Supporting Information

for

"Issues on DFT+*U* calculations of organic diradicals"

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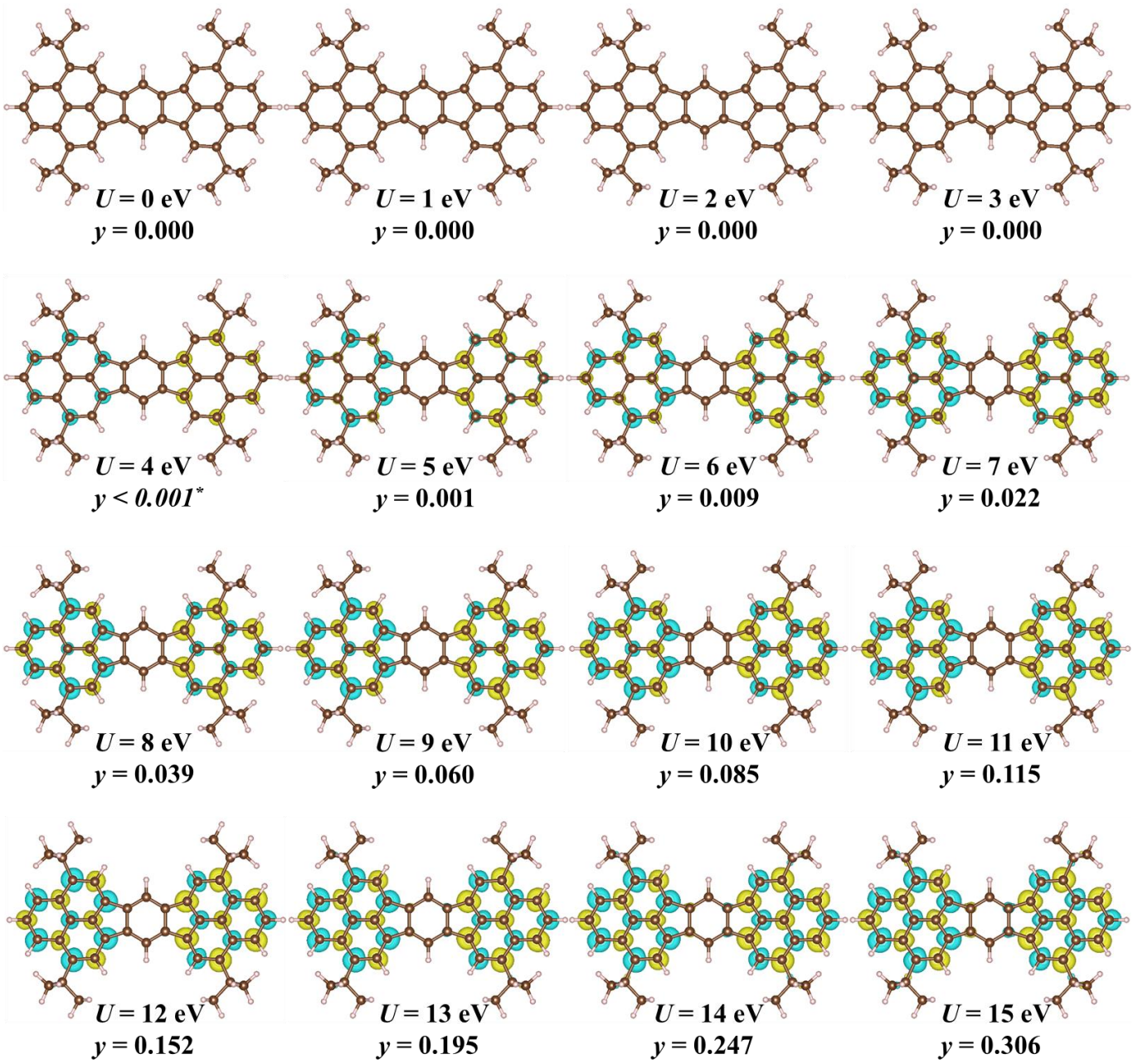


Figure S1. Dependence of spin density distributions of singlet state of Mol 1 on *U* value of DFT+*U* calculation. The threshold is 0.003 e^{-}/Bohr^3. Yellow and blue represent the spin density of major and minor spins, respectively. *Although the spin is polarised in the results by *U* = 4 eV, the *y* value could not be estimated because the spin distribution on the carbons is too small (the detail is described in main text).

