Halogenated MOF-5 variants show new configuration, tunable band gaps and enhanced optical response in the visible and near infrared

Supporting Information

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Table S1. Summary of measurements from molecular dynamics simulations.

Material						**Lattice Constant**		**Lattice Fraction**	
		0 K	**600 K**	**BLE**	**Time**	**0K**	**600 K**	**Linear**	**Volume**
		Å	Å	%	ps	Å	Å		
CdI	C-C	1.43	1.47	3%	7.6	19.70	19.41	0.99	0.96
	C-I	2.13	2.34	10%					
CdF	C-C	1.39	1.49	7%	4.45	19.30	19.20	1.00	0.99
	C-F	1.35	1.43	6%					
ZnI	C-C	1.40	1.47	5%	6.36	18.45	18.43	1.00	1.00
	C-I	2.10	2.30	10%					
ZnF	C-C	1.39	1.47	5%	5.4	18.47	18.61	1.01	1.02
	C-F	1.35	1.45	7%					

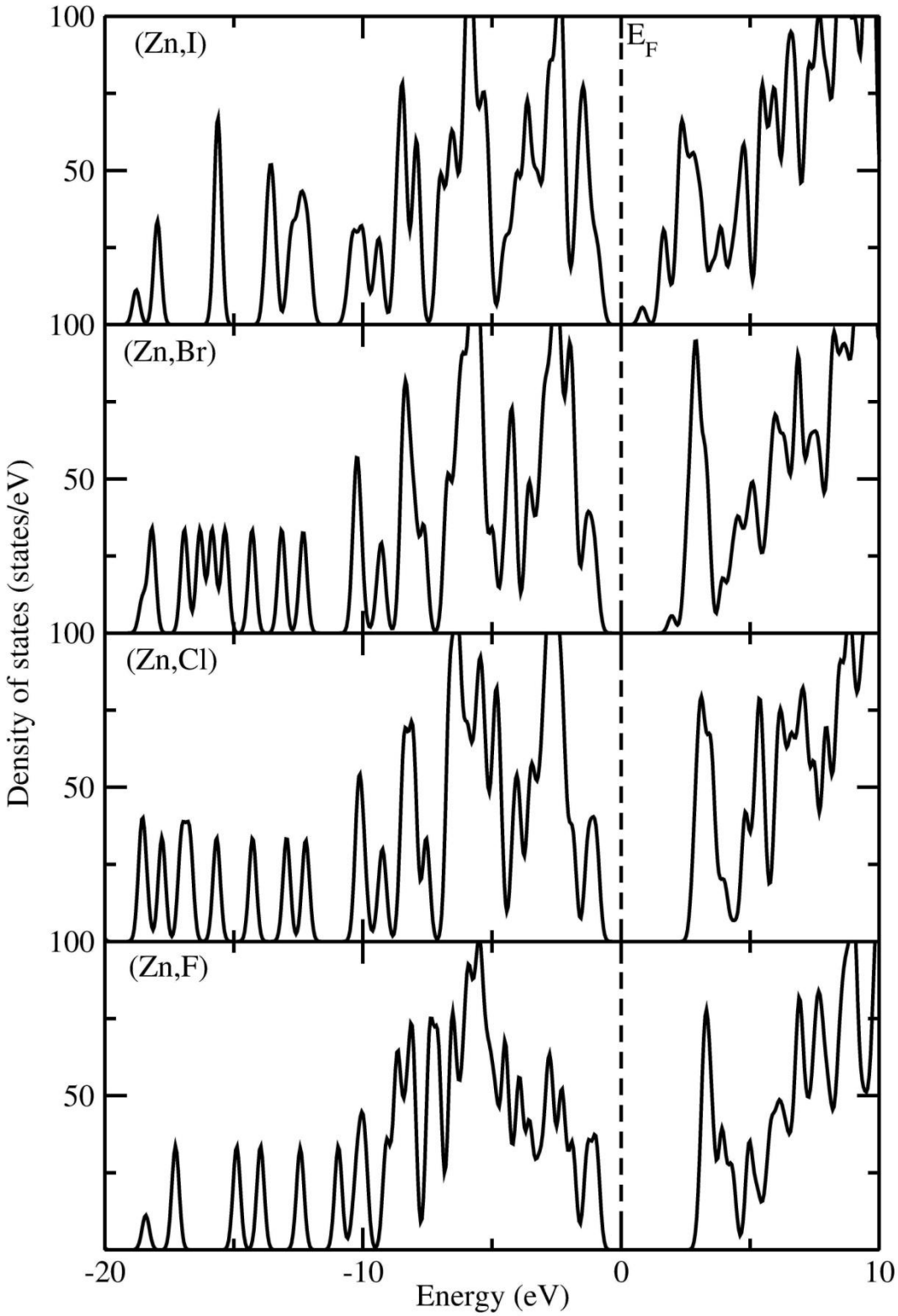

Figure S1. Calculated total density of states (TDOS) for the (Zn, Z)-90 ° subseries, (Z = F, Cl, Br, I) in the equilibrium cubic structure with *Fm-3m* symmetry (no. 225).

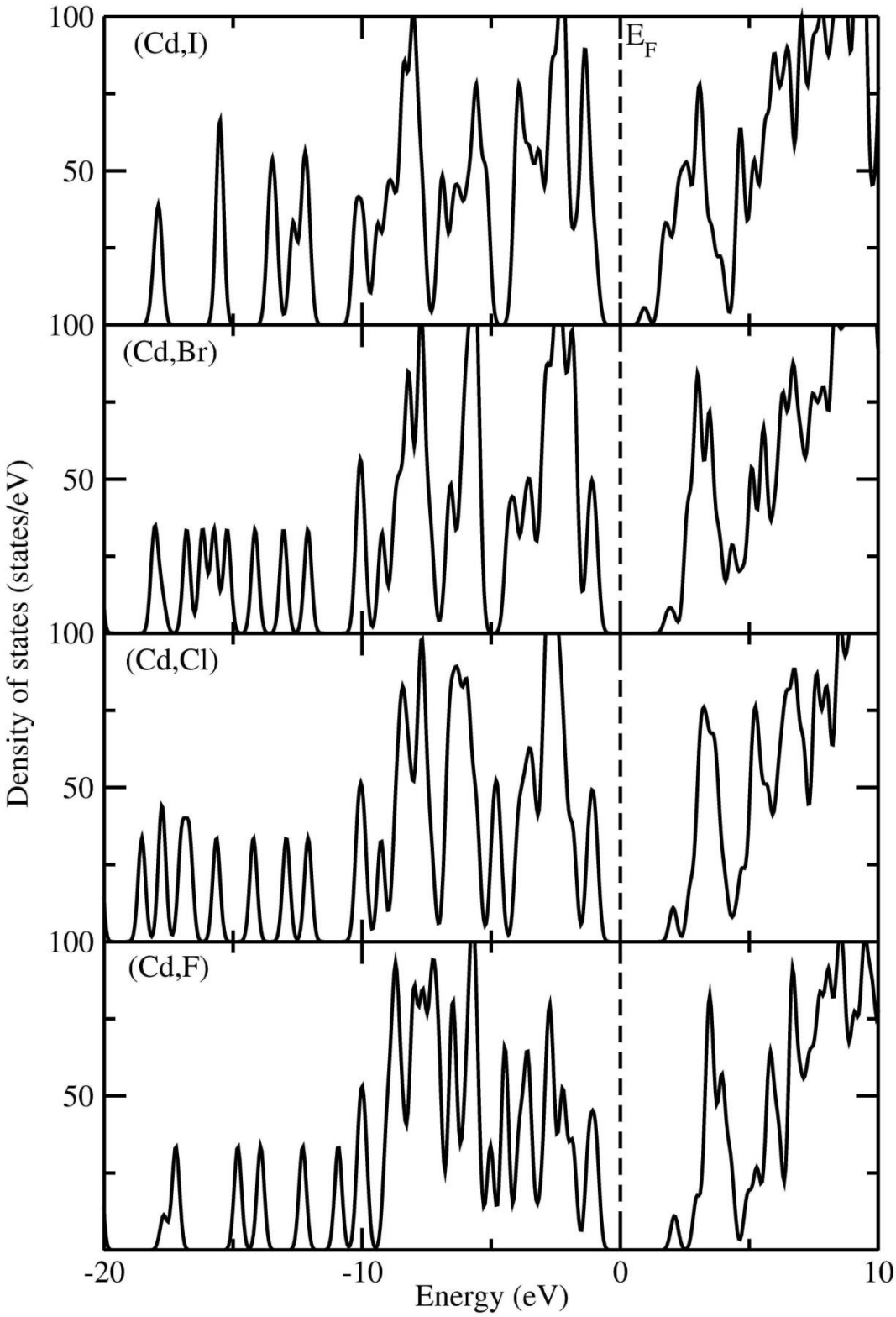

Figure S2. Calculated total density of states (TDOS) for the (Cd, Z)-90 °subseries, (Z = F, Cl, Br, I) in the equilibrium cubic structure with *Fm-3m* symmetry (no. 225).

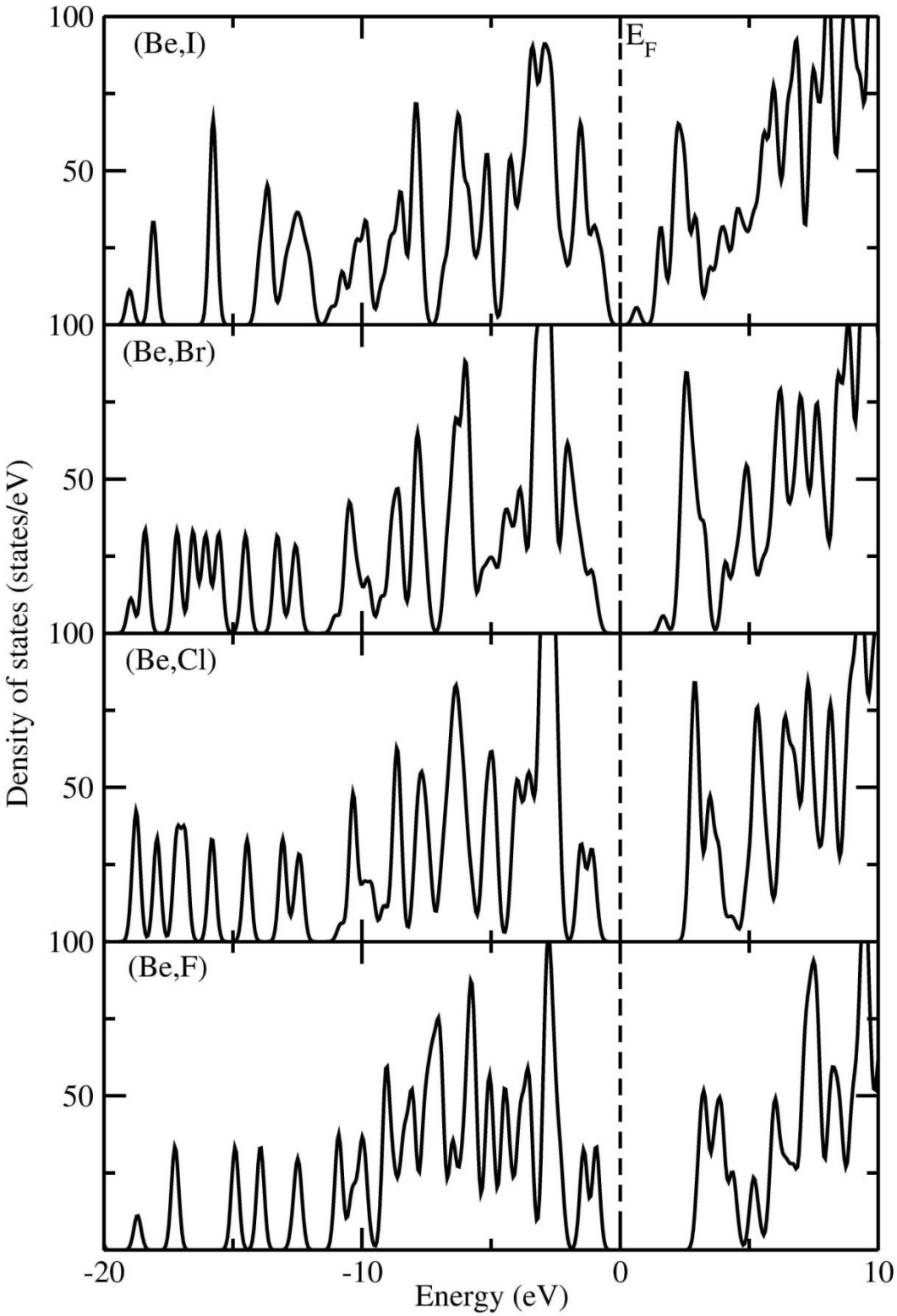

Figure S3. Calculated total density of states (TDOS) for the (Be, Z)-90 °subseries, (Z = F, Cl, Br, I) in the equilibrium cubic structure with *Fm-3m* symmetry (no. 225).

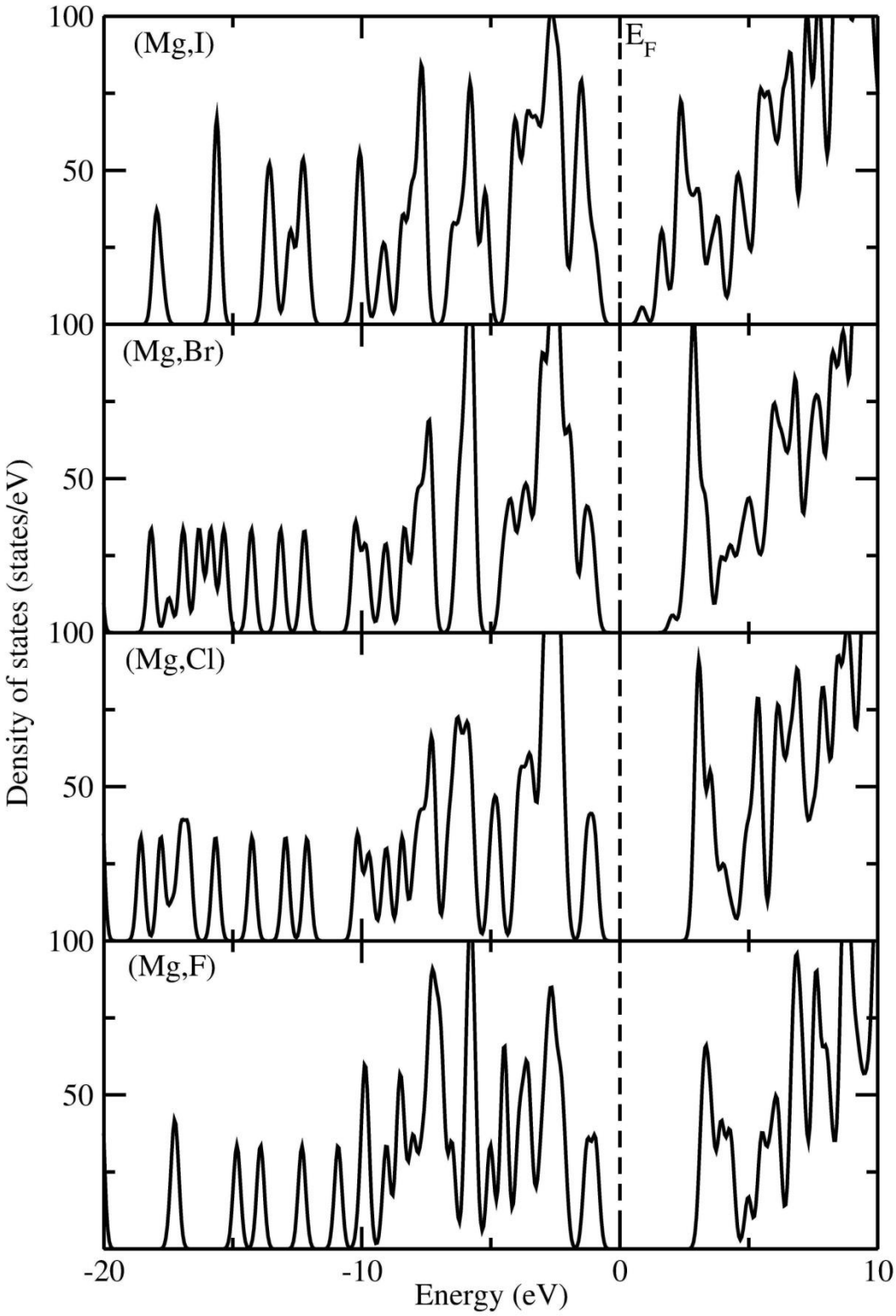

Figure S4. Calculated total density of states (TDOS) for the (Mg, Z)-90 ° subseries, (Z = F, Cl, Br, I) in the equilibrium cubic structure with *Fm-3m* symmetry (no. 225).

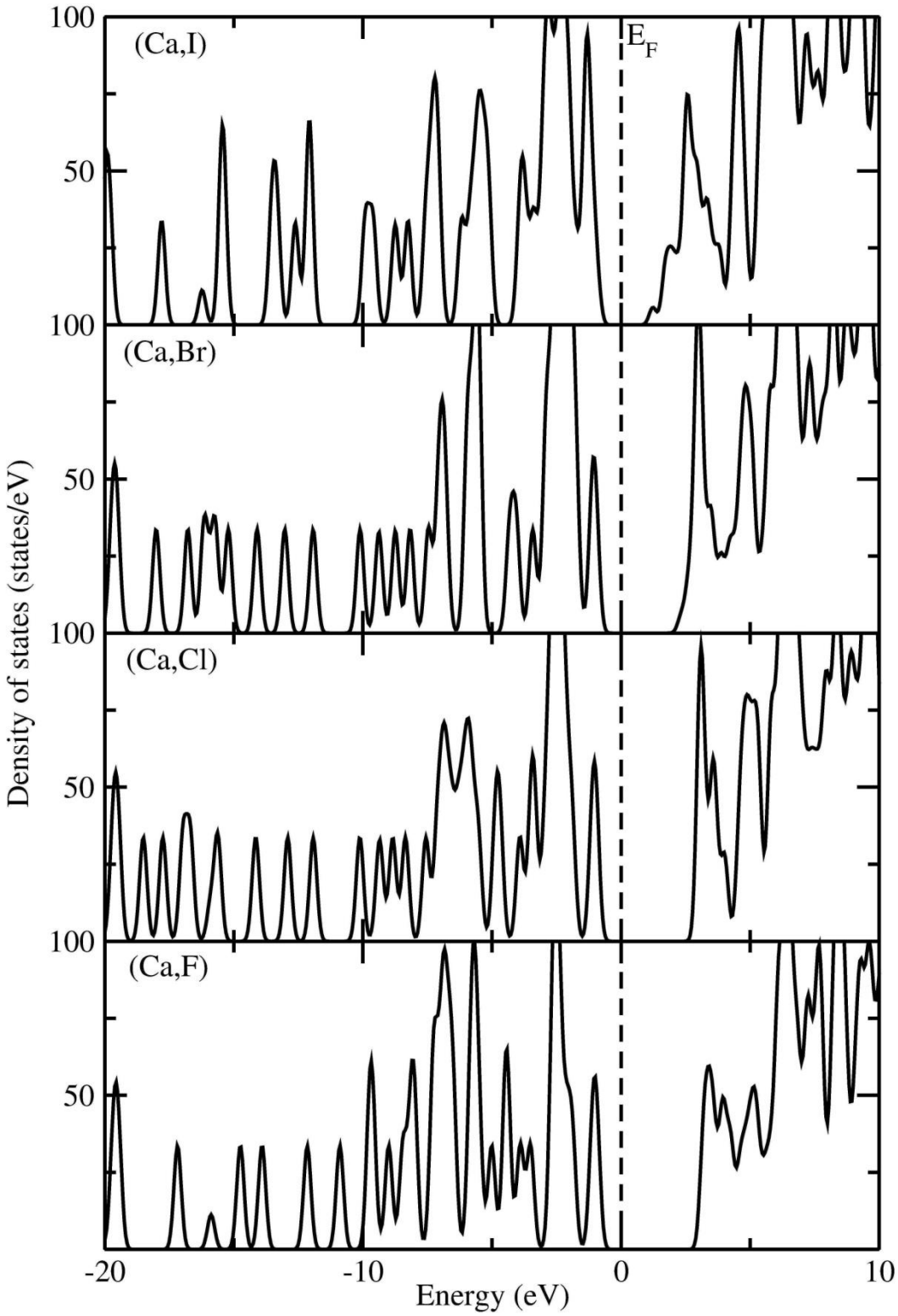

Figure S5. Calculated total density of states (TDOS) for the (Ca, Z)-90 ° subseries, (Z = F, Cl, Br, I) in the equilibrium cubic structure with *Fm-3m* symmetry (no. 225).

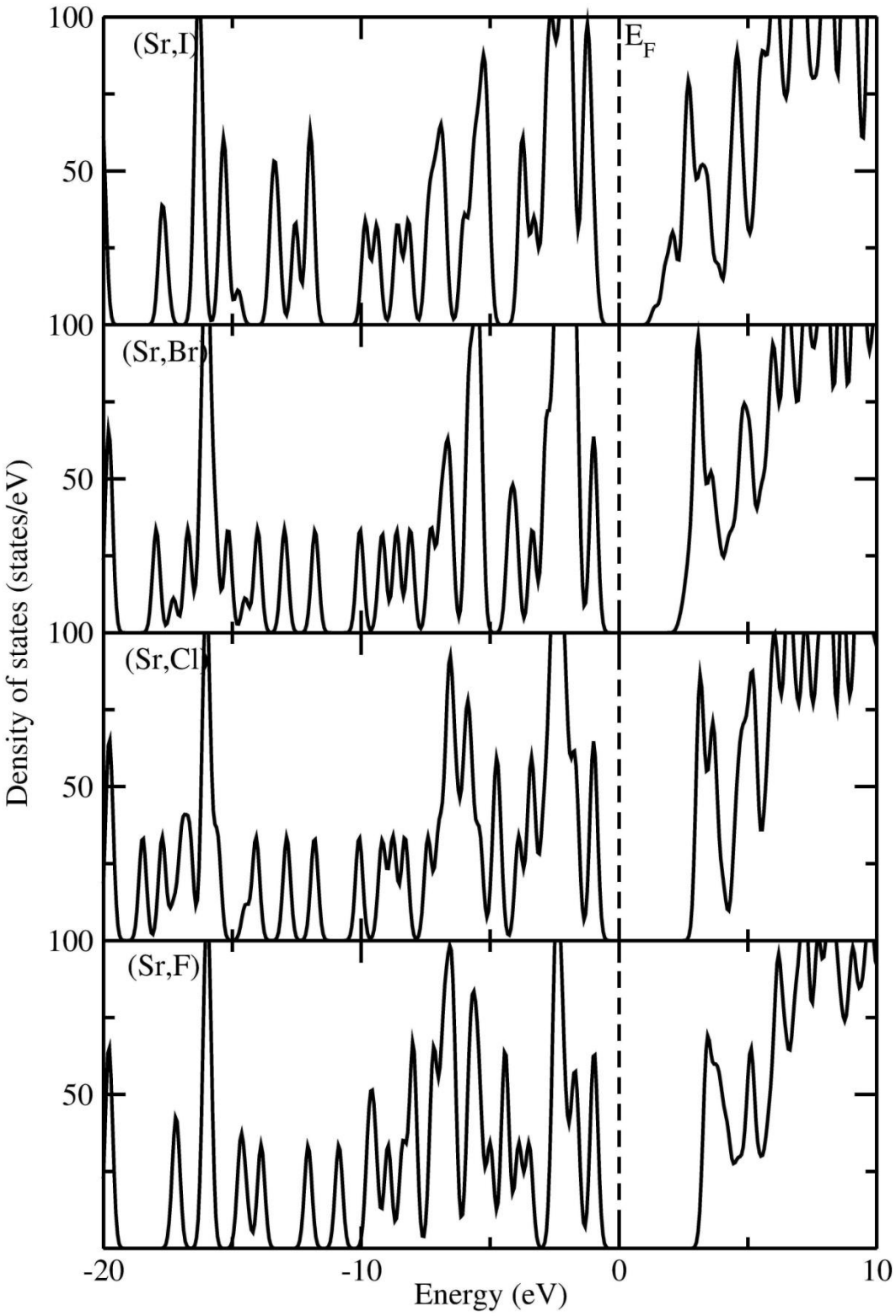

Figure S6. Calculated total density of states (TDOS) for the (Sr, Z)-90 ° subseries, (Z = F, Cl, Br, I) in the equilibrium cubic structure with *Fm-3m* symmetry (no. 225).

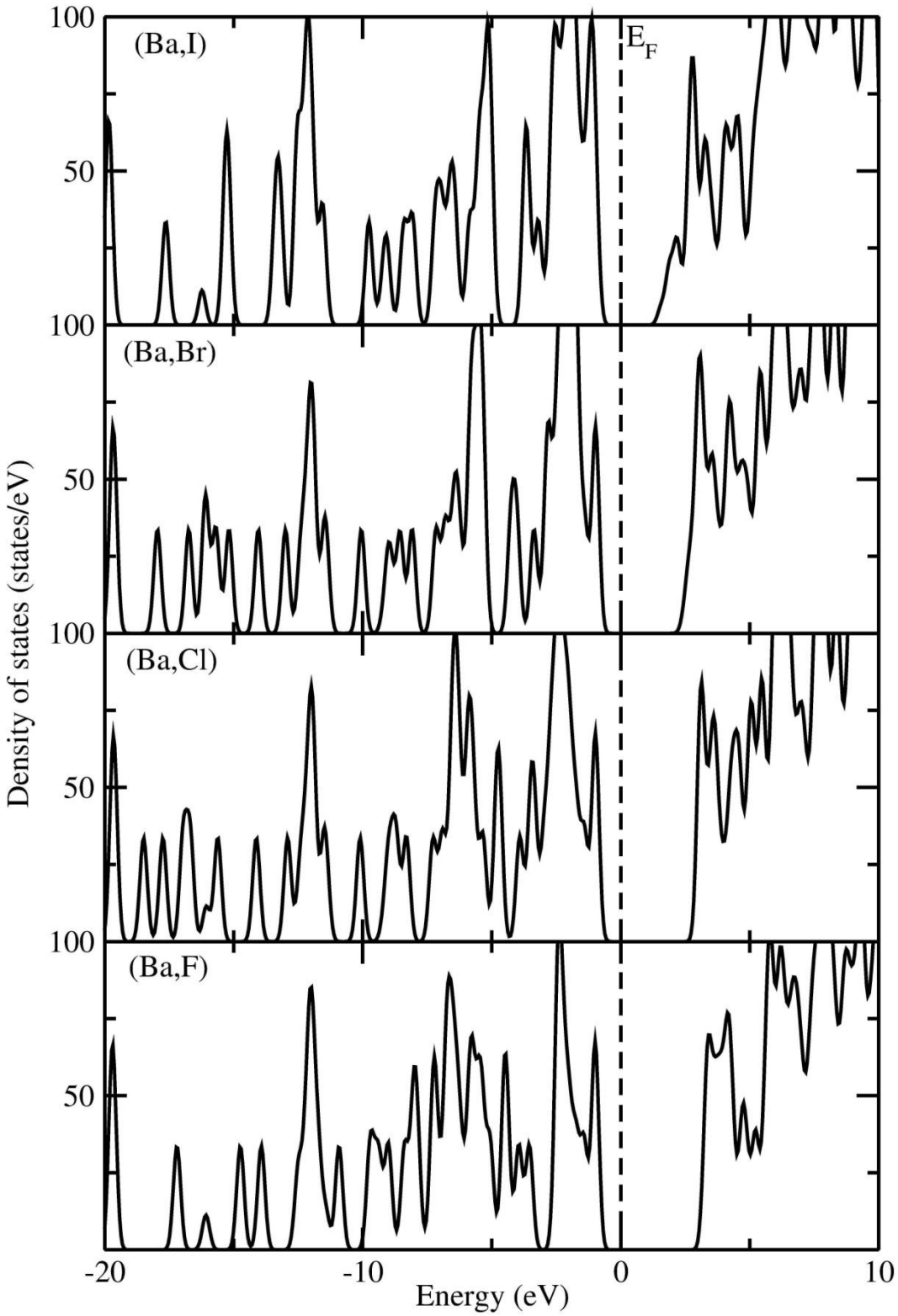

Figure S7. Calculated total density of states (TDOS) for the (Ba, Z)-90 °subseries, (Z = F, Cl, Br, I) in the equilibrium cubic structure with *Fm-3m* symmetry (no. 225).

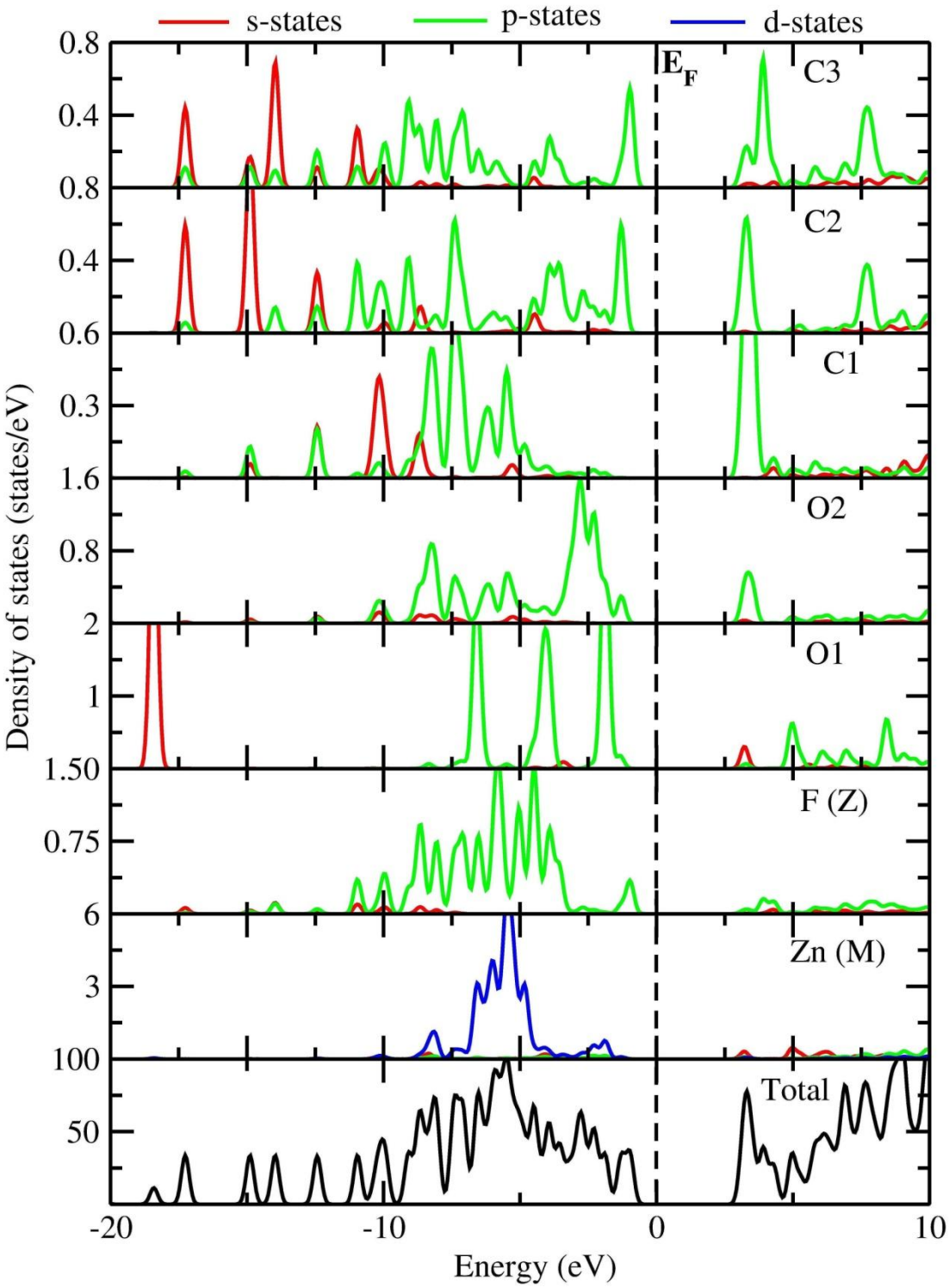

Figure S8. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Zn, F)-90 ° in the cubic *Fm-3m* symmetry (no. 225). Note that the atomic labels of atoms for all PDOS are numbered according to Figure 1. This is also the case for all the PDOS plots.

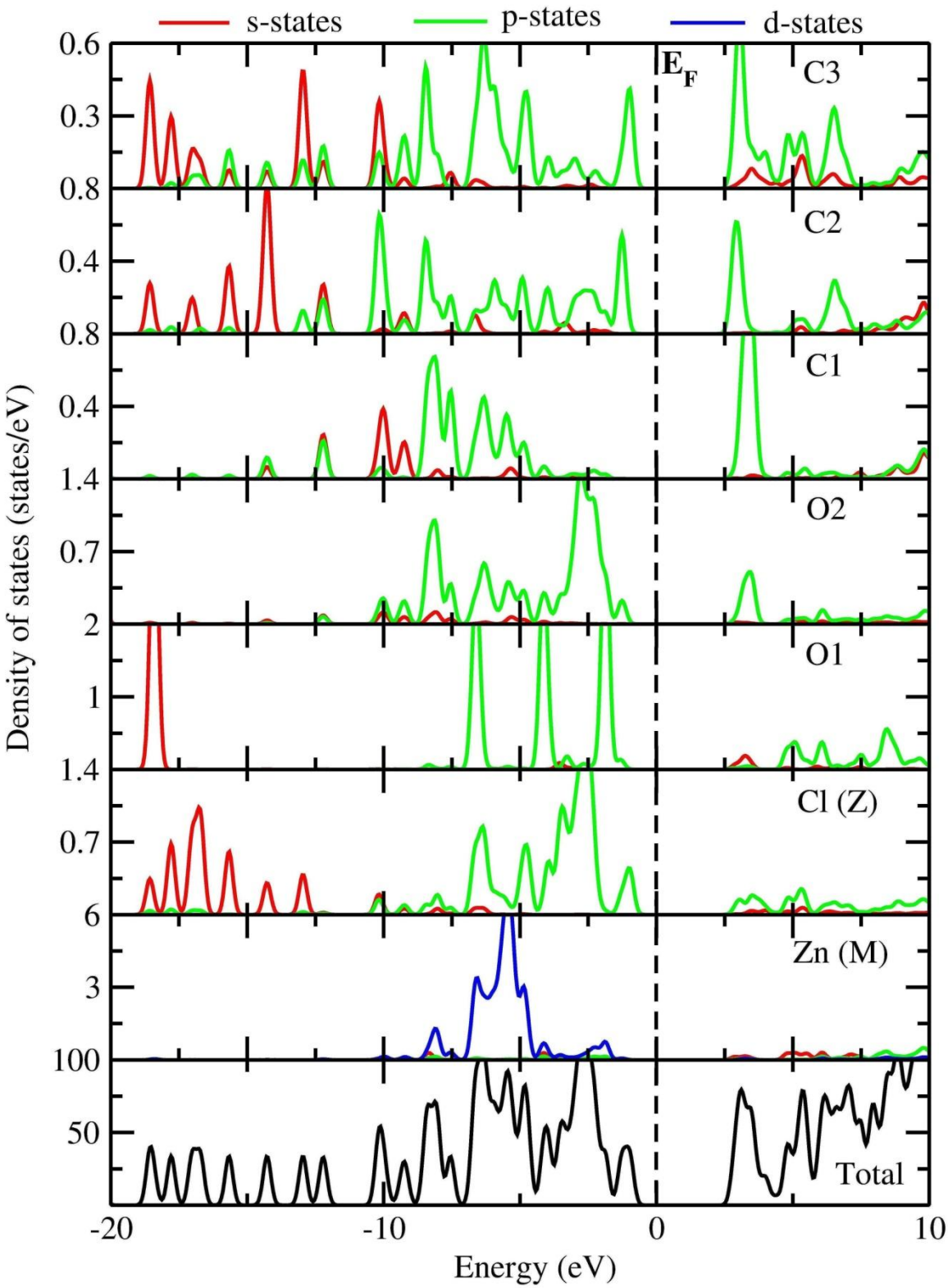

Figure S9. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Zn, Cl)-90 °in the cubic *Fm-3m* symmetry (no. 225).

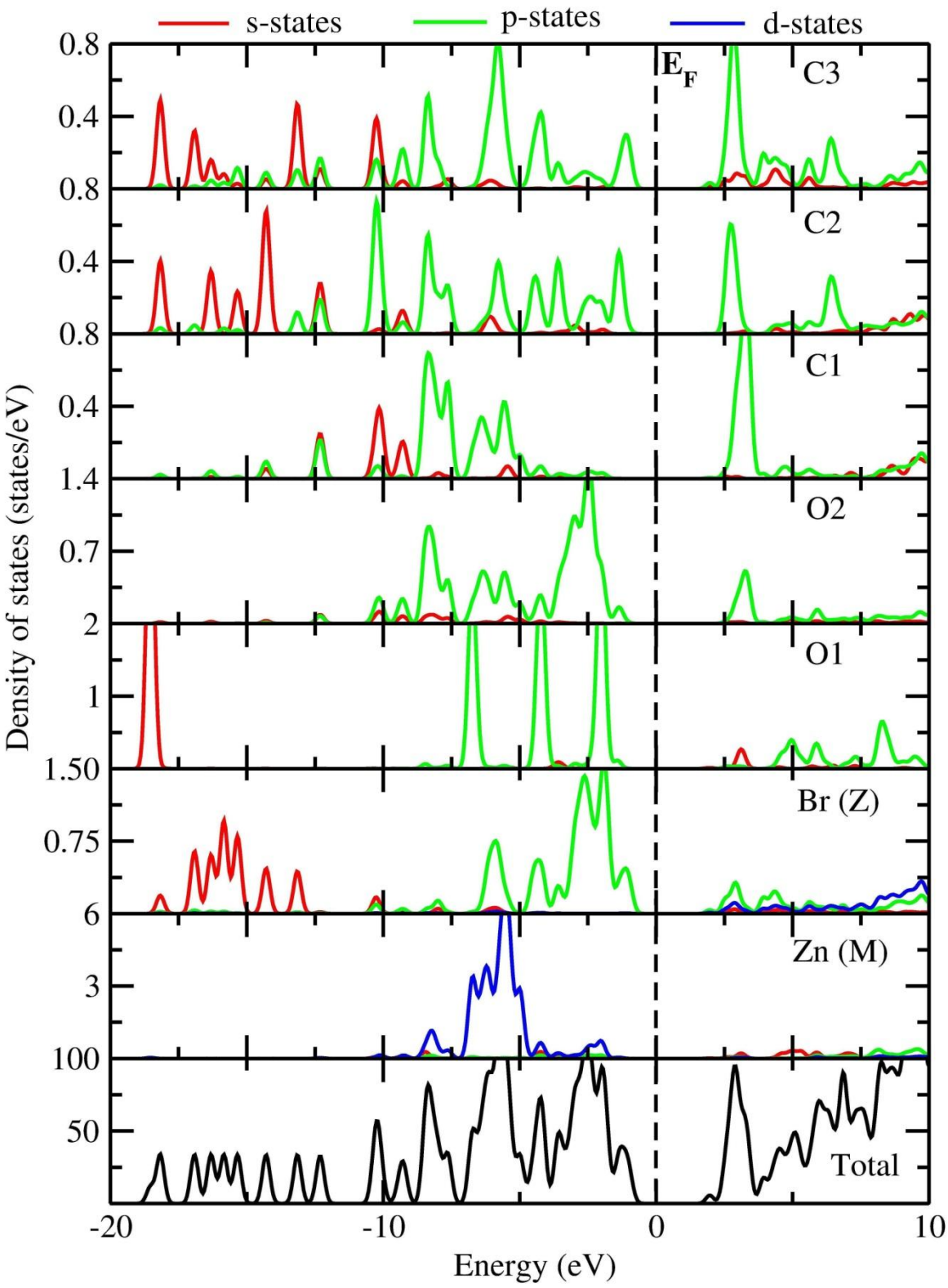

Figure S10. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Zn, Br)-90 °in the cubic *Fm-3m* symmetry (no. 225).

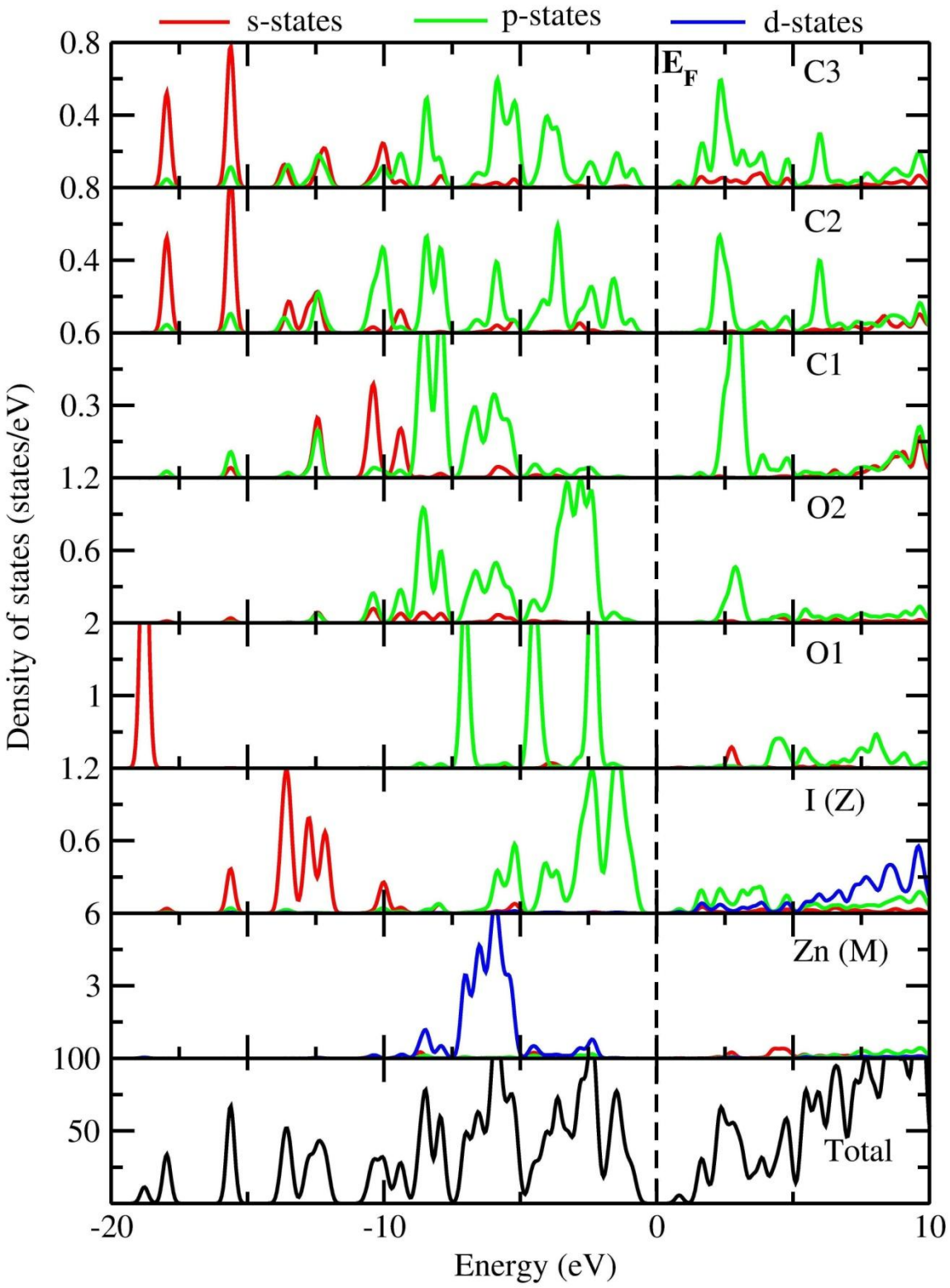

Figure S11. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Zn, I)-90 °in the cubic *Fm-3m* symmetry (no. 225).

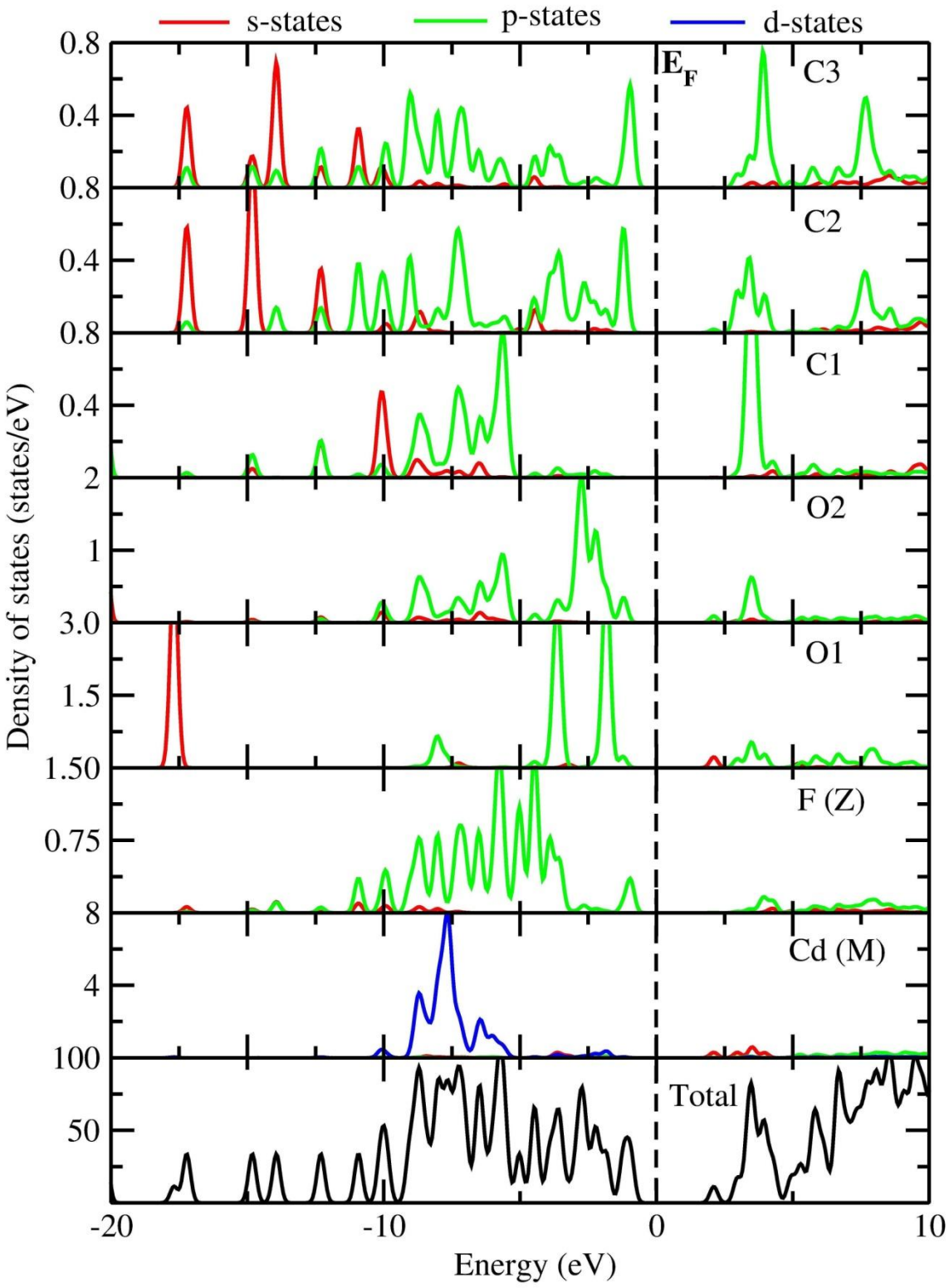

Figure S12. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Cd, F)-90 °in the cubic *Fm-3m* symmetry (no. 225).

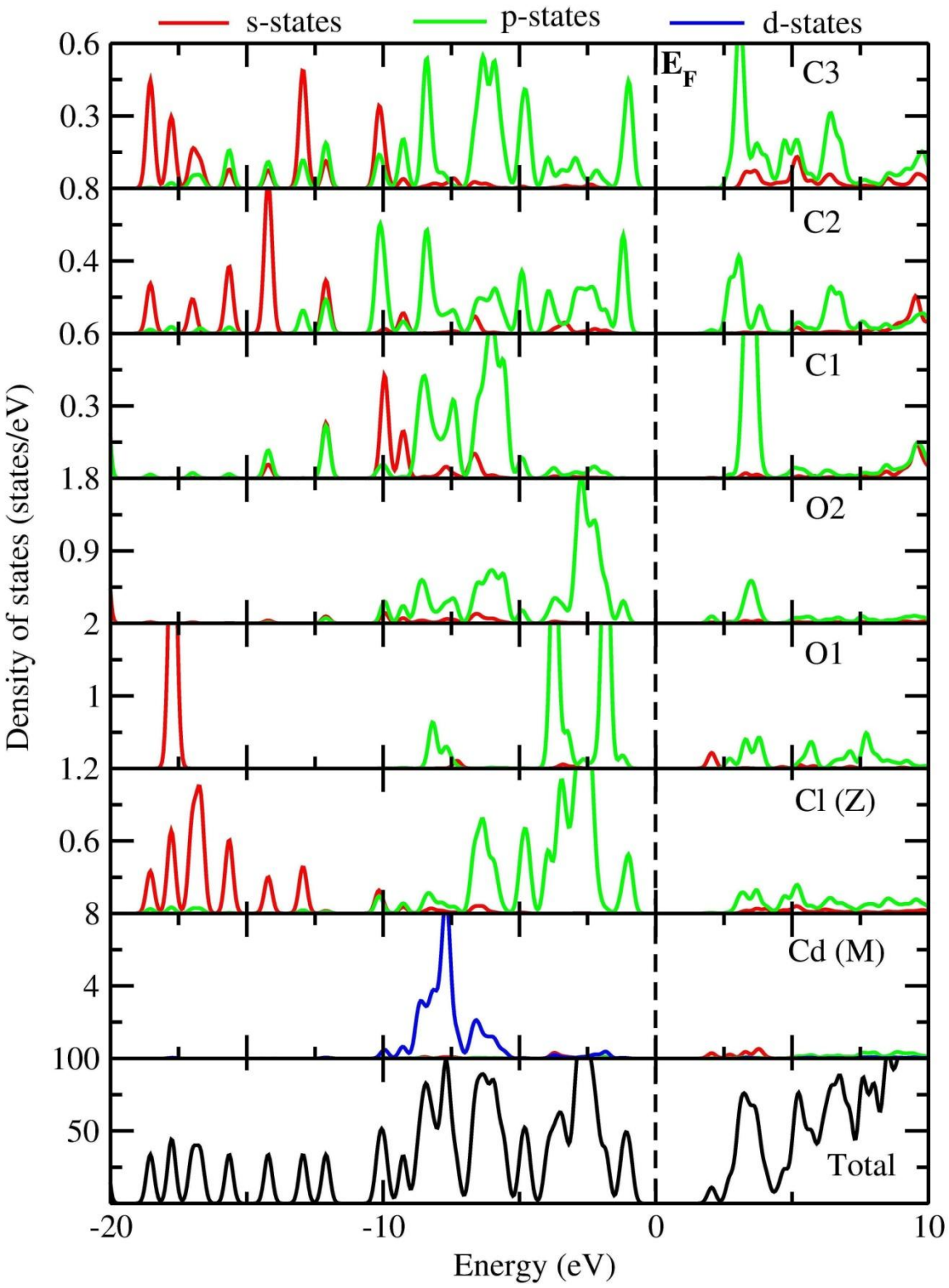

Figure S13. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Cd, Cl)-90 °in the cubic *Fm-3m* symmetry (no. 225).

