



Ab-initio investigation of electronic and magnetic properties of the 18-valence-electron fully-compensated ferrimagnetic (CrV)XZ Heusler compounds: A prototype for spin-filter materials



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ABSTRACT

Spin-filter materials are magnetic semiconductors susceptible of improving the performance of magnetic tunnel junction based spintronic devices. Using state-of-the-art *ab initio* electronic structure calculations, we exploit the electronic and magnetic properties of (CrV)XZ quaternary Heusler compounds having 18 valence electrons per formula unit. All compounds under study have been found to combine fully-compensated ferrimagnetism to magnetic semiconducting behavior and some are even gapless or spin-gapless semiconductors. Our results pave the way for incorporating these compounds in spintronic and magnetoelectronic devices.

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1. Introduction

The design of novel magnetic nanomaterials is primordial for the development of the fields of spintronics and magnetoelectronics [1]. In this respect, *ab initio*, also known as *first-principles*, electronic band structure calculations play a key role since they allow the study of such materials prior to their experimental growth leading to an à-la-carte design of materials for specific applications [2]. Among Heusler compounds several have been extensively studied for their half-metallic properties [3–7], but still novel properties can be discovered among them. A case of particular interest is the so-called spin-filter materials (SFMs) [8,9]. These materials are magnetic semiconductors aiming to maximize the efficiency of devices based on magnetic tunnel junctions (MTJs) [1], like the recently proposed spin-current diodes [10]. To achieve spin-polarized current through a MTJ either (a) one should use magnetic electrodes separated by an insulating barrier and ballistic transport is achieved through the tunnelling of the electrons via the barrier, or (b) one could use non-magnetic metallic electrodes separated by a SFM to achieve the spin-polarized current due to the different

potential barrier confronted by the spin-up and spin-down electrons.

The existing SFMs are not suitable for applications either due to their very low Curie temperature (e.g. europium chalcogenides) or their complex lattice structures (e.g. NiFe₂O₄) [11–18], and thus devices based on them are not operational [19–22]. This makes very attracting the quaternary Heusler compounds [23,24], which have been predicted to be SFMs and more precisely the (CoV)XAl and (CrV)XAl compounds where X stands for Ti, Zr, Hf [25,26]. The former compounds are ferromagnetic semiconductors while the latter ones are fully-compensated ferrimagnetic semiconductors since they combine the existence of energy gaps in both spin-directions to zero magnetization [25,26]; note that the use of the term “antiferromagnet” is not adequate since in these compounds there are three magnetic sublattices made of Cr, V and Ti (or Zr or Hf) atoms with antiparallel spin moments while in conventional antiferromagnets there are two equivalent magnetic sublattices with opposite spin magnetic moments. For both families, the estimated Curie temperature T_C is shown to exceed the room temperature and thus they are of interest for room-temperature spintronic/magnetoelectronic applications. The fully-compensated ferrimagnetic character of (CrV)XAl compounds make them even more attractive for applications since the zero net magnetization leads to vanishing stray fields and thus to minimal energy losses.

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2. Motivation and computational details

In the present study we extend our previous work in reference [26] to cover a wider range of potential SFM materials. We have included (a) the (CrV)XZ compounds, where X stands for Ti, Zr or Hf and Z being Al, Ga or In atoms, (b) the (CrV)XZ compounds where X is Sc or Y and Z is Si, Ge or Sn. All these compounds have 18 valence electrons per formula unit (f.u.) and thus, as will be shown and explained in the next section, are fully-compensated ferrimagnets. The lattice structure is that of the LiMgPdSn-type Heusler compounds where the unit cell is a fcc with four atoms as basis along the diagonal and the sequence of the atoms is Cr–X–V–Z since in all studied cases the valence of X is smaller than the valence of Cr and V atoms [24]. In this structure each Cr or V atom has four X and four Z atoms as nearest neighbors where each X or Z atom has four Cr and four V atoms as nearest neighbors. The Cr and V atoms are next-nearest-neighbors between them. We have also studied the (CrV)XS with X being K or Rb, the (CrV)XP with X being Ca or Sr and finally the (CrV)XZn compounds with X being V, Nb or Ta. All these compounds have also 18 valence electrons per f.u. being fully-compensated ferrimagnets, but show even more interesting behavior in electronic states around Fermi energy like the ferrimagnetic gapless-semiconducting [27] or the spin-gapless-semiconducting behavior [24,27–29]. In the former there is a zero energy gap in both spin directions but the bands are different for the two spin-directions, while in the latter there is a zero gap for the one spin-direction and a finite gap for the other spin-direction. To clarify this complex behavior in Fig. 1 we have plotted schematically the density of states (DOS) per f.u. vs the energy for fully-compensated ferrimagnets and simple ferrimagnets. For the former the number of spin-up and spin-down electrons are equal, i.e., $N^\uparrow = N^\downarrow$, while for the latter they are not ($N^\uparrow \neq N^\downarrow$).

