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Journal of Magnetism and Magnetic Materials 290–291 (2005) 385–387
Journal of magnetism and magnetic materials
www.elsevier.com/locate/jmmm

Magnetic exchange coupling and Curie temperature of $Ni_{(1+x)}MnSb(x = 0, 0.25, 0.5, 0.75, 1)$ from first principles

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Available online 10 December 2004

Abstract

We study the dependence of magnetic interactions and Curie temperature in $Ni_{(1+x)}MnSb$ system on the Ni concentration within the framework of the density-functional theory. The calculation of the exchange parameters is based on the super-cell and frozen-magnon approaches. The Curie temperatures, T_C, are calculated within the random-phase approximation. In agreement with experiment we obtain decrease of the Curie temperature with increasing Ni content.

PACS: 75.50.Cc; 75.30.Et; 71.15.Mb

Keywords: Heusler alloys; Ferromagnetism; Electronic structure; Curie temperature; Exchange parameter

Recently rapid development of spin electronics intensified the researches on the ferromagnetic materials that are suitable for the spin injection into a semiconductor [1]. One of the promising classes of these materials is the Heusler alloys. Heusler alloys have been intensively studied theoretically [2,3] and experimentally [4,5] as possible sources of spin-polarized carriers for spintronics applications. Among the properties useful for the applications are high Curie temperature, high electron spin polarization at the Fermi level and very small lattice mismatch with widely employed semiconductors (e.g., Ni_2MnIn and InAs) [6]. Some of the Heusler compounds were found to have half-metallic ground state [2] which is characterized by a 100% carrier spin-polarization. An interesting combination of physical properties makes Heusler alloys the subject of intensive experimental and theoretical investigations [7–9].

In the present work we report the theoretical study of the exchange interactions and Curie temperature of $Ni_{(1+x)}MnSb$ system as a function of Ni concentration. In particular, we focus on the effect of local environment of Mn atoms on half-metallicity and exchange interactions. We show that, occupation of vacant sites in NiMnSb by Ni immediately leads to disappearance of the half-metallicity and to substantial decrease of both inter-sublattice (Mn–Ni) and intra-sublattice (Mn–Mn) exchange interactions.

The calculations are carried out with the augmented spherical waves (ASW) method [10] within the generalized gradient approximation (GGA) [11] for the exchange-correlation potential. In all calculations the experimental values of the lattice parameters are used [12]. The radii of all atomic spheres are chosen to be equal [13]. We use cubic supercell for $0.25 \leqslant x \leqslant 0.75$.

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0304-8853/$ - see front matter
doi:10.1016/j.jmmm.2004.11.267

We describe the interatomic exchange interactions in terms of the classical Heisenberg Hamiltonian

$$H_{\text{eff}} = -\sum_{\mu,\nu} \sum_{\substack{\mathbf{R},\mathbf{R}' \\ (\mu\mathbf{R}\neq\nu\mathbf{R}')}} J^{\mu\nu}_{\mathbf{R}\mathbf{R}'} \mathbf{s}^{\mu}_{\mathbf{R}} \mathbf{s}^{\nu}_{\mathbf{R}'}. \quad (1)$$

In Eq. (1), the indices μ and ν number different sublattices and $\mathbf{R}$ and $\mathbf{R}'$ are the lattice vectors specifying the atoms within sublattices, $\mathbf{s}^{\mu}_{\mathbf{R}}$ is the unit vector pointing in the direction of the magnetic moment at the site $(\mu, \mathbf{R})$.

We employ the frozen-magnon approach [14,15] to calculate interatomic Heisenberg exchange parameters. The calculations involve few steps. In the first step, the exchange parameters between the atoms of a given sublattice μ are computed. The calculation is based on the evaluation of the energy of the frozen-magnon configurations defined by the following atomic polar and azimuthal angles:

$$\theta^{\mu}_{\mathbf{R}} = \theta, \quad \phi^{\mu}_{\mathbf{R}} = \mathbf{q}\cdot\mathbf{R} + \phi^{\mu}. \quad (2)$$

