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# High $T_C$ half-metallic fully-compensated ferrimagnetic Heusler compounds

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Extensive *ab-initio* electronic structure calculations on Heusler alloys suggest that Cr<sub>2</sub>CoGa is the alloy of choice to achieve the half-metallic fully-compensated ferrimagnetism since (1) it has been already grown experimentally [T. Graf *et al.*, *Z. Anorg. Allg. Chem.* **635**, 976 (2009)], (2) half-metallic XA structure is favored energetically over all the studied lattice constant range with respect to the L<sub>21</sub> which is not half-metallic, (3) the half-metallic gap is wide and the Fermi level falls at the middle of the gap and thus, it presents high degree of spin-polarization for a wide range of lattice constants, and (4) the Curie temperature is extremely high reaching the 1520 K.

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Half-metallic alloys have been widely studied due to their potential applications in spintronics.<sup>1,2</sup> These compounds present metallic character for the electrons of one spin direction while their band structure presents a gap in the other spin direction and thus half-metallic properties.<sup>2</sup> Half-metallic Heusler compounds having the formula X<sub>2</sub>YZ, where X and Y transition metal atoms and Z a sp-element, attracted intense interest since: (1) most of them present very high Curie temperatures, (2) they crystallize in a cubic high-symmetry structure closely related to the zincblende of semiconductors, and (3) simply by varying in the periodic table the valence of X, Y, and Z in a continuous way new Heusler alloys with novel properties emerge.<sup>3</sup> Galanakis and collaborators have elucidated the origin of the gap in these alloys and have shown that the total spin-magnetic moments exhibit a Slater-Pauling behavior being per unit cell in  $\mu_B$  the total number of valence electrons minus 24.<sup>4</sup>

A special case in the applications are the compounds made up of magnetic elements with exactly 24 valence electrons which should have a total zero spin magnetic moment in the case of half-metallicity. These alloys should be of special interest for applications since they create no external stray fields and thus exhibit minimal energy losses. They should be named as half-metallic fully-compensated ferrimagnets (HM-FCFs)<sup>5</sup> and not half-metallic antiferromagnets which was the initial term used by van Leuken and de Groot<sup>6</sup> when studying the semi-Heusler compound CrMnSb in 1995. Contrary to conventional antiferromagnets here, the compensation of the spin magnetic moments stems from different magnetic sublattices, e.g., in CrMnSb Cr and Mn atoms have antiparallel spin magnetic moments of about the same magnitude.<sup>6</sup> Due to the origin of the annihilation of the spin moments, the microscopic magnetization at finite temperature is not zero as in conventional antiferromagnets.<sup>7</sup> Heusler alloys with 24 valence electrons which have been predicted to be HM-FCFs include the Mn<sub>3</sub>Ga,<sup>5</sup> Cr<sub>2</sub>MnZ (Z = P, As, Sb, Bi) alloys,<sup>8,9</sup> the Co-doped Mn<sub>2</sub>VZ (Z = Al, Si)

half-metallic ferrimagnetic alloys,<sup>10</sup> and the Cr-doped Co<sub>2</sub>CrAl.<sup>11</sup>

Experimentally Meinert *et al.*<sup>12</sup> have tried unsuccessfully due to disorder to achieve a HM-FCF alloy by growing (Mn<sub>0.5</sub>Co<sub>0.5</sub>)<sub>2</sub> VAl films. An alternative route are the Heusler alloys crystallizing not in the usual L<sub>21</sub> structure (prototype Cu<sub>2</sub>MnAl) but in the so-called XA structure (prototype Hg<sub>2</sub>TiCu) susceptible to occur when the valence of the X atom is smaller than the valence of the Y transition metal atom.<sup>3</sup> In the latter structure, the four sites (A, B, C, and D) in the unit cell are occupied in the sequence X-X-Y-Z instead of the X-Y-X-Z sequence in the L<sub>21</sub> structure and the two X atoms are no more equivalent (see Fig. 1 in Ref. 9 for the structure). Recently Graf *et al.*<sup>13</sup> have grown bulk samples of the Cr<sub>2</sub>CoGa alloy made up from polycrystalline ingots in the XA structure, which has 24 valence electrons and in accordance to the Slater-Pauling rule, it is expected to be a HM-FCF. The aim of the present letter is to study the 24-valence compounds Cr<sub>2</sub>CoGa, Cr<sub>2</sub>FeGe, Cr<sub>2</sub>MnAs, and Cr<sub>3</sub>Se alloys in both the XA and L<sub>21</sub> structures using *ab-initio* calculations. Our results show that for Cr<sub>2</sub>CoGa and Cr<sub>2</sub>FeGe, the

