Towards half-metallic interfaces: the Co$_2$CrAl/InP contacts

Iosif Galanakis §
Institut für Festkörperforschung, Forschungszentrum Jülich, D-52425 Jülich, Germany

Abstract. Although the interest on half-metallic Heusler alloys, susceptible to be used in spintronic applications, has considerably grown, their interfaces with semiconductors show very low spin-polarization. I identify mechanisms which can keep the high spin-polarization at the interface (more than 80% of the electrons at the Fermi level are of majority spin) although the half-metallicity is lost. The large enhancement of the Cr moment at the interface between a CrAl terminated Co$_2$CrAl(001) spacer and the InP(001) semiconductor weakens the effect of the interface states resulting in this high spin-polarization. On the other hand the Co$_2$CrAl/InP interfaces made up by a Co layer and either an In or a P one show a severe decrease of the Co spin moment but Cr in the subinterface layer is bulklike and the resulting spin-polarization is similar to the CrAl-based interfaces.

PACS numbers: 73.20.-r, 73.20.At, 71.20.-b, 71.20.Lp

§ To whom correspondence should be addressed, e-mail: I.Galanakis@fz-juelich.de
Towards half-metallic interfaces: the Co$_2$CrAl/InP contacts

1. Introduction

A central problem in the field of magneto- or spin–electronics \cite{1} is the spin-injection from a metal into a semiconductor \cite{2}. In principle it is possible to achieve 100% spin-polarized injected current if the magnetic lead is a half-metallic material. These compounds are ferromagnets where there is a band gap at the Fermi level ($E_F$) for the minority spin band while the majority spin band is metallic. In such a compound the behavior of the interface between the half-metal and the semiconductor is of great importance since interface states can kill the half-metallicity. Although from point of view of transport a single interface state does not affect the magnetoconductance since the wavefunction is orthogonal to all bulk states incident to the interface, its interaction with other defect states makes the interface states conducting.

NiMnSb, a member of the Heusler alloys, was the first material to be predicted to be a half-metal in 1983 by de Groot and his collaborators \cite{3}. There exist several other ab-initio calculations on NiMnSb reproducing the results of de Groot \cite{2} and Galanakis et al. showed that the gap arises from the hybridization between the $d$ orbitals of the Ni and Mn atoms \cite{5}. Its half-metallicity seems to be well-established experimentally in the case of single crystals \cite{6}. Also the so-called full-Heusler alloys like Co$_2$MnGe or Co$_2$CrAl were predicted to be half-metals \cite{7} and the gap in the case of these materials arises from states located exclusively at the Co states which are non-bonding with respect to the other atoms \cite{8}.

Although films of both half- and full-Heusler alloys attracted a lot of experimental attention \cite{9, 10, 11}, theoretical calculations for the interfaces of these materials with the semiconductors are few. All ab-initio results agree that half-metallicity is lost at the interface between the Heusler alloy and the semiconductor \cite{12, 13, 14} but the interface dependence of the spin-polarization has not been studied in detail. Even if half-metallicity is lost it is possible that a high degree of spin polarization remains at the interface, as it will be shown in this contribution, and these structures remain attractive for realistic applications.

In this communication I study the (001) interfaces of the half-metallic Co$_2$CrAl Heusler alloy with InP. This Heusler alloy has the same experimental lattice constant with the InP within 1%. I take into account all possible interfaces and show that in all cases a high degree of spin-polarization remains at the interface. In section 2 I discuss the structure of the interface and the computational details and in section 3 I present and analyze my results. Finally in section 4 I summarize and conclude.

