Supplementary Information: Screening of Metallic Single-Replacements for Lead-free Perovskites with Intrinsic Photovoltaic Functionalities

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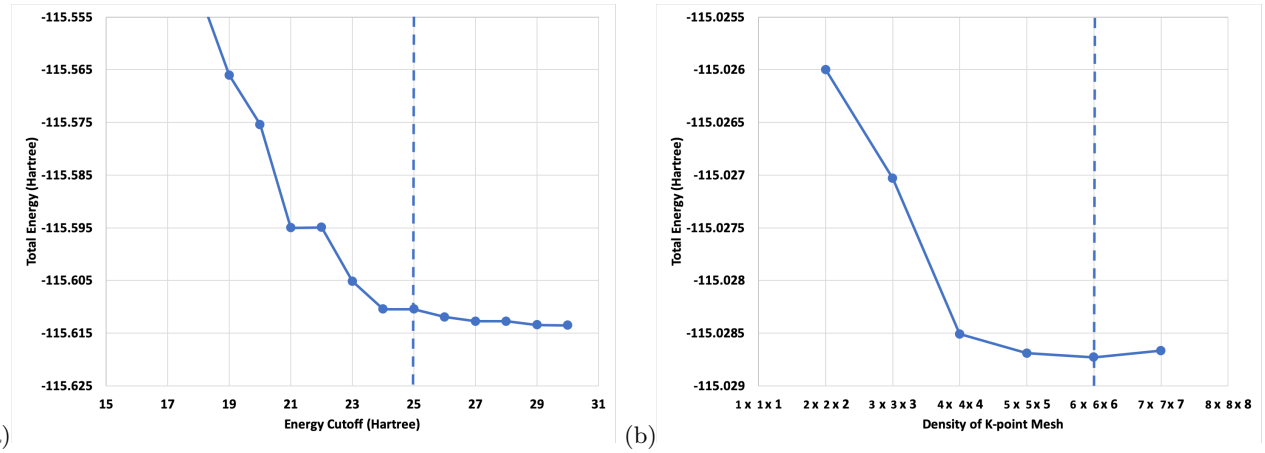
PACS numbers:



(a) (b)

FIG. S1: Convergence studies in the (a) energy cutoff and (b) K-point mesh density. The dotted lines represent the values at which the total energy was converged to five decimal places.

TABLE S1: Structural and Thermal Properties of Calculated Perovskites (PBE unless specified)

Material	Tolerance Factor	Tolerance Factor (prev.)	Octahedral Factor	ΔH (eV/f.u.)	ΔH (eV/f.u.) (prev.)
$MAPbI_3$	0.912	0.9[1]	0.541	-0.381	0.097[2]
$MASiI_3$	1.188	1.2[1]	0.182	-2.101	-2.1[1]
$MAVI_3$	1.033	-	0.359	-4.272	-
$MACrI_3$	1.055	-	0.332	-3.238-	
$MAMnI_3$	1.077	-0.305	0.163	-	
$MAFeI_3$	1.100	-	0.277	1.850	-
$MACoI_3$	1.084	-	0.295	-1.905	-
$MANiI_3$	1.069	-	0.314	-0.626	-
$MACuI_3$	1.055	-	0.332	4.716	-
$MAZnI_3$	1.003	1.1[1]	0.400	-3.129	-0.3[1]
$MAGaI_3$	1.096	1.1[1]	0.282	-3.075	0.2[1]
$MAGeI_3$	1.055	1.0[1]	0.332	-3.537	0.1[1]
$MAMoI_3$	1.069	-	0.314	-2.612	-
$MARhI_3$	1.079	-	0.302	6.095	-
$MARuI_3$	1.073	-	0.309	-4.680	-
$MAPdI_3$	1.010	-	0.391	-1.116	-
$MAInI_3$	1.030	-	0.364	-2.612	-
$MASnI_3$	1.069	0.9[1]	0.314	-3.342	-0.2[1]
$MASbI_3$	0.957	-	0.468	3.401	-
$MAWI_3$	1.044	-	0.345	-3.401	-
$MAReI_3$	1.092	-	0.286	2.014	-
$MAIrI_3$	1.073	-	0.309	-0.299	-
$MAPtI_3$	1.030	-	0.468	-4.626	-
$MABiI_3$	0.957	-	0.468	-117.662	-
$MAZnCl_3$	1.065	-	0.527	-13.70	-

[1] R. Ali, G.-J. Hou, Q.-B. Zhu, Z.-G. Yan, Q.-R. Zheng and G. Su, *J. Mater. Chem. A*, 2018, **6**, 9220–9227.
[2] G. P. Nagabhushanaa, R. Shivaramaiaha and A. Navrotsky, *J. of App. Physics.*, 2016, **28**, 77177721.

