Supplementary Information for

Electronic Structures and Charge Transport Mobilities in Hybrid Organic-inorganic Mixed Sn-Pb Alloyed Perovskites

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S1. Structural properties

Table S1. Calculated lattice constants (a, b, and c), angles between the base vectors ($\alpha, \beta,$ and γ), and structural volume (V) in the $FA_{0.75}Cs_{0.25}Sn_{1-x}Pb_xI_3$ mixed perovskites with a 2×2×2 supercell.

Pb content	a (Å)	b (Å)	c (Å)	α (°)	β (°)	γ (°)	V (Å^3)
x = 0.0	12.780	12.864	12.775	90.00	90.00	90.00	2100.2345
x = 0.125	14.355	12.763	12.671	89.50	85.65	90.64	2321.4902
x = 0.25	14.337	12.752	12.643	89.43	86.47	91.34	2311.4618
x = 0.375	14.351	12.744	12.634	89.28	86.72	91.08	2310.6214
x = 0.5	14.392	12.900	12.518	90.00	88.89	89.99	2324.0518
x = 0.625	14.329	12.938	12.559	89.60	85.79	90.57	2328.2954
x = 0.75	14.666	12.661	12.623	90.79	89.09	90.53	2343.9172
x = 0.875	14.680	12.621	12.657	90.55	89.08	90.57	2345.0418

Table S2 Estimated B-I-B angles of the $FA_{0.75}Cs_{0.25}Sn_{1-x}Pb_xI_3$ mixed perovskites with a 2×2×2 supercell in the *ab* direction (θ_{ab}) and *z* direction (θ_z).

Pb content	θ_{ab} (°)	θ_z (°)
x = 0.0	162.0012	159.3035
	162.0012	159.3035
	162.8436	163.6155
	162.8436	163.6155
x = 0.125		151.7160
		157.4875
x = 0.25	162.9221	148.3039
	172.7045	155.9225
		158.6629
x = 0.375	164.8589	147.9664
		158.4977
		158.6280
x = 0.5	156.5134	165.8015
	156.5134	166.9284
x = 0.625	158.5985	142.1254
	158.5985	151.7415
	161.5770	
x = 0.75	145.275	146.4781
	155.5197	146.5527
		148.7081
		160.6034
x = 0.875	146.8620	146.4402
	153.5496	147.3735
		148.1203
		162.1327

Table S3. Estimated B-I bond lengths of the $FA_{0.75}Cs_{0.25}Sn_{1-x}Pb_xI_3$ mixed perovskites with a 2×2×2 supercell. The B-I bond length is in Å unit.