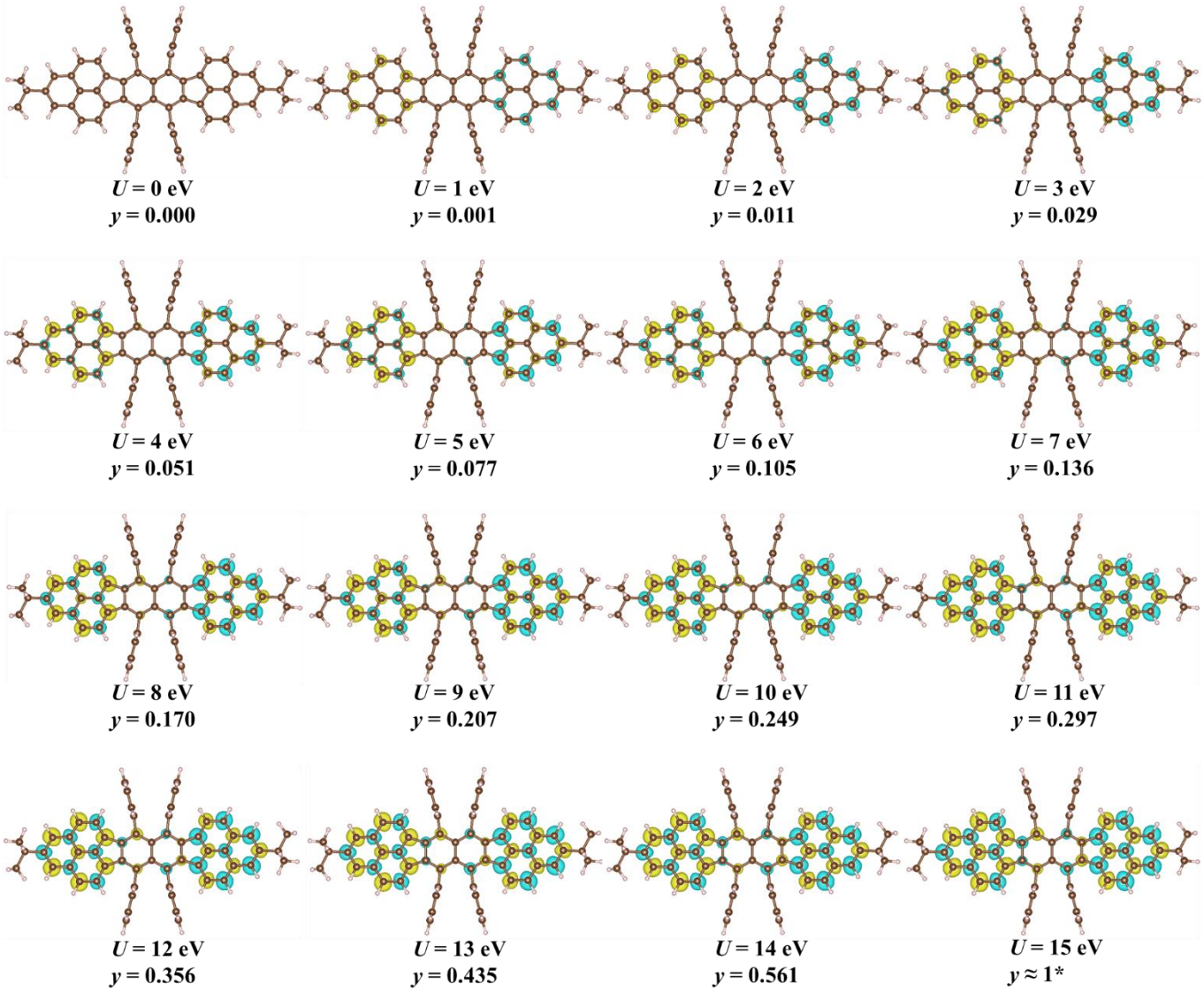


Figure S2. Dependence of spin density distributions of singlet state of Mol 2 on *U* value of DFT+*U* calculation. The threshold is 0.003 e^{-}/Bohr^3. Yellow and blue represent the spin density of major and minor spins, respectively. *Spin polarisation in the results by*U* =15 eV is too large to estimate the *y* value using electron density.

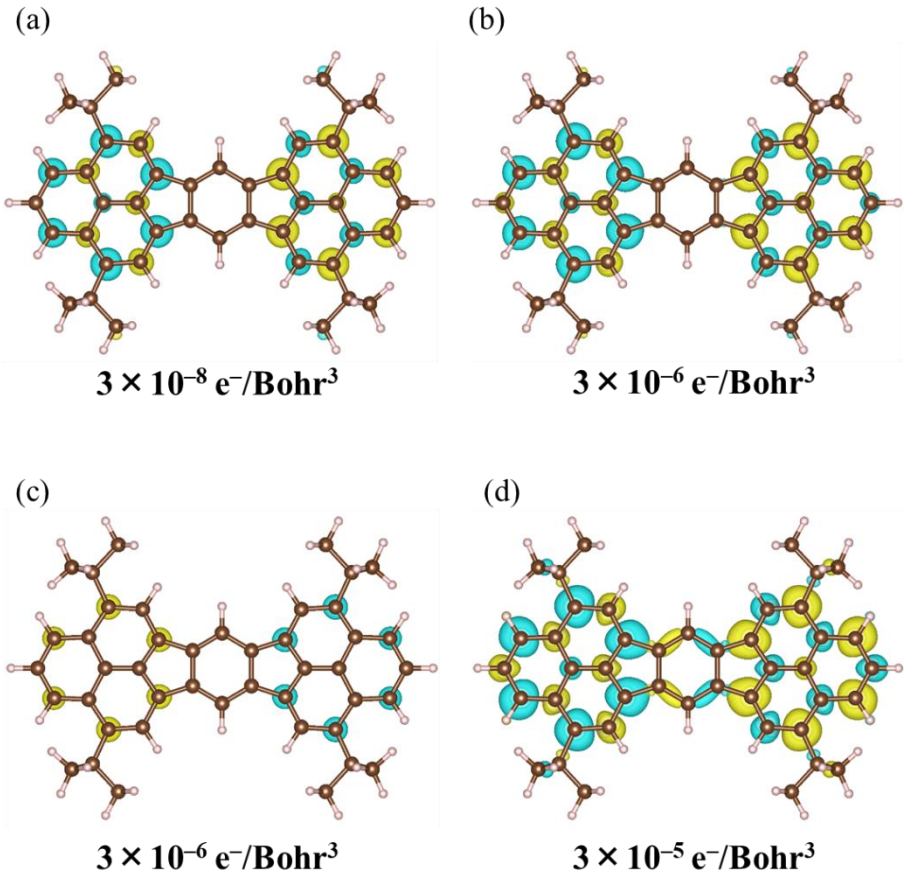


Figure S3. Spin density distributions of singlet state of Mol 1 estimated by DFT+*U*/plane-wave with low threshold. (a) $U = 0$ eV (threshold is 3×10^{-8} a.u.), (b) $U = 1$ eV (threshold is 3×10^{-6} a.u.), (c) $U = 2$ eV (threshold is 3×10^{-6} a.u.), and (d) $U = 3$ eV (threshold is 3×10^{-5} a.u.). Yellow and blue represent the spin density of major and minor spins, respectively.

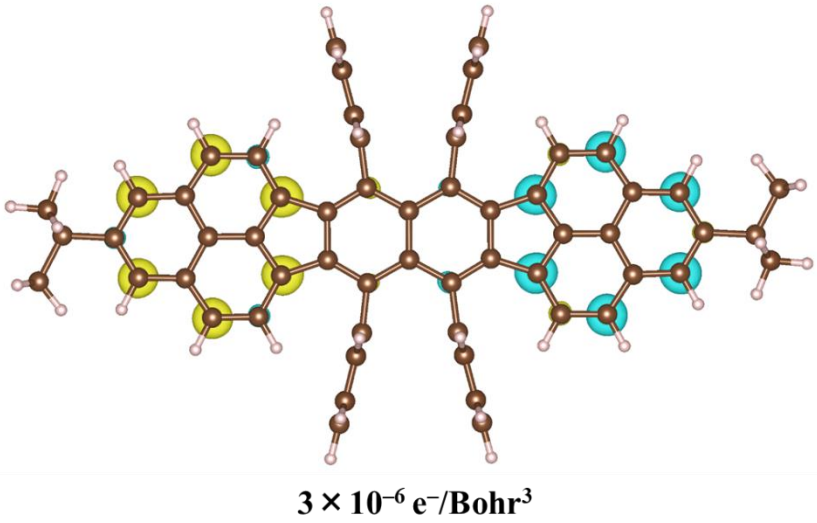


Figure S4. Spin density distributions of singlet state of Mol 2 estimated by pure-DFT/plane-wave with low threshold (3×10^{-6} a.u.). Yellow and blue represent the spin density of major and minor spins, respectively.