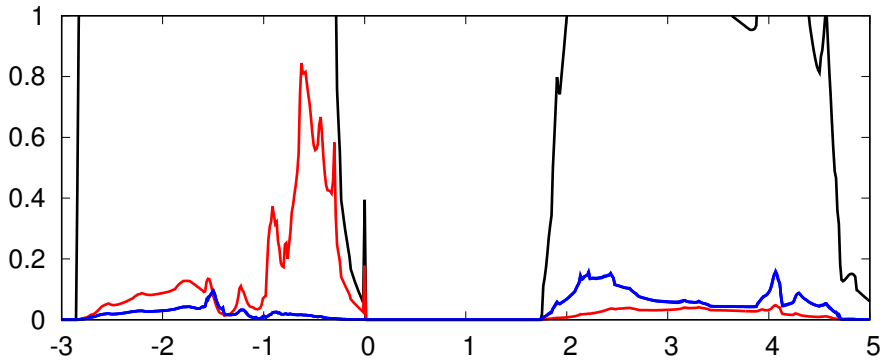


FIG. S2: Total Density of States for $MAPbI_3$ with the partial Density of States of Pb in red and I in blue.

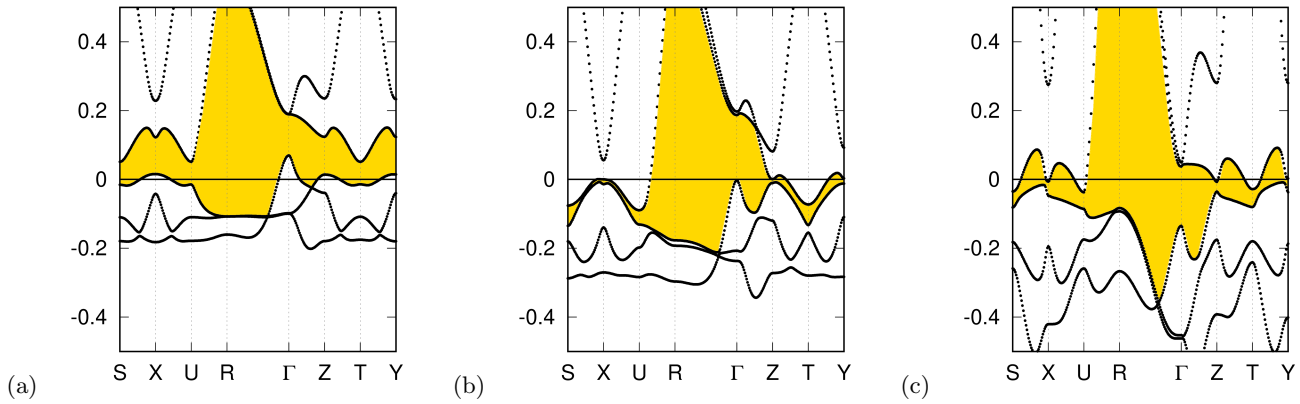


FIG. S3: A close up on (a) $MAMnI_3$, (b) $MAFeI_3$, (c) $MARuI_3$ whose band gaps opened up when triiodide is replaced with tribromide. The yellow region represents the band gap.

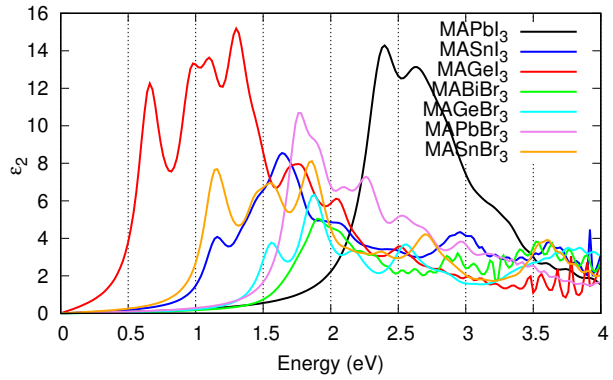


FIG. S4: Imaginary part of the calculated dielectric function, ε_2 of $MAPbI_3$ in black, $MASnI_3$ in blue, $MAGeI_3$ in red, $MABiBr_3$ in green, $MAGeBr_3$ in cyan, $MAPbBr_3$ in pink, $MASnBr_3$ in orange

