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Role of the magnetic impurities in $Ni_2Ti_{1-x}Mn_xSn$ Heusler type alloys. Ab-initio calculations

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We have studied electronic and magnetic properties of $Ni_2Ti_{1-x}Mn_xSn$ Heusler type alloys. The calculations were carried out within the density functional theory using ab-initio TB LMTO-ASA method. We found that $Ni_2Ti_{1-x}Mn_xSn$ becomes magnetic for a small manganese concentration $x = 0.0625$.

Key words: *electronic structure; magnetic moment; Heusler alloy; LMTO*

1. Introduction

The electronic and magnetic properties of Ni_2TiSn and Ni_2MnSn Heusler type alloys have been studied in the last years experimentally and theoretically [1–8]. An interesting physical problem is the dependence of the magnetic moment on the distribution of atoms in the unit cell as well as the influence of the impurities on the magnetic and electronic properties. Ni_2TiSn is a Pauli paramagnet, however, in Ni_2MnSn the total magnetic moment is close to 4.0 μ_B [8]. We have analyzed the role of the magnetic impurities on the electronic structure and magnetic properties of the of $Ni_2Ti_{1-x}Mn_xSn$ Heusler type alloys by ab-initio TB LMTO-ASA [9, 10] method. The recent ab-initio calculations [1–3, 5–6] have shown that chemical and atomic disorder strongly modify the electronic and magnetic properties of Heusler-type alloys. In this work, we present the electronic and magnetic properties of $Ni_2Ti_{1-x}Mn_xSn$ as a function of Mn concentration.

2. The model and method of calculations

The electronic and magnetic properties were calculated using the spin polarized tight binding linearised muffin tin orbital (TB LMTO) method [9, 10] within the

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atomic sphere approximation (ASA). The band calculations were performed for the theoretical lattice parameters estimated from the minimum of the total energy (Table 1).

Table 1. Lattice parameters for $Ni_2Ti_{1-x}Mn_xSn$ alloys

Parameter	Mn concentration x				
	0.00	0.25	0.50	0.75	1.00
Exp. lattice parameter [a.u.] [4]	11.47	11.45	11.44	11.43	11.42
Lattice parameter para [a.u.]	11.46	11.44	11.42	11.32	11.24
Lattice parameter ferro [a.u.]	11.55	11.32	10.88	10.76	10.39

In the TB LMTO-ASA method, the atomic spheres were chosen in such a way that the sum of the volumes of all the atomic spheres of the unit cell and the overlapping of the atomic spheres was less then 10%. The exchange correlation potential was assumed in the form proposed by von Barth and Hedin [11] and we included in the calculations also the non-local corrections [12]. In the LMTO method, the scalar relativistic approximation for the band electrons and the fully relativistic treatment of the frozen core electrons were assumed during the self-consistent band calculations. The band calculations were performed for 256 k points in the irreducible wedge of the Brillouin zone. Ni_2TiSn and Ni_2MnSn crystallize into $L2_1$ type structure consisting of four interpenetrating *fcc* sublattices. Each *fcc* sublattice can be divided into four simple cubic lattices and hence the $L2_1$ type structure can be considered as a 16-atom unit cell. In the case of the spin polarized system, we have performed the band calculations for 32- and 64-atom unit cells in order to get a smaller value of Mn concentration $x = 0.125$ and $x = 0.0625$, respectively. The titanium and manganese atoms are distributed randomly in Ti–Mn sublattices.

3. Results

Ab-initio band calculations were performed for paramagnetic and ferromagnetic $Ni_2Ti_{1-x}Mn_xSn$ alloys. The theoretical lattice parameters were estimated from the minimum of the total energy and the values are listed in Table 1 for paramagnetic and ferromagnetic systems. The theoretical values of the lattice parameters for the paramagnetic alloy are very close to the experimental results, however, the values obtained for the ferromagnetic systems are lower than the experimental ones. In the supercell model, Ti and Mn atoms were distributed in different sites of Ti–Mn sublattice. The density of states was calculated for the distribution of atoms for which the total energy had a minimum. In Figure 1, we present the total density of states for paramagnetic $Ni_2(Ti_{1-x}Mn_x)Sn$ alloys for $x = 0.0$, 0.25, 0.50, 0.75 and 1.0. The density of states at the Fermi energy $N(E_F)$ increases with the increasing Mn concentration.

We have also performed the spin-polarized calculations for the theoretical lattice parameters (Table 1) from which we estimated the magnetic moment.

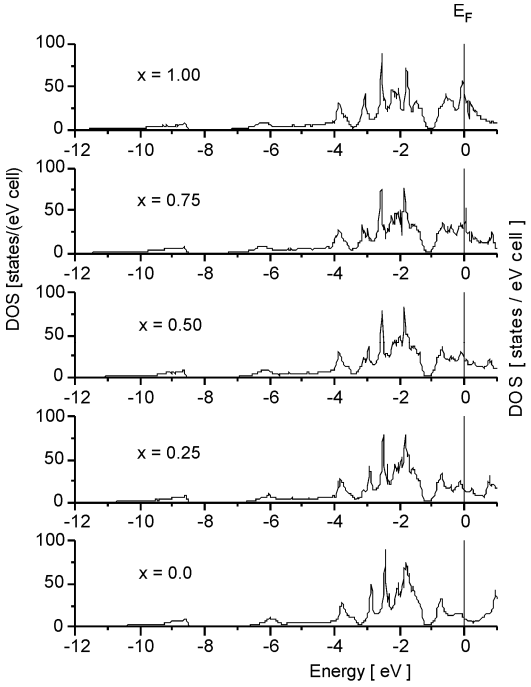

Fig. 1. The total density of states for paramagnetic $Ni_2Ti_{1-x}Mn_xSn$ alloys. The Fermi level is located at $E = 0$

