

Supporting Information :
Mn-Doped Sr/Si(111)-(3×2) HCC Surfaces:
Antiferromagnetic Semiconductors for Spintronic
Applications

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(1) Structural Stability for Mn Atoms on (Ca, Ba)/Si(111)-(3×2) HCC Surfaces

The total energy in FM and AFM states ($E_{\text{FM}}^{\text{total}}$ and $E_{\text{AFM}}^{\text{total}}$) for Mn atoms at site M_6 and H_4 on (Ca, Ba)/Si(111)-(3×2) HCC surfaces are calculated and listed in Table S1. The most stable adsorption positions in both FM and AFM states are the *insertion* site M_6 on (Ca, Ba)/Si(111)-(3×2) HCC surfaces, except for the FM state of Mn-doped Ca/Si(111)-(3×2) HCC surfaces, where site H_4 is the most stable adsorption position. Meanwhile, the total energy differences ($\Delta E_{\text{AFM-FM}}^{\text{total}}$) show that AFM states are more favorable than FM states, showing an antiferromagnetic ground state as well as the findings for M_6 and H_4 sites on Sr/Si(111)-(3×2) HCC surface.

Table S1. The total energy in AFM and FM states ($E_{\text{AFM}}^{\text{total}}$ and $E_{\text{FM}}^{\text{total}}$, in eV) and their total energy difference ($\Delta E_{\text{AFM-FM}}^{\text{total}}$, in eV) for Mn atoms at sites M_6 and H_4 on (Ca, Ba)/Si(111)-(3×2) HCC surfaces. These magnetic structures correspond to a double supercell along the $y[110]$ direction.

Substrate	Site	$E_{\text{AFM}}^{\text{total}}$	$E_{\text{FM}}^{\text{total}}$	$\Delta E_{\text{AFM-FM}}^{\text{total}}$
Ca/Si(111)-(3×2)	M_6	-659.875	-659.829	-0.046
	H_4	-659.834	-659.832	-0.002
Ba/Si(111)-(3×2)	M_6	-660.377	-660.319	-0.058
	H_4	-660.204	-660.196	-0.008

(2) Total and Projected Density of States for Mn Atoms at M_6 and H_4 Sites on Sr/Si(111)-(3×2) HCC Surface

In order to understand the origin of magnetism of Mn atom at sites M_6 and H_4 on Sr/Si(111)-(3×2) HCC surface, we have calculated the total and projected density of states (TDOS and PDOS) using PBE+U functional. The calculated results for structure M_6 are plotted in Figure S1. As shown in Figure S1a, there is a large band splitting between spin-up and spin-down TDOS curves near the E_{F} , where the spin-down TDOS

peak below the E_F is mainly from the d band of Mn atom (see Figure S1b). Meanwhile, as shown in Figure S1c-f, the p bands of Si atoms (Si_1 , Si_2 , Si_4 and Si_9) around Mn atom have a little splitting near the E_F , which give rise to the small moment $-0.026 \mu_B$ for Si_1 , $-0.058 \mu_B$ for Si_2 , $-0.012 \mu_B$ for Si_4 , and $-0.011 \mu_B$ for Si_9 . Because Si_6 and Si_2 atoms have the equivalent position and have the similar moment, as well as Si_9 and Si_{10} , their DOS results are not shown in Figure S1. For Sr atom as shown in Figure S1g, it is almost no contribution for the TDOS peak near the E_F .

The calculated results for structure H_4 are plotted in Figure S2. As shown in Figure S2a, there is a large band splitting between spin-up and spin-down TDOS peaks near the E_F , where the spin-down TDOS peak is mainly from the d band of Mn atom (see Figure S2b). Meanwhile, as shown in Figure S2c-h, the p bands of Si atoms (Si_3 , Si_7 , Si_8 , Si_9 , Si_{10} and Si_{11}) around Mn atom have splitting near the E_F , which give rise to a small moment $-0.027 \mu_B$ for Si_3 , $-0.048 \mu_B$ for Si_7 , $-0.01 \mu_B$ for Si_8 , $0.009 \mu_B$ for Si_9 , $-0.011 \mu_B$ for Si_{10} , and $-0.012 \mu_B$ for Si_{11} . These Si band splittings are caused by Mn atoms, which also make the small contribution for the TDOS peak near the E_F . As for Sr atom in Figure S2i, it is almost no contribution for the TDOS peak near the E_F .

