Electronic supplementary information – First-principles study on structural, electronic and optical properties of halide double perovskite Cs$_2$AgBX$_6$ (B = In, Sb; X = F, Cl, Br, I)

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Table S1. Total energy (E_{tot}) and total energy per formula unit (fu) of elementary and binary solids with crystalline phase and space group.

Solid	Phase	Space group	Ref.	E_{tot} (Ry)	E_{tot} per fu (Ry)
Cs	fcc	$Fm\bar{3}m$	[1]	-252.261140	-63.065285
	bcc	$Im\bar{3}m$	[1]	-126.130728	-63.065364
Ag	fcc	$Fm\bar{3}m$	[2]	-1182.487992	-295.621998
In	tetragonal	$I4/MMM$	[3]	-266.963872	-133.481936
	orthorhombic	$Fmmm$	[4]	-533.916447	-133.479112
Sb	fcc	$Fm\bar{3}m$	[5]	-739.705388	-184.926347
	tetragonal	$I4/MMM$	[5]	-369.909949	-184.954974
	hexagonal	$R3mh$	[6]	-1109.711095	-184.951849
AgF	cubic	$Fm\bar{3}m$		-1376.882710	-344.220678
AgCl	cubic	$Fm\bar{3}m$	[7]	-1315.916179	-328.979045
AgBr	cubic	$Fm\bar{3}m$	[8]	-1370.075394	-342.518849
AgI	cubic	$Fm\bar{3}m$	[9]	-1445.211938	-361.302985
	hexagonal	$P63mc$	[10]	-722.632490	-361.316245
	zinc blende	$F\bar{4}3m$	[9]	-1445.267447	-361.316862
CsF	cubic	$Fm\bar{3}m$	[12]	-447.652490	-111.913122
	cubic	$Pm\bar{3}m$	[11]	-111.897148	-111.897148
CsCl	cubic	$Fm\bar{3}m$	[12]	-386.585878	-96.646470
	cubic	$Pm\bar{3}m$	[11]	-96.641591	-96.641591
CsBr	cubic	$Fm\bar{3}m$	[12]	-440.706264	-110.176566
	cubic	$Pm\bar{3}m$	[11]	-110.172869	-110.172869
CsI	cubic	$Fm\bar{3}m$	[12]	-515.767250	-128.941813
	cubic	$Pm\bar{3}m$	[11]	-128.940813	-128.940813
InF$_3$	monoclinic	$C12/m1$	[13]	-1118.322334	
InCl$_3$	monoclinic	$C12/m1$		-934.731573	
InBr$_3$	monoclinic	$C12/m1$		-1097.071959	
InI$_3$	monoclinic	$P121/c1$	[14]	-1322.317962	
SbF$_3$	orthorhombic	$Pnma$	[15]	-1324.036195	
SbCl$_3$	orthorhombic	$Pnma$		-1140.329477	
SbBr$_3$	orthorhombic	$Pbnm$		-1302.674655	
SbI$_3$	monoclinic	$P121/c1$		-1527.973965	

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Table S2. Total energy (E_{tot}) of halide double perovskites CsAgBX$_6$ (B = In, Sb; X = F, Cl, Br, I) and elementary substances of Cs, Ag, In, Sb and X$_2$, and the elementary formation energy (E_f) per formula unit (fu).

