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Influence of Intrinsic Defects on Structure and Dynamics of Mixed Pb-Sn Perovskite: First-Principles DFT and NAMD Simulations

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Table S1: The main geometric parameters and DFEs of mix Pb-Sn perovskite $MASn_{0.03125}Pb_{0.96875}I_3$ with and without defects. Notice that the defect-induced geometric data are only available for the defect-surrounded structures.

Structures	pristine	V_{Sn}	V_{Pb}	I_i
d_{Sn-I}/Å	3.157	3.119	3.117	3.081
d_{Pb-I}/Å	3.202	3.206	3.212	3.204
min(∠M-X-M/)°	143.3	145.3	148.0	106.3
DFE/(eV/cell)	-	+0.0934	+0.0907	+0.0653

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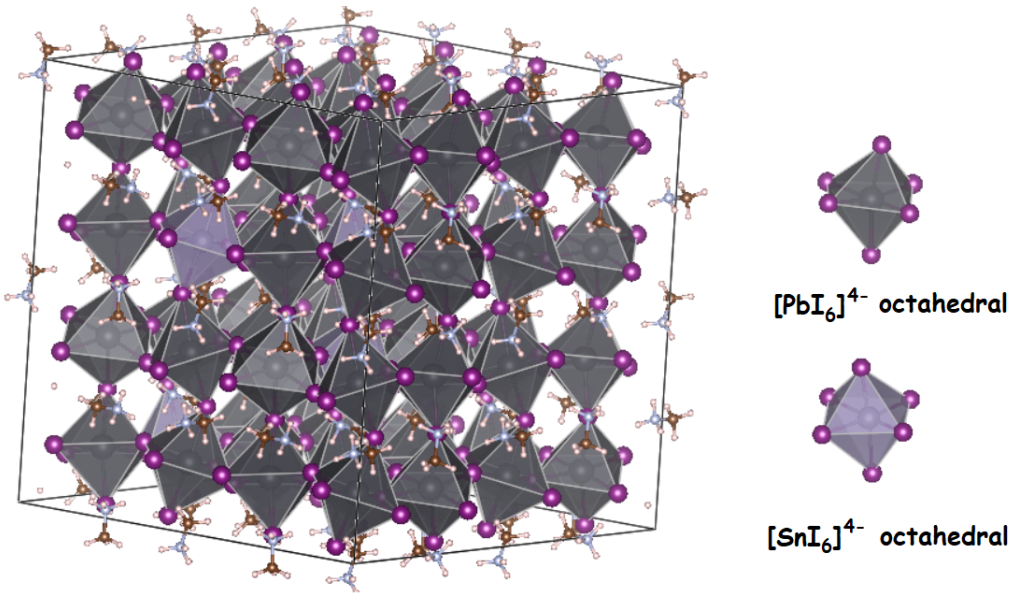

Figure S1: (a) Optimized alternative structures and (b) sketchmap of the $MASn_{0.03125}Pb_{0.96875}I_3$ perovskite without defects.

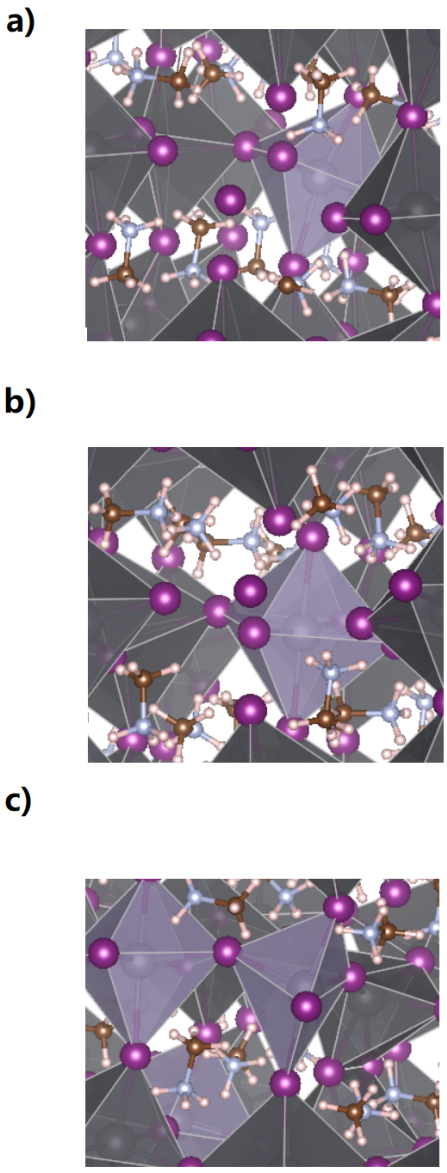

Figure S2: Optimized defect-bearing (a) V_{Pb} (b) V_{Sn} and (c) i_I structure in $MASn_{0.03125}Pb_{0.96875}I_3$ lattice.

