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Half-metallic interface between a Heusler alloy and Si

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Abstract

The interface between the half-Heusler alloy CoFeSi and Si is investigated by using first-principles density-functional calculations. Although CoFeSi has not been fabricated yet, its formation energy turns out to be negative. Within the generalized gradient approximation, CoFeSi shows nearly half-metallic properties, and its lattice constant is about 5.38 Å; this value is relatively close to the lattice constant of Si. We here chiefly investigate the CoFeSi/Si (110) interface, and find that the half-metallic properties are almost preserved at a specific (110) interface. Furthermore, the interfacial structure which leads to the high spin polarization has the lowest energy of the (110) interfacial patterns examined in this work. The half-metallicity at the interfaces is similarly observed in the densities of states projected onto delocalized sp states, and this suggests the relevance of the high spin polarization to transport properties.

1. Introduction

Designing half-metal/semiconductor interfaces where spin polarization at the Fermi energy remains very high is of considerable interest because highly spin-polarized character at an interface is expected to facilitate the efficiency of spin injection into semiconductors [1–3], especially when the injection is carried out via tunneling through a Schottky barrier [4, 5]. However, first-principles calculations have shown that finding half-metallic interfaces is not easy. Indeed, only a few interfaces have been predicted to possess half-metal-like electronic structures from first principles [6–11]. In particular, with respect to the heterostructures consisting of a ferromagnet and *silicon*, no half-metallic interface has been proposed, while injection into Si is quite significant for developing spintronics devices which are well integrated into conventional Si-based electronics system.

In this paper, looking for a possible half-metallic interface, we investigate half-metal/Si heterostructures using first-principles density-functional calculations. Here, as a half-metal, we pick out the half-Heusler alloy CoFeSi (though, strictly, this substance is not completely half-metallic). The lattice constant of CoFeSi calculated within the generalized gradient approximation (GGA) is about 5.38 Å, which is relatively close to the lattice constant of Si (5.43 Å at room temperature), while most half-metals so far proposed have a little larger lattice mismatch with Si [12]. It should be noted at this point that CoFeSi is thermodynamically unstable and has not been fabricated yet. Nonetheless, by considering the fact that the full Heusler alloy Co_2FeSi does exist, it still looks possible to grow CoFeSi by choosing the concentration of Co. Particularly, growth on Si substrates seems to be helpful for fabricating CoFeSi from the viewpoint of lattice mismatch; the lattice constant of Co_2FeSi is 5.64 Å [13] and leads to somewhat large lattice mismatch with Si (∼4%).

Our calculations focus on the CoFeSi/Si (110) interface, and show that high spin polarization is preserved at a certain interfacial pattern. Furthermore, the interfacial pattern with high spin polarization turns out to be energetically favorable. These findings are considered encouraging for further study of CoFeSi as a source substance for spin injection into Si.

2. Computational details

Our first-principles density-functional calculations are performed within the GGA by using the exchange–correlation energy form parameterized by Perdew *et al* [14]. We use a plane-wave basis set with a 270 eV cutoff and the projector augmented-wave (PAW) method [15, 16]. These calculations

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are implemented within the Vienna *ab initio* Simulation Program (VASP) [17–19].

To deal with a CoFeSi/Si (110) multilayer, which consists of eight-atomic-layer CoFeSi and ten-atomic-layer Si, the size of the supercell is set to be 3.80 Å × 5.37 Å× ∼ 34 Å; the in-plane lattice parameters are in accordance with those of bulk CoFeSi, and the length of the c-axis is optimized depending on the interfacial structures. The number of $\boldsymbol{k}$-point mesh is chosen to be $n_k = 6 \times 4 \times 1$ when carrying out structural optimization. But it is increased to $n_k = 13 \times 9 \times 1$ for calculating the local density of state (LDOS), spin polarization, and so on, with the broadening parameter of the size of 0.02 eV.

Regarding the calculations of bulk properties of Co_xFeSi, the above computational conditions are slightly changed. When the volume optimization is performed, the cutoff energy is increased to 335 eV, and the number of $\boldsymbol{k}$-point mesh is set to $n_k \sim 1.5 \times 10^5$ Å$^3/v_c$, where v_c is the volume of unit cell. For the calculations of density of state, n_k is further increased to $n_k \sim 4.0 \times 10^5$ Å$^3/v_c$.