x = 0.0		x = 0.125		x = 0.25		x = 0.375		x = 0.5		x = 0.625		x = 0.75		x = 0.875	
Sn-I	Pb-I	Sn-I	Pb-I	Sn-I	Pb-I	Sn-I	Pb-I	Sn-I	Pb-I	Sn-I	Pb-I	Sn-I	Pb-I	Sn-I	Pb-I
2.95392		2.91928	3.02175	2.93954	3.02811	2.99497	2.98423	2.95449	3.15751	3.00279	3.00128	3.05219	3.00036	3.11295	3.02139
2.95392		2.97706	3.16172	2.94403	3.03547	2.99685	3.02303	2.9681	3.18228	3.04048	3.01909	3.10951	3.00761	3.12458	3.04271
2.95392		2.98984	3.22605	2.96601	3.15124	3.01837	3.02315	2.9681	3.18233	3.08336	3.03438	3.12239	3.01489	3.28188	3.05835
2.95877		3.00754	3.3446	2.96816	3.15311	3.044	3.12096	2.96892	3.19087	3.19731	3.04614	3.14265	3.03679	3.31064	3.0606
2.95877		3.02694	3.40114	2.98155	3.20547	3.08568	3.15424	2.969	3.19096	3.24798	3.05715	3.24487	3.06188		3.07756
2.95877		3.03697	3.67987	2.98526	3.22062	3.09056	3.16765	3.14818	3.20457	3.43002	3.07952	3.3063	3.06383		3.09381
2.95877		3.0395		2.99249	3.33381	3.11483	3.22941	3.14823	3.20461	3.48018	3.09578	3.30825	3.0754		3.10938
3.19927		3.0534		3.0074	3.36832	3.13563	3.26865	3.15381	3.31303		3.12179	3.40452	3.09056		3.1469
3.19927		3.05355		3.02278	3.40341	3.30986	3.28448	3.15384	3.31312		3.15314		3.10342		3.15535
3.20702		3.05922		3.0334	3.41663	3.3332	3.28532	3.31832	3.33585		3.18462		3.10448		3.1776
3.20702		3.09531		3.07609	3.66052	3.33832	3.29448	3.31833	3.33596		3.21167		3.13497		3.18373
3.20702		3.33626		3.09844	3.68909	3.35352	3.30915	3.31943	3.3457		3.22109		3.15818		3.18846
3.20702		3.35705		3.12615		3.40514	3.37239	3.31951	3.34576		3.23496		3.18422		3.20333
3.21036		3.39576		3.26728		3.46837	3.39613	3.48119	3.35515		3.23915		3.20739		3.21181
3.21036		3.4115		3.32624		3.47506	3.41324	3.48125	3.35523		3.2784		3.2116		3.21978
3.21036		3.44383		3.33678		3.50541					3.28091		3.22691		3.23243
3.21036		3.45076		3.38488							3.28636		3.2323		3.23824
3.21696		3.45608		3.43087							3.33238		3.25395		3.24457
3.21696		3.46017		3.45229							3.33673		3.26412		3.26188
3.21696				3.47626							3.35099		3.26735		3.26513
3.24084				3.50889							3.36524		3.27457		3.2676
3.24084				3.51504							3.37892		3.28693		3.2718
3.24084											3.38967		3.30979		3.27296
3.24084											3.41798		3.32600		3.30973
3.24128											3.48184		3.36432		3.3174
3.24128											3.62627		3.43915		3.31944
3.24128													3.43948		3.3583
3.24235													3.44231		3.36052
3.24235													3.53936		3.3934
3.24235													3.57092		3.4200
3.24235													3.59681		3.5238
3.2622															3.55709
3.2622															3.59164
3.2622															
3.51533															
3.51533															
3.51533															
3.2001		3.2162		3.2207		3.2256		3.2227		3.2335		3.2303		3.2439	
Average value (Å)															

Table S4. Experimental results of lattice constants (*a*, *b*, and *c*) and structural phases in the $FASn_{1-x}Pb_xI_3$, $FA_{0.75}Cs_{0.25}Sn_{1-x}Pb_xI_3$ and $FA_{0.83}Cs_{0.17}Sn_{1-x}Pb_xI_3$ alloyed perovskites measured at room temperature [S1-S4].

Pb content	$FASn_{1-x}Pb_xI_3$ [S1]				$FASn_{1-x}Pb_xI_3$ [S3]			
	a (Å)	*b* (Å)	*c* (Å)	Phase	*a* (Å)	*b* (Å)	*c* (Å)	Phase
x = 0.0	6.313	8.936	8.913	Amm2	6.3074	6.3074	6.3074	$Pm\bar{3}m$
x = 0.125	6.322	8.938	8.921	Amm2				
x = 0.25	6.321	8.934	8.912	Amm2	6.3158	6.3158	6.3158	$Pm\bar{3}m$
x = 0.5	6.416	8.965	8.950	Amm2	6.344	6.344	6.344	$Pm\bar{3}m$
x = 0.625	6.415	8.960	8.942	Amm2				
x = 0.75	6.415	8.960	8.942	Amm2	6.3401	6.3401	6.3401	$Pm\bar{3}m$
Pb content	$FA_{0.75}Cs_{0.25}Sn_{1-x}Pb_xI_3$ [S1,S4]				$FA_{0.83}Cs_{0.17}Sn_{1-x}Pb_xI_3$ [S2]			
	a (Å)	*b* (Å)	*c* (Å)	Phase	*a* (Å)	*b* (Å)	*c* (Å)	Phase
x = 0.0					6.278	6.278	6.278	$Pm\bar{3}m$
x = 0.5	6.449	8.925	8.903	Amm2	6.292	6.292	6.292	$Pm\bar{3}m$

Table S5. Experimental data of some prominent XRD peaks in pure-Sn and mixed Sn-Pb perovskites [S1-S4].

