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Supporting Information

Electronic and optical properties of mixed Sn / Pb organohalide perovskites: A first principles investigation

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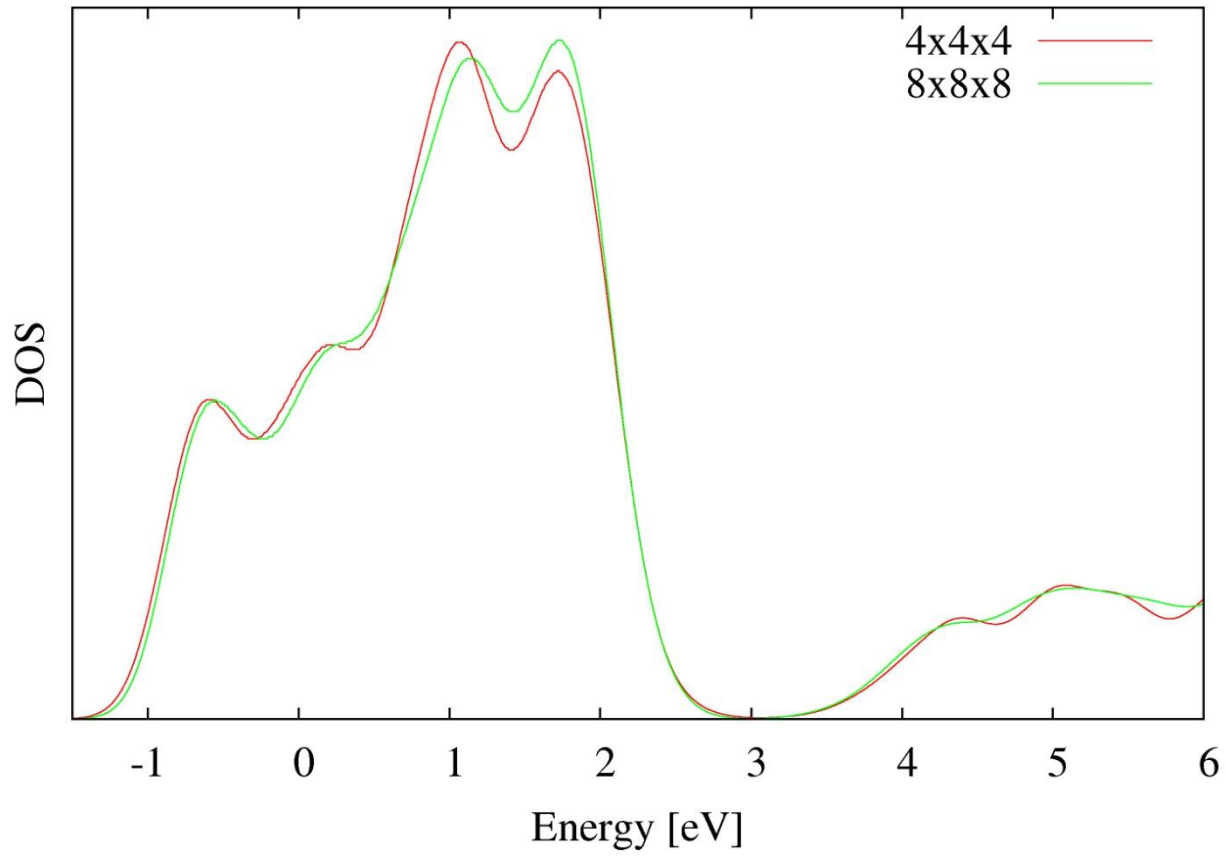


Figure S1. SR-DFT DOS using 4x4x4 and 8x8x8 k-point grid of MAPbI3 I4cm perovskite.

Table S1. HOMO and LUMO energy levels shifts (eV) by varying the Sn/Pb ratio calculated at the SOC-GW level and compared to the experimental measurements by Ogomi et al. VB and CB shifts evaluated from the SOC-GW DOS are also reported. Energy shifts are referred to the pure $MAPbI_3$ perovskites (set as zero). VB and CB values are calculated at 1.4 % of the maximum VB peak of the $MAPbI_3$ calculated SOC-GW DOS.