2. Computational method and structure

In the calculations I used the full-potential version of the screened Korringa-Kohn-Rostoker (KKR) Green’s function method \cite{16} in conjunction with the local spin-density approximation \cite{17} for the exchange-correlation potential \cite{18}. The results of Picozzi et al. \cite{14} and Debernardi et al. \cite{13} have shown that atomic positions scarcely change at the interface and the dominant effect is the expansion or the contraction of the lattice along the growth axis to account for the in-plane change of the lattice parameter. In the case of the interfaces presented here the compounds have similar lattice parameters and thus perfect epitaxy can be assumed. To simulate the interface I used a multilayer consisted of 15 layers of the half-metal and 9 semiconductor layers. This thickness is enough so that the layers in the middle of both the half-metallic part and the semiconducting one exhibit bulk properties. I have also converged the
Towards half-metallic interfaces: the Co$_2$CrAl/InP contacts

Figure 1. Schematic representation of the (001) interface between Co$_2$CrAl and InP. There are several different combinations at the interface which can be either Co/In, Co/P, CrAl/In (shown in the figure) or CrAl/P. Note that there are two inequivalent cobalt atoms at the interface layer or the subinterface layer. One is sitting at the “bridge” site, continuing the zinc-blende structure of the semiconductor, and the other at the “antibridge” site.

Co$_2$CrAl crystallizes in the $L2_1$ structure. The structure of the interface is shown in figure 1. $L2_1$ structure is similar to the zinc-blende structure and thus perfect epitaxy at the interface can be considered. There are several combinations at the interface, e.g. at the Co$_2$CrAl/InP contact the interface can be either a Co/In one, Co/P, CrAl/In or CrAl/P. I will keep this definition throughout the paper to denote different interfaces. Finally I should mention that since my multilayer contains 15 half-metal and 9 semiconductor layers, I have two equivalent surfaces at both sides of the half-metallic spacer.

3. Results and discussion

Interfaces with respect to simple surfaces are more complex systems due to the hybridization between the orbitals of the atoms of the metallic alloy and the semiconductor at the interface. Thus results obtained for the surfaces as the ones in reference 21 cannot be easily generalized for interfaces since for different semiconductors different phenomena can occur. In Heusler alloys (001) surfaces the appearance of surface states kills the half-metallicity 21 but there are cases like the CrAl-terminated (001) surface of Co$_2$CrAl where spin-polarization is as high as...
Towards half-metallic interfaces: the Co$_2$CrAl/InP contacts

3.1. CrAl/In and CrAl/P interfaces

Firstly I will concentrate my study on the case of the CrAl-terminated Co$_2$CrAl(001) film. In a previous article (see reference [21]) I had shown that the CrAl (001) terminated surface was showing a very high degree of spin-polarization compared to all other surfaces. The mechanism was quite simple: Cr was loosing 4 out of the 8 first neighboring Co atoms and regained the charge it was giving away to cobalts in the bulk case. Most of this charge filled up Cr majority states (in figure 3 the majority peak at the Fermi level moves lower in energy) and its spin moment was strongly enhanced and due to the stronger exchange splitting at the surface the unoccupied Cr states were pushed higher in energy and only the surface state due to the Al atoms survived. Actually a similar phenomenon happens at the interface but now the increase of the spin moment is smaller since Cr d-orbitals hybridise also with the In or P p-states at the interface. This is clearly seen in figure 2 where I have gathered the spin moments for the Cr and Co atoms for both interfaces with In and P. Cr spin moments at the interface are enhanced and reach 2.8$\mu_B$ in the case of the interface with In and 2.6$\mu_B$ in the case of the P interface as compared with the 3.1$\mu_B$ of the Cr in the CrAl-terminated surface. The Cr atoms deeper in the half-metallic spacer have bulklike
Towards half-metallic interfaces: the $\text{Co}_2\text{CrAl/In}_P$ contacts

Figure 3. Spin and atom-resolved DOS for the Cr and Al atoms at the interface with In (long dashed line filled with grey) or P (thick solid line) and the Co atoms at the subinterface layer. With solid line the (001) CrAl surface and with the dashed line the bulk results from references [21] and [8], respectively. The zero of the energy is chosen to correspond to the Fermi level. Positive values of the DOS correspond to the majority spin and negative to the minority.