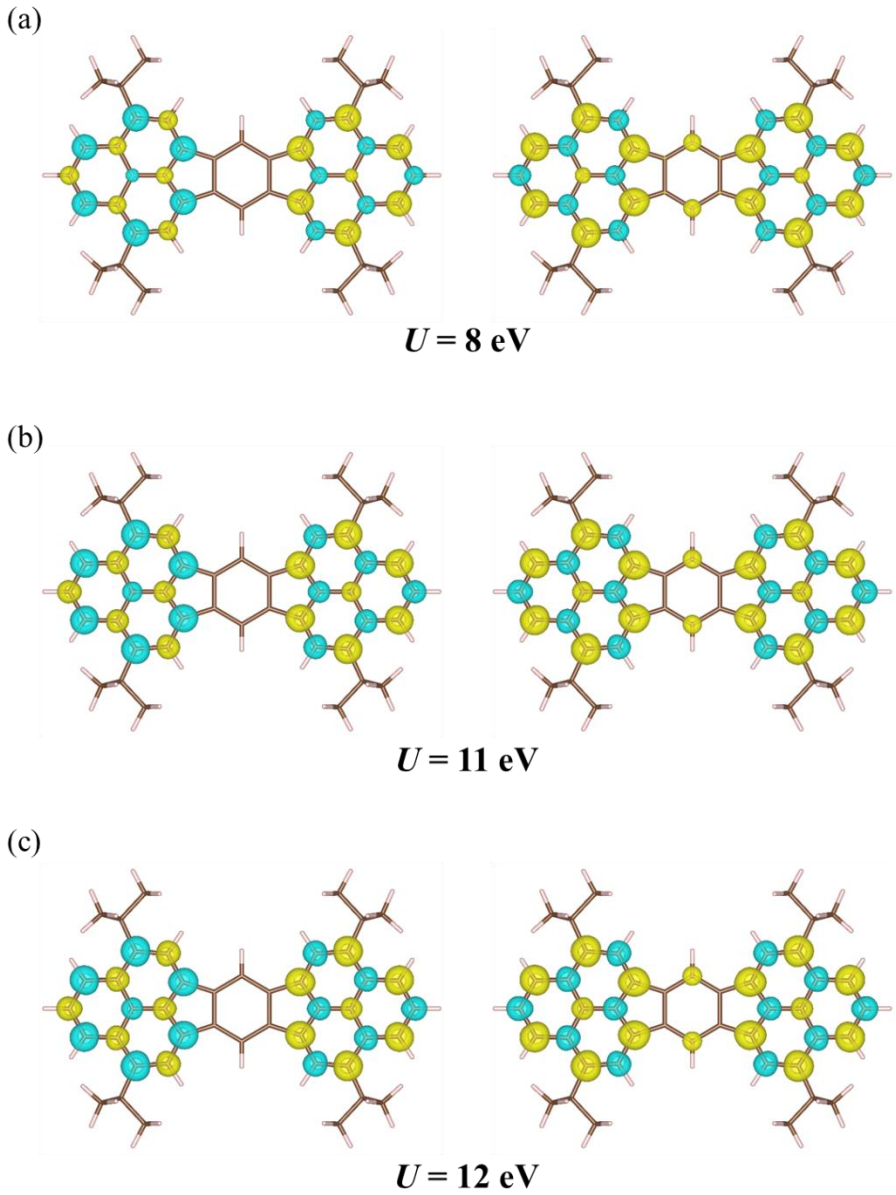


Figure S5. Spin density distributions of Mol 1 estimated by DFT+*U*/plane-wave. (a) U = 8 eV, (b) U = 11 eV, and (c) U = 12 eV. Left and right panels are LS and HS states, respectively. The threshold is 0.003 e^{-}/Bohr^3. Yellow and blue represent the spin density of major and minor spins, respectively.