	Sn content				
	0.00	0.25	0.5	0.75	1.00
Exp.					
ΔE^{HOMO}	0.00	0.27	0.44	0.47	0.66
ΔE^{LUMO}	0.00	0.07	0.21	0.19	0.25
Gap^{EXP}	1.51	1.31	1.28	1.23	1.10
SOC-GW					
ΔE^{HOMO}	0.00	0.34	0.46	0.76	0.99
ΔE^{LUMO}	0.00	0.08	0.23	0.32	0.35

Gap^{H-L}	1.64	1.37	1.41	1.20	1.00
ΔE^{VB}	0.00	0.20	0.35	0.53	0.64
ΔE^{CB}	0.00	0.08	0.17	0.27	0.19
Gap^{VB-CB}	1.74	1.62	1.56	1.48	1.29
SOC-DFT					
ΔE^{HOMO}	0.00	0.20	0.28	0.50	0.87
ΔE^{LUMO}	0.00	0.06	0.18	0.30	0.50
Gap^{H-L}	0.53	0.39	0.43	0.36	0.16
ΔE^{VB}	0.00	0.12	0.19	0.30	0.47
ΔE^{CB}	0.00	0.04	0.10	0.18	0.29
Gap^{VB-CB}	0.76	0.68	0.67	0.64	0.56
SOC-DFT VC-RELAX					
ΔE^{HOMO}	0.00	0.27	0.38	0.61	0.82
ΔE^{LUMO}	0.00	0.06	0.22	0.30	0.50
Gap^{H-L}	0.53	0.34	0.39	0.23	0.21
ΔE^{VB}	0.00	0.17	0.27	0.38	0.52
ΔE^{CB}	0.00	0.06	0.17	0.20	0.31
Gap^{VB-CB}	0.76	0.65	0.65	0.58	0.55
SR-DFT VC-RELAX					
ΔE^{HOMO}	0.00	0.21	0.33	0.58	0.81
ΔE^{LUMO}	0.00	-0.12	-0.08	-0.19	-0.19
Gap^{H-L}	1.43	1.11	1.03	0.66	0.43
ΔE^{VB}	0.00	0.13	0.25	0.35	0.51
ΔE^{CB}	0.00	-0.03	-0.03	-0.10	-0.12
Gap^{VB-CB}	1.41	1.26	1.14	0.96	0.78

