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I. Galanakis, K. Özdoğan and E. Şaşıoğlu

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Spin-filter and spin-gapless semiconductors: The case of Heusler compounds

I. Galanakis,[1] K. Özdoğan,[2] and E. Şaşıoğlu[3]
[1]*Department of Materials Science, School of Natural Sciences, University of Paras, GR-26504 Patra, Greece*
[2]*Department of Physics, Yildiz Technical University, 34210 Istanbul, Turkey*
[3]*Peter Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany and Department of Physics, Fatih University, 34500, Büyükçekmece, Istanbul, Turkey*

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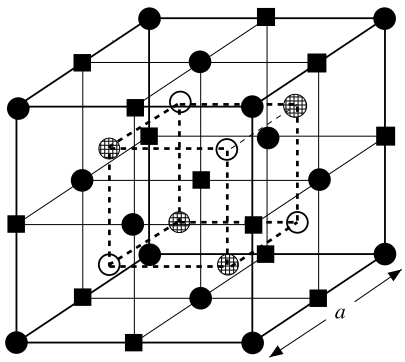
We review our recent first-principles results on the inverse Heusler compounds and the ordered quaternary (also known as LiMgPdSn-type) Heusler compounds. Among these two subfamilies of the full-Heusler compounds, several have been shown to be magnetic semiconductors. Such material can find versatile applications, *e.g.* as spin-filter materials in magnetic tunnel junctions. Finally, a special case are the spin-gapless semiconductors, where the energy gap at the Fermi level for the one spin-direction is almost vanishing, offering novel functionalities in spintronic/magnetoelectronic devices. © *2016 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution 3.0 Unported License.* [http://dx.doi.org/10.1063/1.4943761]

I. INTRODUCTION

Heusler compounds, named after von Heusler,[1] consist a large family of ternary and quaternary intermetallic compounds.[2,3] Among them several are well-known to exhibit diverse magnetic phenomena like itinerant and localized magnetism, antiferromagnetism, helimagnetism, Pauli paramagnetism or heavy-fermionic behavior.[2–9] Initially, the early Heusler compounds studied were crystallizing in the $L2_1$ structure which consists of 4 fcc sublattices presented in Fig. 1. Afterwards, it was discovered that it is possible to leave one of the four sublattices unoccupied ($C1_b$ structure). The latter compounds are often called half- or semi-Heusler compounds or simply Heuslers with NiMnSb being the most-known,[10] while the $L2_1$ compounds are known as full-Heusler compounds. Among both the full and semi Heusler compounds several have been predicted and then proved experimentally to present the so-called half-metallic property, i.e. the majority-spin electronic band structure is a usual metallic one while the minority band is semiconducting with a gap at the Fermi level E_F, leading to 100% spin polarization of conduction electrons at E_F.[11–15] Heusler compounds are particular interesting for applications due to their relative high Curie temperatures and an extensive review on potential applications can be found in Ref. 16.

In this short review, we will discuss from a theoretical point of view the properties of two distinct families of full-Heusler compounds, the so-called inverse full-Heuslers and LiMgPdSn-type quaternary Heuslers. Presented results have been obtained using the state-of-the-art ab-initio electronic band structure full-potential nonorthogonal local-orbital minimum-basis band structure scheme (FPLO)[17,18] method within the generalized gradient approximation (GGA) to the exchange-correlation energy.[19] Among the inverse and LiMgPdSn-type Heusler, there are case of particular interest: the spin-gapless semiconductors (SGS) and spin-filter materials (SFM). Both are in reality magnetic semiconductors. In SGS the gap in one of the spin-electronic band structures is zero and thus in this band structure holes and electrons can be excited with extremely low energies. SFM are usual magnetic semiconductors which present a different gap band-width in the two spin-electronic band structures and thus can act as spin filters finding application as barriers in magnetic tunnel junctions. We also review the properties of both SGS and SFM in Sec. IV.

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	●	○	■	▨
Full–Heuslers ($L2_1$) : X_2YZ	X	Y	X	Z
Quaternary Heuslers : (XX')YZ	X	Y	X'	Z
Inverse Heuslers (XA) : X_2YZ	X	X	Y	Z

FIG. 1. Schematic representation of the structures of the various variants of the full-Heusler compounds. In all cases the lattice is consisted of 4 interpenetrating fcc lattices.

