SUPPLEMENTARY INFORMATION FOR:

Controlling the half-metallicity of Heusler/Si(111) interfaces by a monolayer of Si-Co-Si

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In order to provide further insight of the contribution of different atomic species to the electronic structure at the abrupt CFS/Si interface we present the PDOS by atomic species for the Co, Fe and Si atoms at the interface compared to those in the bulk-like environment, i.e. away from the interface region.

**Supplementary Figure S1.** PDOS by atomic species for the abrupt CFS/Si model, calculated for representative a) Co, b) Fe and c) Si atoms located at the interface (region s4 in Fig. 1) (shown with red solid line) compared with PDOS from atoms in the bulk-like CFS environment (black dotted line).

Next, in order to demonstrate that the reversal of the SP observed at the abrupt CFS/Si interface is not only limited to CFS electrodes, we present (Supplementary Figure S2 and S3) the results for the abrupt CFAS/Si and CMS/Si interfaces. Both show SP reversal at the interface indicating that the observed effect is robust against the exact chemical nature of the Co$_2$(Fe,Mn)(Al,Si) electrode.

**Supplementary Figure S2.** First-principles study of abrupt CFAS/Si interface. a) Structural model shown along the [110] viewing direction. Red balls represent Si atoms, white – Al, blue - Co, brown - Fe. b) - g) Spin polarized PDOS for the regions outlined in (a) and labelled as (s1)-(s6). Positive PDOS are for the spin-up states; negative for spin-down.
Supplementary Figure S3. First-principles study of abrupt CMS/Si interface. a) Structural model shown along the [110] viewing direction. Red balls represent Si atoms, blue - Co, yellow - Mn. b) - g) Spin polarized PDOS for the regions outlined in (a) and labelled as (i)-(vi). Positive PDOS are for the spin-up states; negative for spin-down.

Further images showing the presence of the CoSi$_2$ with thicknesses of 1-2 Si-Co-Si monolayers are given in Supplementary Figure S4.

Supplementary Figure S4. HAADF STEM images of the CFAS/Si interface showing the presence of the CoSi$_2$ nano-island, outlined by the yellow dashed rectangles.

In addition to the calculations with a single Si-Co-Si monolayer, we also performed calculations for a double monolayer (presented below) and a whole unit cell of CoSi$_2$. The layer by layer PDOS for the model with two monolayers (Supplementary Figure S5) show that the SP values for the regions (iii), (iv), (v) and (vi) are -17%, -3%, +17% and 44%, respectively. These demonstrate that the interfacial Si bilayer (region (iii)) as well as interfacial CFS layers (region (vi)) still have much better SP compared to those at the abrupt interface case. However, as the number of Si-Co-Si monolayers increases, these monolayers gradually recover their bulk-like metallic structure (e.g. PDOS for region (iv), Supplementary Figure S5e).
**Supplementary Figure S5.** First-principles study of the CFS/SiCoSi-SiCoSi/Si interface, i.e. with two Si-Co-Si monolayers (regions (iv) and (v)). a) Structural model shown along the [110] viewing direction. Red balls represent Si atoms, blue - Co, brown Fe. b) - g) Spin polarized PDOS for the regions outlined in (a) and labelled as (i)-(vii). Positive PDOS are for the spin-up states; negative for spin-down.

Similarly to the considerations for the abrupt interfaces, in order to reveal whether the beneficial effects of the Si-Co-Si layer is limited only to CFS electrode, we perform the same sets calculations for CFAS and CMS electrodes. The results (**Supplementary Figures S6 and S7**) demonstrate that the continuously high positive SP also takes place at these interfaces.
Supplementary Figure S6. First-principles study of CFAS/Si-Co-Si/Si interface, where the Si-Co-Si monolayer is in the region n4. a) Structural model shown along the [110] viewing direction. Red balls represent Si atoms, white – Al, blue - Co, brown - Fe. b) - g) Spin polarized PDOS for the regions outlined in (a) and labelled as (n1)-(n6). Positive PDOS are for the spin-up states; negative for spin-down.

Supplementary Figure S7. First-principles study of the CMS/Si-Co-Si/Si interface, where the Si-Co-Si monolayer is in region (iii). a) Structural model shown along the [110] viewing direction. Red balls represent Si atoms, blue - Co, yellow - Mn. b) - g) Spin polarized PDOS for the regions outlined in (a) and labelled as (i)-(vi). Positive PDOS are for the spin-up states; negative for spin-down.

In Supplementary Figure S8 we present the potential (for both abrupt CFS/Si and CFS/Si-Co-Si/Si interface models), which is the sum of the local part of the pseudo potential, Hartree term and exchange term. These potentials were averaged in-plane (i.e. planes...
parallel to the interface plane) followed by double averaging along z over the unit cells of Si and CFS, and plotted along the direction normal to the interface plane. The calculated potential difference between Si and CFS is utilized to find the valence band offsets.

Supplementary Figure S8. Average potentials across the a) abrupt CFS/Si and b) CFS/Si-Co-Si/Si interface models, which show lowering of the potential difference upon inclusion of a Si-Co-Si layer (model in (b)). Blue curve represents the average potential for spin-up while red for spin-down electrons. Horizontal axes represent the position along the direction perpendicular to the interface plane and directly correlate to the structural models shown just above the plots.