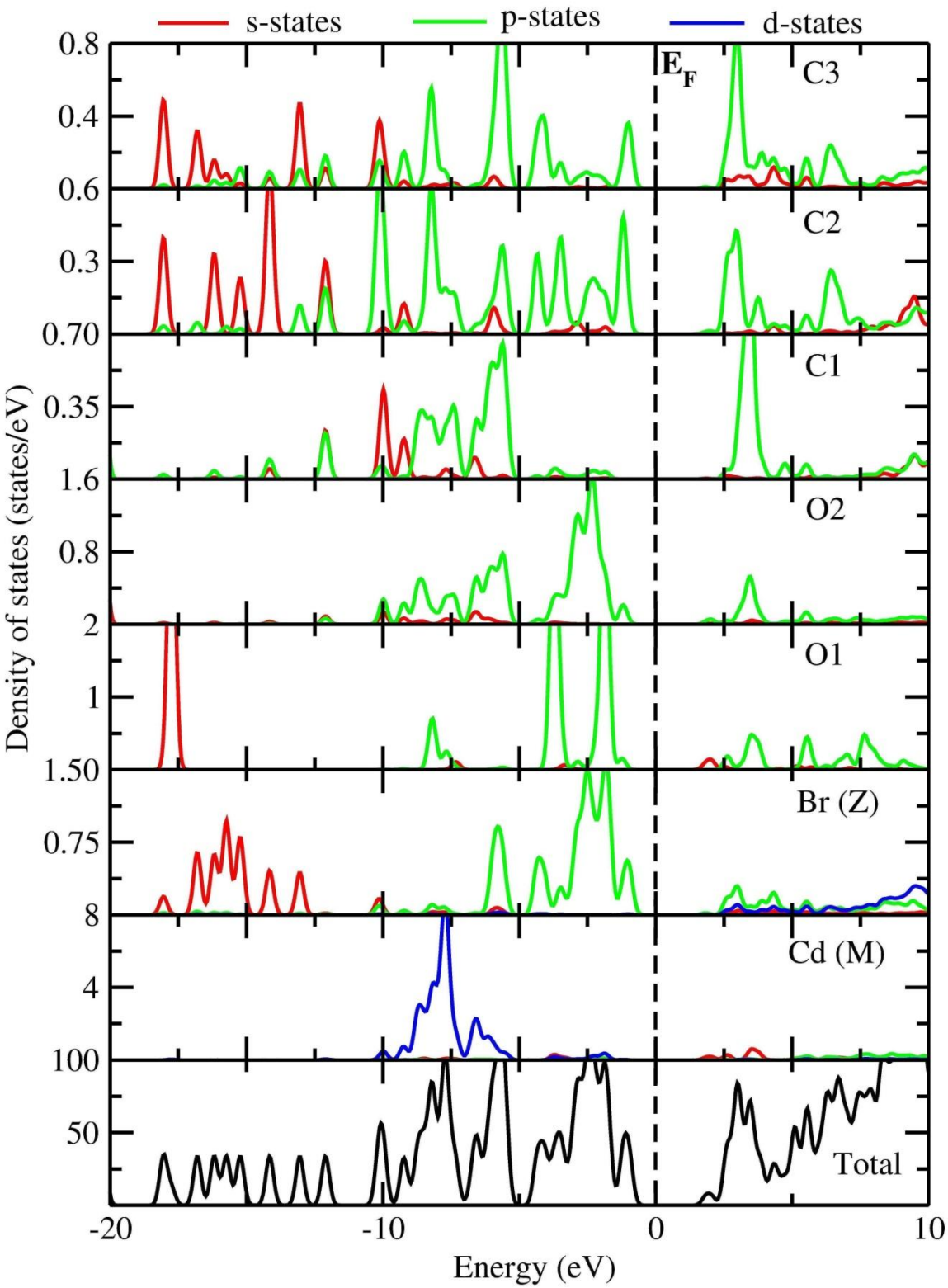

Figure S14. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Cd, Br)-90 °in the cubic *Fm-3m* symmetry (no. 225).

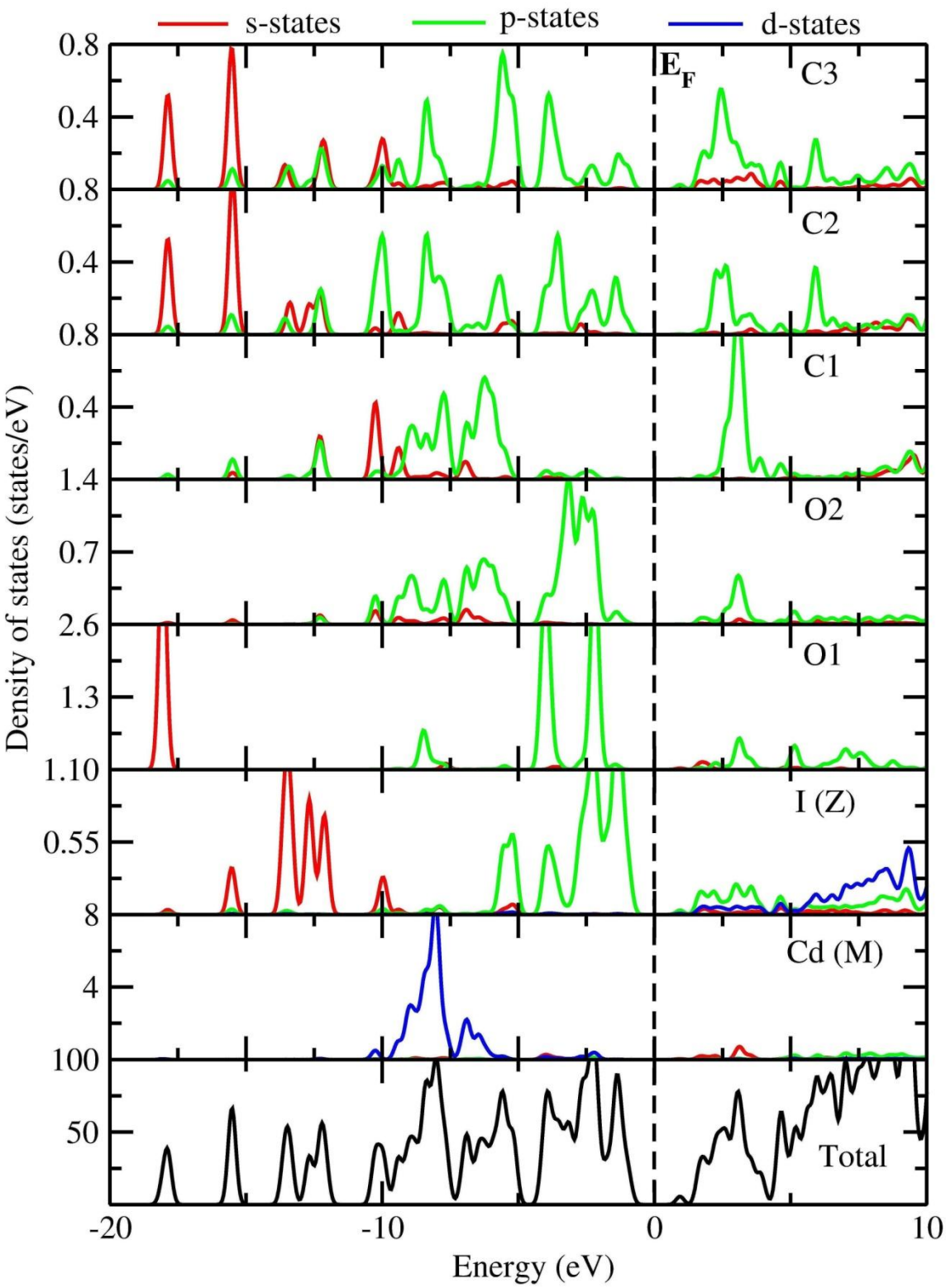

Figure S15. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Cd, I)-90 °in the cubic *Fm-3m* symmetry (no. 225).

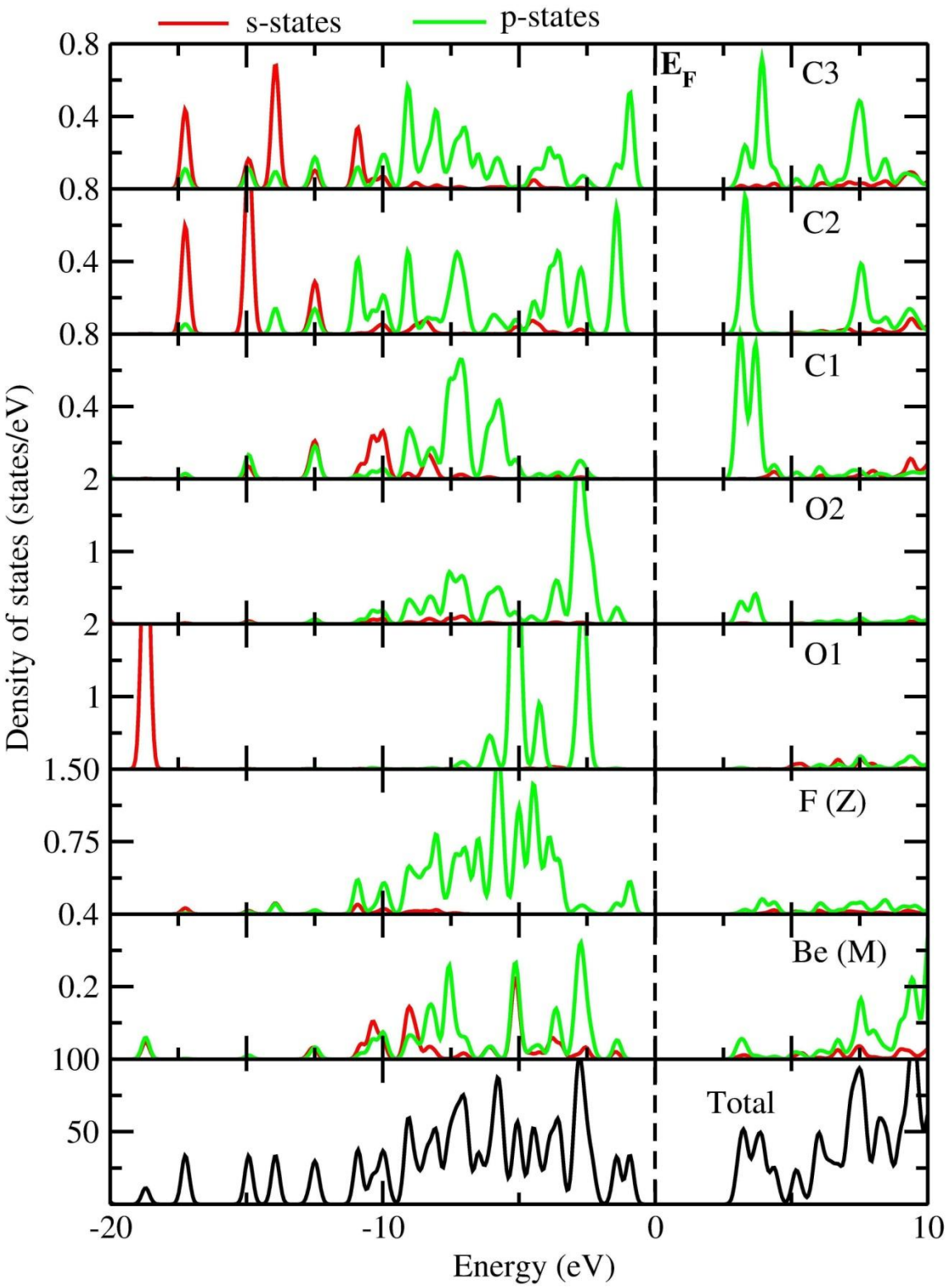

Figure S16. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Be, F)-90 °in the cubic *Fm-3m* symmetry (no. 225).

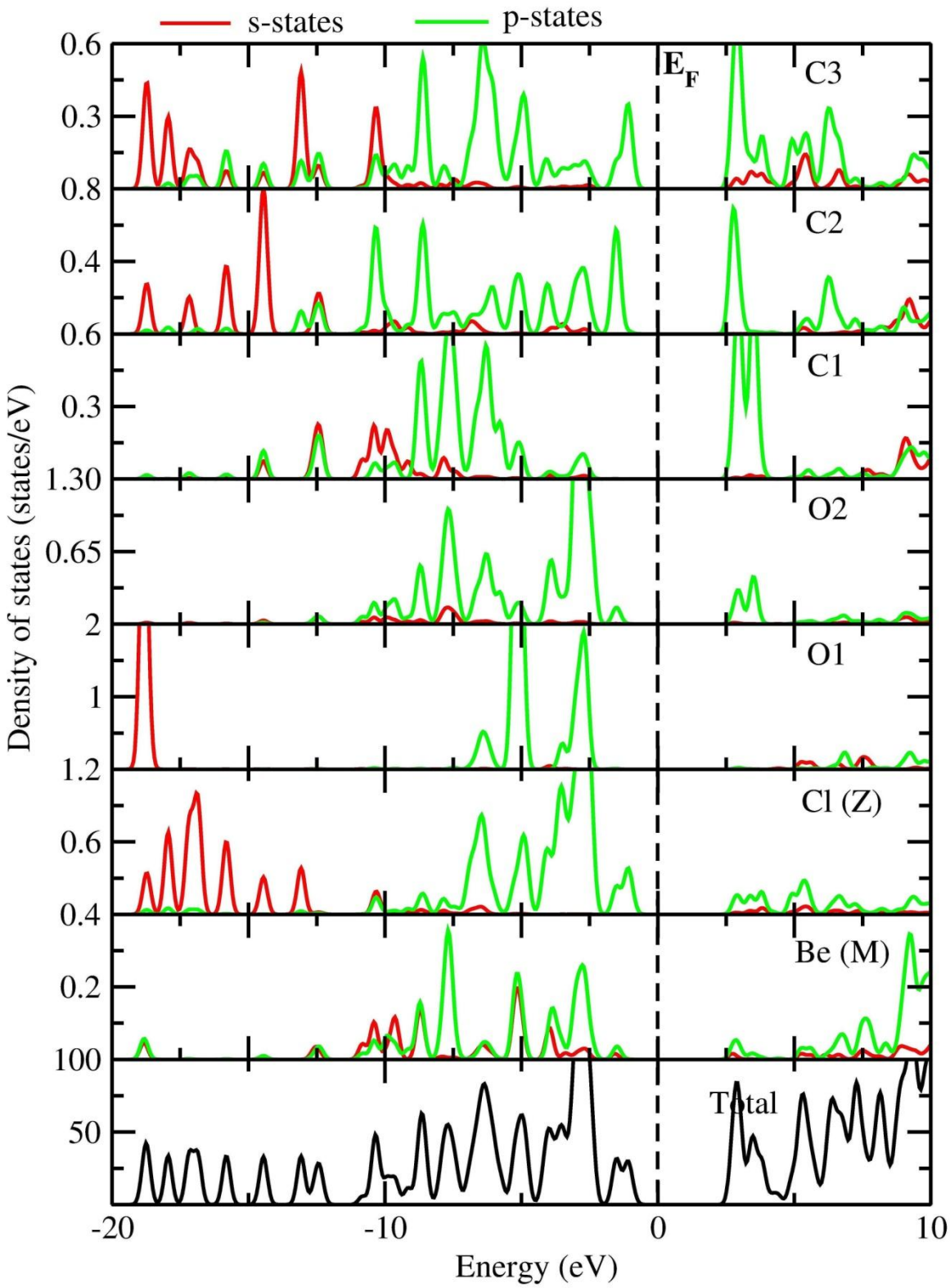

Figure S17. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Be, Cl)-90 °in the cubic *Fm-3m* symmetry (no. 225).

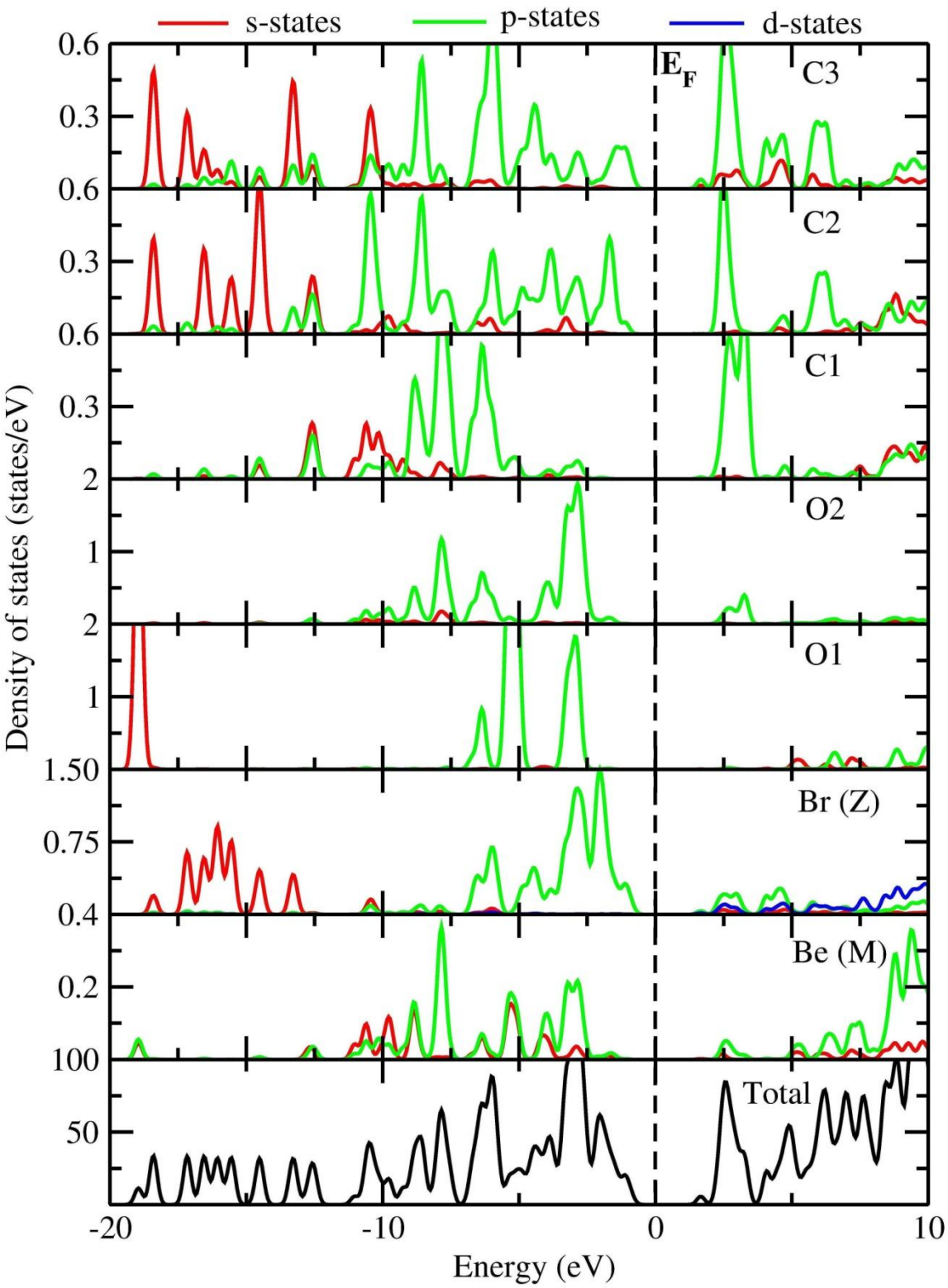

Figure S18. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Be, Br)-90 °in the cubic *Fm-3m* symmetry (no. 225).

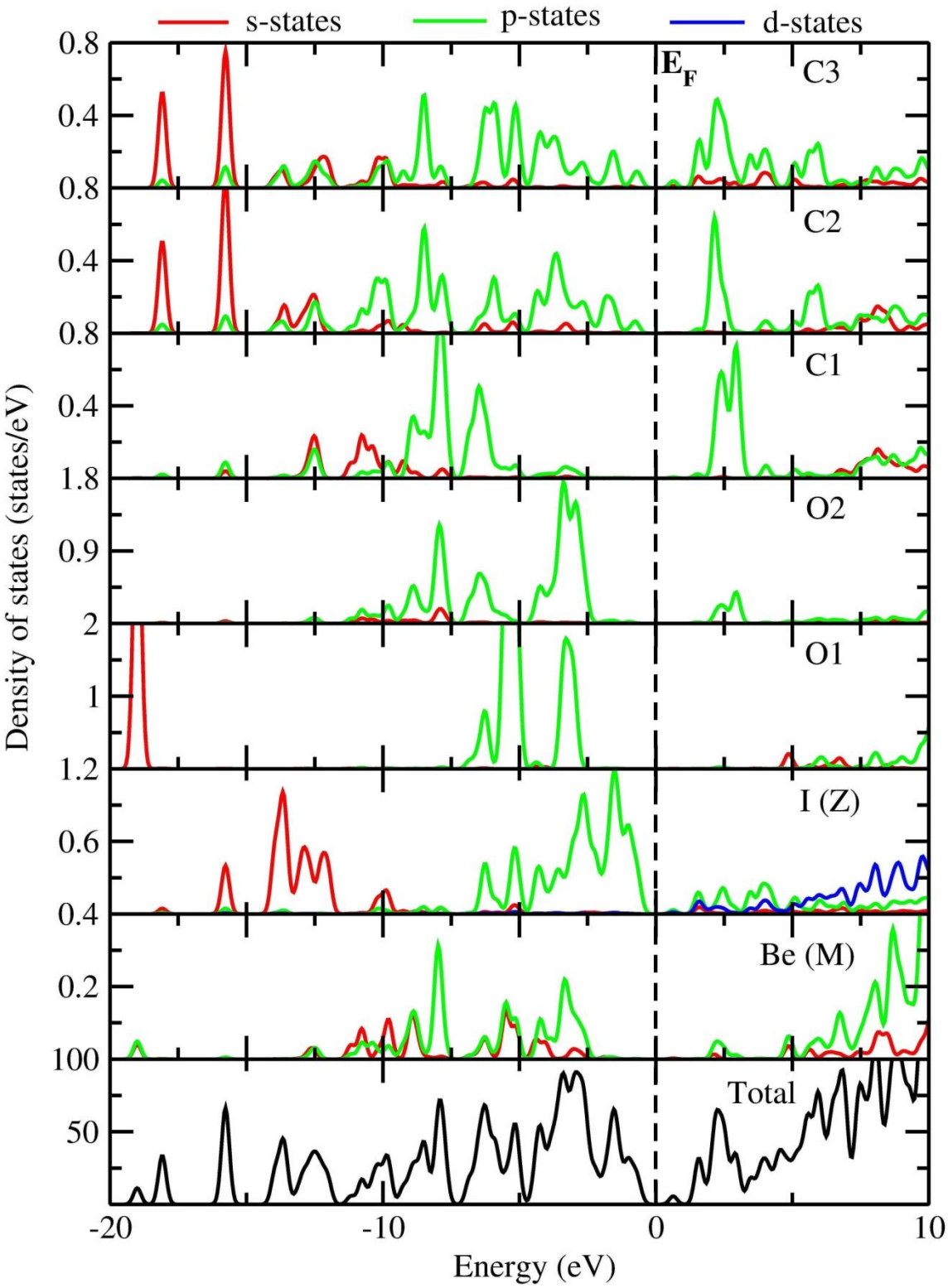

Figure S19. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Be, I)-90 °in the cubic *Fm-3m* symmetry (no. 225).

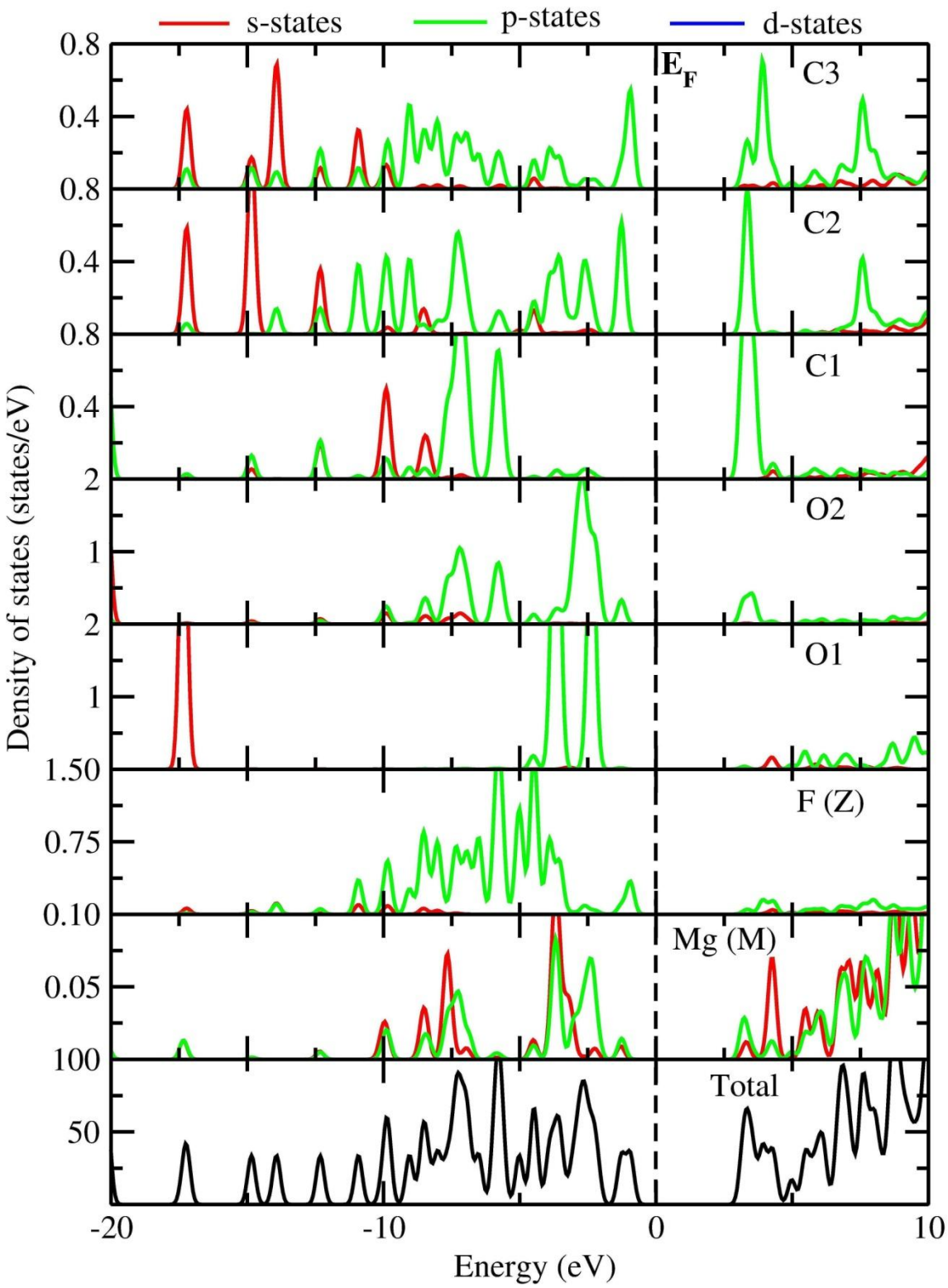

Figure S20. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Mg, F)-90 °in the cubic *Fm-3m* symmetry (no. 225).

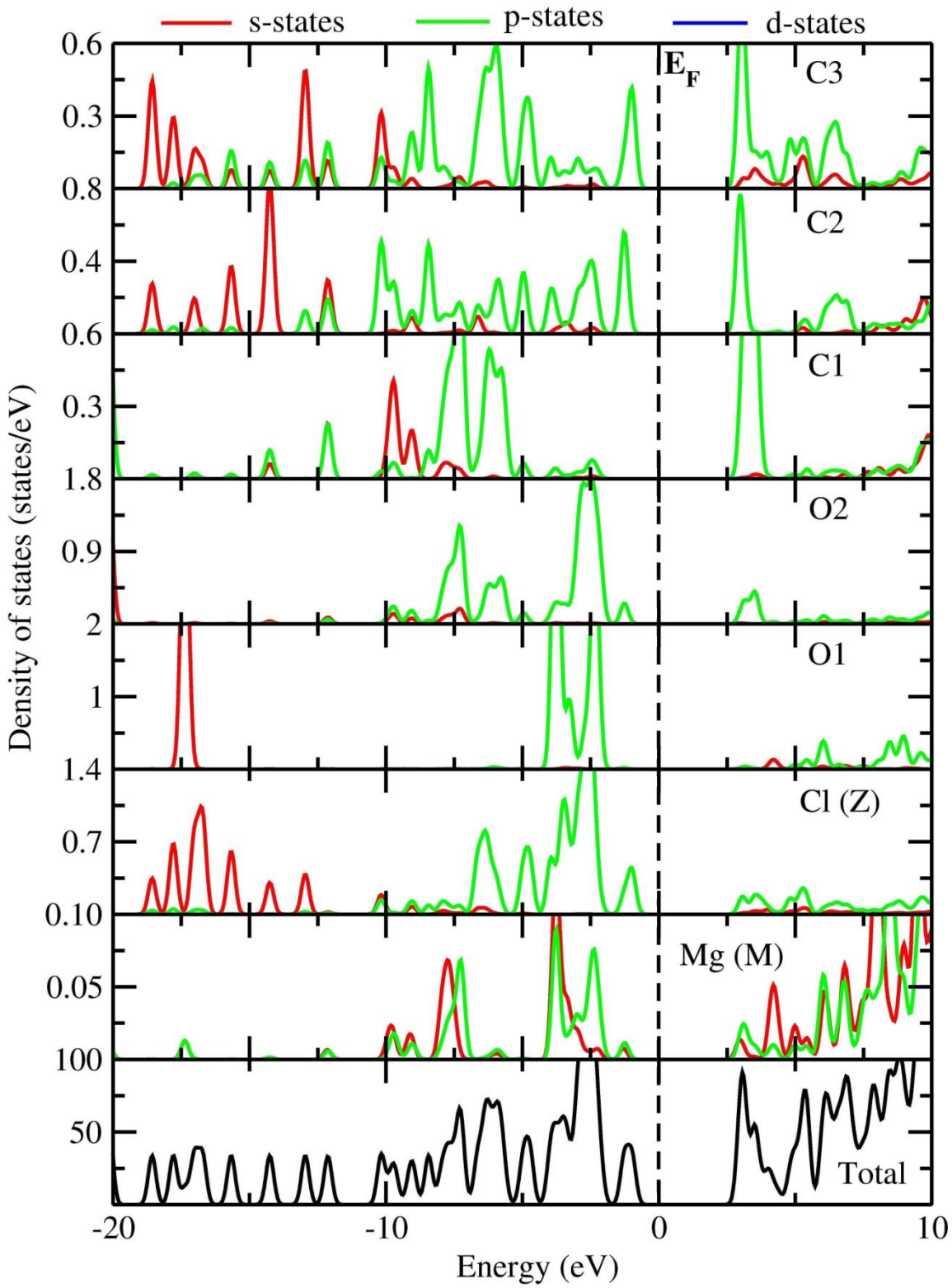

Figure S21. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Mg, Cl)-90 °in the cubic *Fm-3m* symmetry (no. 225).

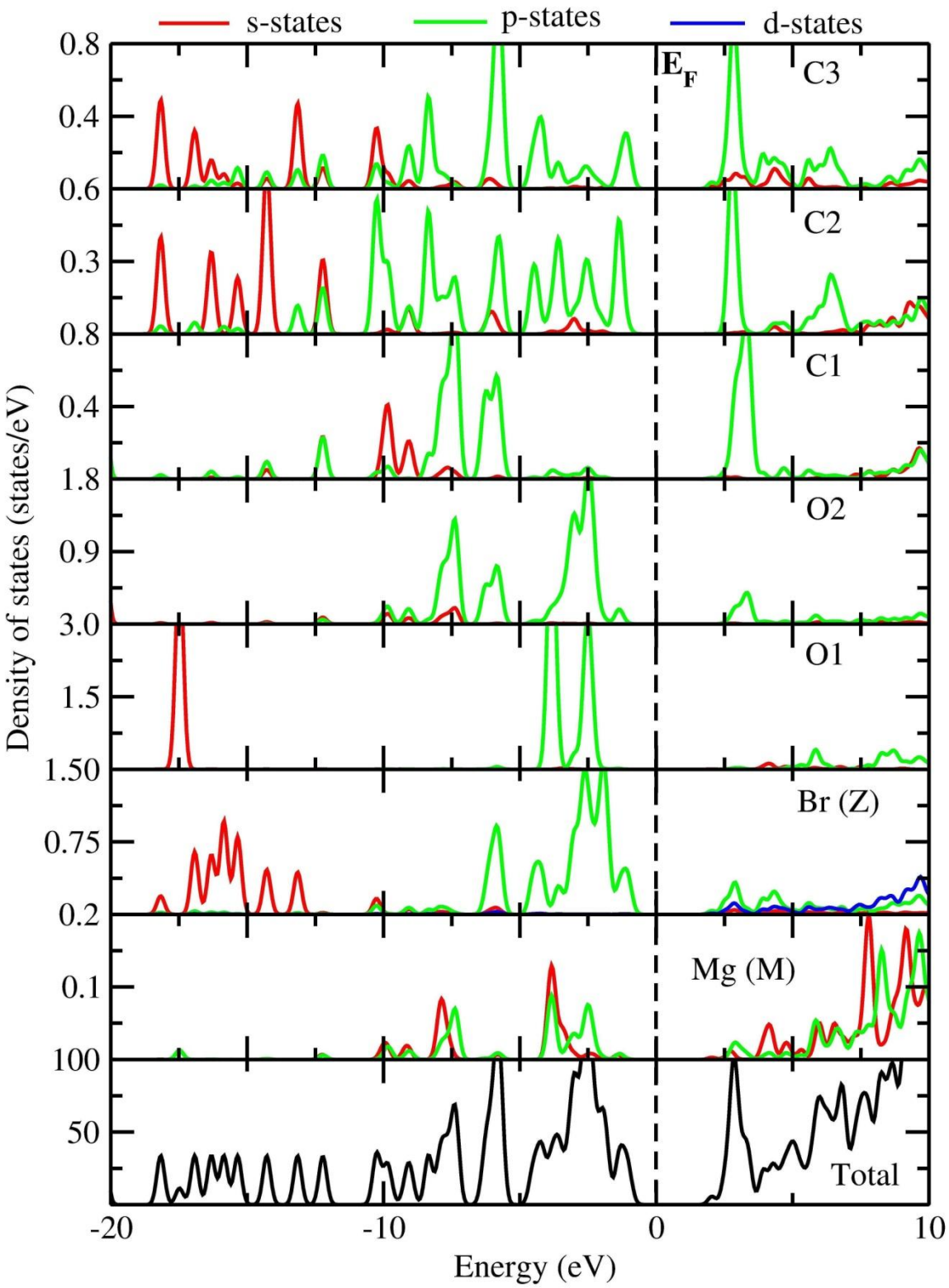

Figure S22. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Mg, Br)-90 °in the cubic *Fm-3m* symmetry (no. 225).

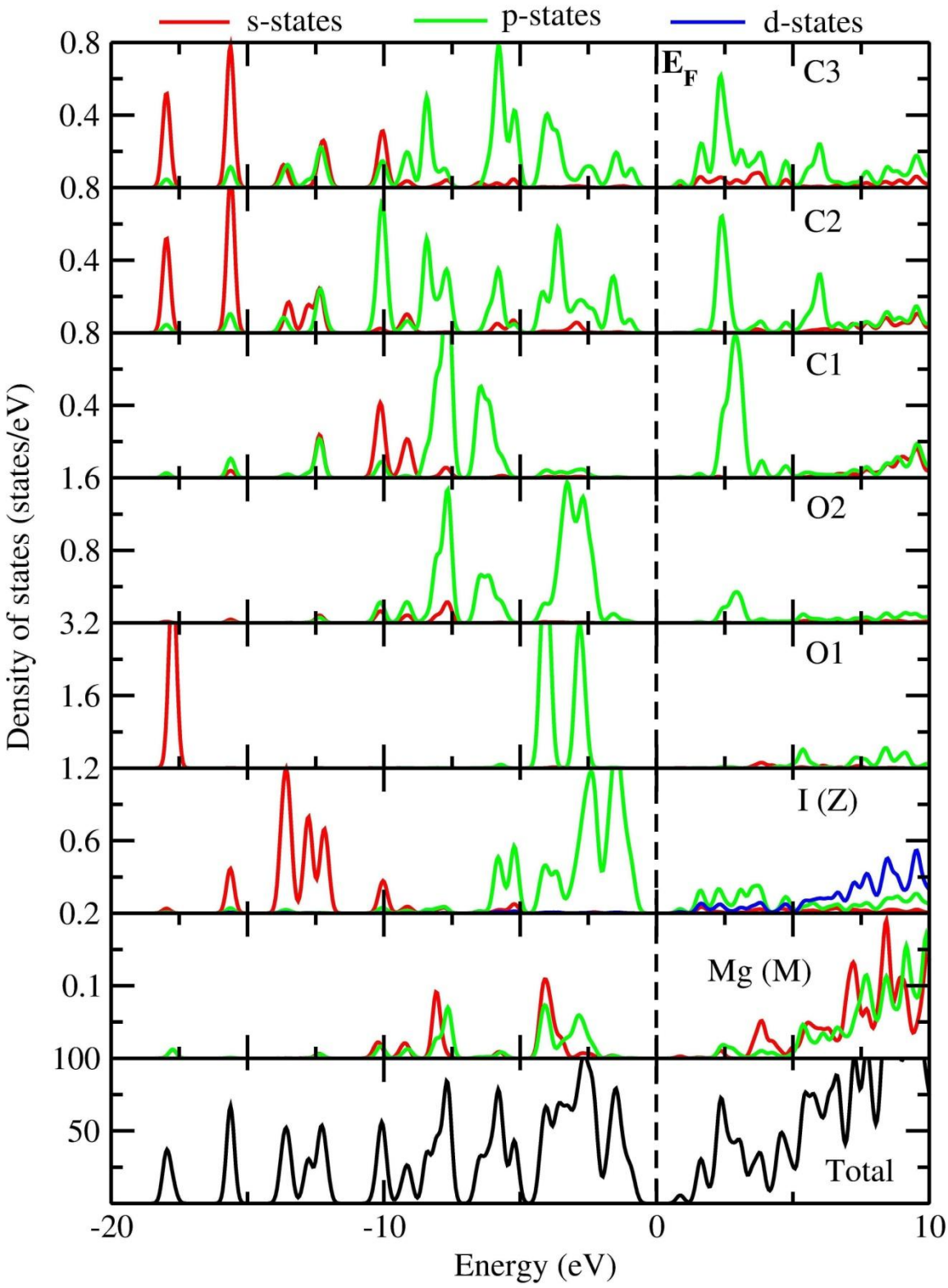

Figure S23. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Mg, I)-90 °in the cubic *Fm-3m* symmetry (no. 225).

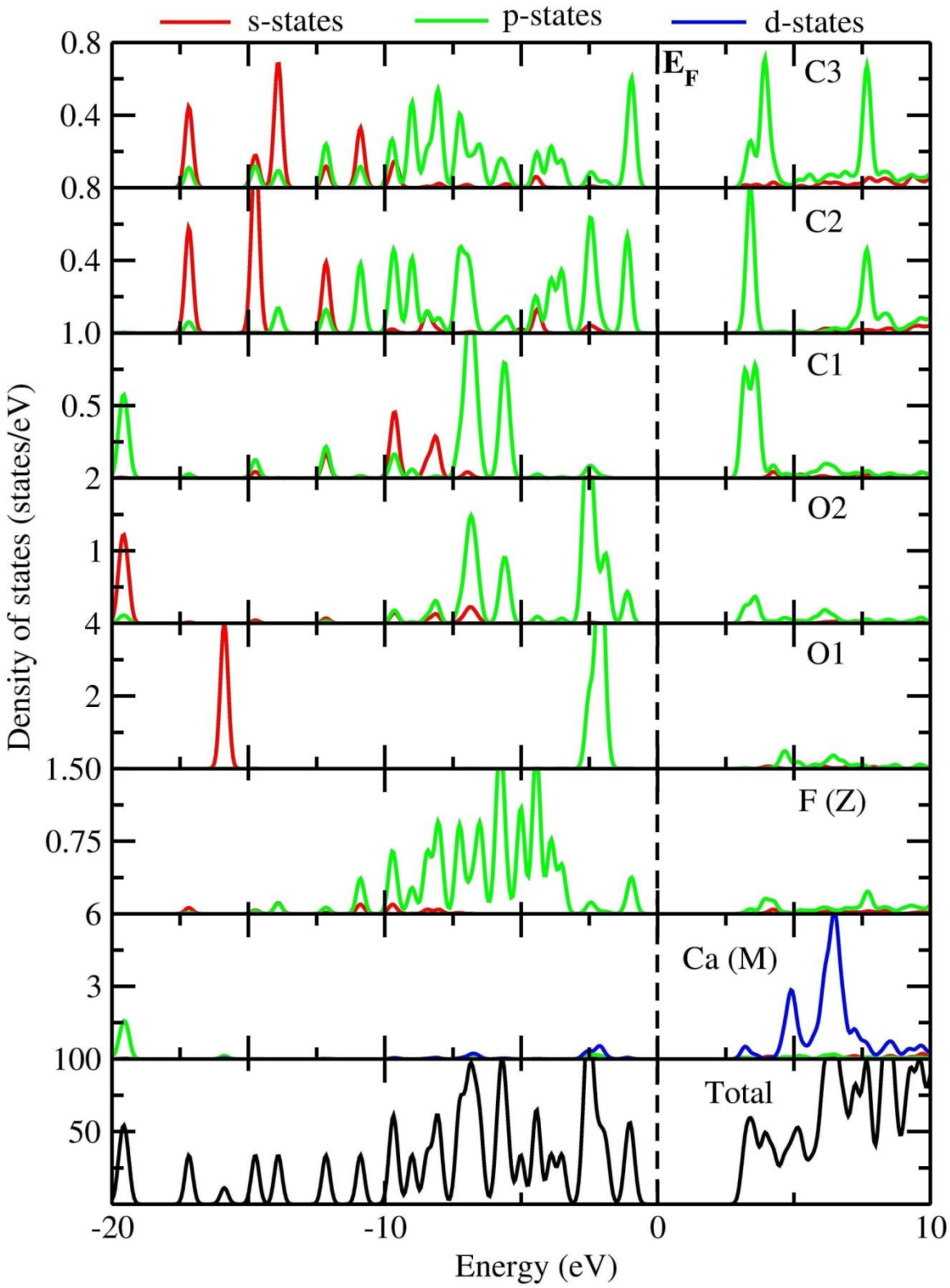

Figure S24. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Ca, F)-90 °in the cubic *Fm-3m* symmetry (no. 225).

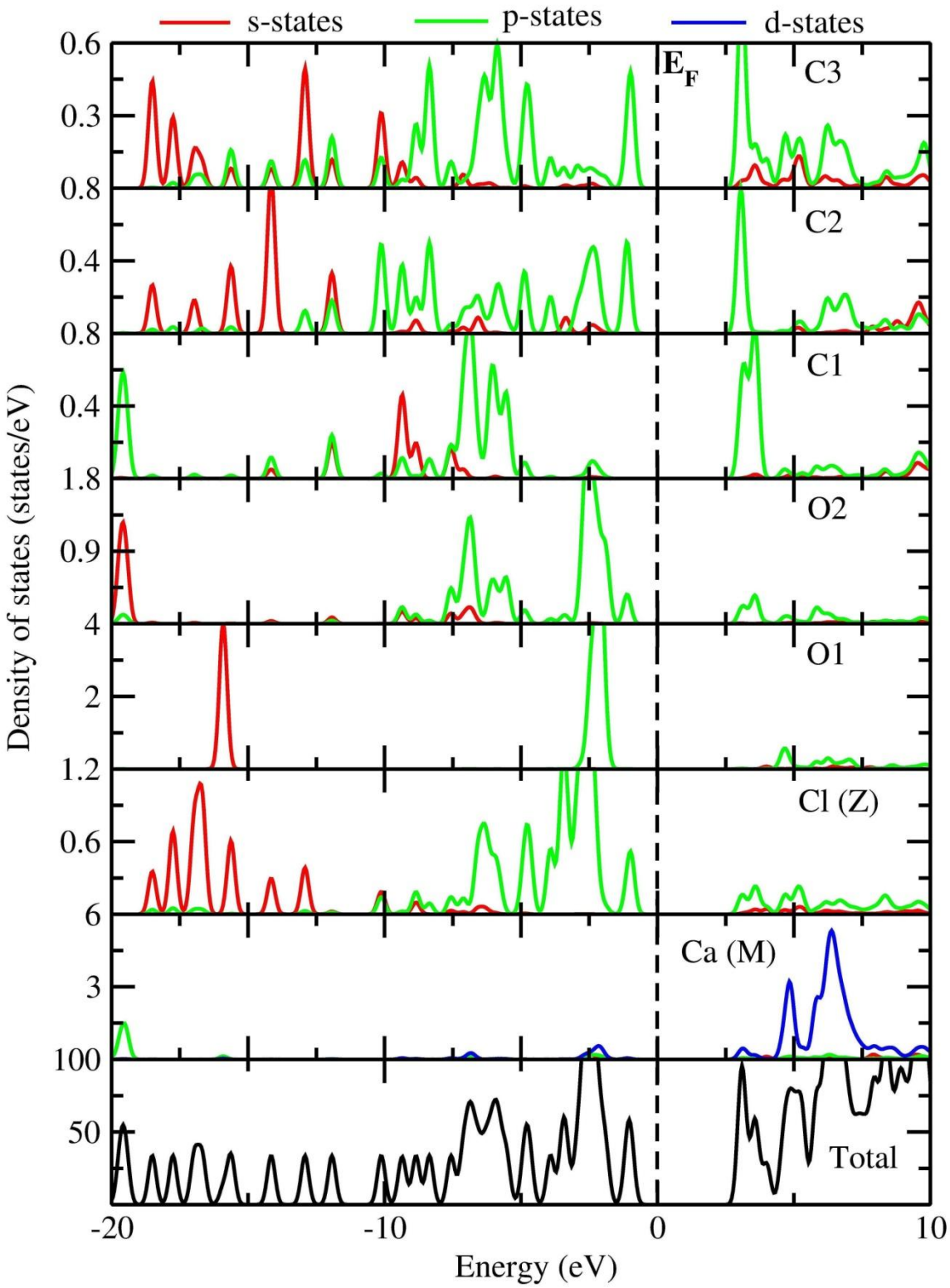

Figure S25. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Ca, Cl)-90 °in the cubic *Fm-3m* symmetry (no. 225).

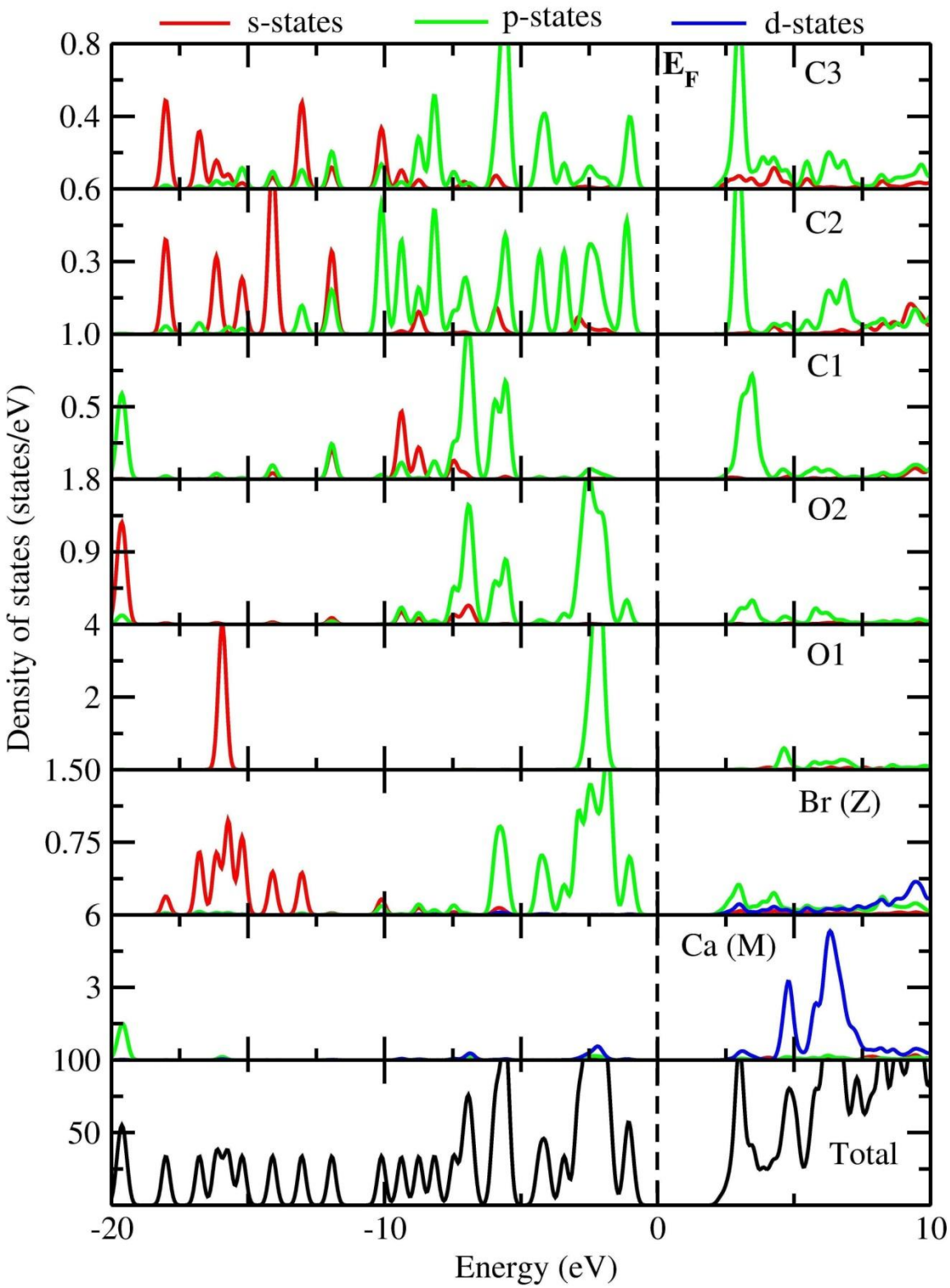

Figure S26. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Ca, Br)-90 °in the cubic *Fm-3m* symmetry (no. 225).

