

Supplementary Information

Two-dimensional robust topological insulator with coexisting ferroelectric and valley polarization

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Computational details on Z_2 topological invariant

The topological Z_2 invariant is confirmed by using the method which uses the Wilson loop methods¹ base on the $U(2N)$ non-Abelian Berry connection by Yu et al². In this method, each state of n -th occupied band is indexed by $|n, k_x, k_y\rangle$, and a square matrix $F(k_x, k_y)$ containing the overlap integrals

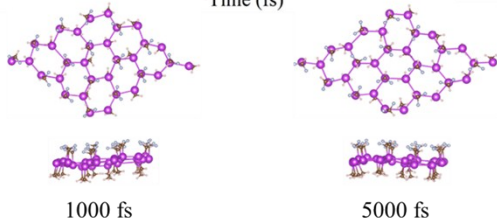
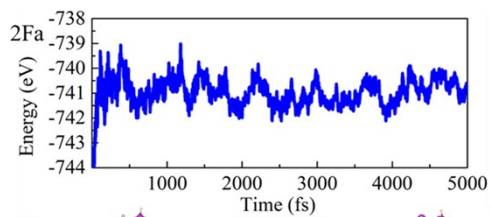
$$[F(k_x, k_y)]_{mn} = \langle m, k_x, k_y | n, k_x, k_y \rangle \quad (1)$$

is defined. The complex unitary square matrix is calculated by

$$D(k_y) = \prod_{j=0}^{N_x-1} F(j\Delta k_x, k_y), \quad (2)$$

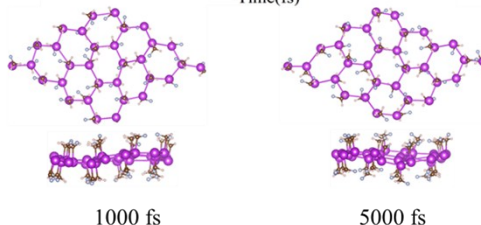
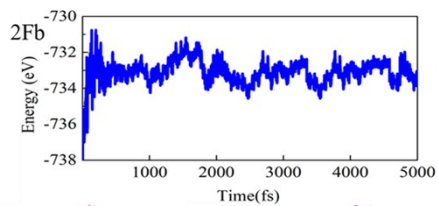
whose complex eigenvalues $\lambda(k_y)$ have phase angle θ . Here $\Delta k_x = \frac{2\pi}{N_x a}$ is the discrete spacing of N_x points along k_x direction. The Wannier center evolutions $\theta(k_y)$ of 3Fa is plotted in Figure 4(b), and the Z_2 invariant can be obtained by counting the even or odd number of crossings of any arbitrary horizontal reference line.

