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Ab initio study of effect of Co substitution on the magnetic properties of Ni and Pt-based Heusler alloys

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ABSTRACT

Using density functional theory based calculations, we have carried out in-depth studies of effect of Co substitution on the magnetic properties of Ni and Pt-based shape memory alloys. We show the systematic variation of the total magnetic moment, as a function of Co doping. A detailed analysis of evolution of Heisenberg exchange coupling parameters as a function of Co doping has been presented here. The strength of RKKY type of exchange interaction is found to decay with the increase of Co doping. We calculate and show the trend, how the Curie temperature of the systems vary with the Co doping.

1. Introduction

Full Heusler alloys (with typical formula A_2BC) have drawn considerable attention of the researchers over the last decades because of their possible technological applications. Upon cooling, some of the Heusler alloys undergo a structural transition from a high temperature cubic phase, namely austenite phase to a lower symmetry phase, called martensite phase below a certain temperature. This type of structural transition is referred as martensite transition, and the particular temperature at which the transition takes place is called martensite transition temperature. Ni_2MnGa belongs to this category of Heusler alloys [1–3]. The Heusler alloys of this category may find their application as various devices, such as actuator, antenna, sensor etc. For the application purpose it is always desired that martensitic transition temperature is above the room temperature. In case of conventional shape memory effect, which is governed by the temperature, the actuation process is much more slow compared to a magnetically controlled actuation. So it is desirable to have a magnetic shape memory alloy with the Curie temperature (T_C) higher than the room temperature. It has been observed that both T_M and T_C values are very much dependent on the composition of a particular Heusler alloy [4–16].

There is also another category of full Heusler alloys, which are known to be metallic for one kind of spin channel and insulator for the other kind of spin channel because of their very high spin polarization (HSP) at the Fermi level. They are often called as half metallic Heusler alloys [17]. Most of the Co-based Heusler alloys, like Co_2MnSn, Co_2MnGa belong to this category [18,19]. These Heusler alloys may have potential application in spintronic devices.

Apart from the technological application, these Heusler alloys are very interesting because of their wide diversity in terms of magnetic property. These alloys may be ferromagnetic, ferrimagnetic, anti-ferromagnetic and also non-magnetic depending on the chemical composition. So it is of immense interest to have an in depth study on the magnetic interactions present in these systems. In most of the full Heusler alloys, A_2BC, B is the primary moment carrying atom. In many of the Heusler alloys, A_2BC, there is presence of a delocalized-like common d-band formed by the d-electrons of the A and B atoms, which are both typically first-row transition metal atoms [20]. Additionally, there is also an indirect RKKY-type exchange mechanism [21] between the B atoms, primarily mediated by the electrons of the C atoms, which also plays an important role in defining the magnetic properties of these materials [20,22]. Staunton et al. [23] reported the role of RKKY interaction behind the origin of magnetic anisotropy of a system. For the magnetic shape memory alloys, magnetic anisotropy energy plays an important role. In this regard also, it will be interesting to study the RKKY interaction in detail in these systems.

In a very recent paper, we have shown in detail the similarities and differences between the Heusler alloys which are likely to show shape memory alloy (SMA) property and which are not, in terms of the electronic, magnetic as well as mechanical prop-

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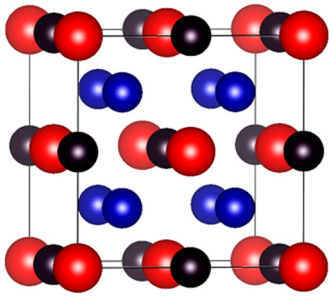

Fig. 1. (Color online.) Structure of conventional Heusler alloy (A_2BC). Blue, black and red balls represent *A*, *B* and *C* atoms respectively.

erties [14]. The main focus of our study here is to show how the magnetic exchange interactions of *B* atom between *B* atom itself, and with other magnetic atoms of the A_2BC systems, are evolving in going from the materials which are prone to martensite transition (which are generally metallic in nature) to the other class of Heusler alloys (which are typically half-metallic in nature) i.e. which do not show SMA property. Here we study about the nature of indirect type RKKY interaction and direct exchange interaction as well, for four sets of materials $Ni_{2-x}Co_xMnGa$, $Ni_{2-x}Co_xFeGa$, $Pt_{2-x}Co_xMnGa$, $Pt_{2-x}Co_xMnSn$ as a function of x (x = 0.00, 0.25, 0.50, 0.75, 1.25, 1.50, 1.75, 2.00). In all the cases the material is likely to show SMA property for x = 0.00 and is predicted to be half-metallic for x = 2.00. In the section following the methodology, the results of the work and the relevant discussion are presented. Finally, we summarize and conclude in the last section.

