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Intermartensitic transformations in $Ni_2Mn_{1-x}Co_xGa$ Heusler alloys

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Ni_2MnGa that possesses a Heusler $L2_1$ structure undergoes a martensitic transformation from the parent cubic (austenitic) phase to a low temperature complex tetragonal structure at $T_M=202$ K and has a Curie temperature of $T_C=376$ K. Some research groups have observed an intermartensitic transformation at a temperature $T_I<T_M$, in Ni_2MnGa single crystals. In this work detailed studies on the influence of substitution in the Mn subsystem by magnetic Co on the intermartensitic transformation properties of $Ni_2Mn_{1-x}Co_xGa$ compounds have been done by magnetization (5–400 K) and thermal expansion (80–300 K) measurements. The samples $Ni_2Mn_{0.92}Co_{.08}Ga$, $Ni_2Mn_{0.91}Co_{.09}Ga$, and $Ni_2Mn_{0.90}Co_{.10}Ga$ were studied. The intermartensitic transformation is observed in all of these alloys. As the Co concentration increases, the transformation becomes more pronounced and the temperature range for which the alloys stay in the intermartensitic state decreases as Co concentration increases. Similarities were observed between the magnetization and thermal expansion curves. The results are discussed in terms of the internal stress produced as a result of the Co substitution. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2176054]

INTRODUCTION

The Heusler alloy Ni_2MnGa undergoes a martensitic transformation from the parent cubic (austenitic) phase to a low temperature complex tetragonal martensitic structure at $T_M=202$ K and has a Curie temperature of $T_C=376$ K.[1] The low temperature martensitic phase is ferromagnetic and exhibits a shape memory effect that can be controlled by the application of magnetic field.[2] This phenomenon indicates that Ni_2MnGa is a very promising material for applications in magnetic sensors and actuators. In addition to the martensitic transformation, some groups have observed another transition known as the intermartensitic transformation in single crystals of Ni_2MnGa.[3,4]. According to previous investigations, the intermartensitic transformation is a first order phase transformation between martensites with different structures at temperatures $T_I<T_M$.[5,6] Several intermartensitic phases have been found so far, and their structures and transformation temperatures T_1 are found to depend on the levels of applied stress and on the chemical composition of the sample.[2]

The observation of the intermartensitic transformations in polycrystalline Ni_2MnGa is not mentioned in the literature. Some groups have studied doped systems of Ni_2MnGa and did not observe any evidence of an intermartensitic transformation.[7] However, in one recent study the Mn sites in Ni_2MnGa were partially substituted with magnetic Co, and $Ni_2Mn_{1-x}Co_xGa$ $(0.08\leqslant x\leqslant 0.10)$ were the only doped alloys in which the intermartensitic transformations were observed.[8]

In our present work, detailed studies of the influence of substitution in the Mn subsystem by magnetic Co on the intermartensitic transformation properties of $Ni_2Mn_{1-x}Co_xGa$ compounds have been done by magnetization (5–400 K) and thermal expansion (80–300 K) measurements. The samples $Ni_2Mn_{0.92}Co_{.08}Ga$, $Ni_2Mn_{0.91}Co_{.09}Ga$, and $Ni_2Mn_{0.90}Co_{.10}Ga$ are studied. The intermartensitic transformation is observed in all of these alloys. As the Co concentration increases, the transformation becomes more pronounced and the temperature range for which the alloys stay in the intermartensitic state decreases as Co concentration increases. A similarity between the magnetization and thermal expansion curves is observed. The results are discussed in terms of internal stress produced due to the Co substitution.

EXPERIMENTAL PROCEDURE

Approximately 5 g of stoichiometric polycrystalline ingots of $Ni_2Mn_{1-x}Co_xGa$ $(0.08\leqslant x\leqslant 0.10)$ were prepared by conventional arc melting in an argon atmosphere using 4*N* purity of Ni, Mn, Co, and Ga. The weight loss after melting was found to be less than 0.3%. For homogenization, the samples were annealed in vacuum for 72 h at 900 °C and slowly cooled down to room temperature.