Compound	E_{tot} (Ry)	E_{tot} (Ry)				ΔE (Ry)	E_f (eV/fu)
		Cs	Ag	In/Sb	X$_2$		
Cs$_2$AgInF$_6$	-847.752926	-126.130728	-1182.487992	-266.963872	-96.918306	-1.7633	-23.9915
Cs$_2$AgInCl$_6$	-756.016824	-126.130728	-1182.487992	-266.963872	-66.573755	-1.0609	-14.4342
Cs$_2$AgInBr$_6$	-837.175734	-126.130728	-1182.487992	-266.963872	-93.671579	-0.9263	-12.6034
Cs$_2$AgInI$_6$	-949.782210	-126.130728	-1182.487992	-266.963872	-131.271802	-0.7321	-9.9613
Cs$_2$AgSbF$_6$	-899.126947	-126.130728	-1182.487992	-369.909949	-96.918306	-1.6643	-22.6443
Cs$_2$AgSbCl$_6$	-807.416302	-126.130728	-1182.487992	-369.909949	-66.573755	-0.9873	-13.4334
Cs$_2$AgSbBr$_6$	-888.595342	-126.130728	-1182.487992	-369.909949	-93.671579	-0.8729	-11.8764
Cs$_2$AgSbI$_6$	-1001.224795	-126.130728	-1182.487992	-369.909949	-131.271802	-0.7017	-9.5469

Table S3. Total energy (E_{tot}) of halide double perovskites CsAgBX$_6$ and the binary compounds of CsX, AgX and BX$_3$, and the binary formation energy (E_f) per formula unit (fu).

Compound	E_{tot} (Ry)	E_{tot} (Ry)			ΔE (Ry)	E_f (eV/fu)
		CsX	AgX	BX$_3$		
Cs$_2$AgInF$_6$	-847.752926	-447.652490	-1376.882710	-1118.322334	-0.1254	-1.7064
Cs$_2$AgInCl$_6$	-756.016824	-386.585878	-1315.916179	-934.731573	-0.0619	-0.8428
Cs$_2$AgInBr$_6$	-837.175734	-440.706264	-1370.075394	-1097.071959	-0.0358	-0.4866
Cs$_2$AgInI$_6$	-949.782210	-515.767250	-1445.267447	-1322.317962	-0.0022	-0.0304