material	Phase	XRD peak (2θ) corresponding to (h,l,k) plane	Ref.
$FASnI_3$	$Pm\bar{3}m$	14° (100), 28° (200)	[S3]
$FA_{0.83}Cs_{0.17}SnI_3$	$Pm\bar{3}m$	14° (100), 28° (200)	[S2]
$FASn_{0.85}Pb_{0.25}I_3$	$Pm\bar{3}m$	14° (100), 28° (200)	[S3]
$FASn_{0.5}Pb_{0.5}I_3$	$Pm\bar{3}m$	14° (100), 28° (200)	[S3]
$FA_{0.83}Cs_{0.17}Sn_{0.5}Pb_{0.5}I_3$	$Pm\bar{3}m$	14.° (100), 28° (200)	[S2]
$FASn_{0.25}Pb_{0.75}I_3$	$Pm\bar{3}m$	14° (100), 28° (200)	[S3]
$FASnI_3$	Amm2		[S1]
$FASn_{0.875}Pb_{0.125}I_3$	Amm2		[S1]
$FASn_{0.85}Pb_{0.25}I_3$	Amm2	14.07° (100), 20.02° (110), 24.59° (111),	[S1]
$FASn_{0.5}Pb_{0.5}I_3$	Amm2	28.35° (200), 31.93° (210), 40.67° (220)	[S1]
$FA_{0.75}Cs_{0.25}Sn_{0.5}Pb_{0.5}I_3$	Amm2		[S1,S4]
$FASn_{0.375}Pb_{0.625}I_3$	Amm2		[S1]
$FASn_{0.25}Pb_{0.75}I_3$	Amm2		[S1]

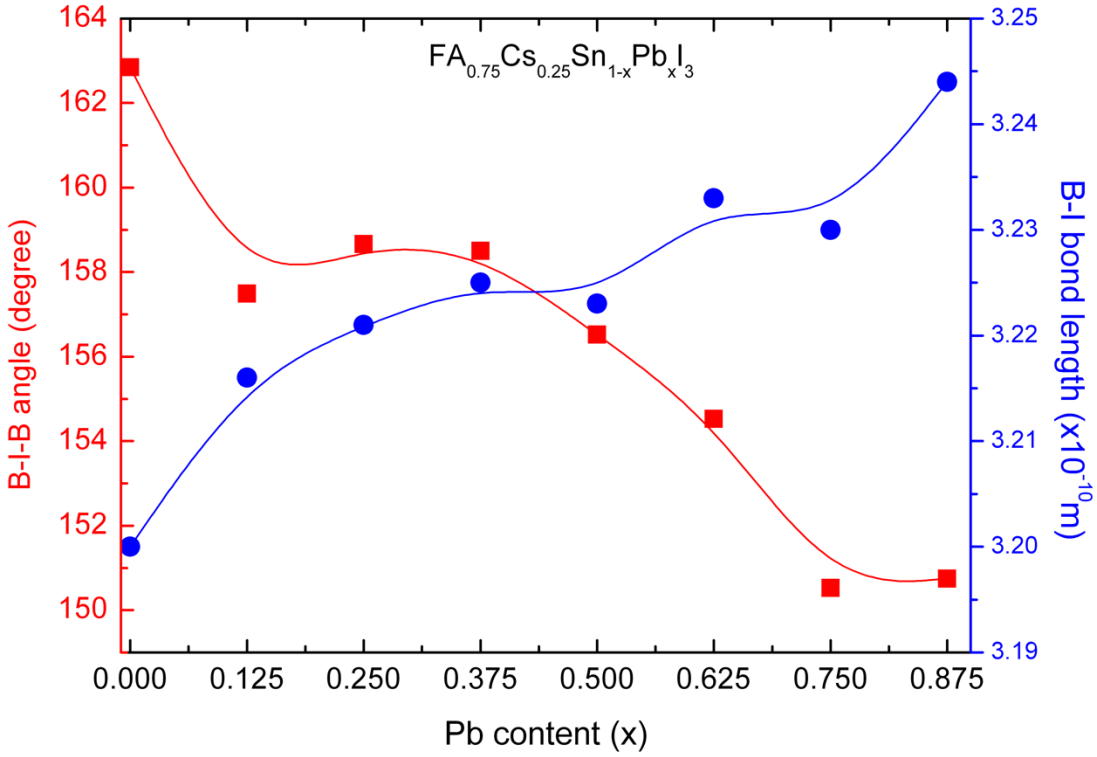

Figure S1. Average B-I-B tilting angles and average B-I bond length in the $FA_{0.75}Cs_{0.25}Sn_{1-x}Pb_xI_3$ mixed perovskites with a 2×2×2 supercell as a function of Pb content.