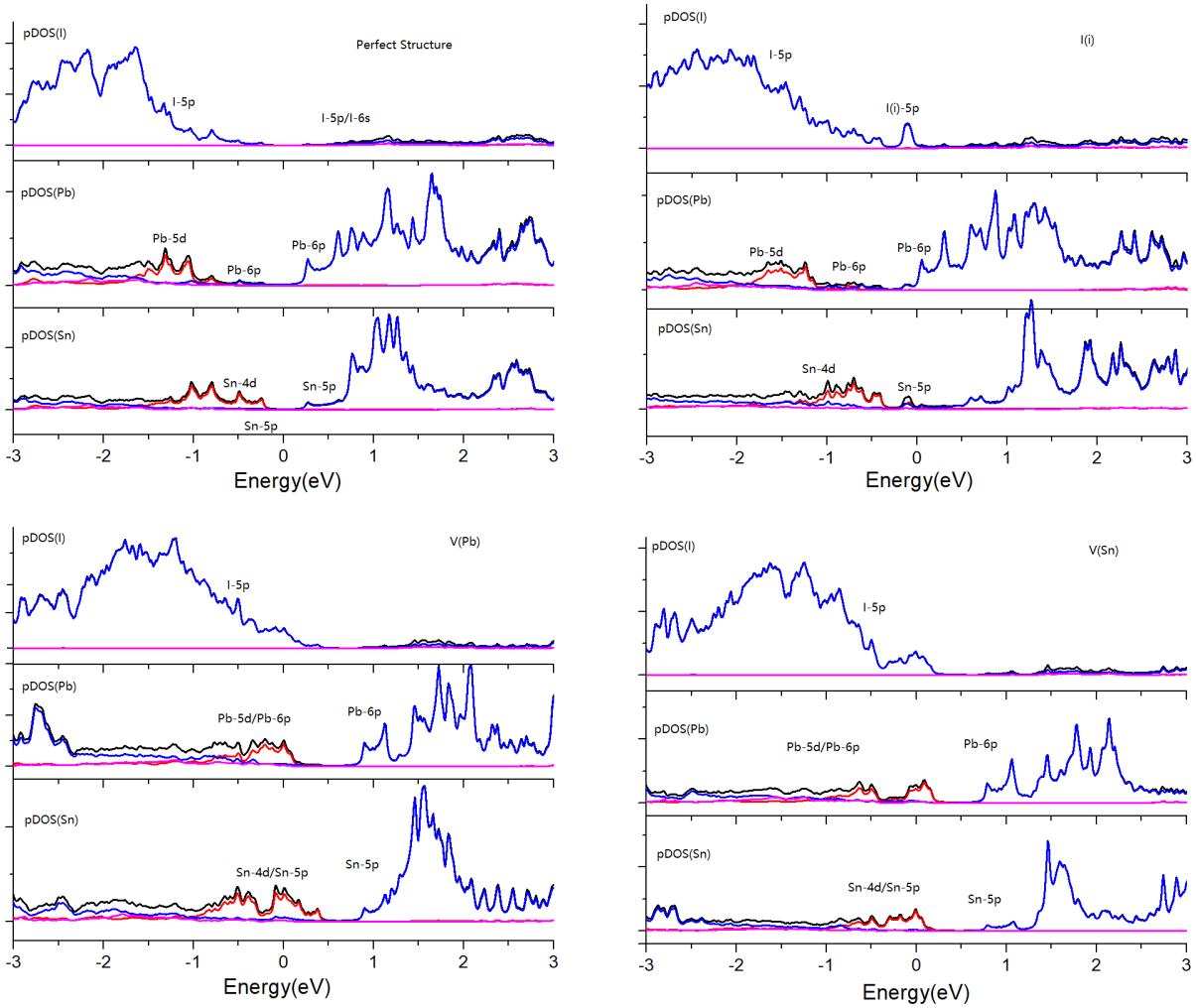

Figure S3: Projected density-of-states of the pristine and defect-bearing MA_2SnPbI_6 perovskites calculated with PBE+SOC.