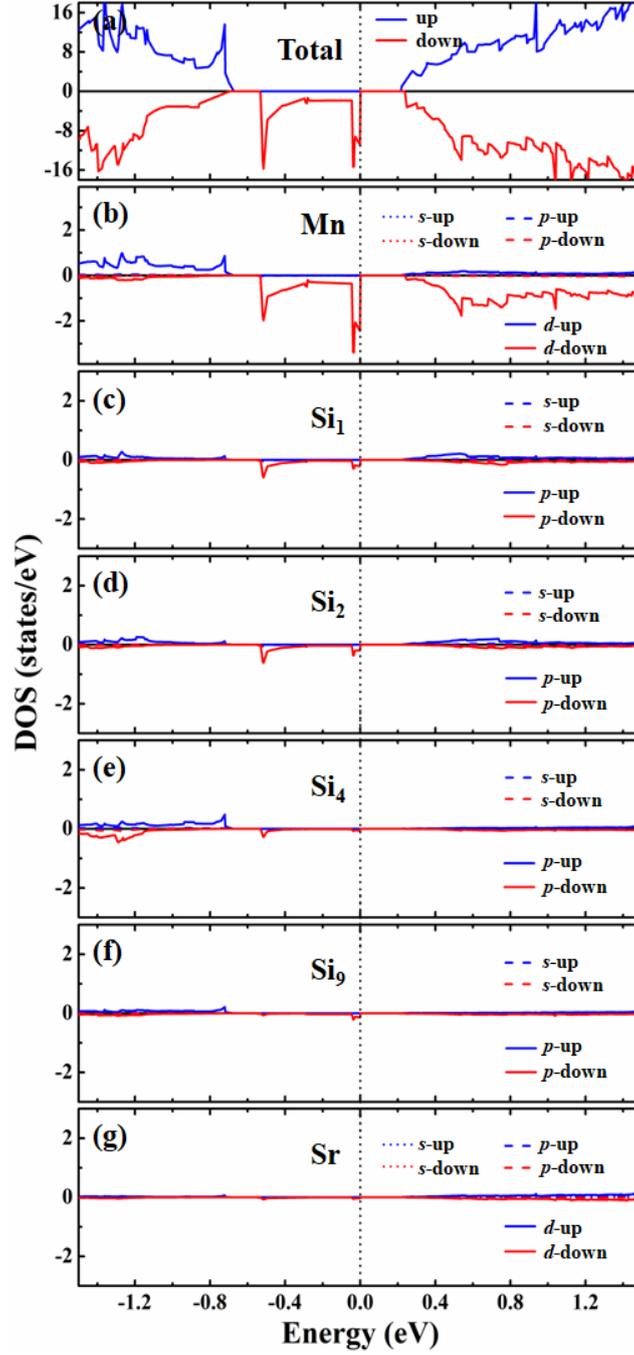


Figure S1. (a) Total density of states of Mn atom at M_6 site on Sr/Si(111)-(3 \times 2) HCC surface. (b-g) Projected density of states for (b) Mn, (c) Si₁, (d) Si₂, (e) Si₄, (f) Si₉ and (g) Sr atoms. Vertical dotted lines give the Fermi level that is set to zero.

