Band Gap Engineering of $Cs_3Bi_2I_9$ Perovskites with Trivalent Atoms through Dual Metal Cation Approach

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Table S1. Lattice structures and band gaps of $P6_3/mmc$ Cs_3BiXI_9.

X	*a* (Å)	*c* (Å)	Vol. (Å^3)	GGA (eV)	SO (eV)	HFSO (eV)
Al	8.195	20.902	1216	1.82	1.49	2.27
As	8.233	20.702	1215	1.86	1.62	2.19
B	8.401	20.529	1254	1.56	1.38	2.12
Bi	8.335	21.326	1283	2.05	1.67	2.25
Co	8.100	20.842	1184	1.01	0.90	1.85
Ga	8.208	20.714	1208	0.95	0.82	1.60
In	8.269	21.021	1244	1.29	1.11	1.90
Ir	8.134	21.022	1204	1.75	1.28	2.18
La	8.402	21.485	1314	2.22	1.70	2.42
P	8.222	20.463	1198	1.74	1.52	2.10
Sb	8.300	21.088	1258	1.75	1.45	1.98
Sc	8.236	21.051	1236	2.20	1.72	2.46
Y	8.320	21.280	1275	2.28	1.70	2.43

Table S2. Lattice structures and band gaps of $P\bar{3}m1$ Cs_3BiXI_9.

X	*a* (Å)	*c* (Å)	Vol. (Å^3)	GGA (eV)	SO (eV)	HFSO (eV)
Al	8.251	10.118	596	1.17	0.85	1.56
As	8.312	10.184	609	1.49	1.20	1.85
B	8.453	10.190	631	1.55	1.35	2.07
Bi	8.453	10.435	646	1.76	1.19	1.74
Co	8.114	9.947	567	0.62	0.44	0.94
Ga	8.270	10.111	599	0.67	0.50	1.20
In	8.343	10.255	618	0.91	0.69	1.41
Ir	8.168	10.054	580	1.03	0.55	1.49
La	8.513	10.560	662	2.40	1.76	2.51
P	8.289	10.070	599	1.47	1.18	1.75
Sb	8.412	10.375	635	1.60	1.24	1.77
Sc	8.304	10.239	611	2.01	1.42	2.14
Y	8.409	10.403	637	2.17	1.57	2.30