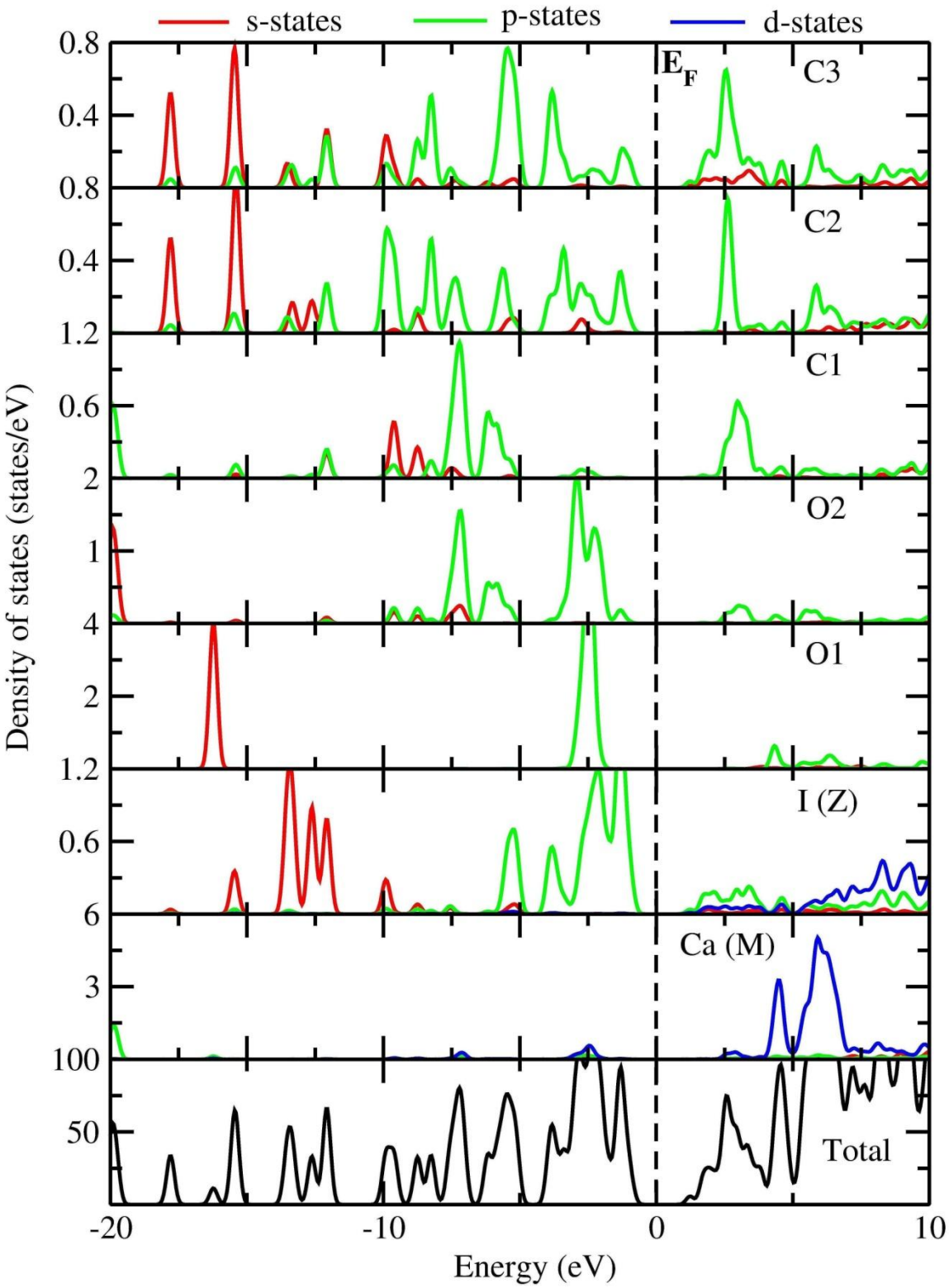

Figure S27. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Ca, I)-90 °in the cubic *Fm-3m* symmetry (no. 225).

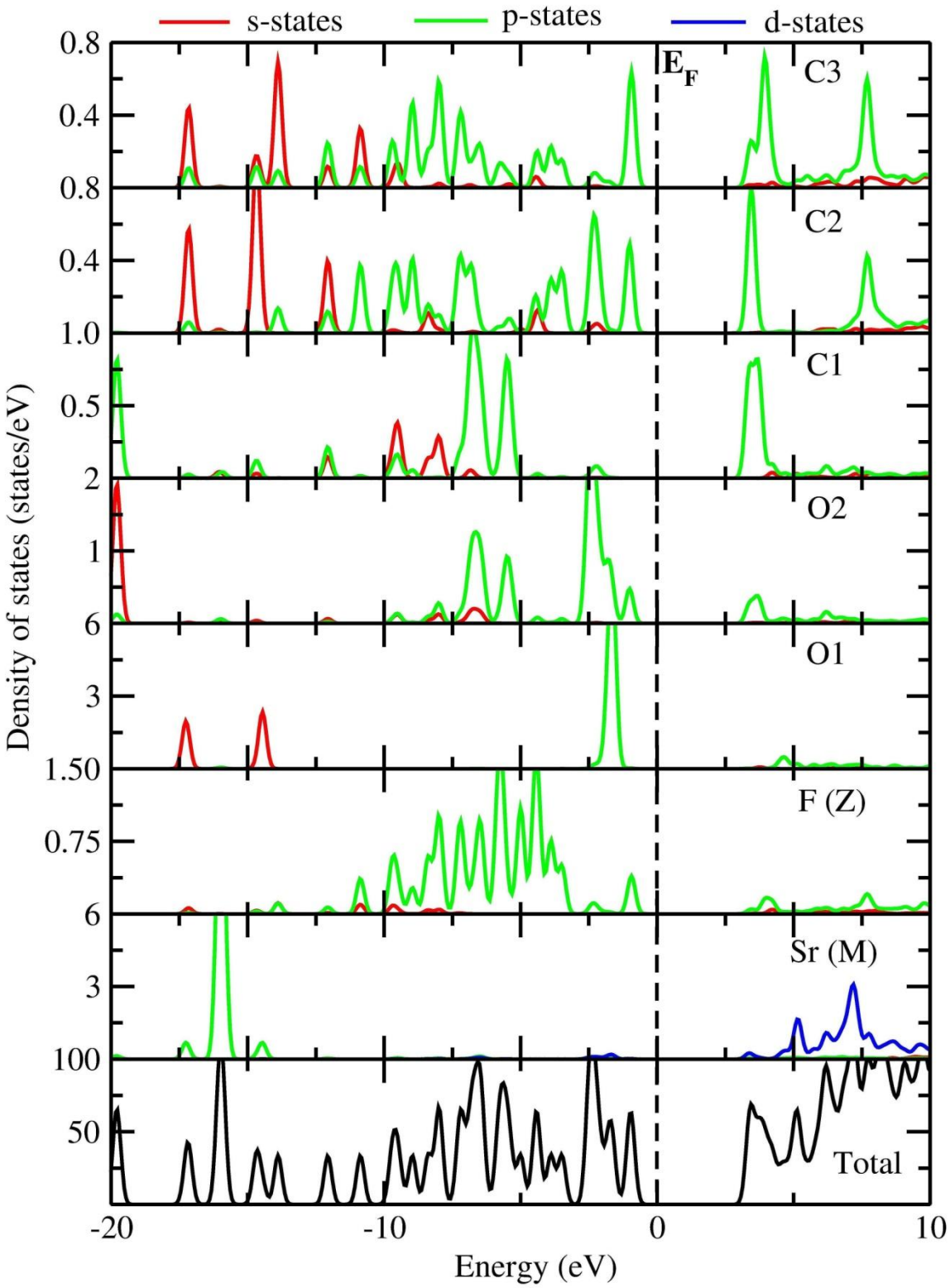

Figure S28. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Sr, F)-90 °in the cubic *Fm-3m* symmetry (no. 225).

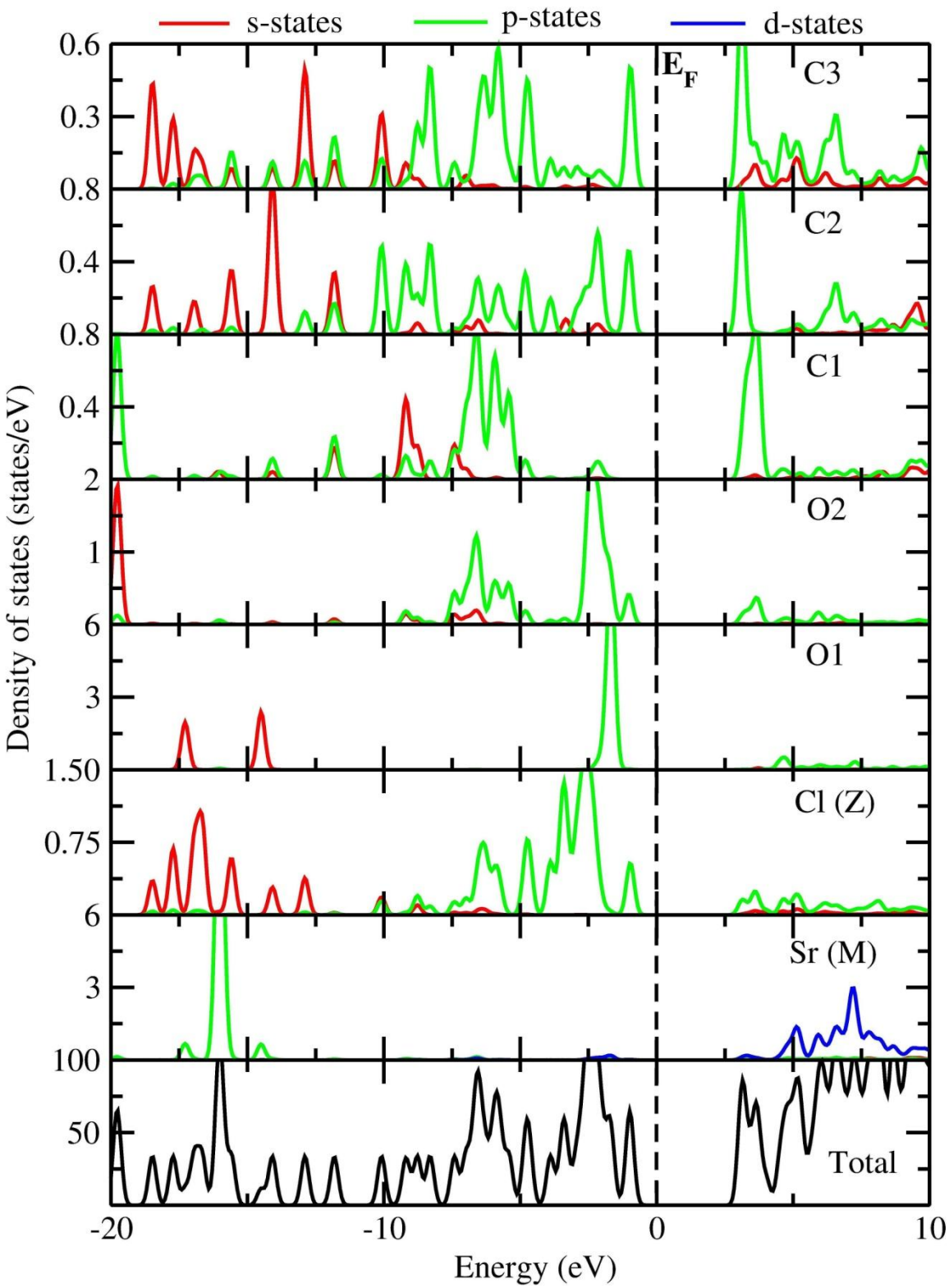

Figure S29. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Sr, Cl)-90 °in the cubic *Fm-3m* symmetry (no. 225).

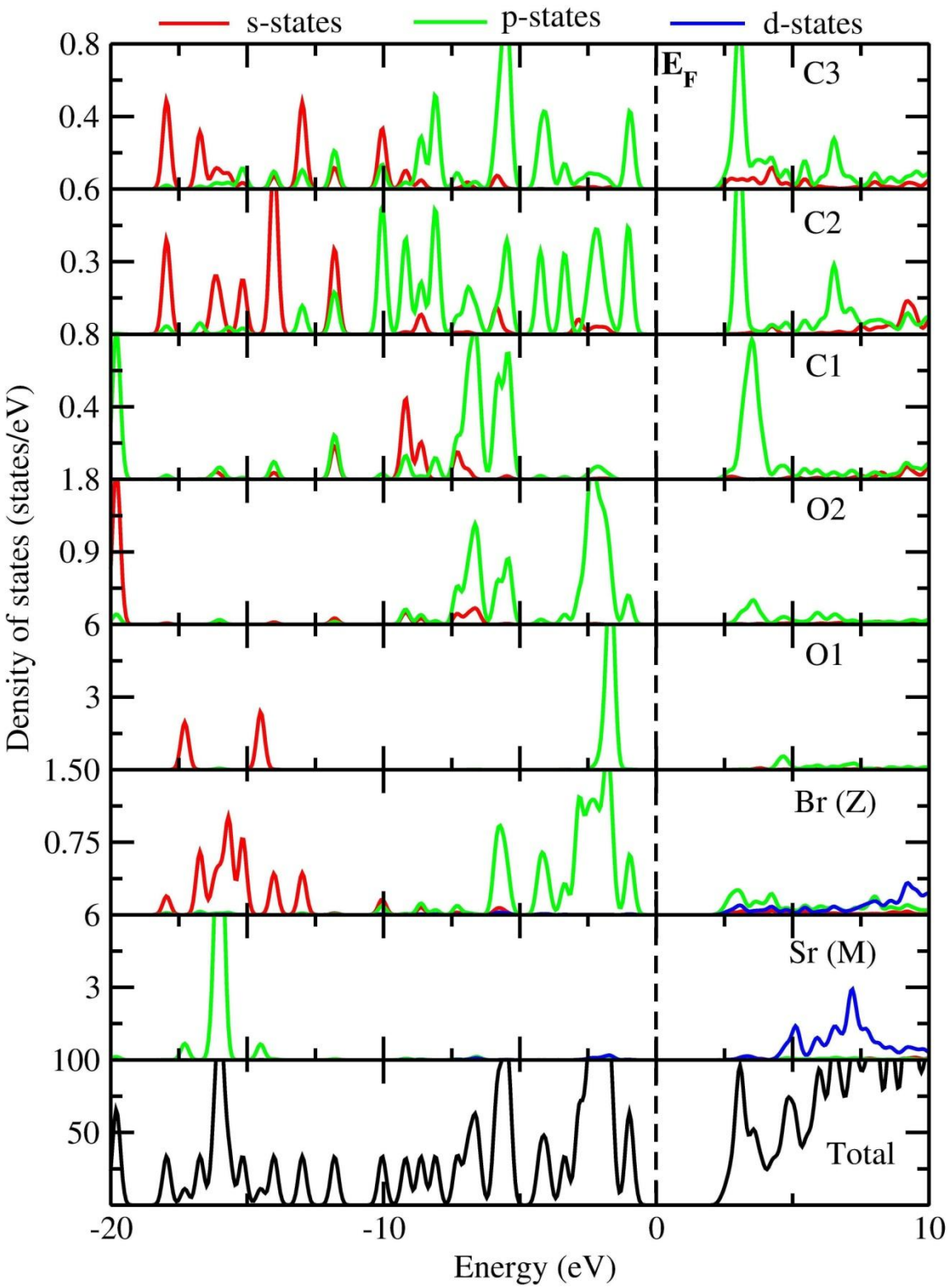

Figure S30. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Sr, Br)-90 °in the cubic *Fm-3m* symmetry (no. 225).

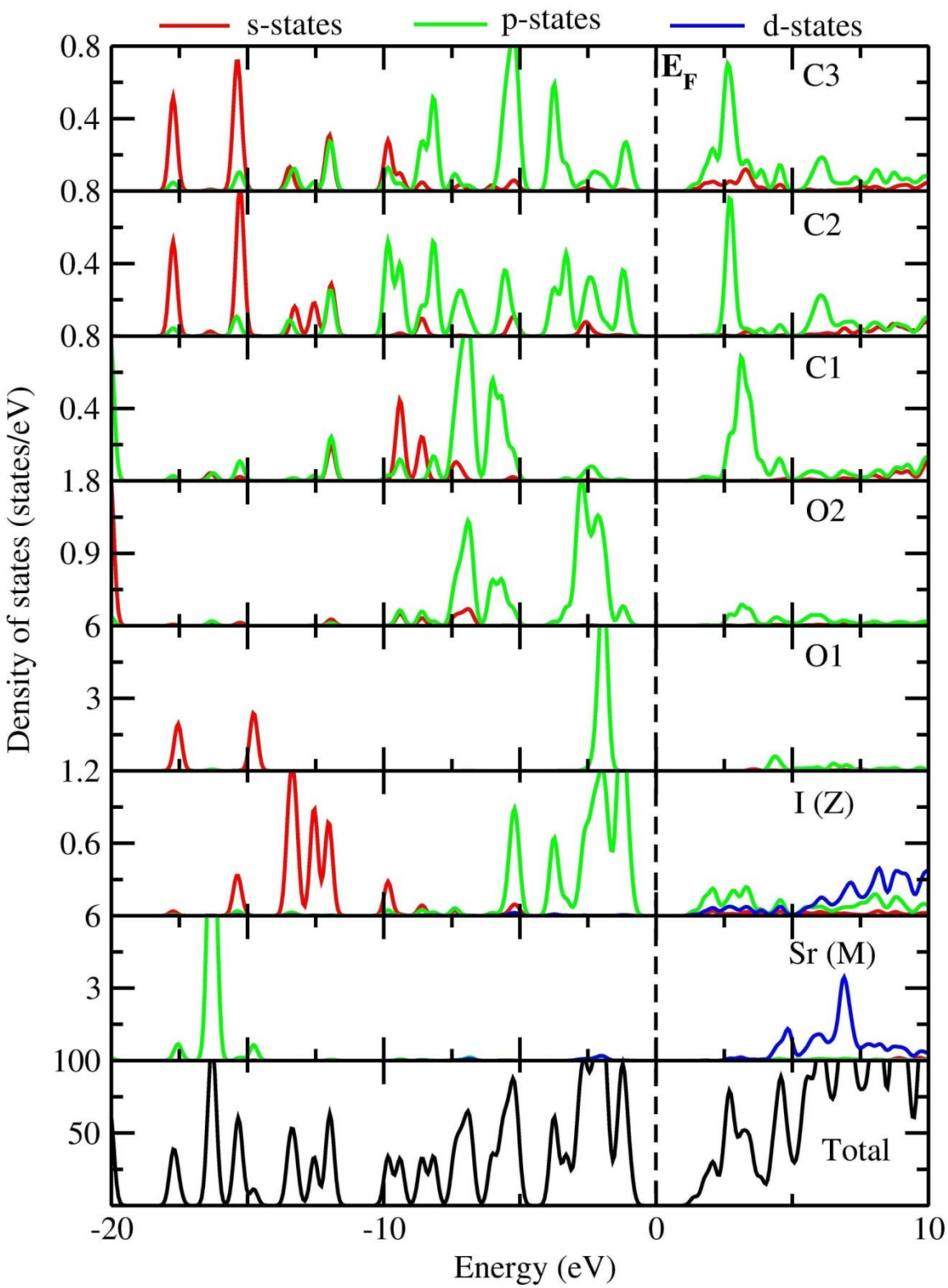

Figure S31. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Sr, I)-90 °in the cubic *Fm-3m* symmetry (no. 225).

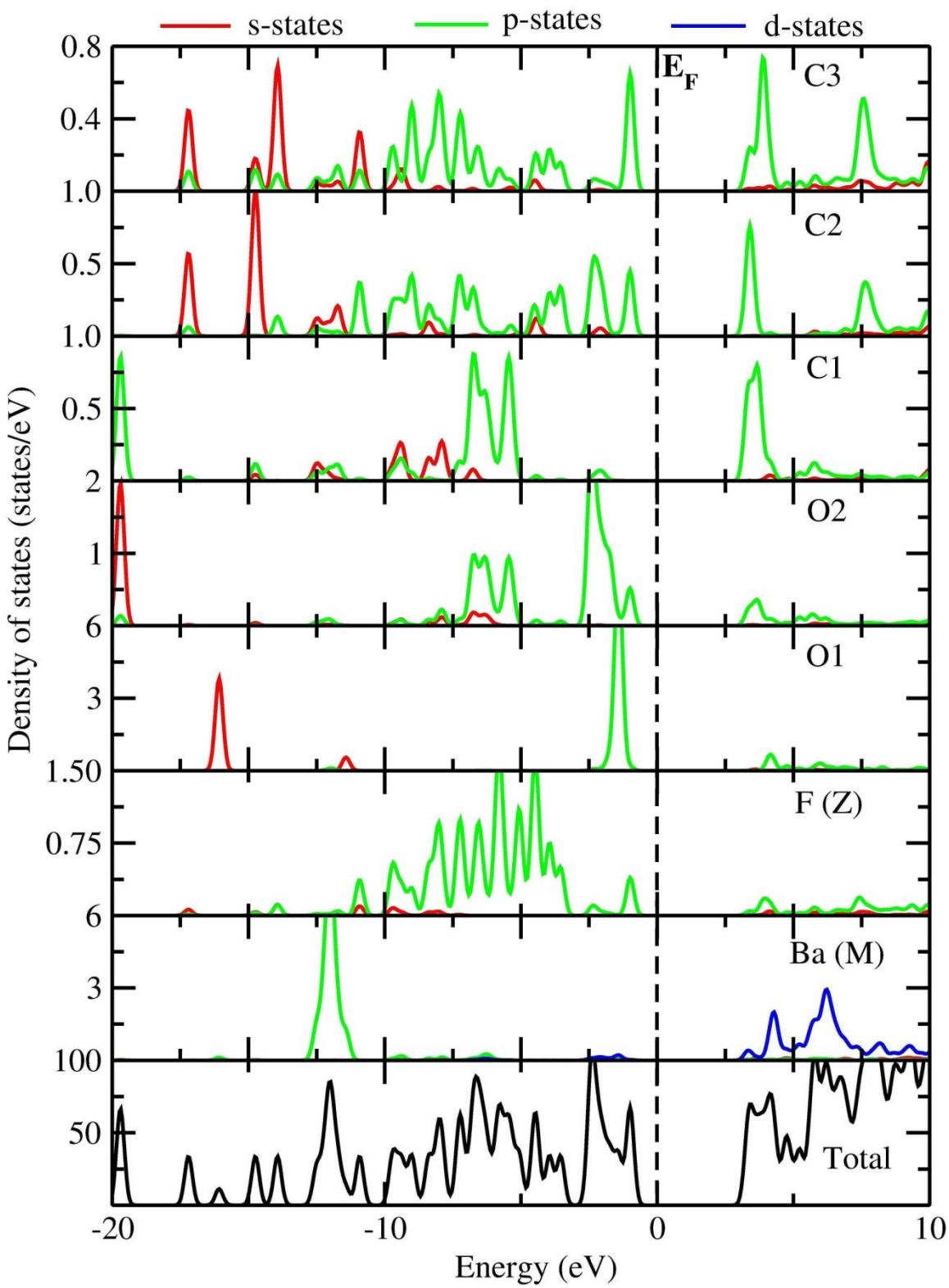

Figure S32. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Ba, F)-90 ° in the cubic *Fm-3m* symmetry (no. 225).

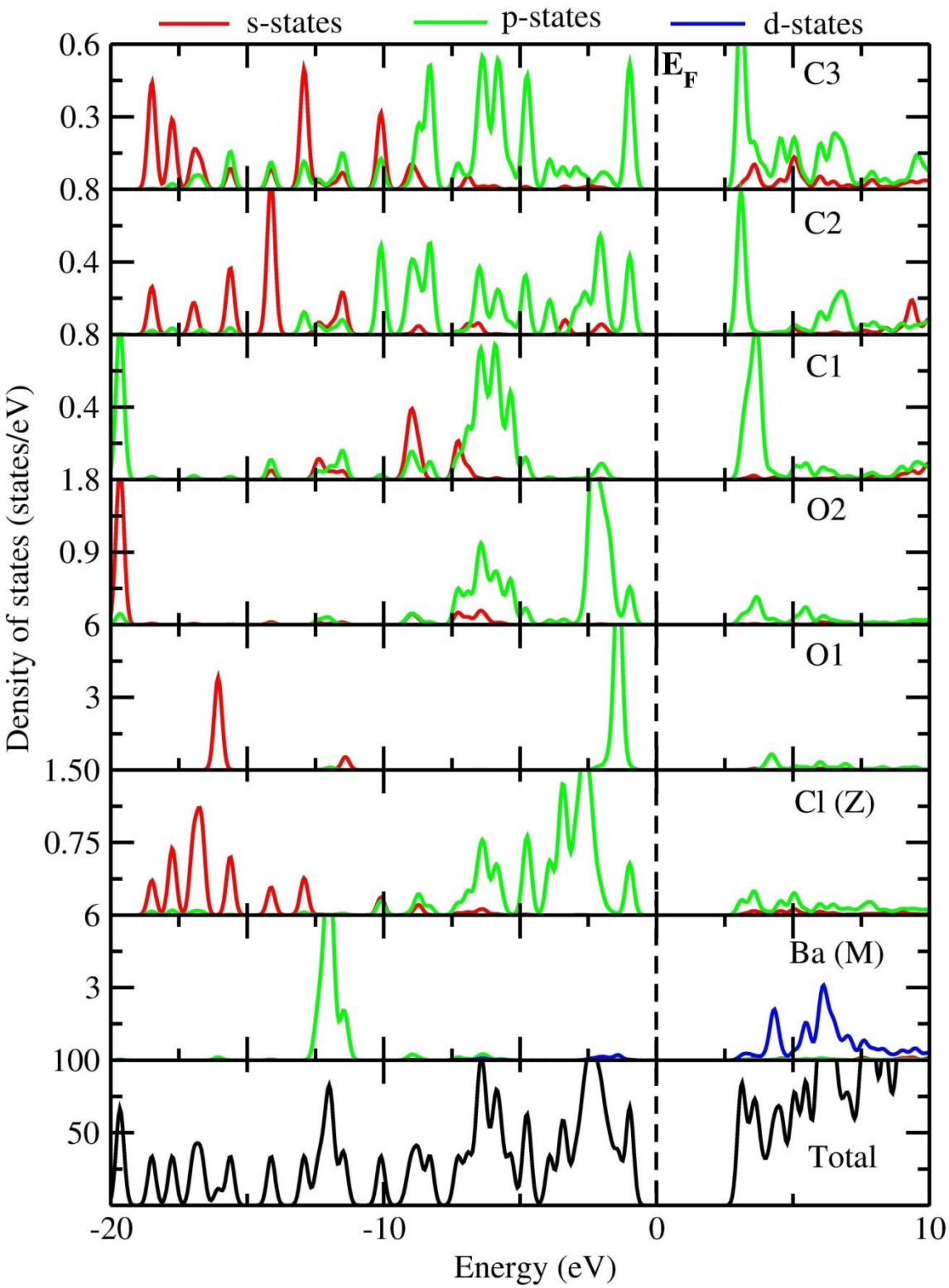

Figure S33. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Ba, Cl)-90 °in the cubic *Fm-3m* symmetry (no. 225).

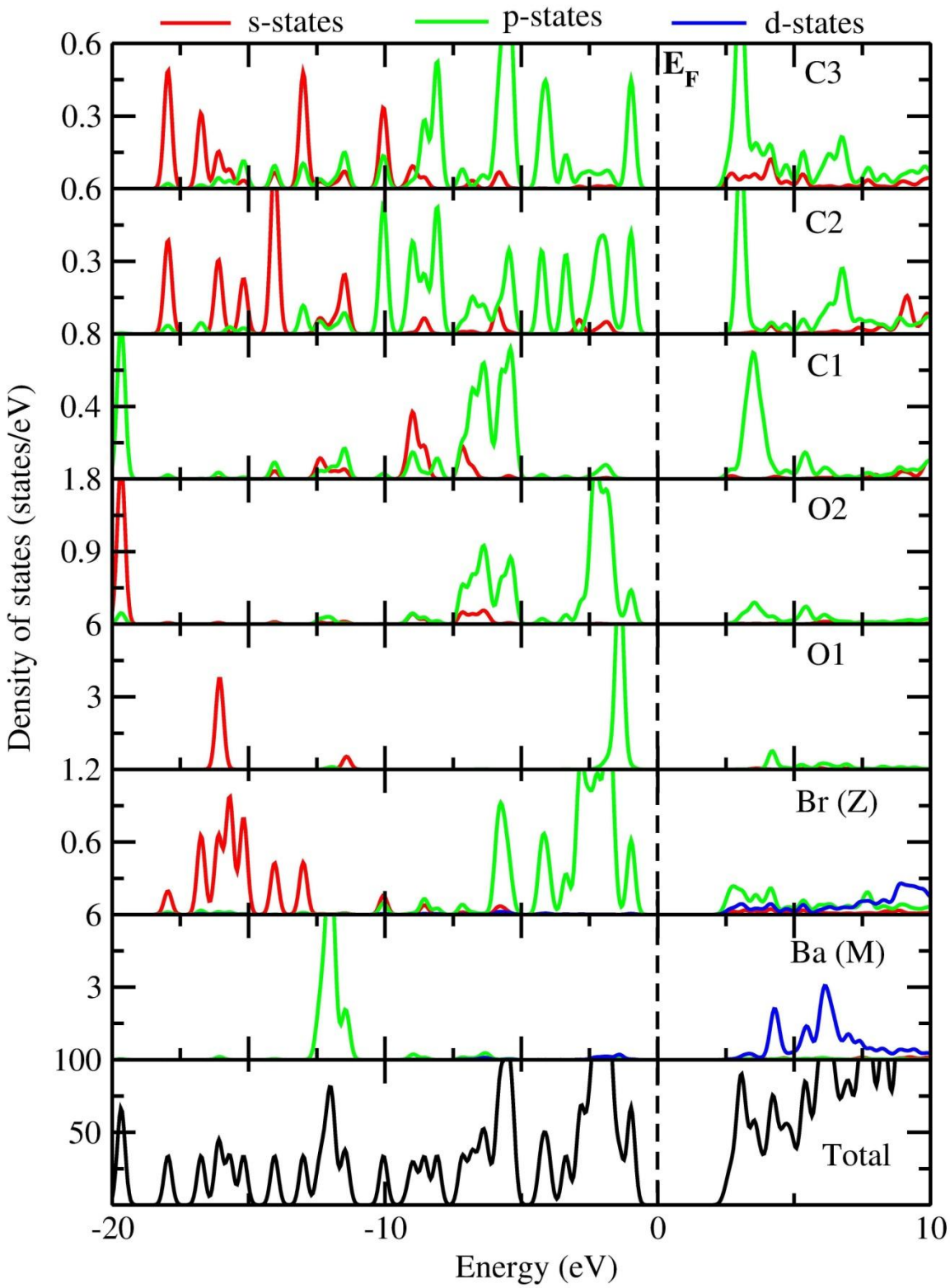

Figure S34. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Ba, Br)-90 °in the cubic *Fm-3m* symmetry (no. 225).

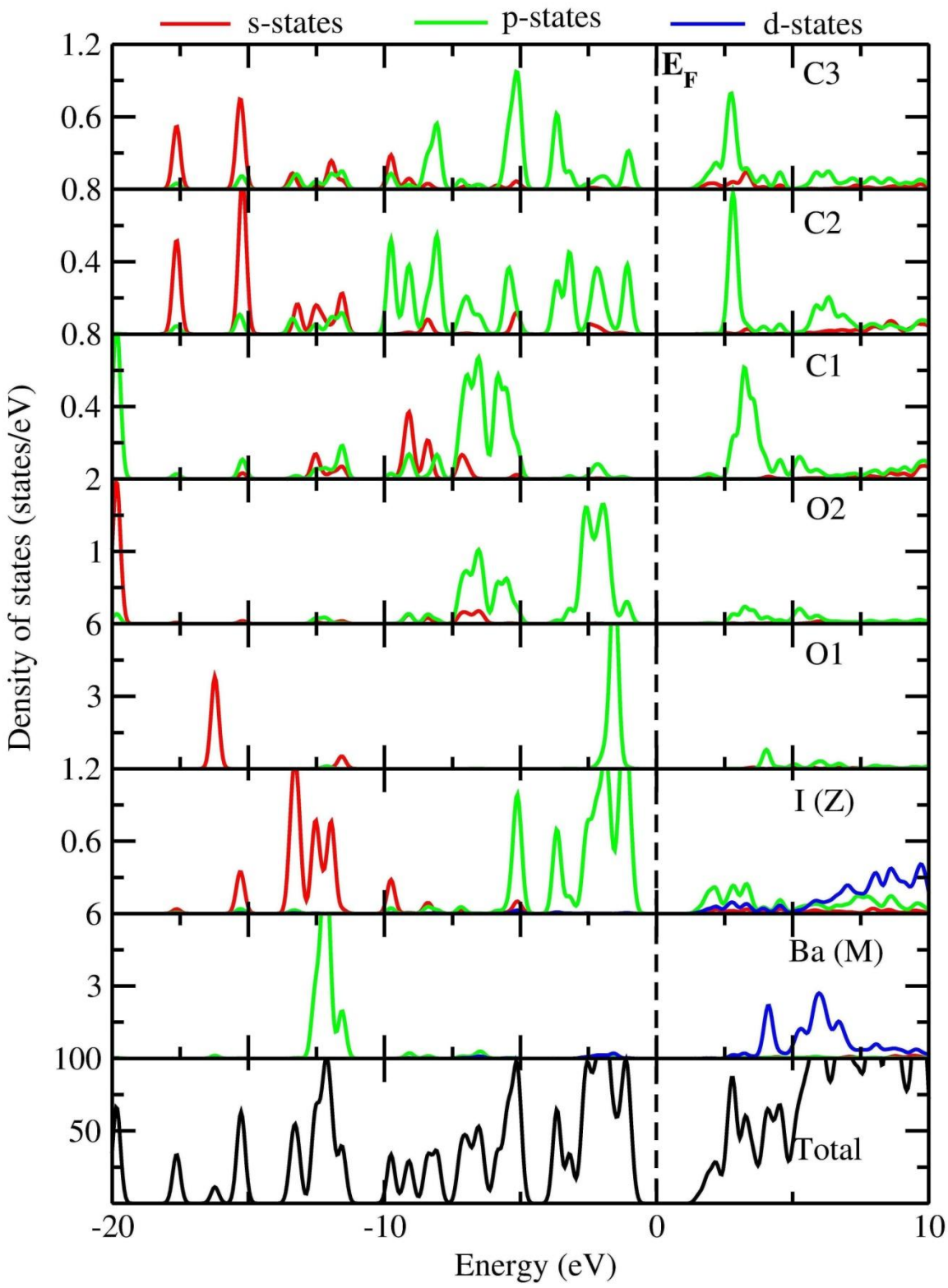

Figure S35. The calculated total density of states (TDOS) and partial density of states (PDOS) for (Ba, I)-90 °in the cubic *Fm-3m* symmetry (no. 225).

Table S2. The calculated Hirschfeld charge (HC, given in terms of *e*), bond overlap populations (BOP) for (Zn, Z)-90° (Z = F, Cl, Br, and I) as well as pristine MOF-5, i.e., (Zn, H)-0 °. Note that the atomic labels of atoms are numbered according to Figure 1.

Materials	Atomic site	HC (*e*)	BOP
(Zn, H)-0 °	Zn (M)	0.43	0.25-0.29 (Zn-O)
	O1	-0.35	0.25 (O1-Zn)
	O2	-0.22	0.29 (O2-Zn)
	C1	0.18	0.91 (C1-O2)
			0.83 (C1-C2)
	C2	-0.01	1.08 (C2-C3)
	C3	-0.03	1.10 (C3-C3)
	H (Z)	0.05	0.89 (H-C3)
(Zn, F)-90 °	Zn (M)	0.42	0.25-0.27 (Zn-O)
	O1	-0.34	0.25 (O1-Zn)
	O2	-0.22	0.27 (O2-Zn)
	C1	0.18	0.93 (C1-O2)
			0.76 (C1-C2)
	C2	-0.04	1.12 (C2-C3)
	C3	0.07	1.12 (C3-C3)
	F (Z)	-0.08	0.47 (F-C3)
(Zn, Cl)-90 °	Zn (M)	0.42	0.25-0.28 (Zn-O)
	O1	-0.34	0.25 (O1-Zn)
	O2	-0.23	0.28 (O2-Zn)
	C1	0.16	0.93 (C1-O2)
			0.78 (C1-C2)
	C2	-0.03	1.10 (C2-C3)
	C3	0.00	1.13 (C3-C3)
	Cl (Z)	0.02	0.59 (Cl-C3)
(Zn, Br)-90 °	Zn (M)	0.42	0.26-0.29 (Zn-O)
	O1	-0.34	0.26 (O1-Zn)
	O2	-0.23	0.29 (O2-Zn)
	C1	0.16	0.93 (C1-O2)
			0.80 (C1-C2)
	C2	-0.02	1.15 (C2-C3)
	C3	-0.01	1.15 (C3-C3)
	Br (Z)	0.03	0.48 (Br-C3)
(Zn, I)-90 °	Zn (M)	0.44	0.28 (Zn-O)
	O1	-0.37	0.28 (O1-Zn)
	O2	-0.22	0.28 (O2-Zn)
	C1	0.16	0.93 (C1-O2)
			0.81 (C1-C2)
	C2	-0.02	1.14 (C2-C3)
	C3	-0.03	1.15 (C3-C3)
	I (Z)	0.06	0.47 (I-C3)

Table S3. The electron configurations of atoms for (Zn, Z)-90° (Z = F, Cl, Br, and I) as well as pristine MOF-5, i.e., (Zn, H)-0 ° from atomic populations (Mulliken) with CASTEP code. Note that the atomic labels of atoms are numbered according to Figure 1.

Materials	Atom	*s*	*p*	*d*	Total	Charge (e)
(Zn, H)-0 °	Zn (M)	0.39	0.35	9.97	10.71	1.29
	O1	1.88	5.19	0.00	7.07	-1.07
	O2	1.80	4.84	0.00	6.64	-0.64
	C1	0.94	2.45	0.00	3.39	0.61
	C2	1.10	2.96	0.00	4.06	-0.06
	C3	1.18	3.08	0.00	4.26	-0.26
	H (Z)	0.72	0.00	0.00	0.72	0.28
(Zn, F)-90 °	Zn (M)	0.43	0.34	9.97	10.74	1.26
	O1	1.88	5.19	0.00	7.07	-1.07
	O2	1.80	4.81	0.00	6.62	-0.62
	C1	0.92	2.44	0.00	3.37	0.63
	C2	1.06	3.04	0.00	4.10	-0.10
	C3	0.98	2.67	0.00	3.65	0.35
	F (Z)	1.93	5.40	0.00	7.33	-0.33
(Zn, Cl)-90 °	Zn (M)	0.43	0.35	9.97	10.75	1.25
	O1	1.88	5.19	0.00	7.07	-1.07
	O2	1.80	4.81	0.00	6.61	-0.61
	C1	0.93	2.45	0.00	3.37	0.63
	C2	1.06	2.96	0.00	4.02	-0.02
	C3	1.10	2.93	0.00	4.03	-0.03
	Cl (Z)	1.90	5.09	0.00	6.99	0.01
(Zn, Br)-90 °	Zn (M)	0.46	0.42	9.97	10.85	1.15
	O1	1.88	5.16	0.00	7.04	-1.04
	O2	1.80	4.80	0.00	6.60	-0.60
	C1	0.93	2.46	0.00	3.40	0.60
	C2	1.07	2.96	0.00	4.03	-0.03
	C3	1.14	2.96	0.00	4.10	-0.10
	Br (Z)	1.87	5.01	0.00	6.88	0.12
(Zn, I)-90 °	Zn (M)	0.47	0.43	9.97	10.87	1.13
	O1	1.89	5.10	0.00	6.99	-0.99
	O2	1.80	4.81	0.00	6.61	-0.61
	C1	0.94	2.47	0.00	3.40	0.60
	C2	1.07	2.95	0.00	4.02	-0.02
	C3	1.17	3.02	0.00	4.18	-0.18
	I (Z)	1.86	4.93	0.00	6.79	0.21

Table S4. The calculated Hirschfeld charge (HC, given in terms of *e*), bond overlap populations (BOP) for (Cd, Z)-90° (Z = F, Cl, Br, and I) as well as Cd-MOF-5, i.e., (Cd, H)-0 °. Note that the atomic labels of atoms are numbered according to Figure 1.

Materials	Atomic site	HC (*e*)	BOP
(Cd, H)-0 °	Cd (M)	0.55	0.21-0.22 (Cd-O)
	O1	-0.40	0.21 (O1-Cd)
	O2	-0.25	0.22 (O2-Cd)
	C1	0.17	0.91 (C1-O2)
			0.83 (C1-C2)
	C2	-0.01	1.08 (C2-C3)
	C3	-0.04	1.10 (C3-C3)
	H (Z)	0.04	0.89 (H-C3)
(Cd, F)-90 °	Cd (M)	0.56	0.21-0.22 (Cd-O)
	O1	-0.40	0.22 (O1-Cd)
	O2	-0.24	0.21 (O2-Cd)
	C1	0.17	0.92 (C1-O2)
			0.75 (C1-C2)
	C2	-0.04	1.12 (C2-C3)
	C3	0.07	1.13 (C3-C3)
	F (Z)	-0.07	0.47 (F-C3)
(Cd, Cl)-90 °	Cd (M)	0.56	0.21-0.22 (Cd-O)
	O1	-0.40	0.22 (O1-Cd)
	O2	-0.24	0.21 (O2-Cd)
	C1	0.15	0.92 (C1-O2)
			0.77 (C1-C2)
	C2	-0.03	1.13 (C2-C3)
	C3	0.00	1.10 (C3-C3)
	Cl (Z)	0.01	0.59 (Cl-C3)
(Cd, Br)-90 °	Cd (M)	0.56	0.22-0.23 (Cd-O)
	O1	-0.40	0.23 (O1-Cd)
	O2	-0.24	0.22 (O2-Cd)
	C1	0.15	0.93 (C1-O2)
			0.79 (C1-C2)
	C2	-0.03	1.14 (C2-C3)
	C3	-0.01	1.15 (C3-C3)
	Br (Z)	0.03	0.50 (Br-C3)
(Cd, I)-90 °	Cd (M)	0.56	0.23 (Cd-O)
	O1	-0.40	0.23 (O1-Cd)
	O2	-0.24	0.23 (O2-Cd)
	C1	0.14	0.92 (C1-O2)
			0.80 (C1-C2)
	C2	-0.03	1.14 (C2-C3)
	C3	-0.03	1.15 (C3-C3)
	I (Z)	0.06	0.50 (I-C3)

Table S5. The electron configurations of atoms for (Cd, Z)-90° (Z = F, Cl, Br, and I) as well as (Cd, H)-0° from atomic populations (Mulliken) with CASTEP code. Note that the atomic labels of atoms are numbered according to Figure 1.

Materials	Atom	*s*	*p*	*d*	Total	Charge (e)
(Cd, H)-0 °	Cd (M)	0.42	0.32	9.98	10.72	1.28
	O1	1.91	5.12	0.00	7.02	-1.02
	O2	1.80	4.83	0.00	6.63	-0.63
	C1	0.94	2.47	0.00	3.41	0.59
	C2	1.09	2.96	0.00	4.06	-0.06
	C3	1.18	3.09	0.00	4.27	-0.27
	H (Z)	0.71	0.00	0.00	0.71	0.29
(Cd, F)-90 °	Cd (M)	0.45	0.32	9.98	10.76	1.24
	O1	1.91	5.11	0.00	7.01	-1.01
	O2	1.81	4.81	0.00	6.62	-0.62
	C1	0.91	2.44	0.00	3.35	0.65
	C2	1.06	3.04	0.00	4.10	-0.10
	C3	0.98	2.67	0.00	3.65	0.35
	F (Z)	1.93	5.41	0.00	7.33	-0.33
(Cd, Cl)-90 °	Cd (M)	0.45	0.33	9.98	10.76	1.24
	O1	1.91	5.10	0.00	7.01	-1.01
	O2	1.81	4.81	0.00	6.62	-0.62
	C1	0.92	2.44	0.00	3.36	0.64
	C2	1.06	2.96	0.00	4.02	-0.02
	C3	1.10	2.93	0.00	4.03	-0.03
	Cl (Z)	1.90	5.09	0.00	6.99	0.01
(Cd, Br)-90 °	Cd (M)	0.48	0.39	9.98	10.85	1.15
	O1	1.91	5.08	0.00	6.99	-0.99
	O2	1.81	4.81	0.00	6.61	-0.61
	C1	0.92	2.46	0.00	3.38	0.62
	C2	1.07	2.96	0.00	4.03	-0.03
	C3	1.14	2.96	0.00	4.10	-0.10
	Br (Z)	1.88	5.01	0.00	6.89	0.11
(Cd, I)-90 °	Cd (M)	0.49	0.40	9.98	10.86	1.14
	O1	1.91	5.08	0.00	6.99	-0.99
	O2	1.81	4.81	0.00	6.61	-0.61
	C1	0.93	2.46	0.00	3.39	0.61
	C2	1.07	2.95	0.00	4.02	-0.02
	C3	1.17	3.02	0.00	4.19	-0.19
	I (Z)	1.87	4.92	0.00	6.80	0.20

Table S6. The calculated Hirschfeld charge (HC, given in terms of *e*), bond overlap populations (BOP) for (Be, Z)-90° (Z = F, Cl, Br, and I) as well as Be-MOF-5, i.e., (Be, H)-0 °. Note that the atomic labels of atoms are numbered according to Figure 1.

Materials	Atomic site	HC (*e*)	BOP
(Be, H)-0 °	Be (M)	0.22	0.36-0.37 (Be-O)
	O1	-0.27	0.37 (O1-Be)
	O2	-0.18	0.36 (O2-Be)
	C1	0.21	0.92 (C1-O2)
			0.84 (C1-C2)
	C2	-0.01	1.08 (C2-C3)
	C3	-0.03	1.10 (C3-C3)
	H (Z)	0.05	0.86 (H-C3)
(Be, F)-90 °	Be (M)	0.09	0.35-0.36 (Be-O)
	O1	-0.27	0.36 (O1-Be)
	O2	-0.18	0.35 (O2-Be)
	C1	0.22	0.94 (C1-O2)
			0.75 (C1-C2)
	C2	-0.03	1.11 (C2-C3)
	C3	0.07	1.12 (C3-C3)
	F (Z)	-0.07	0.47 (F-C3)
(Be, Cl)-90 °	Be (M)	0.08	0.35-0.36 (Be-O)
	O1	-0.26	0.36 (O1-Be)
	O2	-0.18	0.35 (O2-Be)
	C1	0.20	0.94 (C1-O2)
			0.78 (C1-C2)
	C2	-0.02	1.12 (C2-C3)
	C3	0.01	1.09 (C3-C3)
	Cl (Z)	0.02	0.59 (Cl-C3)
(Be, Br)-90 °	Be (M)	0.15	0.36-0.37 (Be-O)
	O1	-0.28	0.37 (O1-Be)
	O2	-0.18	0.36 (O2-Be)
	C1	0.20	0.95 (C1-O2)
			0.81 (C1-C2)
	C2	-0.02	1.14 (C2-C3)
	C3	-0.01	1.15 (C3-C3)
	Br (Z)	0.04	0.45 (Br-C3)
(Be, I)-90 °	Be (M)	0.14	0.33-0.39 (Be-O)
	O1	-0.26	0.33 (O1-Be)
	O2	-0.19	0.39 (O2-Be)
	C1	0.19	0.93 (C1-O2)
			0.80 (C1-C2)
	C2	-0.02	1.13 (C2-C3)
	C3	-0.04	1.15 (C3-C3)
	I (Z)	0.07	0.44 (I-C3)

Table S7. The electron configurations of atoms for (Be, Z)-90° (Z = F, Cl, Br, and I) as well as (Be, H)-0° from atomic populations (Mulliken) with CASTEP code. Note that the atomic labels of atoms are numbered according to Figure 1.

Materials	Atom	*s*	*p*	*d*	Total	Charge (e)
(Be, H)-0 °	Be (M)	2.27	0.60	0.00	2.86	1.14
	O1	1.83	5.12	0.00	6.96	-0.96
	O2	1.79	4.84	0.00	6.63	-0.63
	C1	0.93	2.46	0.00	3.39	0.61
	C2	1.08	2.96	0.00	4.05	-0.05
	C3	1.18	3.08	0.00	4.26	-0.26
	H (Z)	0.69	0.00	0.00	0.69	0.31
(Be, F)-90 °	Be (M)	2.25	0.60	0.00	2.85	1.15
	O1	1.83	5.18	0.00	7.00	-1.00
	O2	1.79	4.81	0.00	6.60	-0.60
	C1	0.92	2.45	0.00	3.37	0.63
	C2	1.06	3.03	0.00	4.09	-0.09
	C3	0.98	2.66	0.00	3.64	0.36
	F (Z)	1.93	5.40	0.00	7.33	-0.33
(Be, Cl)-90 °	Be (M)	2.25	0.61	0.00	2.86	1.14
	O1	1.83	5.17	0.00	7.00	-1.00
	O2	1.79	4.80	0.00	6.60	-0.60
	C1	0.93	2.46	0.00	3.38	0.62
	C2	1.05	2.95	0.00	4.01	-0.01
	C3	1.09	2.93	0.00	4.02	-0.02
	Cl (Z)	1.90	5.09	0.00	6.99	0.01
(Be, Br)-90 °	Be (M)	2.27	0.67	0.00	2.94	1.06
	O1	1.84	5.09	0.00	6.93	-0.93
	O2	1.78	4.81	0.00	6.60	-0.60
	C1	0.94	2.47	0.00	3.41	0.59
	C2	1.07	2.96	0.00	4.03	-0.03
	C3	1.14	2.97	0.00	4.10	-0.10
	Br (Z)	1.85	5.01	0.00	6.86	0.14
(Be, I)-90 °	Be (M)	2.28	0.70	0.00	2.98	1.02
	O1	1.83	5.19	0.00	7.01	-1.01
	O2	1.79	4.79	0.00	6.58	-0.58
	C1	0.95	2.47	0.00	3.42	0.58
	C2	1.09	2.94	0.00	4.02	-0.02
	C3	1.17	3.02	0.00	4.19	-0.19
	I (Z)	1.85	4.92	0.00	6.77	0.23

Table S8. The calculated Hirschfeld charge (HC, given in terms of *e*), bond overlap populations (BOP) for (Mg, Z)-90° (Z = F, Cl, Br, and I) as well as Mg-MOF-5, i.e., (Mg, H)-0 °. Note that the atomic labels of atoms are numbered according to Figure 1.

Materials	Atomic site	HC (*e*)	BOP
(Mg, H)-0 °	Mg (M)	0.48	0.23 (Mg-O)
	O1	-0.44	0.23 (O1-Mg)
	O2	-0.23	0.23 (O2-Mg)
	C1	0.19	0.91 (C1-O2)
			0.84 (C1-C2)
	C2	-0.01	1.08 (C2-C3)
	C3	-0.03	1.11 (C3-C3)
	H (Z)	0.05	0.89 (H-C3)
(Mg, F)-90 °	Mg (M)	0.49	0.20-0.23 (Mg-O)
	O1	-0.45	0.23 (O1-Mg)
	O2	-0.23	0.20 (O2-Mg)
	C1	0.20	0.94 (C1-O2)
			0.76 (C1-C2)
	C2	-0.03	1.12 (C2-C3)
	C3	0.07	1.12 (C3-C3)
	F (Z)	-0.07	0.47 (F-C3)
(Mg, Cl)-90 °	Mg (M)	0.48	0.20-0.23 (Mg-O)
	O1	-0.45	0.23 (O1-Mg)
	O2	-0.23	0.20 (O2-Mg)
	C1	0.18	0.93 (C1-O2)
			0.78 (C1-C2)
	C2	-0.03	1.12 (C2-C3)
	C3	0.00	1.10 (C3-C3)
	Cl (Z)	0.02	0.59 (Cl-C3)
(Mg, Br)-90 °	Mg (M)	0.48	0.23 (Mg-O)
	O1	-0.45	0.23 (O1-Mg)
	O2	-0.23	0.23 (O2-Mg)
	C1	0.18	0.94 (C1-O2)
			0.80 (C1-C2)
	C2	-0.02	1.14 (C2-C3)
	C3	-0.01	1.15 (C3-C3)
	Br (Z)	0.04	0.48 (Br-C3)
(Mg, I)-90 °	Mg (M)	0.48	0.23-0.24 (Mg-O)
	O1	-0.45	0.24 (O1-Mg)
	O2	-0.24	0.23 (O2-Mg)
	C1	0.17	0.94 (C1-O2)
			0.80 (C1-C2)
	C2	-0.02	1.14 (C2-C3)
	C3	-0.03	1.15 (C3-C3)
	I (Z)	0.06	0.48 (I-C3)

Table S9. The electron configurations of atoms for (Mg, Z)-90° (Z = F, Cl, Br, and I) as well as (Mg, H)-0° from atomic populations (Mulliken) with CASTEP code. Note that the atomic labels of atoms are numbered according to Figure 1.