Computational details are similar to the ones in references [25,26]. We employ the full-potential nonorthogonal local-orbital minimum-basis band structure scheme (FPLO) [30] within the local-(spin)-density approximation (L(S)DA) [31] to calculate the ground state properties. For Heusler compounds both LDA and generalized gradient approximation (GGA) [32] yield similar electronic and magnetic properties for the same lattice constant [33]. First, we have calculated the equilibrium lattice constants for the various compounds employing total energy calculations. The calculated values in most cases exceed the 6 Å (see second column in Tables 1 and 2). Using a dense \mathbf{k} -point grid for the equilibrium lattice constants we have calculated both the atomic spin magnetic moments as well as the DOS within the scalar-relativistic formalism.

3. Results and discussion

Prior to presenting our results we should stretch out the fact that they have been produced using a conventional widely-used density functional. Due to the existence of the energy gaps in the electronic band structure one could envisage using also approximations beyond the standard functionals like (a) the orbital dependent functionals [34], (ii) the Hubbard LDA + U schemes [35,36], or (iii) hybrid functionals like the Heyd–Scuseria–Ernzerhof functional [37,38]. Such schemes are known to alter in several cases the results obtained with the standard LDA or GGA schemes, and sometimes they also produce contradictory results. We believe that for the compounds under study the use of standard LSDA or GGA functionals can describe accurately their electronic and magnetic properties since similar calculations reproduce well the experimental results in the case of the spin-gapless semiconducting Mn_2CoAl Heusler compound [39].

3.1. The (CrV)XZ compounds with X = Sc, Y, Ti, Zr or Hf

We begin with the discussion of the compounds containing the two early transition metal atoms Sc (Y) and Ti (Zr, Hf). Note that Y is isovalent to Sc and Zr and Hf are isovalent to Ti. The Z atoms in the case of Sc (Y) could be Si, Ge or Sn to have in total 18 valence electrons per f.u., and Al, Ga or In in the case of Ti (Zr, Hf) atom in order to have again 18 valence electrons per f.u. The properties of (CrV)TiAl, (CrV)ZrAl and (CrV)Hf compounds have been studied in detail in reference [26] and thus we will mostly concentrate on the effect on the properties of the compounds when either Al is substituted by the heavier isovalent Ga or In, and when Sc and Y substitute Ti and Zr.

All compounds under study in this section possess a ferrimagnetic ground state as shown in Table 1 with the Cr atomic spin magnetic moments being antiparallel to the spin magnetic moments of the other transition metal atoms. Cr atoms carry large negative spin magnetic moments being around $-3 \mu_B$ while the V atoms carry positive spin magnetic moments slightly smaller in absolute terms than the Cr ones. The X atoms possess a small but sizeable positive spin magnetic moment while the sp atoms carry only a negligible spin magnetic moment.

The total spin magnetic moment in the f.u. is zero in all cases and thus the ground states of all compounds in Table 1 is the fully-compensated ferrimagnetic one. In usual full-Heusler compounds, like Co_2MnSi , being half-metals there are exactly twelve occupied states in the minority spin band structure (see lower left panel in Fig. 2): four electrons are accommodated in the low lying s and p bands originating from the Si atom, five electrons are accommodated in the double-degenerate e_g and triple-degenerate t_{2g} bonding hybrids between all transition metal atoms and the gap is created between the triple-degenerate non-bonding t_{1u} and the double-degenerate non-bonding e_u hybrids which both are located at the Co atoms (see reference [4] for an extended discussion). In the case of the (CrV)XZ compounds under study the fact that X is an early transition-metal atom greatly affects the band structure and the position of the gap. Z atom again offers in each spin direction four states low in energy like in full-Heusler compounds (see lower right panel in Fig. 2) but now the Fermi level is located for both spin directions below the non-bonding t_{1u} states and thus falls within the gap separating the occupied t_{2g} and the empty t_{1u} states and as result there are exactly nine occupied states per formula unit. The fact that the DOS is not identical for the two spin-directions leads to the fully-compensated ferrimagnetic state instead of the usual antiferromagnetic state. In this respect, it is very important that are Cr and V atoms in the chemical type. If instead of V, there was a second Cr atom, then the two Cr atoms would have antiparallel spin magnetic moments of the same magnitude and the X and Z atoms would have zero spin magnetic moment leading to a conventional antiferromagnetic state. In the upper panel of Fig. 2 we present the hybridization scheme for the d states between the transition-metal atoms. One should first consider the possible hybridization between orbital of the Cr and V atoms which sit at sites of the same nearest-neighbor environment and then the hybridization between these hybrids and the orbitals of the X atom.

All compounds studied in this section are fully-compensated ferrimagnetic semiconductors presenting an energy gap at the Fermi level in both spin-directions. In Fig. 3 we have presented the total DOS per f.u. as a function of the energy for four compounds and compared with the well-studied (CrV)TiAl (see reference [26]). When we substitute Ga or In for Al in (CrV)TiAl, the shape of the DOS does not change considerably (see right panels in Fig. 3) but the energy gaps become smaller since the heavier

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