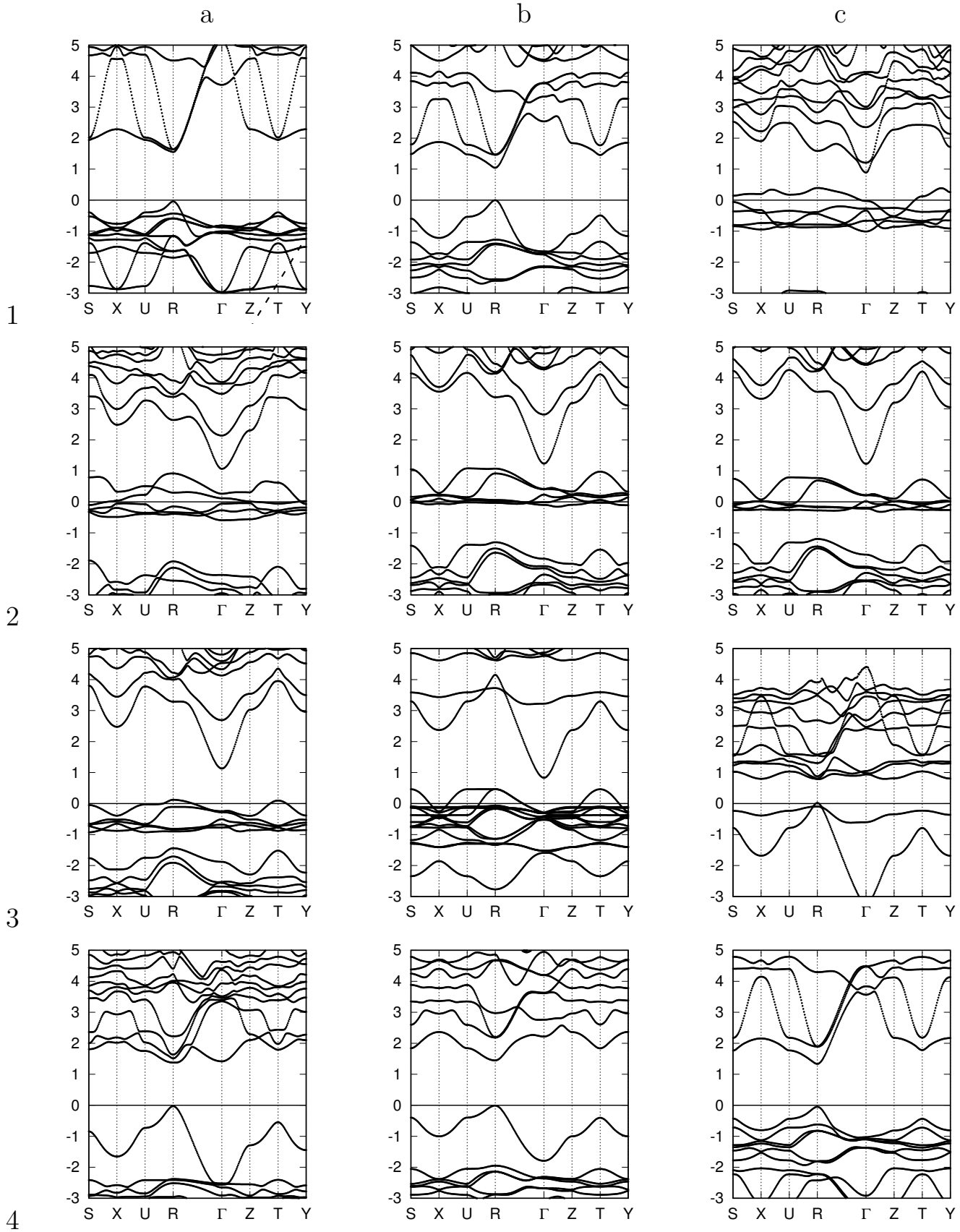


FIG. S5: Band structures shifted so the Fermi energy is at 0 eV
1a: $MAPbI_3$ 1b: $MASiI_3$ 1c: $MAVI_3$
2a: $MACrI_3$ 2b: $MAMnI_3$ 2c: $MAFeI_3$
3a: $MACoI_3$ 3b: $MANiI_3$ 3c: $MACuI_3$
4a: $MAZnI_3$ 4b: $MAGaI_3$ 4c: $MAGeI_3$