The magnetization measurements were performed on a superconducting quantum interference device (SQUID) made by Quantum Design, Inc. The measurements were performed in a temperature range of 5–400 K and in a magnetic field of up to 1 kOe. Direct current resistivity, using the four-probe method, was measured over the same temperature range as the magnetization measurements. To eliminate the contribution of thermoelectric effects, the current direction was reversed and an average of the voltage drops in each direction was taken. Thermal expansion was measured using high-resolution capacitance dialotometry method in the temperature range of 80–295 K.[9]

RESULT AND DISCUSSION

The temperature dependence of the magnetization in a magnetic field of 1 kOe for $Ni_2Mn_{1-x}Co_xGa$ $(0.08\leqslant x\leqslant 0.10)$ is shown in Fig. 1. Upon heating a sharp drop occurs

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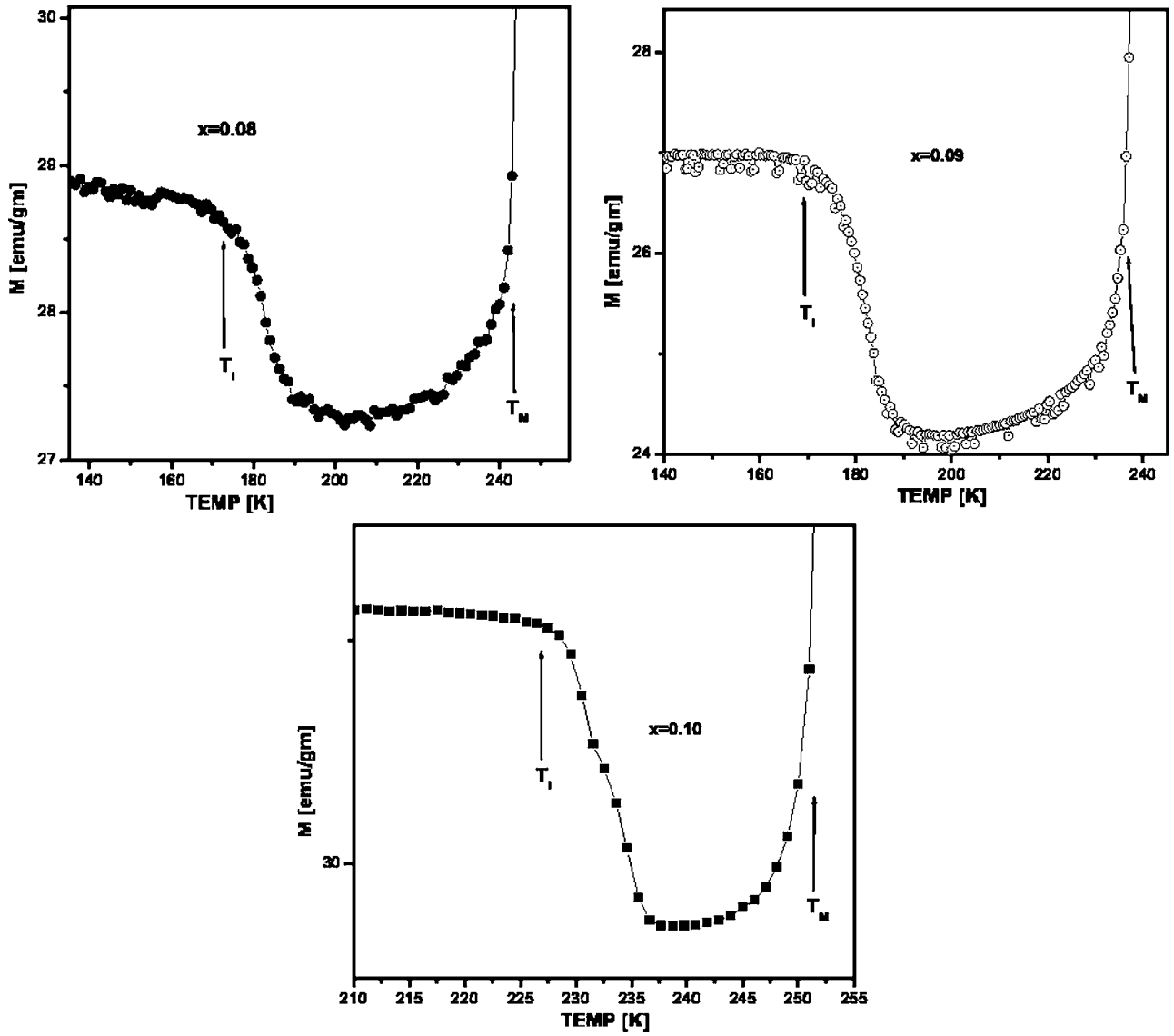