II. INVERSE FULL-HEUSLER COMPOUNDS

The inverse full-Heusler compounds resemble the usual full-Heuslers having also the chemical formula X_2YZ but in their case the valence of the X transition-metal atom is smaller than the valence of the Y transition metal atom. They crystallize in the so-called *XA* or X_α structure, where the sequence of the atoms is X-X-Y-Z and the prototype is Hg_2TiCu; for the lattice structure see Fig. 1. Ab-initio calculations have shown that in that case the *XA* structure is energetically preferred to the $L2_1$ structure of the usual full-Heusler compounds where the sequence of the atoms is X-Y-X-Z.[20] Several inverse Heuslers have been studied using first-principles electronic structure calculations in literature.[20–30] Experiments on Mn_2CoGa and Mn_2CoSn films as well as Co doped Mn_3Ga samples have proven the stability of the *XA* structure in inverse Heusler compounds.[31–34] Inverse Heuslers are interesting for applications since they combine coherent growth on semiconductors with large Curie temperatures which can exceed the 1000 K as in the case of Cr_2CoGa.[35]

In Ref. 36, extensive ab-initio calculations on inverse full-Heusler compounds have been carried out. The inverse full-Heusler under investigation had the chemical formula X_2YZ where (X = Sc, Ti, V, Cr or Mn), (Z = Al, Si or As) and the Y ranged from Ti to Zn. Among them several have been found to be half-metallic magnets presenting simultaneously a Slater-Pauling type behavior for the total spin-magnetic moment. It was shown that when X is Sc or Ti, the total spin magnetic moment per formula unit (or unit cell) in μ_B follows the rule $M_t = Z_t - 18$ where Z_t is the total number of valence electrons in the unit cell. When X = Cr- or Mn, the variant followed by M_t is $M_t = Z_t - 24$, and when X = V the form of the slater-Pauling rule was found to be material specific. The occurrence of these rules has been explained in Ref. 36 using simple hybridization arguments of the transition metal *d*-orbitals. When X is Cr or Mn the situation is similar to the usual Heusler compounds (see Ref. 12), but when X is Sc or Ti, the Fermi level in the minority-spin band structure is located below the non-bonding t_{1u} states leading to the other variant of the Slater-Pauling rule. A schematic representation of the Slater-Pauling rule in inverse Heuslers is presented in Fig. 2 where we plot the calculated total spin-magnetic moments in μ_B per formula unit as a function of the total number of valence electrons. A third variant of the Slater-Pauling rule occurs, as shown in Ref. 36, when X is Cr or Mn and Y is Cu or Zn since in that case the Cu or Zn *d*-states are completely occupied forming a deep energy band, and the half-metallic compounds follow a $M_t = Z_t - 28$ rule.

III. ORDERED QUATERNARY HEUSLER COMPOUNDS

The ordered quaternary full-Heusler compounds are usually named as LiMgPdSn-type full-Heuslers (also known as LiMgPdSb-type Heusler compounds).[38] They have the chemical formula (XX')YZ where X, X' and Y are transition metal atoms where the valence of X' is lower than the valence of X atoms and the valence of the Y element is lower than the valence of both X and X'. In Fig. 1 we present also their structure; the sequence of the atoms along the fcc cube's diagonal