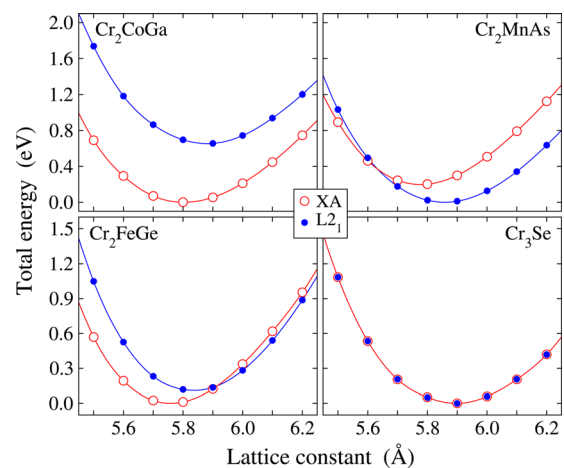


FIG. 1. (Color online) Calculated total energy as a function of the lattice constant for both the XA and L<sub>21</sub> structures. Note that, we have set as zero in each case, the total energy corresponding to the equilibrium lattice constant.

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TABLE I. For all four compounds under study and for both XA and L2<sub>1</sub> lattice structures, we present the calculated equilibrium lattice constant ( $a_{\text{eq}}$ ), the energy difference between the XA and L2<sub>1</sub> structures ( $\Delta E$ ), the absolute value of the spin-polarization at the Fermi level ( $P$ ), and finally, the calculated Curie temperature within the mean-field-approximation taking into account only the inter-sublattice interaction  $T_C^{\text{inter}}$  and both the intra- and inter-sublattice interactions  $T_C^{\text{all}}$ .

	$a_{\text{eq}}$ (Å)		$\Delta E$ (eV)	$P(\%)$		$T_C^{\text{inter}}$ (K)		$T_C^{\text{all}}$ (K)	
	XA	L2 <sub>1</sub>		XA	L2 <sub>1</sub>	XA	L2 <sub>1</sub>	XA	L2 <sub>1</sub>
Cr <sub>2</sub> CoGa	5.80	5.88	-0.70	93	45	1225	210	1520	125
Cr <sub>2</sub> FeGe	5.76	5.84	-0.15	92	24	667	70	748	2
Cr <sub>2</sub> MnAs	5.78	5.87	0.25	95	52	545	600	610	330
Cr <sub>3</sub> Se	5.90		—	75		770		692	

XA structure is the most favorable with a gap in the spin-up band and spin-polarization reaching the 100%. Moreover in the case of XA-Cr<sub>2</sub>CoGa, the estimated value of the Curie temperature exceeds 1500 K and it is expected to be a prototype for achieving a perfect HM-FCF.

Since the equilibrium lattice constants are not known, we calculated them from total energy calculations employing the full-potential linear augmented plane-waves method (FLAPW)<sup>14</sup> in the FLEUR implementation<sup>15</sup> in conjunction to the Perdew-Burke-Ernzerhof formulation of the generalized gradient approximation (GGA) to the exchange-correlation potential.<sup>16</sup> The calculated curves are presented in Fig. 1 for all four compounds under study and for both structures (note that for Cr<sub>3</sub>, Se we cannot distinguish between XA and L2<sub>1</sub> structures). Moreover in Table I, we present the calculated equilibrium lattice constants and the energy difference between the one for the XA and the one for the L2<sub>1</sub> structure. Calculated values are close for both structures which is expected; for XA, they are slightly smaller. Also, we do not remark large deviations among the four compounds since Cr, Mn, Fe, and Co have similar atomic radius and obtained values are close to the lattice constants of various binary semiconductor. When the Y atom is Co or Fe, the XA structure is more stable and especially for Cr<sub>2</sub>CoGa, the energy difference is -0.7 eV, a pretty large value and the lines for the two structures are well separated over the whole studied lattice range contrary to Cr<sub>2</sub>FeGe where XA and L2<sub>1</sub> are degenerated for values larger than 5.9 Å. When we attend Cr<sub>2</sub>MnAs, the L2<sub>1</sub> structure becomes the more favorable one; note that for Cr<sub>2</sub>MnSb studied in Ref. 9, the two structures were almost degenerated. Thus, Cr<sub>2</sub>CoGa seems to be the most well-suited to grow in the XA structure as shown experimentally in Ref. 13. We should also note that in Ref. 13, its lattice constant was determined to be 5.81 Å almost identical to our calculated value of 5.80 Å.