FIG. 1. The temperature variation of the magnetization $M(T)$ of $Ni_2Mn_{1-x}Co_xGa$ obtained at magnetic field of 1 kOe.

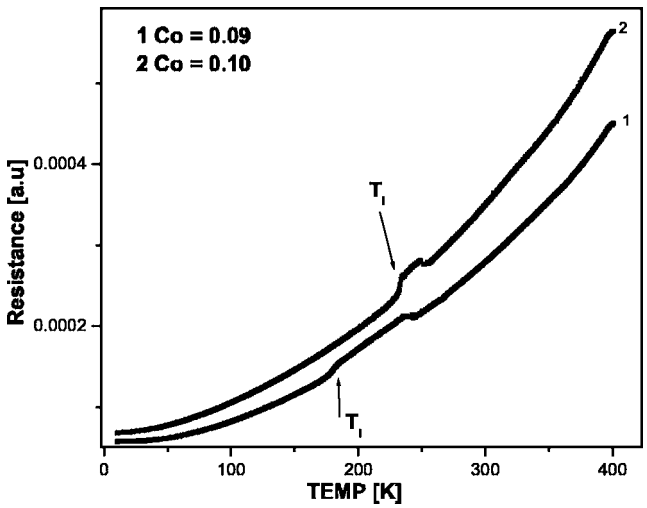

FIG. 2. The temperature variation of the resistance $R(T)$ of $Ni_2Mn_{1-x}Co_xGa$.

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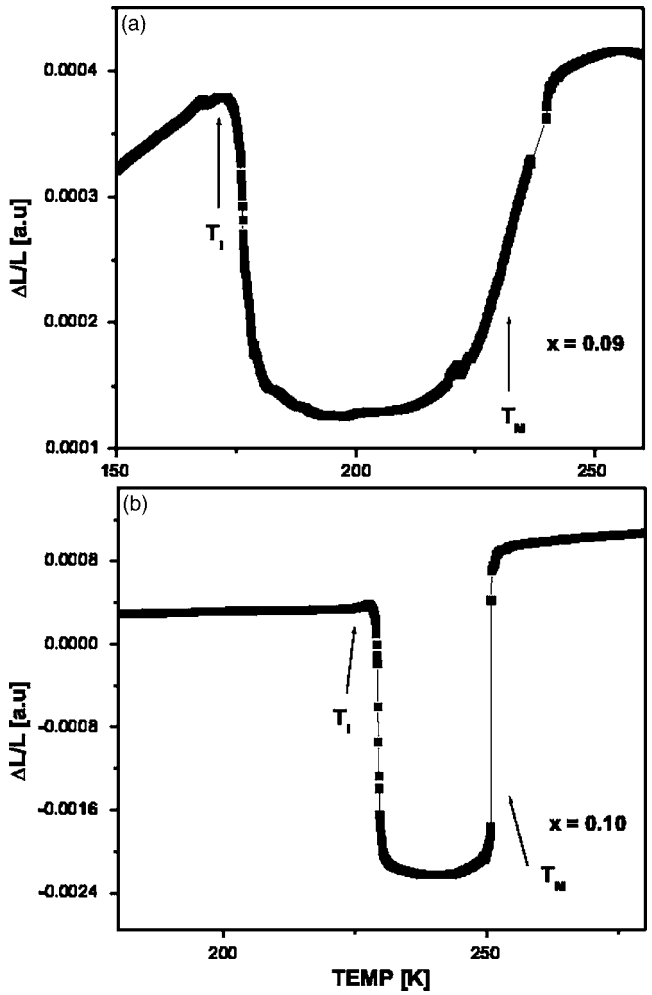