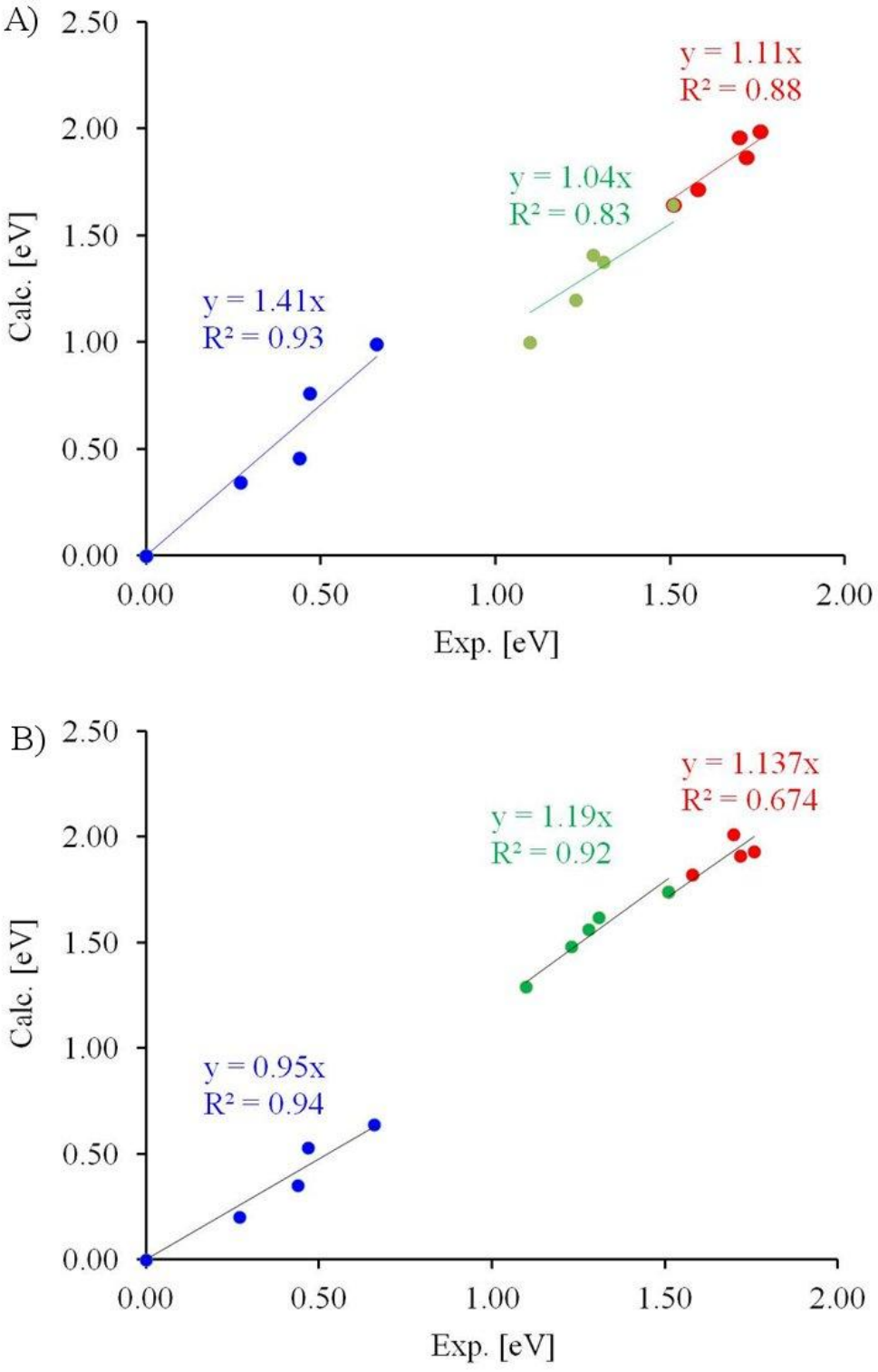


Figure S2. Linear fit of the experimental VB/CB against calculated HOMO/LUMO and CB/VB values, see text for definitions.

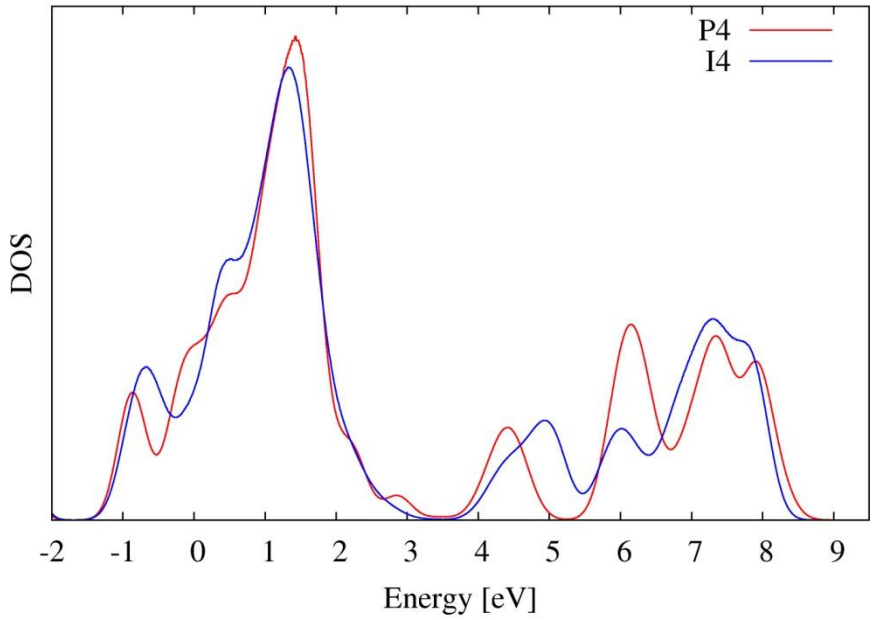


Figure S3. SR-DFT DOS of the I4cm and P4mm phase for the $MASnI_3$ perovskite.

Table S2. Effective masses (m_h, m_e, μ) and Rashba coefficients (A_h, A_e)calculated for electrons and holes by SOC-DFT.