S2. Electronic band structure

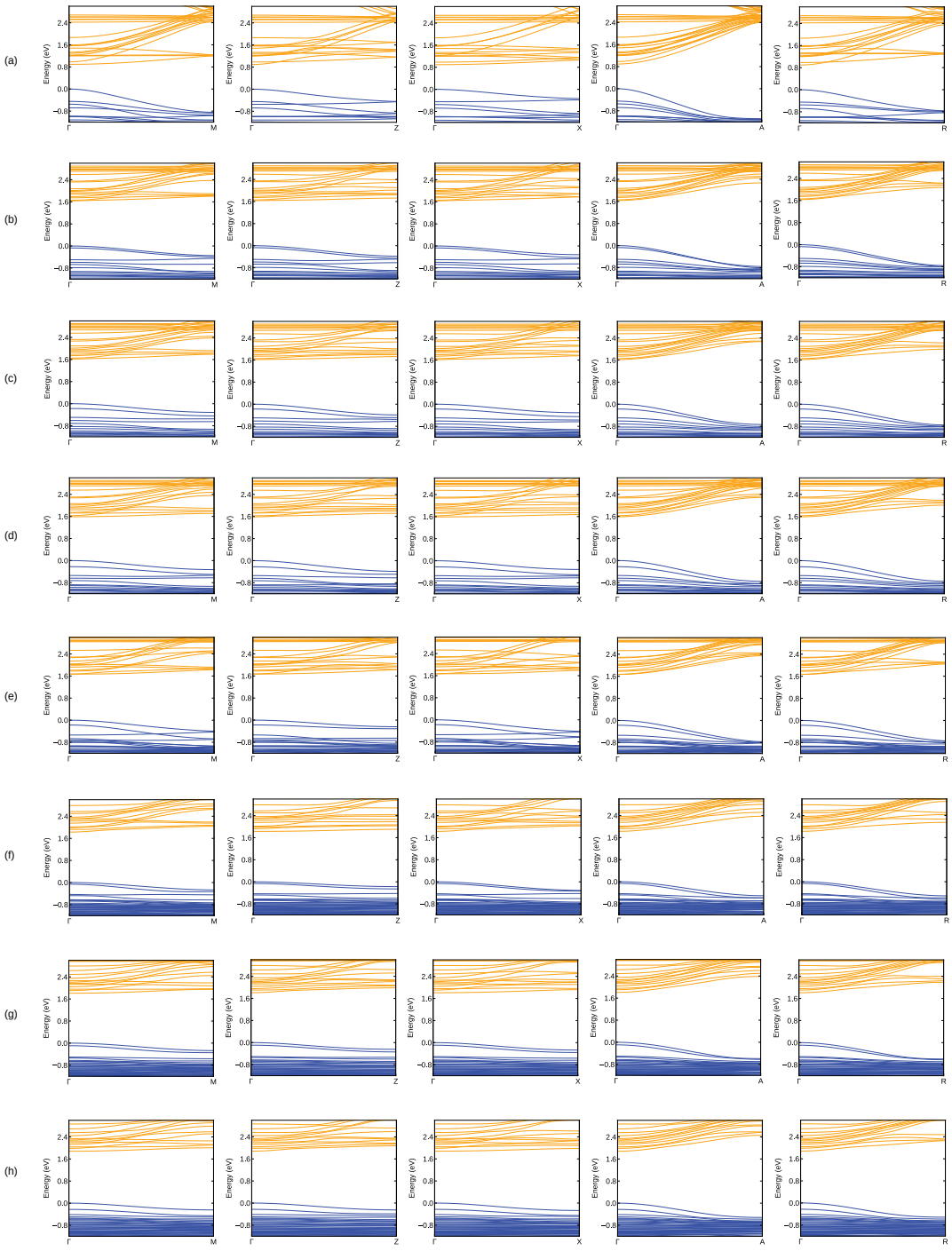

Figure S2. Electronic band structures along the high-symmetry point directions, Γ-M, Γ-Z, Γ-X, Γ-A, and Γ-R, for the mixed tin-lead iodide perovskites with (a) $x = 0.0$, (b) $x = 0.125$, (c) $x = 0.25$, (d) $x = 0.375$, (e) $x = 0.5$, (f) $x = 0.625$, (g) $x = 0.75$, and (h) $x = 0.875$. The VBM is set to zero.