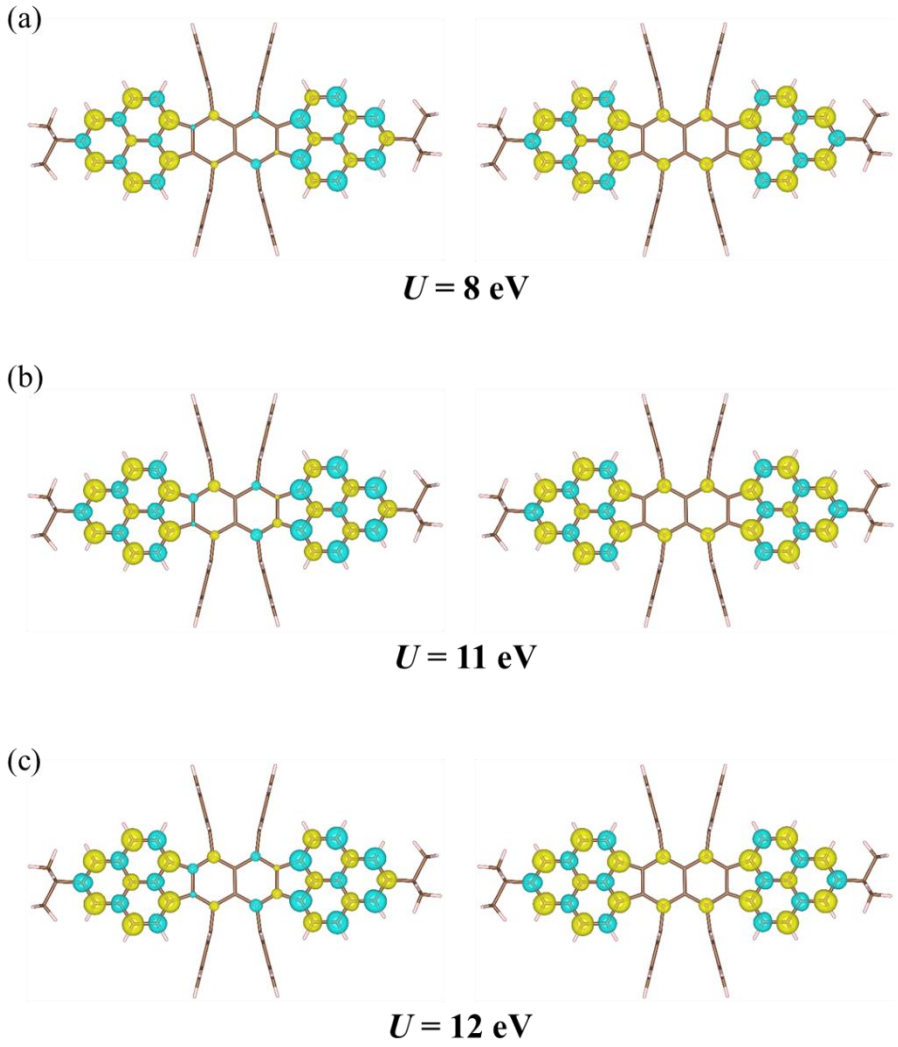
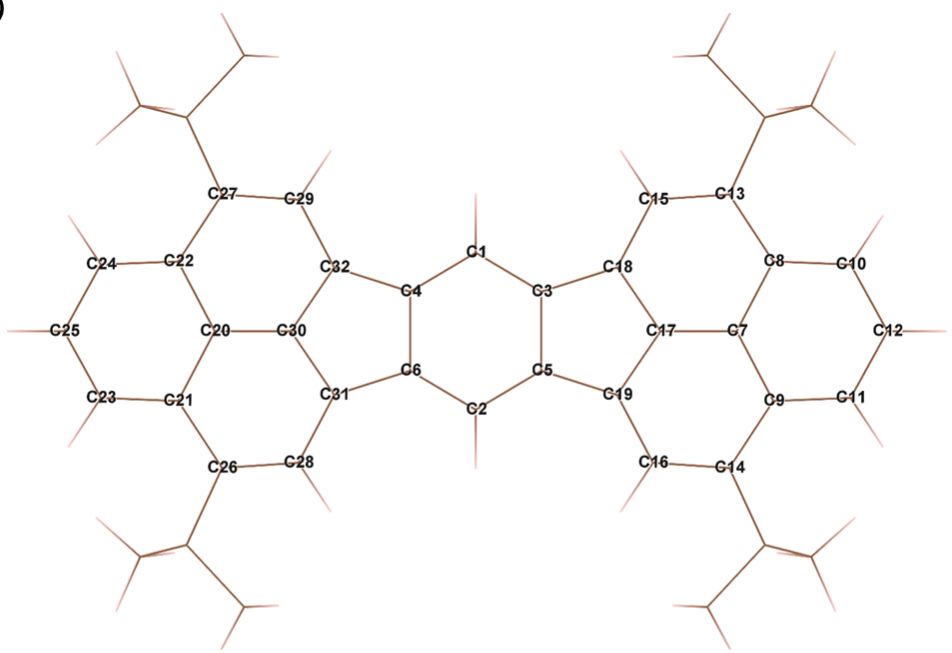


Figure S6. Spin density distributions of Mol 2 estimated by DFT+*U*/plane-wave. (a) $U = 8$ eV, (b) $U = 11$ eV, and (c) $U = 12$ eV. Left and right panels are LS and HS states, respectively. The threshold is 0.003 e^{-}/Bohr^3. Yellow and blue represent the spin density of major and minor spins, respectively.

(a)



(b)

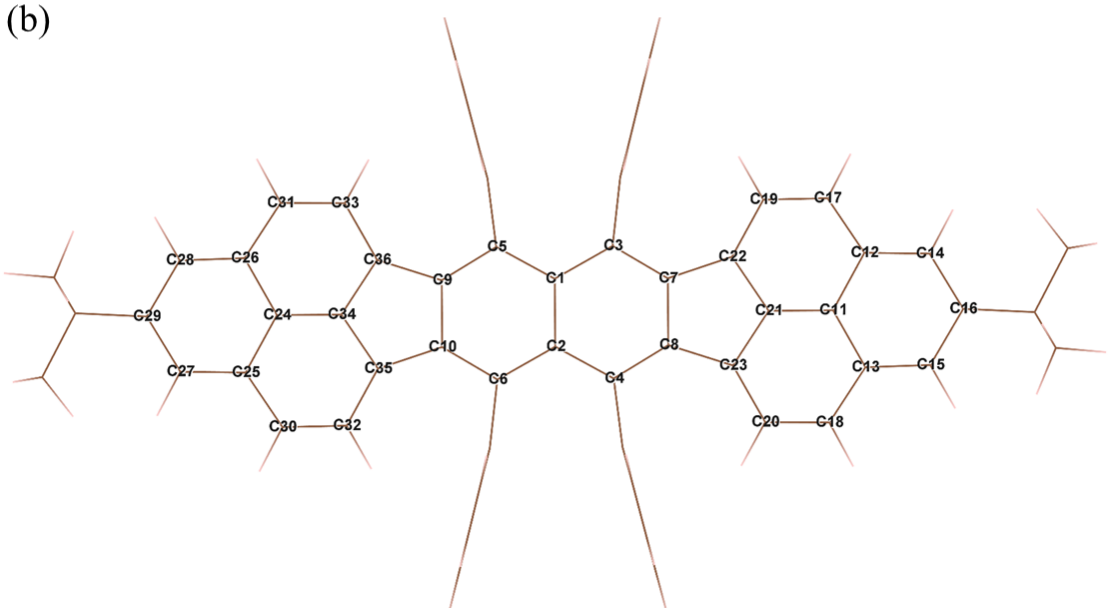


Figure S7. Atom index for carbon atoms in the bisphenarenyl framework.

Table S1. Local magnetic moments of carbon atoms in the bisphenarenyl framework of the LS state of Mol 1.