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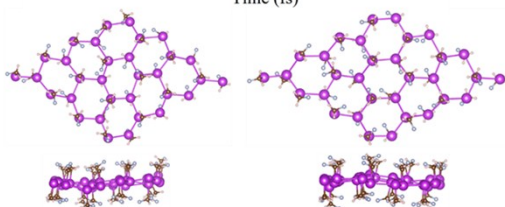
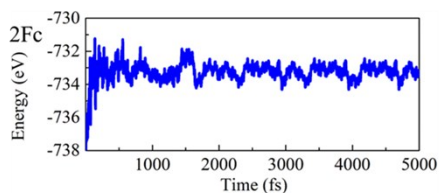
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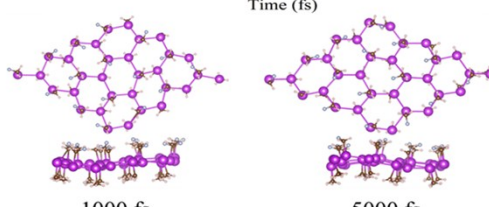
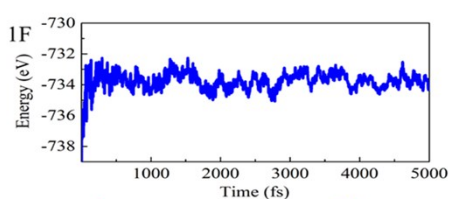
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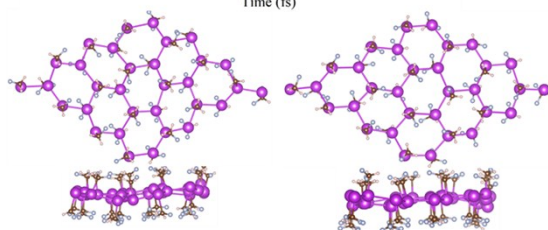
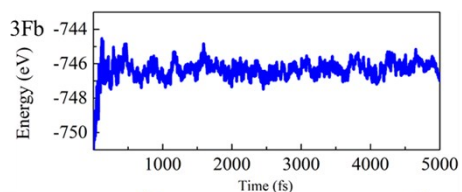
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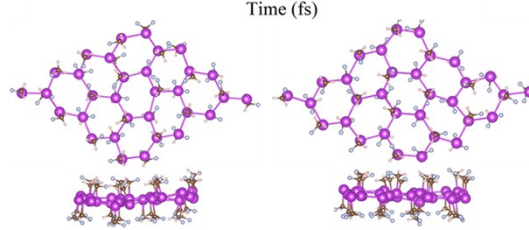
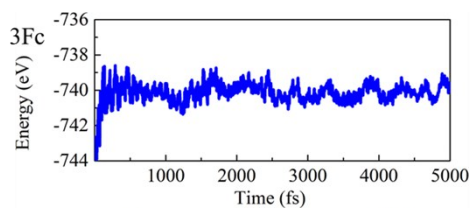
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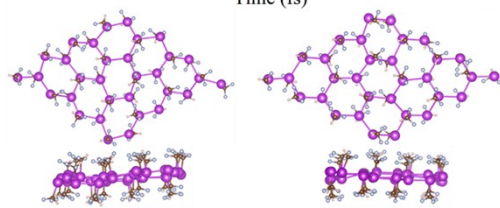
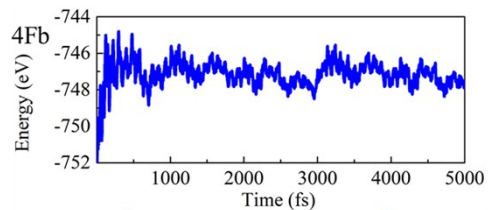
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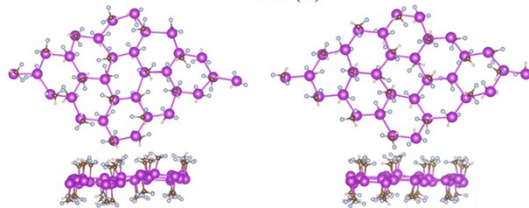
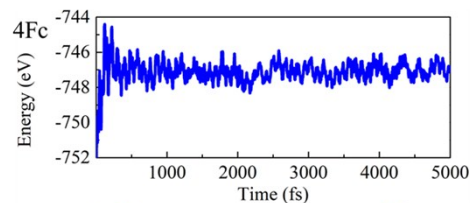
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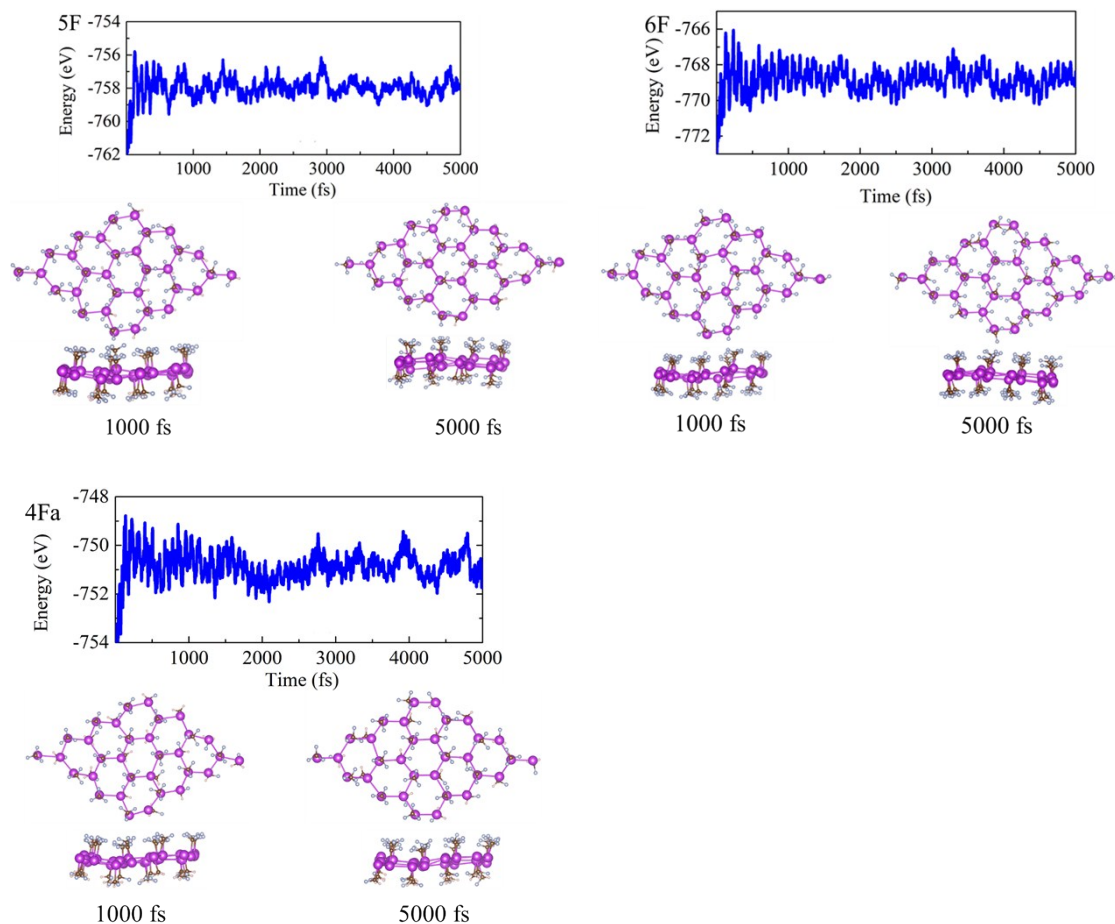