3. Results and discussions

3.1. Bulk properties of CoFeSi

The density of states of CoFeSi is presented in figure 1(a), which shows that the electronic structures are almost half-metallic. Indeed, the magnetic moment per CoFeSi is very close to an integer value multiplied by the Bohr magneton (2.99 $\mu_{\rm B}$).

We also look into the formation energy of Co_xFeSi to examine the stability of CoFeSi. The formation energy is now defined by $F = E - \sum_i \mu_i N_i$, where E is the energy of Co_xFeSi, N_i is the number of the ith element (i = Co, Fe, and Si), and μ_i is the chemical potential of the ith element; the chemical potential is here chosen to be the total energy per atom of each elemental bulk. The unit cell which at most contains $\text{Co}_8\text{Fe}_4\text{Si}_4$ is utilized. Then, the number of Co atoms in this unit cell is changed from eight (full Heusler) to four (half-Heusler), where the configurations of Co vacancies in the unit cell are chosen so that the Co vacancies are as far apart from each other as possible. At each Co concentration, structural relaxation including cell volume (and cell shape for the case of six Co atoms) is carried out. The resulting formation energy is presented in figure 1(b), where x is changed from $x = 1$ (CoFeSi) to $x = 2$ (Co_2FeSi). As is expected, the formation energy of CoFeSi is higher than that of Co_2FeSi. Even so, it is interesting to note that the formation energy of CoFeSi is still negative. This implies that controlling the Co concentration upon growth works out for the fabrication of CoFeSi, especially when the growth is carried out epitaxially on an Si substrate whose lattice mismatch with CoFeSi is rather small.

For the purpose of checking the local stability of CoFeSi with respect to atomic displacements, we have also analyzed the phonon modes. By using a supercell consisting of a $2 \times 2 \times 2$ array of CoFeSi primitive cells, frozen phonon calculations are performed. The resulting phonon modes give rise to no imaginary frequencies. This suggests that half-Heusler CoFeSi is mechanically stable and can continue to exist as a metastable state.

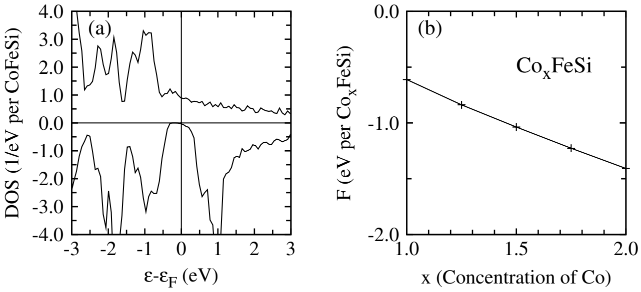

Figure 1. (a) Density of states of CoFeSi. (b) Formation energy per Co_xFeSi as a function of Co concentration, x.

Moreover, the stability regarding the disorder between Co atom and Co vacancy in CoFeSi is investigated by using the Korringa–Kohn–Rostoker method with the coherent potential approximation[1]. We have calculated the energy for ordered and disordered structures, that is, $(\text{Co}_{1-y}, \text{v}_y)(\text{v}_{1-y}, \text{Co}_y)\text{FeSi}$ with y set to 0.0 and 0.5, respectively, where 'v' stands for the Co vacancy. The energy comparison shows that the ordered structure is more stable than the disordered one, and their energy difference is ∼0.06 eV per CoFeSi. It is worth noting that this energy difference is comparable to that calculated for the well-known half-Heusler alloy NiMnSb (∼0.05 eV per NiMnSb).

3.2. Structures at interface

We choose four initial structures as CoFeSi/Si (110) interfacial patterns, where the diamond structure of Si is connected to one of the paths inside CoFeSi; these paths are Co–Fe, Co–Si, vacancy–Si, and vacancy–Fe ones. We call those structures Si–(Co–Fe), Si–(Co–Si), Si–(v–Si), and Si–(v–Fe), respectively. The initial and optimized interfacial structures are shown in figure 2. In the cases of Si–(Co–Fe) and Si–(Co–Si), the atomic positions remain similar to the initial ones. On the other hand, in the cases of Si–(v–Si) and Si–(v–Fe), the structural reconstruction is remarkable because one of the Si atoms on the silicon plane does not have a neighboring atom on the CoFeSi plane.