Atom index	Group	PBE0	U = 8 eV	U = 11 eV	U = 12 eV
C1	G3	0.000	0.000	0.000	0.000
C2	G3	0.000	0.000	0.000	0.000
C3	G3	–0.011	–0.011	–0.017	–0.019
C4	G3	0.011	0.011	0.017	0.019
C5	G3	–0.011	–0.011	–0.017	–0.019
C6	G3	0.011	0.011	0.017	0.019
C7	G2	0.028	0.032	0.061	0.073
C8	G2	–0.05	–0.052	–0.084	–0.097
C9	G2	–0.05	–0.052	–0.084	–0.097
C10	G1	0.096	0.096	0.132	0.146
C11	G1	0.096	0.096	0.132	0.146
C12	G2	–0.049	–0.053	–0.087	–0.100
C13	G1	0.117	0.109	0.146	0.159
C14	G1	0.117	0.109	0.146	0.159
C15	G2	–0.058	–0.058	–0.090	–0.102
C16	G2	–0.058	–0.058	–0.090	–0.102
C17	G2	–0.046	–0.046	–0.075	–0.086
C18	G1	0.115	0.100	0.131	0.142
C19	G1	0.115	0.100	0.131	0.142
C20	G2	–0.028	–0.032	–0.061	–0.073
C21	G2	0.050	0.052	0.084	0.097
C22	G2	0.050	0.052	0.084	0.097
C23	G1	–0.096	–0.096	–0.132	–0.146
C24	G1	–0.096	–0.096	–0.132	–0.146
C25	G2	0.049	0.053	0.087	0.100
C26	G1	–0.117	–0.109	–0.146	–0.159
C27	G1	–0.117	–0.109	–0.146	–0.159
C28	G2	0.058	0.058	0.090	0.102
C29	G2	0.058	0.058	0.090	0.102
C30	G2	0.046	0.046	0.075	0.086
C31	G1	–0.115	–0.100	–0.131	–0.142
C32	G1	–0.115	–0.100	–0.131	–0.142

Table S2. Local magnetic moments of carbon atoms in the bisphenarenyl framework of the HS state of Mol 1.

Atom index	Group	PBE0	U = 8 eV	U = 11 eV	U = 12 eV
C1	G3	0.036	0.042	0.046	0.048
C2	G3	0.036	0.042	0.046	0.048
C3	G3	0.010	0.010	0.008	0.007
C4	G3	0.010	0.010	0.008	0.007
C5	G3	0.010	0.010	0.008	0.007
C6	G3	0.010	0.010	0.008	0.007
C7	G2	0.032	0.041	0.066	0.077
C8	G2	–0.058	–0.066	–0.092	–0.103
C9	G2	–0.058	–0.066	–0.092	–0.103
C10	G1	0.117	0.128	0.152	0.162
C11	G1	0.117	0.128	0.152	0.162
C12	G2	–0.059	–0.071	–0.098	–0.109
C13	G1	0.138	0.145	0.168	0.177
C14	G1	0.138	0.145	0.168	0.177
C15	G2	–0.060	–0.067	–0.092	–0.103
C16	G2	–0.060	–0.067	–0.092	–0.103
C17	G2	–0.054	–0.061	–0.084	–0.094
C18	G1	0.132	0.134	0.153	0.161
C19	G1	0.132	0.134	0.153	0.161
C20	G2	0.032	0.041	0.066	0.077
C21	G2	–0.058	–0.066	–0.092	–0.103
C22	G2	–0.058	–0.066	–0.092	–0.103
C23	G1	0.117	0.128	0.152	0.162
C24	G1	0.117	0.128	0.152	0.162
C25	G2	–0.059	–0.071	–0.098	–0.109
C26	G1	0.138	0.145	0.168	0.177
C27	G1	0.138	0.145	0.168	0.177
C28	G2	–0.060	–0.067	–0.092	–0.103
C29	G2	–0.060	–0.067	–0.092	–0.103
C30	G2	–0.054	–0.061	–0.084	–0.094
C31	G1	0.132	0.134	0.153	0.161
C32	G1	0.132	0.134	0.153	0.161

Table S3. Local magnetic moments of carbon atoms in the bisphenarenyl framework of the LS state of Mol 2.

atom index	group	PBE0	U = 8 eV	U = 11 eV	U = 12 eV
C1	G3	–0.003	–0.003	–0.005	–0.005
C2	G3	0.003	0.003	0.005	0.005
C3	G2	–0.023	–0.023	–0.027	–0.029
C4	G2	–0.031	–0.030	–0.038	–0.042
C5	G2	0.031	0.03	0.038	0.042
C6	G2	0.023	0.023	0.027	0.029
C7	G3	0.015	0.016	0.023	0.025
C8	G3	0.021	0.023	0.032	0.036
C9	G3	–0.021	–0.023	–0.032	–0.036
C10	G3	–0.015	–0.016	–0.023	–0.025
C11	G2	–0.031	–0.039	–0.067	–0.079
C12	G2	0.053	0.057	0.086	0.098
C13	G2	0.055	0.059	0.088	0.100
C14	G1	–0.112	–0.116	–0.148	–0.159
C15	G1	–0.125	–0.129	–0.163	–0.175
C16	G2	0.060	0.068	0.101	0.114
C17	G1	–0.117	–0.116	–0.146	–0.157
C18	G1	–0.124	–0.122	–0.153	–0.164
C19	G2	0.062	0.065	0.095	0.107
C20	G2	0.065	0.069	0.099	0.111
C21	G2	0.050	0.053	0.080	0.091
C22	G1	–0.119	–0.110	–0.136	–0.145
C23	G1	–0.127	–0.117	–0.144	–0.153
C24	G2	0.031	0.039	0.067	0.079
C25	G2	–0.053	–0.057	–0.086	–0.098
C26	G2	–0.055	–0.059	–0.088	–0.100
C27	G1	0.112	0.116	0.148	0.159
C28	G1	0.125	0.129	0.163	0.175
C29	G2	–0.060	–0.068	–0.101	–0.114
C30	G1	0.117	0.116	0.146	0.157
C31	G1	0.124	0.122	0.153	0.164
C32	G2	–0.062	–0.065	–0.095	–0.107
C33	G2	–0.065	–0.069	–0.099	–0.111
C34	G2	–0.050	–0.053	–0.080	–0.091

C35	G1	0.119	0.110	0.136	0.145
C36	G1	0.127	0.117	0.144	0.153

Table S4. Local magnetic moments of carbon atoms in the bisphenarenyl framework of the HS state of Mol 2.

