Supporting information for: Electrical Control of Spin States of Ferrocene on Cu(111)

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1 Optimized C-C bond lengths of Fc by LDA+U

Table S1: Optimized C-C bond lengths $d_{\text{C-C}}$ (Å) of the free and adsorbed Fc.

		bond index				
$d_{\text{C-C}}$	[Å]	1	2	3	4	5
		free Fc				
LS	top			1.432		
	bot			1.432		
HS	top	1.432	1.425	1.420	1.425	1.432
	bot	1.420	1.425	1.431	1.431	1.425
		adsorbed Fc				
LS	top			1.431		
	bot	1.446	1.446	1.450	1.447	1.450
HS	top	1.426	1.426	1.425	1.425	1.426
	bot	1.438	1.439	1.448	1.437	1.449

2 Structural properties of Fc by PBE-D+U

Table S2: Optimized geometries of the free and adsorbed (ads) Fc in the low-spin state and the binding energy E_{b} by the PBE-D+U approach.

	d_{top} (Å)	d_{bot} (Å)	d (Å)	E_{b} (eV)
free	1.64	1.64		
ads	1.66	1.63	2.73	1.09

3 xyz files of the isolated Fc optimized by LDA+U

Fc in the low-spin state

```
21

C       1.833314    2.172296   11.906969
C       3.158226    3.807212    8.392729
C       1.477513    3.557551   11.833152
```

C	2.683311	4.317372	11.694205
C	3.513180	2.421931	8.465137
C	2.306731	1.662257	8.597741
C	1.732307	3.903780	8.480159
C	1.206031	2.578108	8.607046
C	3.259017	2.076272	11.813744
C	3.784179	3.401867	11.682080
H	1.129410	1.324033	11.992200
H	3.863078	4.655198	8.313749
H	0.450772	3.966750	11.851591
H	2.750366	5.415472	11.583884
H	4.540179	2.013134	8.451973
H	2.239486	0.563640	8.703664
H	1.143186	4.839481	8.478704
H	0.139394	2.310997	8.720734
H	3.849199	1.141355	11.813278
H	4.849723	3.668788	11.559643
Fe	2.492966	2.988803	10.148037

Fc in the high-spin state

21

C	1.913112	2.244873	12.120752
C	3.147911	3.765091	8.195998
C	1.568843	3.634299	12.092636
C	2.777771	4.384177	12.017791
C	3.498177	2.378134	8.216654
C	2.291822	1.620786	8.242066

```
C       1.719538    3.853684    8.216903
C       1.198276    2.527409    8.242489
C       3.339872    2.149433   12.053574
C       3.865927    3.471915   11.993688
H       1.207001    1.399793   12.221380
H       3.853602    4.614541    8.137824
H       0.545467    4.051541   12.142427
H       2.853901    5.486700   11.958861
H       4.524826    1.966811    8.200545
H       2.220197    0.516977    8.281271
H       1.125633    4.786626    8.194616
H       0.126669    2.252476    8.278403
H       3.929650    1.213535   12.065325
H       4.936421    3.740394   11.911840
Fe      2.493648    2.988959   10.143491
```