Figure S1 Variation of energy with increasing time obtained from MD simulation at 300 K for $\text{Bi}_2\text{C}_2\text{H}_{6-x}\text{F}_x$ films.

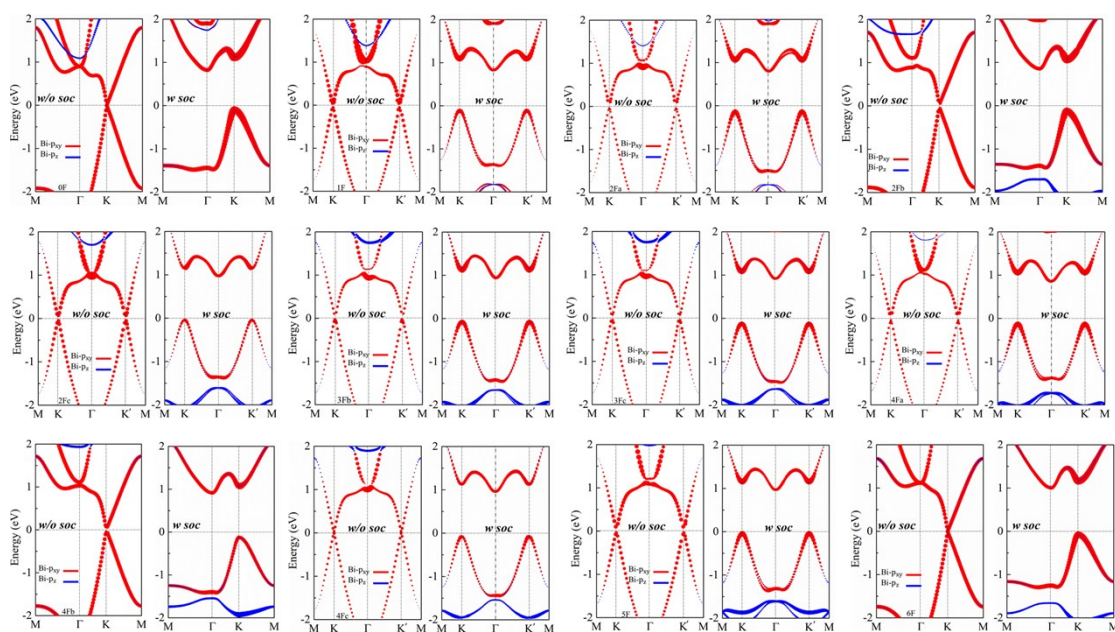


Figure S2 Band structure of $\text{Bi}_2\text{C}_2\text{H}_{6-x}\text{F}_x$ films monolayer without and with SOC. The blue, red dots present the weights of the Bi- p_{xy} and Bi- p_z character, respectively.