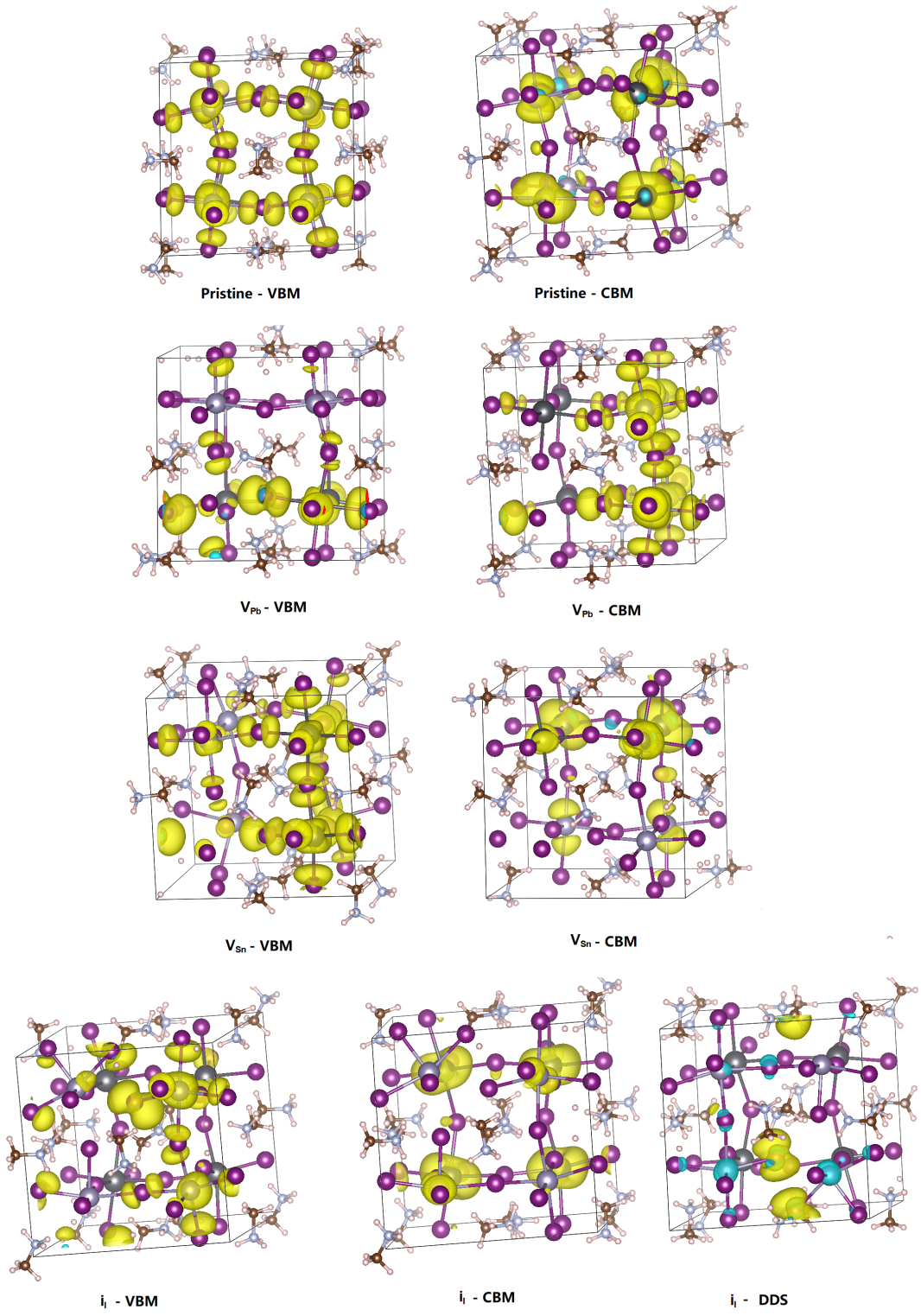

Figure S4: Charge distributions of the key states in the pristine and defect-bearing MA_2SnPbI_6 perovskites calculated with PBE+SOC.

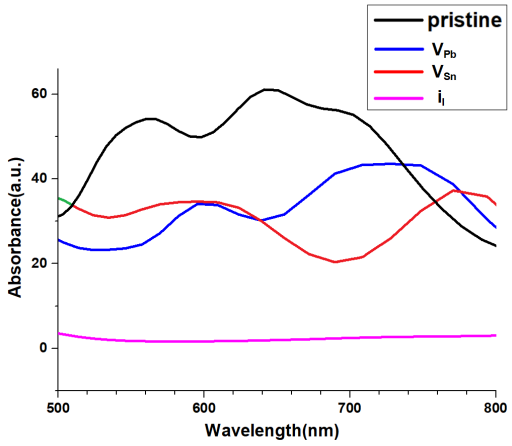

Figure S5: Calculated absorption spectra of the pristine and defect-bearing MA_2SnPbI_6 perovskite in visible region.