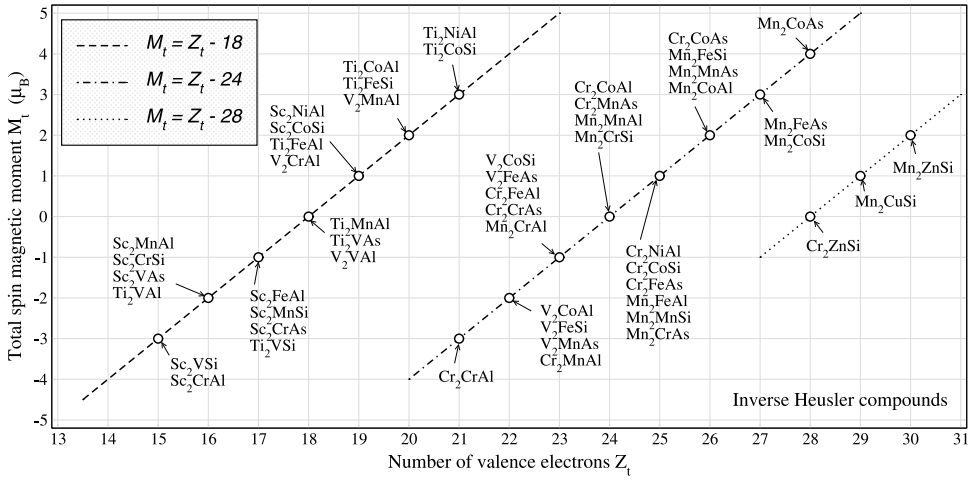


FIG. 2. Calculated total spin magnetic moments for several inverse Heusler compounds studied in Ref. 36 as a function of the total number of valence electrons in the unit cell, Z_t.

is X-Y-X'-Z which is energetically the most stable.[37] Recently they have also attracted interest and several studies have appeared.[38–42]

In Ref. 42, we have employed first-principles electronic structure calculations to study the electronic and magnetic properties of 60 LiMgPdSn-type quaternary Heusler compounds. It was shown that most of the compounds were half-metals obeying the same Slater-Pauling rule for full-Heusler compounds, $M_t = Z_t$-24,[12] with only few exceptions since the hybridization scheme is the same as in full-Heusler compounds. In Table I, we have gathered some of the calculated total and atom-resolved spin magnetic moments presented in Ref. 42.

IV. MAGNETIC SEMICONDUCTORS

In Heusler compounds there are three magnetic sublattices resulting to all kind of magnetic orders (ferro-, ferri- and antiferro-magnetism) in magnetic semiconductors depending on their chemical composition. Moreover, contrary to usual magnetic semiconductors, Heusler compounds exhibit large values of the Curie temperature and compatible crystal structure to the usual semiconductors -like Si, GaAs etc- making their incorporation in realistic devices more promising.

TABLE I. Calculated equilibrium lattice constant and spin magnetic moments in μ_B for the ordered quaternary Heusler compounds using the FPLO method.[17] All studied compounds follow the $M_t = Z_t - 24$ Slater-Pauling rule. The total spin magnetic moment is given per formula unit (which coincides with the per unit cell value).

(XX')YZ	a(Å)	m^X	$m^{X'}$	m^Y	m^{total}
(CoCr)TiAl	5.95	-0.23	-2.15	0.28	-2.00
(CoV)TiSi	5.90	-0.26	-1.67	-0.12	-2.00
(CoCr)VAl	5.82	-0.34	-1.28	0.59	-1.00
(CoMn)TiAl	5.86	-0.23	-1.03	0.23	-0.98
(CoCr)TiSi	5.80	-0.21	-0.92	0.09	-1.00
(CoMn)CrAl	5.71	0.66	-1.09	1.50	1.00
(CoFe)VAl	5.73	0.61	0.51	-0.09	0.97
(CoMn)VSi	5.65	0.67	-0.05	0.38	0.97
(CoFe)TiSi	5.73	0.58	0.65	-0.19	1.00
CoFe)MnAl	5.68	0.73	-0.18	2.59	3.00
(CoFe)CrSi	5.61	1.00	0.33	1.79	3.00
(CoFe)MnSi	5.60	0.79	0.55	2.81	4.00
(CoFe)MnAs	5.74	0.94	0.99	3.12	5.00