Table 1 shows the energy of each interface relative to that of the Si–(Co–Si) interface. The most stable structure of the four is the Si–(Co–Si), the second most stable is the Si–(v–Si), and the other two structures have much higher energy. One possible reason for the stability of Si–(Co–Si) is that the atomic configurations at the interface are similar to those in the bulk states. Actually, there appears a Si–Si bonding between CoFeSi and Si slabs at the interface. Also, there is a Co–Si bonding at the interface which looks naturally connected to the Co–Si path inside CoFeSi.

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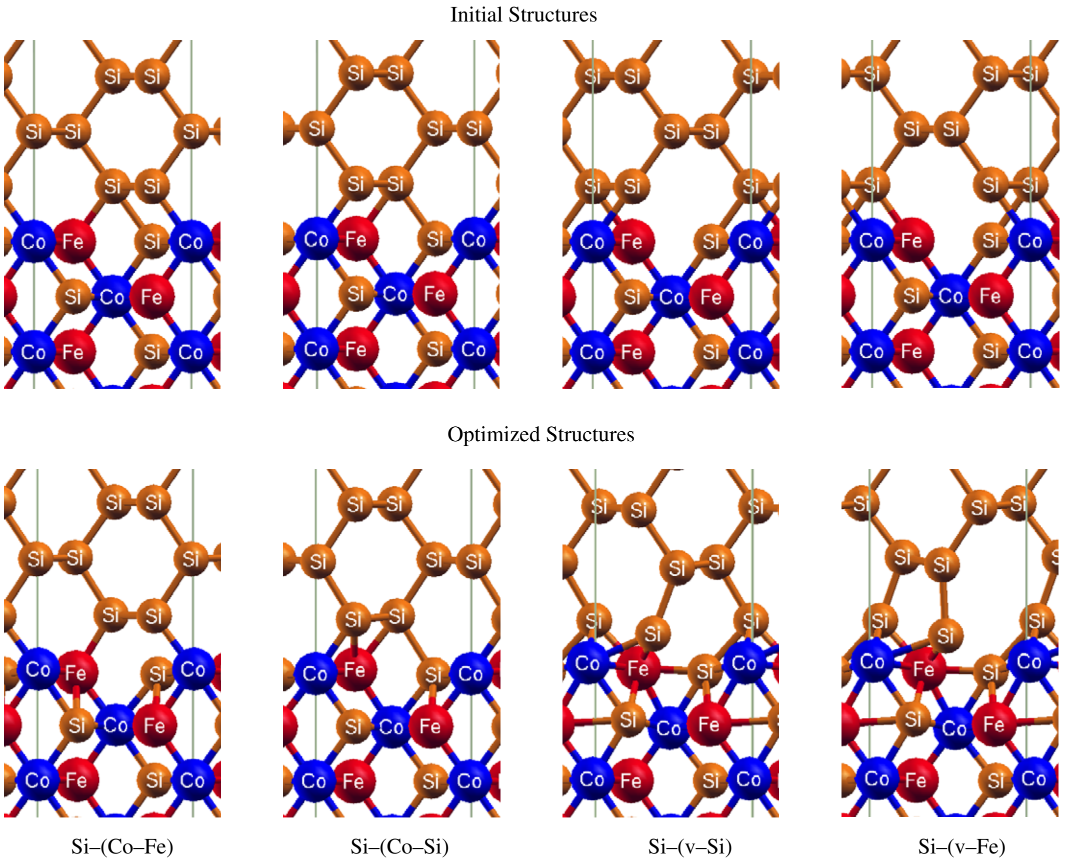
[1] The calculations are performed using the *ab initio* calculation code developed by Akai *et al*. See for example [20]. For these computations, another form of the exchange–correlation energy [21] is used.


Initial Structures

Optimized Structures

Si–(Co–Fe) Si–(Co–Si) Si–(v–Si) Si–(v–Fe)

Figure 2. Interfacial patterns investigated. Initial structures are shown in the upper row, and their optimized structures in the lower row. See the text for the way of denoting each interfacial pattern.