Materials	Atom	*s*	*p*	*d*	Total	Charge (e)
(Mg, H)-0 °	Mg (M)	0.20	6.22	0.00	6.42	1.58
	O1	1.90	5.38	0.00	7.28	-1.28
	O2	1.80	4.91	0.00	6.71	-0.71
	C1	0.94	2.46	0.00	3.40	0.60
	C2	1.09	2.97	0.00	4.06	-0.06
	C3	1.18	3.08	0.00	4.26	-0.26
	H (Z)	0.72	0.00	0.00	0.72	0.28
(Mg, F)-90 °	Mg (M)	0.28	6.18	0.00	6.46	1.54
	O1	1.90	5.41	0.00	7.31	-1.31
	O2	1.81	4.88	0.00	6.69	-0.69
	C1	0.92	2.45	0.00	3.37	0.63
	C2	1.06	3.04	0.00	4.10	-0.10
	C3	0.98	2.67	0.00	3.65	0.35
	F (Z)	1.93	5.41	0.00	7.33	-0.33
(Mg, Cl)-90 °	Mg (M)	0.28	6.19	0.00	6.47	1.53
	O1	1.90	5.41	0.00	7.31	-1.31
	O2	1.81	4.88	0.00	6.69	-0.69
	C1	0.93	2.45	0.00	3.38	0.62
	C2	1.06	2.96	0.00	4.01	-0.01
	C3	1.09	2.93	0.00	4.02	-0.02
	Cl (Z)	1.90	5.09	0.00	6.99	0.01
(Mg, Br)-90 °	Mg (M)	0.28	6.28	0.00	6.56	1.44
	O1	1.90	5.39	0.00	7.29	-1.29
	O2	1.81	4.87	0.00	6.68	-0.68
	C1	0.93	2.47	0.00	3.40	0.60
	C2	1.07	2.96	0.00	4.03	-0.03
	C3	1.14	2.96	0.00	4.10	-0.10
	Br (Z)	1.87	5.01	0.00	6.88	0.12
(Mg, I)-90 °	Mg (M)	0.29	6.29	0.00	6.58	1.42
	O1	1.90	5.38	0.00	7.28	-1.28
	O2	1.81	4.87	0.00	6.67	-0.67
	C1	0.94	2.48	0.00	3.41	0.59
	C2	1.07	2.95	0.00	4.02	-0.02
	C3	1.17	3.02	0.00	4.19	-0.19
	I (Z)	1.87	4.92	0.00	6.79	0.21

Table S10. The calculated Hirschfeld charge (HC, given in terms of *e*), bond overlap populations (BOP) for (Ca, Z)-90° (Z = F, Cl, Br, and I) as well as Ca-MOF-5, i.e., (Ca, H)-0 °. Note that the atomic labels of atoms are numbered according to Figure 1.

Materials	Atomic site	HC (*e*)	BOP
(Ca, H)-0 °	Ca (M)	0.55	0.14-0.18 (Ca-O)
	O1	-0.48	0.18 (O1-Ca)
	O2	-0.26	0.14 (O2-Ca)
	C1	0.19	0.89 (C1-O2)
			0.82 (C1-C2)
	C2	-0.01	1.07 (C2-C3)
	C3	-0.04	1.09 (C3-C3)
	H (Z)	0.04	0.86 (H-C3)
(Ca, F)-90 °	Ca (M)	0.56	0.13-0.18 (Ca-O)
	O1	-0.48	0.18 (O1-Ca)
	O2	-0.25	0.13 (O2-Ca)
	C1	0.20	0.91 (C1-O2)
			0.73 (C1-C2)
	C2	-0.04	1.11 (C2-C3)
	C3	0.07	1.12 (C3-C3)
	F (Z)	-0.07	0.46 (F-C3)
(Ca, Cl)-90 °	Ca (M)	0.55	0.13-0.18 (Ca-O)
	O1	-0.48	0.18 (O1-Ca)
	O2	-0.25	0.13 (O2-Ca)
	C1	0.18	0.91 (C1-O2)
			0.76 (C1-C2)
	C2	-0.03	1.12 (C2-C3)
	C3	0.00	1.10 (C3-C3)
	Cl (Z)	0.02	0.58 (Cl-C3)
(Ca, Br)-90 °	Ca (M)	0.54	0.14-0.18 (Ca-O)
	O1	-0.48	0.18 (O1-Ca)
	O2	-0.25	0.14 (O2-Ca)
	C1	0.17	0.92 (C1-O2)
			0.78 (C1-C2)
	C2	-0.02	1.14 (C2-C3)
	C3	-0.01	1.15 (C3-C3)
	Br (Z)	0.04	0.50 (Br-C3)
(Ca, I)-90 °	Ca (M)	0.53	0.14-0.18 (Ca-O)
	O1	-0.48	0.18 (O1-Ca)
	O2	-0.25	0.14 (O2-Ca)
	C1	0.17	0.92 (C1-O2)
			0.79 (C1-C2)
	C2	-0.02	1.14 (C2-C3)
	C3	-0.03	1.15 (C3-C3)
	I (Z)	0.07	0.49 (I-C3)

Table S11. The electron configurations of atoms for (Ca, Z)-90° (Z = F, Cl, Br, and I) as well as (Ca, H)-0° from atomic populations (Mulliken) with CASTEP code. Note that the atomic labels of atoms are numbered according to Figure 1.

Materials	Atom	*s*	*p*	*d*	Total	Charge (e)
(Ca, H)-0 °	Ca (M)	2.13	6.00	0.53	8.65	1.35
	O1	1.90	5.24	0.00	7.14	-1.14
	O2	1.80	4.89	0.00	6.70	-0.70
	C1	0.92	2.43	0.00	3.34	0.66
	C2	1.09	2.97	0.00	4.06	-0.06
	C3	1.18	3.08	0.00	4.27	-0.27
	H (Z)	0.69	0.00	0.00	0.69	0.31
(Ca, F)-90 °	Ca (M)	2.13	6.00	0.51	8.64	1.36
	O1	1.91	5.25	0.00	7.15	-1.15
	O2	1.80	4.87	0.00	6.67	-0.67
	C1	0.89	2.42	0.00	3.31	0.69
	C2	1.06	3.04	0.00	4.10	-0.10
	C3	0.98	2.67	0.00	3.64	0.36
	F (Z)	1.93	5.41	0.00	7.34	-0.34
(Ca, Cl)-90 °	Ca (M)	2.13	6.00	0.51	8.64	1.36
	O1	1.91	5.25	0.00	7.15	-1.15
	O2	1.80	4.87	0.00	6.67	-0.67
	C1	0.90	2.42	0.00	3.32	0.68
	C2	1.06	2.96	0.00	4.01	-0.01
	C3	1.09	2.93	0.00	4.02	-0.02
	Cl (Z)	1.90	5.10	0.00	7.00	0.00
(Ca, Br)-90 °	Ca (M)	2.15	6.00	0.52	8.67	1.33
	O1	1.90	5.24	0.00	7.15	-1.15
	O2	1.80	4.86	0.00	6.66	-0.66
	C1	0.91	2.44	0.00	3.35	0.65
	C2	1.07	2.96	0.00	4.03	-0.03
	C3	1.13	2.96	0.00	4.09	-0.09
	Br (Z)	1.88	5.01	0.00	6.90	0.10
(Ca, I)-90 °	Ca (M)	2.16	6.00	0.51	8.66	1.34
	O1	1.90	5.24	0.00	7.14	-1.14
	O2	1.80	4.86	0.00	6.66	-0.66
	C1	0.92	2.45	0.00	3.36	0.64
	C2	1.07	2.95	0.00	4.03	-0.03
	C3	1.16	3.01	0.00	4.18	-0.18
	I (Z)	1.89	4.93	0.00	6.82	0.18

Table S12. The calculated Hirschfeld charge (HC, given in terms of *e*), bond overlap populations (BOP) for (Sr, Z)-90° (Z = F, Cl, Br, and I) as well as Sr-MOF-5, i.e., (Sr, H)-0 °. Note that the atomic labels of atoms are numbered according to Figure 1.

Materials	Atomic site	HC (*e*)	BOP
(Sr, H)-0 °	Sr (M)	0.64	0.14-0.17 (Sr-O)
	O1	-0.52	0.17 (O1-Sr)
	O2	-0.27	0.14 (O2-Sr)
	C1	0.18	0.89 (C1-O2)
			0.81 (C1-C2)
	C2	-0.01	1.08 (C2-C3)
	C3	-0.04	1.09 (C3-C3)
	H (Z)	0.04	0.86 (H-C3)
(Sr, F)-90 °	Sr (M)	0.65	0.12-0.18 (Sr-O)
	O1	-0.53	0.18 (O1-Sr)
	O2	-0.26	0.12 (O2-Sr)
	C1	0.19	0.92 (C1-O2)
			0.73 (C1-C2)
	C2	-0.04	1.11 (C2-C3)
	C3	0.06	1.12 (C3-C3)
	F (Z)	-0.08	0.46 (F-C3)
(Sr, Cl)-90 °	Sr (M)	0.63	0.12-0.18 (Sr-O)
	O1	-0.53	0.18 (O1-Sr)
	O2	-0.27	0.12 (O2-Sr)
	C1	0.17	0.91 (C1-O2)
			0.76 (C1-C2)
	C2	-0.03	1.12 (C2-C3)
	C3	0.00	1.10 (C3-C3)
	Cl (Z)	0.01	0.58 (Cl-C3)
(Sr, Br)-90 °	Sr (M)	0.62	0.13-0.18 (Sr-O)
	O1	-0.53	0.18 (O1-Sr)
	O2	-0.27	0.13 (O2-Sr)
	C1	0.16	0.92 (C1-O2)
			0.78 (C1-C2)
	C2	-0.03	1.14 (C2-C3)
	C3	-0.01	1.15 (C3-C3)
	Br (Z)	0.03	0.50 (Br-C3)
(Sr, I)-90 °	Sr (M)	0.60	0.14-0.18 (Sr-O)
	O1	-0.53	0.18 (O1-Sr)
	O2	-0.27	0.14 (O2-Sr)
	C1	0.16	0.92 (C1-O2)
			0.78 (C1-C2)
	C2	-0.03	1.14 (C2-C3)
	C3	-0.03	1.15 (C3-C3)
	I (Z)	0.07	0.50 (I-C3)

Table S13. The electron configurations of atoms for (Sr, Z)-90° (Z = F, Cl, Br, and I) as well as (Sr, H)-0° from atomic populations (Mulliken) with CASTEP code. Note that the atomic labels of atoms are numbered according to Figure 1.

Materials	Atom	*s*	*p*	*d*	Total	Charge (e)
(Sr, H)-0 °	Sr (M)	2.11	5.99	0.51	8.61	1.39
	O1	1.90	5.24	0.00	7.14	-1.14
	O2	1.80	4.89	0.00	6.70	-0.70
	C1	0.92	2.44	0.00	3.36	0.64
	C2	1.09	2.97	0.00	4.06	-0.06
	C3	1.18	3.08	0.00	4.27	-0.27
	H (Z)	0.69	0.00	0.00	0.69	0.31
(Sr, F)-90 °	Sr (M)	2.11	5.99	0.49	8.60	1.40
	O1	1.91	5.24	0.00	7.15	-1.15
	O2	1.81	4.86	0.00	6.67	-0.67
	C1	0.90	2.43	0.00	3.33	0.67
	C2	1.06	3.04	0.00	4.10	-0.10
	C3	0.98	2.67	0.00	3.65	0.35
	F (Z)	1.93	5.41	0.00	7.34	-0.34
(Sr, Cl)-90 °	Sr (M)	2.11	5.99	0.50	8.60	1.40
	O1	1.91	5.24	0.00	7.15	-1.15
	O2	1.81	4.86	0.00	6.67	-0.67
	C1	0.90	2.43	0.00	3.34	0.66
	C2	1.06	2.96	0.00	4.02	-0.02
	C3	1.09	2.93	0.00	4.02	-0.02
	Cl (Z)	1.90	5.10	0.00	7.00	0.00
(Sr, Br)-90 °	Sr (M)	2.13	5.99	0.51	8.64	1.36
	O1	1.91	5.24	0.00	7.14	-1.14
	O2	1.81	4.86	0.00	6.66	-0.66
	C1	0.92	2.45	0.00	3.36	0.64
	C2	1.07	2.96	0.00	4.03	-0.03
	C3	1.14	2.96	0.00	4.09	-0.09
	Br (Z)	1.89	5.02	0.00	6.91	0.09
(Sr, I)-90 °	Sr (M)	2.13	5.99	0.51	8.64	1.36
	O1	1.91	5.23	0.00	7.14	-1.14
	O2	1.81	4.85	0.00	6.66	-0.66
	C1	0.92	2.45	0.00	3.38	0.62
	C2	1.07	2.95	0.00	4.03	-0.03
	C3	1.17	3.01	0.00	4.18	-0.18
	I (Z)	1.89	4.93	0.00	6.82	0.18

Table S14. The calculated Hirschfeld charge (HC, given in terms of *e*), bond overlap populations (BOP) for (Ba, Z)-90° (Z = F, Cl, Br, and I) as well as Ba-MOF-5, i.e., (Ba, H)-0 °. Note that the atomic labels of atoms are numbered according to Figure 1.

Materials	Atomic site	HC (*e*)	BOP
(Ba, H)-0 °	Ba (M)	0.53	0.11-0.16 (Ba-O)
	O1	-0.54	0.16 (O1-Ba)
	O2	-0.28	0.11 (O2-Ba)
	C1	0.18	0.89 (C1-O2)
			0.81 (C1-C2)
	C2	-0.01	1.08 (C2-C3)
	C3	-0.04	1.09 (C3-C3)
	H (Z)	0.04	0.86 (H-C3)
(Ba, F)-90 °	Ba (M)	0.55	0.10-0.16 (Ba-O)
	O1	-0.54	0.16 (O1-Ba)
	O2	-0.27	0.10 (O2-Ba)
	C1	0.18	0.91 (C1-O2)
			0.72 (C1-C2)
	C2	-0.04	1.11 (C2-C3)
	C3	0.06	1.12 (C3-C3)
	F (Z)	-0.08	0.46 (F-C3)
(Ba, Cl)-90 °	Ba (M)	0.52	0.10-0.16 (Ba-O)
	O1	-0.54	0.16 (O1-Ba)
	O2	-0.27	0.10 (O2-Ba)
	C1	0.16	0.91 (C1-O2)
			0.75 (C1-C2)
	C2	-0.03	1.12 (C2-C3)
	C3	0.00	1.10 (C3-C3)
	Cl (Z)	0.01	0.58 (Cl-C3)
(Ba, Br)-90 °	Ba (M)	0.51	0.11-0.16 (Ba-O)
	O1	-0.54	0.16 (O1-Ba)
	O2	-0.27	0.11 (O2-Ba)
	C1	0.16	0.92 (C1-O2)
			0.77 (C1-C2)
	C2	-0.03	1.14 (C2-C3)
	C3	-0.01	1.15 (C3-C3)
	Br (Z)	0.03	0.50 (Br-C3)
(Ba, I)-90 °	Ba (M)	0.48	0.11-0.15 (Ba-O)
	O1	-0.54	0.15 (O1-Ba)
	O2	-0.27	0.11 (O2-Ba)
	C1	0.15	0.92 (C1-O2)
			0.78 (C1-C2)
	C2	-0.02	1.14 (C2-C3)
	C3	-0.03	1.15 (C3-C3)
	I (Z)	0.06	0.50 (I-C3)

Table S15. The electron configurations of atoms for (Ba, Z)-90° (Z = F, Cl, Br, and I) as well as (Ba, H)-0° from atomic populations (Mulliken) with CASTEP code. Note that the atomic labels of atoms are numbered according to Figure 1.

Materials	Atom	*s*	*p*	*d*	Total	Charge (e)
(Ba, H)-0 °	Ba (M)	2.08	5.98	0.57	8.64	1.36
	O1	1.91	5.17	0.00	7.08	-1.08
	O2	1.81	4.89	0.00	6.69	-0.69
	C1	0.92	2.44	0.00	3.36	0.64
	C2	1.09	2.96	0.00	4.06	-0.06
	C3	1.18	3.09	0.00	4.27	-0.27
	H (Z)	0.70	0.00	0.00	0.70	0.30
(Ba, F)-90 °	Ba (M)	2.08	5.99	0.54	8.61	1.39
	O1	1.92	5.18	0.00	7.09	-1.09
	O2	1.81	4.86	0.00	6.67	-0.67
	C1	0.90	2.43	0.00	3.33	0.67
	C2	1.06	3.04	0.00	4.10	-0.10
	C3	0.98	2.67	0.00	3.65	0.35
	F (Z)	1.93	5.42	0.00	7.34	-0.34
(Ba, Cl)-90 °	Ba (M)	2.08	5.99	0.54	8.62	1.38
	O1	1.92	5.18	0.00	7.09	-1.09
	O2	1.81	4.86	0.00	6.67	-0.67
	C1	0.90	2.43	0.00	3.33	0.67
	C2	1.06	2.96	0.00	4.02	-0.02
	C3	1.09	2.93	0.00	4.02	-0.02
	Cl (Z)	1.90	5.10	0.00	7.01	-0.01
(Ba, Br)-90 °	Ba (M)	2.10	5.99	0.55	8.64	1.36
	O1	1.92	5.17	0.00	7.09	-1.09
	O2	1.81	4.85	0.00	6.66	-0.66
	C1	0.91	2.45	0.00	3.36	0.64
	C2	1.07	2.96	0.00	4.03	-0.03
	C3	1.13	2.96	0.00	4.09	-0.09
	Br (Z)	1.89	5.02	0.00	6.91	0.09
(Ba, I)-90 °	Ba (M)	2.10	5.99	0.56	8.65	1.35
	O1	1.91	5.17	0.00	7.09	-1.09
	O2	1.81	4.85	0.00	6.66	-0.66
	C1	0.91	2.46	0.00	3.37	0.63
	C2	1.08	2.95	0.00	4.03	-0.03
	C3	1.17	3.01	0.00	4.17	-0.17
	I (Z)	1.90	4.93	0.00	6.83	0.17

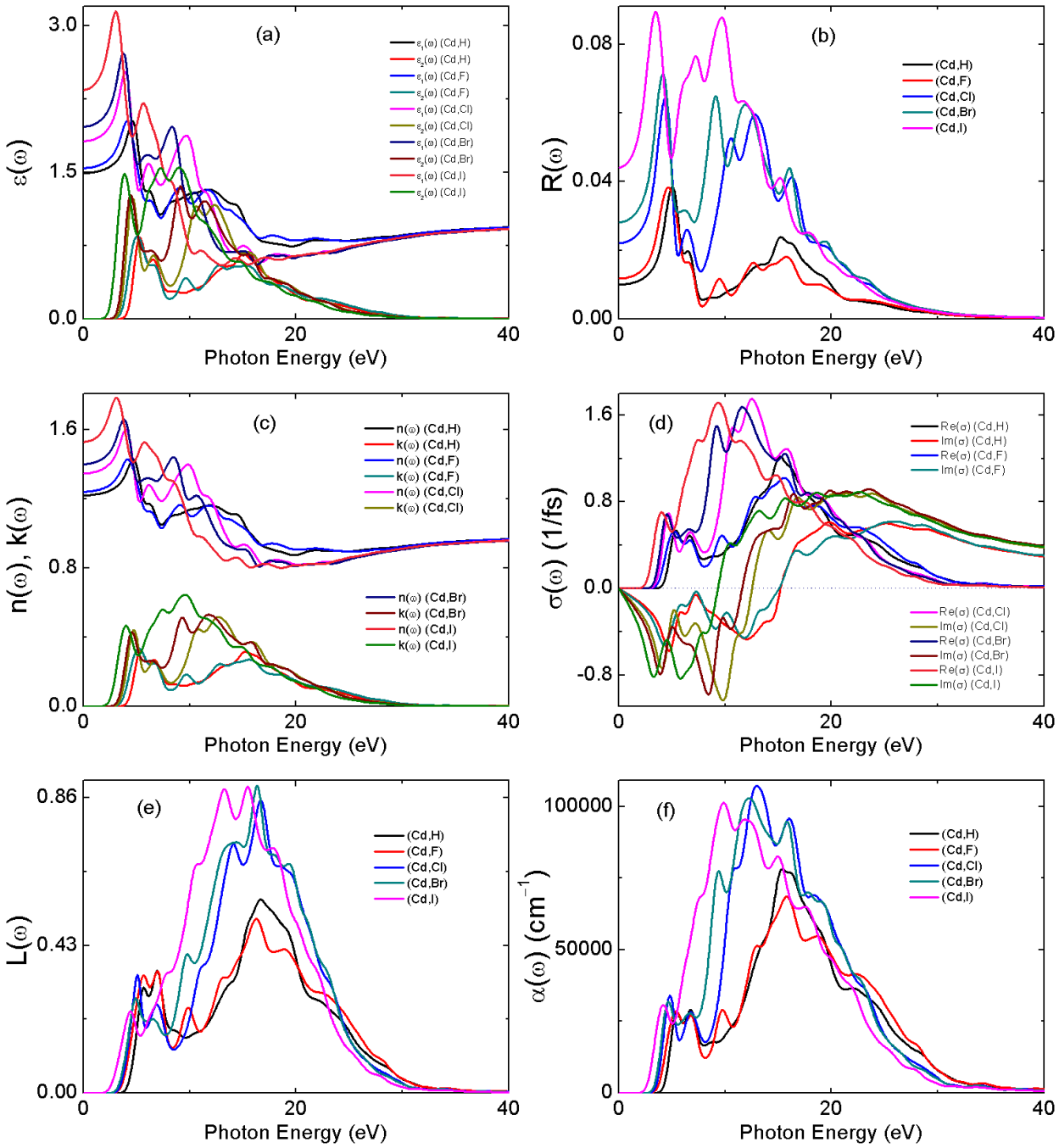

Figure S36. Calculated optical properties for (Cd, Z)-90 °, (Z = F, Cl, Br, I) as well as (Cd, H)-0 °: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

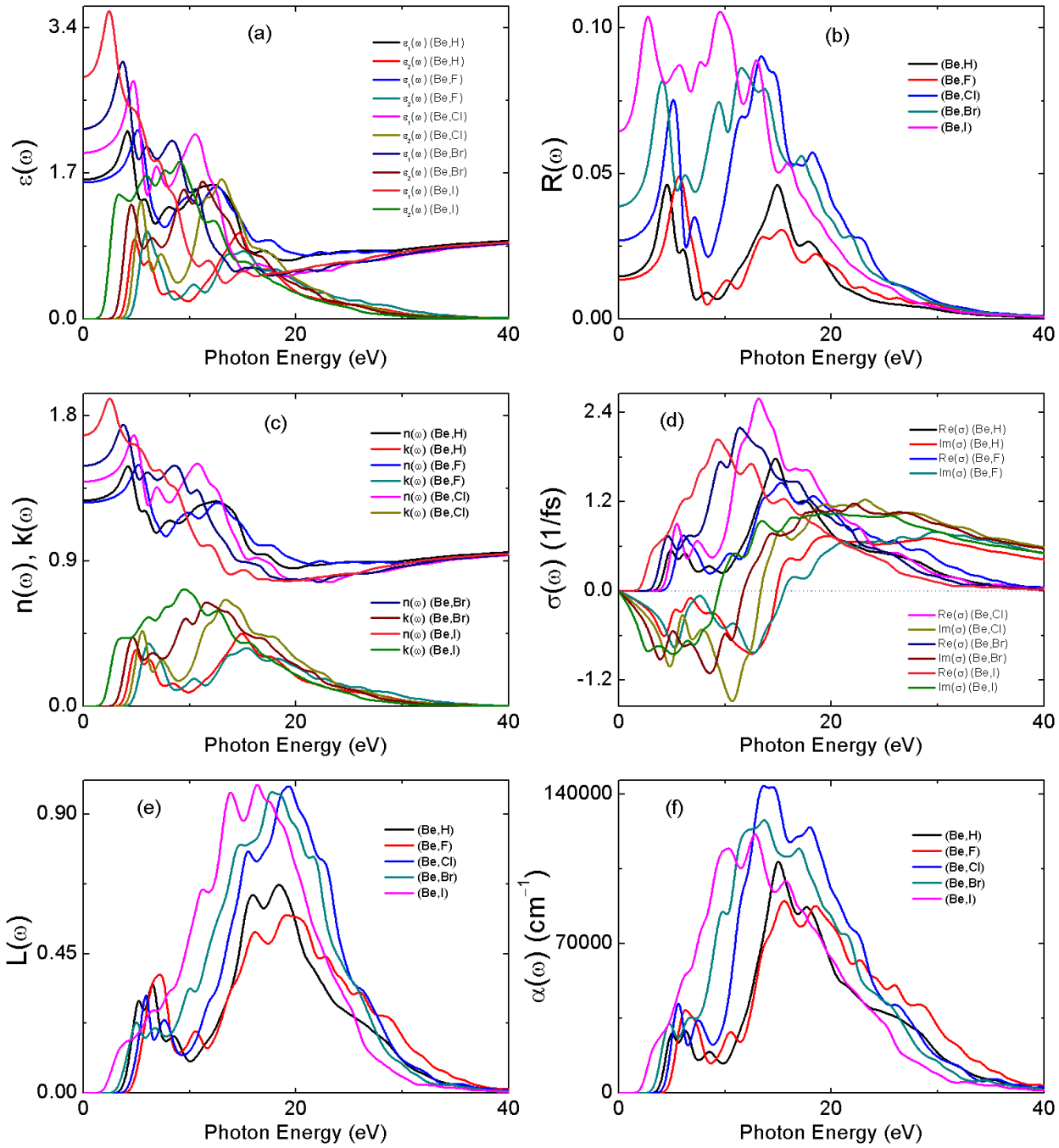

Figure S37. Calculated optical properties for (Be, Z)-90 °, (Z = F, Cl, Br, I) as well as (Be, H)-0 °: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

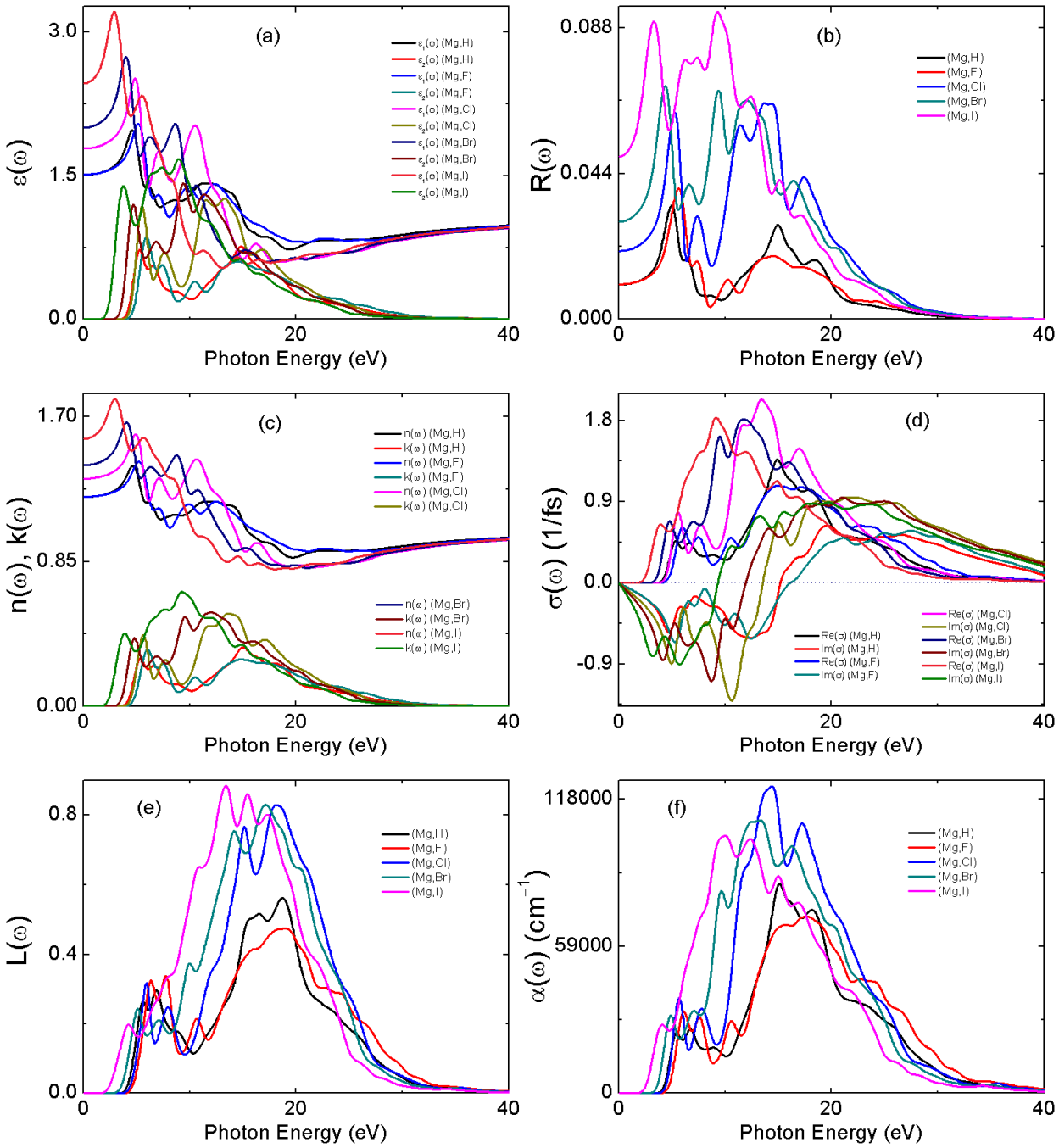

Figure S38. Calculated optical properties for (Mg, Z)-90 °, (Z = F, Cl, Br, I) as well as (Mg, H)-0 °: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

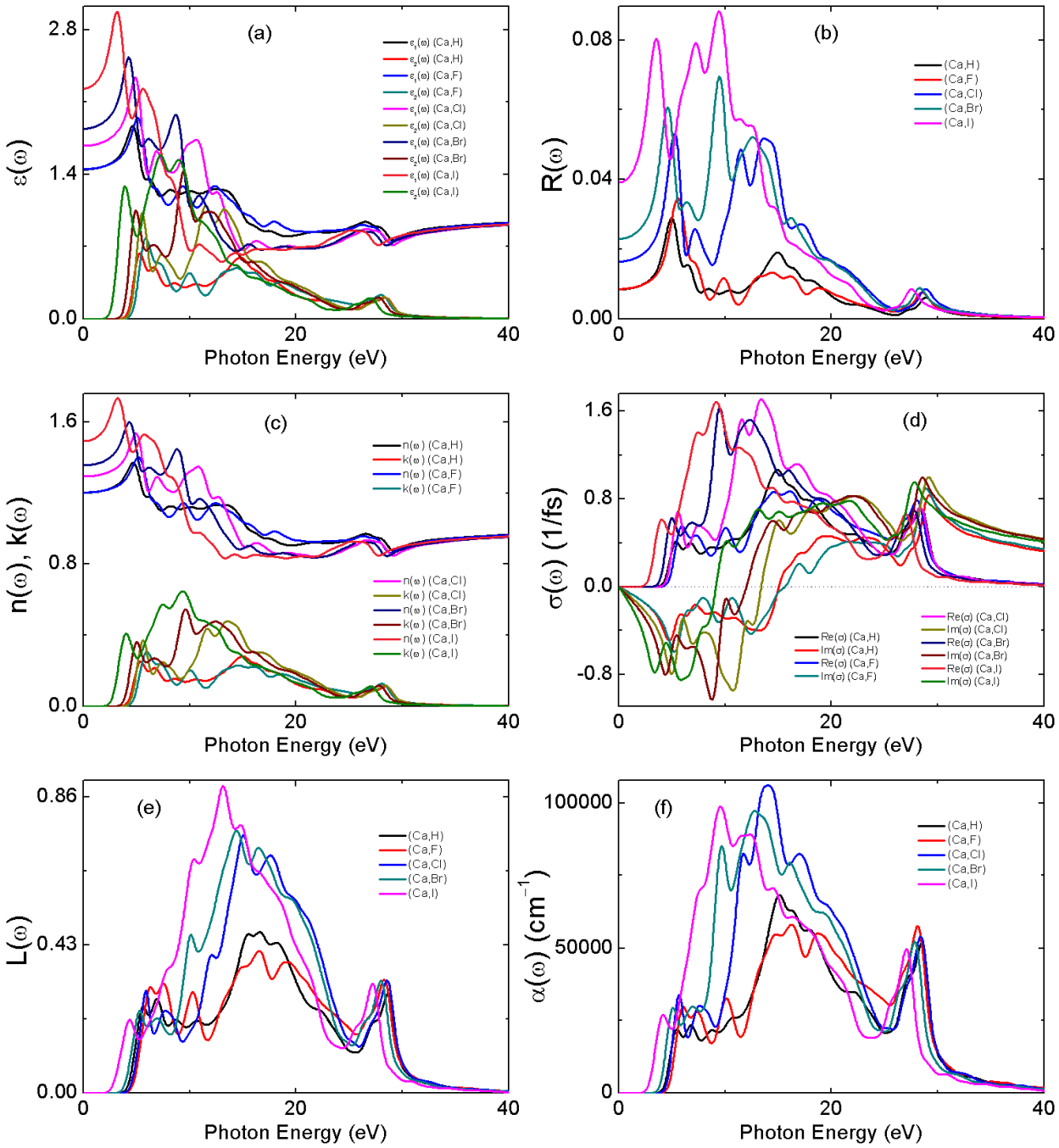

Figure S39. Calculated optical properties for (Ca, Z)-90 °; (Z = F, Cl, Br, I) as well as (Ca, H)-0 °: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

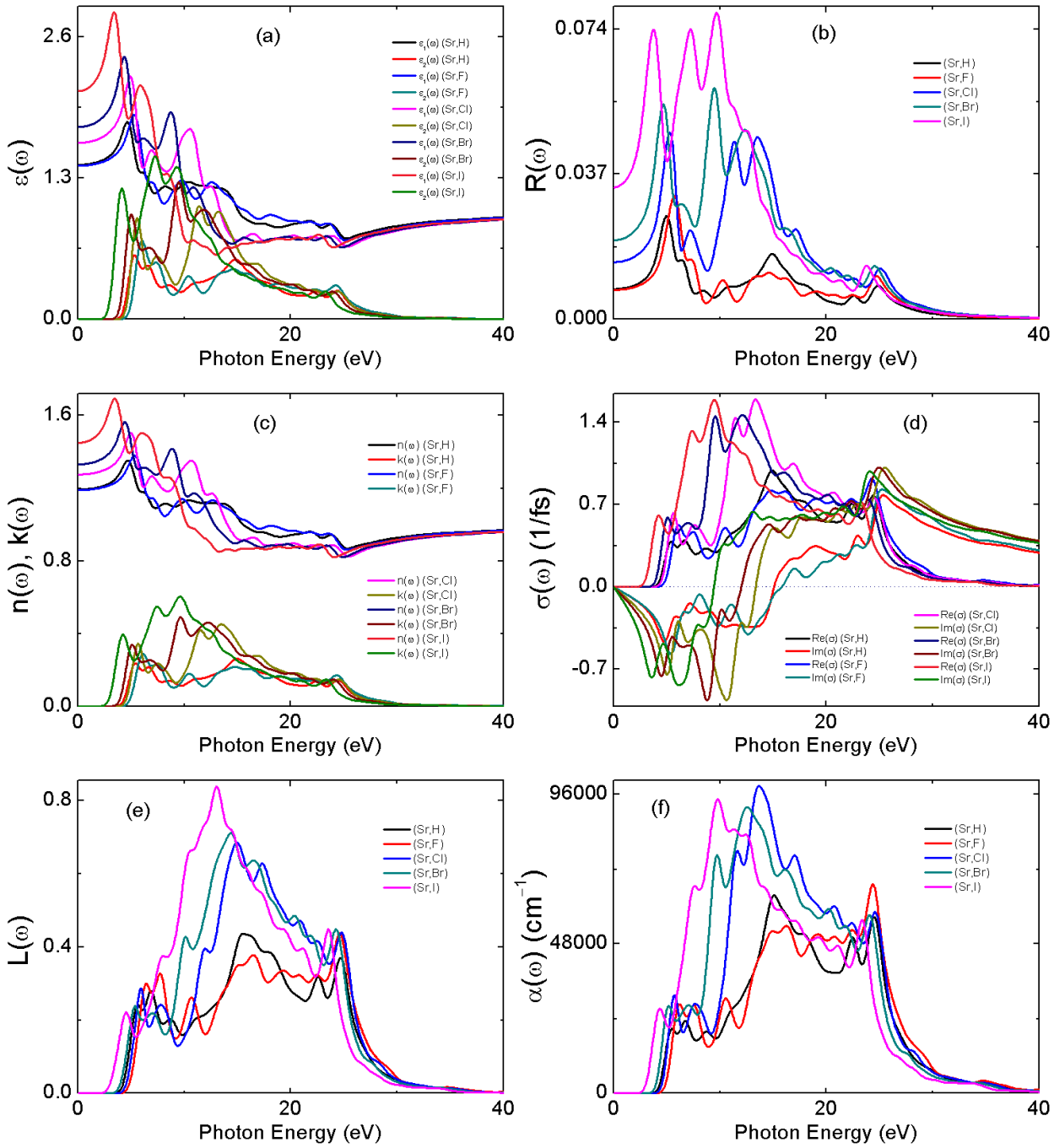

Figure S40. Calculated optical properties for (Sr, Z)-90 °, (Z = F, Cl, Br, I) as well as (Sr, H)-0 °: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

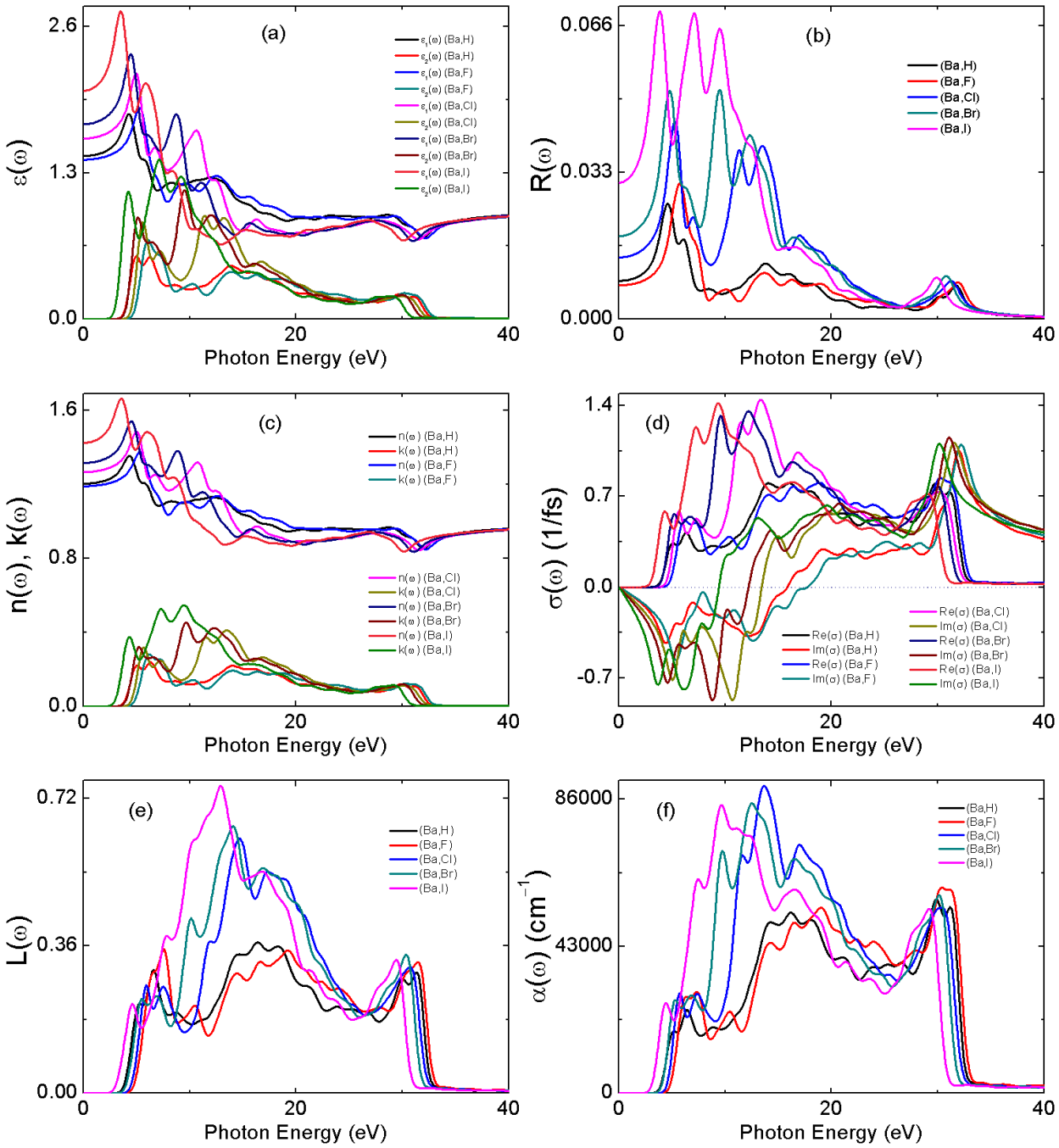

Figure S41. Calculated optical properties for (Ba, Z)-90 °, (Z = F, Cl, Br, I) as well as (Ba, H)-0 °: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

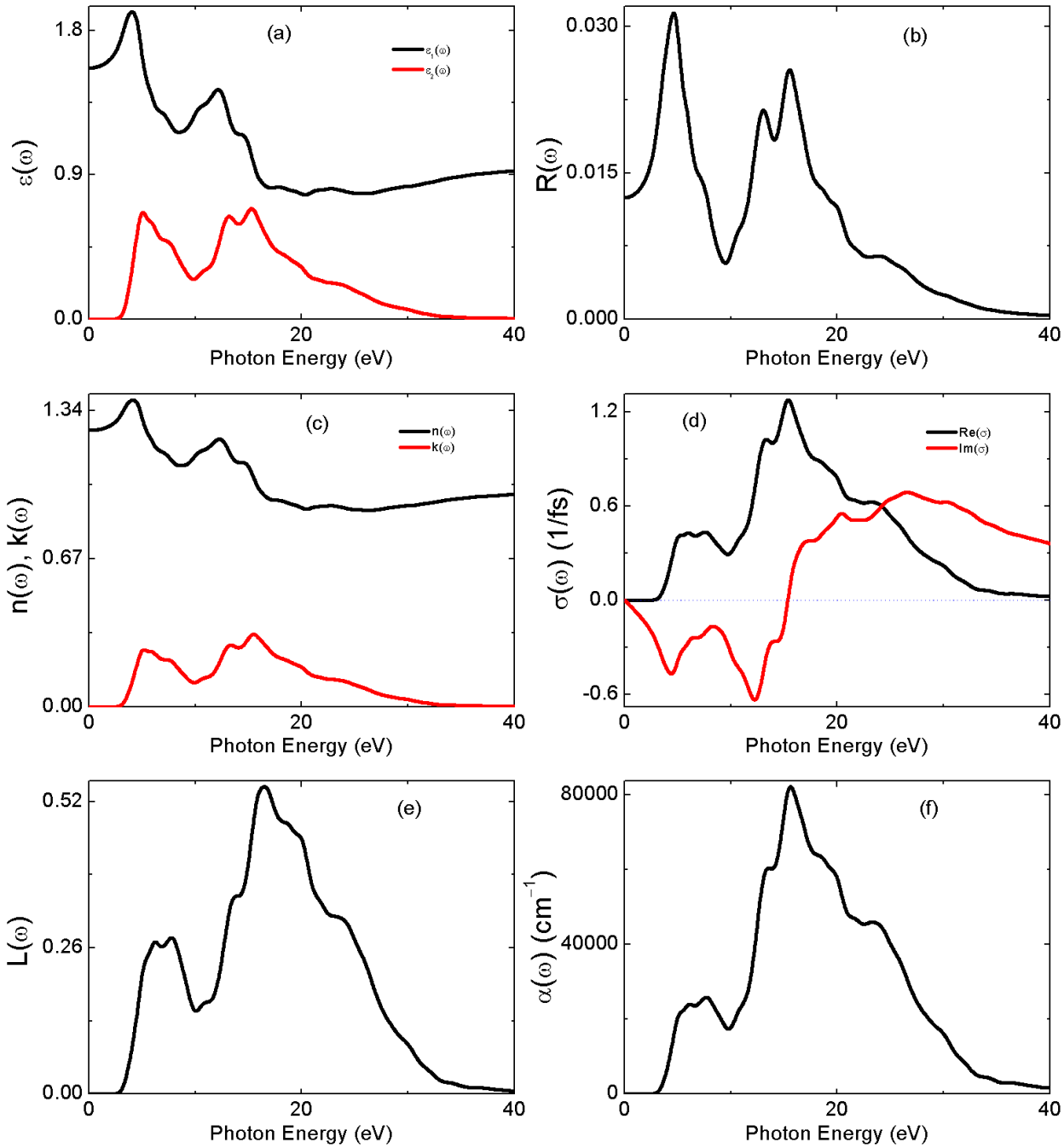

Figure S42. Calculated optical properties for (Zn, F)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

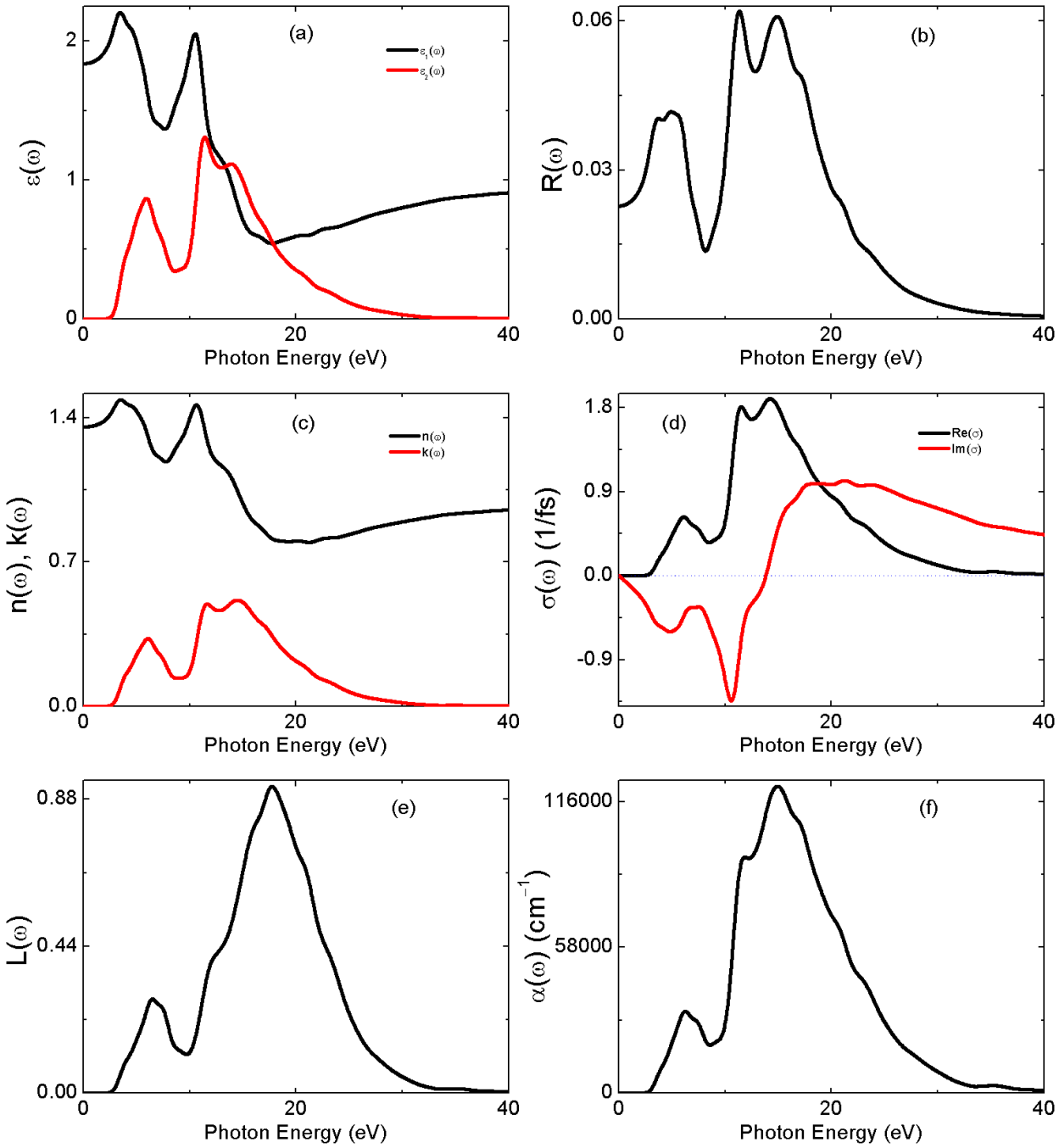

Figure S43. Calculated optical properties for (Zn, Cl)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