S3. Carrier effective masses

Table S6. Calculated effective masses of electrons and holes along the Γ-high-symmetry point directions in $FA_{0.75}Cs_{0.25}Sn_{1-x}Pb_xI_3$ mixed perovskites, where m_0 is the free electron mass.

Pb content	(Γ-Z)		(Γ-X)		(Γ-M)		(Γ-R)		(Γ-A)	
	$m_e^*(m_0)$	$m_h^*(m_0)$	$m_e^*(m_0)$	$m_h^*(m_0)$	$m_e^*(m_0)$	$m_h^*(m_0)$	$m_e^*(m_0)$	$m_h^*(m_0)$	$m_e^*(m_0)$	$m_h^*(m_0)$
x = 0.0	0.076	0.123	0.980	0.199	1.044	0.139	0.141	0.153	0.204	0.135
x = 0.125	0.715	0.216	0.197	0.229	0.348	0.408	0.258	0.221	0.342	0.278
x = 0.25	0.473	0.200	0.233	0.233	0.397	0.421	0.258	0.213	0.345	0.281
x = 0.375	0.180	0.197	1.032	0.227	1.479	0.411	0.270	0.210	0.360	0.276
x = 0.5	0.644	0.305	0.843	0.182	1.197	0.362	0.344	0.231	0.454	0.303
x = 0.625	3.009	0.438	0.140	0.246	0.264	0.591	0.255	0.303	0.341	0.438
x = 0.75	0.164	0.264	1.379	0.282	1.703	0.585	0.296	0.279	0.363	0.412
x = 0.875	0.158	0.292	1.387	0.259	1.659	0.494	0.284	0.277	0.346	0.387

Table S7. Calculated effective masses along the Γ-R and Γ-A directions, average effective masses and reduced masses in $FA_{0.75}Cs_{0.25}Sn_{1-x}Pb_xI_3$ mixed perovskites.

Pb content	Effective mass (Γ-R)		Effective mass (Γ-A)		Average mass		Reduced mass
	$m_e^*(m_0)$	$m_h^*(m_0)$	$m_e^*(m_0)$	$m_h^*(m_0)$	$\overline{m}_e^*(m_0)$	$\overline{m}_h^*(m_0)$	$\mu_r(m_0)$
x = 0.0	0.141	0.153	0.204	0.135	0.172	0.144	0.078
x = 0.125	0.258	0.221	0.342	0.278	0.300	0.249	0.136
x = 0.25	0.258	0.213	0.345	0.281	0.302	0.247	0.136
x = 0.375	0.270	0.210	0.360	0.276	0.315	0.243	0.137
x = 0.5	0.344	0.231	0.454	0.303	0.399	0.267	0.160
x = 0.625	0.255	0.303	0.341	0.438	0.298	0.370	0.165
x = 0.75	0.296	0.279	0.363	0.412	0.329	0.345	0.168
x = 0.875	0.284	0.277	0.346	0.387	0.315	0.332	0.162