The next question which arises is if this enhancement of the spin moment of Cr is enough to guarantee a high degree of spin polarization. In figure 3 I have plotted the DOS for the Cr and Al atoms at the interface and the Co atoms at the subinterface layer for both CrAl/In (dashed line filled with grey) and CrAl/P (thick solid line) contacts with respect to the surface (solid line) and bulk calculations (dashed line). At the Cr site the spin-polarization is almost 100% for the CrAl/P case and there is a small DOS for the CrAl/In. For the other three atoms the differences are small between the two different interfaces. Al atom shows a much higher spin polarization at the Fermi level with respect to the surface results while $\text{Co}^{ab}$ shows the inverse behavior. Notice that the scale along the DOS axis for the Al atom is different than for the other three. To make all this more clear, in table 1 I have gathered the spin moments. In the case of the Co atoms the situation is more complicated. There are two inequivalent Co atoms: the one at the “bridge” site ($\text{Co}^b$) and the one at the “antibridge” site ($\text{Co}^{ab}$). At the subinterface layer in general Co spin moments are strongly enhanced and the moments are larger for the Co atoms at the “antibridge” sites. If I add the spin moments of both inequivalent Co’s I notice that the sum is the same for both the CrAl/In and CrAl/P interfaces and around 2 $\mu_B$. If I take into account the band structure analysis for the bulk $\text{Co}_2\text{CrAl}$ presented in reference [8] that means that both majority $e_u$ states are occupied leading to a total Co spin moment of 2 $\mu_B$, while these non-bonding states are unoccupied for the minority band. In the case of the I-3 layer the average Co spin moment is equal to the bulk one and deeper in the film one finds again the bulk values.

The next question which arises is if this enhancement of the spin moment of Cr is enough to guarantee a high degree of spin polarization. In figure 3 I have plotted the DOS for the Cr and Al atoms at the interface and the Co atoms at the subinterface layer for both CrAl/In (dashed line filled with grey) and CrAl/P (thick solid line) contacts with respect to the surface (solid line) and bulk calculations (dashed line). At the Cr site the spin-polarization is almost 100% for the CrAl/P case and there is a small DOS for the CrAl/In. For the other three atoms the differences are small between the two different interfaces. Al atom shows a much higher spin polarization at the Fermi level with respect to the surface results while $\text{Co}^{ab}$ shows the inverse behavior. Notice that the scale along the DOS axis for the Al atom is different than for the other three. To make all this more clear, in table 1 I have gathered the...
Towards half-metallic interfaces: the Co$_2$CrAl/InP contacts

Table 1. Number of states at the Fermi level in states/eV units for the atoms at the interface for the case of the CrAl/In and CrAl/P interfaces as ratios between majority (↑) and minority (↓) spins together with the results for the CrAl-terminated (001) surfaces. The last line is the spin-polarization $P$ taking into account the interface layers and the subinterface ones.

|        | CrAl/In | CrAl/P | CrAl-surf |
|--------|---------|--------|-----------|
| Co$^b$ (↑/↓) | 1.18/0.13 | 0.91/0.10 | 1.03/0.06 |
| Co$^{ab}$ (↑/↓) | 0.72/0.12 | 0.49/0.26 | 1.03/0.06 |
| Cr (↑/↓) | 1.39/0.43 | 1.34/0.09 | 1.48/0.03 |
| Al (↑/↓) | 0.15/0.05 | 0.12/0.05 | 0.01/0.15 |
| In (↑/↓) | 0.09/0.25 | 0.07/0.06 | – |
| Void (↑/↓) | 0.09/0.08 | 0.02/0.01 | – |
| P (↑−↓) | 0.15/0.08 | 0.11/0.08 | – |
| Void (↑/↓) | 0.02/0.10 | 0.07/0.02 | – |