FIG. 3. Thermal expansion as a function of temperature of $Ni_2Mn_{1-x}Co_xGa$ (a) $x=0.09$ and (b) $x=0.10$.

in the M-T curve at T_I, and at the same temperature a steplike jump is observed in the resistivity curve (see Fig. 2). As usual for these alloys, upon further heating an abrupt increase of magnetization is observed at T_M, which is in accordance with a pronounced decrease in the resistivity curve. In order to verify the intermartensitic transformation further we performed thermal expansion measurements. The evidence of the intermartensitic transformation in $Ni_2Mn_{1-x}Co_xGa$ being a first order phase transition can be found from the thermal expansion curves, as shown in Fig. 3. Upon heating there is a sharp decrease in the thermal expansion curve. Such a steplike variation in the thermal expansion curve is very typical for a first order phase transition. The intermartensitic transformation starts appearing in the sample with $x=0.08$ and as the Co concentration increases, the transformation becomes more pronounced and the temperature range for which the alloys stay in the intermartensitic decreases as the Co concentration increases (see Fig. 3). As shown in Fig. 4, similarities were observed between the magnetization and thermal expansion curves.

According to Ref. 3, an average internal stress of 13.8 ± 1.08 MPa must be stored in the distorted lattice in order for an intermartensitic transformation to take place. It is therefore possible that the substitution of Co in the Mn sites in $Ni_2Mn_{1-x}Co_xGa$ results in an increase in the internal stress

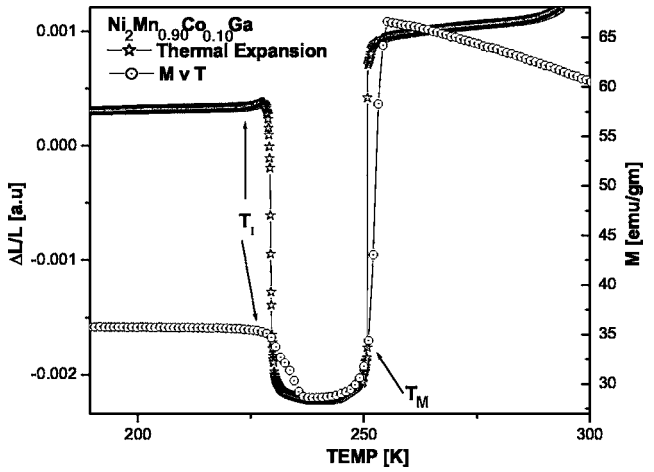

FIG. 4. Thermal expansion and magnetization as a function of temperature of $Ni_2Mn_{0.90}Co_{0.10}Ga$.

with increasing Co concentration. The minimum stress needed for the intermartensitic transformation to occur is produced at $x=0.08$, and the maximum internal stress at which the intermartensitic transformation can still exist is produced at $x=0.10$. Beyond $x=0.10$ the internal stress value exceeds the suggested value of 13.8 ± 1.08 and hence in Ref. 8 the intermartensitic transformation is no longer observed for $0.08\geqslant x\geqslant 0.10$.

CONCLUSION

The intermartensitic transformation in $Ni_2Mn_{1-x}Co_xGa$ system has been studied. The results of these experimental studies suggest that the $Ni_2Mn_{1-x}Co_xGa$ system undergoes an intermartensitic transformation for $0.08\leqslant x\leqslant 0.10$. The transformation is initially observed at $x=0.08$ and becomes more pronounced as the Co concentration increases. As presented in Ref. 8, for $x>0.10$ the intermartensitic transformation no longer exists in the $Ni_2Mn_{1-x}Co_xGa$ system. In light of previous experimental results, the intermartensitic transformation in this system could be due to the internal stress produced by the substitution of Co on the Mn sites. The minimum stress required to facilitate an intermartensitic transformation occurs at $x=0.08$, and the stress value exceeds the maximum stress allowable for an intermartensitic transformation to occur when $x>0.10$.

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