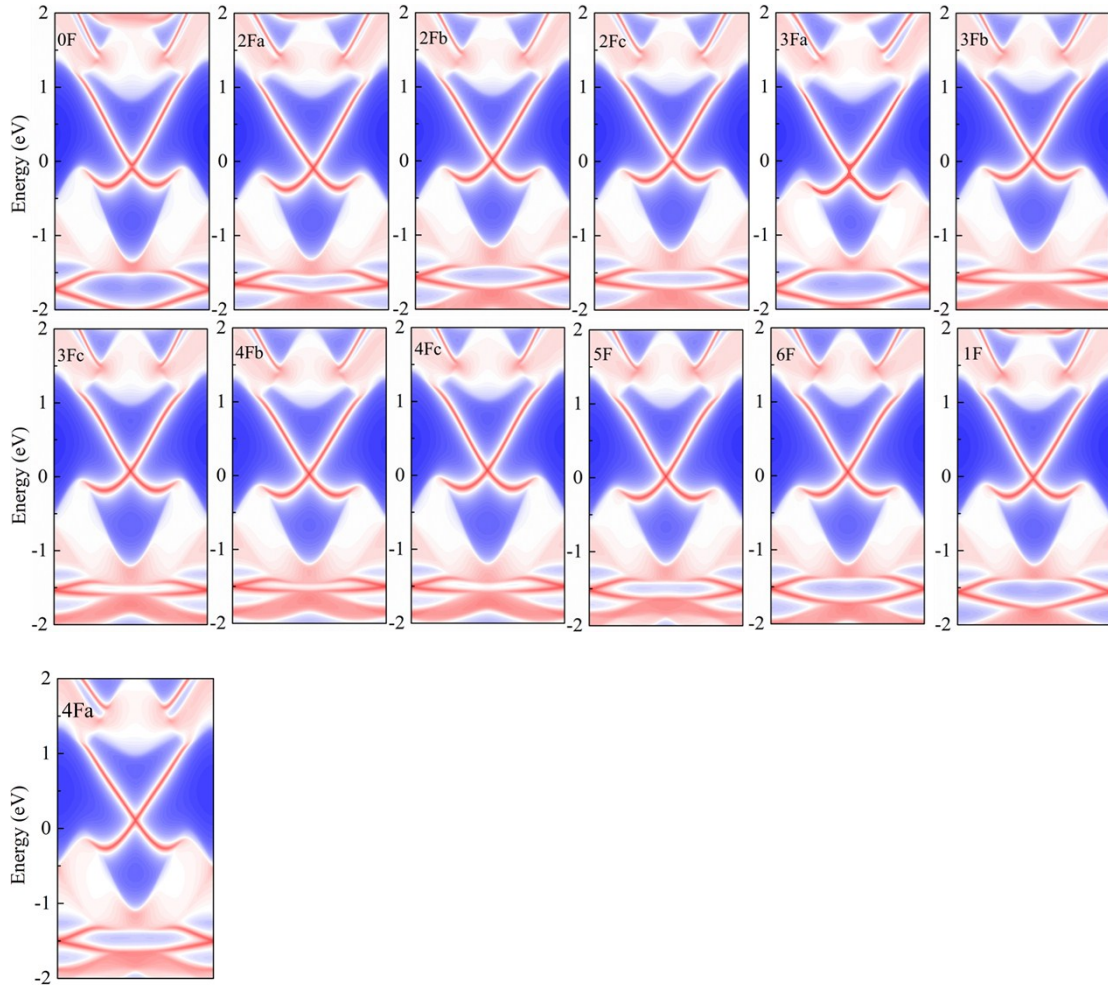


Figure S3 Total edge density of states for $\text{Bi}_2\text{C}_2\text{H}_{6-x}\text{F}_x$. Evolutions of Wannier centers for $\text{Bi}_2\text{C}_2\text{H}_6$ $x\text{F}_x$ along k_y .