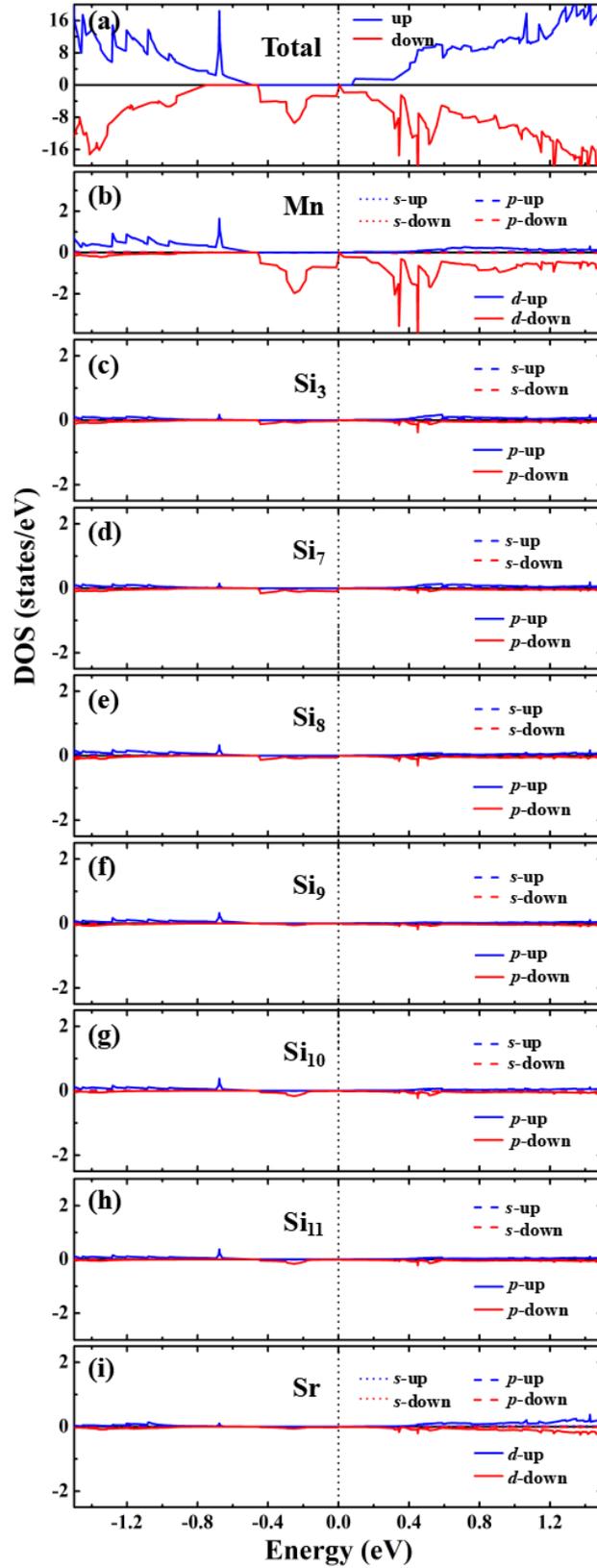


Figure S2. (a) Total density of states of Mn atom at H₄ site on Sr/Si(111)-(3×2) HCC surface. (b-i) Projected density of states for (b) Mn, (c) Si₃, (d) Si₇, (e) Si₈, (f) Si₉, (g) Si₁₀, (h) Si₁₁, and (i) Sr atoms. The Fermi level is set to zero.

(3) Néel Temperature and Magnetic Anisotropy Energy for Structures of M₆ and H₄ on Sr/Si(111)-(3×2) HCC Surface in AFM State

For the AFM materials, an important parameter is the Néel temperature (T_N), below which the system develops long range AFM ordering. Here, we adopt the mean field theory (MFT) to estimate the T_N of structures M₆ and H₄ with equation: $(3/2)k_B T_N = -\Delta E_{\text{total}}^{\text{AFM-FM}}/n$, where k_B and $\Delta E_{\text{total}}^{\text{AFM-FM}}$ are Boltzmann's constant and the total energy difference between AFM and FM states, and $n=2$ is the number of Mn atom in the supercell. As listed in Table S2, the estimated T_N values are about 240 K for structure M₆ and 54 K for structure H₄, below the room temperature.

Table S2. Néel temperature (T_N , in K), magnetic anisotropy energy ($E_{[010]}-E_{[001]}$, in meV/Mn) and easy axis (EA) for both structures M₆ and H₄ in a double supercell along the $y[\bar{1}10]$ direction.

		T_N	$E_{[010]}-E_{[001]}$	EA
M ₆	FM		0.07	[001]
	AFM	240	0.11	[001]
H ₄	FM		0.07	[001]
	AFM	54	0.05	[001]

In order to analyze the easy axis of magnetization, we further calculate the magnetic anisotropy energy (MAE) using non-collinear magnetic order with spin-orbit coupling (SOC) for the FM and AFM states of structures M₆ and H₄. The relative stabilities along two magnetization directions are studied, i.e., [010] direction parallel to the channel and [001] direction perpendicular to the Mn chain, out-of-plane. The calculated MAEs are summarized in Table S2. It shows that the easy axes of FM and AFM states for both structures M₆ and H₄ are parallel to the [001] direction, out-of-plane.

(4) Electronic Band Structures for Mn at H₄ Site on Sr/Si(111)-(3×2) HCC

Surface

The calculated spin-polarized band structures in FM and AFM states for Mn atoms at H_4 site are given in Figure S3a,b, respectively. It is shown that FM state has half-metallic character, while AFM state presents the semiconducting character with the band gap of 0.21 eV. Moreover, there are some new surface states c_{1d} and v_{1d} for FM state and c_1 and v_1 for AFM state near the E_F , compared with the band structure of Sr/Si(111)-(3 \times 2) HCC surface (see Figure 3a). The corresponding surface band gaps for AFM state is between states c_1 and v_1 . Thus, the above calculated band gaps 0.21 eV for AFM is the surface band gaps, which is smaller than the bulk band gap 0.75 eV.

