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Non-monotonous variation of structural instability in self-doped Ni–Mn–Sn based shape memory alloys



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ABSTRACT

A series of Ni-doped Ni_{50+x}Mn_{35-x}Sn₁₅ samples for x = -1, 0, 1, 2, 3 and 4 have been investigated by transport and dc magnetization measurements. The parent sample (x = 0) undergoes martensitic phase transition around 180 K during cooling along with ferromagnetic Curie point (T_c) to be 340 K. Interestingly, Ni doping at the Mn site leads to rather unusual non-monotonous variation of martensitic transition temperature (T_{MS}) which is precisely corroborated by the similar non-monotonous variation of lattice volume (V_{300K}). Between $-1 \le x \le 2$, the variation of various physical parameters in the studies alloys is conventional and can be explained on the basis of e/a ratio of the samples. However, for x > 2, parameters such as T_c , T_{MS} , V_{300K} etc. show sudden change in the nature of their variation which is anomalous and cannot be accounted by the e/a ratio formalism. Present study reveals that possible reason behind this non-monotonous variation is closely connected with a self-sustained mechanism originating from the change in the strength of hybridization between Ni $3d e_g$ state and the excess Mn at the Sn site on Ni substitution. The samples also show large negative magnetoresistance near T_{MS} with its magnitude showing similar non-monotonous variation with Ni doping.

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1. Introduction

Ni–Mn–Z (Z = Sb, In, Sn) based off-stoichiometric Heusler alloys have gained considerable attention in recent times due to their numerous magneto-functional properties [1–5]. These materials are all characterized by a shear dominated structural phase transition from the high temperature (*T*) austenite to low temperature martensite, known as martensitic phase transition (MPT). It is experimentally well established that the region across the MPT is highly sensitive to external parameters such as pressure and magnetic field (*H*) and this is the key factor towards their functionality [6–8]. The full Heusler compounds in this series (Ni₂MnZ or Ni₅₀Mn₂₅Z₂₅ in the present notation) do not undergo MPT. Structural instability can only be induced in off-stoichiometric alloys such as Ni₅₀Mn_{25+p}Z_{25-p} (for certain values of *p*), where excess Mn is doped at the Z site of the Heusler composition.

The electronic, magnetic and structural properties of these materials strongly depend upon the alloy composition. In particular, the MPT temperature in a specific series of alloys shows a steep

* Corresponding author. E-mail address: sspsm2@iacs.res.in (S. Majumdar). change with valence electron concentration (e/a ratio), which implies importance of electronic band filling in the material. Apart from e/a ratio, T_{MS} is also found to be related to the lattice volume of the cubic austenite [9] which in its turn signifies the importance of Mn–Mn magnetic interaction towards martensitic instability. In these off-stoichiometric alloys, Mn can occupy either its regular site (we denote it by Mn^Y) or the Z site (denoted by Mn^Z). The magnetic interaction between Mn^Y-Mn^Z is found to be antiferromagnetic (AFM) below a critical separation of 3 Å [10], and the signature of AFM correlation between Y and Z sites are established below MPT [11–13].

It is to be noted that Ni plays an important role in deciding the electronic as well as the structural properties of the alloys. Ni-3*d* level hybridizes with the excess Mn at the Z site to produce a peak structure in the minority spin states near Fermi level and such peak structure is intimately related to the observed MPT in the materials [10,14].

Considering the importance of both Mn and Ni in these offstoichiometric alloys, we studied the effect of introducing Ni at the expense of Mn in the parent alloy $Ni_{50}Mn_{35}Sn_{15}$. Though doping with transition metal at the Mn site other than the parent one (*i.e.* Ni) is well documented in the literature [15–17], such self doping



study is relatively uncommon among Ni–Mn–Z based alloys [18,19]. The parent alloy undergoes MPT at about 180 K along with ferromagnetic (FM) Curie point $T_C = 340$ K and it is a metamagnetic shape memory alloy (MSMA) showing large magnetoresistance (MR) and magnetocaloric effect, which arise due to magnetic field induced phase transition (metamagnetism) [20]. The primary goal of Ni variation was to see how magnetism and T_{MS} can get affected by Ni concentration and subsequently understand the role of Ni in deciding the ground state properties of the alloys. In the present work we have prepared and investigated Ni_{50+x}Mn_{35-x}Sn₁₅ samples with x = -1, 0, 1, 2, 3 and 4.