Atom index	group	PBE0	U = 8 eV	U = 11 eV	U = 12 eV
C1	G3	0.009	0.008	0.007	0.006
C2	G3	0.009	0.008	0.007	0.006
C3	G2	0.035	0.042	0.047	0.049
C4	G2	0.038	0.045	0.050	0.053
C5	G2	0.038	0.045	0.050	0.053
C6	G2	0.035	0.042	0.047	0.049
C7	G3	–0.006	–0.007	–0.012	–0.014
C8	G3	–0.008	–0.009	–0.014	–0.016
C9	G3	–0.008	–0.009	–0.014	–0.016
C10	G3	–0.006	–0.007	–0.012	–0.014
C11	G2	0.033	0.043	0.069	0.081
C12	G2	–0.057	–0.064	–0.090	–0.101
C13	G2	–0.059	–0.066	–0.092	–0.103
C14	G1	0.122	0.133	0.158	0.168
C15	G1	0.136	0.148	0.175	0.185
C16	G2	–0.065	–0.078	–0.107	–0.119
C17	G1	0.126	0.133	0.157	0.166
C18	G1	0.133	0.140	0.163	0.173
C19	G2	–0.062	–0.069	–0.097	–0.108
C20	G2	–0.065	–0.073	–0.100	–0.111
C21	G2	–0.054	–0.061	–0.085	–0.095
C22	G1	0.126	0.127	0.147	0.155
C23	G1	0.134	0.135	0.155	0.162
C24	G2	0.033	0.043	0.069	0.081
C25	G2	–0.057	–0.064	–0.090	–0.101
C26	G2	–0.059	–0.066	–0.092	–0.103
C27	G1	0.122	0.133	0.158	0.168
C28	G1	0.136	0.148	0.175	0.185
C29	G2	–0.065	–0.078	–0.107	–0.119
C30	G1	0.126	0.133	0.157	0.166
C31	G1	0.133	0.140	0.163	0.173
C32	G2	–0.062	–0.069	–0.097	–0.108
C33	G2	–0.065	–0.073	–0.100	–0.111
C34	G2	–0.054	–0.061	–0.085	–0.095

C35	G1	0.126	0.127	0.147	0.155
C36	G1	0.134	0.135	0.155	0.162

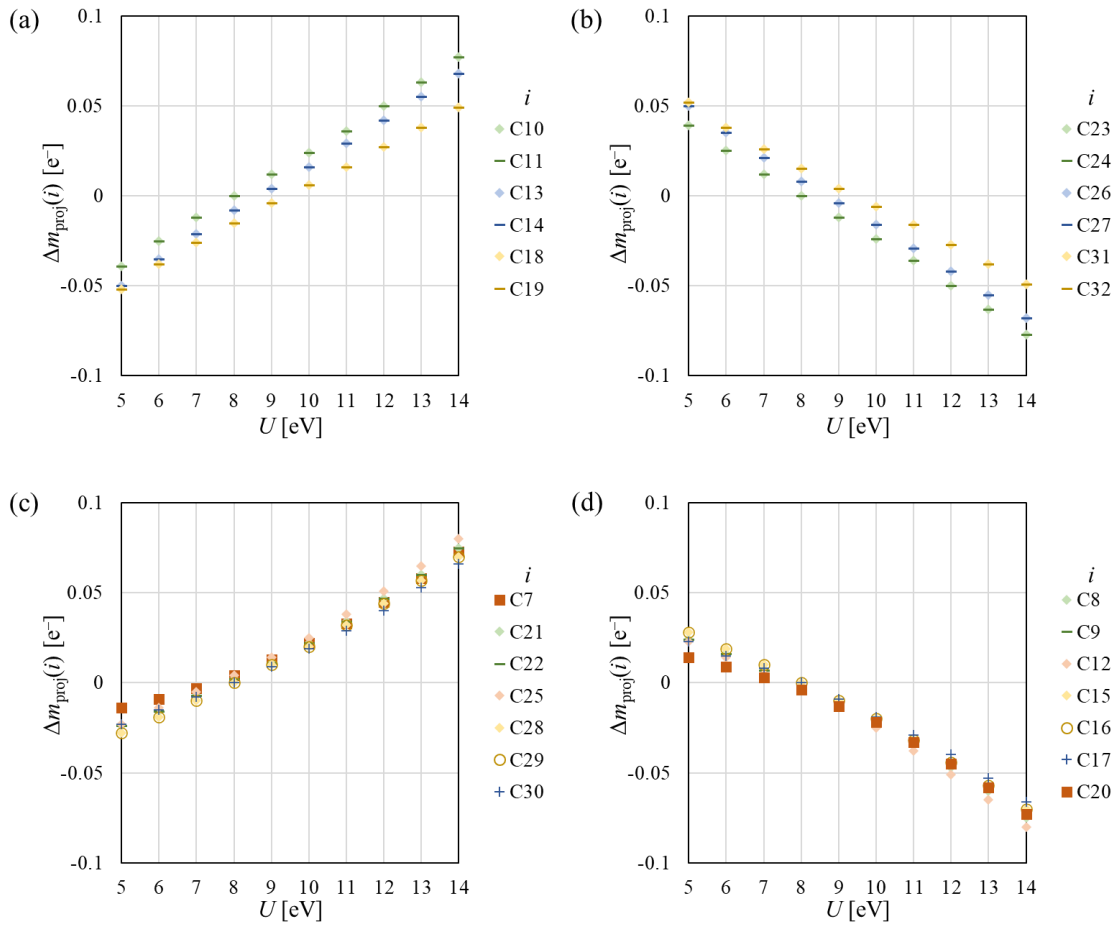


Figure S8. Local magnetic moment dependences on the *U* values of Mol 1 with LS state. (a) carbon atoms of G1 with up spin (C10, C11, C13, C14, C18, and C19 atoms), (b) carbon atoms of G1 with down spin (C23, C24, C26, C27, C31, and C32 atoms), (c) carbon atoms of G2 with up spin (C7, C21, C22, C25, C28, C29, and C30 atoms), and (d) carbon atoms of G2 with down spin (C8, C9, C12, C15, C16, C17, and C20 atoms).