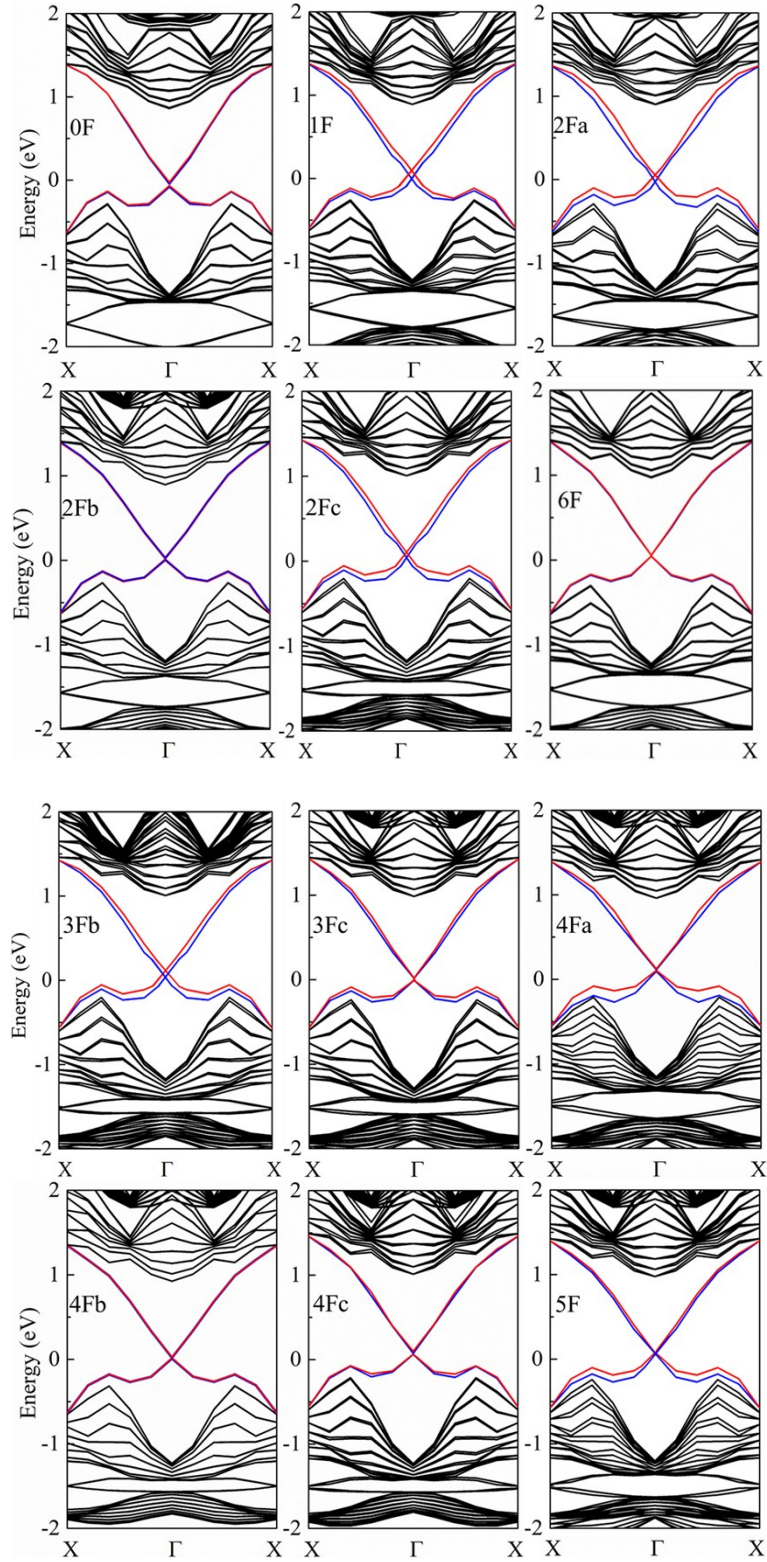


Figure S4 The edge states. The red and blues lines correspond to the edge state from the right and left sides, respectively,

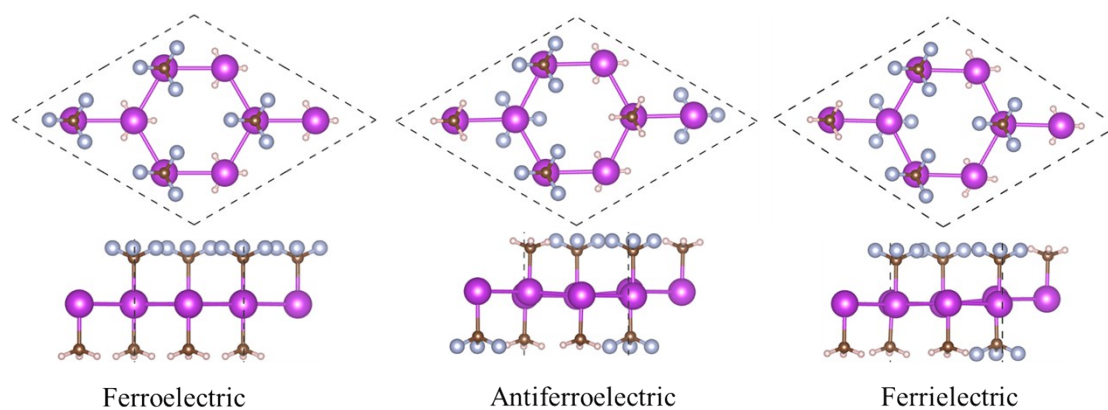


Figure S5 Three structures of 3Fa, ferroelectric, antiferroelectric, ferrielectric.

Table S1 The charge transfer situation of fluorination-methyl-functionalized bismuth ($\text{Bi}_2\text{C}_2\text{H}_6$ - xF_x) films.

atom (0F)	CHARGE	atom (1F)	CHARGE	atom (2Fa)	CHARGE
H	0.9327	H	0.9044	H	0.8854
H	0.9327	H	0.8657	H	0.9438
H	0.932	H	0.9450	H	0.9172
H	0.932	H	0.9162	H	0.9204
H	0.8870	H	0.9096	C	3.2093
H	0.8870	C	3.9003	C	3.9003
C	4.5534	C	4.4759	F	7.6257
C	4.5534	F	7.6416	F	7.6268
Bi	4.7077	Bi	4.6854	Bi	4.7185
Bi	4.6821	Bi	4.7558	Bi	4.6368

atom (2Fb)	CHARGE	atom (2Fc)	CHARGE	atom (3Fa)	CHARGE
H	0.8557	H	0.9589	H	0.9454
H	0.8986	H	1.0654	H	0.9365
H	0.9075	H	1.0633	H	0.9338

