Interface structure and magnetism of Fe$_3$Si/GaAs(110) multilayers: An ab-initio study

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Ferromagnet-semiconductor (FM/SC) hybrid structures have attracted much interest in view of materials being suitable for spintronic devices. Such devices demand a high spin-polarization at the FM/SC interface, as is provided by Fe$_3$Si. Here, we investigate the hybrid system consisting of Fe$_3$Si and GaAs which is of special interest due to its tiny lattice mismatch. The multilayers containing 1–3 layer thick Fe$_3$Si films are oriented in the (110) direction. Using the (110) orientation of GaAs has the advantage that no surface reconstruction has to be considered. Free GaAs(110) surfaces show only a rippling but no reconstruction. We investigate the structural and magnetic properties of thin layers of Fe$_3$Si on GaAs(110) using density functional theory. Therefore we choose a multilayer geometry in order to avoid additional contributions from the surface. The ground state structure turns out to be the same, independently of the number of FM layers, and the rippled GaAs(110) surface is flattened if overgrown by Fe$_3$Si. Moreover, we find the system being stable against diffusion effects which leads to average magnetic moments of the magnitude of bulk Fe$_3$Si. Concerning the magnitude of the magnetic moments, our calculations suggest that Fe$_3$Si/GaAs(110) may be a good candidate for magnetoelectronics.

Keywords: ab-initio; electronic structure; Fe-based alloys; GaAs; interfaces; magnetic films; multilayer

1. Introduction

The search for materials that are appropriate for magnetoelectronic devices is still intensively pursued. Suitable ferromagnets should have a high spin-polarization like many Heusler alloys. These systems are often half-metallic in their bulk phase, which naturally leads to a high spin-polarization. Besides the ‘classical’ ternary A$_2$BC Heusler compounds with L1$_2$ structure, also the quasi-Heusler system Fe$_3$Si with DO$_3$ symmetry is of interest for spintronic injectors, because its spin-polarization is about 43% and can be regarded as nearly half-metallic [1]. In its bulk phase, Fe$_3$Si has two types of Fe atoms with magnetic moments of 1.2–1.35μ$_B$ and 2.2–2.4μ$_B$, respectively, and a Curie temperature of 840 K [1,2]. Furthermore, it has been found that ferromagnetism can be conserved even in very thin Fe$_3$Si layers with a nominal thickness of two monolayers [3]. Altogether, these features make this system an interesting candidate for spintronic applications [4].

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First experimental realizations of heterostructures with Fe$_3$Si were made on GaAs(001) and Si(111). GaAs is of special interest, because the lattice mismatch between GaAs and Fe$_3$Si amounts only to 0.1% which should support epitaxial growth of the multilayers. However, it is well known that GaAs(001) surfaces are highly reconstructed. The reconstruction depends on the preparation of the system; i.e., in case of As-rich surfaces a $\beta(2 \times 4)$ reconstruction is observed [5] while for Ga-rich surfaces ($4 \times 6$) or ($2 \times 4$) surface reconstructions are common [6]. The growth properties and the interface magnetism seem to depend on the details of the GaAs(001) surface [7,8]. A promising alternative in which the problem of complex surface preparation and reconstruction can be avoided, is the use of (110) oriented GaAs instead. This surface does not show any reconstruction; only surface relaxation effects can be observed. Typically, the GaAs(110) surface has a rippled structure, which means that the Ga and As atoms on the surface layer are shifted in the growth direction. From experiments with low-energy electron diffraction (LEED) techniques, it is known that the Ga atoms of the surface layer tend to move into the GaAs film layer the As atoms move outwards [9,10]. This can be related to a rotation angle of about 27–28° between the ideal and actual As-Ga bond (projection onto the (001) plane) [9]. GaAs(110) is obtained experimentally by cleaving such that the surface has the same structure as bulk GaAs and can then be overgrown by Fe$_3$Si which also has its bulk composition in the (110) planes. So far, there exist some experimental results for Fe/GaAs(110) [11,12], but not much work has been done to study the magnetic properties of Fe$_3$Si/GaAs(110) interfaces. In this paper, we investigate the interface structure and the magnetic configuration of thin Fe$_3$Si films on GaAs(110) by using multilayers instead of a single FM film on a GaAs(110) substrate. This factors out the influence of the surface on the magnetic moments and on the structure of the FM/SC system and we will be able to discuss interface effects only. The calculations are performed using density functional theory as implemented in the Vienna ab-initio Simulation Package (VASP) [13,14]. Ionic relaxation has been taken into account for one and three layers of Fe$_3$Si to follow up the question of whether the rippling of the GaAs surface survives if overgrown by Fe$_3$Si. Furthermore, it is known that Fe does not necessarily grow layer-wise on GaAs. Therefore, we performed calculations for (Fe$_3$Si)$_1$/GaAs(110) including diffusion at the interfaces.