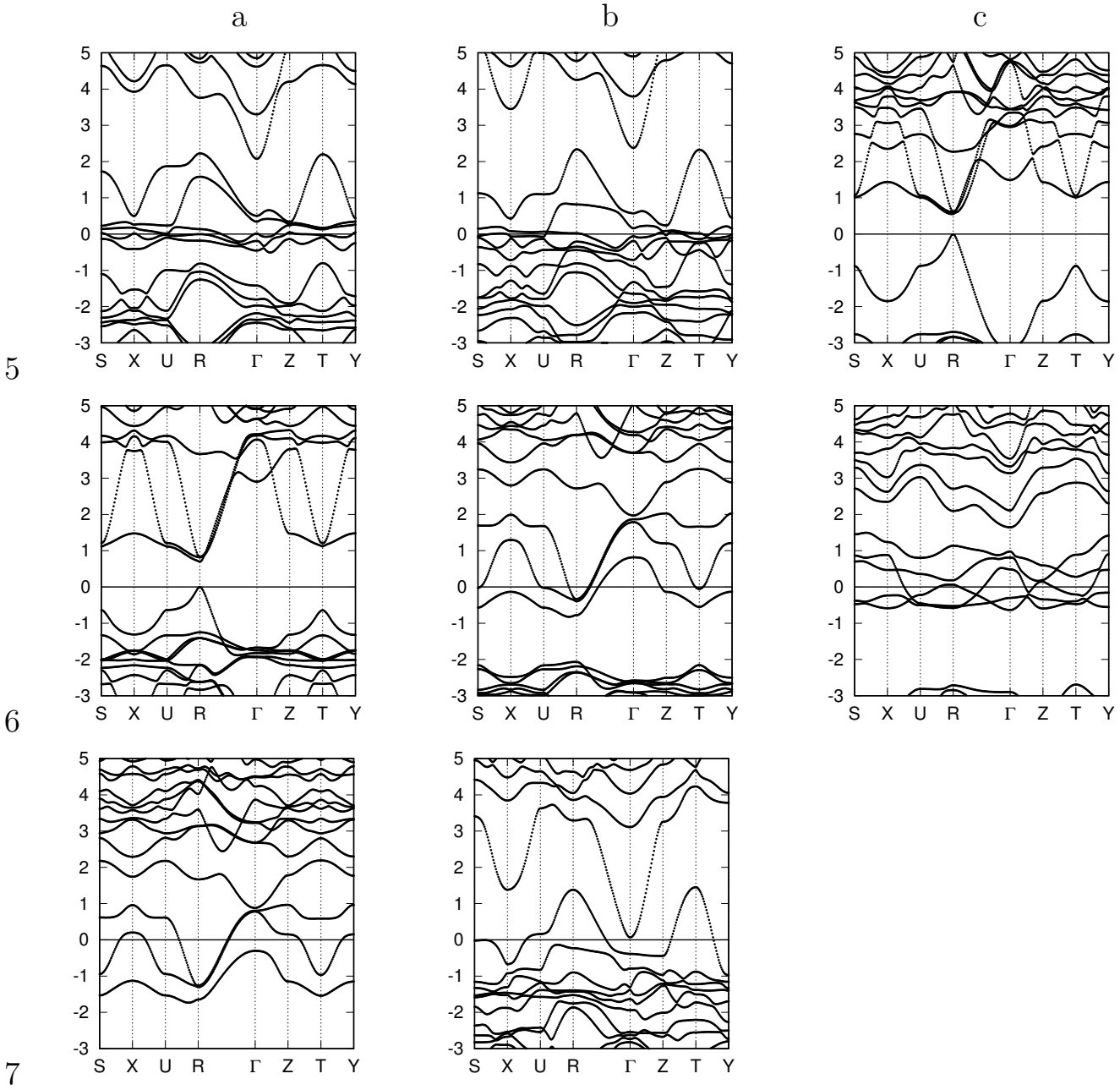


FIG. S6: Band structures shifted so the Fermi energy is at 0 eV
5a: $MARuI_3$ 5b: $MARhI_3$ 5c: $MAInI_3$
6a: $MASnI_3$ 6b: $MASbI_3$ 6c: $MAWI_3$
7a: $MABiI_3$ 7b: $MAPtI_3$