H	0.9220	H	0.9738	C	2.5268
C	3.9189	C	3.3827	C	4.4495
C	3.8436	C	3.3717	F	7.6131
F	7.6408	F	7.7812	F	7.6167
F	7.6411	F	7.7783	F	7.6204
Bi	4.7108	Bi	4.8218	Bi	4.7402
Bi	4.6612	Bi	4.8029	Bi	4.6176

atom (3Fb)	CHARGE	atom (3Fc)	CHARGE	atom (4Fa)	CHARGE
H	0.8978	H	0.9201	H	0.8763
H	0.8995	H	0.9024	H	0.8783
H	0.8724	H	0.8714	C	2.5373
C	3.2141	C	3.2108	C	3.8946
C	3.8692	C	3.8471	F	7.6194
F	7.6314	F	7.6324	F	7.6244
F	7.6289	F	7.6321	F	7.6402
F	7.6406	F	7.6362	F	7.6210
Bi	4.6971	Bi	4.6920	Bi	4.6920
Bi	4.6490	Bi	4.6555	Bi	4.6555

atom (4Fb)	CHARGE	atom (4Fc)	CHARGE	atom (5F)	CHARGE
H	0.8588	H	0.8604	H	0.8868
H	0.8644	H	0.8496	C	2.5346

C	3.2292	C	3.2384	C	3.2187
C	3.2417	C	3.1981	F	7.6180
F	7.6328	F	7.6297	F	7.6190
F	7.6282	F	7.6314	F	7.6263
F	7.6271	F	7.6333	F	7.6276
F	7.6274	F	7.6369	F	7.6243
Bi	4.6461	Bi	4.6451	Bi	4.6355
Bi	4.6444	Bi	4.6772	Bi	4.6091

atom (6F)	CHARGE
C	0.9780
C	0.9780
F	3.8973
F	3.8951
F	3.8993
F	3.8973
F	3.8950
F	3.8994
Bi	7.3306
Bi	7.3300

The corresponding structural information on POSCAR in our calculations:

(a) POSCAR of 0F

1.000000000

4.7181260097862250 -2.7240113647305648 0.0000000000000000

0.0000000371610732	5.4480226650962580	0.0000000000000000
0.0000000000000000	0.0000000000000000	20.0000000000000000

H	C	Bi
6	2	2

Direct

0.2193028727447784	0.7728810842834658	0.6488934572188833
0.2271189157165347	0.4464217884613127	0.6488934572188833
0.5535782115386870	0.7806971272552223	0.6488934572188833
0.7806971272552223	0.2271189157165347	0.3511065427811157
0.7728810842834658	0.5535782115386870	0.3511065427811157
0.4464217884613127	0.2193028727447784	0.3511065427811157
0.3333333333333357	0.6666666666666643	0.6294593440074644
0.6666666666666643	0.3333333333333357	0.370540655925351
0.3333333333333357	0.6666666666666643	0.5015406473196501
0.6666666666666643	0.3333333333333357	0.4984593526803493

(b) POSCAR of 1F

1.0000000000000000		
4.8090390672149868	-2.7765000000000040	-0.0000000000000000
0.0000000000000000	5.5530000000000008	-0.0000000000000000
0.0000000000000000	0.0000000000000000	20.0000000000000000

H	C	F	Bi
5	2	1	2

Direct

0.5502048785471771	0.7549210418996850	0.6322300965614502
0.2228096086498041	0.4304856289161094	0.6323387927314243
0.7597760722187559	0.2076702897368179	0.3592557980366138
0.4380355562533964	0.2115760327561702	0.3616130794226819
0.7637618849130661	0.5333789282751553	0.3615004235920551
0.3346683610224161	0.6458120230818380	0.6139692432161823
0.6549979171542404	0.3165323911189262	0.3784098984267601
0.1990773039391769	0.7853572308817718	0.6386591396223480
0.3330599461910779	0.6466595231460062	0.4973433873008446
0.6626867552344606	0.3093723250014122	0.4940186582647144

(c) POSCAR of 2Fa

1.0000000000000000		
4.7406230603160138	-2.7370000000000019	-0.0000000000000000
0.0000000000000000	5.4740000000000002	-0.0000000000000000
0.0000000000000000	0.0000000000000000	20.0000000000000000

H C F Bi
4 2 2 2

Direct

0.2365761051838135	0.7887849980052264	0.6251886808876270
0.7800463583919807	0.2509648891273764	0.3514156031520402
0.4504320711141268	0.2483559096663905	0.3511529279938828
0.7774288222884279	0.5779667573844508	0.3511673071696360
0.3432362234174346	0.6859861610327681	0.6012575565411251
0.6691722063234761	0.3592030187262015	0.3689717652051874
0.2156158538088632	0.4098271206909048	0.6238440451622211
0.6198061198894917	0.8193317837813664	0.6237067153119300
0.3349434723259901	0.6932490939317238	0.4834392065447406
0.6682089693881349	0.3599352033148962	0.4847687241434264

(d) POSCAR of 2Fb

1.0000000000000000

4.7774118194687780	-2.7582400000000029	0.0000000000000000
0.0000000000000000	5.5164799999999996	0.0000000000000000
0.0000000000000000	0.0000000000000000	20.0000000000000000