2. Method

The Heusler alloys (A_2BC) studied here possess $L2_1$ structure that consists of four interpenetrating face-centered-cubic (fcc) sublattices with origin at fractional positions, (0.25, 0.25, 0.25), (0.75, 0.75, 0.75), (0.5, 0.5, 0.5), and (0.0, 0.0, 0.0). For the conventional Heusler alloy structure, the first two sub-lattices are occupied by *A* atom and the third by *B* and fourth by *C* atom. In total, there are 16 atoms in the cell. Fig. 1 depicts the structure of a conventional Heusler alloy. While we study the Co substitution in A_2BC systems, the Co atom substitutes the *A* atom only. First we carry out full geometry optimization of the materials, of all the materials corresponding to x = 0.00, 0.25, 1.75, 2.00, using the 16 atom cell. For the geometry optimization, we employ the Vienna Ab Initio Simulation Package (VASP) [24] in combination with the projector augmented wave method [25]. We use an energy cutoff of minimum 500 eV for the planewave basis set. The calculations have been performed with a *k* mesh of 15 × 15 × 15. The energy and force tolerance used were 10 μeV and 10 meV/Å, respectively. After obtaining the equilibrium lattice constants of the four above-mentioned materials by using the VASP package, we plot the same. A linear variation of the lattice constant is observed. We deduce the lattice constants of the other materials, corresponding to x = 0.50, 0.75, 1.25, 1.50 by the method of interpolation. To gain insight into the magnetic interactions of these materials, we calculate and discuss their Heisenberg exchange coupling parameters. We use the spin-polarized-relativistic Korringa–Kohn–Rostoker method (SPR-KKR) to calculate the Heisenberg exchange coupling parameters, J_{ij}, as implemented in the SPR-KKR programme package [26]. The mesh of *k* points for the SCF cycles has been taken as 21 × 21 × 21 in the BZ. The angular momentum expansion for each atom is taken such that $l_{max} = 3$. The partial and total moments have also been calculated for all the materials studied. We use local density approximation (LDA) for exchange correlation functional [27]. For composition with x ≠ 0 or 2 we use SPR-KKR method with coherent potential approximation.

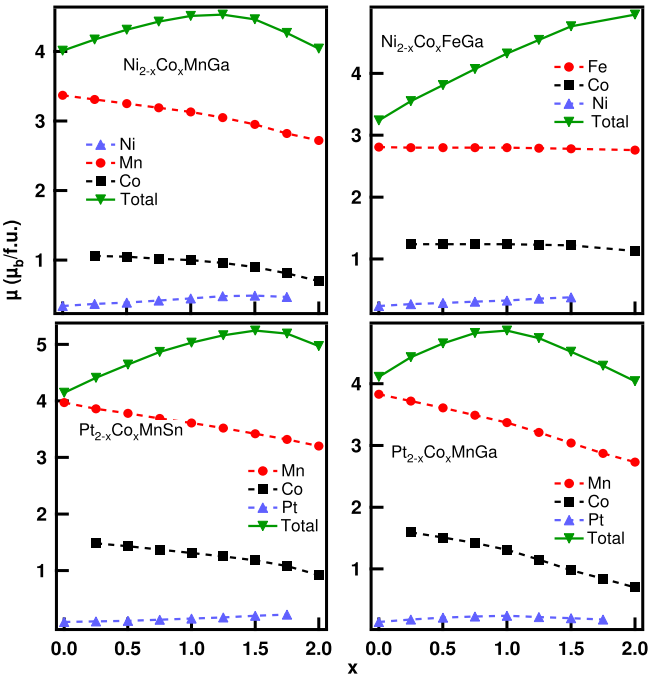

Fig. 2. (Color online.) x dependence of magnetic moments for $Ni_{2-x}Co_xMnGa$, $Ni_{2-x}Co_xFeGa$, $Pt_{2-x}Co_xMnSn$, $Pt_{2-x}Co_xMnGa$. The line is only guide to the eyes.

The calculation of Heisenberg exchange coupling parameter (J_{ij}) is based on the real space approach following the method proposed by Lichtenstein et al. [28]. It involves the magnetic force theorem to evaluate the Heisenberg exchange coupling parameter

$$J_{ij} = \frac{1}{4\pi}\int^{E_F} dE Im Tr_L\{\Delta_i \tau_{\uparrow}^{ij} \Delta_j \tau_{\downarrow}^{ij}\} \quad (1)$$

where τ is the scattering path operator. Difference in the inverse single site t matrices of up and down spin has been presented by Δ. Tr_L represents the trace of scattering matrices over the orbital indices L(l, m). The calculation of J_{ij} has been carried out with a cluster radius of 3a, a is the lattice parameter of the particular system. T_C has been calculated from Heisenberg exchange coupling parameters using mean field approximation [29]. Convergence of the T_C has been tested with respect to cluster radius.

For the calculation of density of states using SPR-KKR, we use following parameters NKTAB = 2500, NE = 550, ImE = 0.001 Ry [30].

3. Results and discussion

Total and partial moments As mentioned above, we studied here four sets of materials, $Ni_{2-x}Co_xMnGa$, $Ni_{2-x}Co_xFeGa$, $Pt_{2-x}Co_xMnGa$, $Pt_{2-x}Co_xMnSn$ with x = 0.00, 0.25, 0.50, 0.75, 1.25, 1.50, 1.75, 2.00. At the two ends of the composition, i.e. for x = 0.00 and x = 2.00, all the materials except Pt_2MnSn are already reported in the literature. We note that, all the materials, corresponding to x = 0.00, are likely to exhibit martensite transition. We predict here that Pt_2MnSn also possesses conventional Heusler alloy structure in its ground state and exhibits the martensite transition. All the studied materials here are taken to be ferromagnetic in nature.

We observe from Fig. 2 that for the three sets of materials namely, $Ni_{2-x}Co_xMnGa$, $Pt_{2-x}Co_xMnGa$, $Pt_{2-x}Co_xMnSn$ the variation of total moment (μ_T) follows the same trend, which is for lower value of x, μ_T increases and then starts to fall at a higher x value, attaining a maximum value in between the range of

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