A comparative study of (Fe, Fe$_3$Si)/GaAs and Heusler/MgO for spintronics applications

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Abstract. Many Heusler-like compounds as well as elemental Fe show large spin polarization in their bulk phase, which makes them possible candidates for spintronic applications. However, it has turned out that the magnetic properties may change significantly if the ferromagnet is grown on a semiconductor. Here, we investigate from first principles the magnetic properties of thin Fe, Fe$_3$Si, and Heusler films on GaAs(110) and MgO(001) substrates, which show rather flat interfaces due to the absence of reconstruction. We observe high spin polarization for the Fe containing systems and both Heusler alloys considered here, even for ultra thin layers. These systems are mainly unaffected by the substrate.

1. Introduction

Hybrid structures consisting of alternating metallic and semiconducting layers are of large interest for modern spintronic devices. Important aspects for the suitability of a layered material system are stable magnetism in the vicinity of the interfaces and a suitable density of states (DOS) at the Fermi level $E_F$, $N_{\uparrow \downarrow}(E_F)$ which however strongly depend on the interface structure. The spin polarization indicates its quality for use in transport devices,

$$P = \frac{N_{\uparrow}(E_F) - N_{\downarrow}(E_F)}{N_{\uparrow}(E_F) + N_{\downarrow}(E_F)}. \quad (1)$$

For ballistic transport, knowledge of the complex band structure is necessary to make accurate predictions about transport properties, which is beyond the scope of this paper. Heusler alloys like Co$_2$MnGe seem to be promising ferromagnets (FM) for spintronic devices, because they are half-metallic in their bulk phases [1] and previous investigations have shown a promising large spin polarization of 60.8% at the MgO-interface [2]. Furthermore, epitaxial growth is possible due to the small lattice mismatch of 3.6% and a high TMR of 83% has been measured at room temperature [3]. However, one disadvantage of this ternary system is that annealing is necessary to receive a well ordered L2$_1$ structure [3], because this may cause interdiffusion with the MgO substrate. Recently, Fe-based systems have attracted interest, too. Spin injection up to 30% has been measured [4] for Fe/GaAs(001) as well as 10% [5] for Fe$_3$Si/GaAs(001). Since the free GaAs(110) surface does not reconstruct [6] epitaxial growth can be expected for Fe and the quasi-Heusler Fe$_3$Si, for which the lattice mismatch amounts only to 0.1%. To study the influence of the MgO substrate on Heusler alloys in general, also the Ni$_2$MnGa/MgO system has been investigated, which exhibits a pseudogap in the minority spin channel 0.5 eV below $E_F$ [7] that may be shifted favorably by a suitable tetragonal distortion. As
Figure 1: (a): Schematic view of an (Fe₃Si)₅/(GaAs)₅ multilayer oriented along the (110)-direction. In case of Fe/GaAs(110), the Si atoms have been replaced by Fe. (b): Layer resolved DOS of GaAs at the Fe-interface.

Ni₂MnGa Heusler systems undergo martensitic transformation and are subject to shape memory behavior [7], tetragonally or orthorhombic structures are likely stabilized on corresponding substrates, motivating a closer look on this material.

2. Computational details
All results were obtained by self-consistent density functional theory calculations using the plane wave pseudopotential code VASP [8]. Pseudopotentials generated by the projector augmented wave method [9] were used. For the GaAs/Fe system and the Heusler compounds the generalized gradient approximation of Perdew, Burke and Ernzerhof (PBE) [10] was used to describe the exchange correlation functional. Since PBE slightly overestimates the magnetic moments in Fe₃Si, the PW91 functional [11] was used for the GaAs/Fe₃Si system instead. The ionic positions have been relaxed in all investigated systems. For the two GaAs-based systems the supercell in Fig. 1(a) has been used for our investigations, with varying the number of layers. For the case of pure Fe the Si atoms have to be replaced by Fe. For the Heusler-based systems a similar supercell as described in [2] has been used.

3. (Fe, Fe₃Si)/GaAs
Previous investigations have shown that the spin polarization strongly depends on the interface structure [2]. In case of flat interfaces, a finite spin polarization inside the Fe layer appears even directly at the interface. Additionally, interface states are induced in the semiconducting substrate near E_F by Fe, see Fig. 1(b). These states are highly polarized and are therefore favorable for injection of a spin polarized current. Since in case of the Fe₃Si based system, diffusion at the interfaces is only of minor importance and flat interfaces are energetically favorable [12], we focus on the ideal interface. In contrast to pure Fe bulk, the Fe₃Si system has a negative polarization due to hybridization effects. The quasi-Heusler possesses two inequivalent Fe-positions: Fe II with a bulk-Fe-like chemical environment
Table 1: Polarization $P$ for $(\text{Fe}_3\text{Si})_n/\text{GaAs}(110)$ multilayers of different thicknesses of the $\text{Fe}_3\text{Si}$ film $n$. For each configuration layer resolved $P$ are listed in the columns. Hereby, the layers are numbered relative to the central $\text{Fe}_3\text{Si}$ layer "0". The bulk value of $P$ is given in the last column.