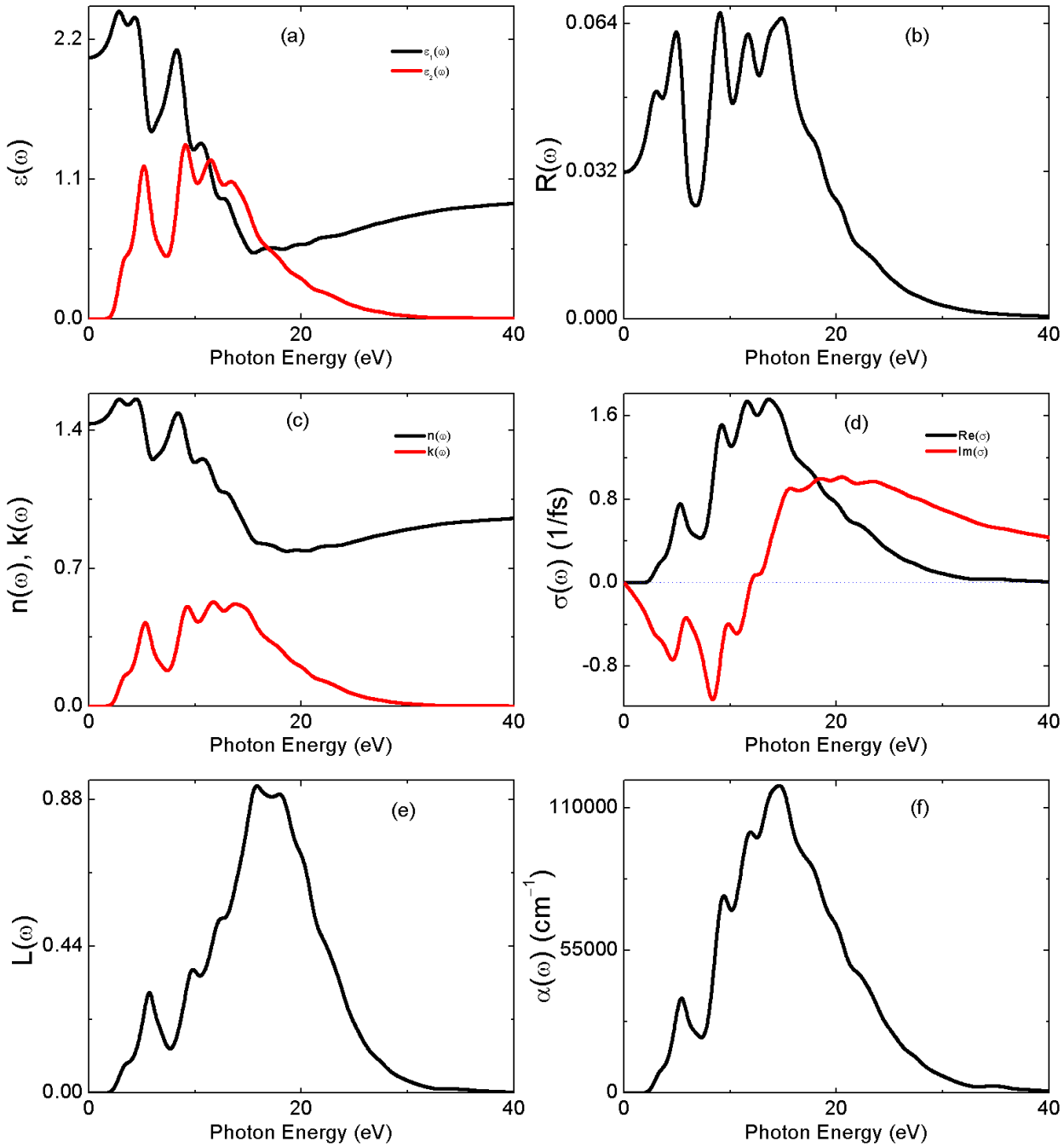

Figure S44. Calculated optical properties for (Zn, Br)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

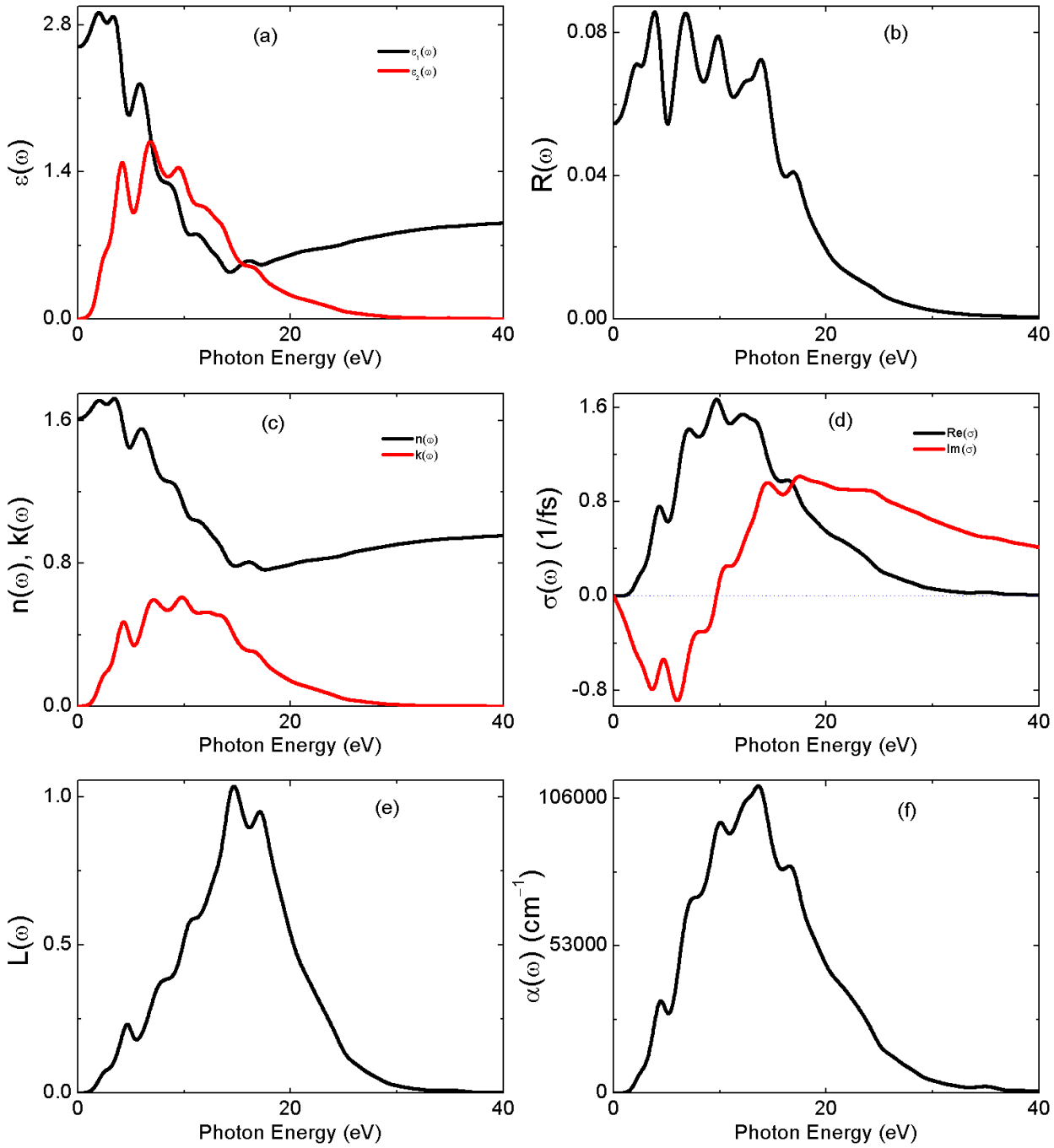

Figure S45. Calculated optical properties for (Zn, I)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

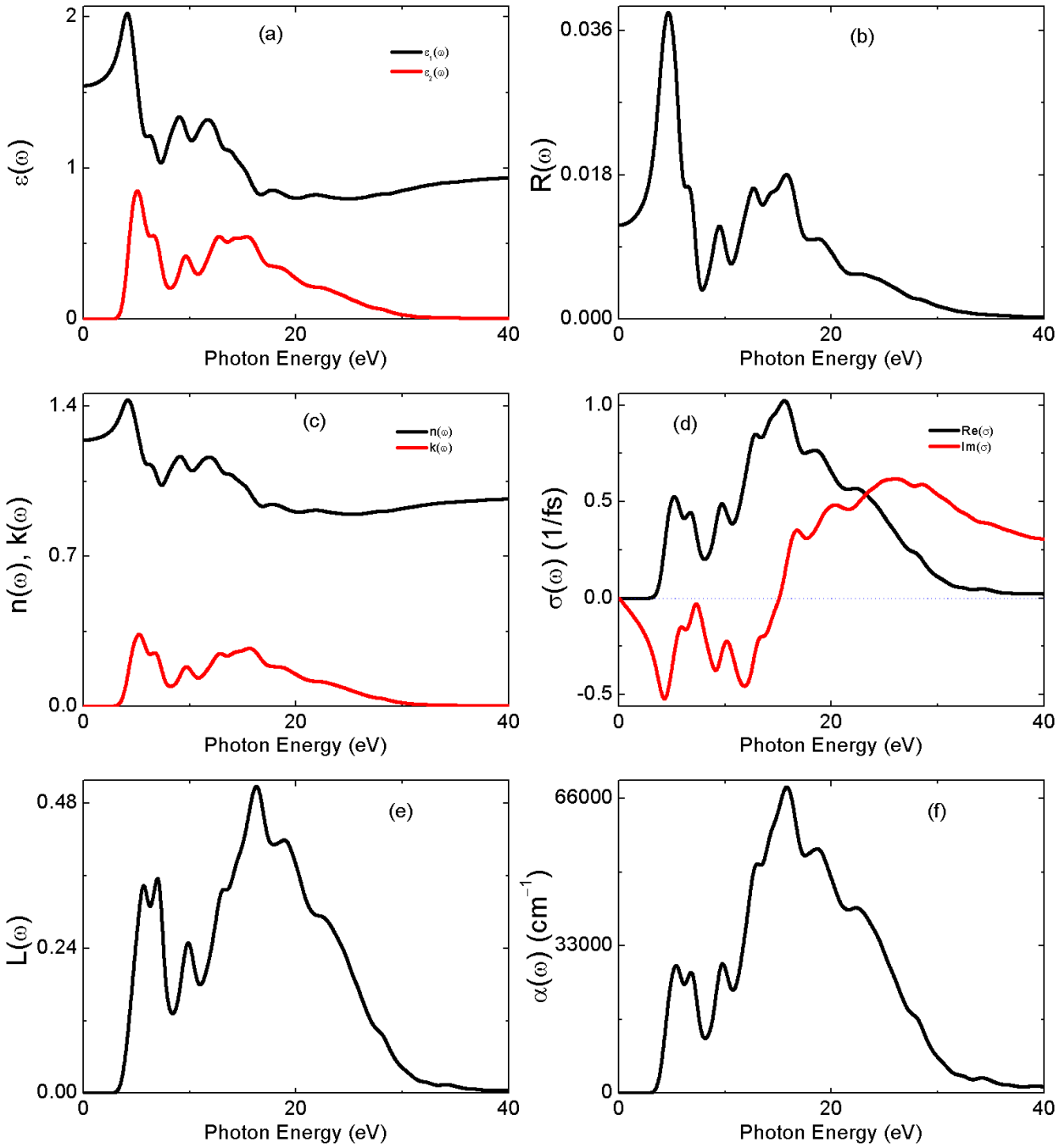

Figure S46. Calculated optical properties for (Cd, F)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

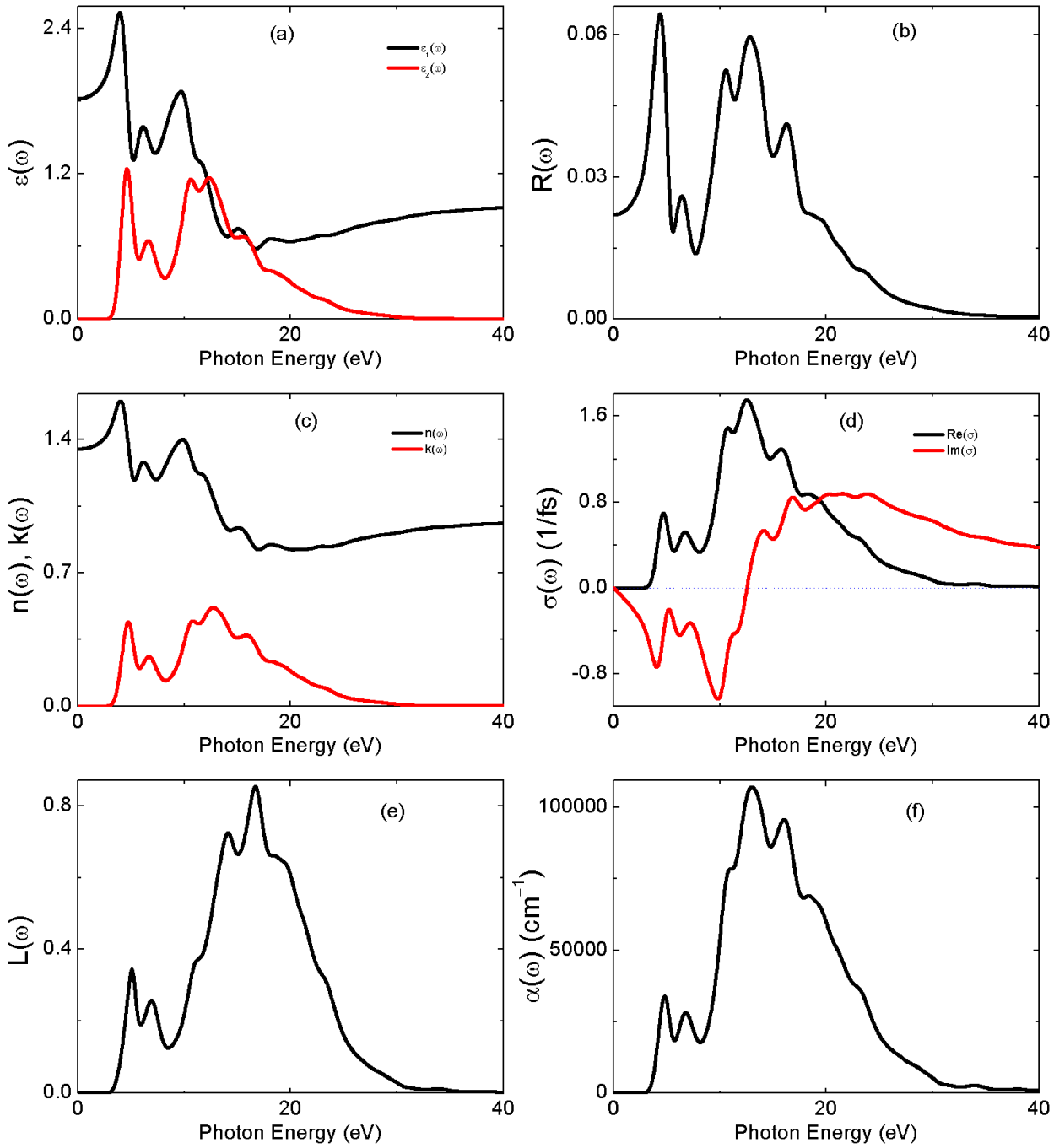

Figure S47. Calculated optical properties for (Cd, Cl)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

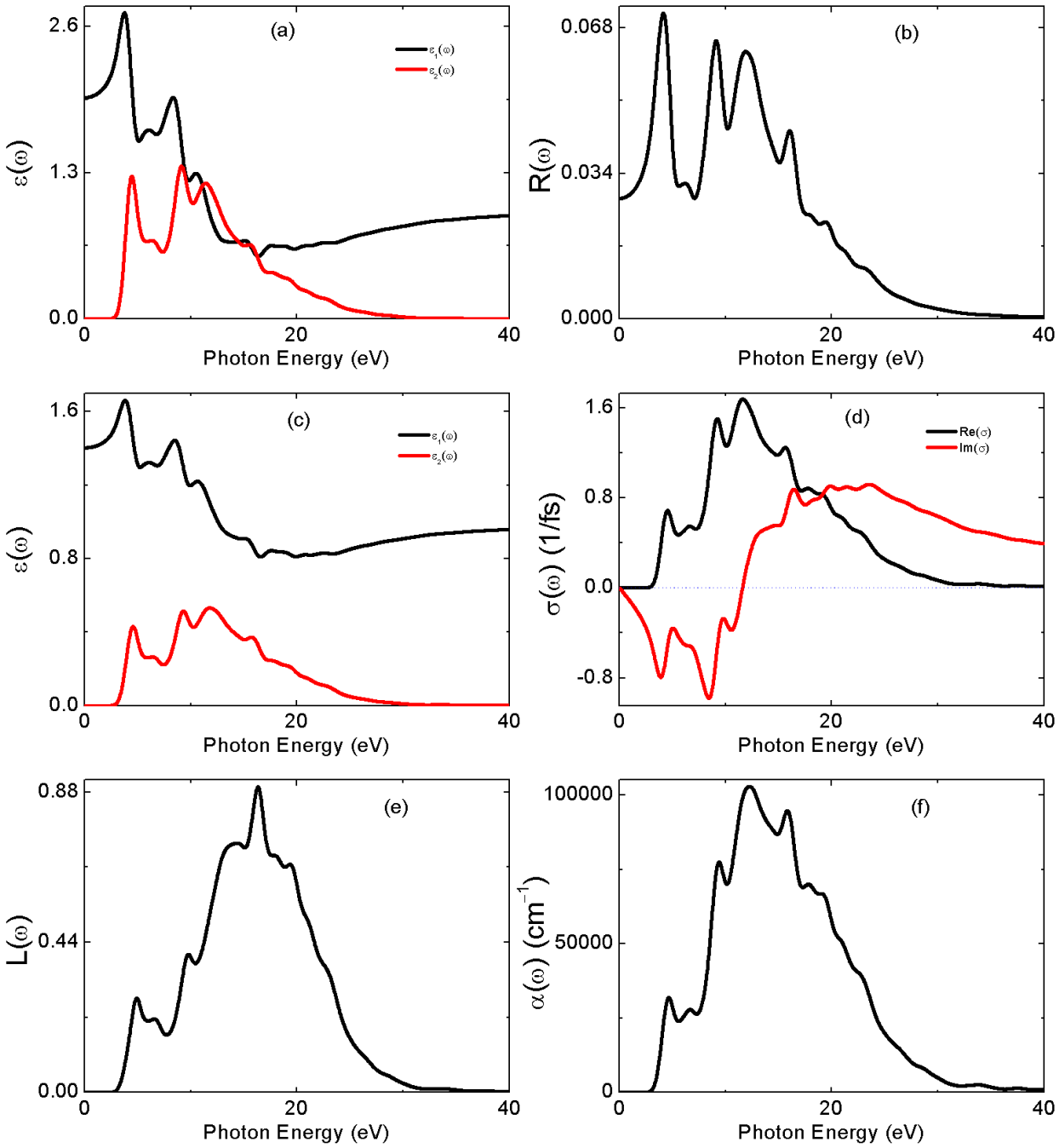

Figure S48. Calculated optical properties for (Cd, Br)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

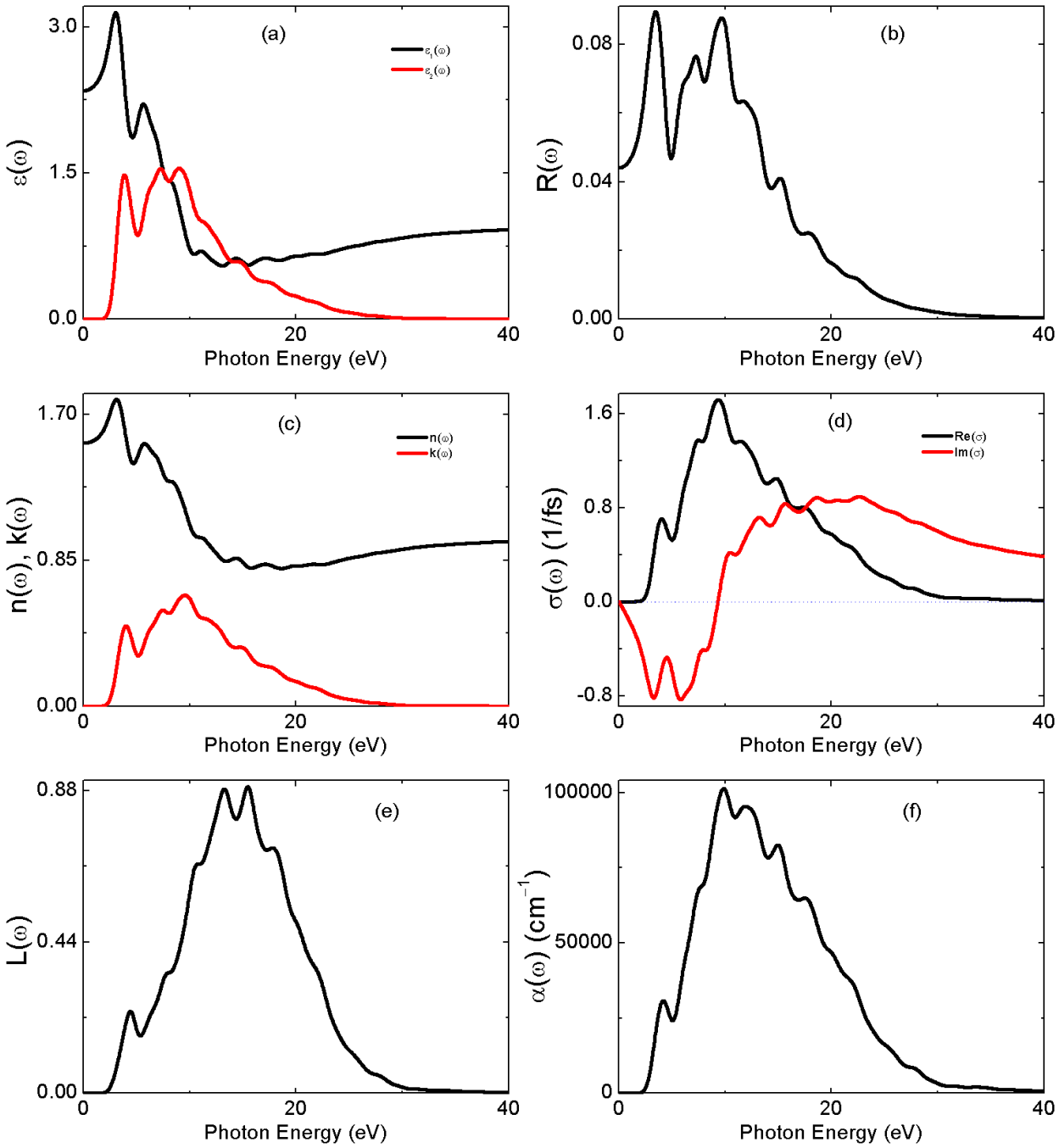

Figure S49. Calculated optical properties for (Cd, I)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

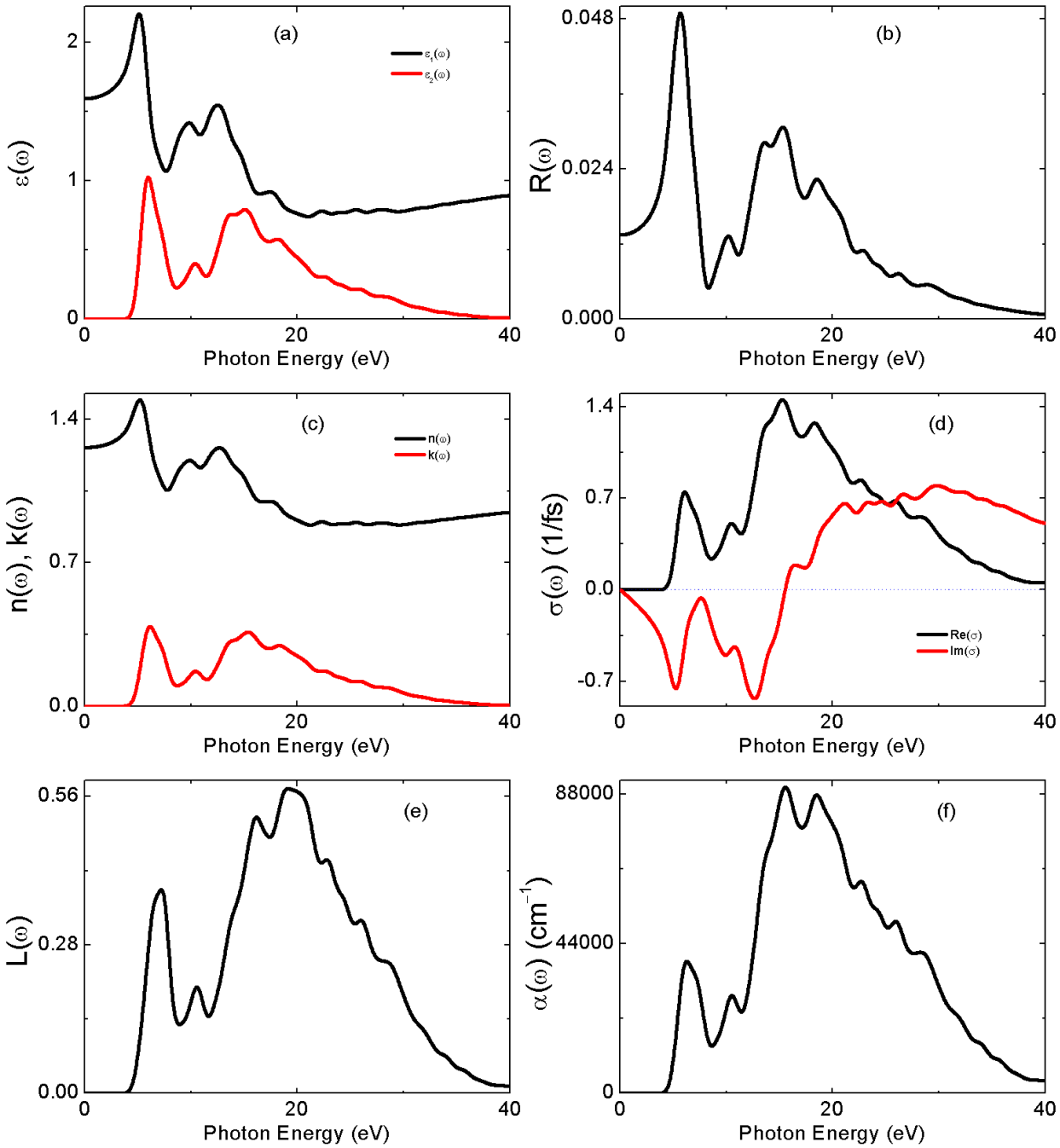

Figure S50. Calculated optical properties for (Be, F)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

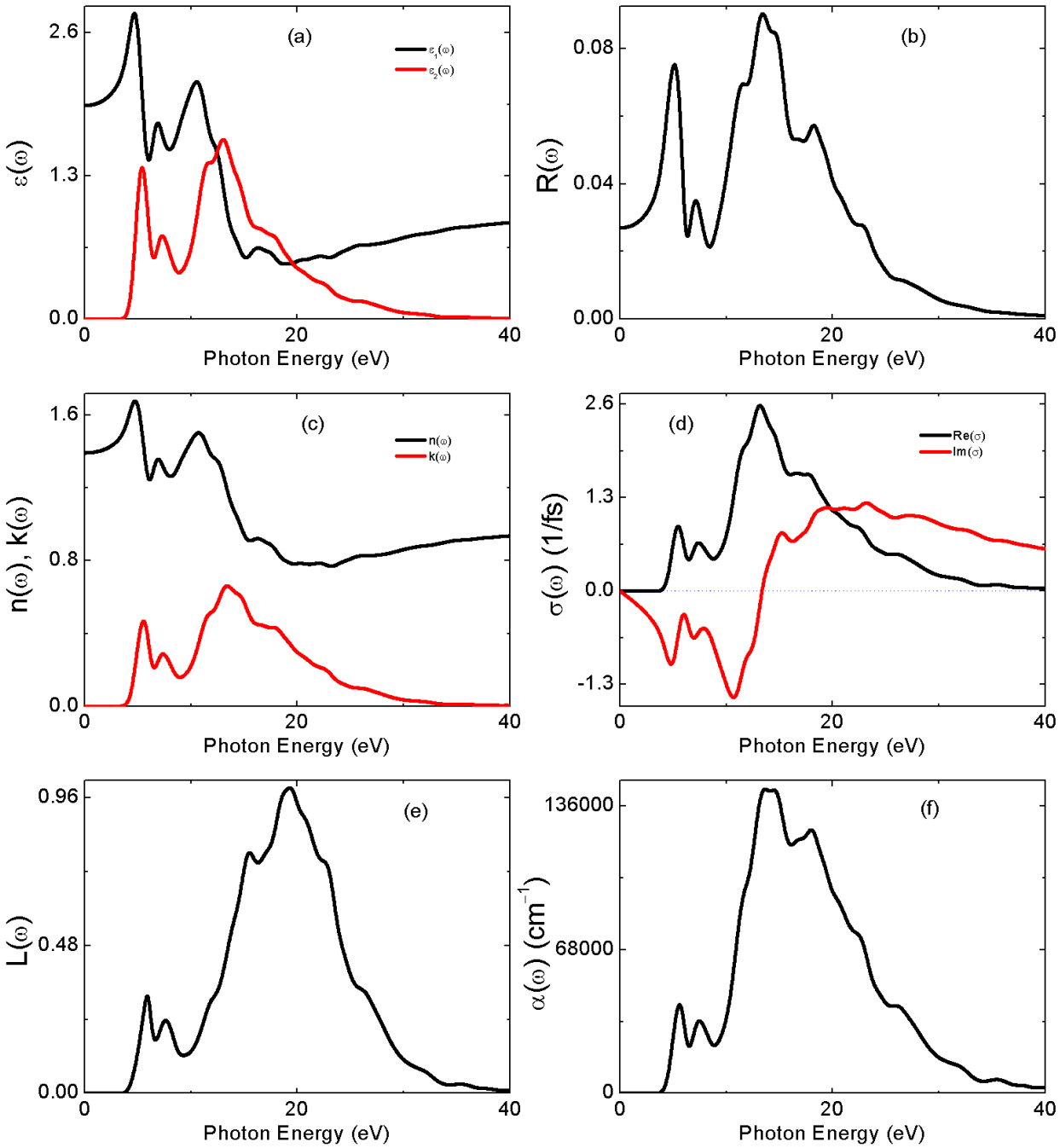

Figure S51. Calculated optical properties for (Be, Cl)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

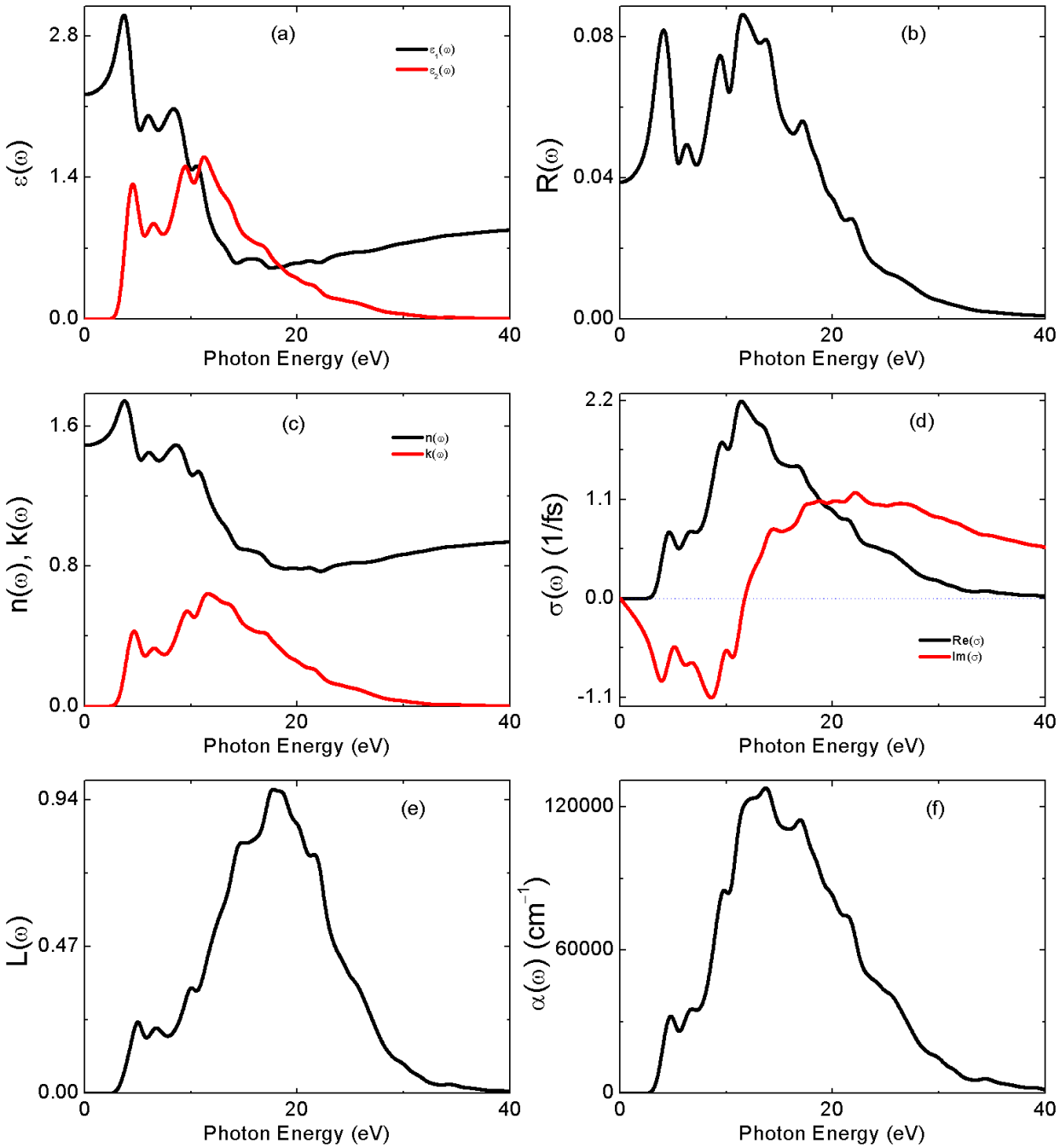

Figure S52. Calculated optical properties for (Be, Br)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

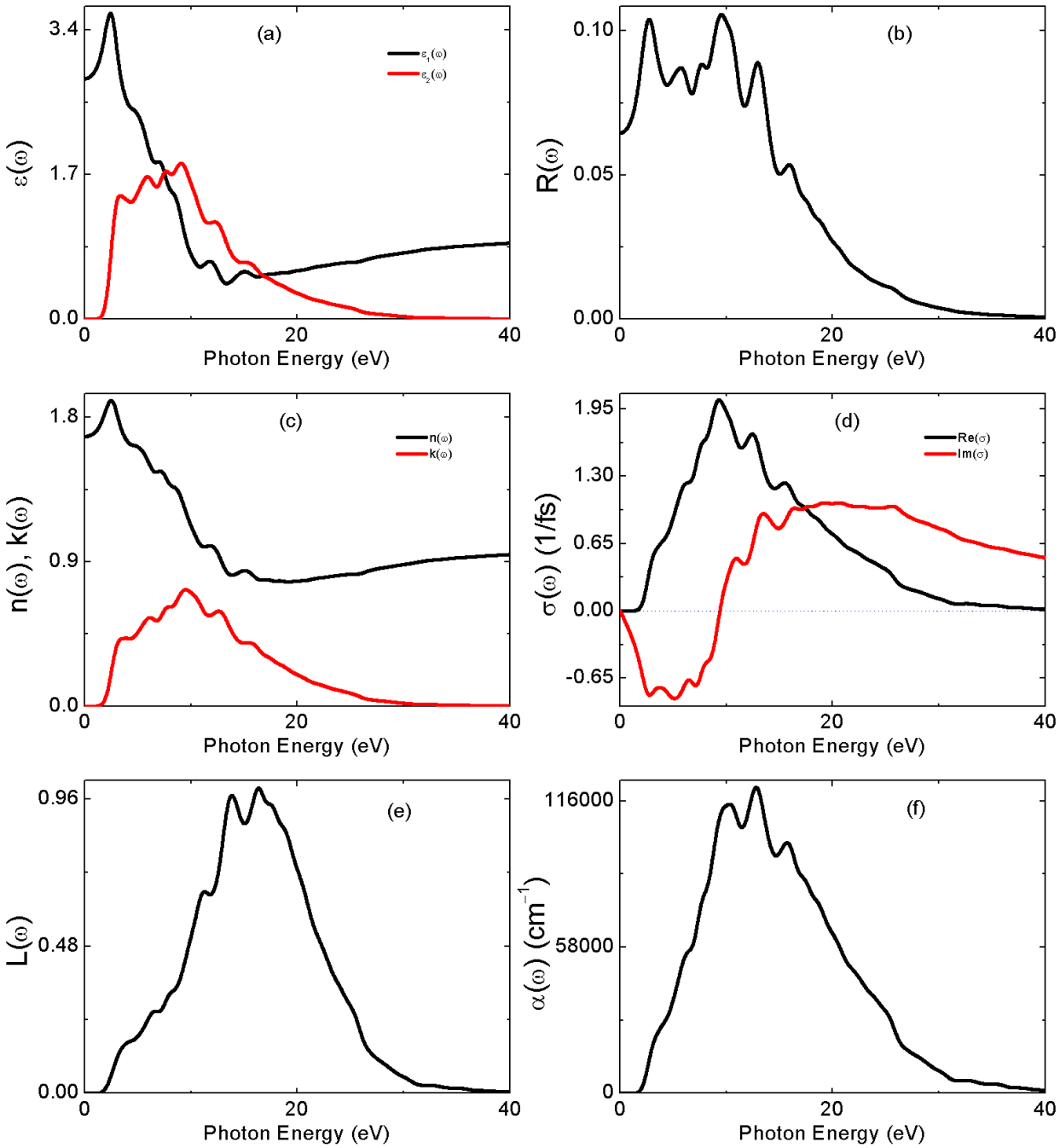

Figure S53. Calculated optical properties for (Be, I)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

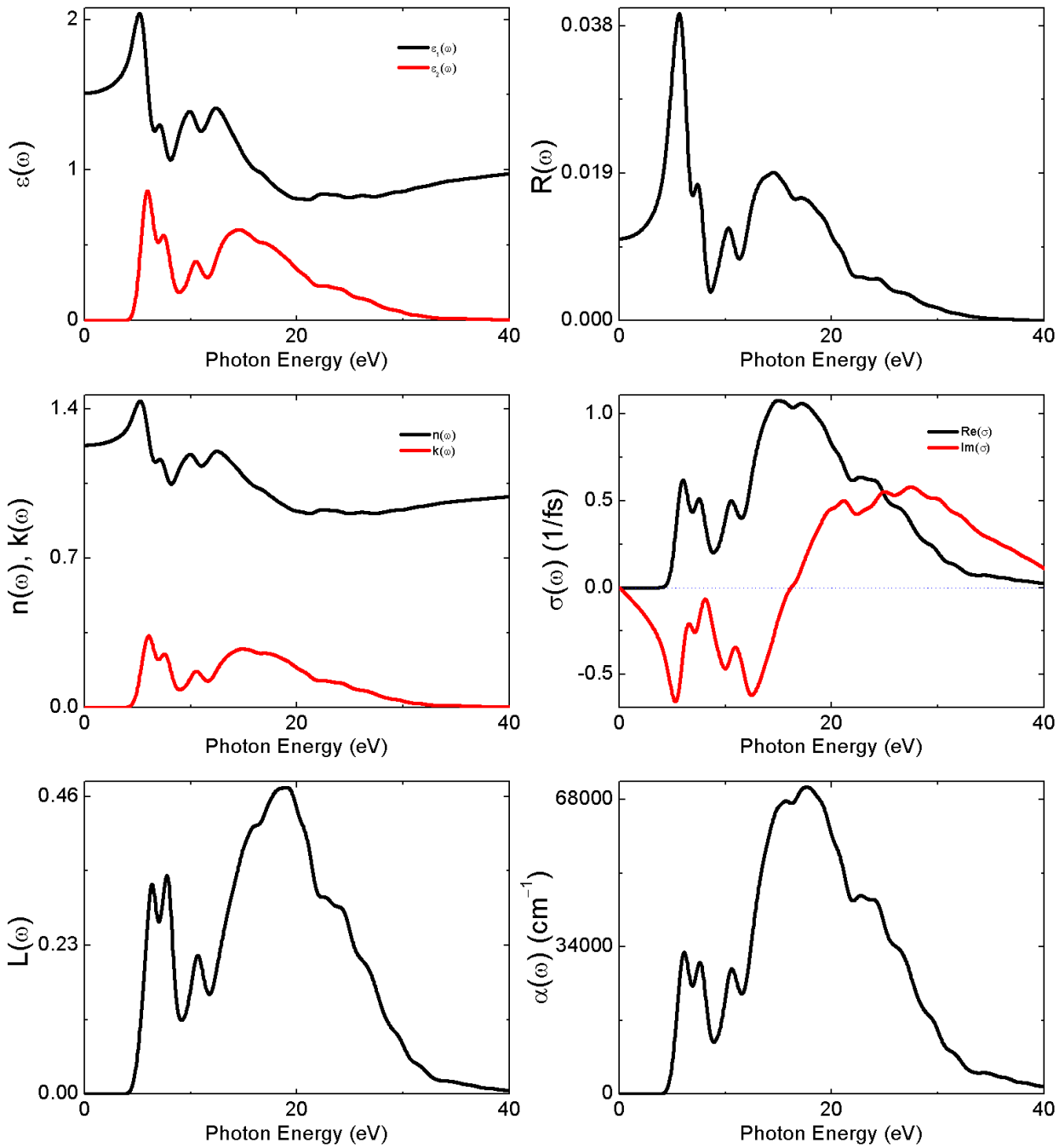

Figure S54. Calculated optical properties for (Mg, F)-90 °: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

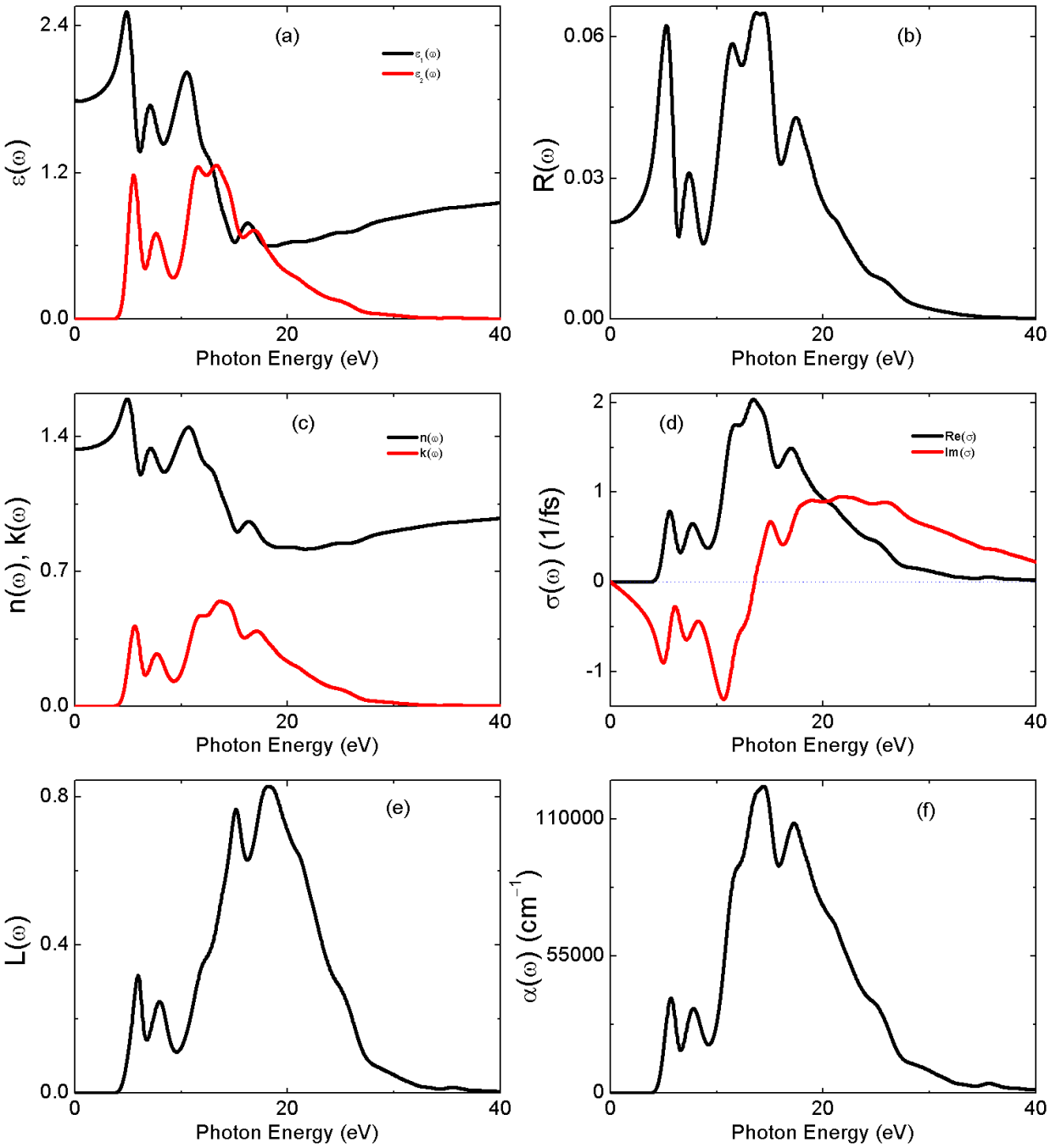

Figure S55. Calculated optical properties for (Mg, Cl)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

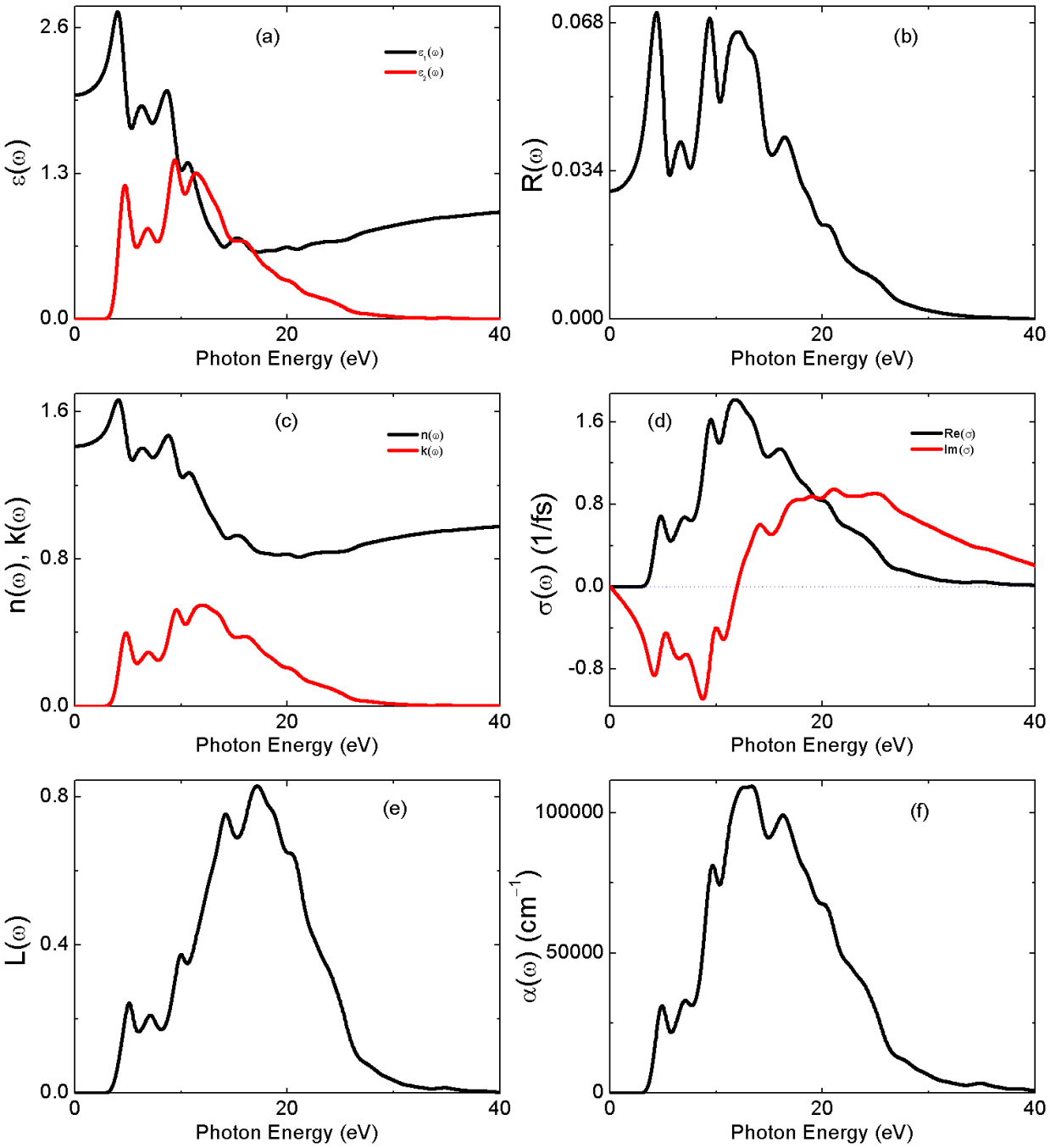

Figure S56. Calculated optical properties for (Mg, Br)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

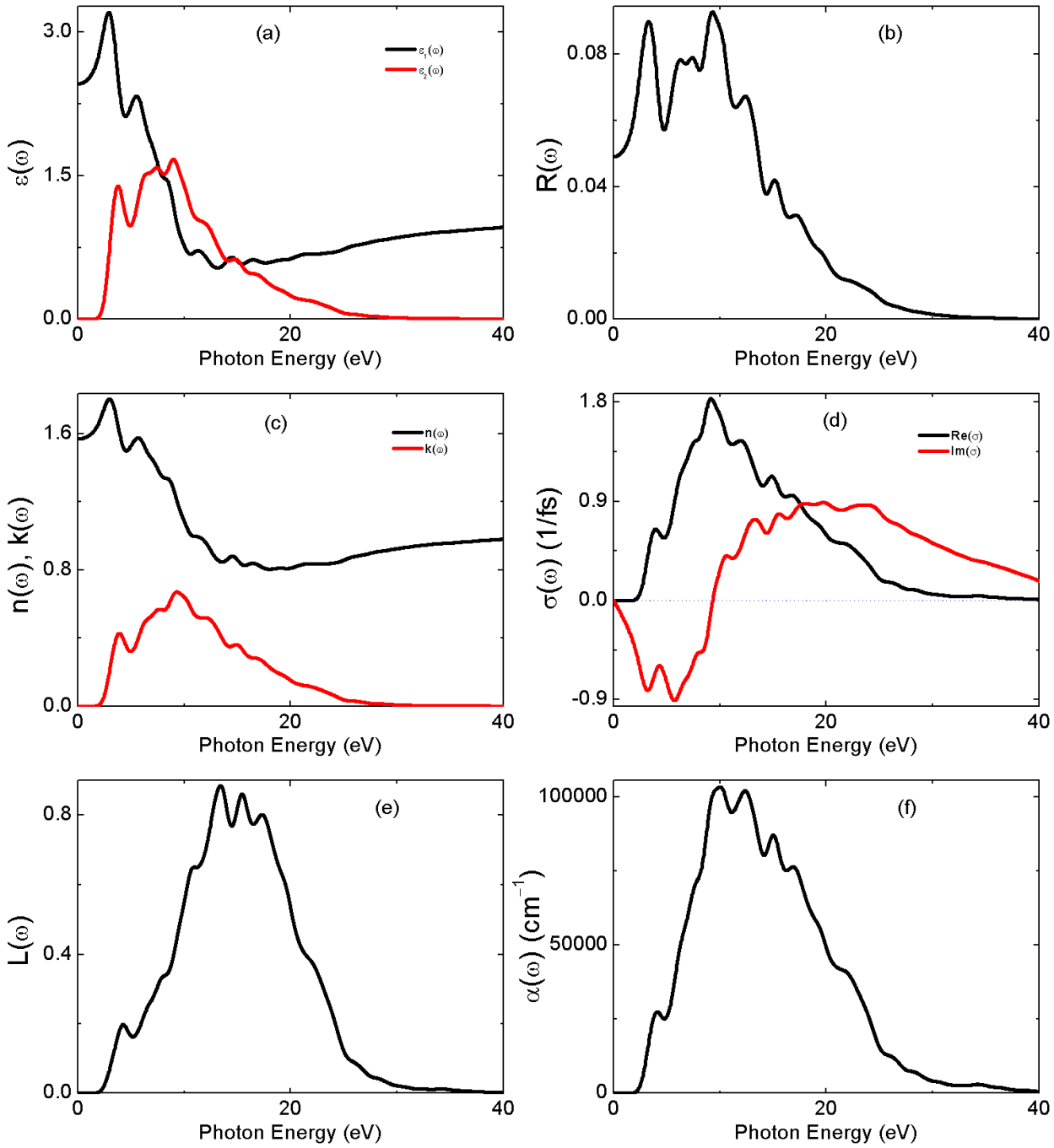

Figure S57. Calculated optical properties for (Mg, I)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