	Structure from Ref. 17					Structure 1				
Sn/Pb	0/4					0/4				
	m_h	m_e	μ	A_h	A_e	m_h	m_e	μ	A_h	A_e
M	0.28	0.20	0.12	3.18	7.61	0.18	0.16	0.08	1.93	3.87
Z	0.24	0.17	0.10	1.66	3.37	0.23	0.16	0.09	0.15	0.68
X	0.21	0.17	0.09	2.34	5.10	0.17	0.15	0.08	2.09	3.87
A	0.25	0.18	0.11	3.69	7.81	0.17	0.15	0.08	1.78	3.81
R	0.19	0.15	0.08	3.32	6.29	0.17	0.14	0.07	1.75	3.50
AVG	0.23	0.17	0.10	2.84	6.04	0.18	0.15	0.08	1.54	3.15
	Structure 1					Structure 1				
	1/3					2/2				
	m_h	m_e	μ	A_h	A_e	m_h	m_e	μ	A_h	A_e
M	0.16	0.15	0.08	1.87	3.93	0.15	0.16	0.08	2.00	4.05
Z	0.20	0.15	0.09	0.72	1.67	0.17	0.16	0.08	0.12	0.49
X	0.15	0.15	0.07	2.09	3.94	0.14	0.15	0.07	2.21	4.00
A	0.15	0.14	0.07	0.98	3.51	0.14	0.14	0.07	1.72	3.81
R	0.15	0.13	0.07	1.11	2.45	0.13	0.13	0.07	1.75	3.50
AVG	0.16	0.14	0.08	1.35	3.10	0.15	0.15	0.07	1.56	3.17
	Structure 1					Structure 1				
	3/1					4/0				
	m_h	m_e	μ	A_h	A_e	m_h	m_e	μ	A_h	A_e
M	0.13	0.17	0.07	2.49	3.99	0.09	0.19	0.06	3.13	5.90
Z	0.15	0.16	0.08	1.44	2.61	0.11	0.15	0.06	0.22	0.67

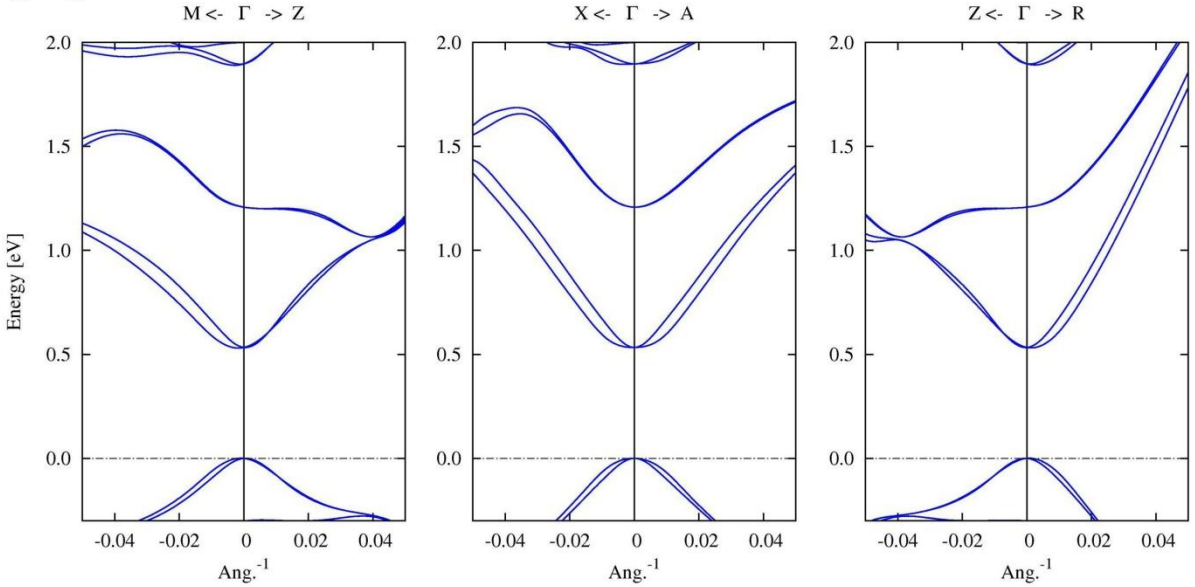
X	0.12	0.16	0.07	2.03	4.12	0.09	0.16	0.06	3.32	6.33
A	0.12	0.14	0.06	1.84	2.03	0.08	0.13	0.05	2.61	5.39
R	0.11	0.13	0.06	0.12	1.52	0.08	0.11	0.05	2.47	5.45
AVG	0.13	0.15	0.07	1.58	2.85	0.09	0.15	0.06	2.35	4.75
	Structure from Ref. 16									
	4/0									
	m_h	m_e	μ	A_h	A_e					
M	0.11	0.19	0.07	3.44	5.35					
Z	0.11	0.16	0.07	1.68	2.87					
X	0.09	0.16	0.06	2.03	4.43					
A	0.10	0.14	0.06	3.52	6.00					
R	0.08	0.12	0.05	2.83	6.12					
AVG	0.10	0.15	0.06	2.70	4.95					