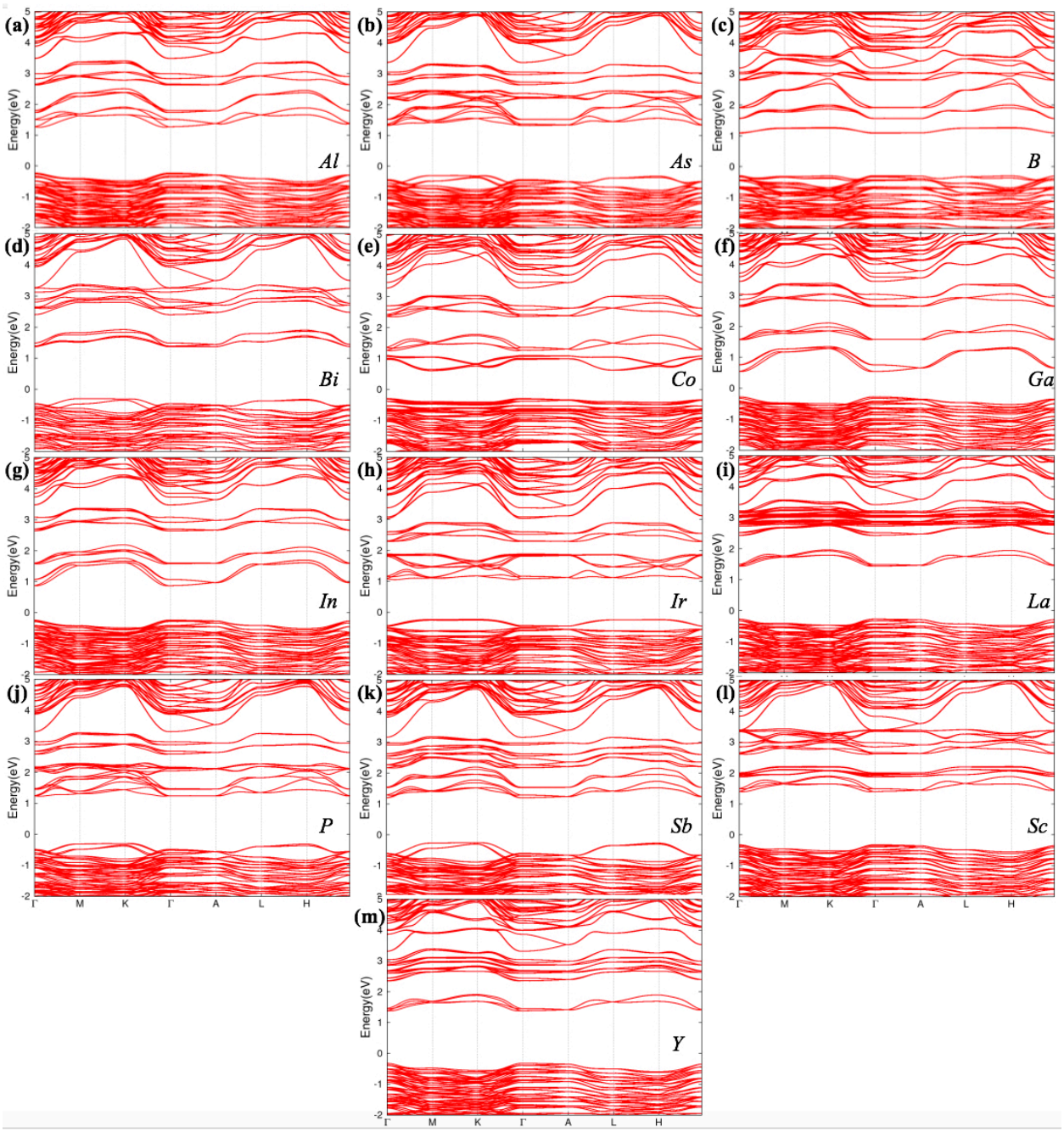

Figure S1. Band structures of $P6_3/mmc$ Cs_3BiXI_9 when X is (a) Al, (b) As, (c) B, (d) Bi, (e) Co, (f) Ga, (g) In, (h) Ir, (i) La, (j) P, (k) Sb, (l) Sc, and (m) Y.