In Fig. 2, we present the calculated total density of states (DOS) for all four compounds and for both structures at the equilibrium lattice constants. We have focused around the half-metallic gap and the insets present the DOS over a wider energy range. For the L2<sub>1</sub> structure, the Cr<sub>2</sub>CoGa and Cr<sub>2</sub>FeGe alloys show no gap which has been discussed in Ref. 17 in terms of the exchange splitting of the Co(Fe) d-states. Cr<sub>2</sub>MnAs presents a gap in the spin-up band but the Fermi level falls slightly below the gap as in Cr<sub>2</sub>MnSb

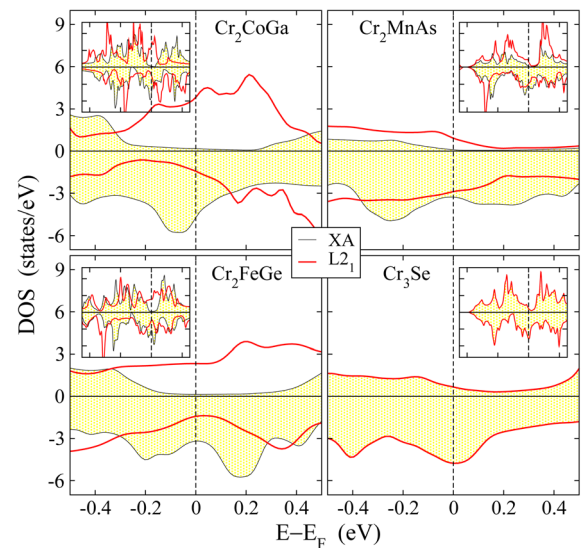


FIG. 2. (Color online) Total DOS for both XA and L2<sub>1</sub> structures for a region  $\pm 0.5$  eV around the Fermi level. Insets present the DOS over a much wider energy region. The zero energy is assigned to the Fermi level and the positive DOS values correspond to the spin-up electrons and negative DOS values to the spin-down electrons.

alloy.<sup>9</sup> As a result, the absolute value of the spin-polarization ( $P$ ) at the Fermi level defined as the ratio  $|\frac{N^{\uparrow} - N^{\downarrow}}{N^{\uparrow} + N^{\downarrow}}|$  where  $N^{\uparrow}$  is the number of electrons at the Fermi level with spin-up and  $N^{\downarrow}$  the electrons of spin-down character, presented in Table I for the L2<sub>1</sub> structure is very small. When we move to the XA structure now at the C site is a Cr atom and the large exchange splitting of the Cr d-states restores the half-metallic gap in the spin-up band (we cannot use the terms majority and minority spins since both spin-directions contain the same number of electrons) and the Fermi level falls within the half-metallic gap. The gap is pretty large being about 0.5–0.6 eV. Cr<sub>2</sub>MnAs in the XA structure also shows a half-metallic gap and similar is the situation for Cr<sub>3</sub>Se but in the latter, the Fermi level falls near the left edge of the gap and  $P$  presented in Table I is 75% far below the values for the other three alloys in the XA structure. Note that, we do not have a real gap and thus,  $P$  is not exactly 100% but exceeds 90% meaning that more than 95% of the electrons are of spin-down character. Finally, since the Fermi level falls for both Cr<sub>2</sub>CoGa and Cr<sub>2</sub>FeGe in the middle of the gap, we expect to retain a large value of  $P$  upon compression or expansion since these result to a shift of the Fermi level towards smaller or larger values of the energy, respectively.<sup>18</sup> Calculations for both alloys at the experimental lattice constants of zinc-blende CdS (5.65 Å), InP (5.87 Å), and InAs (6.06 Å) semiconductors have produced for  $P$  values of 95%, 92%, and 91%, respectively, for Cr<sub>2</sub>CoGa and 92%, 97%, and 96% for Cr<sub>2</sub>FeGe. Thus, an extremely high spin-polarization is preserved over a wide lattice constant region.