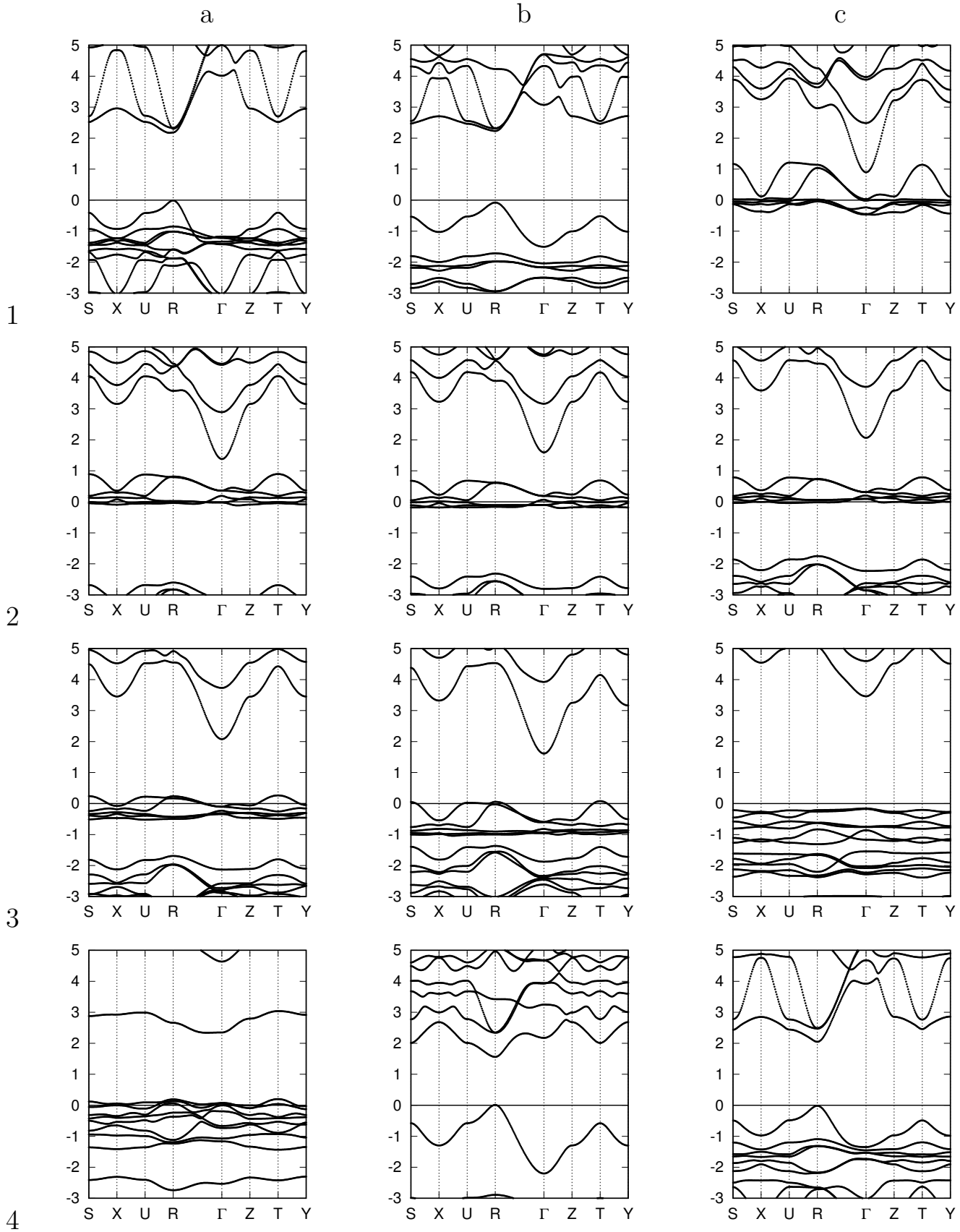


FIG. S7: Band structures shifted so the Fermi energy is at 0 eV
1a: $MAPbBr_3$ 1b: $MASiBr_3$ 1c: $MAVBr_3$
2a: $MACrBr_3$ 2b: $MAMnBr_3$ 2c: $MAFeBr_3$
3a: $MACoBr_3$ 3b: $MANiBr_3$ 3c: $MACuBr_3$
4a: $MAZnBr_3$ 4b: $MAGaBr_3$ 4c: $MAGeBr_3$