Cs$_2$AgSbF$_6$	-899.126947	-447.652490	-1376.882710	-1324.036195	-0.0710	-0.9657
Cs$_2$AgSbCl$_6$	-807.416302	-386.585878	-1315.916179	-1140.329477	-0.0619	-0.8429
Cs$_2$AgSbBr$_6$	-888.595342	-440.706264	-1370.075394	-1302.674655	-0.0547	-0.7442
Cs$_2$AgSbI$_6$	-1001.224795	-515.767250	-1445.267447	-1527.990350	-0.0267	-0.3636

Table S4. Effective mass of electron (m_e) and hole (m_h) in the three Cartesian directions (x, y, z), and their harmonic mean values calculated by $m^* = 3/(1/m_x + 1/m_y + 1/m_z)$.

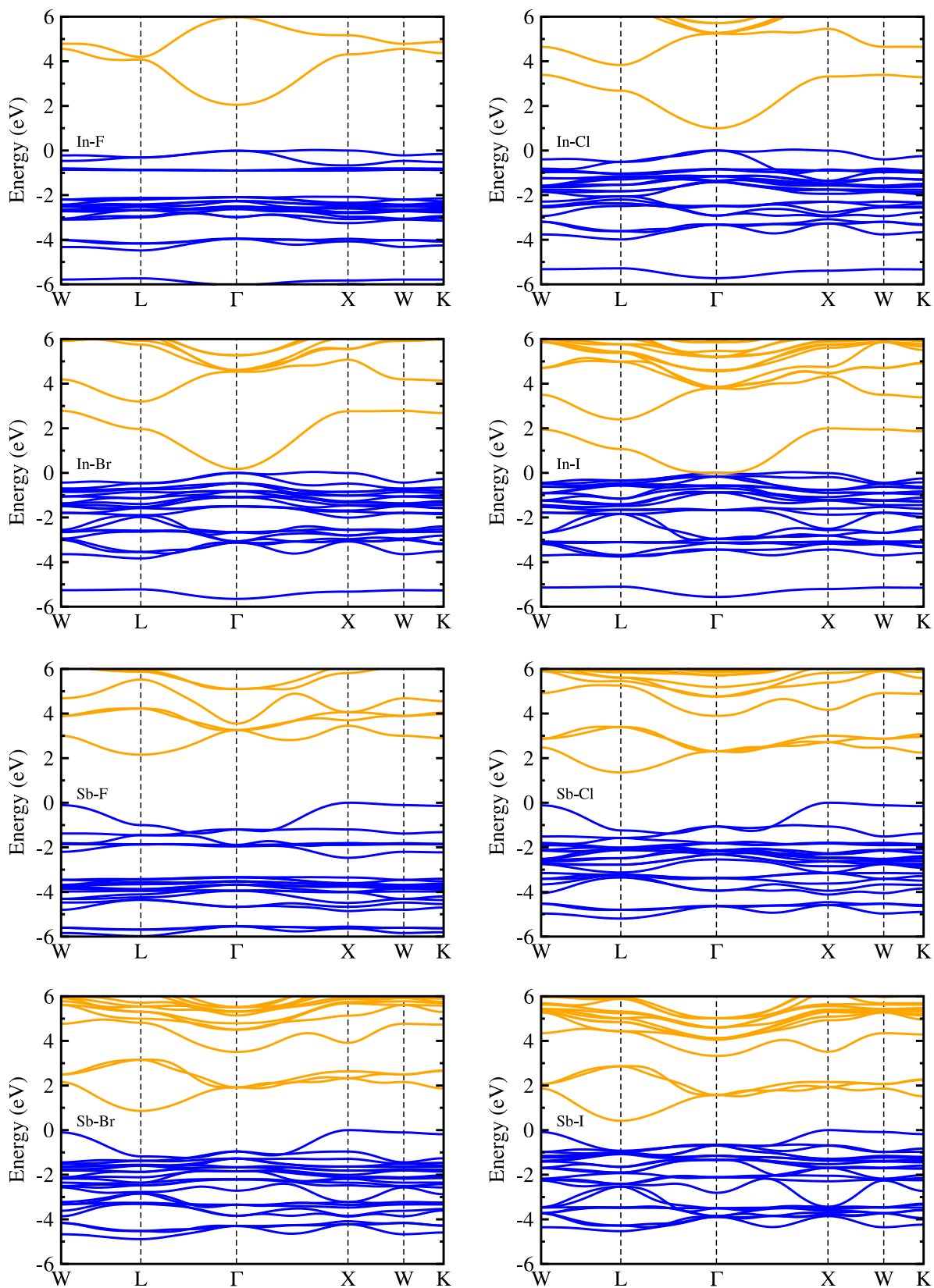| Compound	m_e				m_h			
	x	y	z	m^*	x	y	z	m^*
Cs$_2$AgInF$_6$	0.4090	0.4090	0.4090	0.4090	1.1829	1.1829	1.7791	1.3317
Cs$_2$AgInCl$_6$	0.2716	0.2716	0.2716	0.2716	0.4626	0.4626	0.6030	0.5015
Cs$_2$AgInBr$_6$	0.1614	0.1614	0.1614	0.1614	0.3392	0.3392	0.5077	0.3814
Cs$_2$AgInI$_6$	0.1062	0.1062	0.1062	0.1062	0.2433	0.2433	0.4212	0.2832
Cs$_2$AgSbF$_6$	0.6062	0.5826	0.5769	0.5883	0.5974	1.0447	0.7601	0.7601
Cs$_2$AgSbCl$_6$	0.3646	0.3511	0.3479	0.3544	0.3382	0.5879	0.4294	0.4294
Cs$_2$AgSbBr$_6$	0.2803	0.2659	0.2625	0.2693	0.2911	0.5181	0.3728	0.3728
Cs$_2$AgSbI$_6$	0.2088	0.1895	0.1852	0.1940	0.2536	0.4508	0.3246	0.3246

Figure S1. Electronic band structures of all-inorganic halide double perovskites Cs_2AgBX_6 (B = In, Sb; X = F, Cl, Br, I), calculated with PBE functional.