| $n$ | 3       | 2       | 1       | 0   | bulk |
|-----|---------|---------|---------|-----|------|
| 3   | 3       | -11.8   | +35.7   | -40.0 |
| 5   | -41.5   | -14.6   | -28.1   | -40.0 |
| 7   | -38.5   | +1.0    | -25.0   | -37.0 | -40.0 |

Figure 2: Layer resolved magnetic moments of the $(\text{Fe}, \text{Fe}_3\text{Si})/\text{GaAs}(110)$ system. Bulk values at the lattice constant of GaAs are marked with dotted lines.

and Fe I with a B2 FeSi environment. Fe I atoms show an enhancement of the minority DOS due to hybridization with Si states, whereas for Fe II atoms, the DOS at $E_F$ is small as compared to bulk Fe resulting in a polarization of ca 50% as measured experimentally [13, 14]. If $\text{Fe}_3\text{Si}$ is placed onto GaAs(110), the spin polarization $P$ is still large, see Table 1. However, it shows a distinct dependence on the distance from the interface. In the center of the $\text{Fe}_3\text{Si}$ layer and at the interface, bulk-like polarization is obtained, whereas the polarization in the second layer shows strong modification. The polarization is smaller or in some cases positive. This seems to be related to a reduction of the spin-down DOS of the Fe-atoms and decreases with increasing thickness of the FM layer, i.e., for bulk-like layers the effect may be less dramatic.

Besides the polarization, the magnetic moments of the $(\text{Fe}, \text{Fe}_3\text{Si})/\text{GaAs}$ systems have been investigated. In case of 3 Fe-layers between the GaAs substrate, the magnetic moments in the center layer are increased due to ionic relaxations which lead to an increase of the atomic volume in the center layer. With at least 5 Fe-layers, this finite size effect is suppressed indicating a proper modeling of the system. The magnetic moments at the direct interface layer are enlarged in comparison to the bulk value [15] as the Fe-coordination at the interface is reduced.

The magnetic moments obtained for bulk $\text{Fe}_3\text{Si}$ are 1.34 and 2.57 $\mu_B$, i.e., the average magnetic moment corresponds to 1.75 $\mu_B$, which is in agreement with previous calculations [16]. The experimental magnetic moment is slightly smaller being 1.6 $\mu_B$ [17]. As in case of pure Fe, enhanced magnetic moments are observed at the GaAs-interface, see Fig. 2. Besides small oscillations due to local volume changes, the inner layers show mainly bulk behavior.

4. $(\text{Co}_2\text{MnGe}/\text{Ni}_2\text{MnGa})/\text{MgO}$

For both Heusler/MgO systems we obtained as lowest-energy configurations Co/Ni-terminated surfaces with the Co/Ni-atoms posed on top of the O-atoms in the underlaying MgO-layer. The behavior of the magnetic moments is similar in both systems, see Table 2. For the Co-based system, we find a reduction of the magnetic moments only in the direct surface region, while at two monolayer distance towards the surface the magnetic moments approach their bulk value. This is in close agreement with previous measurement of the magnetic moments at the $\text{Co}_2\text{MnGe}/\text{MgO}$ interface [18]. In contrast, the magnetic moments in the Ni-based system are slightly enlarged in comparison to the bulk value. Thereby an artificial increase of the moments
Table 2: Magnetic moments of Co$_2$MnGe/Ni$_2$MnGa on top of MgO in $\mu_B$. The layers are numbered with respect to their interface distance. The bulk values are given for the lattice constant of MgO.

| layer       | 5  | 4  | 3  | 2  | 1  | 0  | bulk     |
|-------------|----|----|----|----|----|----|----------|
| Co/Ni       | —  | 0.92/0.42 | —  | 0.85/0.4 | —  | 0.47/0.43 | 0.96/0.36 |
| Mn/Mn       | 3.11/3.45 | —  | 3.09/3.44 | —  | 2.85/3.48 | —  | 3.10/3.39 |
| Ge/Ga       | -0.06/-0.05 | —  | -0.06/-0.05 | —  | -0.05/-0.05 | —  | -0.06/-0.06 |

in the central layer as in the Fe-based system appears. In summary, only little modification of the magnetic properties due to the MgO-interface is found for both systems. The polarization of the Ni-atoms at the interface is about -37% and is therefore not comparable with Co$_2$MnGe which shows a polarization of ca 61% at the MgO interface [2].

5. Conclusions
The magnetic moments at the GaAs(110) interface are stable for both Fe-based systems. Even in case of very thin films, finite moments are obtained at the interface. For the Fe/GaAs(110) system the investigated aspects are promising for spintronic applications. In case of flat interfaces the system does not only show a finite polarization at the direct interface, also the interface-induced states in the bandgap of GaAs are largely polarized. The polarization of the Fe$_3$Si-system is less stable for thin films and shows oscillation with the interface distance. Therefore, the ideal Fe$_3$Si system is less advantageous than the Fe-system. Future work has to show if variation of composition or any alloying can circumvent this problem. Also the oscillation may be less dramatic for thicker Fe$_3$Si layers. The Co-based Heusler system is even more promising for applications as finite magnetic moments and a huge polarization appear at the MgO interface. In contrast, the Ni-based system does not show any advantage over the Fe-based systems.

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