Magnetic properties, electronic structure and stability of Heusler alloys $Mn_{2-x}Fe_{1+x}Al$

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Heusler Mn-based alloys are being intensively studied because of complex magnetic properties [1], spintronic applications, electronic structure and band-gap design. We present an investigation of the electronic structure and magnetic properties of the novel composition $Mn_{1.5}Fe_{1.5}Al$ [2].

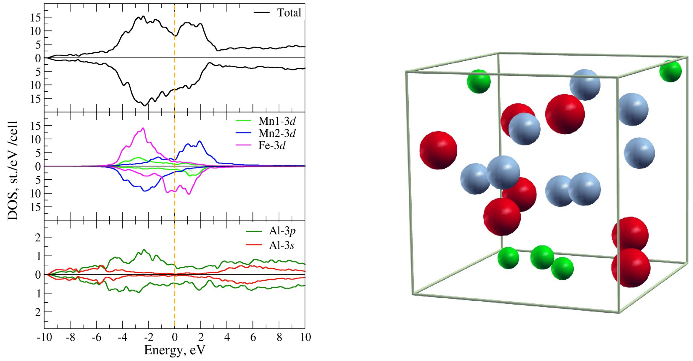
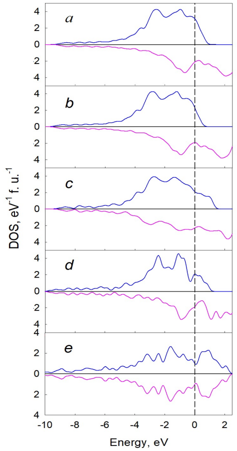


Fig. 1. Total and partial densities of states of the Heusler alloy $Mn_{1.5}Fe_{1.5}Al$ (a). Optimized crystal structure with Al shown as small green spheres, Fe as big red spheres, Mn shown as grey spheres (b).

The electronic structure of the Heusler alloy were calculated within the Quantum ESPRESSO (QE) package [3] with the exchange-correlation potential generalized gradient approximation (GGA) in the Perdew-Burke-Ernzerhof (PBE) version. The wave functions were expanded in plane waves. In the calculations, we used the same standard ultrasoft pseudopotentials from the QE library, as in [4].

$Mn_{1.5}Fe_{1.5}Al$ was found to be stabilized in a cubic β-Mn-type crystal structure [2] with an antiferromagnetic ordering of the Mn and Fe magnetic moments, similar to the recently reported Mn_2FeAl Heusler alloy [4]. The detailed theoretical study was done to obtain the optimized atomic positions for $Mn_{1.5}Fe_{1.5}Al$ in the β-Mn-type structure for the first time. The calculated total magnetic moment is found to be 1.76 μ_B per formula unit of $Mn_{1.5}Fe_{1.5}Al$. The magnetic ordering in this configuration is composed of the ferro and antiferromagnetically arranged Mn ions, being antiferromagnetically ordered mostly in the 8c-type positions. The average magnetic moment of Mn, Fe and Al are 3.1 (Mn2) and 2.2 (Mn1) μ_B, 0.7 and 0.2 μ_B. These contributions of the Mn ions give the largest contributions to the densities of states and magnetic properties of the $Mn_{1.5}Fe_{1.5}Al$ Heusler alloy.

The curves of the density of states (DOS) of the alloy are shown in Fig. 1a. Let us note significantly different patterns of the density of the 3d states of Mn2, Fe, Mn1 atoms for spins up and for spins down. As a result, the magnetic moments of Fe, Mn1 atoms are directed in one direction, and Mn2 atoms are directed in the opposite direction (ferrimagnet). DOS at the Fermi level in both spin sub-systems is relatively high. In comparison to Mn_2FeAl in the same crystal structure [4], both spin projections are more symmetrical because the most spin-polarized states of the Mn2 ions in Mn_2FeAl are less intense in $Mn_{1.5}Fe_{1.5}Al$ being partially replaced by Fe. On the other hand, the larger number of the Fe ions with spin polarization provide additional electronic states to the occupied part of the spin up and unoccupied part of the spin down in comparison to Mn_2FeAl. For spins up, the contribution to the density of states comes from the 3d-states of the Mn2, Fe, Mn1 atoms. For spins down the 3d-states of Fe atoms make the main contribution. The density of states of Al is low and is distributed over a wide range of energies.



Structural modification	Total density of spin up states at the Fermi energy, st/ eV Mn_3Al f.u.	Total density of spin down states at the Fermi energy, st/ eV Mn_3Al f.u.	Summed total density of states at the Fermi energy, st/ eV Mn_3Al f.u.	Total magnetic moment, μ_B / Mn_3Al f.u.	Cohesive energy, eV / f.u.
Mn_3Al with 5Al in 8c	3.1	2.1	5.2	6.9	-13.8
Mn_3Al with 5Al in 12d	1.8	1.9	3.7	4.1	-14.7
Mn_3Al with 5Al in both 8c and 12d	2.1	2.3	4.4	5.1	-14.3
$Mn_{1.5}Al$, i.e. all 8c are occupied with 8Al	2.1	1.9	4.0	4.0	-
$MnAl_{1.5}$, i.e. all 12d are occupied with 12Al	1.0	1.00	2.0	0	-

Fig. 2. The total density of electronic states calculated for the Mn_3Al alloy in the β-Mn type structure when aluminum atoms are: only in 8c (a), only in 12d (b) positions or simultaneously in both 8c and 12d types of positions (c). Figures (d) and (e) correspond to the limiting cases of full filling of all 8c are occupied with 8Al - $Mn_{1.5}Al$, (d) or all 12d positions are occupied with 12Al - $MnAl_{1.5}$ (e). The calculated parameters for the Mn-Al alloys with the β-Mn structure (table).

Conclusions

The calculations of the electronic structure and studies of the Heusler alloy $Mn_{1.5}Fe_{1.5}Al$ were carried out. The optimized β-Mn-type crystal structure is reported for the first time for $Mn_{1.5}Fe_{1.5}Al$. In the electronic structure of $Mn_{1.5}Fe_{1.5}Al$ in the β-Mn type structure, we obtained the metallic type of the density of states with the lower density in one spin projection corresponding to the ferrimagnetic ordering of the magnetic moments. Some Mn ions are found to form an antiferromagnetic ordering with the other Mn and Fe magnetic moments similar to the previously reported Mn_2FeAl. The calculations for the Mn-Al alloys show that with the occupation of the 12d positions with Al results in the decreasing total magnetic moment with the total moment equal to zero in some cases. Our theoretical calculations demonstrate the complex character of the magnetic properties and electronic structure of the Heusler $Mn_{2-x}Fe_{1+x}Al$ alloys.

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References

[1] V. Alijani, J. Winterlik, G.H. Fecher, C. Felser, Appl. Phys. Lett. **99**, 222510 (2011).
[2] E.I. Shreder, A.V. Lukoyanov, A.A. Makhnev, Yu.I. Kuz'min, S. Dash, A.K. Patra, M. Vasundhara, Electronic structure and optical properties of Heusler allow $Mn_{1.5}Fe_{1.5}Al$, JETP (2021, in press).
[3] P. Giannozzi, O. Andreussi, T. Brumme et al., J. Phys.: Condens. Matter. **29**, 465901 (2017).
[4] S. Dash, A.V. Lukoyanov, Nancy, D. Mishra, U.P.M. Rasi, R.B. Gangineni, M. Vasundhara, A.K. Patra, J. Magn. Magn. Mater. **513**, 167205 (2020).