2. Computational details

In order to study the interface magnetism and the structure of Fe$_3$Si on (110)-oriented GaAs, multilayers consisting of alternating five layers GaAs and 1–3 layers Fe$_3$Si have been investigated using periodic super-cells with 14–22 atoms. We make use of the VASP code [13,14] with the implementation of the projector augmented wave (PAW) method [15] and the generalized gradient approximation (GGA) in the formulation of Perdew and Wang (PW91) [16] for the exchange correlation potential. The calculations have been performed with a periodicity of one lattice constant in the lateral directions using a tetragonal unit cell. A top view of the FM/SC interface layers is shown in Figure 1. In all calculations, the experimental lattice constant of GaAs $a = 5.65 \, \text{Å}$ has been used. The theoretical lattice constant of bulk GaAs is generally underestimated by density functional theory using the GGA [17]; however, LDA is not suitable for Fe, and the use of the too-large theoretical lattice constant may lead to artifacts in the magnetic moments.
In order to determine the ground state configuration of the FM on the semiconducting layer, we first kept the positions of the two subsystems fixed and placed the FM on a regular grid of $6 \times 6$ positions for one layer and of $4 \times 4$ positions for the systems with two and three layers of Fe$_3$Si GaAs(110). Ionic relaxations have been performed by using a Davidson Block iteration scheme, subsequently keeping the volume and cell shape fixed.

3. Results

The energies of the investigated interface structures of Fe$_3$Si on GaAs(110) are summarized in Figure 2. The results are plotted over the area of an in-plane unit cell with the Ga atoms sitting at the corner positions where the numbers on the axis mark the position of the Fe$_1$ atom, cf. Figure 1. In the case of an odd number of layers, the two FM/SC interfaces are equal and we expect similar energy surfaces in both cases whereas

Figure 1. Schematic top view of the zinc-blende GaAs(110) (left) and Fe$_3$Si (right) unit cells. The numbers in the right panel refer to chemically inequivalent positions when Fe is placed on GaAs(110); for details see text.

Figure 2. Constant energy surfaces of Fe$_3$Si/GaAs(110) for 1, 2, and 3 FM layers as shown in (a), (b), and (c), respectively. The coordinates of the [001] and [−110] axis are the positions of the Fe atoms marked by ‘1’ in Figure 1. The energy increases with the brightness of the colour, i.e., dark colours denote low energies. The distance of the contour lines is 0.4 eV and the minimum positions ($E = 0$ eV) are located in the middle of the [−110] axis. The plot range covers the surface of one in-plane unit cell with Ga atoms sitting at the corner positions and As on the (0.5, 0.25) in-plane position.
for two layers of Fe$_3$Si, the two FM/SC interfaces are different. However, without relaxation the most stable structure is the same for all investigated systems, see Figure 2. The atoms of the FM layer are placed in the interstitial positions of the GaAs(110) layer as shown schematically in Figure 3a. For two layers of Fe$_3$Si, a second minimum occurs at the centre of the energy surface, due to symmetry reasons, cf. Figure 2b. This minimum corresponds to a configuration where the Si and Fe$_2$ positions of the interface layer are reversed and the second Fe$_3$Si layer has the configuration shown in Figure 3a. The most unlikely configurations occur for Si and Fe atoms sitting on the same positions as the Ga and As atoms in the outermost GaAs layer. These configurations have 5–7 eV higher energy than the ground state configuration and correspond to the white areas in Figure 2. The energy contribution of the interface can be estimated from