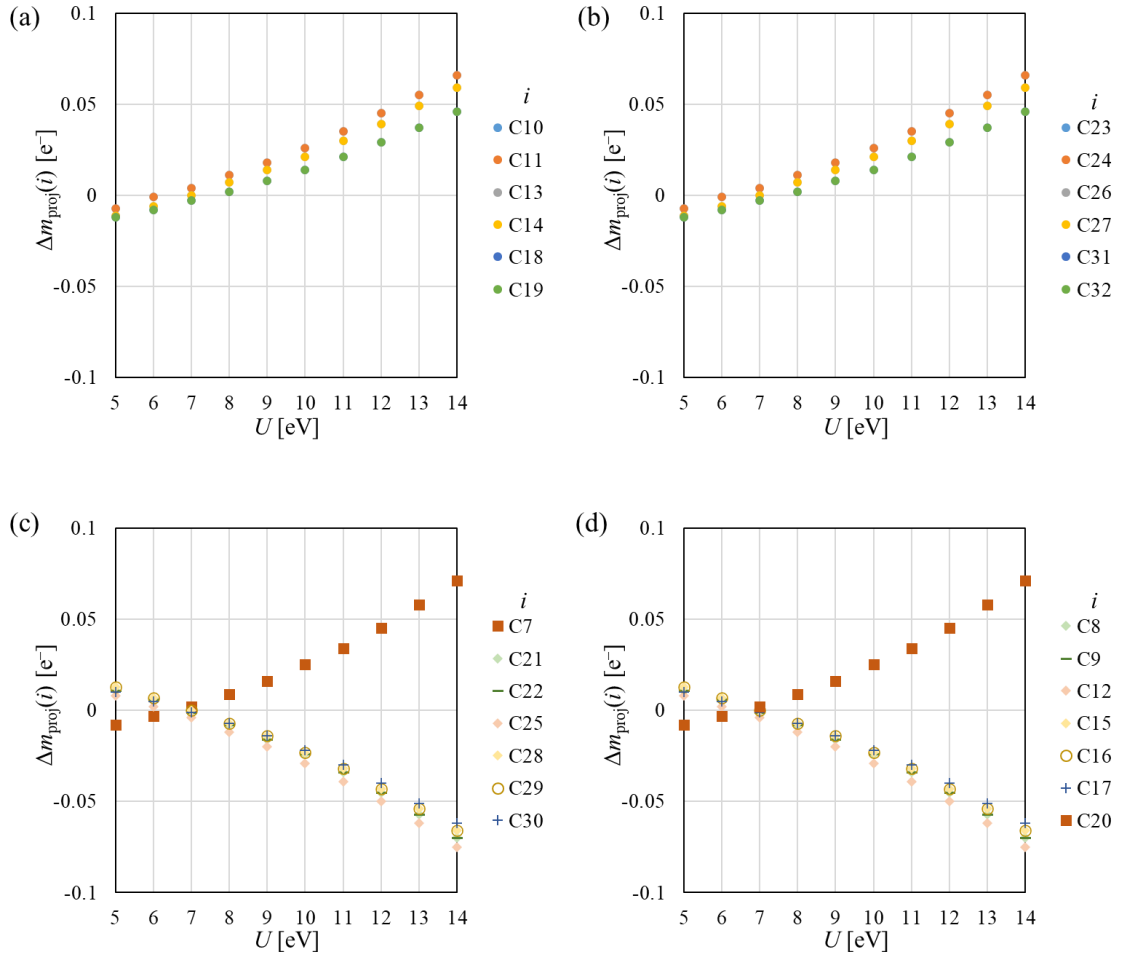


Figure S9. Local magnetic moment dependences on the *U* values of Mol 1 with HS state. (a) carbon atoms of G1 (C10, C11, C13, C14, C18, and C19 atoms) which have up spin when it is in LS state, (b) carbon atoms of G1 (C23, C24, C26, C27, C31, and C32 atoms) which have down spin when it is in LS state, (c) carbon atoms of G2 (C7, C21, C22, C25, C28, C29, and C30 atoms) which have up spin when it is in LS state, and (d) carbon atoms of G2 (C8, C9, C12, C15, C16, C17, and C20 atoms) which have down spin when it is in LS state.