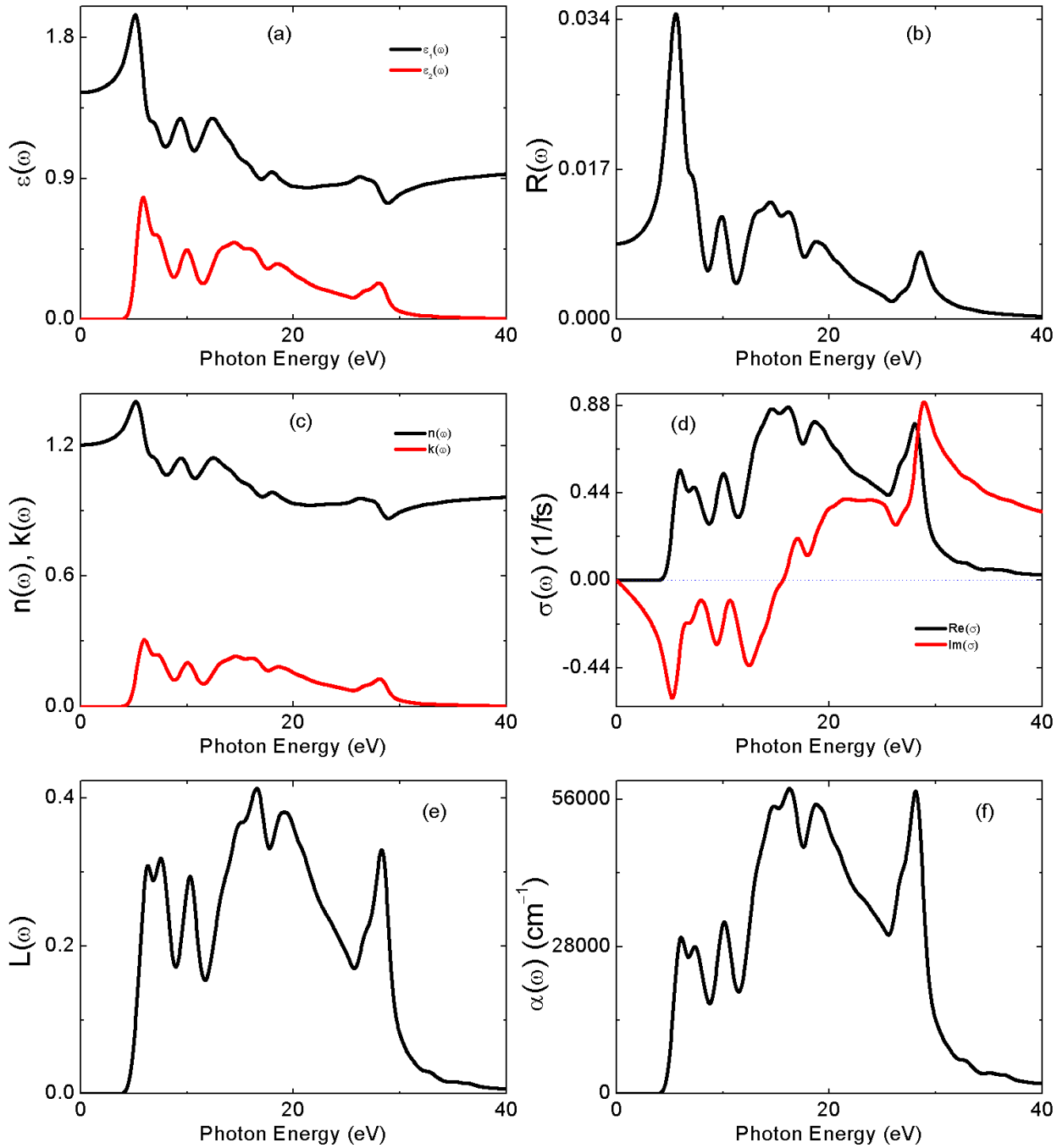

Figure S58. Calculated optical properties for (Ca, F)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

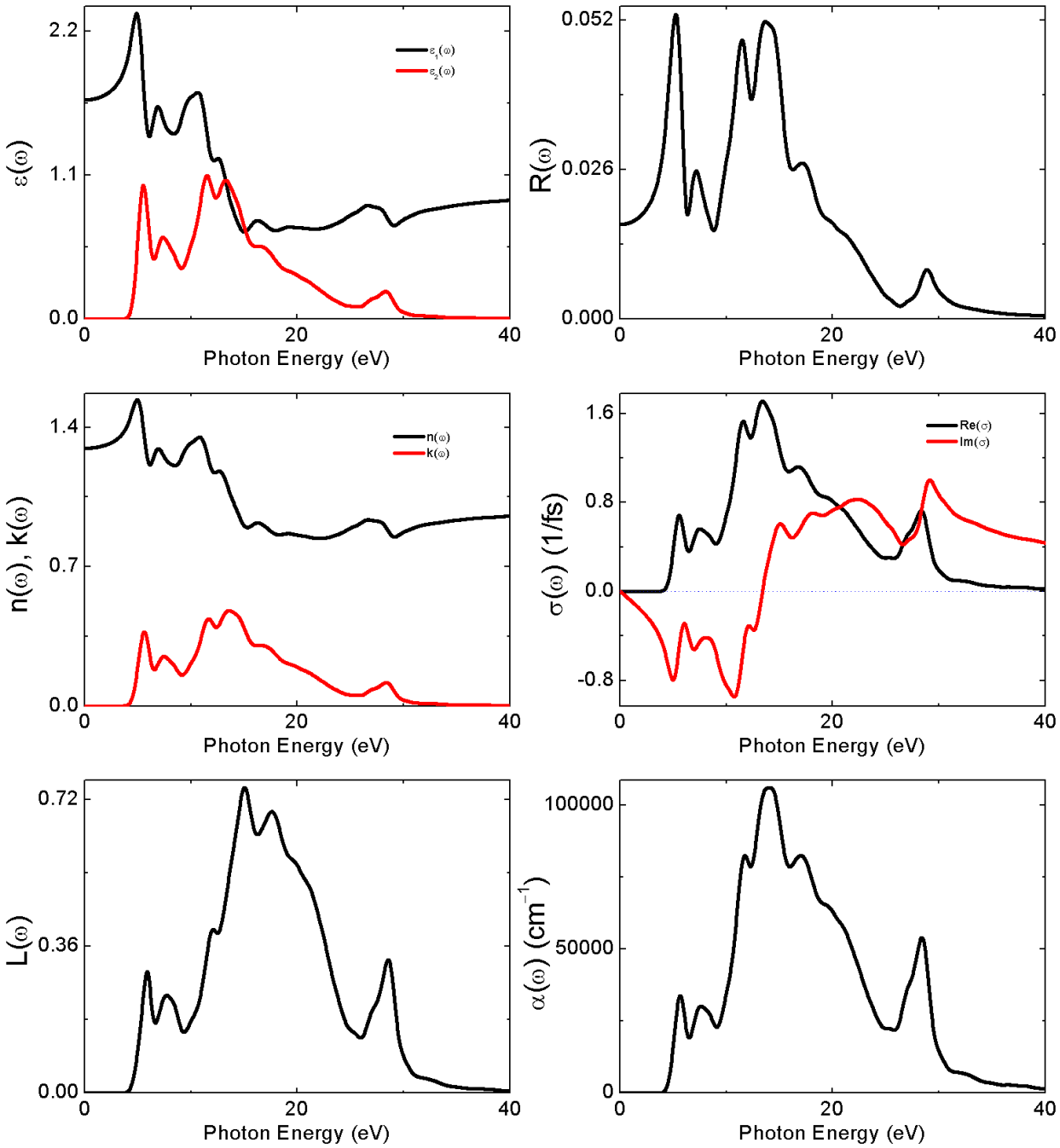

Figure S59. Calculated optical properties for (Ca, Cl)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

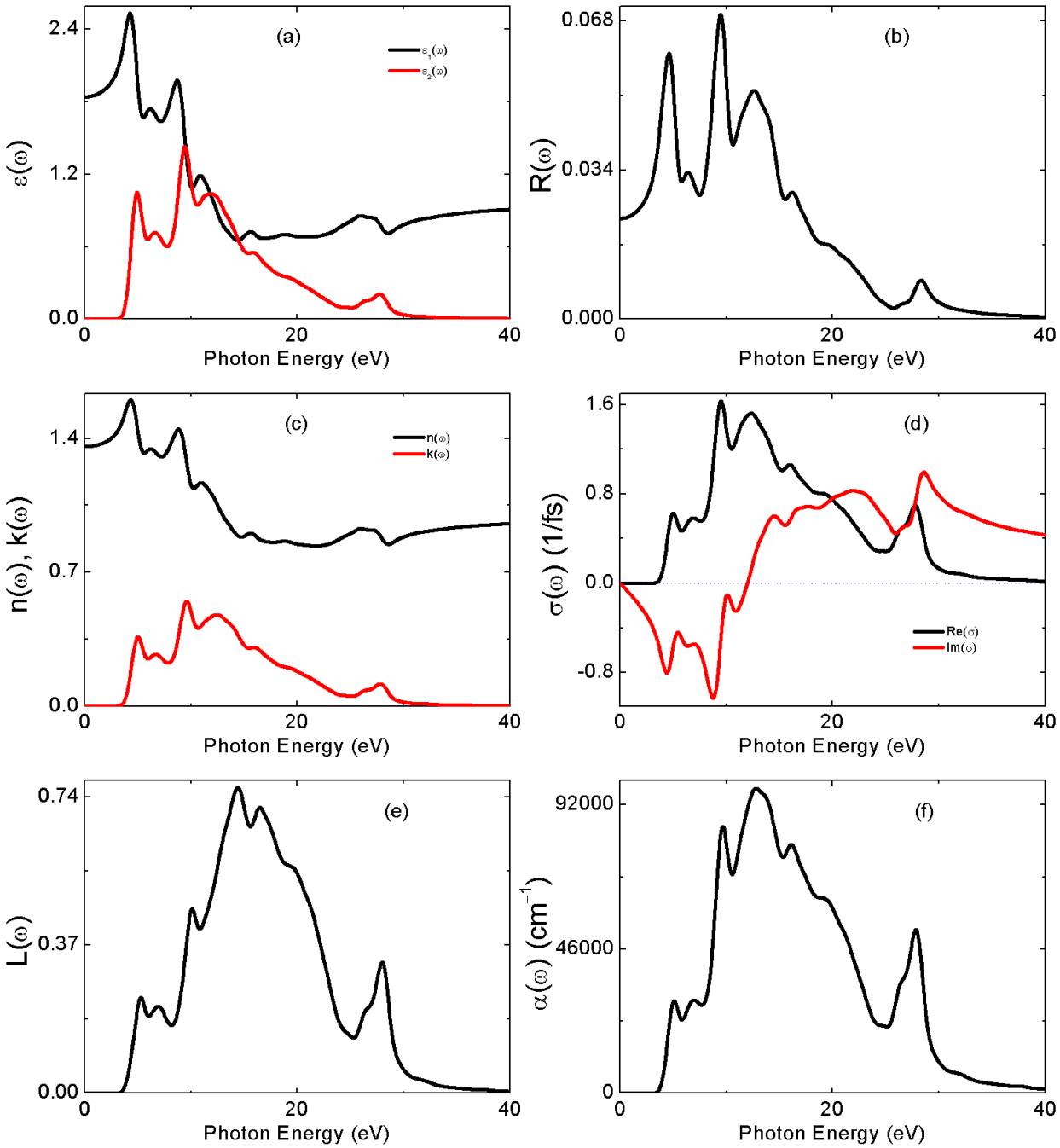

Figure S60. Calculated optical properties for (Ca, Br)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

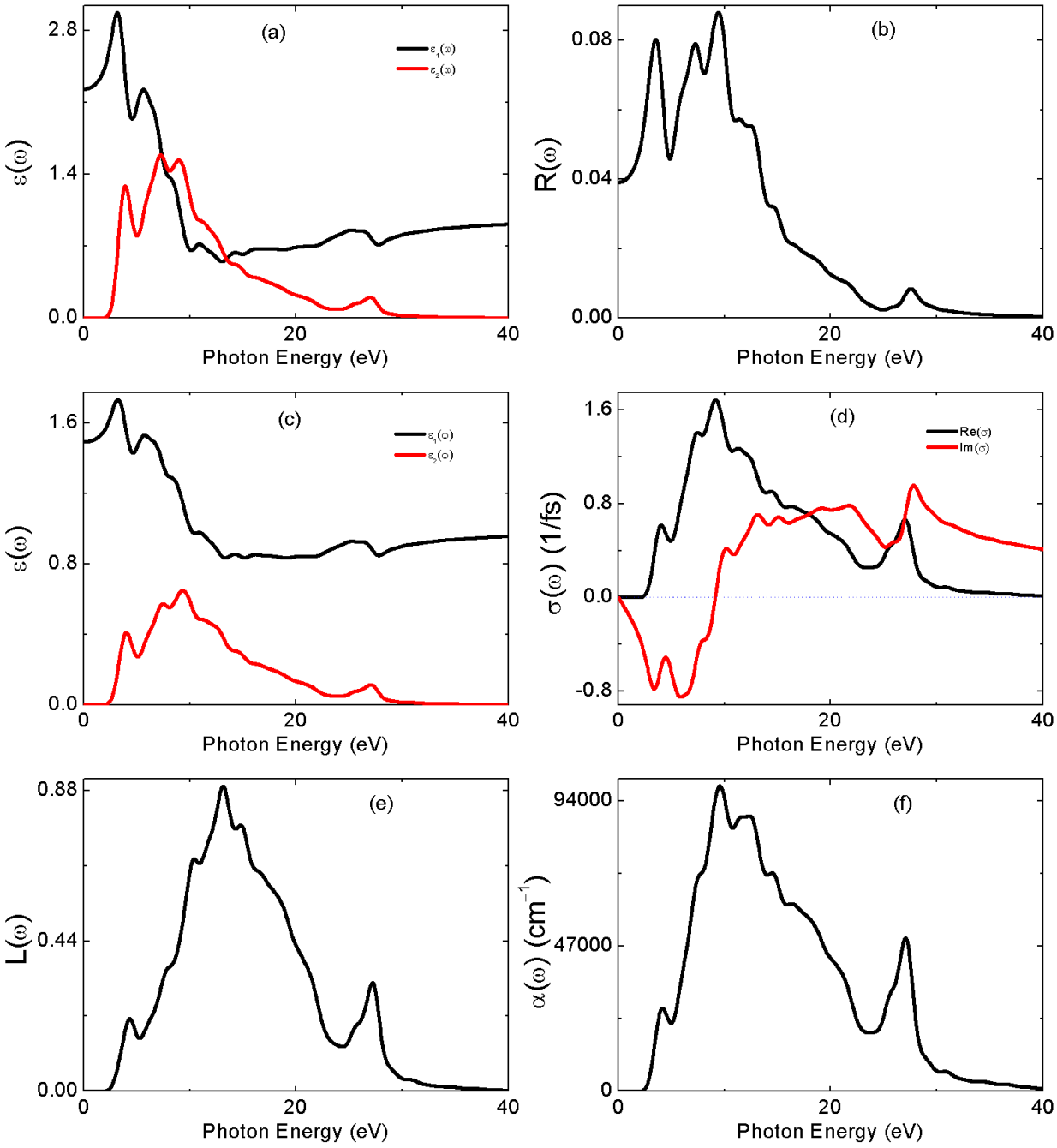

Figure S61. Calculated optical properties for (Ca, I)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

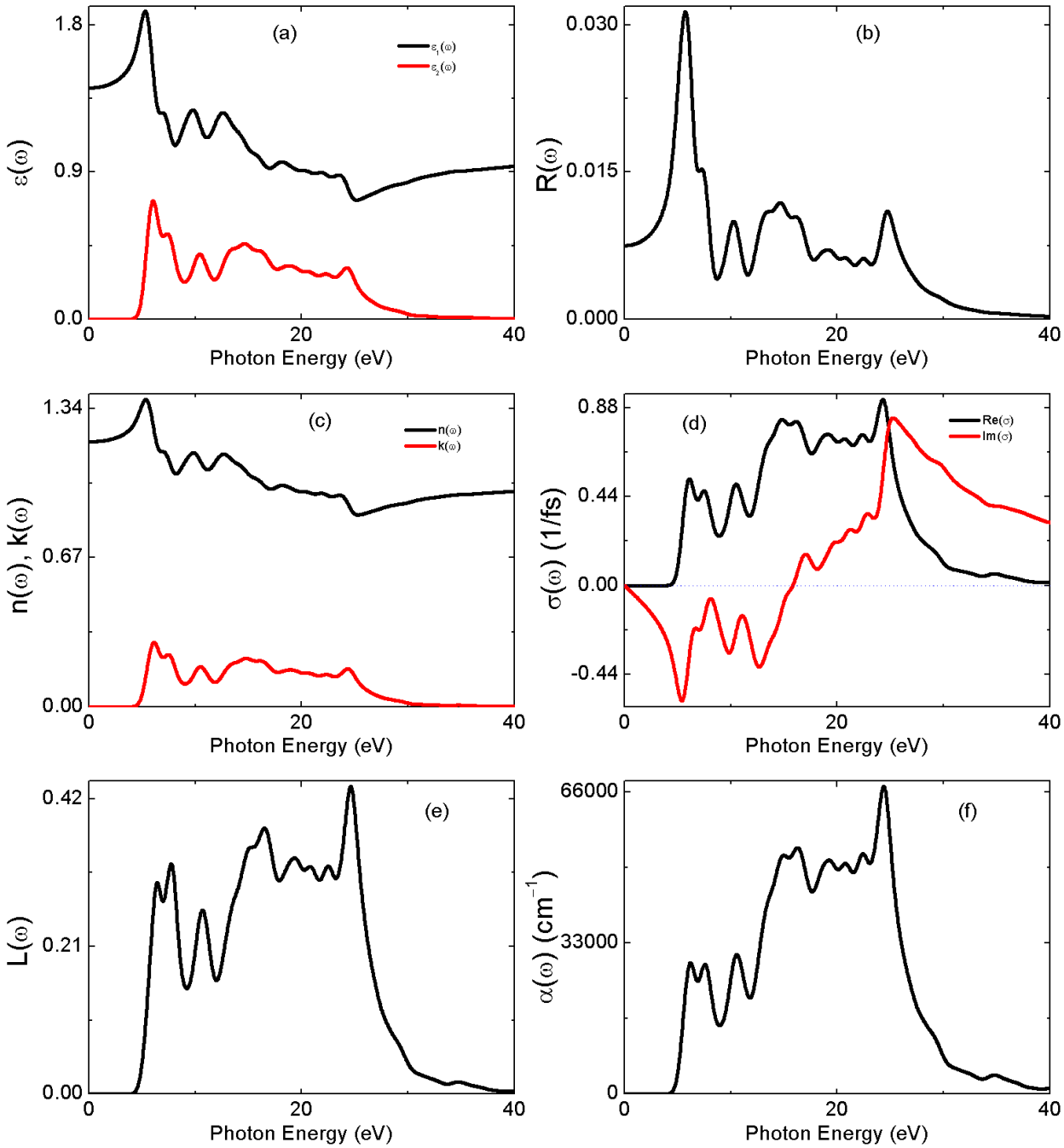

Figure S62. Calculated optical properties for (Sr, F)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

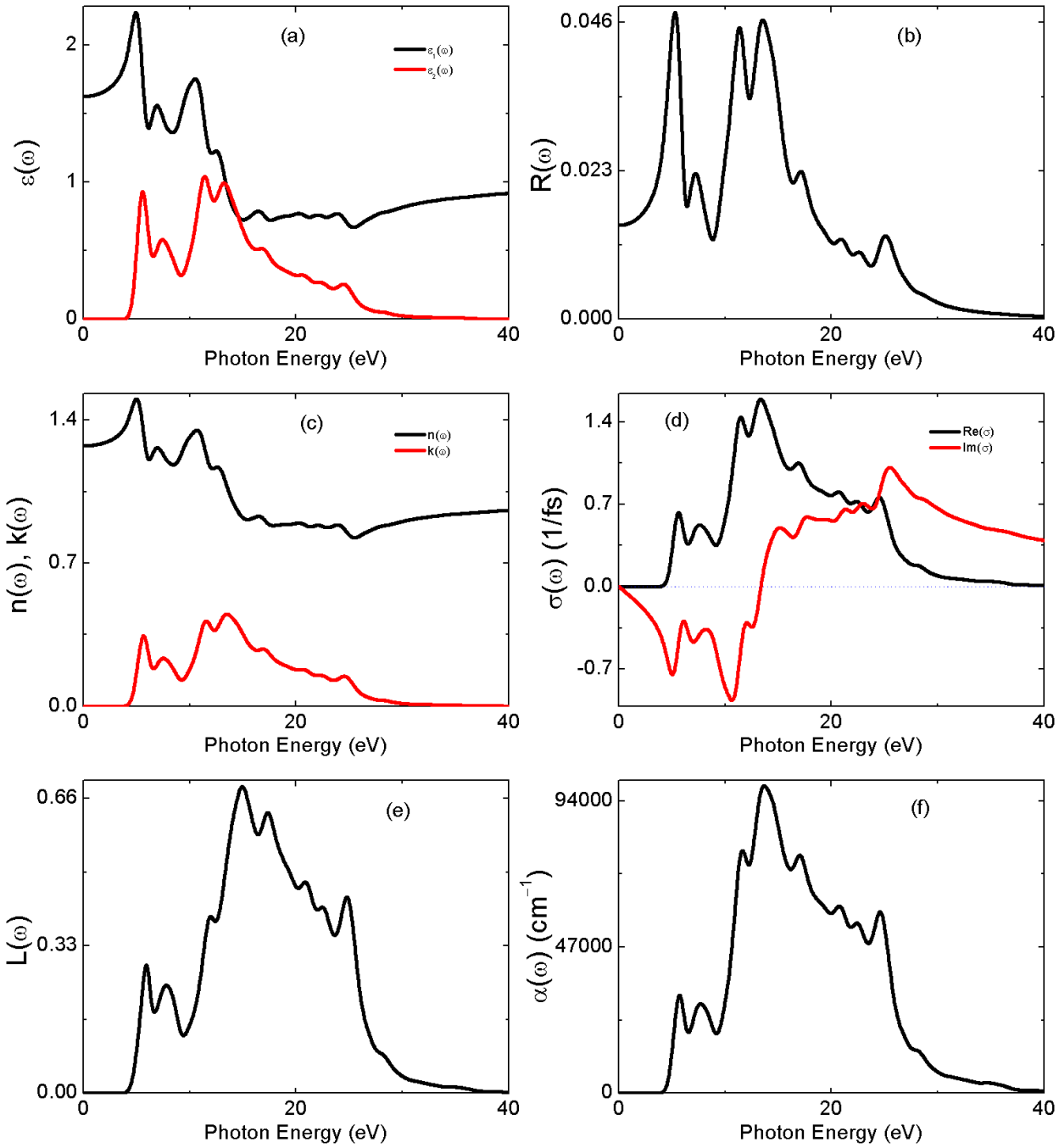

Figure S63. Calculated optical properties for (Sr, Cl)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

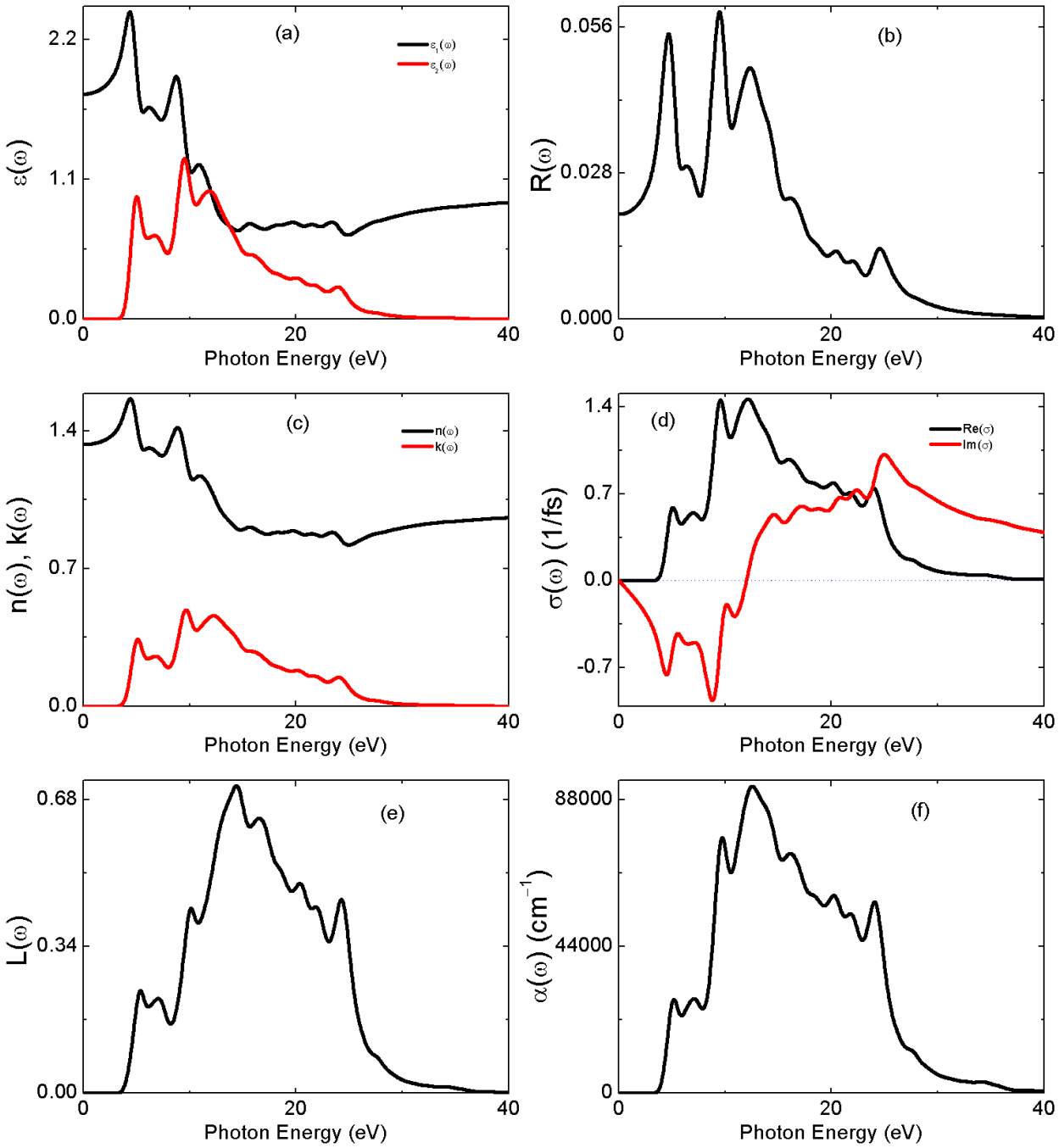

Figure S64. Calculated optical properties for (Sr, Br)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

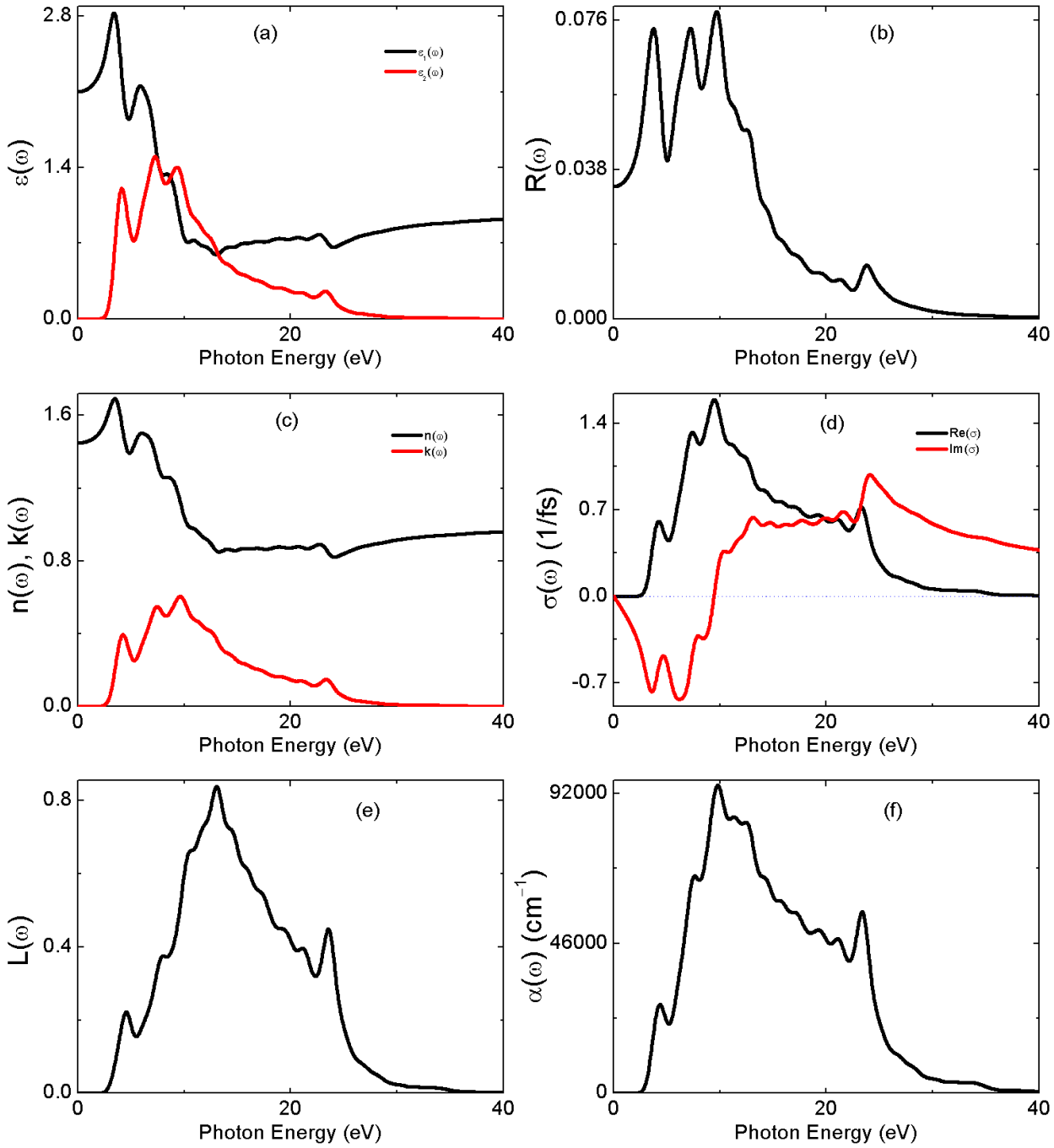

Figure S65. Calculated optical properties for (Sr, I)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

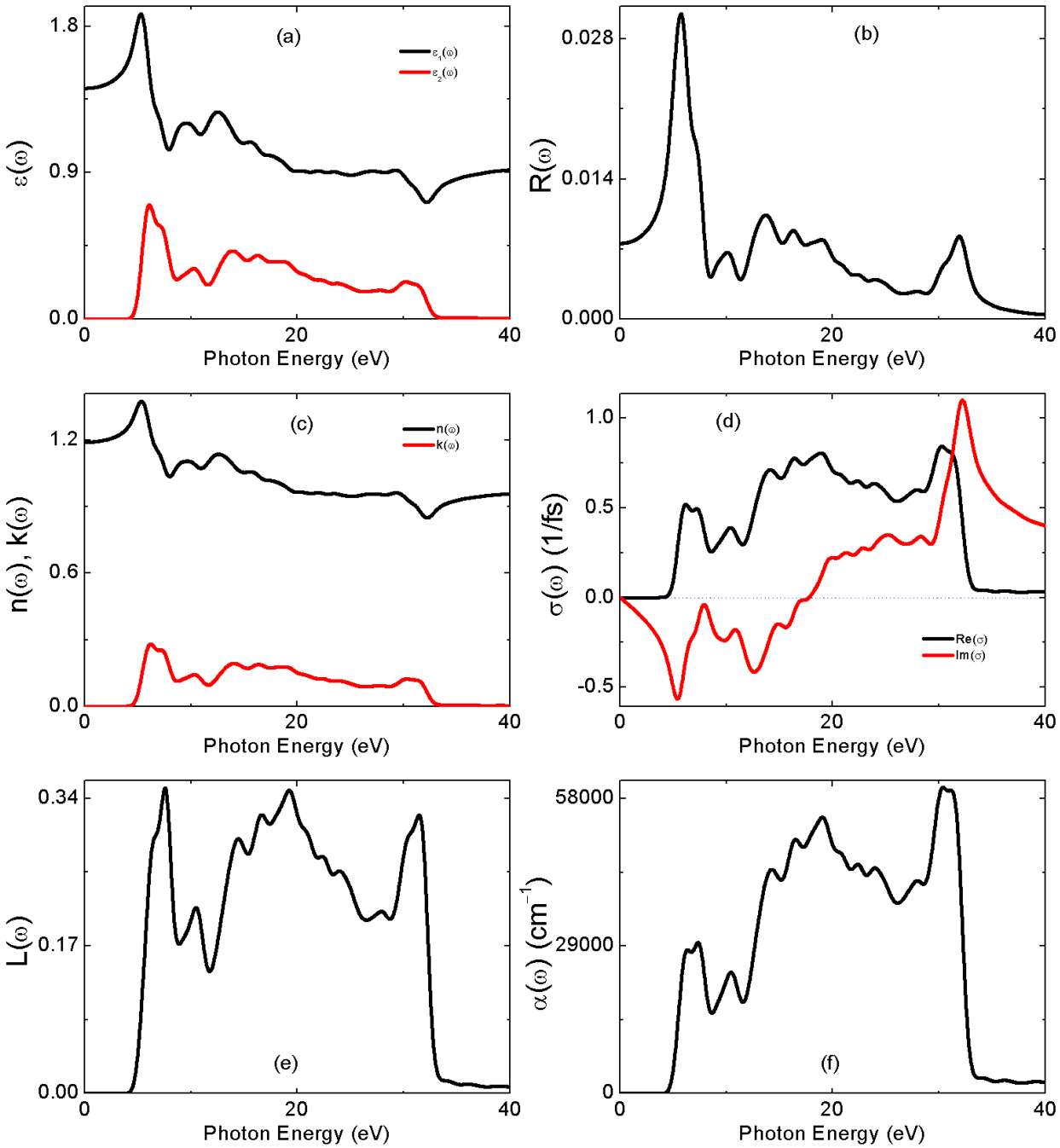

Figure S66. Calculated optical properties for (Ba, F)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

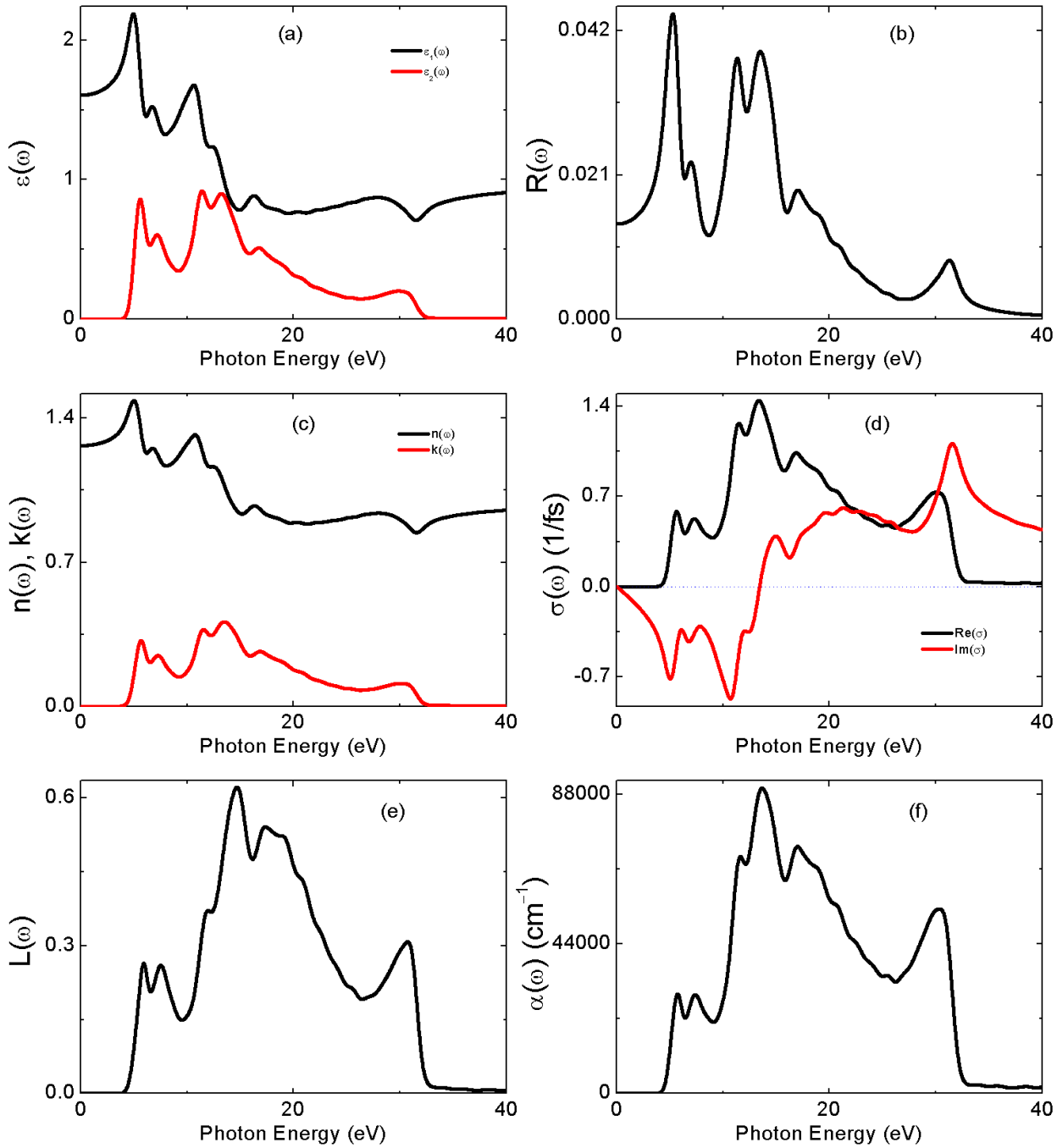

Figure S67. Calculated optical properties for (Ba, Cl)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

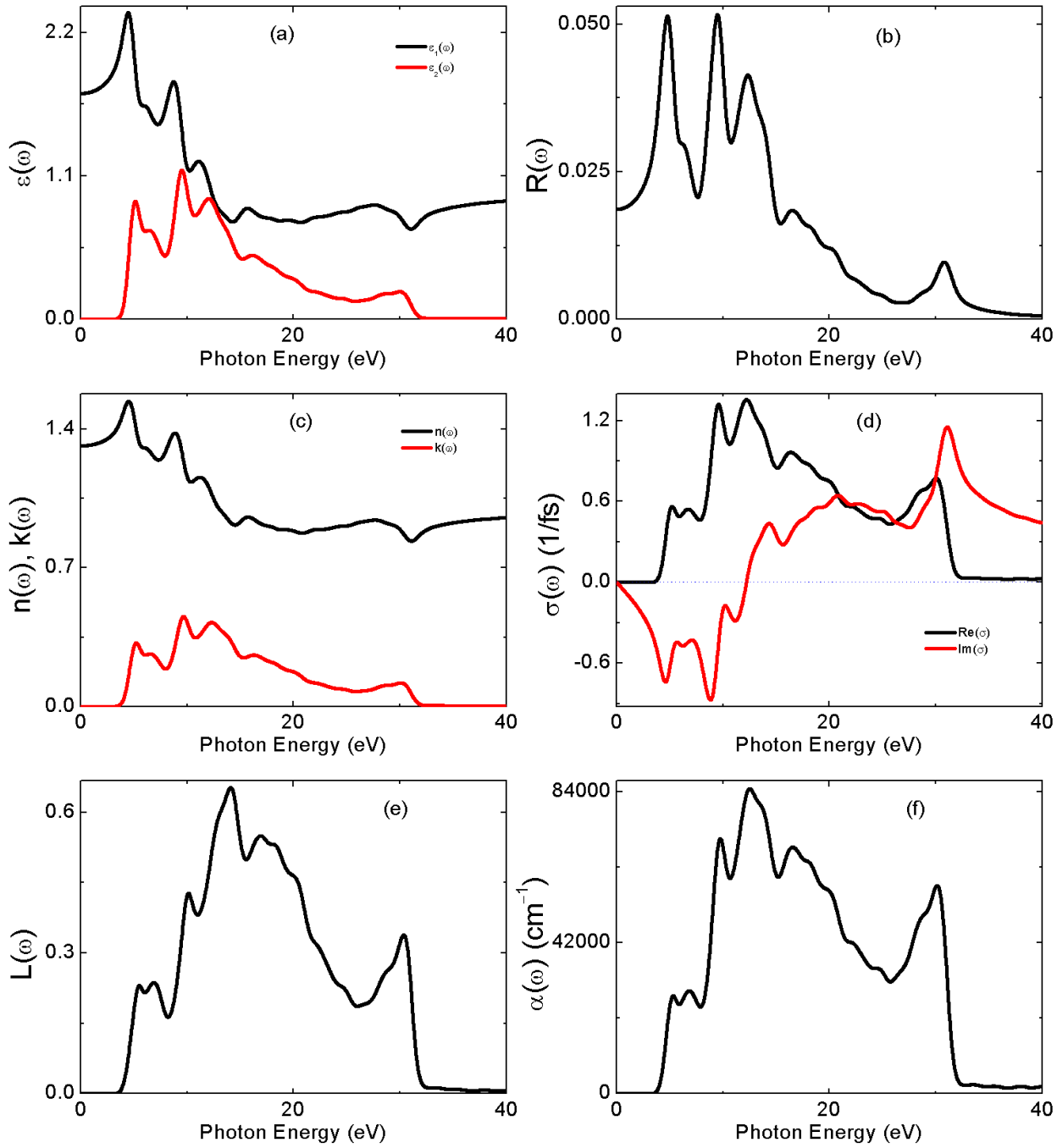

Figure S68. Calculated optical properties for (Ba, Br)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

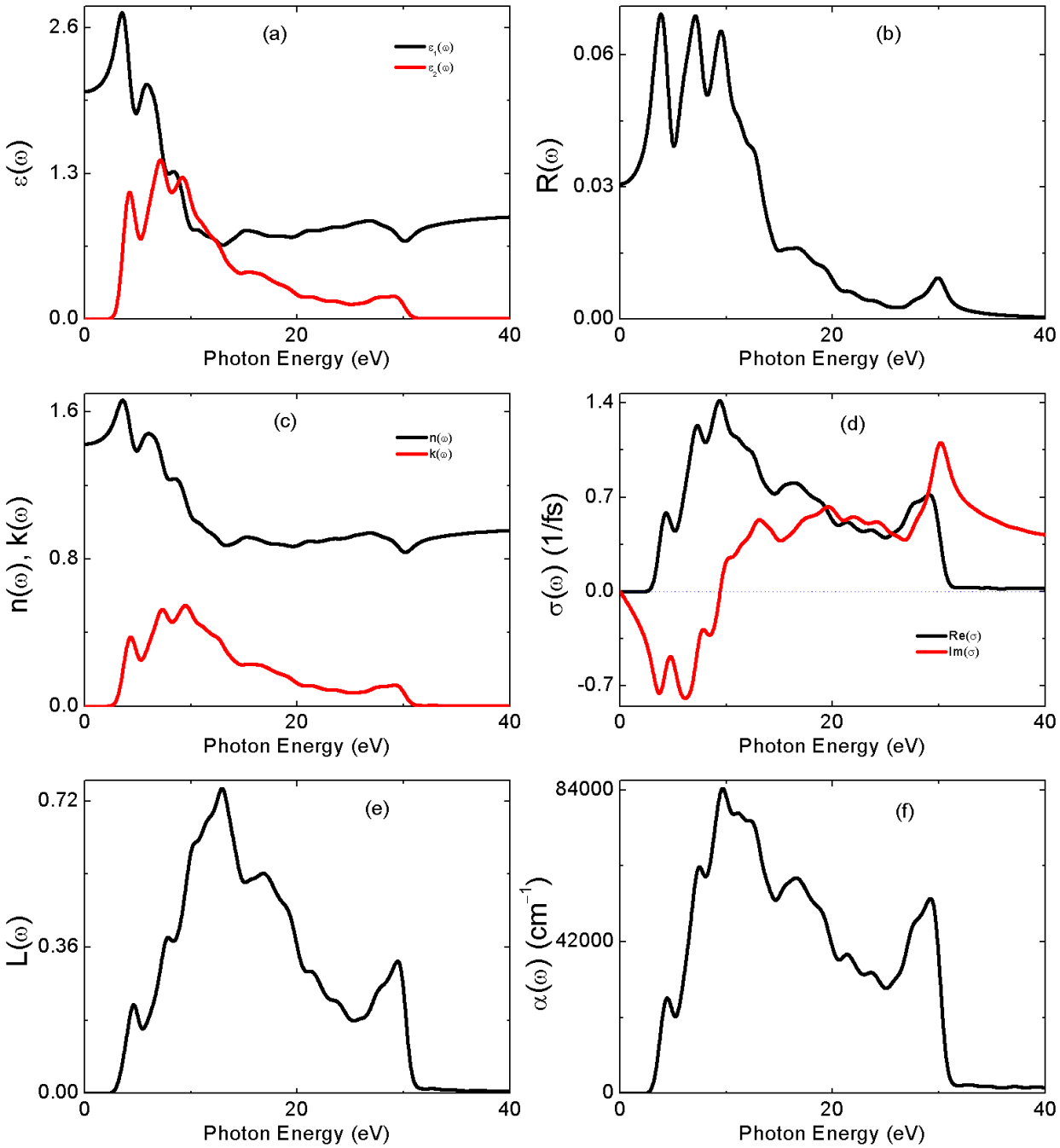

Figure S69. Calculated optical properties for (Ba, I)-90°: (a) dielectric function ε(ω), (b) reflectivity R(ω), (c) refractive index **n**(ω); extinction coefficient **k**(ω), (d) optical conductivity σ(ω), (e) energy loss function L(ω), (f) absorption α(ω).

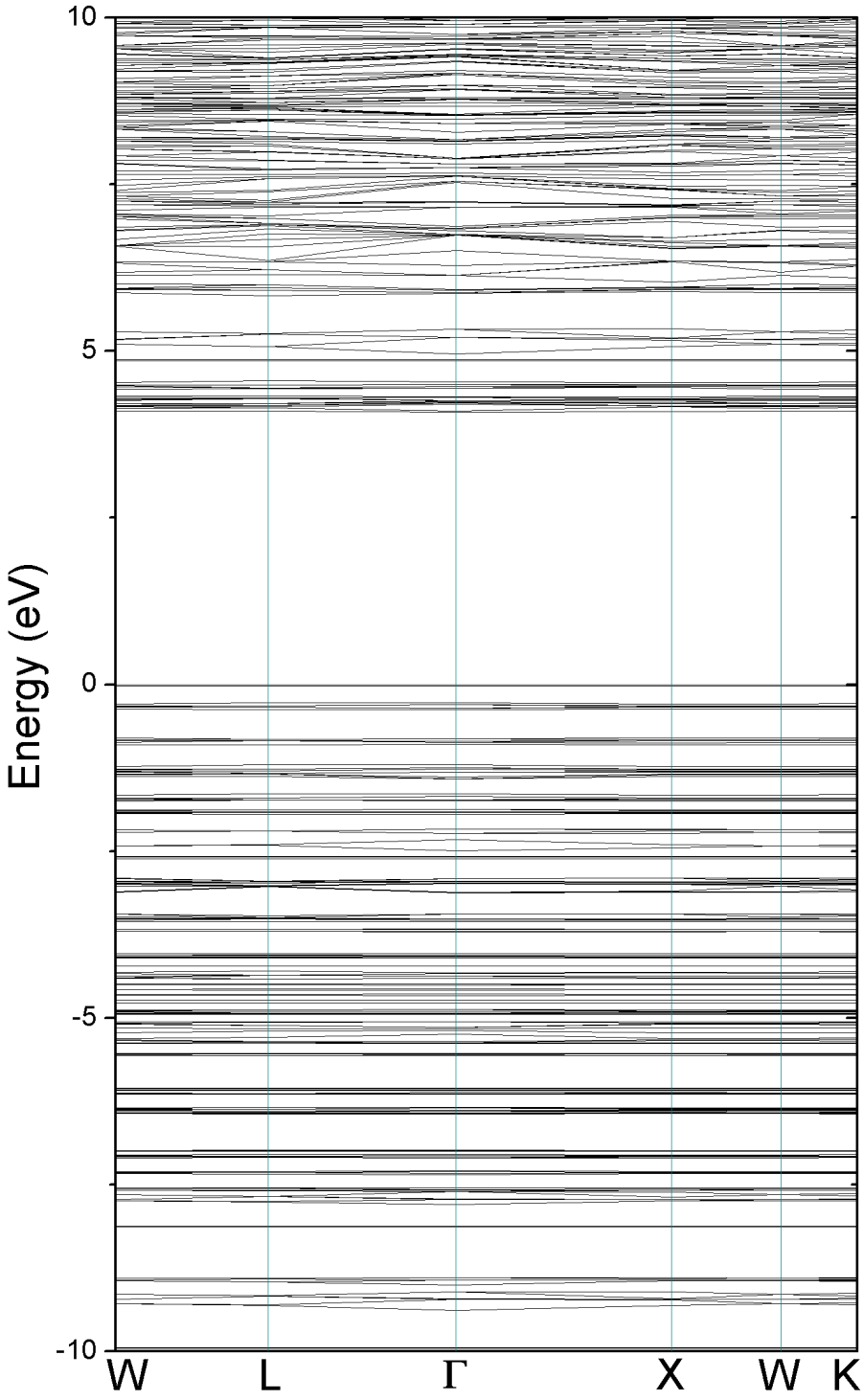

Figure S70. The electronic band structure of (Zn, F)-90°. The Fermi level is set to zero and placed in the valence band maximum.