4 Simplified cif files of Fc/Cu(111) optimized by LDA+U

Fc/Cu(111) in the low-spin state

```
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_cell_length_a    10.224501
_cell_length_b    10.224497
_cell_length_c    21.262054
_cell_angle_alpha    90.000000
_cell_angle_beta     90.000000
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   Cu    Cu     0.000000     0.250000     0.000000
   Cu    Cu     0.000000     0.500000     0.000000
   Cu    Cu     0.000000     0.750000     0.000000
   Cu    Cu     0.250000     0.000000     0.000000
   Cu    Cu     0.250000     0.250000     0.000000
   Cu    Cu     0.250000     0.500000     0.000000
   Cu    Cu     0.250000     0.750000     0.000000
   Cu    Cu     0.500000     0.000000     0.000000
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   Cu    Cu     0.500000     0.750000     0.000000
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Cu	Cu	0.750000	0.250000	0.000000
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Cu	Cu	0.750000	0.750000	0.000000
Cu	Cu	0.997714	0.998952	0.291869
Cu	Cu	0.997530	0.250158	0.291650
Cu	Cu	0.999941	0.500708	0.292478
Cu	Cu	0.000107	0.750880	0.292536
Cu	Cu	0.248765	0.998366	0.291150
Cu	Cu	0.245042	0.248813	0.293620
Cu	Cu	0.248463	0.502448	0.290376
Cu	Cu	0.250031	0.750948	0.292549
Cu	Cu	0.500323	0.999255	0.292002
Cu	Cu	0.502500	0.249056	0.293596
Cu	Cu	0.501521	0.503842	0.293729
Cu	Cu	0.500433	0.752574	0.291698
Cu	Cu	0.749808	0.000548	0.292887
Cu	Cu	0.751355	0.250315	0.291861
Cu	Cu	0.753078	0.502310	0.290557
Cu	Cu	0.751187	0.752300	0.291786
Cu	Cu	0.166921	0.084362	0.195314
Cu	Cu	0.166877	0.333206	0.195094
Cu	Cu	0.166479	0.584044	0.194585
Cu	Cu	0.166579	0.833574	0.194761
Cu	Cu	0.416973	0.084369	0.195297
Cu	Cu	0.416394	0.333440	0.197158
Cu	Cu	0.417202	0.583317	0.195151

Cu	Cu	0.416177	0.833479	0.194557
Cu	Cu	0.666655	0.083583	0.195017
Cu	Cu	0.665870	0.333258	0.195127
Cu	Cu	0.665674	0.583254	0.195192
Cu	Cu	0.666745	0.833823	0.194946
Cu	Cu	0.916419	0.083519	0.194945
Cu	Cu	0.916858	0.334261	0.194489
Cu	Cu	0.917052	0.584004	0.194598
Cu	Cu	0.916864	0.833369	0.194550
Cu	Cu	0.083330	0.166670	0.098160
Cu	Cu	0.083330	0.416670	0.098160
Cu	Cu	0.083330	0.666670	0.098160
Cu	Cu	0.083330	0.916670	0.098160
Cu	Cu	0.333330	0.166670	0.098160
Cu	Cu	0.333330	0.416670	0.098160
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Cu	Cu	0.333330	0.916670	0.098160
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Cu	Cu	0.583330	0.416670	0.098160
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Cu	Cu	0.583330	0.916670	0.098160
Cu	Cu	0.833330	0.166670	0.098160
Cu	Cu	0.833330	0.416670	0.098160
Cu	Cu	0.833330	0.666670	0.098160
Cu	Cu	0.833330	0.916670	0.098160
C	C	0.284683	0.235201	0.557431
C	C	0.540129	0.443039	0.401439

C	C	0.329124	0.391974	0.556057
C	C	0.490633	0.477540	0.555521
C	C	0.500531	0.285639	0.402084
C	C	0.337638	0.194570	0.403166
C	C	0.401522	0.448748	0.401274
C	C	0.276124	0.294798	0.401752
C	C	0.418642	0.223966	0.557730
C	C	0.546002	0.373787	0.556531
H	H	0.167423	0.139417	0.556657
H	H	0.656330	0.541445	0.404048
H	H	0.252254	0.438707	0.554192
H	H	0.560420	0.601985	0.552968
H	H	0.580841	0.243101	0.406961
H	H	0.271148	0.069322	0.405914
H	H	0.392243	0.552227	0.403824
H	H	0.155148	0.260761	0.406371
H	H	0.423000	0.118054	0.557268
H	H	0.666001	0.404010	0.555069
Fe	Fe	0.412416	0.336736	0.478778

Fc/Cu(111) in the high-spin state

```
data_a


_cell_length_a    10.224501
_cell_length_b    10.224497
_cell_length_c    21.262054
_cell_angle_alpha    90.000000
_cell_angle_beta     90.000000
```

```
_cell_angle_gamma   120.000012
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_symmetry_Int_Tables_number      1