The constant phase ϕ^{μ} is always chosen equal to zero. The magnetic moments of all other sublattices are kept parallel to the z-axis. Within the Heisenberg model (1) the energy of such configuration takes the form

$$E^{\mu\mu}(\theta, \mathbf{q}) = E^{\mu\mu}_{0}(\theta) + \sin^2\theta\, J^{\mu\mu}(\mathbf{q}), \quad (3)$$

where $E^{\mu\mu}_{0}$ does not depend on $\mathbf{q}$ and the Fourier transform $J^{\mu\nu}(\mathbf{q})$ is defined by

$$J^{\mu\nu}(\mathbf{q}) = \sum_{\mathbf{R}} J^{\mu\nu}_{0\mathbf{R}} \exp(i\mathbf{q}\cdot\mathbf{R}). \quad (4)$$

In the case of $\nu = \mu$ the sum in Eq. (4) does not include $\mathbf{R} = 0$. Calculating $E^{\mu\mu}(\theta, \mathbf{q})$ for a regular $\mathbf{q}$-mesh in the Brillouin zone of the crystal and performing back Fourier transformation one gets exchange parameters $J^{\mu\mu}_{0\mathbf{R}}$ for sublattice μ.

The determination of the exchange interactions between the atoms of two different sublattices μ and ν is discussed in Ref. [16].

The Curie temperature is estimated within the random phase approximation (RPA)

$$\frac{1}{k_{\text{B}} T_{\text{c}}^{\text{RPA}}} = \frac{6\mu_{\text{B}}}{M}\frac{1}{N}\sum_{q}\frac{1}{\omega(\mathbf{q})}, \quad (5)$$

where $\omega(\mathbf{q})$ is the spin-wave dispersion.

We begin with the discussion of the structural properties. NiMnSb and Ni_2MnSb crystalize in $C1_b$-type and in $L2_1$-type structures, respectively. According to our calculations for the intermediate compositions $(0<x<1)$ lower total energy corresponds to the $C1_b$-type structure with one Ni sublattice completely and another Ni sublattice partially filled. This result is in good agreement with the experiment [12].

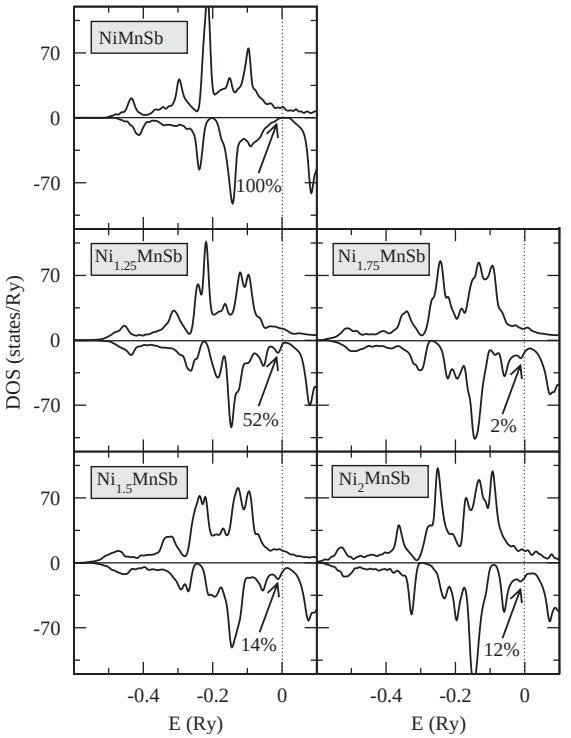

Fig. 1. Spin-projected total DOS of $Ni_{(1+x)}MnSb$. The spin polarization at the Fermi level is shown.

We present in Fig. 1, calculated total density of states (DOS). NiMnSb is found to have half-metallic ground state which is characterized by a 100% carrier spin-polarization at the Fermi level. The occupation of vacant sites in NiMnSb by Ni atoms destroys the half-metallicity and reduces the spin polarization. The loss of the half-metallicity with increasing Ni concentration can be attributed to the increased Mn–Ni hybridization [3]. In Table 1 we present calculated magnetic moments. The Mn and Ni[1] magnetic moments decreases with increasing Ni concentration. On the other hand the Ni[2] moment increases. Total magnetic moment slightly decreases.