(This figure is in colour only in the electronic version)

Table 1. Energy of the interface per cell area (3.8 Å × 5.37 Å) relative to that of the Si–(Co–Si) interface. See the text for the way of denoting each interfacial pattern.

Interfacial pattern	ΔE (eV)
Si–(Co–Fe)	0.685
Si–(Co–Si)	0.000
Si–(v–Si)	0.008
Si–(v–Fe)	0.343

3.3. Density of states and spin polarization at the interface

LDOS at the Fermi energy and local spin polarization are examined with respect to the most stable Si–(Co–Si) interface. Now, local spin polarization is defined by $P = [D_{\uparrow}(\epsilon_F) - D_{\downarrow}(\epsilon_F)]/[D_{\uparrow}(\epsilon_F) + D_{\downarrow}(\epsilon_F)]$, where $D_{\sigma}(\epsilon_F)$ is the LDOS at the Fermi energy for spin direction σ. It is clearly seen in figure 3(a) that $D_{\downarrow}(\epsilon_F)$ is almost zero even in the vicinity of the interface, namely, highly spin-polarized properties are well preserved there. Besides, figure 3(a) tells that gap states inside Si decay swiftly, for $D_{\uparrow}(\epsilon_F)$ shows a rapid decrease toward zero. Yet, notice that $D_{\uparrow}(\epsilon_F)$ is much larger than $D_{\downarrow}(\epsilon_F)$ inside Si, and spin polarization is, therefore, kept markedly high (figure 3(b)). Spin polarization of gap states is thought to be closely related to tunneling current through Si. Thus, the behavior of spin polarization inside Si suggests that the interface is suitable for achieving efficient spin injection by using tunneling through a Schottky barrier.

Now, it is worth pointing out that the Si–(Co–Si) interface is *not completely* half-metallic although spin polarization remains very high there. In fact, for the Fe atom at the interface, spin polarization seems to be only slightly deviated from unity, being lowered to ∼0.8. The reason for this slight deterioration of the half-metallicity can be understood by looking at the LDOS at the interface, which is presented in figure 4. For all the atoms at the interface, the down-spin LDOS has a clear dip at the Fermi energy, and a down-spin gap seems almost to appear. However, the up-spin LDOS is found to be rather small at the Fermi energy, and this tendency is prominent in particular for the Fe atom. This small size of the up-spin LDOS leads to the lowering of the Fe spin polarization in spite of the distinct dip at the Fermi energy in the down-spin LDOS.

Although the spin polarization of Fe is slightly decreased at the interface, it is still unclear how much the decrease affects tunneling current. To look into this point in a little more detail, we have checked the spin polarization only from sp-LDOS because sp states are ordinarily more delocalized and thought to be more relevant to transport properties than d states [9]. The resulting spin polarization turns out to be extremely high. It is beyond 0.99 for all interfacial atoms except for one Si atom, yet

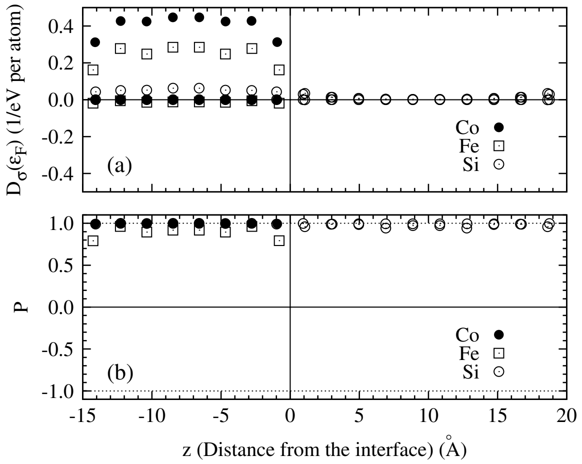


Figure 3. (a) LDOS at the Fermi energy $D_\sigma(\epsilon_F)$ as a function of the distance from the interface, where the LDOS for down-spin is shown with negative sign (though it looks almost zero all over the cell in this scale of the vertical axis). (b) Local spin polarization P as a function of the distance from the interface. Notice that the multilayer is used for the calculations, and therefore both the ends ($z \sim -15$ Å and $z \sim 20$ Å) also correspond to the interface, where the interfacial pattern is the Si–(Co–Si).

the Si atom still keeps spin polarization of the size of ~0.96. From these results, it is expected that the deviation from half-metallicity at the interface does not significantly lower the spin polarization of the tunneling current.