Table S2. Effective masses (m_h, m_e and μ) calculated by SOC-GW.

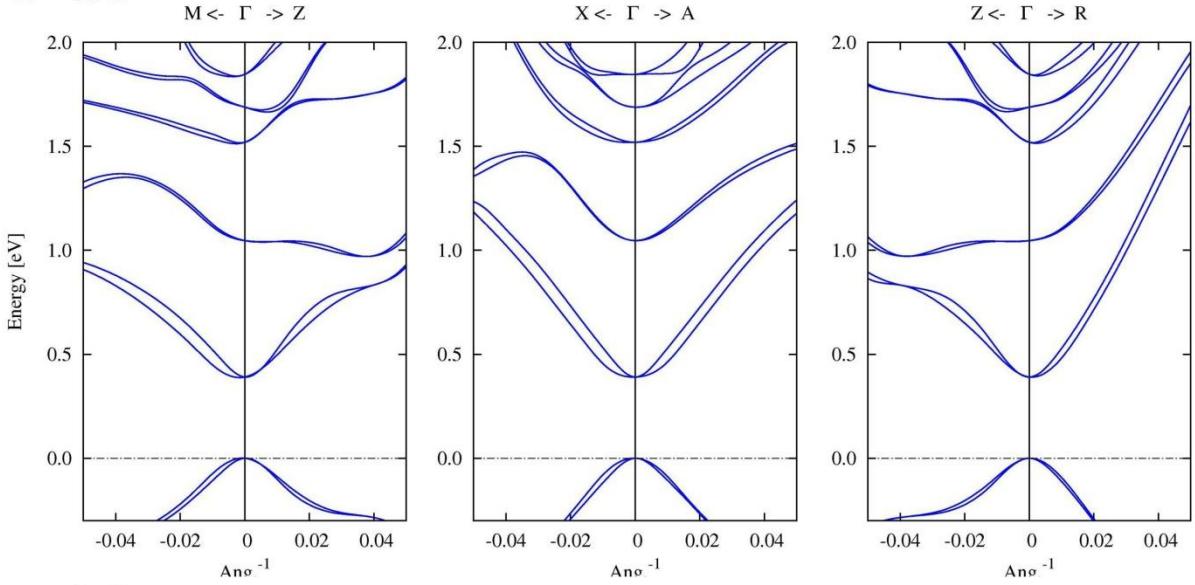
GW	Structure from Ref. 17			Structure 1		
Sn/Pb	0/4			0/4		
	m_h	m_e	μ	m_h	m_e	μ
M	0.26	0.21	0.12	0.17	0.17	0.09
Z	0.21	0.19	0.10	0.21	0.17	0.09
X	0.19	0.18	0.09	0.16	0.16	0.08
A	0.23	0.20	0.11	0.16	0.16	0.08
R	0.17	0.17	0.08	0.15	0.15	0.07
AVG	0.21	0.19	0.10	0.17	0.16	0.08
	Structure 1			Structure 1		
Pb/Sn	1/3			2/2		
	m_h	m_e	μ	m_h	m_e	μ
M	0.12	0.16	0.07	0.14	0.17	0.07
Z	0.15	0.16	0.08	0.15	0.16	0.08
X	0.12	0.15	0.07	0.13	0.16	0.07
A	0.12	0.14	0.06	0.13	0.15	0.07
R	0.11	0.13	0.06	0.12	0.13	0.06
AVG	0.13	0.15	0.07	0.13	0.15	0.07
	Structure 1			Structure 1		
Pb/Sn	3/1			4/0		
	m_h	m_e	μ	m_h	m_e	μ
M	0.11	0.16	0.06	0.08	0.22	0.06
Z	0.12	0.15	0.07	0.09	0.17	0.06
X	0.10	0.14	0.06	0.07	0.19	0.05
A	0.10	0.13	0.06	0.07	0.16	0.05
R	0.09	0.12	0.05	0.06	0.13	0.04
AVG	0.10	0.14	0.06	0.07	0.17	0.05

Figure S4. Calculated SOC-GW band structures for the investigated series.