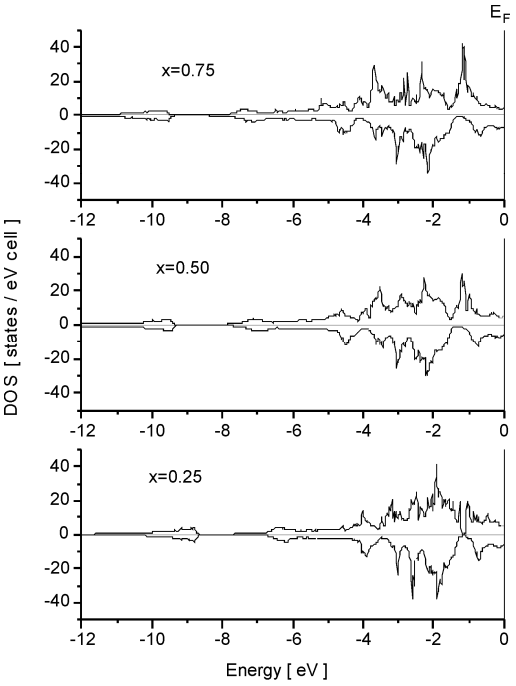

Fig. 2. The total density of states for ferromagnetic $Ni_2Ti_{1-x}Mn_xSn$ alloys. The Fermi level is located at $E = 0$

The total density of states for the ferromagnetic $Ni_2(Ti_{1-x}Mn_x)Sn$ is presented in Fig. 2 for $x = 0.25$, 0.5 and 0.75. We observe the variation of positions of the peaks in the majority spin band. The change of the density of states at the Fermi level for the ferromagnetic alloys is listed in Table 2.

Table 2. Total $N(E_F)$ and partial $N_T(E_F)$ (T = Mn, Ni, Ti, Sn) density of states at the Fermi level (states/eV) for the ferromagnetic $Ni_2Ti_{1-x}Mn_xSn$ alloys

x	$N(E_F)$	Mn	Ni	Ti	Sn
0.00	9.7	0.0	5.75	3.20	0.75
0.25	9.88	0.79	5.79	2.46	0.83
0.50	10.15	1.94	5.69	1.64	0.87
0.75	11.54	2.94	5.78	0.92	0.90
1.00	10.85	3.31	5.78	0.0	0.73

The maximum of $N(E_F)$ is observed for $x = 0.75$. The contribution from Ni and Sn to the total $N(E_F)$ is almost constant, however the contributions from Mn and Ti de-

pend on the concentration x. The total magnetic moment increases with the increase of the Mn concentration (Fig. 3).

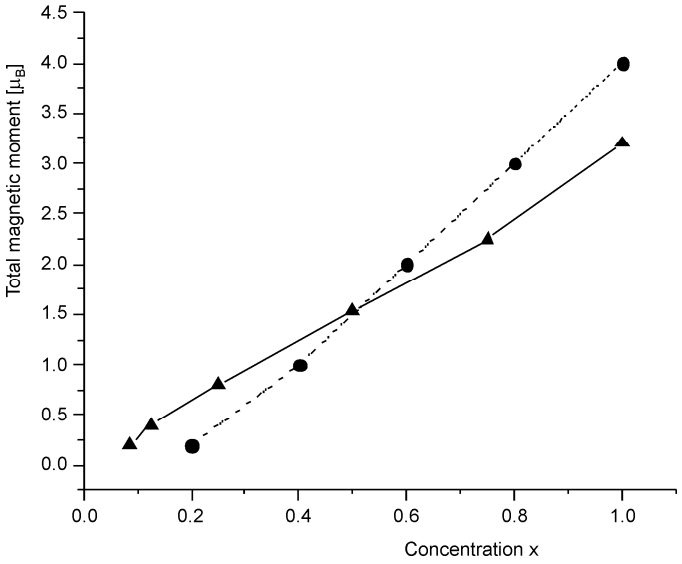

Fig. 3. The dependence of the total magnetic moment on manganese concentration in $Ni_2Ti_{1-x}Mn_xSn$ alloys. The dots denote the experimental data [4], and triangles denote the theoretical values obtained for the theoretical lattice parameters

The triangles denote the theoretical values and the dots represent the experimental data [4]. We observe that $Ni_2(Ti_{1-x}Mn_x)Sn$ becomes magnetic for $x > 0.0625$. The theoretical dependence of the magnetic moments is in a good agreement with the dependence $M_t = (Z_t - 24)\ \mu_B$ for the full-Heusler alloys, where Z_t is the total number of valence electrons per unit cell. Worse agreement between experimental and theoretical values of the total magnetic moments may be connected to the lower values of the theoretical lattice parameter (Table 1).

4. Conclusions

The electronic and magnetic properties of $Ni_2Ti_{1-x}Mn_xSn$ change with the increase of manganese concentration. The spin-polarized ab-initio calculations indicate that a small concentration of manganese ($x = 0.0625$) leads to ferromagnetic ground states of the $Ni_2Ti_{0.94}Mn_{0.06}Sn$ alloy. The theoretical values of the total magnetic moment change linearly with the increase of manganese concentration.

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