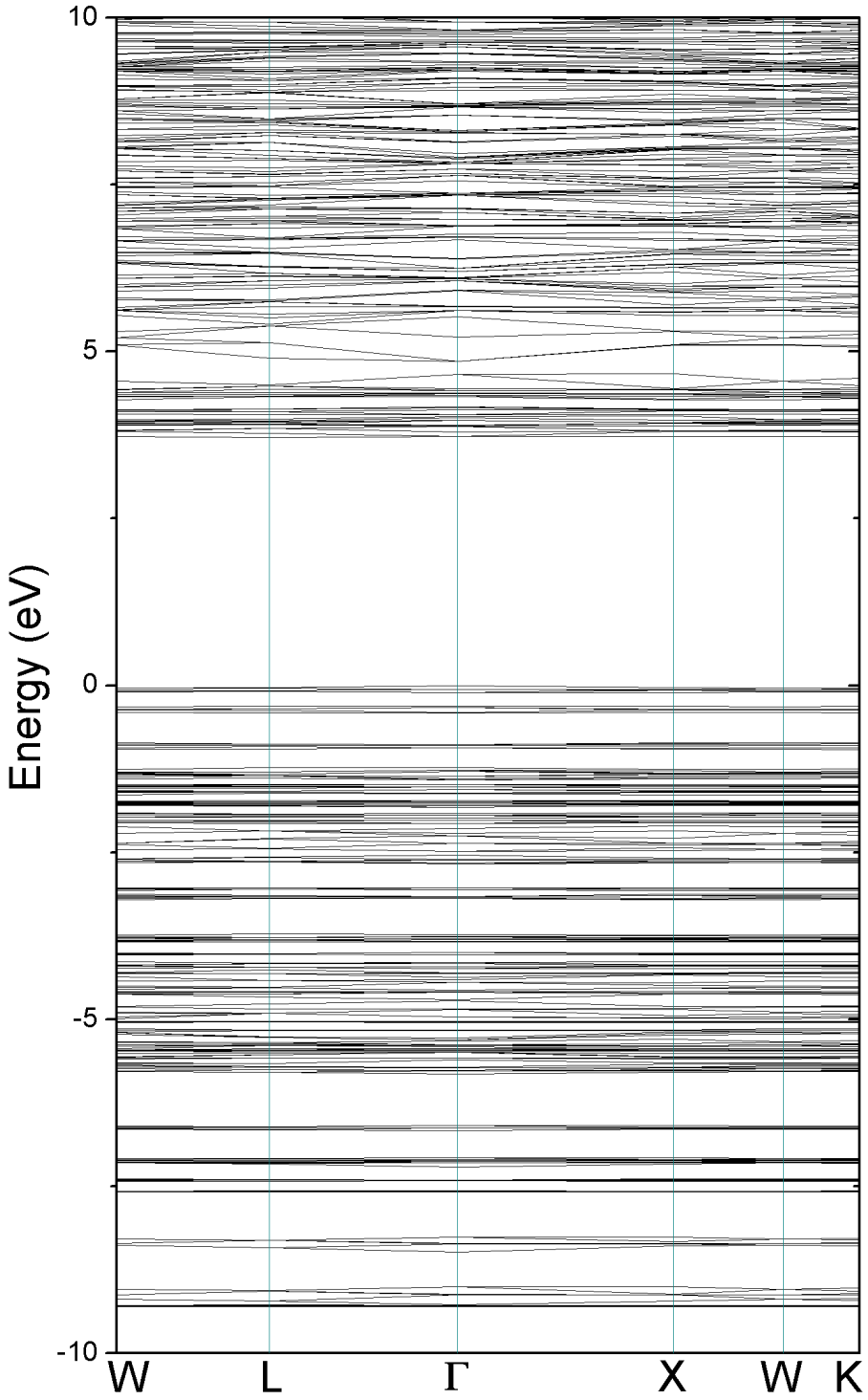

Figure S71. The electronic band structure of (Zn, Cl)-90°. The Fermi level is set to zero and placed in the valence band maximum.

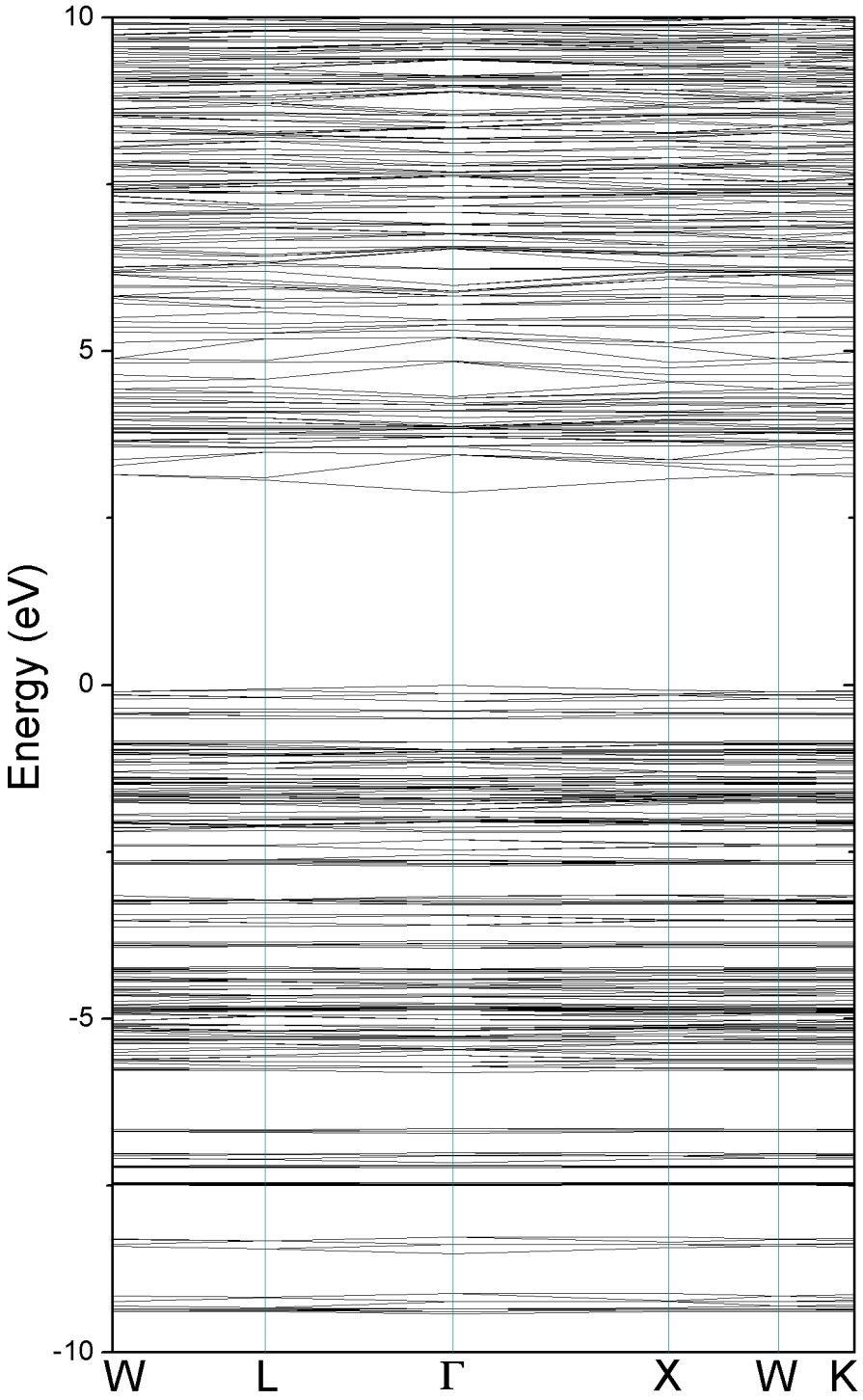

Figure S72. The electronic band structure of (Zn, Br)-90°. The Fermi level is set to zero and placed in the valence band maximum.

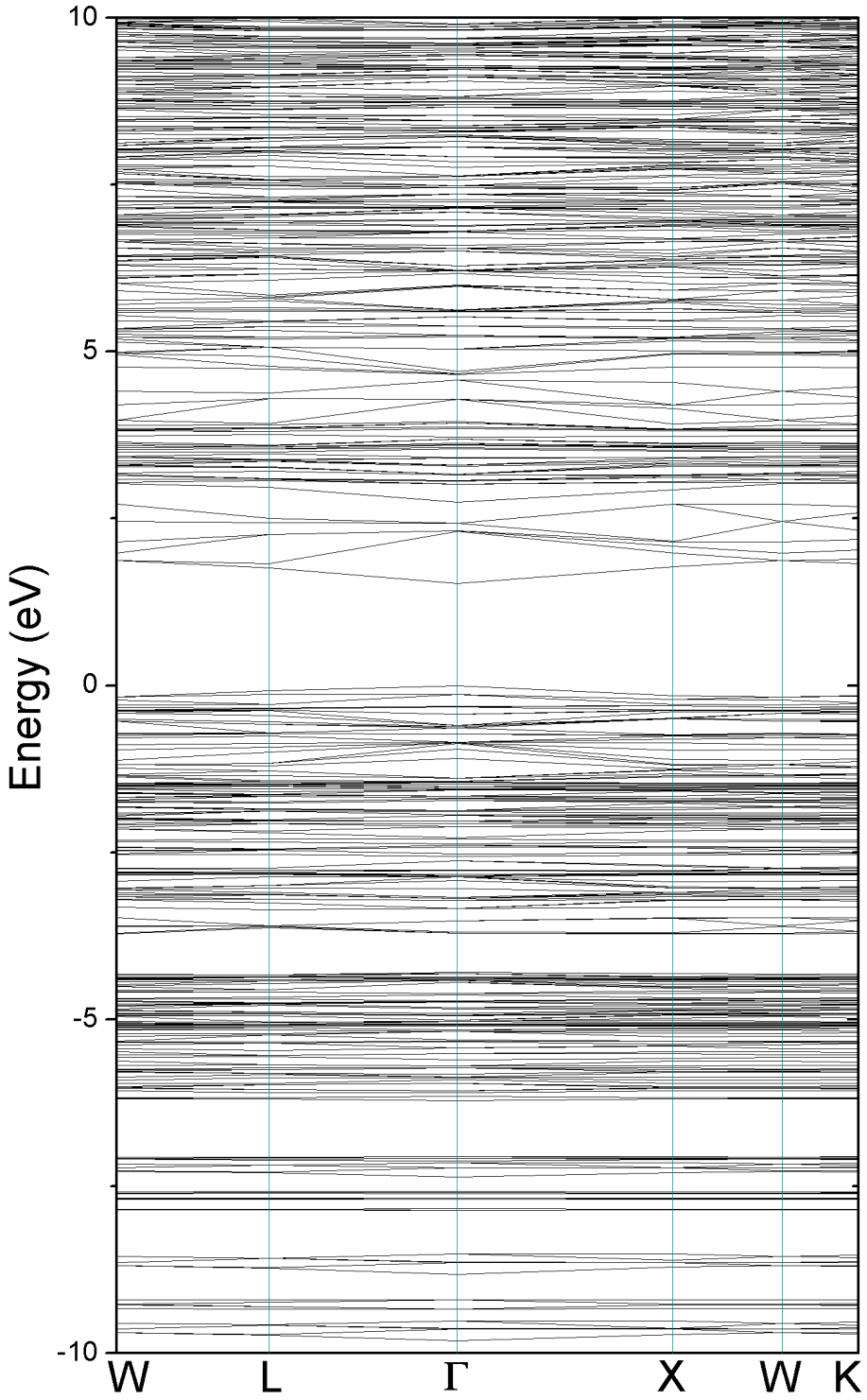

Figure S73. The electronic band structure of (Zn, I)-90°. The Fermi level is set to zero and placed in the valence band maximum.

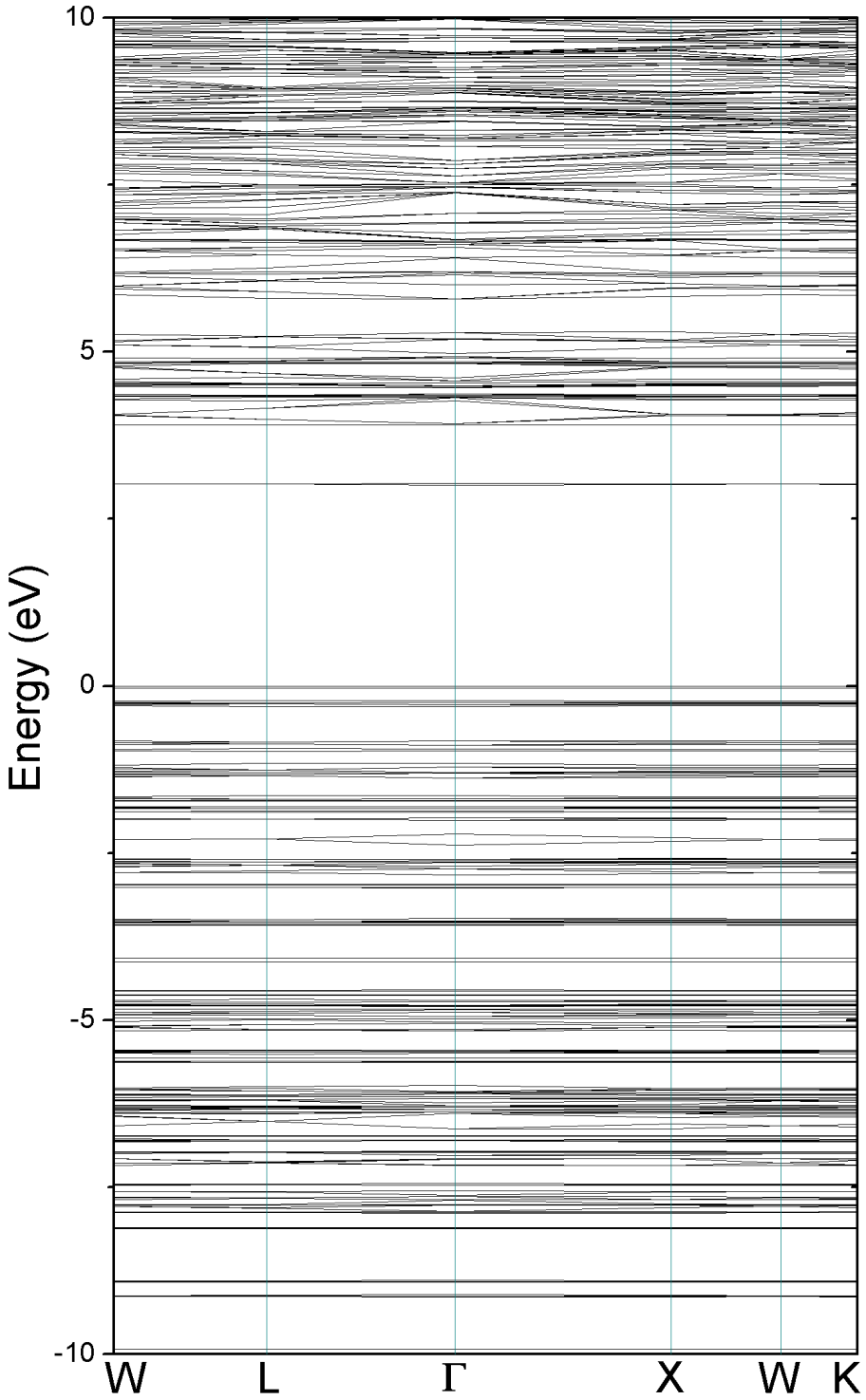

Figure S74. The electronic band structure of (Cd, F)-90°. The Fermi level is set to zero and placed in the valence band maximum.

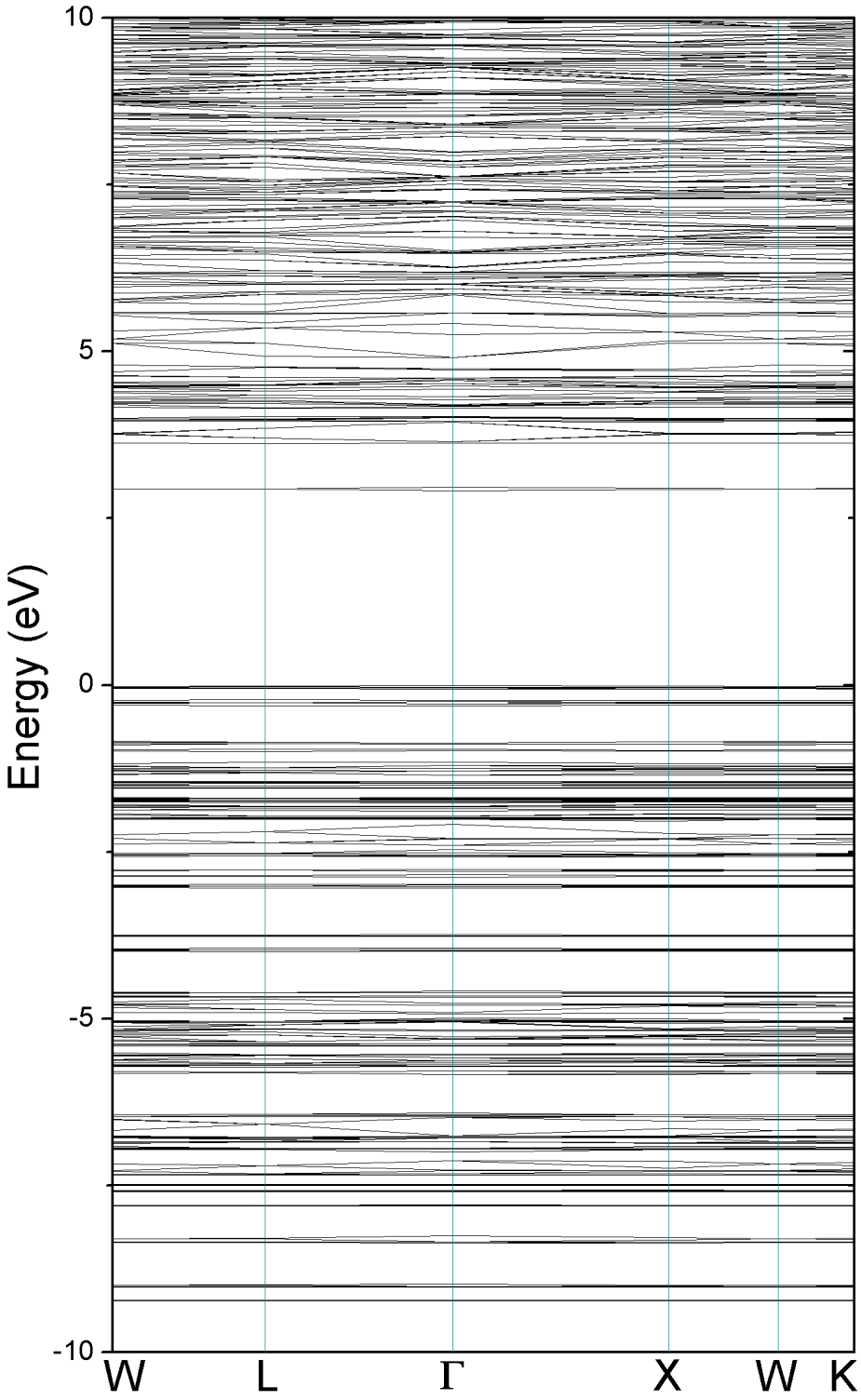

Figure S75. The electronic band structure of (Cd, Cl)-90°. The Fermi level is set to zero and placed in the valence band maximum.

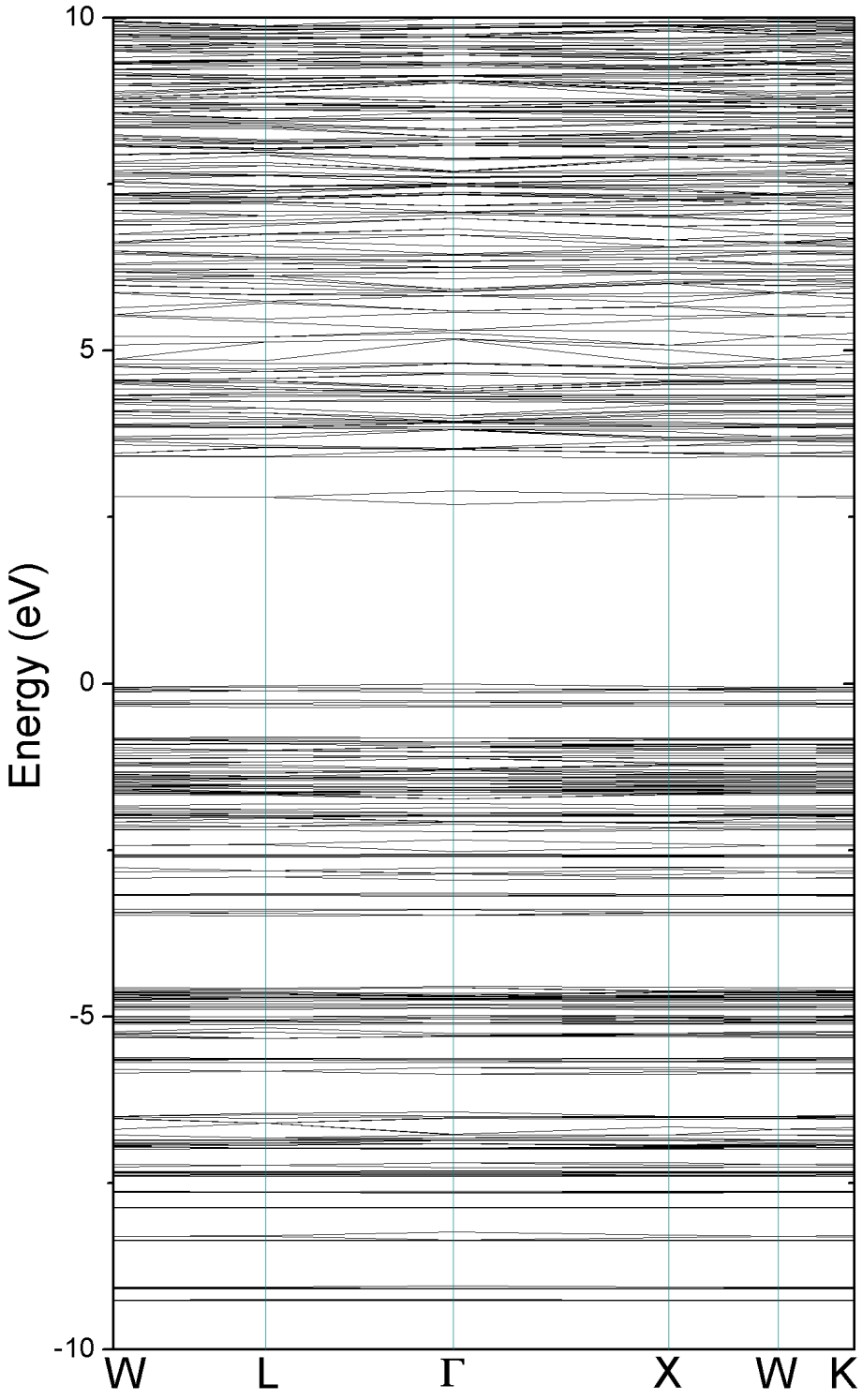

Figure S76. The electronic band structure of (Cd, Br)-90°. The Fermi level is set to zero and placed in the valence band maximum.

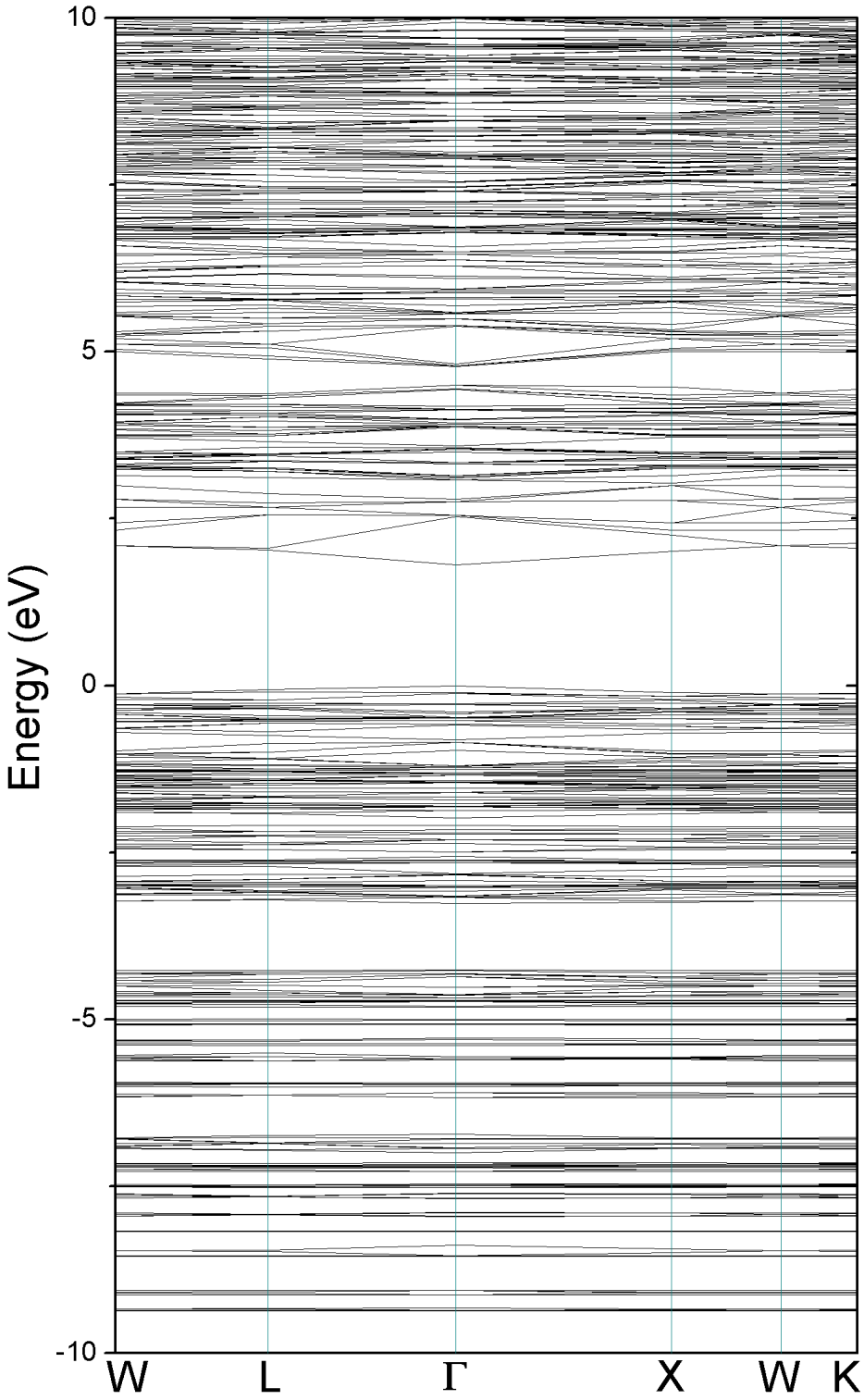

Figure S77. The electronic band structure of (Cd, I)-90°. The Fermi level is set to zero and placed in the valence band maximum.

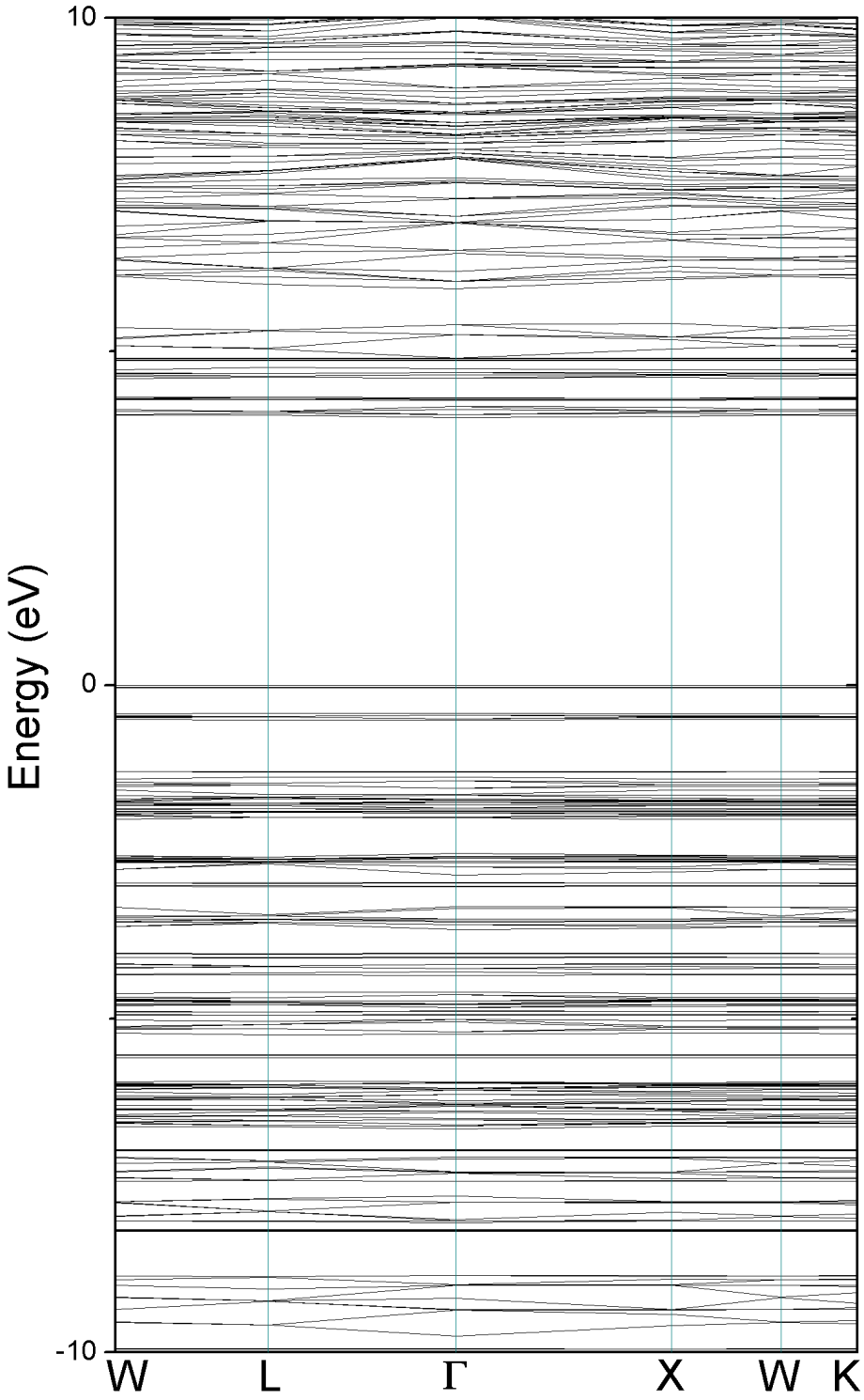

Figure S78. The electronic band structure of (Be, F)-90°. The Fermi level is set to zero and placed in the valence band maximum.

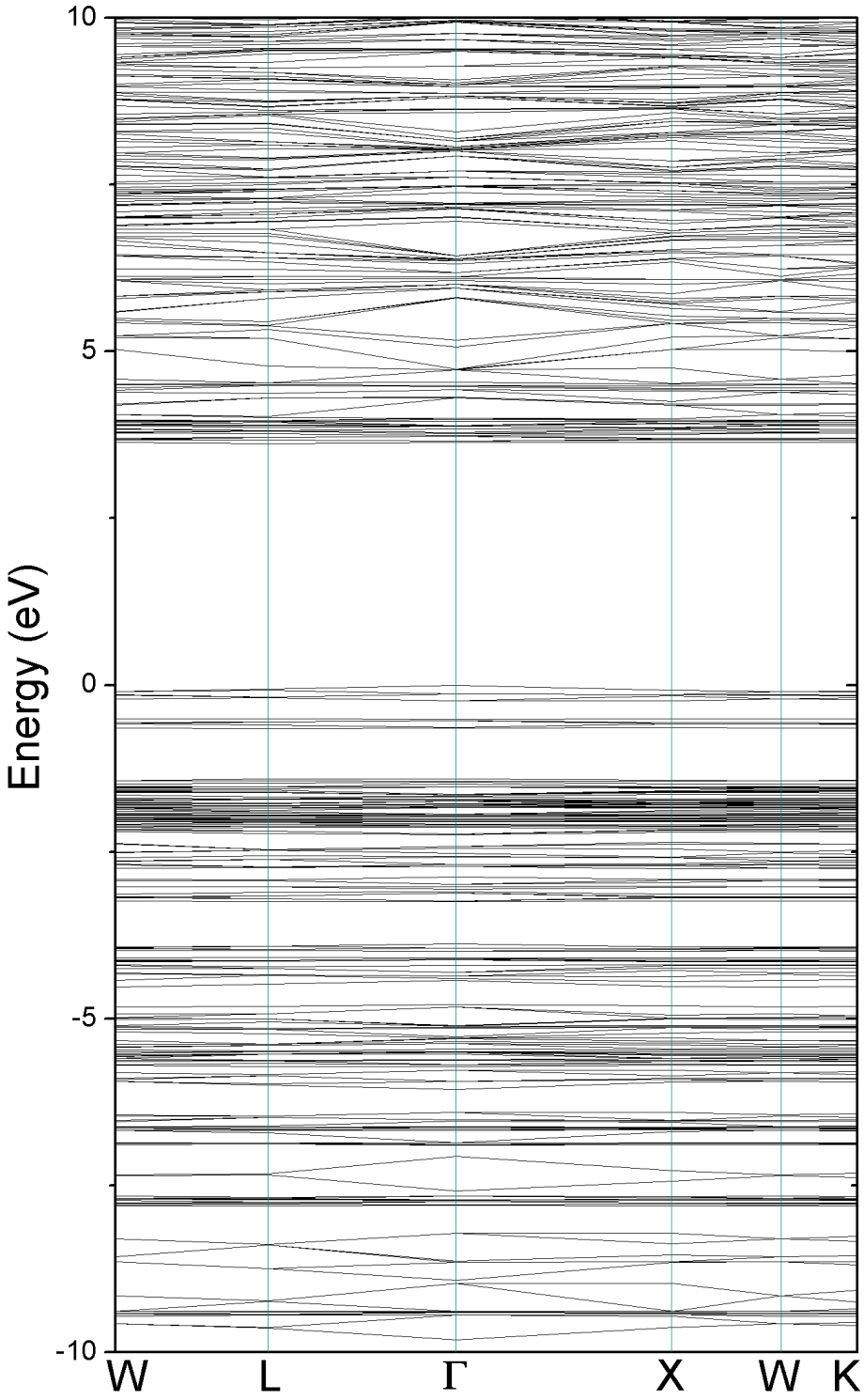

Figure S79. The electronic band structure of (Be, Cl)-90°. The Fermi level is set to zero and placed in the valence band maximum.

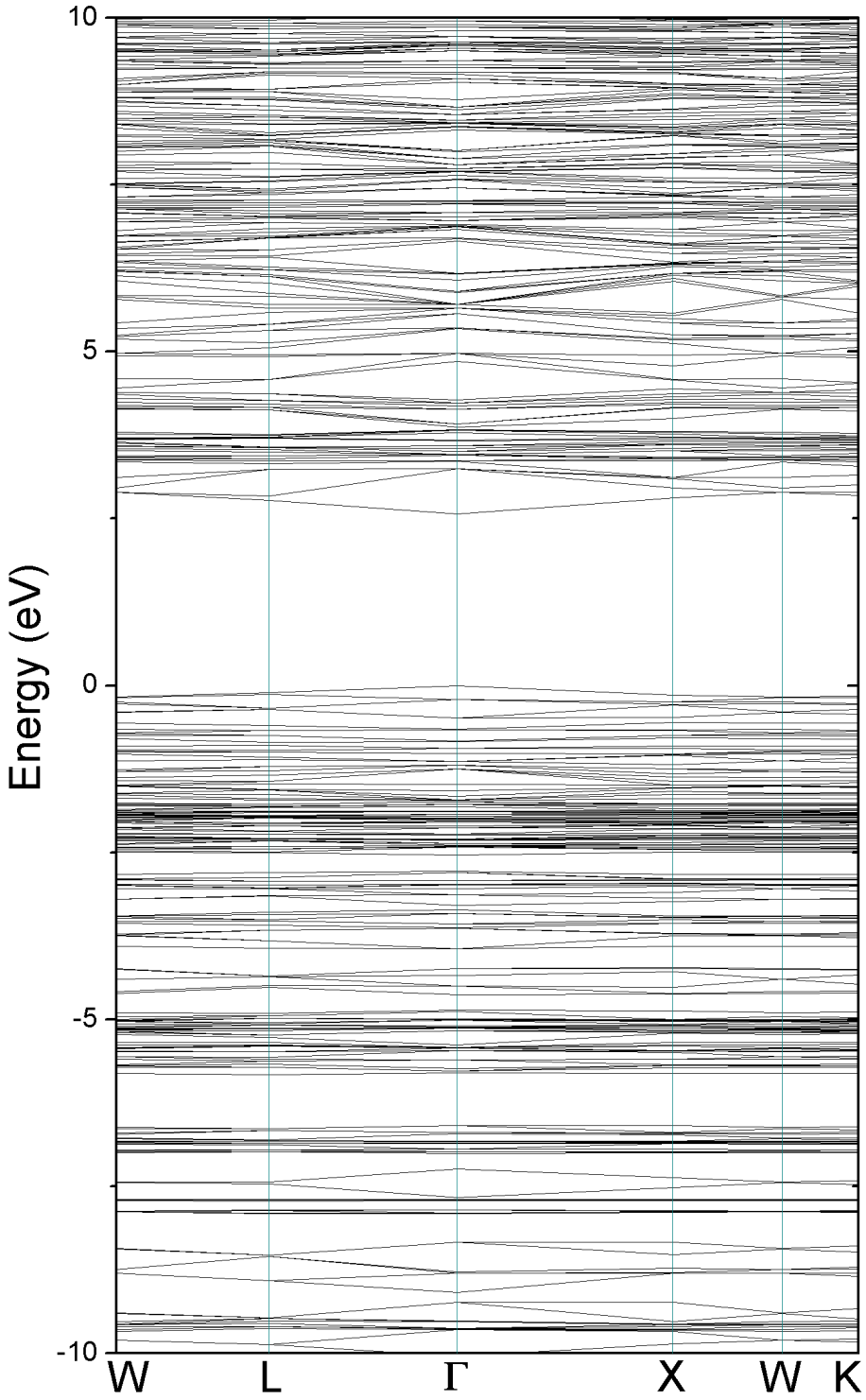

Figure S80. The electronic band structure of (Be, Br)-90°. The Fermi level is set to zero and placed in the valence band maximum.

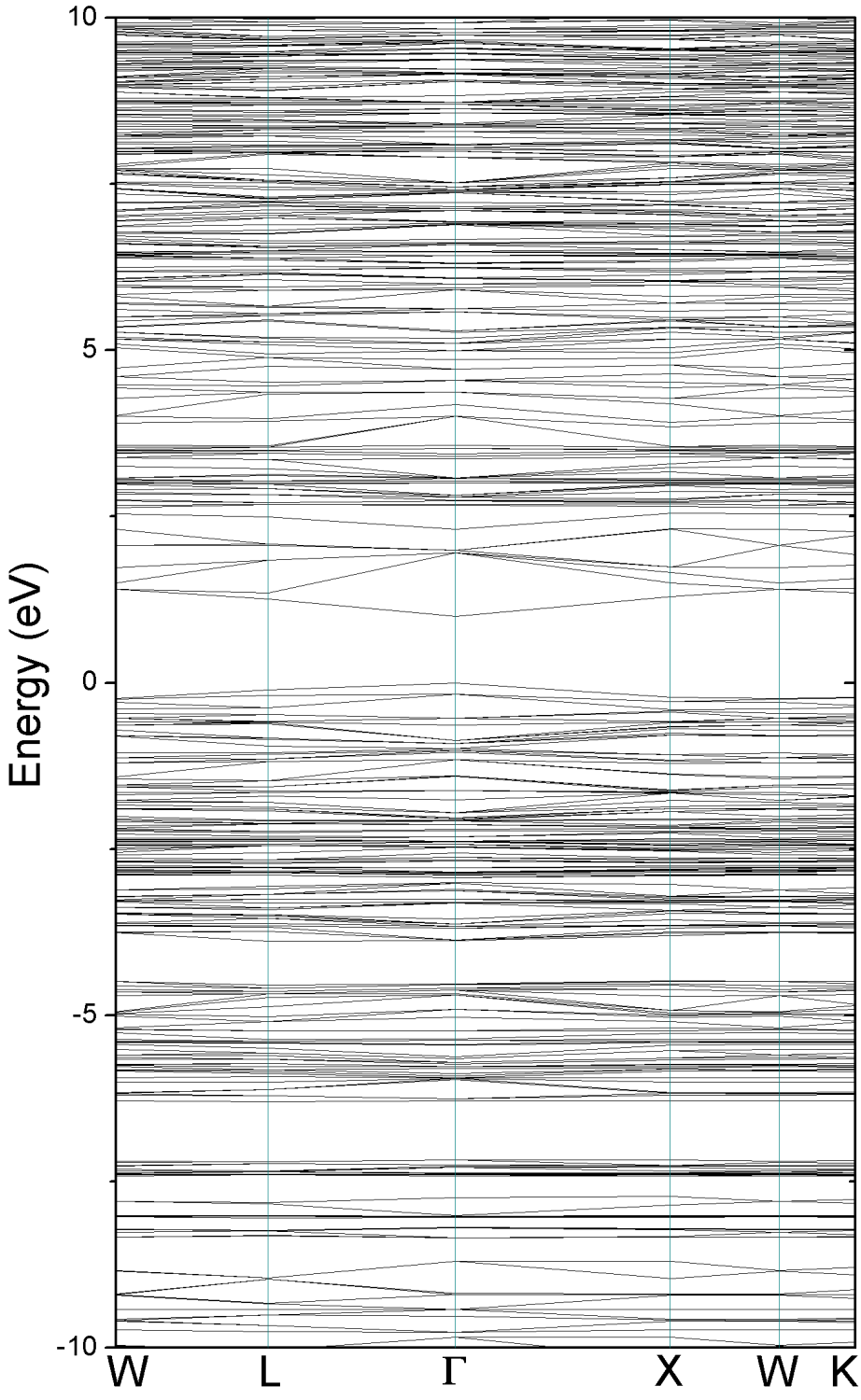

Figure S81. The electronic band structure of (Be, I)-90°. The Fermi level is set to zero and placed in the valence band maximum.

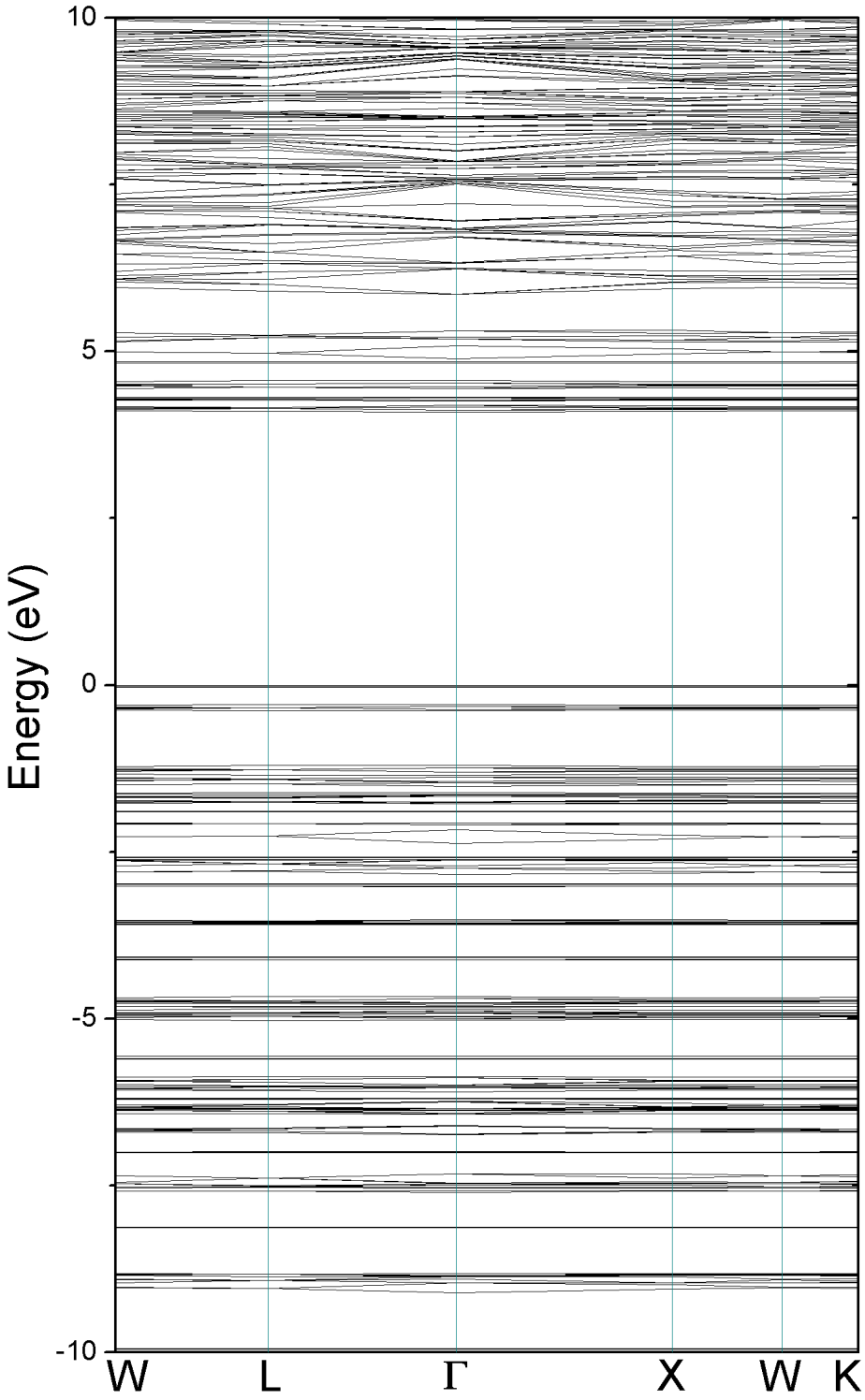

Figure S82. The electronic band structure of (Mg, F)-90°. The Fermi level is set to zero and placed in the valence band maximum.

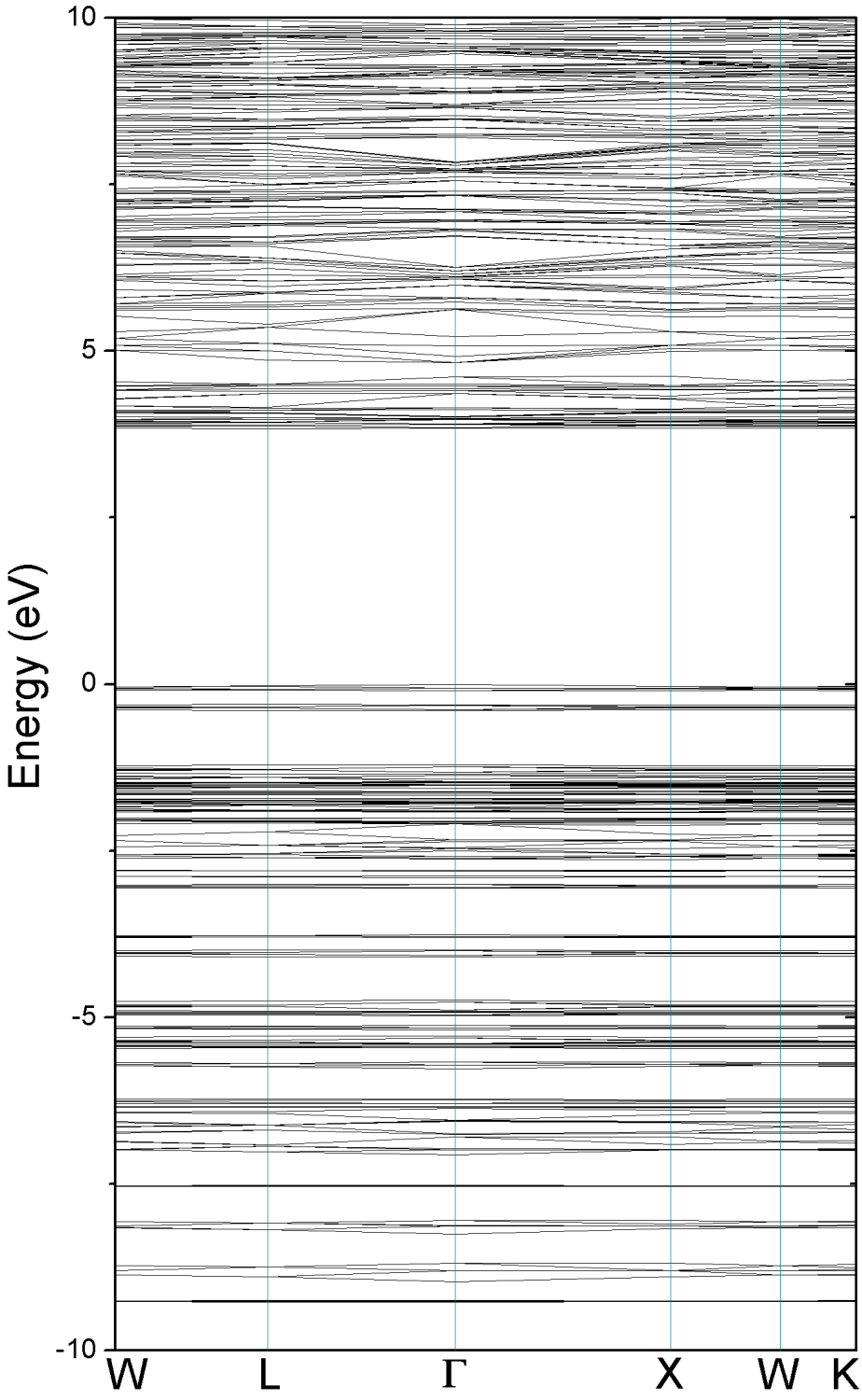

Figure S83. The electronic band structure of (Mg, Cl)-90°. The Fermi level is set to zero and placed in the valence band maximum.

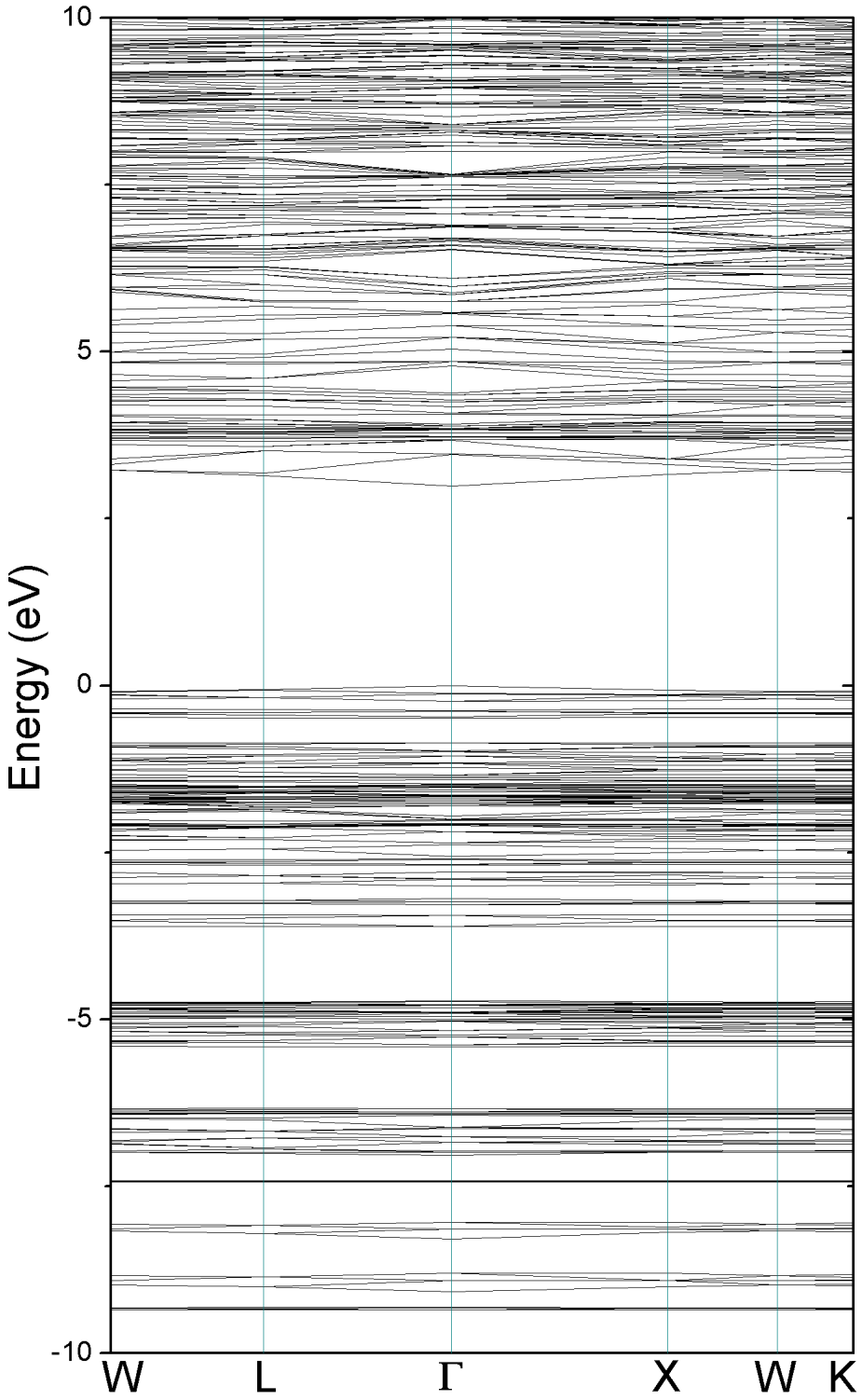

Figure S84. The electronic band structure of (Mg, Br)-90°. The Fermi level is set to zero and placed in the valence band maximum.