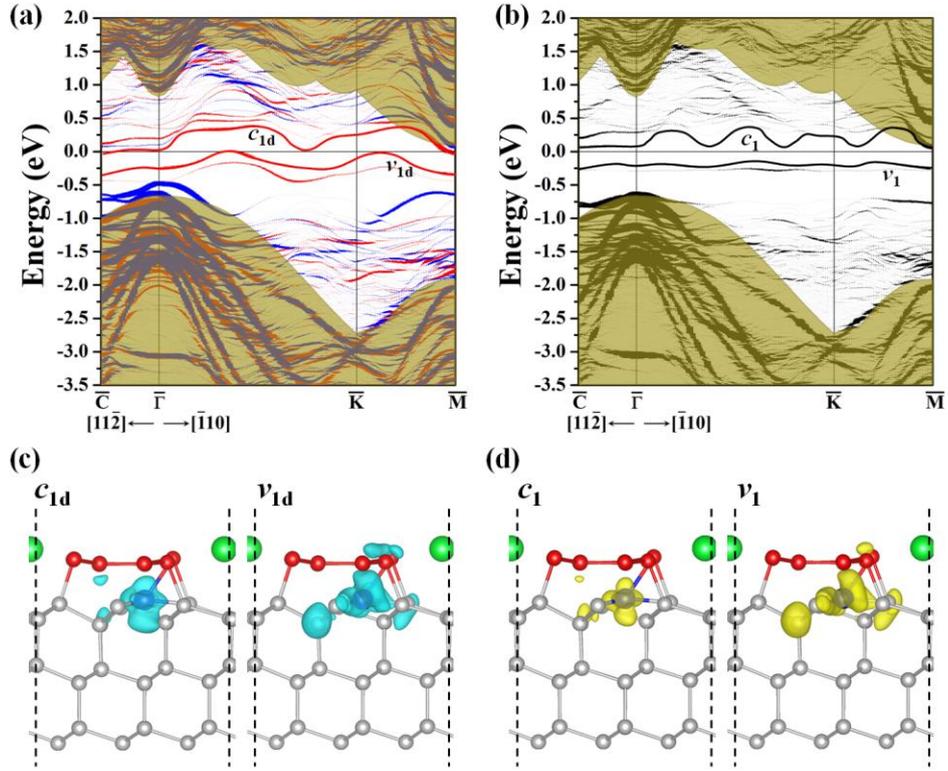


Figure S3. The unfolded electronic band structures of FM (a) and AFM (b) states for Mn atom at H_4 site on Sr/Si(111)-(3 \times 2) HCC surface. Blue and red lines in (a) represent spin-up and spin-down bands of FM state. c_{1d} , v_{1d} , c_1 and v_1 are four surface states near the E_F , c_{1d} and v_{1d} are marked in red, and c_1 and v_1 are black. (c) The band-decomposed charge density distributions of surface state c_{1d} , v_{1d} in FM band structure (a). (d) The band-decomposed charge density distributions of surface state c_1 and v_1 in AFM band structure (b). The iso-surface value is $0.01 \text{ e}/\text{\AA}^3$. Dashed lines represent unit cell in (c,d).

The calculated band-decomposed charge density distributions (BDCDDs) of FM state for H_4 structure are plotted in Figure S3c. It shows that state c_{1d} is almost entirely originated from the d orbitals of Mn atom, while for v_{1d} , the main contribution is from the d orbitals of Mn atom and small contribution is from the p orbitals of neighboring Si atoms ($Si_3, Si_7, Si_8, Si_9, Si_{10}$ and Si_{11}). Meanwhile, the BDCDDs of AFM state are plotted in Figure S3d. The distribution of state c_1 and v_1 are similar to state c_{1d} and v_{1d} of FM state, respectively; that is, c_1 is contributed by the Mn d orbitals, and v_1 is from Mn d and Si p orbitals.

REFERENCES

- [1] Sakamoto K.; Zhang H. M.; Uhrberg R. I. G. Electronic Structure of the Ca/Si(111)-(3 \times 2) Surface. *Phys. Rev. B* **2004**, 69, 125321.