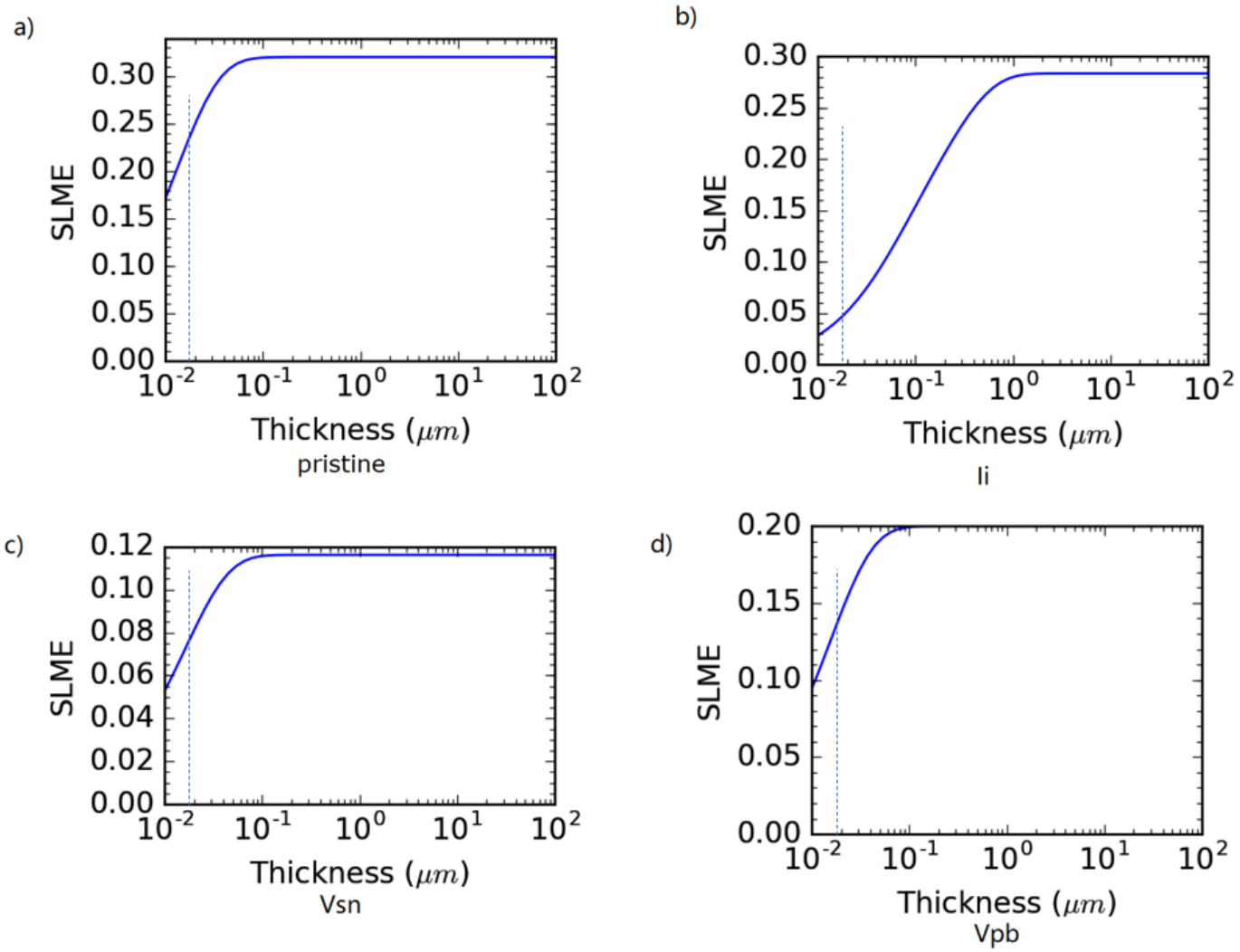

Figure S6: Calculated spectral-limited maximum efficiency of the pristine and defect-bearing MA_2SnPbI_6 perovskites.