$$E/N = a + b/N$$

where $N$ corresponds to the number of atoms in the unit cell and $E$ is the total energy of the system ($a$ and $b$ are fit parameters). Then the slope of the straight line $E/N$ determines the interface contribution to the energy. Here, we obtain approximately 23 eV, which is only a rough estimate because this formula is properly defined only in the limit of infinitely thick layers.

In order to determine the deviations from the ideal structure, we restrict the discussion to systems with identical interfaces; i.e., one and three layers of Fe$_3$Si. It turns out that the ground state structure is not much effected by relaxation. The interface is basically the same except for some relaxation effects. The structure being lowest in energy after ionic relaxation is sketched schematically in Figure 3b. The As atom of the interface layer is moved in the [001] direction by 13% from the ideal position whereas the As atoms in the layer lying below move in the opposite direction, but this effect is already extremely small (0.05%). The relaxation effects are not limited to atoms moving in the in-plane directions. We observe an additional displacement of the Ga and As atoms perpendicular to the surface. In the case of a free GaAs(110) surface, it is expected that the Ga atoms move inwards and the As atoms move outwards from the surface, which leads to the rippled surface mentioned in the introduction. A displacement of the top GaAs layer can also be observed in the Fe$_3$Si/GaAs(110) system. However, in our case both atoms – Ga and As – move outwards whereby the displacement of the As atom is more expressed being 2.5 times larger as compared to the displacement of the Ga atom. The Ga-As bond length is thereby enlarged by 9.4% which is not assumed to occur for a free GaAs(110) surface [18]. It should be noticed that this change cannot result from fixing the lattice constant to the experimental value, because the relaxation effect is much larger than the lattice mismatch. The change of the bond length and the displacements cause the rippling of the surface.

Figure 3. Schematic top view of the Fe$_3$Si/GaAs(110) interface without ionic relaxation (a) and including relaxation (b). The relaxation mainly affects the As atom.
nearly to vanish. Experiments have shown that rippling is related to a rotation angle which measures the angle between the Ga-As bonds projected in the (001) plane relative to the ideal orientation of the bond. Instead of a rotation angle of $27°$–$28°$ observed in [9,18], a rather small angle of approximately $2°$ results from the present calculations, which means that the rippling or buckling is destroyed if the surface is overgrown by Fe$_3$Si at least in the case of thin FM layers separating the GaAs layers.

Besides the previously discussed relaxation effect, we observe a lateral adjustment of the interface atoms. The Ga, Fe$_2$ and Si interface atoms are moved in the minus $[-110]$ direction, whereas the other Fe atoms and As are shifted to the opposite direction. This seems to be an artifact of the lattice constant being fixed.

For all systems discussed so far we have neglected diffusion effects. Experiments with Fe/GaAs(110) indicate that good epitaxial growth can be expected [19]. However, we deal with very thin FM layers and we could not naively assume perfect layer by layer growth. In order to study the influence of possible interface roughness, we have systematically interchanged the positions of Ga and As with the Fe atoms of the FM layer. As we use only a small in-plane unit cell, any interchange of atomic positions presents a large effect which may not occur experimentally. However, it can give a rough estimate of the importance of disorder in the system.

Here, we present only results for the case of one layer of Fe$_3$Si for which we expect the effects discussed above to be largest. In Figure 4 we compare the energy of the ideal system to the energy of the systems where Fe atoms are interchanged with Ga or As. Independently of the Fe position (cf. Figure 3a), replacing Ga atoms by Fe, the required energy input is lower by a factor of three compared to the case in which As atoms are interchanged with Fe atoms. This may be understood from the fact that interchanging Ga and Fe creates zinc-blende Fe-As layers and it has been shown that in a zinc-blende lattice FeAs is more stable than FeGa [17]. However, none of the calculated configurations is lower in energy than the ideal structure discussed previously. From all the investigated