S4. Charge-carrier mobility

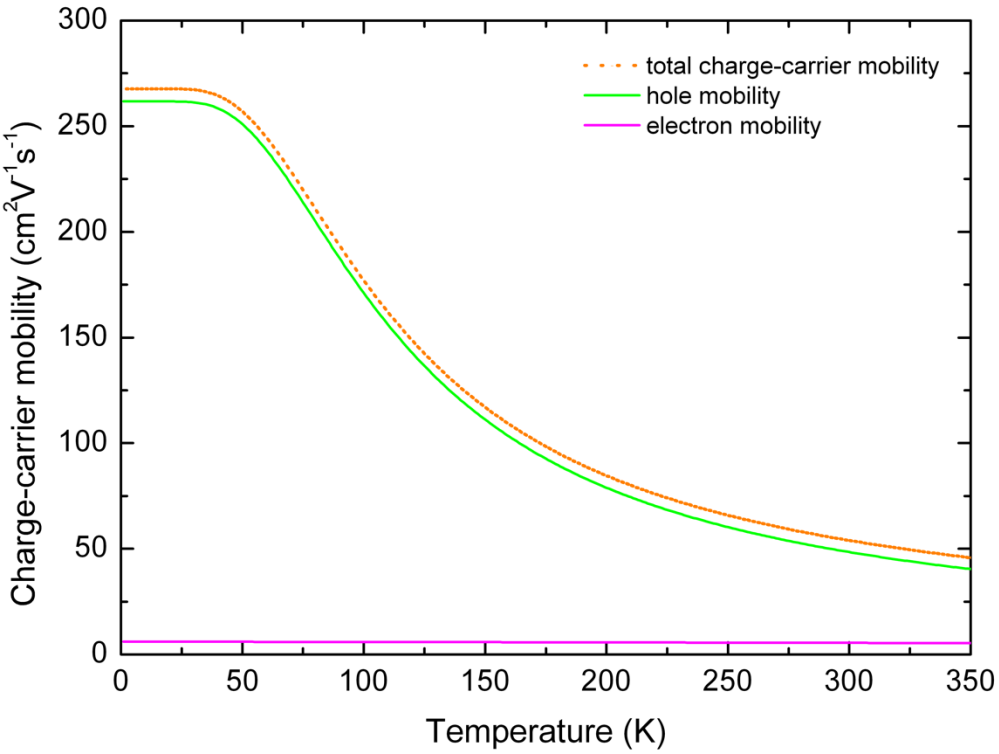

Figure S3. Plot of total charge-carrier mobility (dotted orange line), hole mobility (solid green line) and electron mobility (solid magenta line) as a function of temperature, calculated using $N_I^{(e)} = N_I^{(h)} = 1.0 \times 10^{19}$ cm^{-3}, as a function of temperature.

S5. Comparisons of the carrier effective masses using DFT calculations

Table S8. DFT calculations of electron and hole effective masses (m_e^* and m_h^*) with different functionals in pure-Sn, mixed Sn-Pb, and pure-Pb perovskites.

Material	**Functional**	$\boldsymbol{m_e^*}$ **(m_0)**	$\boldsymbol{m_h^*}$**(m_0)**	**Ref.**
$FA_{0.75}Cs_{0.25}SnI_3$	PBE	0.172	0.144	This work
$FASnI_3$	PBE	0.29	0.14	[S5]
$FASnI_3$	PBE	0.18	0.12	[S5]
$FASnI_3$	HSE06	0.28	0.11	[S6,S7]
$MASnI_3$	GW+SOC	0.36	0.16	[S8]
$FA_{0.75}Cs_{0.25}Sn_{0.75}Pb_{0.25}I_3$	PBE	0.302	0.247	This work
$FA_{0.875}Cs_{0.125}Sn_{0.75}Pb_{0.25}I_3$	PBE+SOC	0.1	0.08	[S9]
$MASn_{0.75}Pb_{0.25}I_3$	GW+SOC	0.23	0.14	[S8]
$FA_{0.75}Cs_{0.25}Sn_{0.5}Pb_{0.5}I_3$	PBE	0.399	0.267	This work
$FA_{0.75}Cs_{0.25}Sn_{0.5}Pb_{0.5}I_3$	PBE	0.33	0.296	[S10]
$FA_{0.875}Cs_{0.125}Sn_{0.5}Pb_{0.5}I_3$	PBE+SOC	0.1	0.11	[S9]
$MASn_{0.5}Pb_{0.5}I_3$	GW+SOC	0.32	0.21	[S8]
$FA_{0.75}Cs_{0.25}Sn_{0.25}Pb_{0.75}I_3$	PBE	0.329	0.345	This work
$FA_{0.875}Cs_{0.125}Sn_{0.25}Pb_{0.75}I_3$	PBE+SOC	0.13	0.14	[S9]
$MASn_{0.25}Pb_{0.75}I_3$	GW+SOC	0.37	0.26	[S8]
$FAPbI_3$	HSE06+SOC	0.184	0.213	[S11]
$FA_{0.875}Cs_{0.125}PbI_3$	PBE+SOC	0.13	0.19	[S9]
$FA_{0.83}Cs_{0.17}PbI_3$	GW+SOC	0.218	0.273	[S12]