2. Experimental details

Polycrystalline samples of Ni-Mn-Sn alloys of nominal compositions $Ni_{50+x}Mn_{35-x}Sn_{15}$ (*x* = -1, 0, 1, 2, 3 and 4) were prepared by argon arc melting the constituent metals on a water-cooled copper hearth. In order to get proper homogeneity, the samples were remelted several times turning back to back. The arc molted ingots were vacuum sealed in a quartz tube and annealed at 900 °C for 45 h followed by a rapid ice water quenching [3,21]. Room temperature powder x-ray diffraction (XRD) data were recorded on the samples using Cu K_{α} radiation. The zero-field and in-field resistivity (ρ) were measured by four-probe method on a homemade setup fitted in a nitrogen cryostat (between 77 and 400 K) as well as on a cryogen-free high magnetic field system (Cryogenic Ltd., U.K.) with *H* as high as 90 kOe and within the *T* range between 5 and 300 K. The magnetic measurements were performed on a vibrating sample magnetometer from Cryogenic Ltd. UK and by a commercial Quantum Design SQUID magnetometer (MPMS XL Ever Cool model).

3. Results

3.1. Powder X-ray diffraction

The room temperature powder XRD patterns of the studied samples are shown in the main panel of Fig. 1. All the alloys are found to crystallize in cubic Heusler structure with $L2_1$ ordering. The signature of $L2_1$ ordering is evident from the observation of reflections such as (111), (311), (331) etc. [22] Even after large Ni doping, the samples retain the parent ordering as no extra unindexable reflection was found in the XRD patterns. Since Ni has



Fig. 1. XRD patterns for x = -1, 1, 2, 3 and 4 recorded at room temperature using Cu K α radiation. Inset shows variation of a_c with x.

smaller covalent radius than Mn, Ni doping at the expense of Mn is supposed to reduce the cubic lattice parameter (a_c). However, the observed room temperature a_c is found to show rather unusual variation with x. Initially with increasing Ni concentration, a_c decreases up to x = 2 and further doping causes increase in a_c . Variation of a_c with doping concentration is plotted in the inset of Fig. 1 and the values are found to be between 5.963 Å and 5.983 Å having a maximum deviation of ± 0.002 Å. Such non-monotonous variation of a_c keeping the overall symmetry unchanged may indicate an isostructural phase transition in the system as a function of doping concentration. Such transition as a function of x takes place in the austenite phase as evident from the variation of a_c at 300 K.

3.2. Magneto-transport

We measured the thermal variation of ρ in zero field as well as in presence of 50 kOe applied field for all the samples and the data have been shown for two representative samples (x = 2 and 4) in Fig. 2 (a) and (b) respectively. Clear signature of MPT is observed in all the samples which is typically characterized by large increase in ρ on cooling along with the signature of thermal hysteresis. The MPT start temperature (T_{MS}) is denoted by the point where ρ increases sharply. The FM T_C is also evident from the change in slope. The values of T_{MS} are tabulated in Table 1 (in the later part of the text) for all the compositions, and interestingly T_{MS} shows a nonmonotonous variation with Ni concentration. With initial Ni doping T_{MS} increases and attains its maximum value of 240 K for x = 2. On further increase of Ni, T_{MS} starts to drop and attains a value



Fig. 2. (a) and (b) shows thermal variation of ρ for x = 2 and 4 samples (both during heating and cooling) respectively in presence of H = 0 and 50 kOe. Ferromagnetic T_C and T_{MS} are indicated by arrows. Insets in (a) and (b) show M vs T data for FC and FCH protocols of the same samples respectively.

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