![Figure 4. Increase of the total energy $\Delta E$ of $(\mathrm{Fe}_3\mathrm{Si})_1/\mathrm{GaAs}(110)$ due to interchange of Ga and As atoms with Fe in the interface layers relative to the energy of the unperturbed system with ideal interfaces. The $x$-axis is the same as in Figure 1. Circles mark the interchange of Ga with Fe and squares denote the results for interchange of As and Fe, respectively. Filled symbols quote results without relaxation whereas open symbols mark the energy difference for relaxed structure. The lines are only guides for the eyes.](image-url)
disordered structures the interchange of Fe$_2$ with Ga requires the smallest energy. This configuration is only 51 meV/atom higher in energy compared to the ground state energy for the case without interdiffusion. The energy of 51 meV/atom corresponds to 590 K which is in the range of the substrate temperatures during growth [3]; thus, this configuration may eventually be stabilized thermodynamically.

Taking into account ionic relaxation does not change the picture in an essential way. The trends remain mostly the same; only the energy differences are slightly changed, see Figure 4. The energy costs of interchanging Ga with Fe$_2$ amounts to 32.82 meV/atom for the relaxed structures and is therefore still not favourable at $T=0$ K. In conclusion, we expect no significant contributions from interdiffusion or roughness at low temperatures.

In experiments, however, multilayers are grown at elevated temperatures which may involve interdiffusion at the interfaces to become important.

As expected, the interchange of Ga and Fe$_2$ leads to a shift of the atomic positions. However, for most atoms this effect is only about 0.06 Å except for the Ga atom in the FM layer where it shows a significant in-plane distortion. In contrast to that, the rearrangements in the FeAs (the former GaAs interface) layer are small. The displacements in the perpendicular direction are rather small except for the As and Ga atoms. For the Ga atom in the FM layer we observe a shift of 2% outwards of the plane, i.e., in the $-[110]$ direction. This seems to be correlated to a displacement in the second GaAs layer in which As atoms are shifted in the $[110]$ direction. Furthermore, the largest change is observed for the configuration with the interchange of As and Fe$_3$, see Figure 4. A detailed analysis of the structure shows that in this case Ga and As atoms move out of the interface layer of the GaAs film, i.e., the two subsystems try to separate.

As mentioned before, in the context of spintronic applications it is important to find FM materials with high spin polarization which remains unaffected if contacted with a SC material. In Figure 5 the average magnetic moments of the Fe atoms of the ground state configurations of $(\text{Fe}_2\text{Si})_n$/GaAs(110) ($n=1–3$) systems are summarized. The magnetic

![Figure 5](image_url)  

Figure 5. Average magnetic moments of the Fe atoms in $(\text{Fe}_2\text{Si})_n$/GaAs(110) with $n=1–3$ for the respective ground states. Circles denote the magnetic moments for systems without relaxation and squares give the magnetic moments after relaxation of the multilayer. The dashed and dotted lines mark the bulk values of the magnetic moments for bcc Fe and DO$_3$ Fe$_3$Si, respectively. The full lines are only guides for the eyes.
moments show a distinct dependence on the thickness of the FM layer such that they decrease with increasing number of Fe$_3$Si layers. In the case of a single FM layer, the average Fe spin moment corresponds to the value obtained for bulk bcc Fe $\bar{m} = 2.2 \mu_B$. Without lattice relaxation, the magnetic moments immediately drop down to $\bar{m} = 1.74 \mu_B$ or $\bar{m} = 1.46 \mu_B$ for two and three layers of Fe$_3$Si, respectively. If we include lattice relaxation, the magnetic moments recover, see Figure 5. For the system with $n = 3$ we obtain a magnetic moment of $\bar{m} = 1.8 \mu_B$, i.e., 18% higher as for the non-relaxed structure. The main contributions to the enhanced magnetic moment stem from the two interface layers, whereas the moments of the central FM layer stick to the experimental values for Fe$_3$Si. Until now, there are no experimental values for magnetic moments in (Fe$_3$Si)$_n$/GaAs(110) with thin FM layers; results are only reported for the (001) orientation [3]. For the (001) direction the magnetic moments determined by a magnetic force magnetometer are close to the value of bulk Fe$_3$Si, cf. Figure 5. The magnetic moments obtained from the present calculations are enhanced compared to the moments of bulk Fe$_3$Si, which may depend on the geometry of the system and the tiny thickness of the film. Since the magnetic moments decrease with increasing number of FM layers, we assume that the bulk value will be approached for thick enough FM layers. This has to be verified in future investigations for larger super-cells.