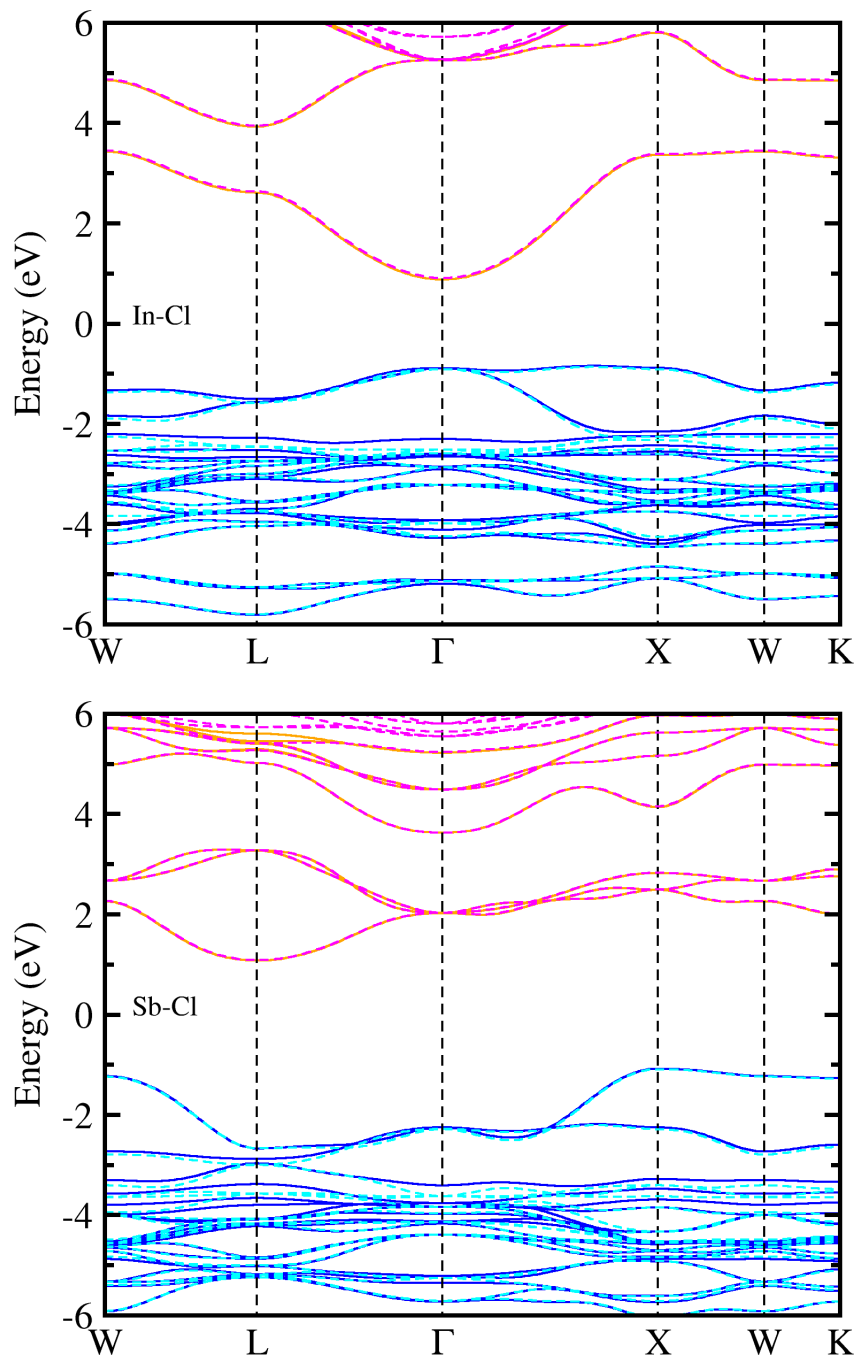
Figure S2. Electronic band structures of all-inorganic halide double perovskites $Cs_2AgInCl_6$ (top) and $Cs_2AgSbCl_6$ (bottom), calculated with PBE (solid lines) and PBE+SOC (dashed lines).