$P$ (↑−↓) | 63% | 65% | 84%

density of states at the Fermi level for all atoms at the interface for both CrAl/In and CrAl/P interfaces together with the results for the CrAl surface. Cobalt has a different behavior depending on which site it sits at and the ones at the “bridge” site behave like in the surface showing a higher spin polarization. As already mentioned the Cr spin-polarization is higher for the case of the contact with P than with In. In the semiconductor film the only noticeable effect is when the In atom is at the interface and it has a large negative spin-polarization while when it sits at the subinterface layer in the case of the CrAl/P contact its net spin-polarization is almost zero. In total the CrAl/In interface shows a spin polarization of 63% and the CrAl/P of 65% as compared to the 84% of the CrAl surface case. This means that in both interfaces more than 80% of the electrons at the Fermi level are of majority spin character and the interface holds a very high degree of spin-polarization.

3.2. Co/In and Co/P interfaces

In the second part of my study I will discuss the case of the interfaces made up by Co and either an In or a P layer. In figure 4 I have gathered the atomic spin moments for the Co atoms at the interface and the Cr atoms at the subinterface layers and their variation in the film. Spin moments at the interface are strongly reduced especially for the Co atoms sitting at the ideal zinc-blende positions, the so-called “bridge” site. The Co$^b$ spin moment decreases down to $\sim 0.3\mu_B$ for the Co/P interface and the quenching of the Co$^b$ spin moment is almost complete in the case of the Co/In interface. On the other hand the Co$^{ab}$ atoms show a more modest decrease of their spin moment by $\sim 0.15 - 0.2\mu_B$ with respect to the bulk value denoted by a straight line in the figure. The Cr atoms at the interface layer (I-1) follow through hybridization the behavior of the Co spin moments and their spin moment is $\sim 0.15 - 0.25\mu_B$ smaller than the bulk value. As soon as one reaches the second layer below the interface, atoms regain a bulklike behavior and moments are close to their bulk values.

The behavior of the Co spin moments at the interface has been also observed in the case of the Co$_2$MnGe/GaAs contacts studied by Picozzi and collaborators [14]. For this compound Co in the bulk has a spin moment of $\sim 1\mu_B$ but at the Co/Ga or Co/As interfaces the decrease of the Co$^b$ is as much as 0.8$\mu_B$ while for Co$^{ab}$ atoms
Towards half-metallic interfaces: the Co_{2}CrAl/InP contacts

Figure 4. Atom-resolved spin moments in $\mu_B$ for Co at the interface (I) and Cr at the subinterface (I-1) layer and their variation in the spacer. Co atoms can sit either at a “bridge” site (Co^b) or an “antibridge” site (Co^{ab}). With the straight horizontal line the bulk values.

The reduction of the spin moment is only 0.2$\mu_B$. It seems that the reduction of the Co spin moment depends strongly on the hybridization between the Co $d$-orbitals and the $p$-orbitals of the semiconductor. Already for Co at the “bridge” site the orbitals hybridise much stronger than in the case of the Co at the “antibridge” site resulting in a larger decrease of the spin moment. Also in the system which I study hybridization is much more important in the case of an In interface layers than of a P one leading to the complete quenching of the Co^b spin moment. Similar results have been obtained in the case of an Fe film capped by GaAs \[22\]. In this case an ad layer of Ga or As on top of the Fe film suppresses the Fe magnetic moments, the effect being particularly pronounced in As-capped case, due to the stronger covalent bonding between the As and the Fe atoms.

Finally I will discuss the atom-resolved DOS at the interface. The hybridization of the Co $d$-states with the $p$ states of either In or P at the interface not only reduces the spin moment but also kills half-metallicity at the Co sites as can be seen for both Co^b and Co^{ab} in figure 4. Cr and Al atoms at the subinterface layer have an environment very similar to the bulk case and although the spin moment of Cr is slightly decreased, as I have already mentioned above, its DOS remains similar to the bulk one and it keeps a very high majority DOS at the Fermi level similar to the bulk DOS. This effect largely compensates the loss of half-metallicity of the Co atoms and the spin-polarization at the Fermi level, if I take into account the layers close to the interface (as in table 1), is $\sim$56\% for the Co/In interface and $\sim$74\% for the Co/P interface. Thus 78\% of the electrons at the Fermi level for the Co/In interface and
Towards half-metallic interfaces: the $\text{Co}_2\text{CrAl}/\text{InP}$ contacts

87% for the Co/P one are of majority character.

