

Magnetic interactions in the martensitic phase of Mn rich Ni-Mn-In shape memory alloys

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The magnetic properties of $\text{Mn}_2\text{Ni}_{(1+x)}\text{In}_{(1-x)}$ ($x = 0.5, 0.6, 0.7$) and $\text{Mn}_{(2-y)}\text{Ni}_{(1.6+y)}\text{In}_{0.4}$ ($y = -0.08, -0.04, 0.04, 0.08$) shape memory alloys have been studied. Magnetic interactions in the martensitic phase of these alloys are found to be quite similar to those in $\text{Ni}_2\text{Mn}_{(1+x)}\text{In}_{(1-x)}$ type alloys. Doping of Ni for In not only induces martensitic instability in Mn_2NiIn type alloys but also affects magnetic properties due to a site occupancy disorder. Excess Ni preferentially occupies X sites forcing Mn to the Z sites of X_2YZ Heusler composition resulting in a transition from ferromagnetic ground state to a state dominated by ferromagnetic Mn(Y)-Mn(Y) and antiferromagnetic Mn(Y)-Mn(Z) interactions. These changes in magnetic ground state manifest themselves in observation of exchange bias effect even in zero field cooled condition and virgin magnetization curve lying outside the hysteresis loop. © 2013 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4829278>]

I. INTRODUCTION

Interest in ferromagnetic shape memory alloys (FSMAs) due to their technological potential has generated much attention in recent years.¹⁻³ These alloys have the generic formula X_2YZ where the X and Y atoms are 3d elements, while Z is a group IIIA-VA element. They undergo a diffusionless transformation from high temperature austenitic cubic structure to low temperature (martensitic) tetragonal or orthorhombic structure.³ One such system extensively studied is Ni_2MnGa which undergoes martensitic transformation at $T_M \sim 220$ K. Martensitic transformation can also be induced in $Z = \text{In, Sn}$ or Sb but in off-stoichiometric compositions, $\text{Ni}_2\text{Mn}_{(1+x)}\text{Z}_{(1-x)}$.⁴ Apart from the martensitic transformation, these alloys exhibit other interesting properties like large magnetocaloric effects, magnetic superelasticity⁴⁻⁷ and magnetic field induced giant strains,^{8,9} exchange bias,^{10,11} etc.

Alloys of the type Mn_2NiZ have higher Mn content compared to the traditional Ni_2MnZ and is considered beneficial in realizing better magnetic, magnetocaloric, and magnetotransport properties.¹²⁻¹⁴ Mn_2NiGa is one such Mn rich alloy which undergoes martensitic transformation at $T_M \sim 270$ K in an ferrimagnetically ordered state ($T_C = 588$ K).¹⁵ Ferrimagnetic order has also been confirmed from band structure calculations and arise due to unequal magnetic moments of antiferromagnetically coupled Mn atoms occupying the X and Y sites of X_2YZ Heusler structure.¹⁶⁻²⁰ Although martensitic transformations have been theoretically predicted in other Mn_2NiZ ($Z = \text{In, Sn, Sb}$) alloys, experimental investigations have reported these alloys to have stable crystal structures.^{21,22} However, just as

in case of Ni_2MnZ alloys partial substitution of Z atoms by Mn results in martensitic instability increasing Ni content at the expense of Z atoms in Mn_2NiZ results in martensitic alloys.^{23,24} However, realization of such alloys with general composition $\text{Mn}_2\text{Ni}_{(1+x)}\text{Z}_{(1-x)}$ can lead to a structural disorder due to site preferences of transition metal ions. In a $L2_1$ Heusler composition Ni atoms prefer X sites as compared to Z sites²⁵ and therefore doping excess Ni could result in newer magnetic interactions as it would force Mn to occupy the Z sites. These conditions can change the sign of RKKY interaction leading to magnetic frustration or a new type of magnetic order. Local structural disorder is shown to be primarily responsible for martensitic transformation and magnetic interactions in the martensitic state of $\text{Ni}_2\text{Mn}_{1+x}\text{In}_{1-x}$ type alloys.²⁶⁻²⁸ In case of Mn_2NiGa as well, site occupancy disorder has been shown to be an important factor in explaining magnetic properties of the martensitic state.²⁹ The antisite disorder is also shown to be responsible for the zero field cooled exchange bias in Mn_2PtGa .³⁰

In this paper, we focus our attention on understanding the magnetic properties of $\text{Mn}_2\text{Ni}_{(1+x)}\text{In}_{(1-x)}$ ($x = 0.5, 0.6, 0.7$) and $\text{Mn}_{(2-y)}\text{Ni}_{(1.6+y)}\text{In}_{0.4}$ ($-0.08 \leq y \leq 0.08$) type alloys especially in their martensitic state. By comparing the magnetic properties of martensitic and structurally stable alloys having nearly similar compositions, we show that low temperature magnetic properties of the martensitic alloys are dominated by ferromagnetic and antiferromagnetic interactions, arising due to site occupancy disorder in these Mn rich alloys.

II. EXPERIMENTAL

The samples of above composition were prepared by arc melting the weighed constituents in argon atmosphere followed by encapsulating in a evacuated quartz tube and

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