H C F Bi
4 2 2 2

Direct

0.5533158836036165	0.7614535018673294	0.6369175528574829
0.2235234668478014	0.4352552395537757	0.6369175590639955
0.4353060594993110	0.2163962066383860	0.3630824409360045
0.7650974876331844	0.5425954906073969	0.3630824471425171
0.3359535087052379	0.6525741259172193	0.6186883182790623
0.6526684343976896	0.3252765823415800	0.3813116817209377
0.2003936678669618	0.7926635227679242	0.6445103922110462
0.7882282752359657	0.1851871854908751	0.3554896077889538
0.3302541032032451	0.6584639696514998	0.5004558793658305
0.6583678398996753	0.3193867386072995	0.4995441206341695

(e) POSCAR of 2Fc

1.0000000000000000

4.7583765810935947	-2.7472499999999997	0.0000000000000000
0.0000000000000000	5.4945000000000004	0.0000000000000000
0.0000000000000000	0.0000000000000000	20.0000000000000000

H C F Bi
4 2 2 2

Direct

0.5385221723513354	0.7473630425426475	0.6267101603058960
0.1847014078496932	0.7809197130023803	0.6306927676881423
0.7974770519600423	0.5841781787787209	0.3547722420274883
0.7683086627969304	0.1986467696076663	0.3581222205159673
0.3212751945088984	0.6362757668932078	0.6080633720870947
0.6566481168522107	0.3030809446787586	0.3775066990335229
0.4395918326329863	0.1999542569682967	0.3587761031871434
0.2066580345304316	0.4205518084687764	0.6276167849183211
0.3215670568264602	0.6256851363121786	0.4913250149719894
0.6555470380058331	0.2926767438568754	0.4942471369923850

(f) POSCAR of 3Fa

1.0000000000000000

4.7925845845430803	-2.7670000000000021	0.0000000000000000
0.0000000000000000	5.5339999999999989	0.0000000000000000
0.0000000000000000	0.0000000000000000	20.0000000000000000

H	C	F	Bi
3	2	3	2

Direct

0.7700578549932970	0.2166791059235820	0.3529448807784519
0.4458457586528368	0.2172523774899915	0.3530421871180280
0.7706308675353668	0.5414651788626088	0.3529980676683238
0.3292722070959471	0.6581977941308637	0.6033193000957127
0.6622454952009917	0.3251278839927281	0.3708719484015077
0.5941180040293830	0.7832680874540046	0.6287317450848562
0.1895961498723935	0.3933244560241747	0.6287174975023007
0.2041574685899121	0.7978168338314617	0.6287941400437447
0.3292102057388604	0.6584198000448964	0.4852822347895653
0.6625435017237464	0.3250848684997862	0.4867372553669682

(g) POSCAR of 3Fb

1.0000000000000000

4.7279229709745971	-2.7296676000000000	0.0000000000000000
0.0000000000000000	5.4593351999999999	0.0000000000000000
0.0000000000000000	0.0000000000000000	20.0000000000000000

H	C	F	Bi
3	2	3	2

Direct

0.5559756978241107	0.7623640897640869	0.6370108911530561
0.2235974984115364	0.4322090308489313	0.6371313480542417
0.7685473958122006	0.2141789120212845	0.3591388902771296
0.3382676773088059	0.6496637016918581	0.6174719707521309

0.6659759059272748	0.3206628067384888	0.3839020563917054
0.3884440745592505	0.1926524031345446	0.3614101238180116
0.7988190650713543	0.5982037313931883	0.3612906398915764
0.1979854876417685	0.7930380048976238	0.6401825381860259
0.3431687482962103	0.6437208621375585	0.4988529566483209
0.6767870215261503	0.3101297338483664	0.5036105939327478

(h) POSCAR of 3Fc

1.0000000000000000

4.7279229709745971	-2.7296676000000000	0.0000000000000000
0.0000000000000000	5.4593351999999999	0.0000000000000000
0.0000000000000000	0.0000000000000000	20.0000000000000000

H	C	F	Bi
3	2	3	2

Direct

0.7627491990958445	0.2202321544887291	0.3631034704872690
0.7635111530968776	0.5519083548306072	0.3629223293431728
0.2345538936046700	0.7680515190685563	0.6403755735556692
0.3382510817661886	0.6618859914569796	0.6160124804725112
0.6591622390382771	0.3343991321643031	0.3825781017028927
0.6146371987096827	0.7893654246986263	0.6393123768990325
0.2041545215957410	0.3839396499730085	0.6383825858816502
0.3755568334478286	0.1928689766564547	0.3598392184062822
0.3358201463243883	0.6737785621917027	0.4962675535019093
0.6691723056991630	0.3403935109469567	0.5012083188545506