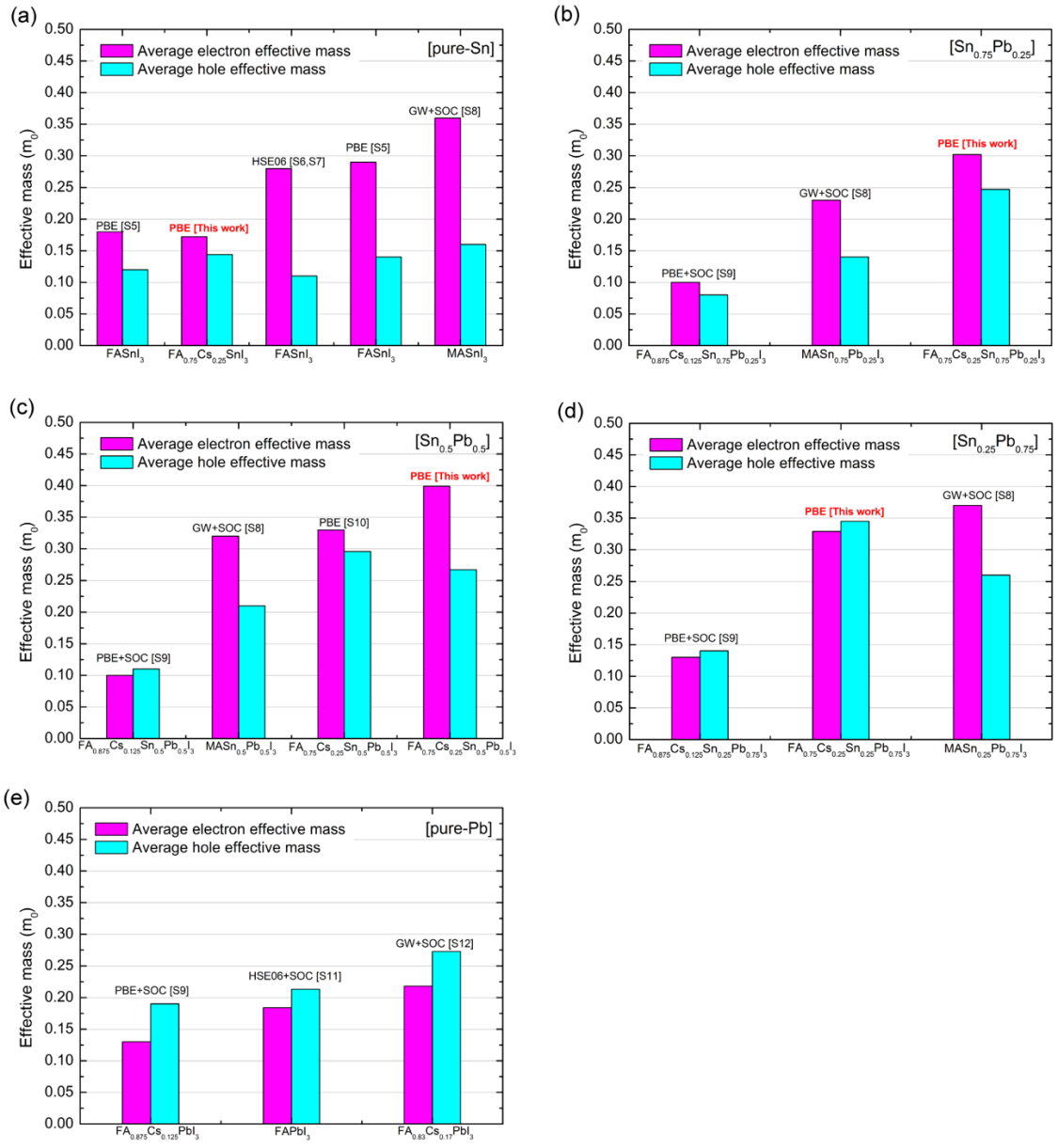

Figure S4. DFT calculations of electron (magenta bars) and hole (cyan bars) effective masses with different functionals in pure-Sn, mixed Sn-Pb, and pure-Pb perovskites, (a) pure-Sn, (b) $Sn_{0.75}Pb_{0.25}$, (c) $Sn_{0.5}Pb_{0.5}$, (d) $Sn_{0.25}Pb_{0.75}$, and (e) pure-Pb perovskites.

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