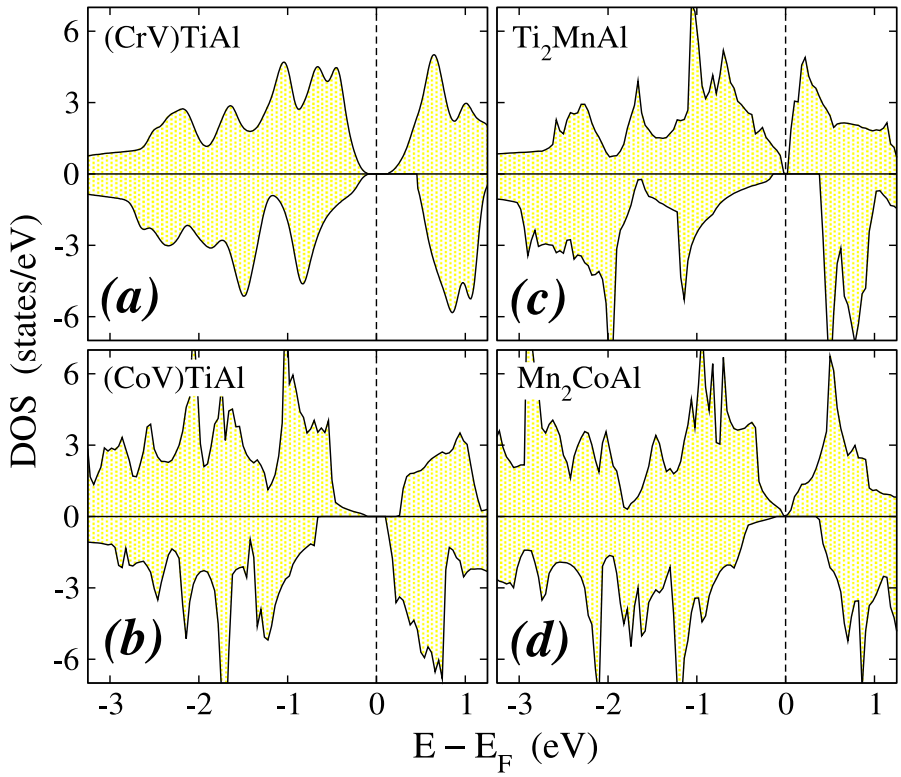


FIG. 3. Total DOS per formula unit around the Fermi level for (a) (CrV)TiAl a fully-compensated ferrimagnetic semiconductor, (b) (CoV)TiAl a ferromagnetic semiconductor, and (c) Ti_2MnAl and (d) Mn_2CoAl two spin-gapless semiconductors.

Thus Heusler compounds which are magnetic semiconductors can offer novel functionalities to spintronic/magnetoelectronic devices; they can act as spin-filter materials (SFM) in magnetic tunnel junctions (MTJ) where the barrier is a SFM and the electrodes are metallic. Then the probability for electrons tunnelling through the spin-filter barrier is different for the two spin-directions and the flow of a spin-polarized current can be achieved.[43,44]

Among Heusler compounds there are two families of compounds studied recently which are magnetic semiconductors and can be used as spin-filter materials: (i) (CoV)XAl with X being Ti, Zr or Hf which are ferromagnetic semiconductors,[45] and (ii) (CrV)XAl with X being Ti, Zr or Hf which are fully-compensated ferrimagnetic semiconductors.[46] Moreover the (CrV)XAl compounds combine the SFM character with a zero net magnetization leading to minimum energy losses in devices. In Fig. 3 we present the total density of states for (CoV)TiAl and (CrV)TiAl where the energy gaps in both spin-directions are present. Moreover in a recent publication (Ref. 47) further SFM have been identified among the quaternary ordered (CrV)XZ compounds Heusler compounds possessing 18 valence electrons per unit cell.

A new special class of magnetic semiconductors discussed above are the so-called spin-gapless semiconductors (SGS). In these magnetic semiconductors, one of the two spin-electronic band structures is gapless, *i.e.* presents an almost vanishing zero-width energy gap at the Fermi level.[48] SGS offer novel functionalists in spintronic/magnetoelectronic devices due to their unique properties: (i) the mobility of carriers is considerably larger than in usual semiconductors, (ii) excited carriers include both electrons and holes which can be 100% spin-polarized simultaneously, and (iii) a vanishing amount of energy is enough to excite majority-spin electrons from the valence to the conduction band.