If epitaxial growth is somehow distorted such that interdiffusion can take place, the magnetic structure changes drastically at least for thin FM layers which is shown here for the case $n = 1$. Keeping the atomic positions fixed, the disordered systems behave similarly to the ideal system, except for the interchange of Ga and the Fe$_2$, which has the lowest energy, see Figure 6. For this configuration the magnetic moment amounts to $\bar{m} = 1.26 \mu_B$ only, being 35% smaller as compared to the system without disorder. In contrast to the previously discussed systems with ideal interfaces, relaxation results in further decrease of

![Figure 6](image_url)
the average magnetic moments having values already below the corresponding bulk value of Fe₃Si. The actual size of the magnetic moment or its reduction depends on the details of the corresponding structure, e.g., in the case of Ga on the Fe₂ position, the magnetic moment of Fe in the zinc-blende FeAs layer amounts to \( m = 0.9 \mu_B \) only. The size of the moments for atoms on the Fe₁ and Fe₃ positions are \( \tilde{m} = 1.1 \mu_B \) and \( \tilde{m} = 1.9 \mu_B \), respectively.

The interdiffusion effects which have been modelled in our calculations are exaggerated, when assuming the complete replacement of Ga by Fe in the interface layer. Nonetheless, we can conclude that if no epitaxial growth of Fe₃Si on GaAs(110) is achieved and Fe penetrates into the GaAs layer, the moments will significantly decrease.

4. Conclusion

We have investigated the interface structure and the magnetic properties of Fe₃Si/GaAs(110) in the limit of thin FM layers. The calculations have been carried out within ab-initio density functional theory using the VASP code with PAW pseudopotentials and the GGA. In all calculations we used the experimental lattice constant of GaAs instead of the calculated minimum lattice constant, because the lattice constant is overestimated by density functional theory using the GGA and therefore, may cause artifacts in the magnetic structure. However, small deviations of the atoms from the ideal positions may be related to the use of the experimental value. Concerning the general trends obtained in our calculations this can be considered a minor influence.

First, the minimum configuration of the Fe₃Si system on GaAs(110) has been determined for 1–3 layer thick films of Fe₃Si assuming ideal interfaces, because GaAs(110) does not show any surface reconstruction. The most stable configuration for all three thicknesses was obtained for Fe and Si atoms sitting on the interstitial positions on top of the GaAs layer. The GaAs interface layers do not show the well-known rippling of the (110) oriented GaAs surfaces. Therefore, the rotation angle amounts only to about 2° instead of 27–28° for the free GaAs(110) surface. The present calculations give no evidence for interdiffusion or disorder at the interfaces at low temperatures. At higher temperatures, disorder may occur where an interchange of Ga and Fe would be favoured. However, it should be kept in mind that we use a relatively small super-cell in which the diffusion concentrations are extremely large. For a more detailed description, i.e., realistic concentrations, larger cells have to be investigated in future work. Nonetheless, the present results point out a trend.

Furthermore, we investigated the magnetic structure of the FM/SC hybrid system. The average magnetic moments are larger than in bulk Fe₃Si, however they decrease with increasing thickness of the FM and seem to approach the bulk value for thicker layers. Lattice relaxation is essential to obtain the correct magnitude of the magnetic moments, because without ionic relaxation the Fe moments turn out to be underestimated by 20% in the case of three FM layers. If the interface structure deviates from the ideal composition the magnetic moments are strongly reduced and relaxing the structure leads to an even stronger reduction of the magnetic moments.
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