystalline sample with $x=15$ is similar to that found in the single crystal sample of Ref. 12. Figure 4(b) shows the typical isothermal $\rho(H)$ curves of the $Ni_{50}Mn_{50-x}In_x$ system around T_M (shown for $x=16$ at $T=107$ K). An increase in the external magnetic field decreases 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.[5] The calculated MR from the $\rho(H)$ curves at the temperature of 107 K was found to be $\approx -77\%$. The maximum value of MR, calculated from the $\rho(T)$, was found to be -80% at $T \approx 125$ K for $\Delta H = 5$ T, which is consistent with that found in $\rho(H)$. In the neutron diffraction study of Ref. 2, it was shown that the alloy with

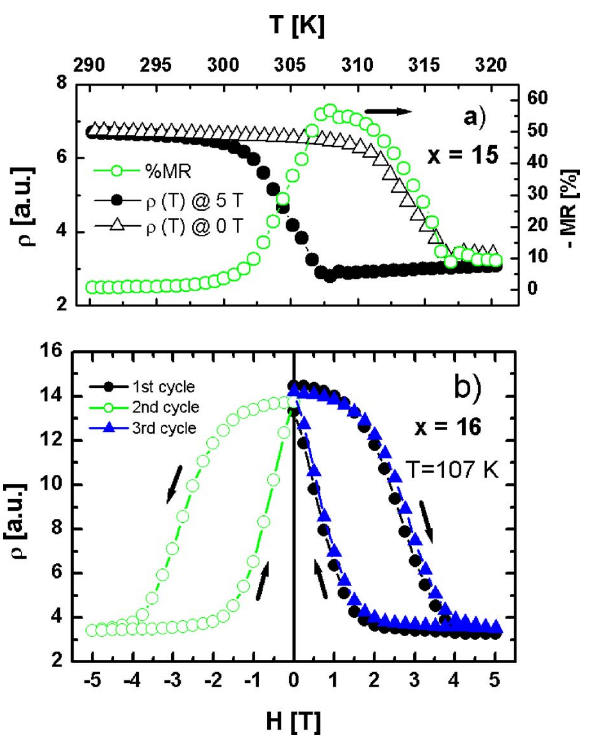

FIG. 4. (Color online) (a) The resistivity (ρ) ($H=0$ and 5 T) and $\Delta\rho/\rho_o$ (%) with $\Delta H = 5$ T for $x=15$ as a function of increasing temperature; (b) Isothermal resistivity (ρ) as a function of applied field (H) for $x=16$.

$x = 16$ undergoes a magnetic-field-induced structural transition from the martensitic state to the austenitic state. Therefore, the large value of MR and the hysteresis of $\rho(H)$ in $Ni_{50}Mn_{34}In_{16}$ are most likely due to a field induced magnetic transition from the martensitic to the austenitic phase. Thus, the underlying cause of the observed MR is likely due to a change in the electronic structure in the austenitic phase compared to that of the martensitic phase, which alters the density of states near the Fermi surface, and therefore affects the electronic transport properties.

In conclusion, we studied the phase transitions and magnetoresistance properties of the $Ni_{50}Mn_{50-x}In_x$ Heusler alloys. We observed three phase transitions: A low temperature transition at T_{CM}, a martensitic transition at T_M, and the ferromagnetic transition at T_C for concentrations in the range $15 \leq x \leq 15.05$. The transition temperature T_{CM} overlaps T_M for concentrations $15.05 < x \leq 16$. Moreover, we observed only the austenitic phase for the sample with $x = 16.2$. A large value of MR (−80%) was observed at $T \approx 125$ K for the sample with $x = 16$. In addition to large MR, the $Ni_{50}Mn_{50-x}In_x$ system exhibits the ferromagnetic shape-memory effect,[3] a large magnetic entropy change,[4] and the exchange bias effect.[11] Hence the Ni–Mn–In system is a multifunctional applied material candidate.

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