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Electronic Structure and Magnetic Properties of $Ni_2MnGa_{1-x}Ge_x$ and Disordered Ni_2MnSn Heusler Alloys

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In this work we present the influence of atomic disorder on the electronic and magnetic properties of $Ni_2MnGa_{1-x}Ge_x$ and Ni_2MnSn Heusler alloys. *Ab initio* band calculations were performed for the experimental lattice parameters. We applied SPR–KKR–CPA methods in the local spin density approximation.

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1. Introduction

The physical properties of the Heusler alloys are of great interest for both basic research and technological applications because of their magnetic properties and structural transformation. Due to potential application as magnetic actuators, the Heusler alloys based on Ni and Mn transition metals have been extensively investigated [1–9]. Ni_2MnGa is a vastly studied ferromagnetic shape memory alloy with martensitic transformation [5–7]. Here, we focused on electronic properties of $Ni_2MnGa_{1-x}Ge_x$, whose magnetic properties and temperature of martensitic transformation have been experimentally studied earlier [8]. The structural transformation of Ni–Mn–Sn from austenite to martensite phase has been studied by various experimental methods [1]. It was observed that the strength of the exchange interaction in both phases change substantially upon transformation from one phase to another. Attention has been devoted to $Ni_{50}Mn_{34}Sn_{16}$ in [4] because of its large magnetocaloric effect. Moreover, film samples of $Ni_{50}Mn_{50-x}Sn_x$ exhibited signs of martensitic transformation [2, 9], too.

2. Computational details

Electronic structure calculations were performed within the Korringa–Kohn–Rostocker (SPR-KKR) method in the atomic sphere approximation (ASA) [10–12]. We used the local spin density approximation (LSDA) in the formulation of Vosko–Wilk–Nusair (VWN) [14]. The effect of disorder was described by the coherent-potential approximation (CPA) as implemented in the framework of the method [10–13]. Computations were done for above than 800 k-points in the irreducible wedge of the first Brillouin zone. As a minimum-basis set for the valence states the $4s$, $4p$, $3d$ states were chosen for Ni, Mn, Ge, Ga atoms. The valence states of Sn were $5s$, $5p$, $4d$.

The studied alloys have cubic $L2_1$ structure. This structure can be described as consisting of four interpenetrating fcc Bravais lattices shifted along the body diagonal, originated at $(0,0,0)$, $(\frac{1}{4},\frac{1}{4},\frac{1}{4})$, $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ and $(\frac{3}{4},\frac{3}{4},\frac{3}{4})$ positions and abbreviated A, B, C and D, respectively. The A and C positions are occupied by Ni atoms. In the calculations for $Ni_2MnGa_{1-x}Ge_x$ alloys Ga atoms were replaced by Ge atoms in D sublattice of the $L2_1$ cell. Concentration of Ge atoms was $x = 0.02$, 0.06 and 0.10. The band calculations were performed for the experimental lattice parameter for Ni_2MnGa ($a = 10.9982$ a.u.).

The notation of $Ni_2(Mn_{1-x}Sn_x)(Mn_xSn_{1-x})$ is used below for describing disorder within the Mn–Sn sublattice in Ni_2MnSn alloy. For this notation at $x = 0$ Mn atoms occupied sublattice B and Sn atoms occupied sublattice D, i.e. the Ni_2MnSn is ordered. However, at the studied concentration $x = 0.15$, 0.36, 0.4, 0.5 some disorder between the Sn and Mn sublattice is assumed: Sn and Mn atoms simultaneously occupied both B and D positions in proportion defined by formula unit.

3. Results and discussion

The total and partial density of states (DOS) for $Ni_2Mn(Ga,Ge)$ for above-mentioned concentration of Ge atoms are presented in Fig. 1. It should be noted that the DOS of $Ni_2Mn(Ga,Ge)$ is formed via the hybridization of $3d$ Ni(A,C) and $3d$ Mn(B) states. The DOS of Mn illustrates separation of $3d$ Mn to bonding and antibonding states due to large exchange splitting. Thus, amount of states for majority spin is much bigger than the number of states for minority spin below the Fermi energy (FE) which is demonstrated by the large magnetic moment of Mn. The contribution to the total moment from Mn moments is 3.38 μ_B. Ni and Mn moment are oriented

parallel to each other. Substitution of Ge in D sublattice does not change the value of the total magnetic moment ($m = 3.896 \div 3.90\ \mu_B$). This theoretical result is not in contradiction with experimental study of Ni_2MnGa with Ga substituted by Ge. $Ni_2MnGa_{1-x}Ge_x$ film obtained by flash-evaporation [8] did not reveal appreciate differences in the magnetization for concentration of Ge $x = 0.08$, 0.24, 0.4.