pristine	vbm-5	vbm-4	vbm-3	vbm-2	vbm-1	vbm	cbm	cbm+1	cbm+2	cbm+3	cbm+4
vbm-5	0	35.902	9.5771	4.3208	2.6885	1.5663	0.5137	0.4358	0.5154	0.5139	0.5077
vbm-4	35.902	0	31.368	6.4004	3.4574	2.0092	0.4321	0.4635	0.549	0.4646	0.4961
vbm-3	9.5771	31.368	0	28.02	5.2666	2.5546	0.4521	0.5757	0.5448	0.5885	0.5441
vbm-2	4.3208	6.4004	28.02	0	21.784	4.49	0.5519	0.6068	0.6815	0.6022	0.5825
vbm-1	2.6885	3.4574	5.2666	21.784	0	18.986	0.5143	0.5831	0.7014	0.7086	0.7109
vbm	1.5663	2.0092	2.5546	4.49	18.986	0	0.5422	0.6074	0.8435	0.7191	0.617
cbm	0.5137	0.4321	0.4521	0.5519	0.5143	0.5422	0	22.831	6.945	3.7015	2.8816
cbm+1	0.4358	0.4635	0.5757	0.6068	0.5831	0.6074	22.831	0	31.344	7.4924	4.617
cbm+2	0.5154	0.549	0.5448	0.6815	0.7014	0.8435	6.945	31.344	0	34.46	8.1881
cbm+3	0.5139	0.4646	0.5885	0.6022	0.7086	0.7191	3.7015	7.4924	34.46	0	33.848
cbm+4	0.5077	0.4961	0.5441	0.5825	0.7109	0.617	2.8816	4.617	8.1881	33.848	0

V(Pb)	vbm-4	vbm-3	vbm-2	vbm-1	vbm	cbm	cbm+1	cbm+2	cbm+3	cbm+4	cbm+5
vbm-4	0	37.255	5.9932	3.1349	1.0337	0.5232	0.6704	0.6561	0.6027	0.6305	0.5243
vbm-3	37.255	0	22.812	5.511	1.1743	0.6111	0.5605	0.6437	0.5341	0.5197	0.6219
vbm-2	5.9932	22.812	0	30.113	1.6419	0.5599	0.5427	0.6466	0.7703	0.672	0.6671
vbm-1	3.1349	5.511	30.113	0	2.1693	0.4834	0.6544	0.5994	0.5372	0.5711	0.5767
vbm	1.0337	1.1743	1.6419	2.1693	0	0.6533	0.6126	0.7148	0.6241	0.5961	0.5936
cbm	0.5232	0.6111	0.5599	0.4834	0.6533	0	15.981	4.7635	2.0771	1.6992	1.2438
cbm+1	0.6704	0.5605	0.5427	0.6544	0.6126	15.981	0	22.62	5.6193	4.0108	2.362
cbm+2	0.6561	0.6437	0.6466	0.5994	0.7148	4.7635	22.62	0	29.322	8.147	4.9664
cbm+3	0.6027	0.5341	0.7703	0.5372	0.6241	2.0771	5.6193	29.322	0	34.812	7.9962
cbm+4	0.6305	0.5197	0.672	0.5711	0.5961	1.6992	4.0108	8.147	34.812	0	27.47
cbm+5	0.5243	0.6219	0.6671	0.5767	0.5936	1.2438	2.362	4.9664	7.9962	27.47	0

I(I)	vbm-7	vbm-6	vbm-5	vbm-4	vbm-3	vbm-2	vbm-1	vbm	tr	cbm	cbm+1
vbm-7	0	59.408	15.248	7.2117	4.1297	2.8308	1.8272	1.5081	0.8275	0.328	0.3008
vbm-6	59.408	0	49.234	11.847	5.9334	3.2885	1.9193	1.7469	0.8102	0.3249	0.3264
vbm-5	15.248	49.234	0	40.367	10.236	4.0499	2.2821	1.5671	1.0219	0.3498	0.3499
vbm-4	7.2117	11.847	40.367	0	24.25	5.8946	2.8497	2.0329	1.0304	0.364	0.3619
vbm-3	4.1297	5.9334	10.236	24.25	0	20.666	3.4127	2.6745	1.1727	0.3269	0.3718
vbm-2	2.8308	3.2885	4.0499	5.8946	20.666	0	8.4177	4.2195	1.3354	0.3954	0.3806
vbm-1	1.8272	1.9193	2.2821	2.8497	3.4127	8.4177	0	29.992	1.9649	0.4422	0.439
vbm	1.5081	1.7469	1.5671	2.0329	2.6745	4.2195	29.992	0	2.2713	0.5183	0.5432
tr	0.8275	0.8102	1.0219	1.0304	1.1727	1.3354	1.9649	2.2713	0	0.7631	0.7622
cbm	0.328	0.3249	0.3498	0.364	0.3269	0.3954	0.4422	0.5183	0.7631	0	22.468
cbm+1	0.3008	0.3264	0.3499	0.3619	0.3718	0.3806	0.439	0.5432	0.7622	22.468	0