(i) POSCAR of 4Fa

1.0000000000000000

4.8151012450414754	-2.7800000000000029	0.0000000000000000
0.0000000000000000	5.5599999999999987	0.0000000000000000
0.0000000000000000	0.0000000000000000	20.0000000000000000

H	C	F	Bi
2	2	4	2

Direct

0.4418013372045806	0.2089960004108775	0.3506225080098204
0.7677566302980239	0.5328610853760480	0.3504869116834257
0.3241263233878300	0.6568981543020911	0.6014905060638966
0.6550023921960459	0.3199498605105120	0.3702374363067378
0.5892382231791444	0.7810218401788447	0.6259960304741412
0.1872825109997791	0.3922556479616333	0.6263404261623862
0.7933881207337379	0.1787037802995286	0.3485228599449570
0.2004879712884247	0.7942250549989254	0.6284090649715282

0.3176048254129995	0.6618822337032171	0.4833803906224574
0.6495782760445792	0.3264099086945080	0.4871107702873729

(j) POSCAR of 4Fb

1.0000000000000000

4.7756797686612096	-2.7572399999999999	0.0000000000000000
0.0000000000000000	5.5144799999999998	0.0000000000000000
0.0000000000000000	0.0000000000000000	20.0000000000000000

H	C	F	Bi
2	2	4	2

Direct

0.7576490289582765	0.2311492197297085	0.3582084233556202
0.2390534502838122	0.7572107896645903	0.6418486953759412
0.3377796084002611	0.6494449755455207	0.6168933471073146
0.6540249537114917	0.3341885169219410	0.3831101343950749
0.6123019966585872	0.7720571886429539	0.6394707330709295
0.2022501966834724	0.3741649524450779	0.6392985382649172
0.3790267948001471	0.2045359407723453	0.3606099599912653
0.7827005204928454	0.6090731871636166	0.3605563159436116
0.3297564258102526	0.6591109132423441	0.4971078692954549
0.6630255965795087	0.3258875923478470	0.5028979923048098

(k) POSCAR of 4Fc

1.0000000000000000

4.7192807302651500	-2.7246780000000026	0.0000000000000000
0.0000000000000000	5.4493559999999990	0.0000000000000000
0.0000000000000000	0.0000000000000000	20.0000000000000000

H	C	F	Bi
2	2	4	2

Direct

0.4436465595065400	0.1995333494177345	0.3479152721220444
0.2038046443850945	0.4372138665182419	0.6266431852564196
0.3120824714747812	0.6461735319531741	0.6020405008472238
0.6527305724155639	0.3042457630524353	0.3724816841585721
0.5907819623106576	0.7884876549919682	0.6240347537808333
0.7999757974194850	0.5825943288952899	0.3503453507854459
0.8007572602346755	0.1741353935753622	0.3502196228245680
0.1874251185441622	0.7992223020631428	0.6243944504944565
0.3010097399302509	0.6273399038472292	0.4839560953155129
0.6340524845239344	0.2942574721216289	0.4905659889416683

(l) POSCAR of 5F

1.0000000000000000
 4.7756797686612096 -2.7572399999999999 0.0000000000000000
 0.0000000000000000 5.5144799999999998 0.0000000000000000
 0.0000000000000000 0.0000000000000000 20.0000000000000000

H C F Bi
 1 2 5 2

Direct

0.7619632551622146 0.2239898318948548 0.3567260719362153
 0.3374408834260620 0.6497417168979922 0.6157241293705340
 0.6606713891244311 0.3269409830228938 0.3827803243729235
 0.6029809204056420 0.7821496291232535 0.6417777483232072
 0.2031381048292999 0.3842532789999993 0.6418033723421459
 0.3847448277126588 0.2000164928457977 0.3608567373843030
 0.7901836212203435 0.6030955478779276 0.3609374099410232
 0.2051191833686801 0.7840535532800814 0.6411469592190713
 0.3390153289237148 0.6479230081584291 0.4956679392095324
 0.6723110582056080 0.3146592343747159 0.5025813170059976

(m) POSCAR of 6F

1.0000000000000000
 4.7798477635464440 -2.7596463929689681 0.0000000000000000
 0.0000000000000000 5.5192927859379397 0.0000000000000000
 0.0000000000000001 0.0000000000000000 20.0000000000000000

C F Bi
 2 6 2

Direct

0.3333333333333357 0.6666666666666643 0.6296988706586149
 0.6666666666666643 0.3333333333333357 0.3703011293413851
 0.2004508646648802 0.7995491353351198 0.6586379599965966
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 0.3333333333333357 0.6666666666666643 0.4953262227995268
 0.6666666666666643 0.3333333333333357 0.5046737772004732

References

1. A. Alexandradinata, X. Dai and B. A. Bernevig, *Physical Review B*, 2012, **89**, 1249-1262.
2. R. Yu, X. L. Qi, A. Bernevig, Z. Fang and X. Dai, *Physical Review B*, 2011, **84**, 2250-2262.