Electronic Supplementary Information

for

Structural, magnetic, and electronic properties of $EuSi_2$ thin films on Si(111) surface

Guang Yang,[1,2] Jun-Shuai Chai,[1,3] Kun Bu,[1,2] Li-Fang Xu, [1,4] and Jian-Tao Wang[1,2,4*]

[1]*Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China*

[2]*School of Physical Sciences, University of Chinese Academy of Sciences, Beijing 100049, China*

[3]*Key Laboratory of Microelectronics & Integrated Technology, Institute of Microelectronics, Chinese Academy of Sciences, Beijing 100029, China*

[4]*Songshan Lake Materials Laboratory, Dongguan 523808, Guangdong, China*

[*]*E-mail address*: wjt@aphy.iphy.ac.cn (Jian-Tao Wang).

(1) Structural stability for the unpassivated $EuSi_2$ thin films on Si(111) surface

We first study the structural stability of 1-3 ML unpassivated $EuSi_2$ thin films on Si(111) surface (see Fig. S1). For 2 ML $EuSi_2$ thin film, the structures in AA and AB stacking [see Fig. S1(b,c)] are simulated with interlayer FM and AFM couplings between the Eu atomic layers. As for 3 ML $EuSi_2$ thin film, AAA and ABC stacking are considered [see Fig. S1(d,e)]. The calculated relative total energy and local magnetic moment on Eu sites for 1-3 ML unpassivated $EuSi_2$ thin films on Si(111) surface are listed in Table S1. Based on the above results, we can see that AA-stacking is the most stable model for multilayer unpassivated $EuSi_2$ thin film on Si(111) surface. The calculated local magnetic moment on Eu site in each layer is 6.96-7.00 μ_B, and the ground state for the multilayer unpassivated $EuSi_2$ thin film on Si(111) surface should be in AA stacking with an AFM interlayer coupling. These structural and magnetic behaviors are in agreement with the cases with the H passivated $EuSi_2$ thin films shown in Fig. 1 in main text.

(2) Electronic properties for the unpassivated $EuSi_2$ thin films on Si(111) surface

We next discuss the electronic properties of 1-3 ML unpassivated $EuSi_2$ thin films on Si(111) surface. The calculated electronic band structure for 1-3 ML unpassivated $EuSi_2$ thin films on Si(111) surface are plotted in Fig. S2(a-c). Shaded areas represent the projected Si(111) bulk bands. We find the surface states S_{up} and S_{dn} crossing the Fermi level (F_E) show metallic feature. In order to show the distribution of 1-3 ML unpassivated $EuSi_2$/Si(111) surface states S_{up} and S_{dn} in real space, the band-decomposed charge density distributions (BDCDDs) are calculated and plotted in Fig. S2(d-f). The surface states S_{up} and S_{dn} for 1-3 ML unpassivated $EuSi_2$ thin films are mainly contributed by the topmost Si atoms of $EuSi_2$ thin films. Their corresponding total and projected density of states (DOS) [see Fig. S2(g-i)] show that the surface states are mainly originated from the Si-*p* orbits. On the basis of these results, we concluded that the topmost Si atoms for $EuSi_2$ thin films are unstable and active. Thus, passivating topmost Si atoms by H atoms is essential for further investigating the electronic properties.

Table S1: The calculated relative total energy and local magnetic moment on Eu sites with FM and AFM interlayer coupling for 1ML, 2 ML in AA and AB stacking, and 3 ML in AAA and ABC stacking unpassivated $EuSi_2$ thin films on Si(111) surface.

Model	E_{tot} (meV)	$M_{Eu\text{-}I}$ (μ_B)	$M_{Eu\text{-}II}$ (μ_B)	$M_{Eu\text{-}III}$ (μ_B)
1 ML		6.99		
2 ML-AA(↑↓)	0	6.98	-6.99	
2 ML-AA(↑↑)	4	6.97	6.97	
2 ML-AB(↓↑)	5	-6.98	7.00	
2 ML-AB(↑↑)	8	6.96	6.97	
3 ML-AAA(↑↓↑)	0	6.99	-7.00	6.99
3 ML-AAA(↑↑↑)	5	6.97	6.96	6.98
3 ML-ABC(↓↑↓)	170	-6.98	7.00	-7.00
3 ML-ABC(↑↑↑)	181	6.99	7.00	7.00