The electronic properties are reflected also on the spin magnetic moments presented in Table II. In the case of the XA structure, the total spin magnetic moment for all compounds is close to zero and Cr atoms are antiferromagnetically coupled between them while Co(Fe,Mn) carry smaller spin moments resulting in the desired half-metallic fully-compensated ferrimagnetism. Cr<sub>2</sub>CoGa and Cr<sub>2</sub>FeGe alloys

TABLE II. Spin magnetic moments (in  $\mu_B$ ) for all four  $\text{Cr}_2\text{YZ}$  compounds under study at the calculated equilibrium lattice constants. The values for the sp atom (Z) are not presented since they are negligible.

Sites	XA structure				L2 <sub>1</sub> structure			
	A	B	C	Total	A	B	C	Total
	Cr	Cr	Y	Total	Cr	Y	Cr	Total
$\text{Cr}_2\text{CoGa}$	2.10	-1.93	-0.29	-0.07	2.29	0.97	2.29	5.56
$\text{Cr}_2\text{FeGe}$	1.28	-1.57	0.26	-0.03	0.66	-2.29	0.66	-1.02
$\text{Cr}_2\text{MnAs}$	1.08	-1.85	0.76	-0.03	1.36	-2.88	1.36	-0.21
$\text{Cr}_3\text{Se}$	1.25	-2.34	1.25	0.09	1.25	-2.34	1.25	0.09

in the L2<sub>1</sub> structure have total spin magnetic moments which considerably deviate from the zero spin moment predicted by the Slater-Pauling rule for perfect half-metals being even about 5.5  $\mu_B$  for  $\text{Cr}_2\text{CoGa}$ .  $\text{Cr}_3\text{Se}$  alloys is at the edge of being half-metallic since the Fermi level is at the left edge of the gap and thus, it also presents a total spin magnetic moment close to the zero value.

For realistic applications, one of the most important features is the Curie temperature  $T_C$ . We have estimated using the methodology developed in Ref. 18 and employing the multi-sublattice mean-field-approximation which was found to give accurate results for Heusler compounds like the ones under study here. Note also that, our formalism is implemented in conjunction to the augmented spherical waves method (ASWs)<sup>19</sup> but results using GGA<sup>20</sup> concerning the electronic and magnetic properties at the equilibrium lattice constants were found to be in perfect agreement to the FLAPW results. In Table I, we have gathered our results for both structures taking into account only intersublattice interactions and both intra- and inter-sublattice interactions (denoted as  $T_C^{\text{all}}$ ). We remark that the former ones give the main contribution to the  $T_C$  as for the half-metallic ferromagnets like  $\text{Co}_2\text{MnSi}$  studied in Ref. 18. As shown in Table I, inclusion of the intrasublattice interactions further increases the  $T_C$  value for the XA compounds while the opposite occurs for the L2<sub>1</sub> ones. For both cases where Y is Co or Fe,  $T_C^{\text{all}}$  is much larger for the half-metallic XA structure than the L2<sub>1</sub> one reaching even a value of 1520 K for  $\text{Cr}_2\text{CoGa}$  and making it a perfect candidate for applications. For  $\text{Cr}_2\text{MnAs}$ ,  $T_C^{\text{all}}$  is also larger for the XA structure, while for  $\text{Cr}_3\text{Se}$ , it is about 700 K. Thus, we remark that the high value of the  $T_C$  is intrinsically related to the existence of half-metallicity since the latter one stabilizes the ferro- or ferri-magnetism in Heusler alloys as discussed extensively in Ref. 18.

We have performed extensive *ab-initio* electronic structure calculations for the 24-valence  $\text{Cr}_2\text{CoGa}$ ,  $\text{Cr}_2\text{FeGe}$ ,  $\text{Cr}_2\text{MnAs}$ , and  $\text{Cr}_3\text{Se}$  alloys in both the XA and L2<sub>1</sub> structures. Our results suggest that, all alloys in the XA structure are half-metallic fully-compensated ferrimagnets with high value of the Curie temperature. For the alloys containing Co and Fe, the XA lattice structure is also energetically favorable with respect to the L2<sub>1</sub> which corresponds to non half-metallic alloys.  $\text{Cr}_2\text{CoGa}$  alloy seems to be the most well suited to achieve a half-metallic fully compensated ferrimagnet (also known as half-metallic antiferromagnet) since it has been already grown experimentally as shown in Ref. 13 and it combines a series of unique features: (1) XA structure is favored over all the studied lattice constant range, (2) the spin-up gap is wide and the Fermi level is at the middle of the gap and thus, it presents high degree of spin-polarization for a wide range of lattice constants, and (3) the Curie temperature is extremely high reaching the 1520 K which is the largest with respect to known Heusler alloys.<sup>3</sup>

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