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_symmetry_equiv_pos_as_xyz
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_atom_site_type_symbol
_atom_site_fract_x
_atom_site_fract_y
_atom_site_fract_z
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   Cu    Cu     0.000000     0.250000     0.000000
   Cu    Cu     0.000000     0.500000     0.000000
   Cu    Cu     0.000000     0.750000     0.000000
   Cu    Cu     0.250000     0.000000     0.000000
   Cu    Cu     0.250000     0.250000     0.000000
   Cu    Cu     0.250000     0.500000     0.000000
   Cu    Cu     0.250000     0.750000     0.000000
   Cu    Cu     0.500000     0.000000     0.000000
   Cu    Cu     0.500000     0.250000     0.000000
   Cu    Cu     0.500000     0.500000     0.000000
```

Cu	Cu	0.500000	0.750000	0.000000
Cu	Cu	0.750000	0.000000	0.000000
Cu	Cu	0.750000	0.250000	0.000000
Cu	Cu	0.750000	0.500000	0.000000
Cu	Cu	0.750000	0.750000	0.000000
Cu	Cu	0.997219	0.998394	0.291690
Cu	Cu	0.997136	0.250098	0.291483
Cu	Cu	0.000159	0.500864	0.292395
Cu	Cu	0.000331	0.750882	0.292475
Cu	Cu	0.249046	0.998731	0.291344
Cu	Cu	0.243067	0.248054	0.293688
Cu	Cu	0.248629	0.502418	0.290485
Cu	Cu	0.250128	0.750979	0.292477
Cu	Cu	0.500641	0.998653	0.291769
Cu	Cu	0.504053	0.248269	0.293826
Cu	Cu	0.502150	0.504799	0.294441
Cu	Cu	0.500575	0.752755	0.291552
Cu	Cu	0.749874	0.000322	0.292879
Cu	Cu	0.752104	0.250156	0.291628
Cu	Cu	0.753262	0.502367	0.290647
Cu	Cu	0.751602	0.752589	0.291619
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Cu	Cu	0.167045	0.333208	0.194972
Cu	Cu	0.166744	0.583943	0.194635
Cu	Cu	0.166840	0.833942	0.194861
Cu	Cu	0.417144	0.084513	0.195270
Cu	Cu	0.416478	0.333300	0.197912

Cu	Cu	0.417410	0.583044	0.195208
Cu	Cu	0.416120	0.833355	0.194442
Cu	Cu	0.666687	0.083634	0.194990
Cu	Cu	0.665844	0.333206	0.195037
Cu	Cu	0.665376	0.583001	0.195243
Cu	Cu	0.666703	0.833600	0.194966
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Cu	Cu	0.917041	0.334437	0.194369
Cu	Cu	0.916809	0.583881	0.194645
Cu	Cu	0.916966	0.833261	0.194433
Cu	Cu	0.083330	0.166670	0.098160
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Cu	Cu	0.083330	0.916670	0.098160
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Cu	Cu	0.333330	0.916670	0.098160
Cu	Cu	0.583330	0.166670	0.098160
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Cu	Cu	0.583330	0.916670	0.098160
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Cu	Cu	0.833330	0.416670	0.098160
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Cu	Cu	0.833330	0.916670	0.098160
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C	C	0.540717	0.440587	0.397065
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C	C	0.339434	0.193153	0.398197
C	C	0.403063	0.446226	0.397159
C	C	0.277706	0.292385	0.397223
C	C	0.431300	0.232121	0.579779
C	C	0.556854	0.381895	0.576455
H	H	0.180257	0.145344	0.585373
H	H	0.656800	0.538979	0.401574
H	H	0.261791	0.443171	0.581359
H	H	0.568899	0.608662	0.574665
H	H	0.581594	0.241413	0.404504
H	H	0.273057	0.067694	0.401762
H	H	0.393886	0.549688	0.401442
H	H	0.156989	0.258264	0.403695
H	H	0.437194	0.126988	0.581140
H	H	0.677294	0.413401	0.574540
Fe	Fe	0.413187	0.337310	0.489177