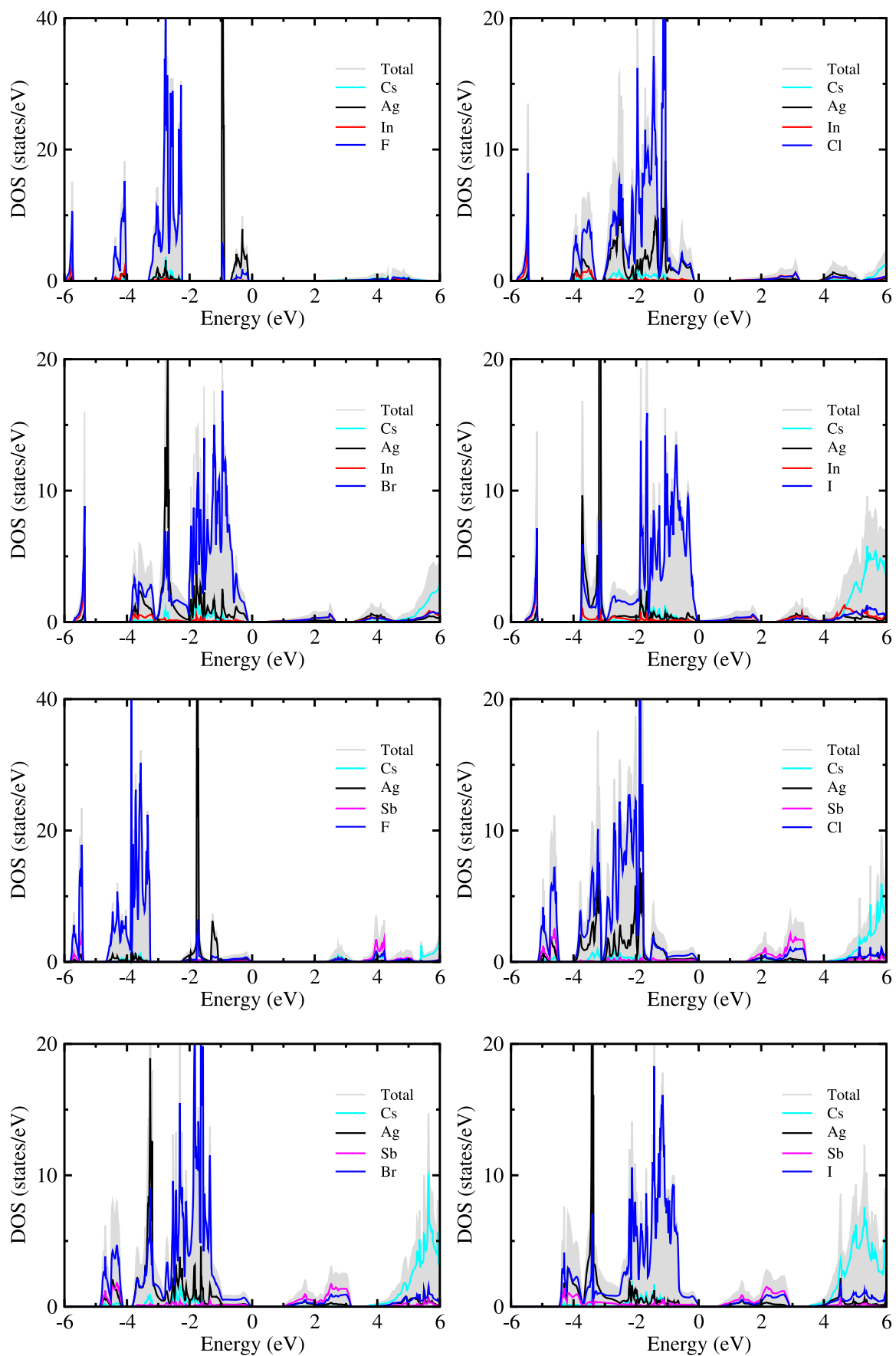
Figure S3. Atom-projected total density of states (DOS) of all-inorganic halide double perovskites Cs_2AgBX_6 (B = In, Sb; X = F, Cl, Br, I), calculated with HSE hybrid functional.

Figure S4. Orbital-resolved partial density of states (PDOS) of all-inorganic halide double perovskites Cs_2AgBX_6 (B = In, Sb; X = F, Cl, Br, I), calculated with HSE hybrid functional.