In the case of the CrAl interfaces the high spin-polarization was due to the large enhancement of the Cr spin moment which weakened the effect of interface states although the Cr majority DOS at the Fermi level was considerably smaller than in the bulk case; the peak moved lower in energy to accommodate the extra electrons (see figure 3). In the case of the Co interfaces, although Co itself presents almost a zero net spin-polarization at the Fermi level, Cr atoms in the subinterface layer keep the high majority DOS of the bulk (see figure 5) and the resulting spin-polarization is similar to the CrAl interfaces.

4. Summary and conclusions

I have studied the electronic and magnetic properties of the (001) interfaces between the half-metal $\text{Co}_2\text{CrAl}$ and the binary semiconductor InP using a full-potential ab-initio technique. When the interface is made up from a CrAl layer then the Cr spin moment is strongly enhanced at the interface as was the case for the CrAl-terminated (001) surfaces. This enhancement limits the effect of the interface states and in both type of contacts (In or P as interface layer) the interface presents a very high spin-polarization of $\sim$63-65%, thus more than 80% of the electrons at the Fermi level are of majority spin character. On the other hand interfaces made up by Co layers present a large decrease of the Co spin moments but, due the bulklike density of states of the Cr atoms in the subinterface layer, they keep a high degree of spin-polarization: 56% for the Co/In interface and 74% for the Co/P one.

Interface states are important because their interaction with defects makes them conducting and lowers the efficiency of devices based on spin-injection. Thus building
Towards half-metallic interfaces: the Co$_2$CrAl/InP contacts

up interfaces with the highest spin-polarization possible like the ones proposed here is a perquisite but not a guarantee to get highly spin-polarized current in spin-injection experiments.