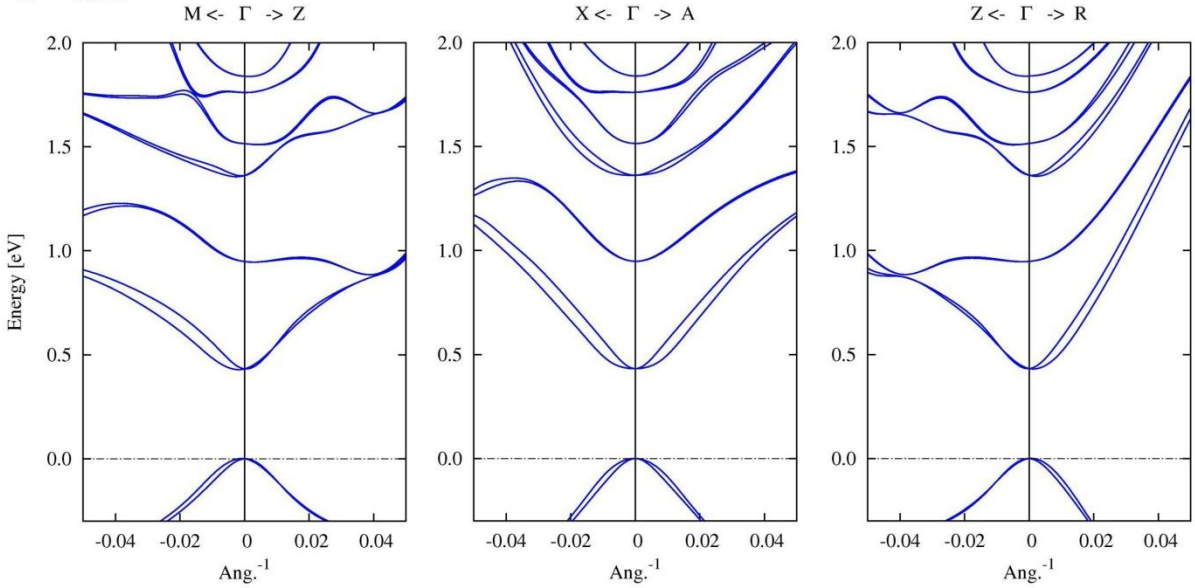
x = 0


M <- Γ -> Z
X <- Γ -> A
Z <- Γ -> R
Energy [eV]
2.0
1.5
1.0
0.5
0.0
-0.04
-0.02
0
0.02
0.04
Ang.-1