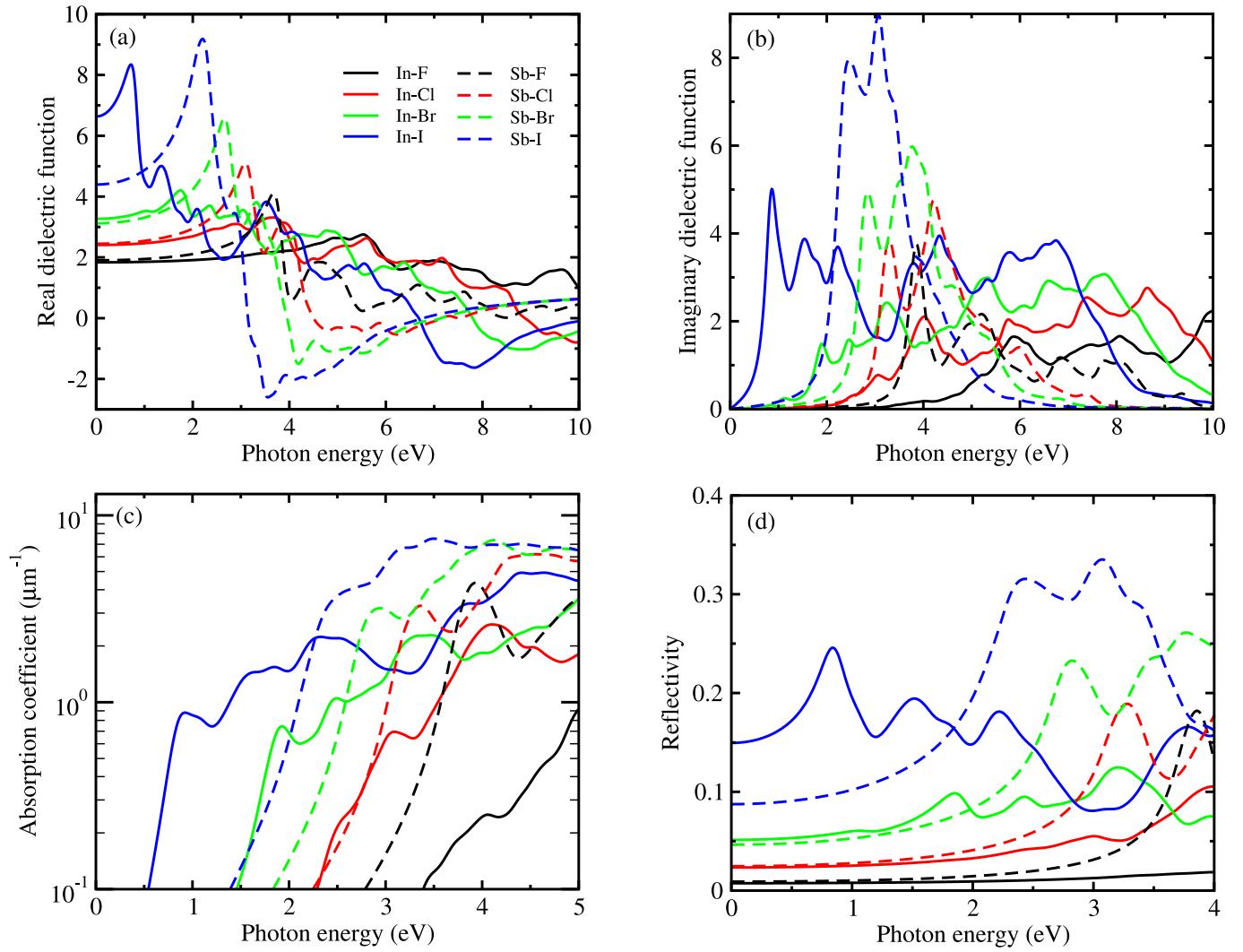
Figure S5. (a) Real and (b) imaginary parts of macroscopic dielectric functions, (c) photo-absorption coefficients, and (d) reflectivity as functions of photon energy for Cs_2AgBX_6 (B = In, Sb; X = F, Cl, Br, I), calculated with the *GW* energies within RPA (i.e., *GW*-RPA).