References

[1] Žutić I, Fabian J and Das Sarma S 2004 Rev. Mod. Phys. 76 323
[2] Wunnicke O, Mavropoulos Ph, Zeller R and Dederichs P H 2004 J. Phys.: Condens. Matter 16 4643; Wunnicke O, Mavropoulos Ph, Zeller R, Dederichs P H and Gröndler D 2002 Phys. Rev. B 65 241306; Mavropoulos Ph, Wunnicke O and Dederichs P H 2002 Phys. Rev. B 66 024416
[3] de Groot R A, Mueller F M, van Engen P G and Buschow K H J 1983 Phys. Rev. Lett. 50 2024
[4] Kulato, V. E. and Mazi, I. 1990 J. Phys.: Condens. Matter 2 343; Halilov S V and Kulatov E T 1991 J. Phys.: Condens. Matter 3 6363; Wang X, Antropov V P and Harmon B N 1994 IEEE Trans. Magn. 30 4458; Youn S J and Min B I 1995 Phys. Rev. B 51 10436; Antonov V N, Oppeener P M, Yaresko A N, Perlov A Ya and Kraft Y 1997 Phys. Rev. B 56 13012; Galanakis I, Ostanin S, Alouani M, Dreyssé H and Wills J M (2000) Phys. Rev. B 61 4093
[5] Galanakis I, Dederichs P H and Papanikolaou N 2002 Phys. Rev. B 66 134428
[6] Kirillova M N, Makhnnev A A, Shreder E I, Dyakina V P and Gorina N B 1995 Phys. Status Solidi (b) 187 231; Hansen K E H M and Mijnarends P E 1990 Phys. Rev. B 34 5009; Hansen K E H M, Mijnarends P E, Rabou L P L M and Buschow K H J 1990 Phys. Rev. B 42 1533
[7] Miura Y, Nagao K and Shirai M 2004 Phys. Rev. B 69 144413; Piccozzi S, Continenza A and Freeman A J 2003 Phys. Rev. B 66 094421; Ishida S, Fuji S, Kashiwagi S and Asano S 1995 J. Phys. Soc. Japan 64 2152
[8] Galanakis I, Dederichs P H and Papanikolaou N 2002 Phys. Rev. B 66 174429; Galanakis I 2004 J. Phys.: Condens. Matter 16 3089
[9] Bach P, Bader A S, Rüster C, Gould C, Becker C R, Schmidt G, Molenkamp L W, Wengand W, Kumpf C, Umbach E, Urban R, Woltersdorf G and Heinrich B 2003 Appl. Phys. Lett. 83 521; Bach P, Rüster C, Gould C, Becker C R, Schmidt G and Molenkamp L W 2003 J. Cryst. Growth 251 323
[10] van Roy W, Wojcik M, Jedryka E, Nadolski S, Jalabert D, Brijs B, Borghs G and De Boeck J 2003 Appl. Phys. Lett. 83 4214; van Roy W, de Boeck J, Brijs B and Borghs G 2000 Appl. Phys. Lett. 77 4190; Schomlka J P, Tolan M and Press W 2000 Appl. Phys. Lett. 76 2005; Ristoiu D, Nouzières J P, Borca C N, Komess T, Jeong H -K and Dowben P A 2000 Europhys. Lett. 49 624; Ristoiu D, Nouzières J P, Borca C N, Borca B and Dowben P A 2000 Appl. Phys. Lett. 76 2349; Giapintzakis J, Grigorescu C, Klini A, Manousakis A, Zorba V, Androulakis J, Viskadourakis Z and Fotakis C 2002 Appl. Phys. Lett. 80 2716
[11] Yang F Y, Shang C H, Chien C L, Ambrose T, Krebs J J, Prinz G A, Nikitenko V I, Gornakov V S, Shapiro A J and Shull R D 2002 Phys. Rev. B 65 174410; Ambrose T, Krebs J J and Prinz G A 2000 Appl. Phys. Lett. 76 3280; Raphael M P, Ravel B, Willard M A, Cheng S F, Das B N, Stroud R M, Bussmann K M, Claassen J H and Harris V G 2001 Appl. Phys. Lett. 70 4396; Chen Y, Basiaga D, O’Brien J R and Heiman D 2002 Appl. Phys. Lett. 83 4301; Elmers H J, Fecher G H, Valdaitsev D, Nepijko S A, Gloskovskii A, Jakob G, Schonhense G, Wurmehl S, Block T, Fehrer C, Hsu P C, Tsai W L and Crann S 2003 Phys. Rev. B 67 104412
[12] Wijs G A and de Groot R A 2001 Phys. Rev. B 64 R202402
[13] Debernardini A, Peressi M and Baldereschi A 2003 Mat. Sci. Eng. C 23 743
[14] Piccozzi S, Continenza A and Freeman A J 2003 J. Phys. Chem. Solids 64 1697; ibid 2003 J. Appl. Phys. 94 4723
[15] Zeller R, Dederichs P H, Újfalussy B, Szunyogh L and Weinberger P 1995 Phys. Rev. B 52 8807
[16] Papanikolaou N, Zeller R and Dederichs P H 2002 J. Phys.: Condens. Matter 14 2799
[17] Vosko S H, Wilk L and Nusair N 1980 Can. J. Phys. 58 1200
[18] Hohenberg P and Kohn W 1964 Phys. Rev. 136 B864; Kohn W and Sham L J 1965 Phys. Rev. 140 A1133
[19] Zeller R 1997 Phys. Rev. B 55 9400
[20] Mavropoulos Ph, Galanakis I and Dederichs P H 2004 J. Phys.: Condens. Matter 16 4261
[21] Galanakis I 2002 J. Phys.: Condens. Matter 14 6329
[22] Erwin S C, Lee S H and Scheffler M (2002) Phys. Rev. B 65 205422