The calculated Heisenberg exchange parameters are presented in Fig. 2. The decrease in the experimental Curie temperatures with increasing Ni concentration is in good agreement with the calculated parameters. Transition from NiMnSb to Ni_2MnSb leads to a strong decrease in all exchange parameters except Mn–Ni[2] which increases slightly. Although the symmetry of the system is reduced at the intermediate compositions, the symmetry of the exchange parameters at small $(x\leqslant 0.25)$ and large $(x\geqslant 0.75)$ values of x is not affected by this reduction. Only at $x = 0.5$ the pattern of exchange interactions shows anisotropic behavior appeared as a splitting in nearest and next-nearest neighborhood

Table 1
Magnetic moments (in μ_B) in $Ni_{(1+x)}MnSb$

	Ni^1	Ni^2	Mn	Total
NiMnSb	0.19	—	3.86	4.000
$Ni_{1.25}MnSb$	(0.20–0.21)	−0.04	3.82	3.971
$Ni_{1.5}MnSb$	(0.19–0.21)	0.03	3.81	3.976
$Ni_{1.75}MnSb$	(0.18–0.19)	0.08	3.76	3.956
Ni_2MnSb	0.14	0.14	3.695	3.945

Ni^1 and Ni^2 denote first and second Ni sublattices, respectively. Different values of magnetic moments in Ni^1 column corresponds to inequivalent atoms in first Ni sublattice.

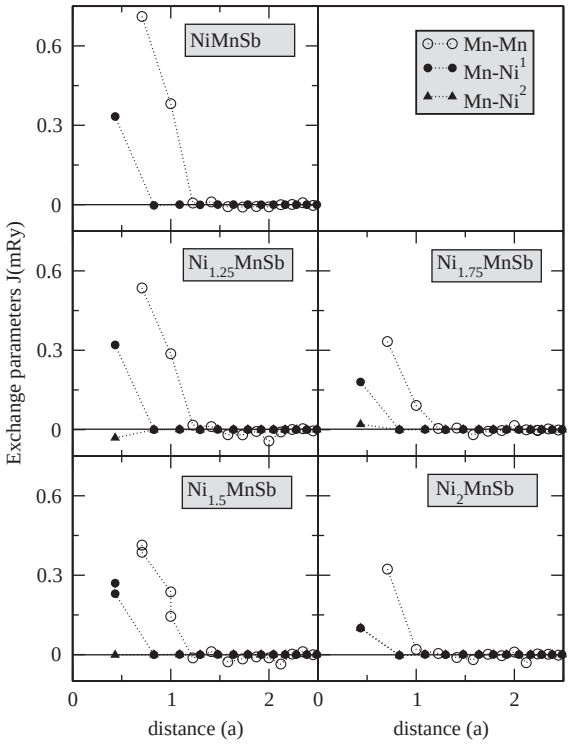

Fig. 2. Intra-sublattice (Mn–Mn) and inter-sublattice (Mn–Ni) exchange interactions in $Ni_{(1+x)}MnSb$. The distances are given in the units of the lattice constant.

exchange parameters. The splitting in further nearest neighborhoods can be neglected.

The interatomic exchange parameters are used to evaluate the Curie temperature. We take only the Mn–Mn interactions and neglect Mn–Ni contribution. As it was shown in Ref. [16] the contribution of the Mn–Ni exchange interaction to the Curie temperature of Ni_2MnSb is much less important than Mn–Mn contribution. The calculated Curie temperatures are presented in Table 2. For low x ($x \leqslant 0.5$) the values of T_C are in good agreement with the experiment. For large x, the calculations underestimate T_C.

Table 2
RPA estimation of the Curie temperature in $Ni_{(1+x)}MnSb$

	T_c(RPA)	T_c(Exp)
NiMnSb	830	730
$Ni_{1.25}MnSb$	550	570
$Ni_{1.5}MnSb$	364	470
$Ni_{1.75}MnSb$	254	405
Ni_2MnSb	190	365

Experimental values of the Curie temperatures are taken from Ref. [12].

In conclusion, we have systematically studied the dependence of magnetic interactions and Curie temperature in $Ni_{(1+x)}MnSb$ system on the Ni content within the parameter-free density functional theory. Our calculations show that Ni substitution destroys the half-metallicity and substantially decreases both inter-sublattice (Mn–Ni) and intra-sublattice (Mn–Mn) exchange interactions. The Curie temperatures are calculated within the RPA to the statistical mechanics of the classical Heisenberg Hamiltonian. In agreement with experiment, we obtain decrease of the Curie temperature with increasing Ni content.

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