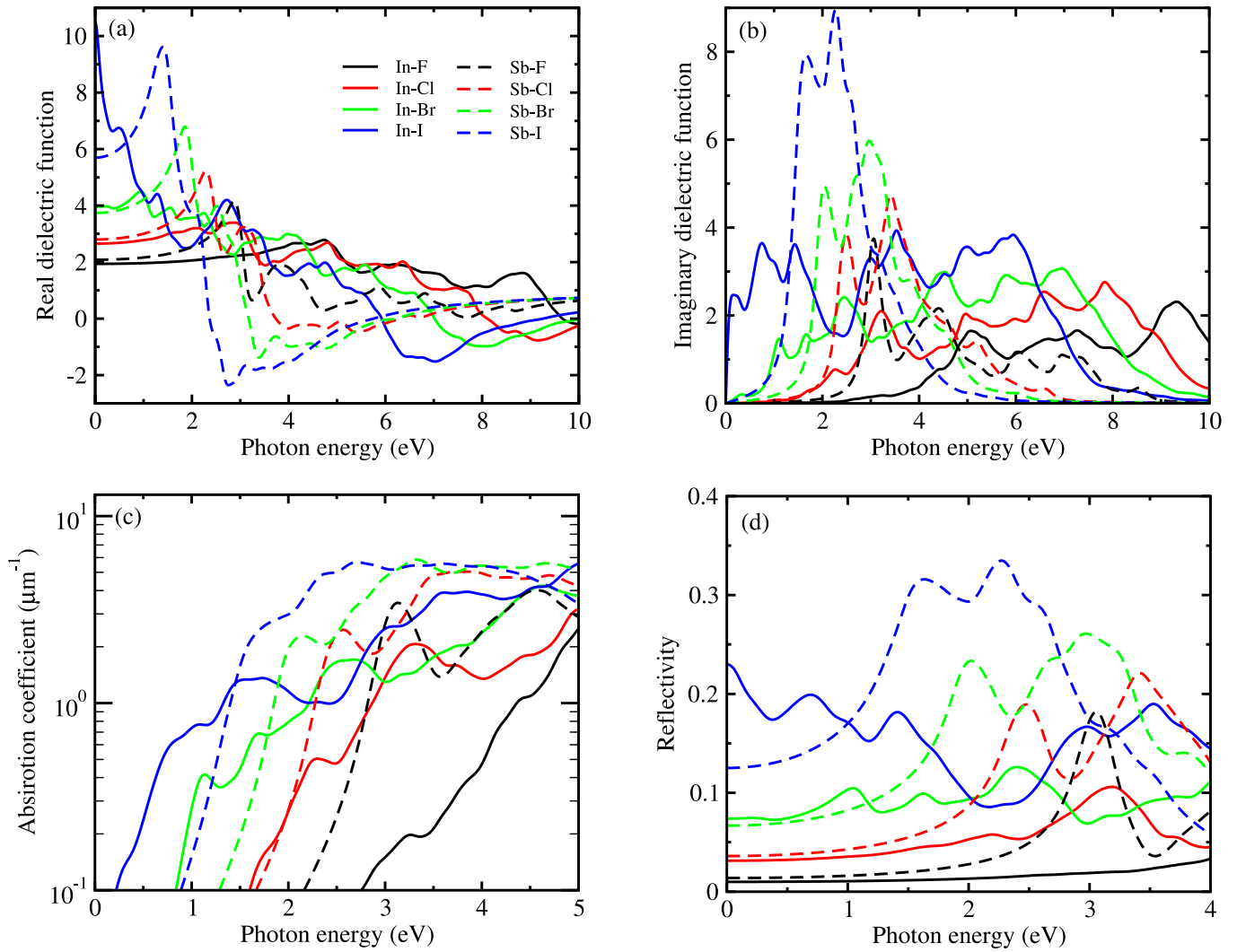
Figure S6. (a) Real and (b) imaginary parts of macroscopic dielectric functions, (c) photo-absorption coefficients, and (d) reflectivity as functions of photon energy for Cs_2AgBX_6 (B = In, Sb; X = F, Cl, Br, I), calculated with the Kohn-Sham energies within RPA (i.e., KS-RPA).

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