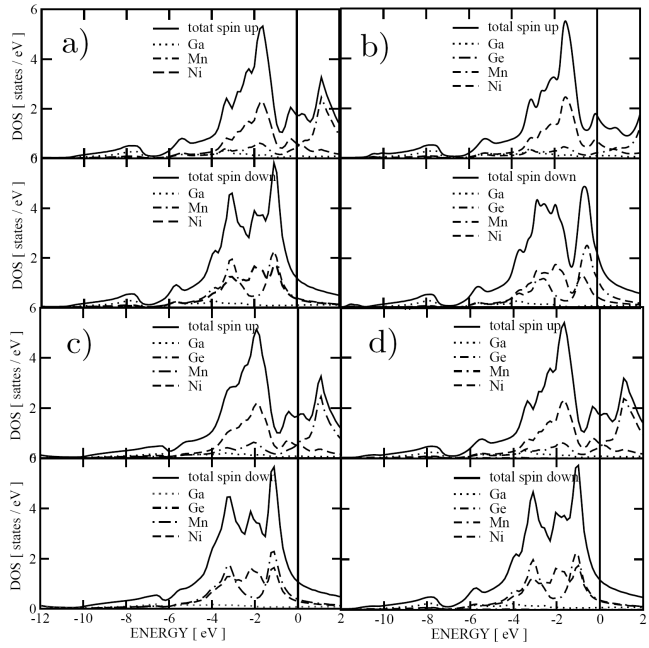

Fig. 1. The spin-projected total and partial DOS for: (a) Ni_2MnGa; (b) $Ni_2MnGa_{0.98}Ge_{0.02}$; (c) $Ni_2MnGa_{0.94}Ge_{0.06}$; (d) $Ni_2MnGa_{0.90}Ge_{0.10}$. The Fermi level is located at $E = 0$ eV.

Figure 2 shows the DOS of Ni_2(Mn,Sn) alloy produced by CPA method. The Mn and Sn atoms in this alloy occupied simultaneously both the B and D position of the $L2_1$ structure. The partial occupation of B-sites by Sn atoms as well of D-sites by Mn atoms changes the DOS at the Fermi level. The first peak for spin-up direction appears below the Fermi level and is shifted slightly towards the lower energy. The intensity of antibonding states of Mn is significantly decreased when the disorder between the Mn–Sn sublattices increases to 50% ($x = 0.5$). The DOS for spin-down direction is rebuilt as a result of the atomic disorder. For spin-down direction the smearing of some initial peaks in the total DOS with increase in the Mn–Sn disorder is observed. The visible changes in the total DOS are mainly determined by Ni and Mn DOS. The electronic states of Ni sublattices (A and C ones) are related to random atomic occupation in B and D sublattices, or to be more precise, to its first and third nearest neighbors. The DOS of Mn is influenced by the second nearest neighbors, while the first nearest neighbors of Mn in B are similar to that for ordered Ni_2MnSn. The increase in disorder between Sn–Mn sublattices leads

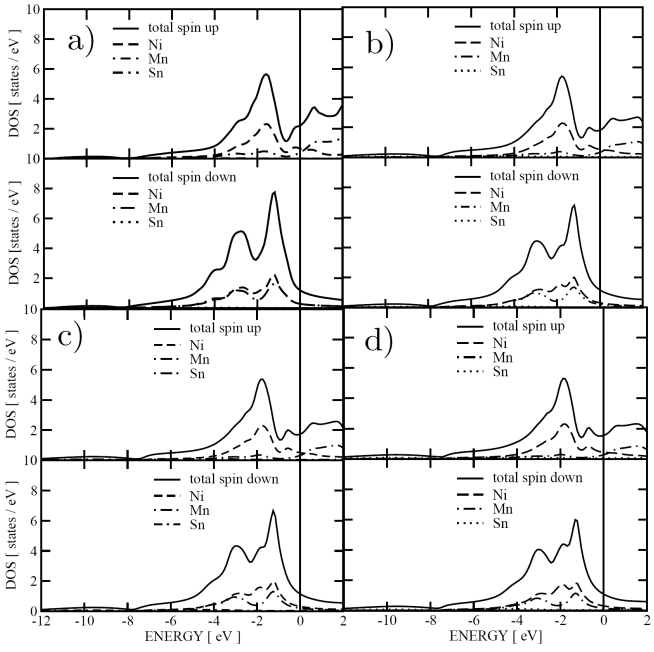

Fig. 2. The spin-projected total and partial DOS for: (a) $Ni_2(Mn_{0.85}Sn_{0.15})(Mn_{0.15}Sn_{0.85})$; (b) $Ni_2(Mn_{0.64}Sn_{0.36})(Mn_{0.36}Sn_{0.64})$; (c) $Ni_2(Mn_{0.6}Sn_{0.4})(Mn_{0.4}Sn_{0.6})$; (d) $Ni_2(Mn_{0.5}Sn_{0.5})(Mn_{0.5}Sn_{0.5})$. The Fermi level is at $E = 0$ eV.

to decrease in total magnetic moment of the alloy from 3.65 μ_B at $x = 0.15$ to 2.05 μ_B at $x = 0.5$. Higher values of the total moment for ordered Ni_2MnSn alloys achieved in *ab initio* calculation have been reported in [15–18]. We notice that the calculated value of DOS at the Fermi energy depends weakly on the alloy composition and the slight decrease in the DOS at the Fermi energy is found.

4. Conclusions

In this paper we present the electronic structure study for $Ni_2MnGa_{1-x}Ge_x$ and disordered Ni_2MnSn Heusler alloys. In the range of studied concentration of Ge our calculations show that magnetic properties of $Ni_2MnGa_{1-x}Ge_x$ does not change. In the studied $Ni_2MnGa_{1-x}Ge_x$ the substitution of Ge for Ga was only assumed in D sublattice of the parent $L2_1$ cubic structure. This result is in a good agreement with experimental data of the Curie temperature and magnetization of flash-evaporated films, in which lower structural ordering should be a natural feature of a sample.

The present study of disordered Ni_2MnSn Heusler alloy demonstrates that the drop of the magnetic moment is related to the disorder between the Mn–Sn sublattices by changing the nearest neighbors of transition metal constituents. This is reflected in the obtained electronic structure pictures by behavior of antibonding states of Mn as well as by reconstruction of spin-down DOS of Ni and Mn atoms.

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