The first reference to SGS materials in literature stems from the work of Wang in 2008, who proposed that the doping of $PbPdO_2$, a gapless semiconductor, with transition metal atoms would lead to a spin gapless semiconductor.[49,50] This prediction was confirmed experimentally several

years after by Kim and collaborators.[51] Among Heusler compounds it was Mn_2CoAl, an inverse full-Heusler compound, which attracted most of the attention. Initially in 2008 Liu and collaborators synthesized Mn_2CoAl and confirmed that it adopts the *XA* structure of inverse Heusler compounds while ab-initio calculations yielded a total spin magnetic moment of 1.95 μ_B per formula unit and an antiparallel coupling between the spin magnetic moments of nearest-neighboring Mn atoms.[21] In 2011 Meinert and collaborators studied again theoretically this compound reproducing the calculated results of Liu *et al*. and also studied the exchange constants revealing that magnetic interactions in these compounds are short range direct interactions.[52] But it was not until 2013 when experiments by Ouardi et al. confirmed the SGS character of Mn_2CoAl in bulk-like pollycrystalline films.[53] Its SGS character was accompanied by a Curie temperature of 720 K and a total spin magnetic per formula unit of 2 μ_B at a temperature of 5 K.[53] Following this breakthrough several groups have proposed new SGS materials based on first-principles electronic structure calculations.[42,54–58] In the same time experimentalists tried to grow thin films of Mn_2CoAl on various substrates; films on top of GaAs were found to deviate from SGS,[59,60] while films on thermally oxidized Si substrates were found to be SGS with a Curie temperature of 550 K.[61] Ab-initio calculations of Skaftouros et al. identified among the inverse Heusler compounds another four potential SGS materials: Ti_2CoSi, Ti_2MnAl, Ti_2VAs and Cr_2ZnSi, with the latter three being also fully-compensated ferrimagnets (we do not use the term antiferromagnets since there are three magnetic sublattices).[54] In Fig. 3 we present the total density of states for Mn_2CoAl and Ti_2MnAl as calculated in Ref. 54. Few theoretical studies have been devoted on the effect of various quantities on the SGS character of Mn_2CoAl. Namely Galanakis and collaborators have shown that defects keep the half-metallic character of Mn_2CoAl but destroy the spin-gapless semiconducting character,[62] and Zhang et al. have studied the effect of doping of Mn_2CoAl with Co, Cu, V and Ti,[63] and also the anomalous Hall effect in Mn_2CoAl has been exploited.[64] A recent publication by Jakobsson and collaborators in 2015 has focused on the five proposed inverse Heusler compounds being SGS (Mn_2CoAl, Ti_2MnAl, Cr_2ZnSi, Ti_2CoSi and Ti_2VAl) and studied the exchange interactions, the magnons and the temperature-dependence of the magnetization in these compounds.[65] They have shown that the exchange interactions are short-range, confirming the conclusions by Meinert et al. on Mn_2CoAl,[52] and all five compounds were found to present Curie temperatures above the room temperature.[65] Finally the calculated magnetization curve and critical temperature for Mn_2CoAl in Ref. 65 reproduced the experimental data in Ref. 53.

V. SUMMARY AND CONCLUSIONS

We have presented a short review of our recent ab-initio results on two distinct subfamilies of the full-Heusler compounds: (a) the so-called inverse Heusler compounds with the chemical formula X_2YZ where X and Y are transition metal atoms with the valence of X being the smallest, and (ii)the ordered quaternary (also known as LiMgPdSn-type) Heusler compounds which have the chemical formula (XX')YZ where X, X' and Y are transition metals.

Among these two subfamilies of the full-Heusler compounds, several are shown to be magnetic semiconductors. Due to the existence of three magnetic sublattices, they present a variety of magnetic orders (ferro- ferri- or antiferromagnetism) depending on their chemical composition. These materials can be used in applications, like magnetic tunnel junctions, as spin-filter materials. The most promising for applications are the ones with 18 valence electrons since they also have a zero net magnetization leading to minimum energy losses in applications.

A special case are the spin-gapless semiconductors, where the energy gap at the Fermi level for the one spin-direction is almost vanishing, offering novel functionalities in spintronic/magnetoelectronic devices. Among the proposed spin-gapless materials Mn_2CoAl is of particular interest since it has been also grown experimentally and its unique character has been confirmed in Ref. 53.

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