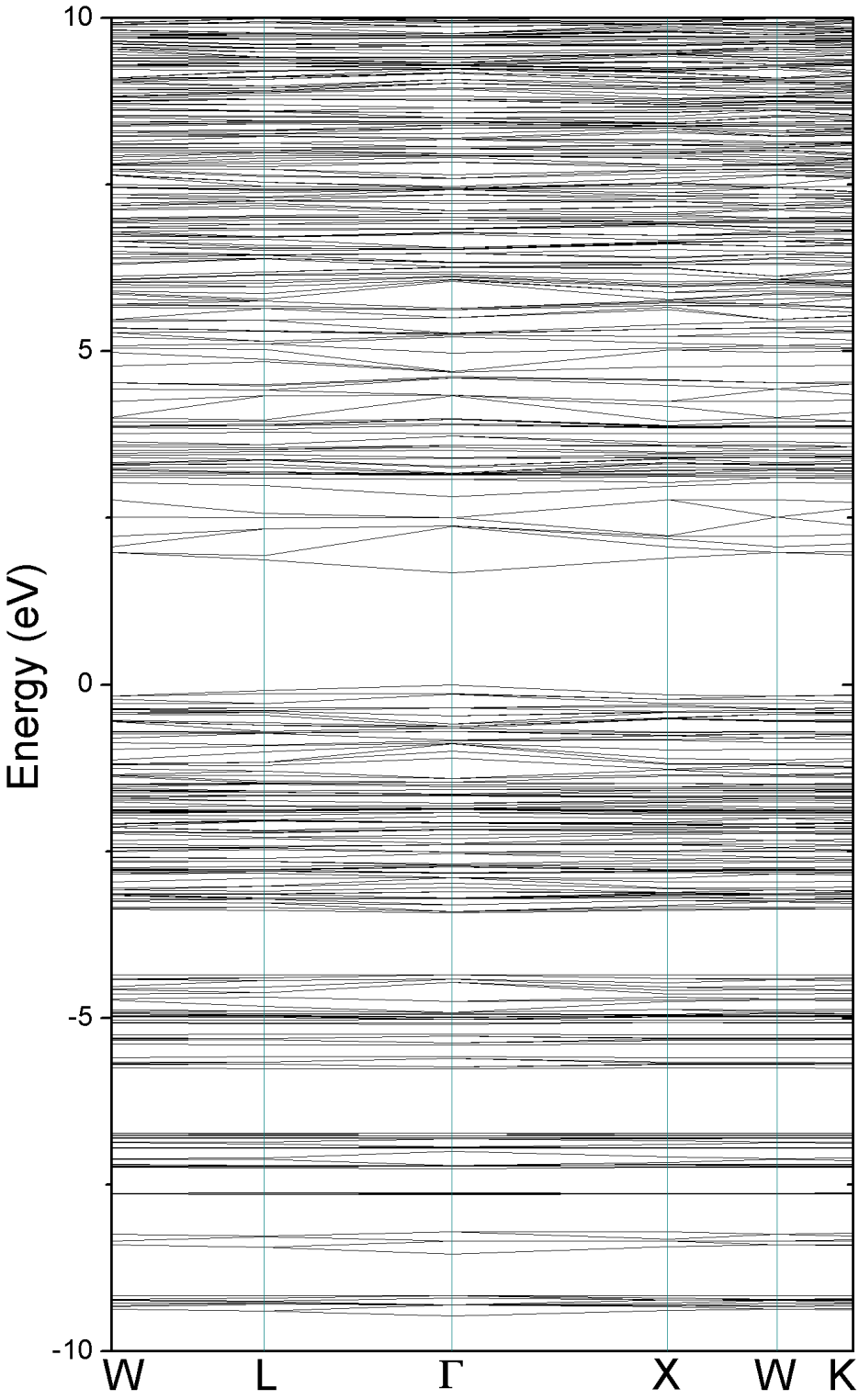

Figure S85. The electronic band structure of (Mg, I)-90°. The Fermi level is set to zero and placed in the valence band maximum.

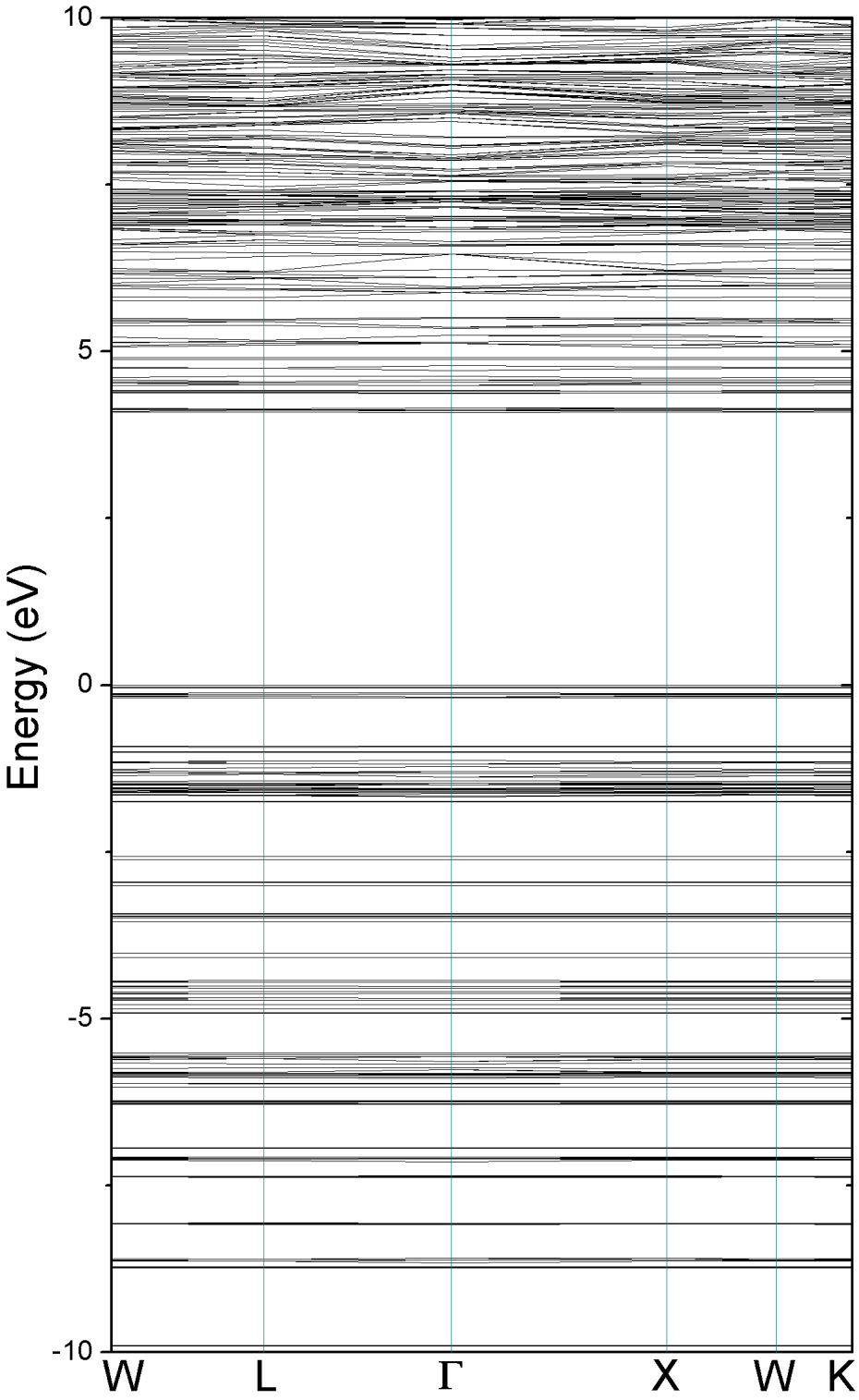

Figure S86. The electronic band structure of (Ca, F)-90°. The Fermi level is set to zero and placed in the valence band maximum.

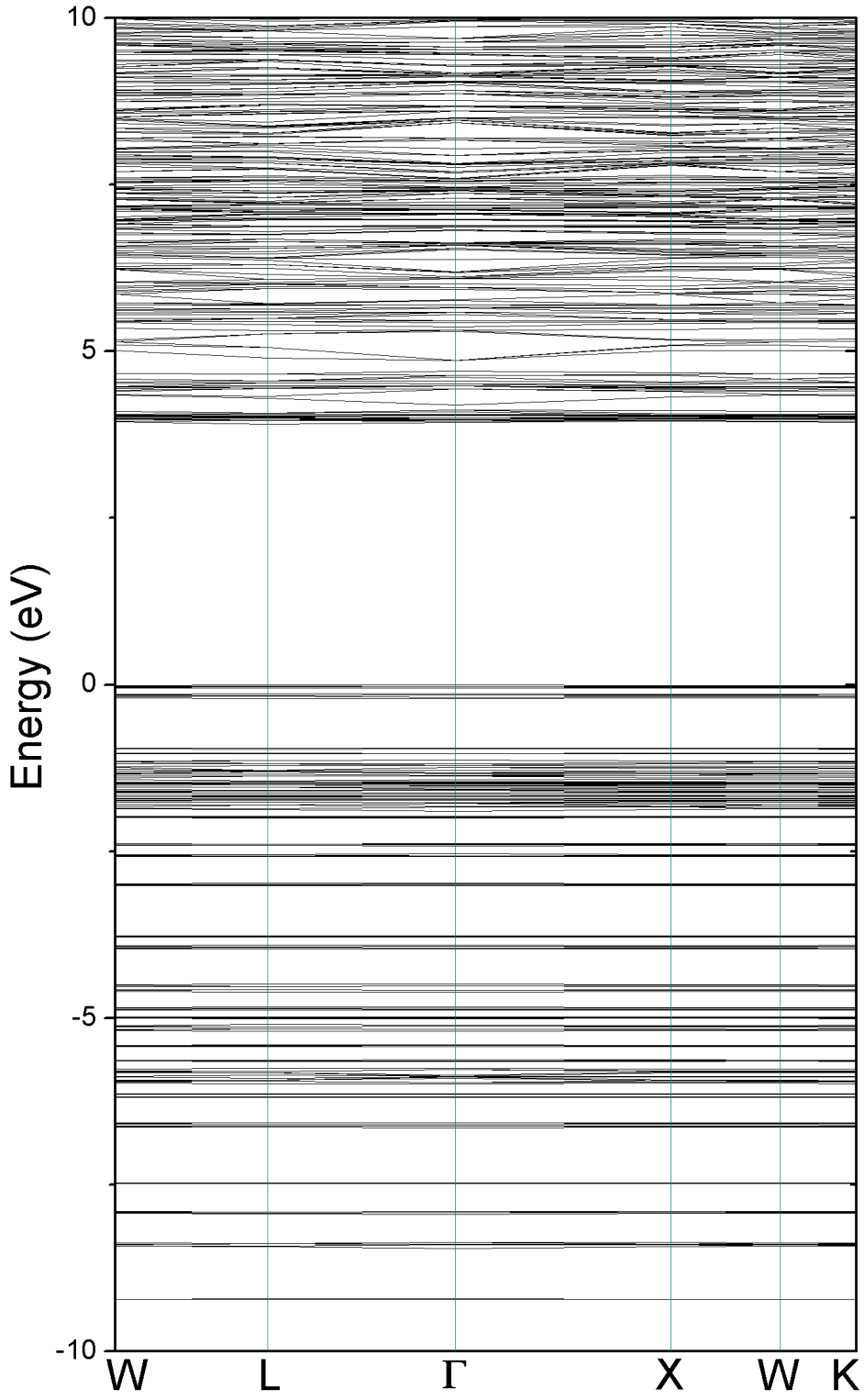

Figure S87. The electronic band structure of (Ca, Cl)-90°. The Fermi level is set to zero and placed in the valence band maximum.

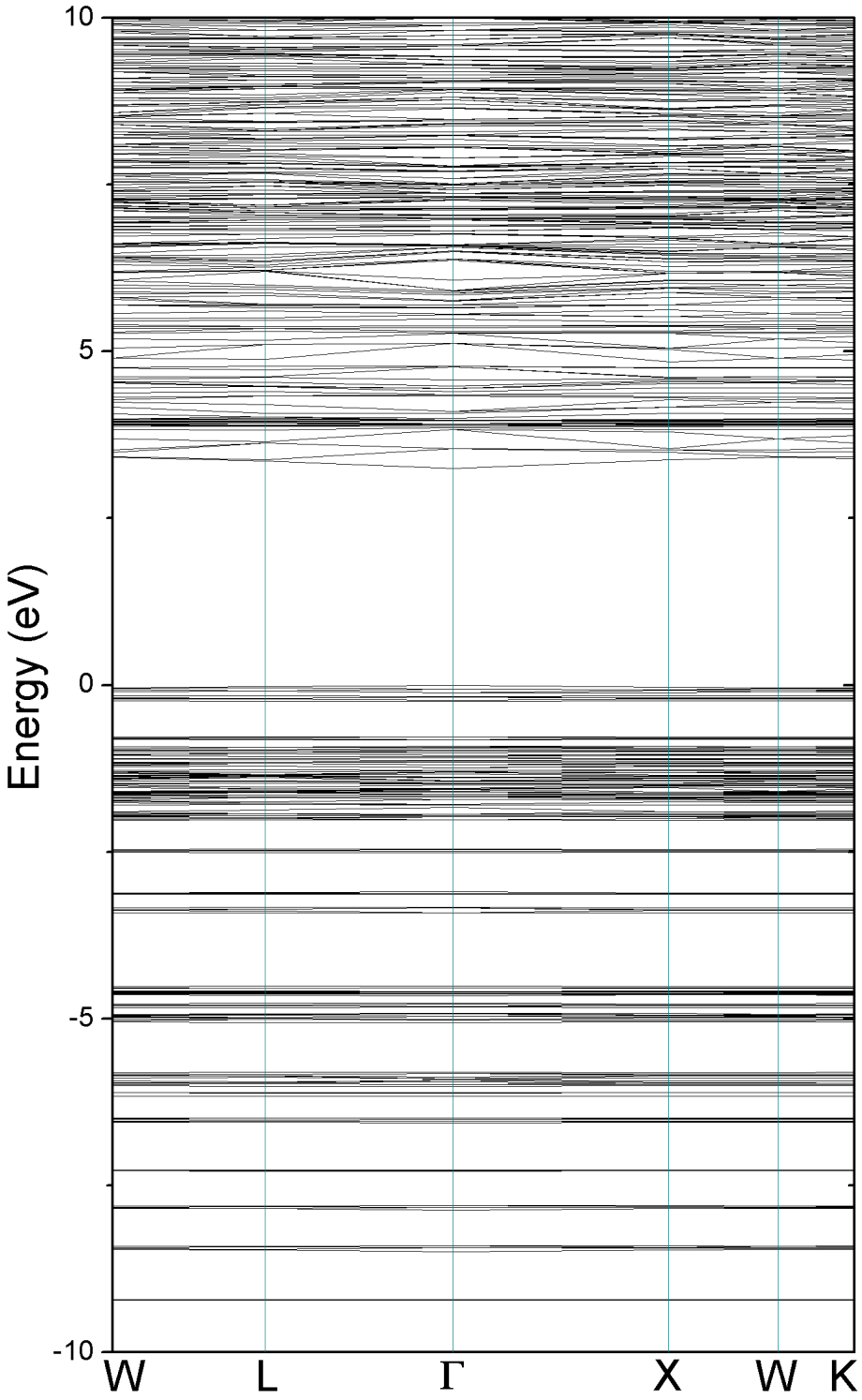

Figure S88. The electronic band structure of (Ca, Br)-90°. The Fermi level is set to zero and placed in the valence band maximum.

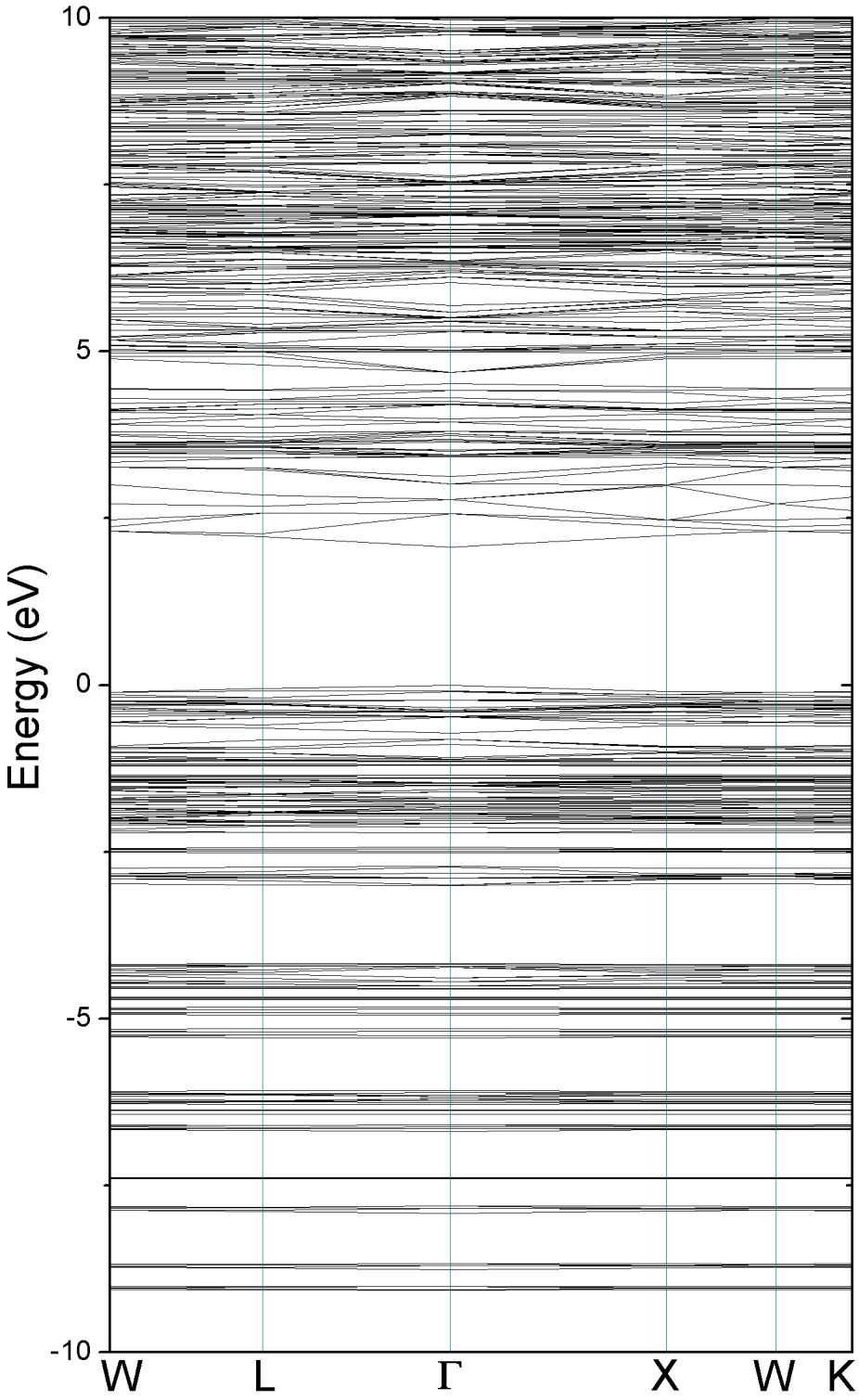

Figure S89. The electronic band structure of (Ca, I)-90°. The Fermi level is set to zero and placed in the valence band maximum.

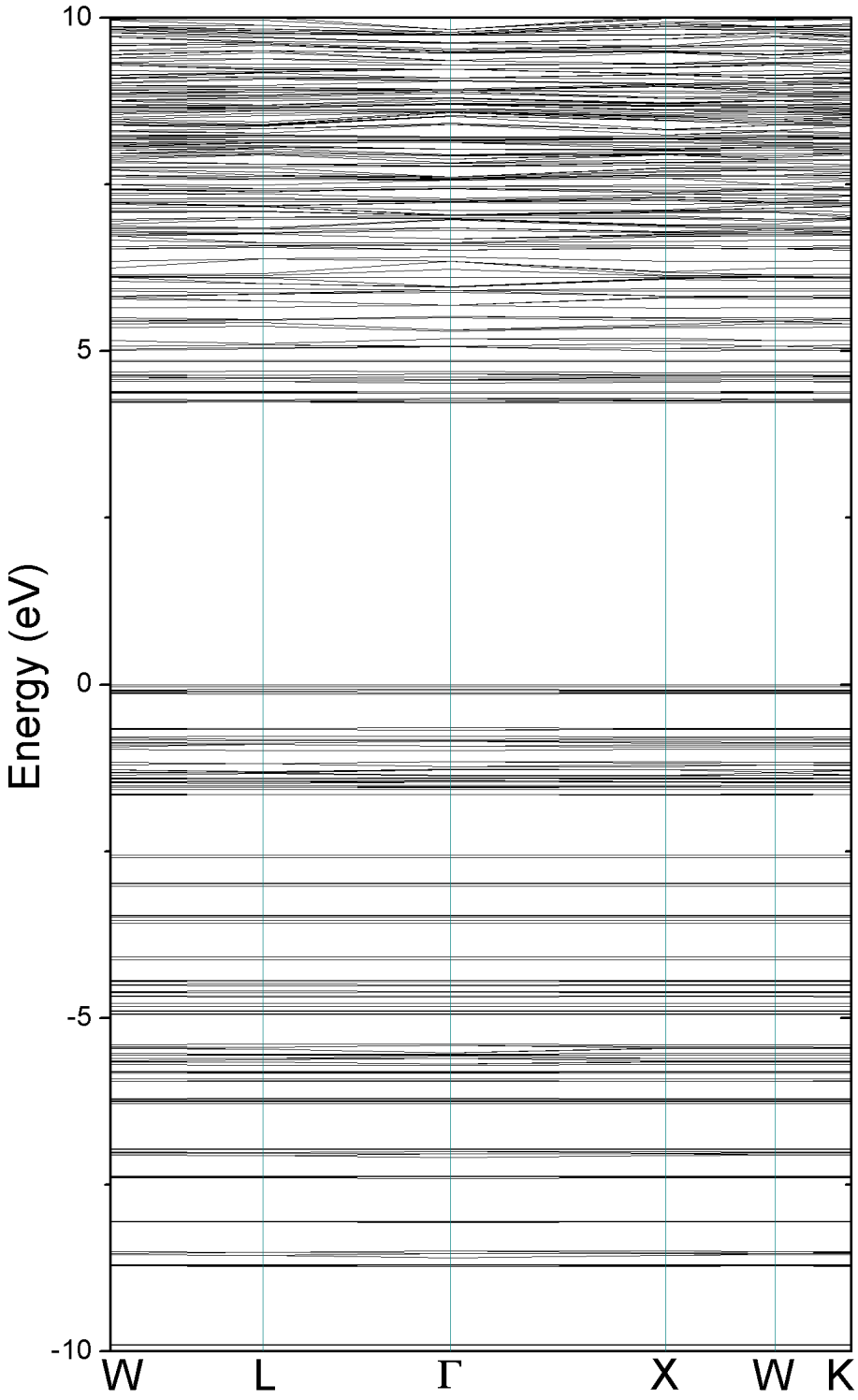

Figure S90. The electronic band structure of (Sr, F)-90°. The Fermi level is set to zero and placed in the valence band maximum.

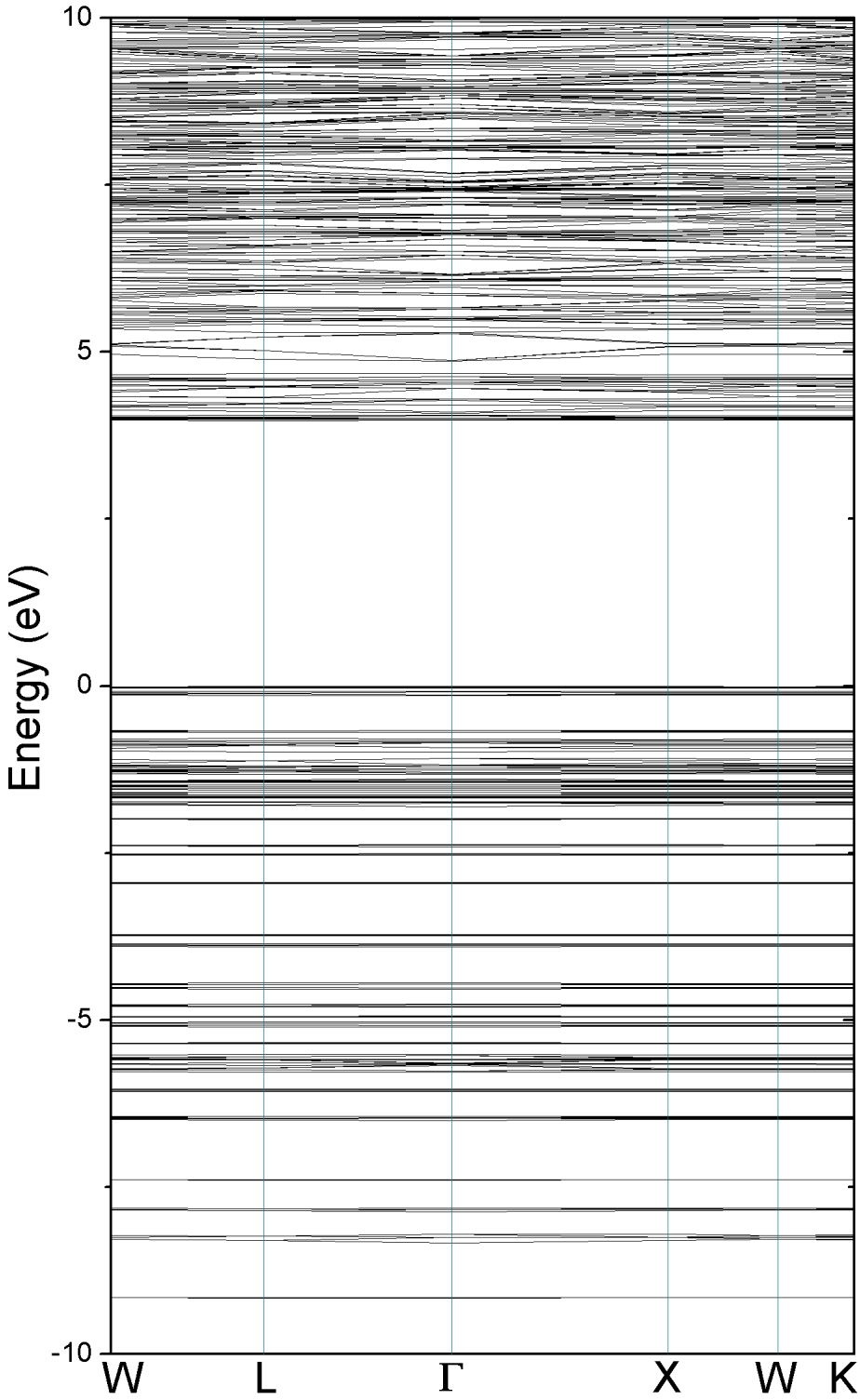

Figure S91. The electronic band structure of (Sr, Cl)-90°. The Fermi level is set to zero and placed in the valence band maximum.

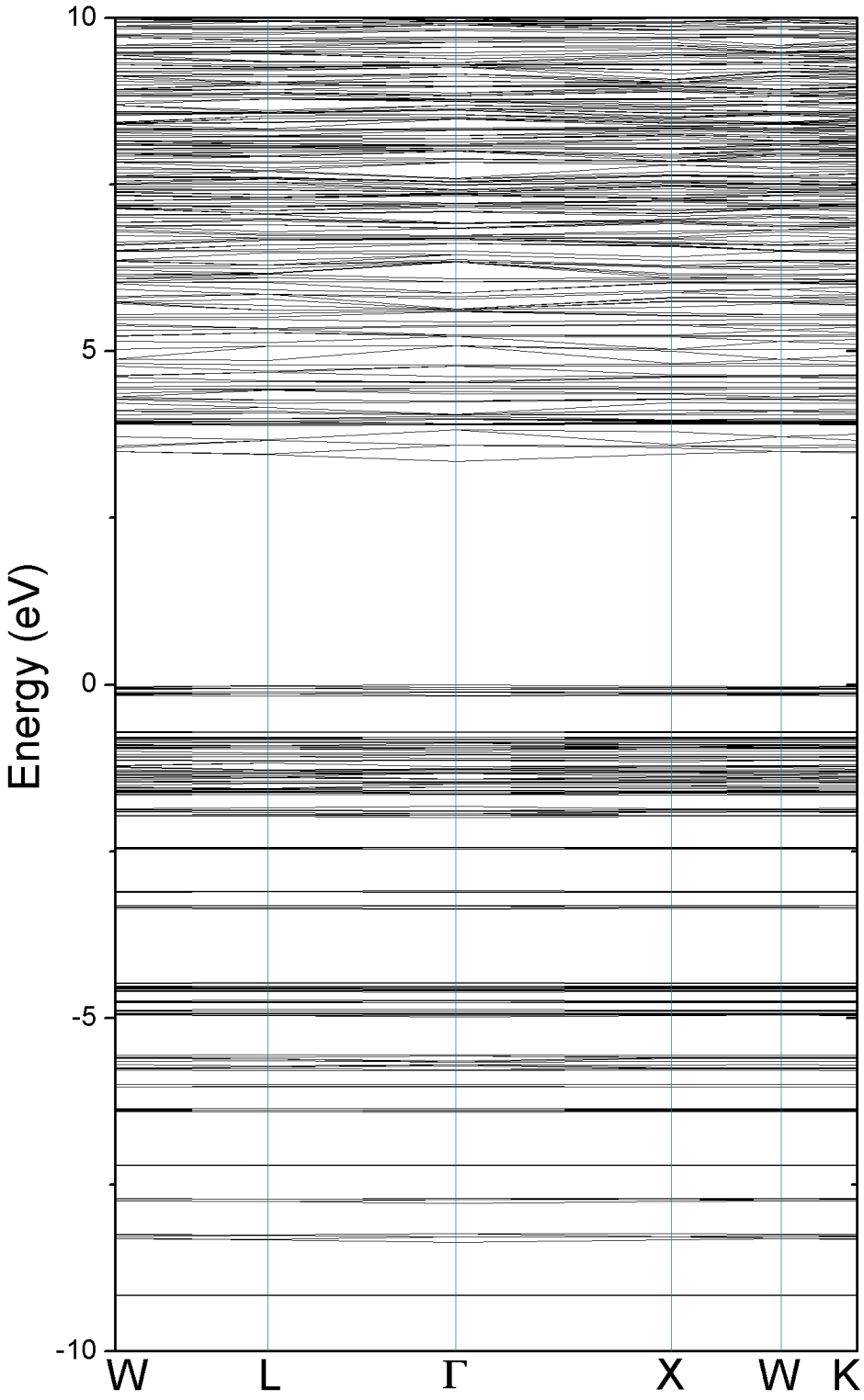

Figure S92. The electronic band structure of (Sr, Br)-90°. The Fermi level is set to zero and placed in the valence band maximum.

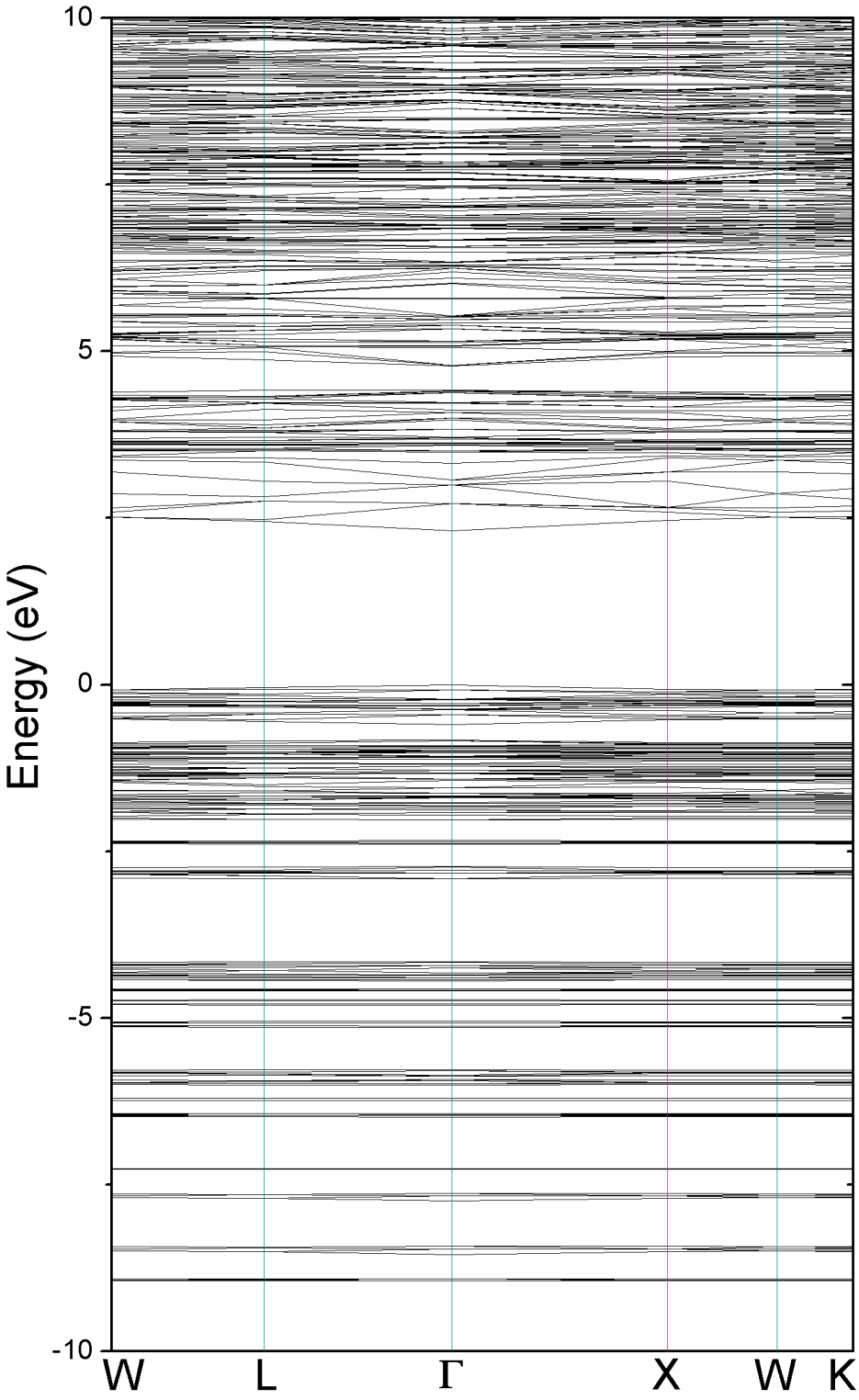

Figure S93. The electronic band structure of (Sr, I)-90°. The Fermi level is set to zero and placed in the valence band maximum.

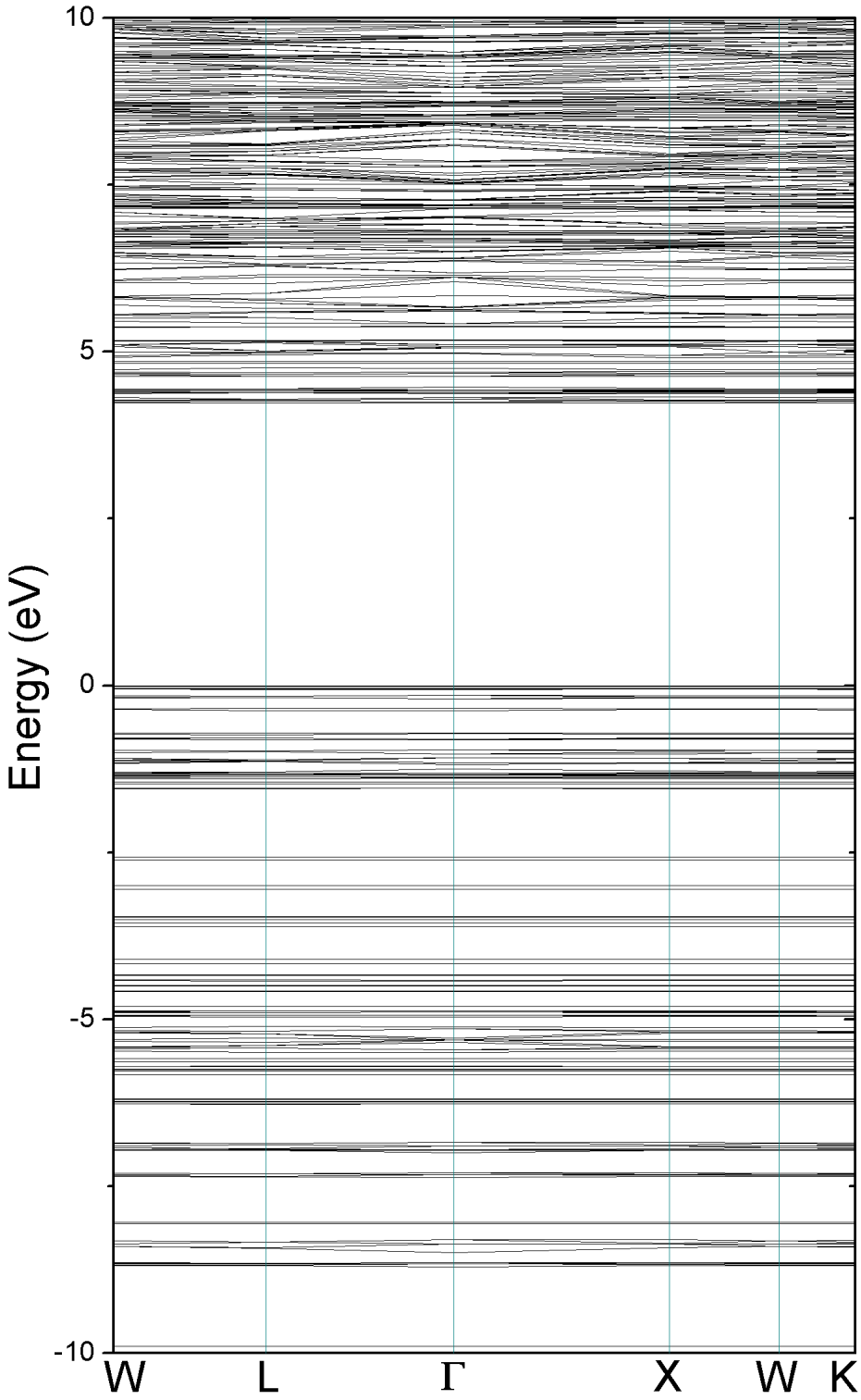

Figure S94. The electronic band structure of (Ba, F)-90°. The Fermi level is set to zero and placed in the valence band maximum.

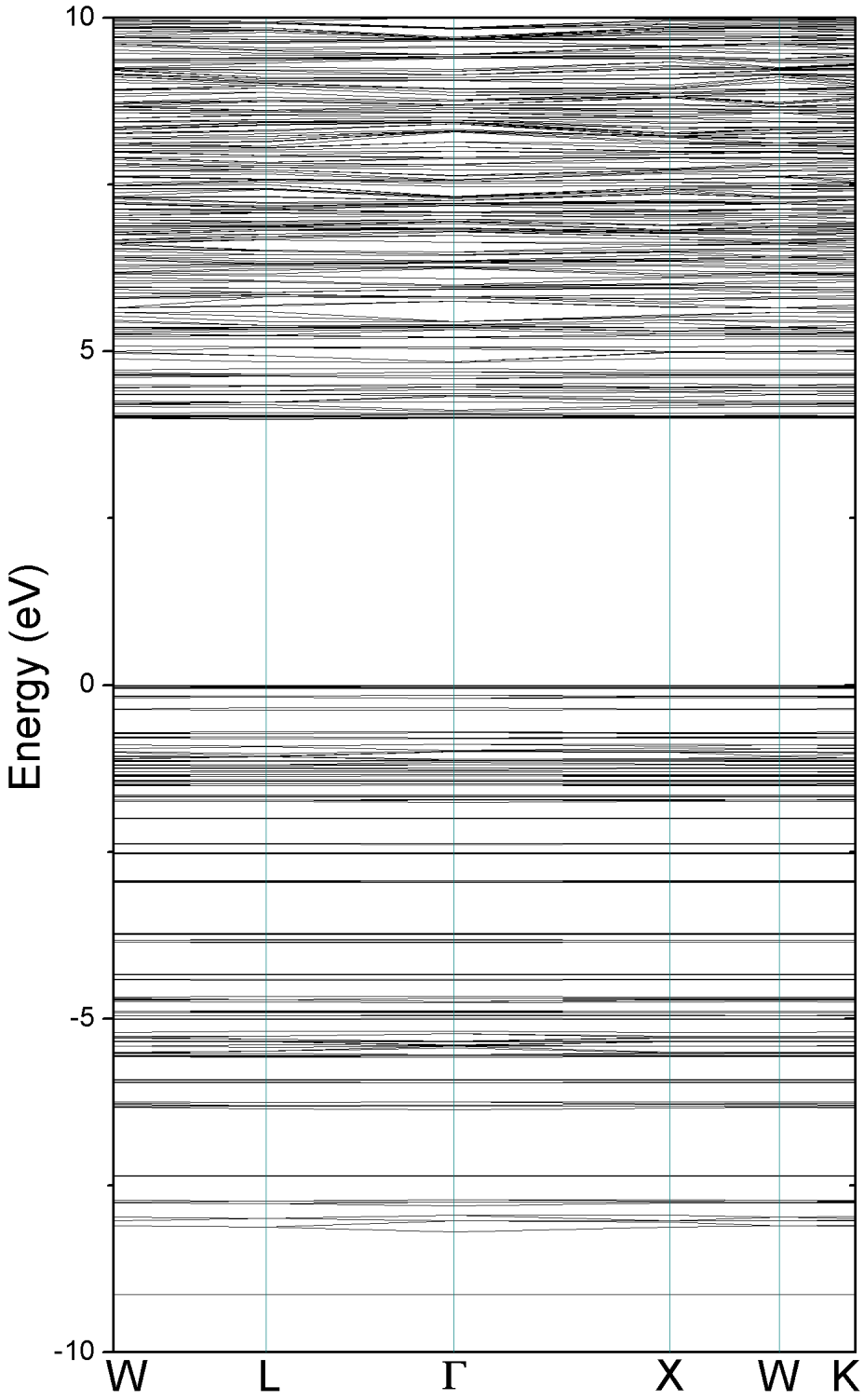

Figure S95. The electronic band structure of (Ba, Cl)-90°. The Fermi level is set to zero and placed in the valence band maximum.

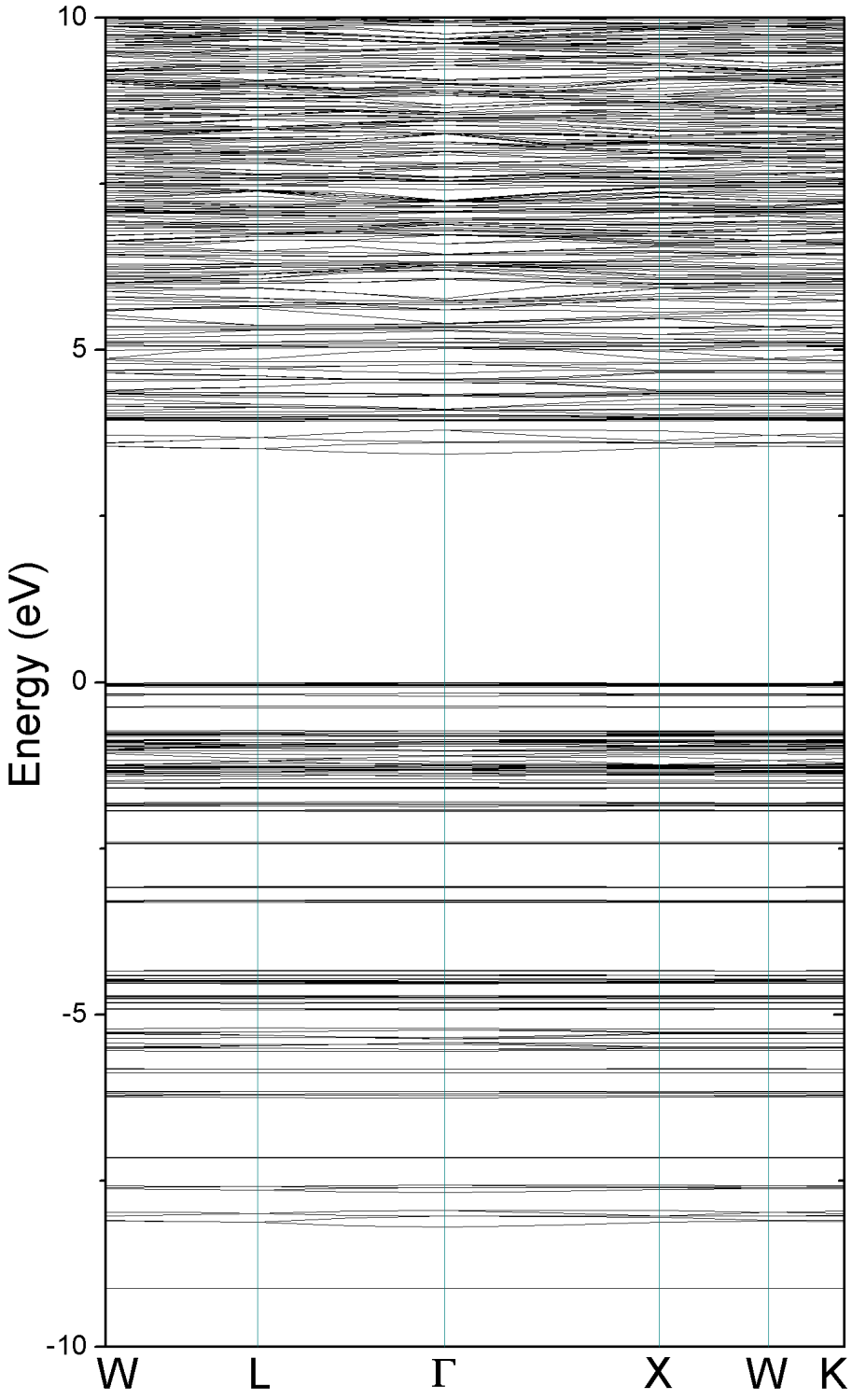

Figure S96. The electronic band structure of (Ba, Br)-90°. The Fermi level is set to zero and placed in the valence band maximum.

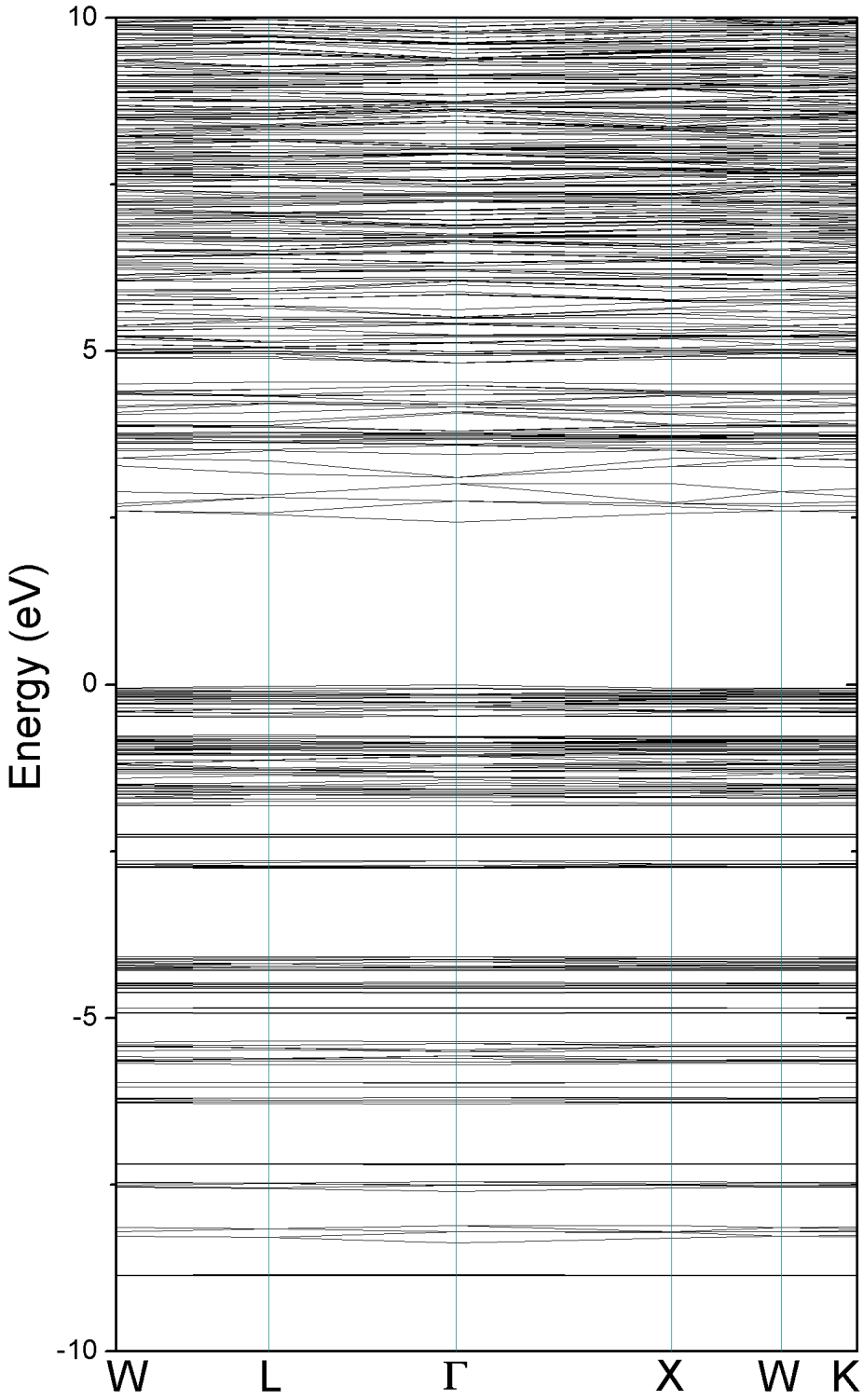

Figure S97. The electronic band structure of (Ba, I)-90°. The Fermi level is set to zero and placed in the valence band maximum.