V(Sn)	vbm-5	vbm-4	vbm-3	vbm-2	vbm-1	vbm	cbm	cbm+1	cbm+2	cbm+3	cbm+4
vbm-5	0	31.913	8.1234	2.8581	1.1803	0.7704	0.3522	0.4049	0.4311	0.3571	0.3967
vbm-4	31.913	0	26.892	4.4115	1.2939	0.9466	0.3896	0.448	0.4888	0.4314	0.4466
vbm-3	8.1234	26.892	0	11.578	1.3627	1.2927	0.5447	0.6203	0.5651	0.4769	0.4971
vbm-2	2.8581	4.4115	11.578	0	3.4393	2.0326	0.5164	0.4742	0.5403	0.4697	0.5961
vbm-1	1.1803	1.2939	1.3627	3.4393	0	7.6303	0.7924	0.7367	0.7342	0.6335	0.6403
vbm	0.7704	0.9466	1.2927	2.0326	7.6303	0	1.2078	1.0741	0.8978	0.6664	0.7397
cbm	0.3522	0.3896	0.5447	0.5164	0.7924	1.2078	0	16.986	3.2859	2.2332	1.3488
cbm+1	0.4049	0.448	0.6203	0.4742	0.7367	1.0741	16.986	0	15.194	2.9743	1.8412
cbm+2	0.4311	0.4888	0.5651	0.5403	0.7342	0.8978	3.2859	15.194	0	7.5185	2.9784
cbm+3	0.3571	0.4314	0.4769	0.4697	0.6335	0.6664	2.2332	2.9743	7.5185	0	15.929
cbm+4	0.3967	0.4466	0.4971	0.5961	0.6403	0.7397	1.3488	1.8412	2.9784	15.929	0

Figure S7: Calculated non-adiabatic coupling matrices of the pristine and defect-bearing MA_2SnPbI_6 perovskites.

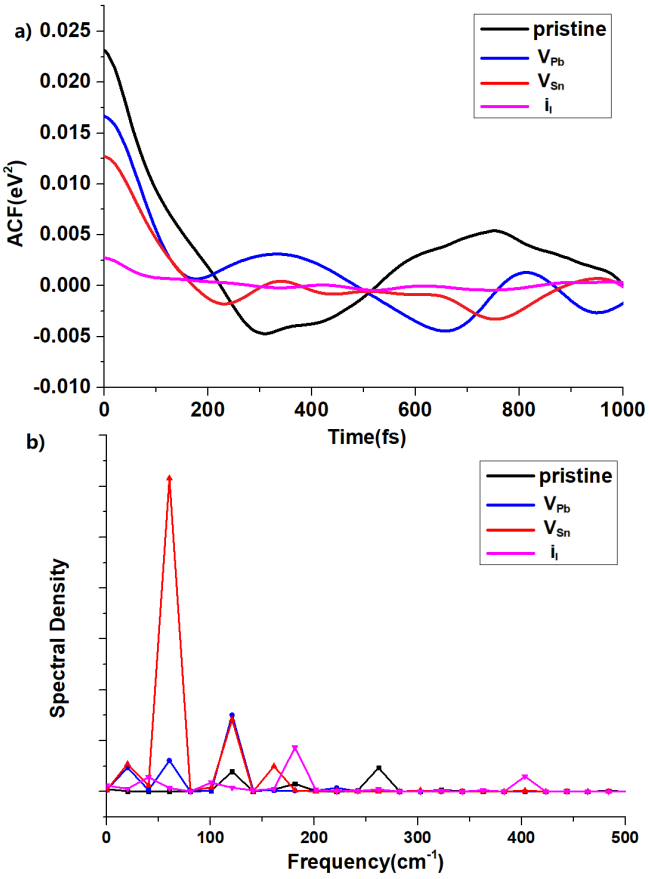

Figure S8: Calculated (a) unnormalized auto-correlation functions and (b) spectral densities of the pristine and defect-bearing MA_2SnPbI_6perovskites.