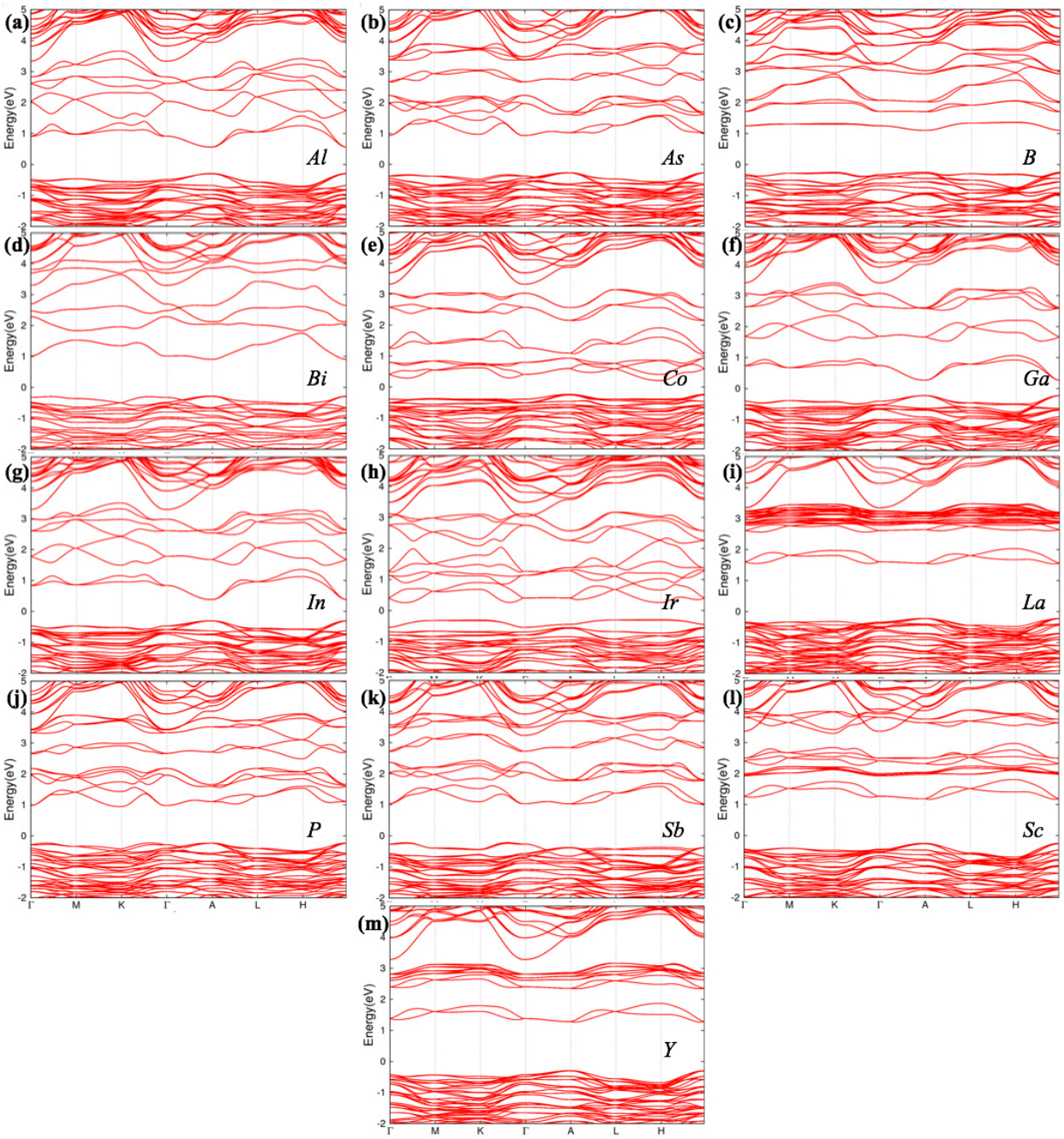

Figure S2. Band structures of $P\bar{3}m1$ Cs_3BiXI_9 when X is (a) Al, (b) As, (c) B, (d) Bi, (e) Co, (f) Ga, (g) In, (h) Ir, (i) La, (j) P, (k) Sb, (l) Sc, and (m) Y.

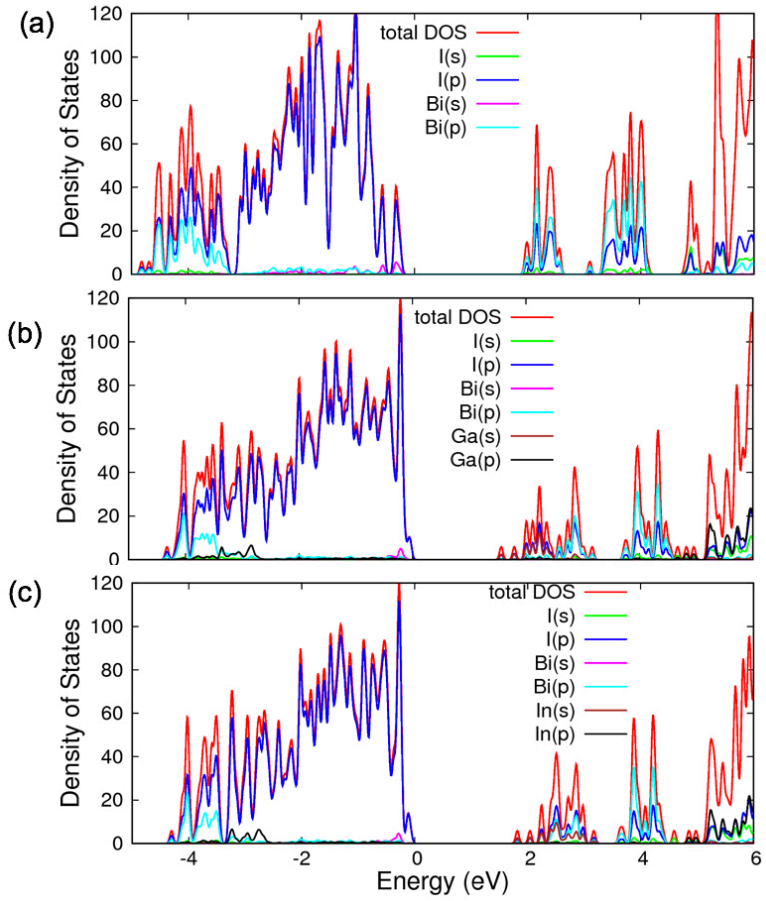

Figure S3. Density of states of (a) $Cs_3Bi_2I_9$ ($P6_3$/mmc) (b) Cs_3BiGaI_9 ($P6_3$/mmc), and (c) Cs_3BiInI_9 ($P6_3$/mmc) with SOC + HSE06.

Table S3. Effective masses of $Cs_3Bi_2I_9$ ($P6_3/mmc$), Cs_3BiGaI_9 ($P6_3/mmc$), and Cs_3BiInI_9 ($P6_3/mmc$) with PBEsol and PBEsol+SOC (SOC)

		$Cs_3Bi_2I_9$	Cs_3BiGaI_9	Cs_3BiInI_9
$m_e^*[100]$	SOC	0.249	0.450	0.319
	PBEsol	0.307	0.404	0.404
$m_e^*[001]$	SOC	1.435	0.375	0.498
	PBEsol	1.772	0.637	0.630
$m_h^*[100]$	SOC	1.114	0.666	0.466
	PBEsol	0.880	0.681	0.590
$m_h^*[001]$	SOC	0.811	0.711	1.059
	PBEsol	0.641	1.622	1.34