Now, we address possible reasons why the (110) Si–(Co–Si) interface gives rise to high spin polarization. The first is that the (110) plane has a relatively high density of atoms. Indeed, the density in the (110) plane is $\sqrt{2}$ times larger than that of (100) plane. In other words, the (110) plane has less out-of-plane bonding than the (100) plane. This implies that the electronic properties in the (110) plane are less affected even if the neighboring plane is replaced with other substances as in the case of the interface. The second is that there exist structural similarities between interface and bulk in the Si–(Co–Si) interface, as already stated in the discussion on the stability of the interface. As a matter of fact, the tendency is also observed in other half-metallic interfaces consisting of full Heusler alloy and GaAs [9]. The similarity, again, helps the electronic structures remain unchanged from those in the bulk states. The last is that Si(110) surfaces tends to make a dip in the LDOS near the Fermi energy after rather simple structural relaxation within a short periodicity. This property seems to work to keep the minority-spin gap of CoFeSi open at the interface. Actually, structural relaxation is indispensable for obtaining the half-metallic properties, and it should be stressed that the optimization only of interlayer distance is not nearly enough for the interface to be highly spin-polarized.

One might think that the above three conditions were likewise satisfied at the Si–(v–Si) interface. In this case, however, the interfacial electronic structures are far from half-metallic. This is probably due to the fact that the notion of a vacancy–Si path is just fictitious since there is, of course, no bonding between vacancy and Si inside CoFeSi. Accordingly, the relation between vacancy and Si at the interface is totally different from that inside the bulk CoFeSi. This causes significant structural reconstruction as shown in figure 2, and also deteriorates the half-metallicity. Lastly, we mention that we have not found any highly spin-polarized (100) interfaces regarding ferromagnet/Si heterostructures. This seems to be because the (100) interface lacks the first and third conditions just mentioned.

4. Summary

In summary, we have performed a first-principles study on the CoFeSi/Si interface, looking for the possibility of a half-metallic interface between a ferromagnet and silicon. Our calculations predict that a highly spin-polarized CoFeSi/Si (110) interface exists, and furthermore the interfacial pattern is energetically favorable. Strictly, the interface does not possess complete half-metallicity. Nevertheless, its transport properties are hopefully half-metallic because spin polarization calculated from sp-LDOS, which will be more relevant to current, reaches almost unity at the interface. Our analyses

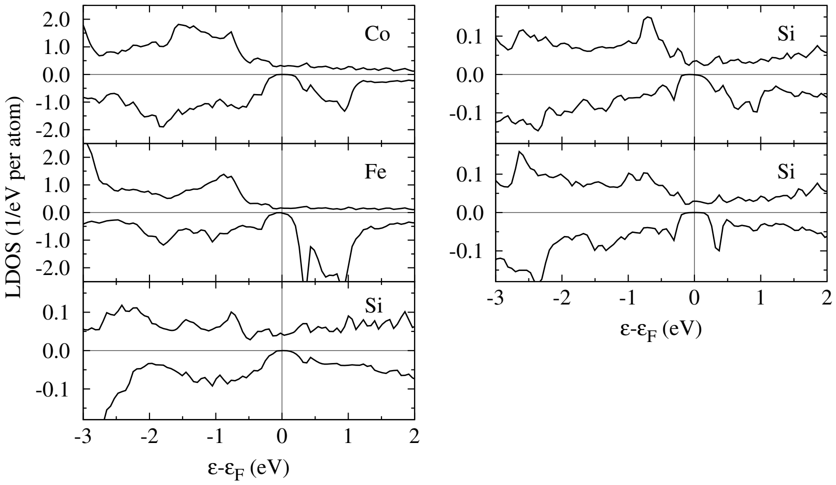


Figure 4. LDOS at the Si–(Co–Si) (110) interface. LDOS for down-spin is shown with negative sign.

suggest that weak interfacial bonding, structural similarity and structural relaxation at the interface are important for attaining half-metallic properties. These findings are expected further to facilitate designing half-metallic interface between a ferromagnet and a semiconductor (or insulator).

Acknowledgments

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