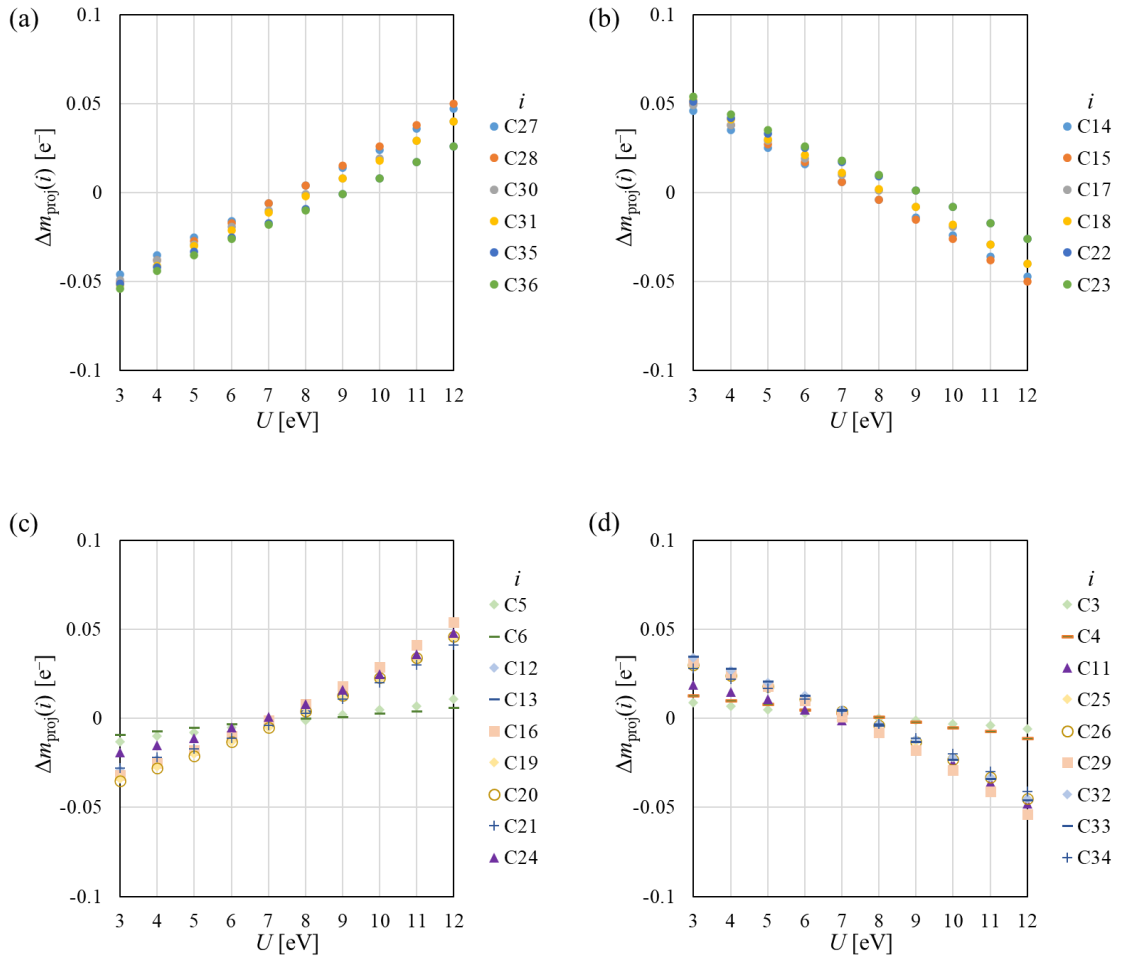


Figure S10. Local magnetic moment dependences on the *U* values of Mol 2 with LS state. (a) carbon atoms of G1 with up spin (C27, C28, C30, C31, C35, and C36 atoms), (b) carbon atoms of G1 with down spin (C14, C15, C17, C18, C22, and C23 atoms), (c) carbon atoms of G2 with up spin (C5, C6, C12, C13, C16, C19, C20, C21, and C24 atoms), and (d) carbon atoms of G2 with down spin (C3, C4, C11, C25, C26, C29, C32, C33, and C34 atoms).

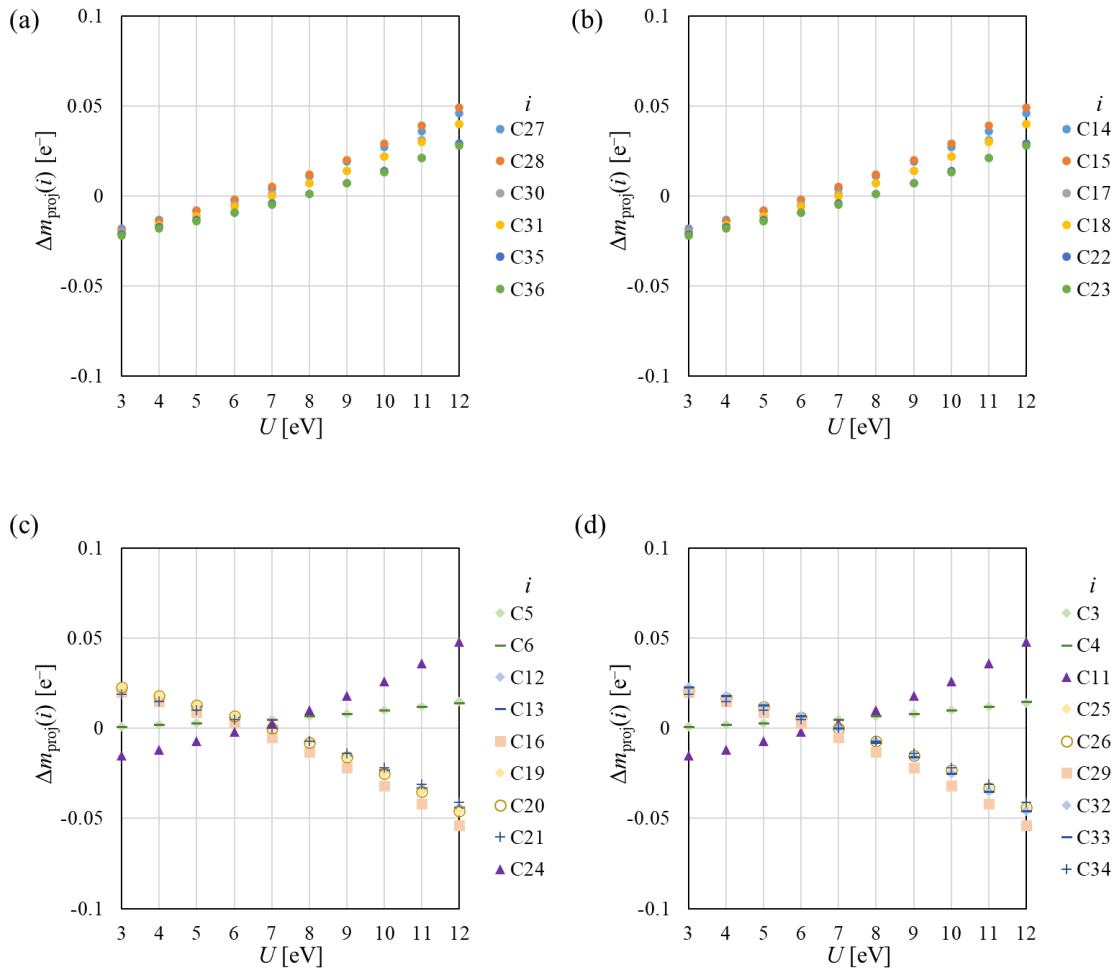


Figure S11. Local magnetic moment dependences on the *U* values of Mol 2 with HS state. (a) carbon atoms of G1 (C27, C28, C30, C31, C35, and C36 atoms) which have up spin when it is in LS state, (b) carbon atoms of G1 (C14, C15, C17, C18, C22, and C23 atoms) which have down spin when it is in LS state, (c) carbon atoms of G2 (C5, C6, C12, C13, C16, C19, C20, C21, and C24 atoms) which have up spin when it is in LS state, and (d) carbon atoms of G2 (C3, C4, C11, C25, C26, C29, C32, C33, and C34 atoms) which have down spin when it is in LS state.