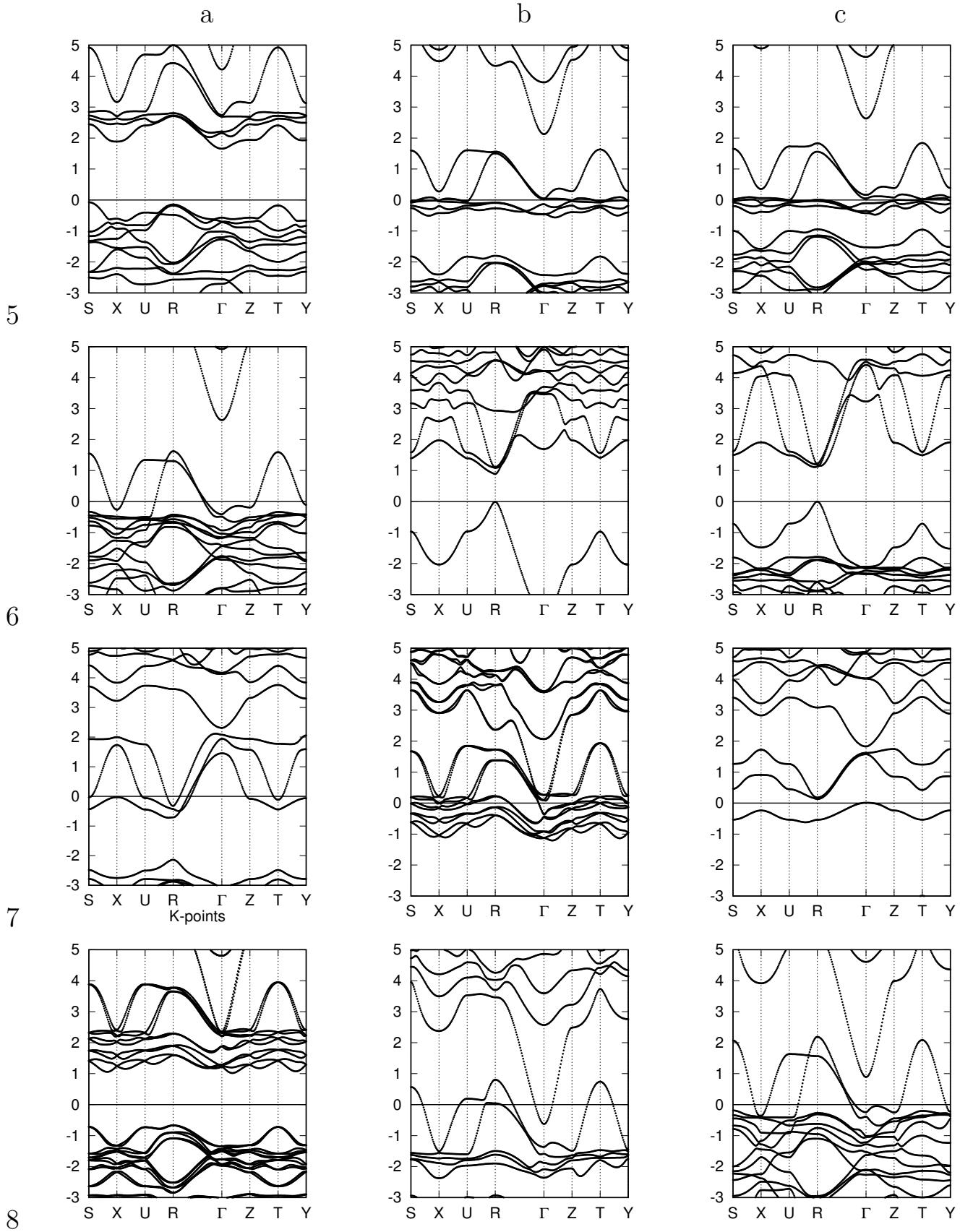


FIG. S8: Band structures shifted so the Fermi energy is at 0 eV
5a: $MAMoBr_3$ 5b: $MARuBr_3$ 5c: $MARhBr_3$
6a: $MAPdBr_3$ 6b: $MAInBr_3$ 6c: $MASnBr_3$
7a: $MASbBr_3$ 7b: $MAWBr_3$ 7c: $MABiBr_3$
8a: $MAReBr_3$ 8b: $MAIrBr_3$ 8c: $MAPtBr_3$