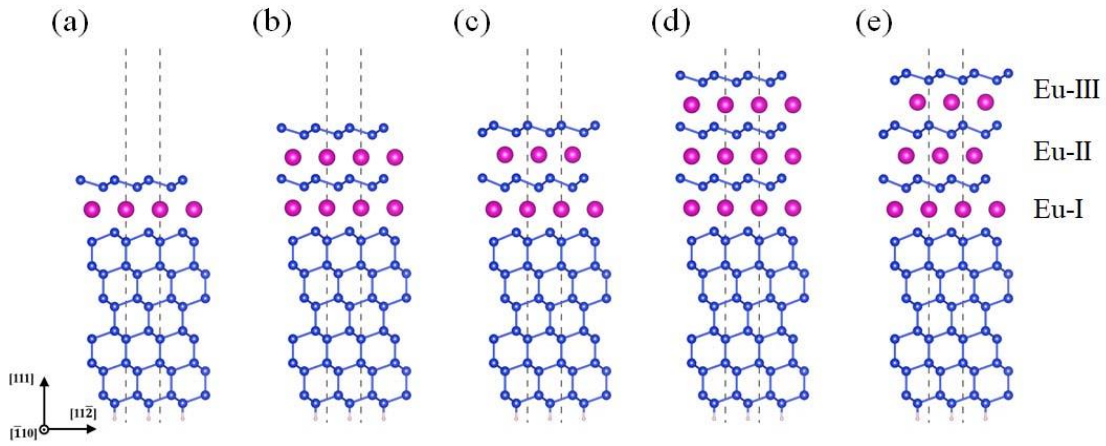

Fig. S1: The side view of unpassivated $EuSi_2$ thin films on Si(111) surface. (a) 1 ML, (b) 2 ML in AA stacking, (c) 2 ML in AB stacking, (d) 3 ML in AAA stacking, and (e) 3 ML in ABC stacking. Dashed lines represent a 1 × 1 hexagonal unit cell. The purple, blue, and white balls represent Eu, Si, and H atoms, respectively. The first, second and third Eu atomic layer are marked as Eu-I, Eu-II, and Eu-III, respectively.

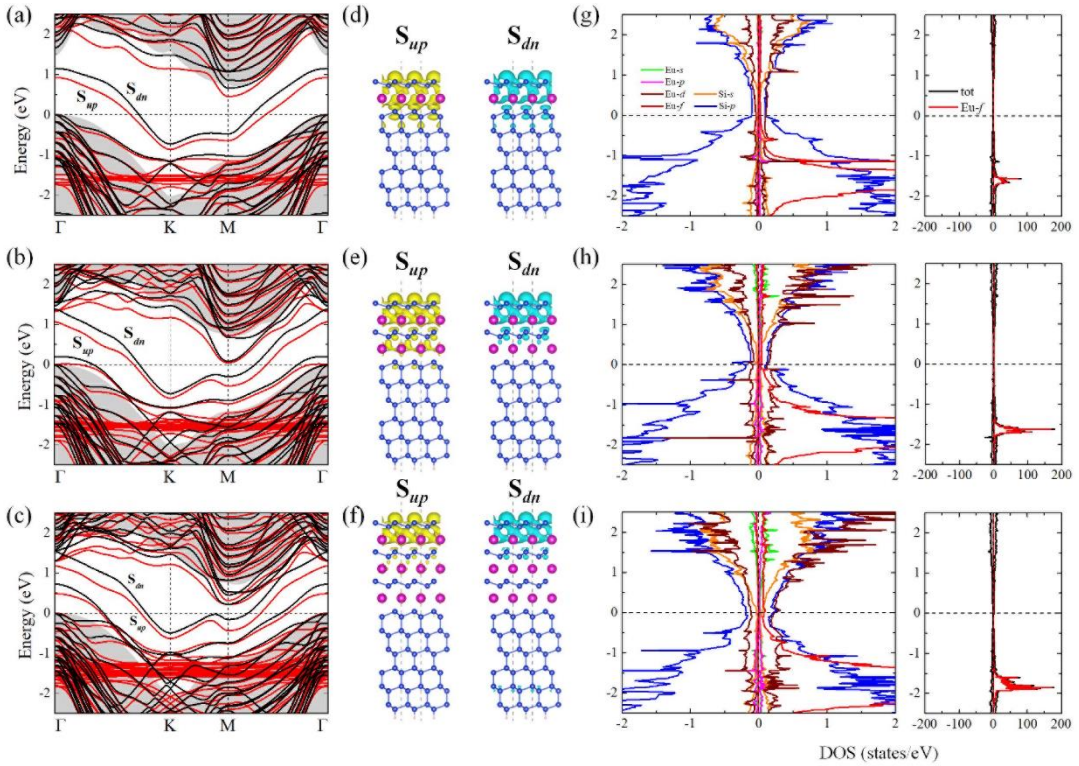

Fig. S2: The electronic band structure for (a) 1 ML, (b) 2 ML and (c) 3 ML unpassivated $EuSi_2$ thin films on Si(111) surface. Shaded areas represent the projected Si(111) bulk bands. The spin-up and spin-down states are represented by red and black lines, respectively. The BDCDDs of S_{up} and S_{dn} for (d) 1 ML, (e) 2 ML and (f) 3 ML unpassivated $EuSi_2$ thin films on Si(111) surface. The iso-surface charge density contour is 0.001 e/Bohr^3. The total and projected DOS for (g) 1 ML, (h) 2 ML and (i) 3 ML unpassivated $EuSi_2$ thin films on Si(111) surface. The spin-up and spin-down states are represented by positive and negative values, respectively. The E_F is set to zero and indicated by the black dashed line.