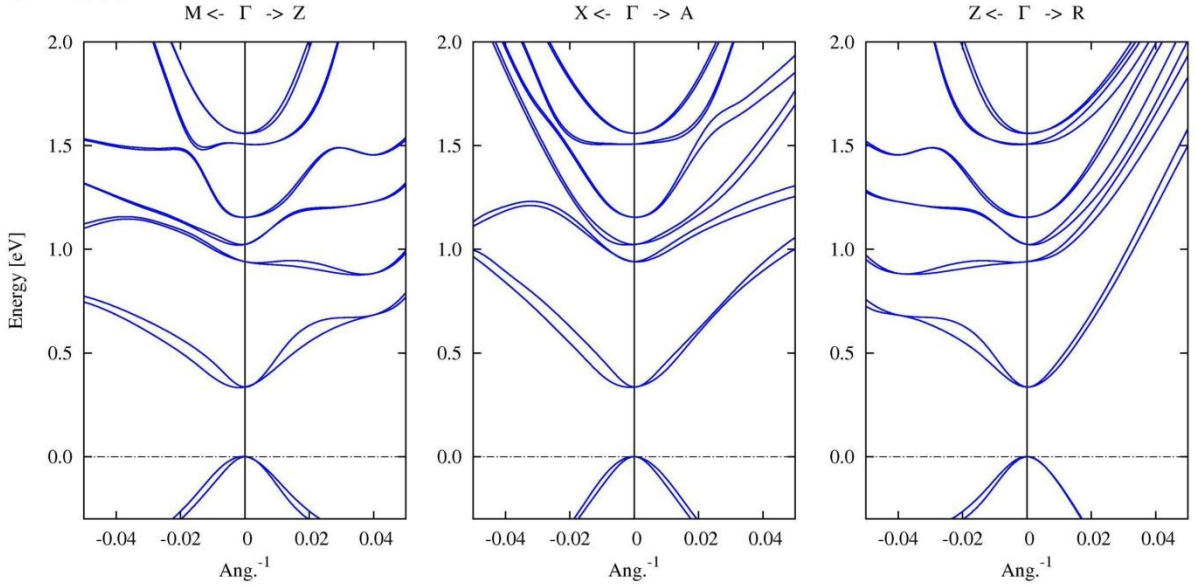
x = 0.25


M <- Γ -> Z
X <- Γ -> A
Z <- Γ -> R
Energy [eV]
2.0
1.5
1.0
0.5
0.0
-0.04
-0.02
0
0.02
0.04
Ang.-1

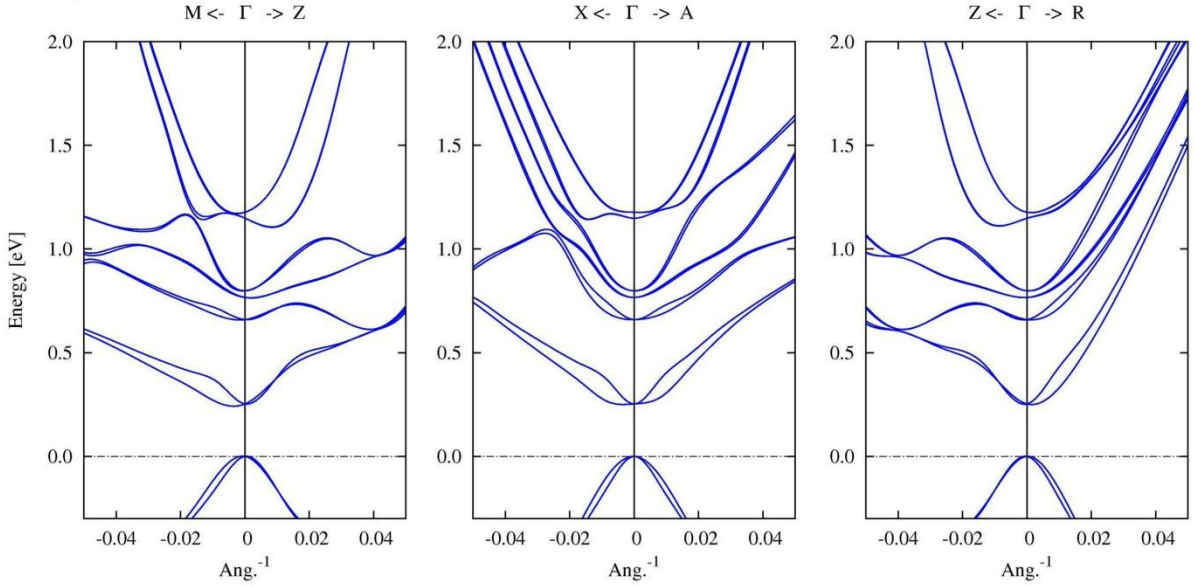
x = 0.50


M <- Γ -> Z
X <- Γ -> A
Z <- Γ -> R
Energy [eV]
2.0
1.5
1.0
0.5
0.0
-0.04
-0.02
0
0.02
0.04
Ang.-1

x = 0.75


M <- Γ -> Z
X <- Γ -> A
Z <- Γ -> R
Energy [eV]
2.0
1.5
1.0
0.5
0.0
-0.04
-0.02
0
0.02
0.04
Ang.-1

x = 1


M <- Γ -> Z
X <- Γ -> A
Z <- Γ -> R
Energy [eV]
2.0
1.5
1.0
0.5
0.0
-0.04
-0.02
0
0.02
0.04
Ang.-1