Supplementary Information for: Frustrated van der Waals heterostructures

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S1 Phonon band structures for monolayers

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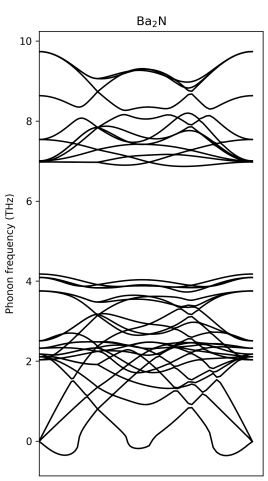	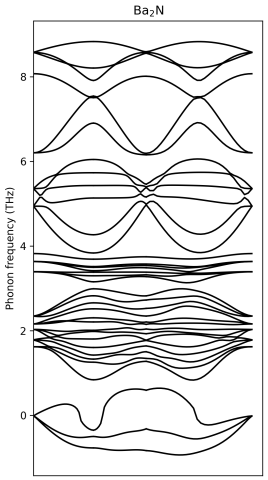	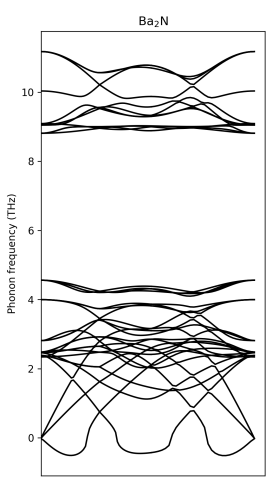
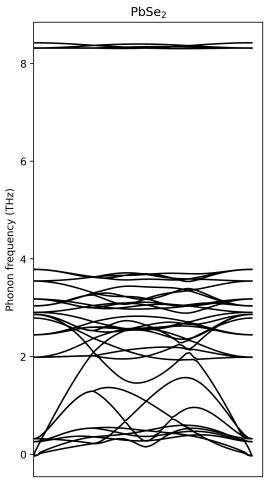	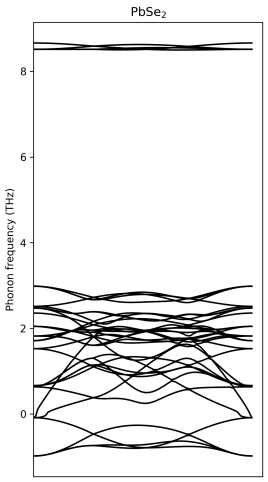	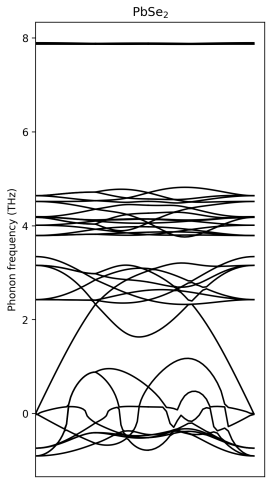

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TABLE S1: The phonon band structures for monolayers examined in this work.

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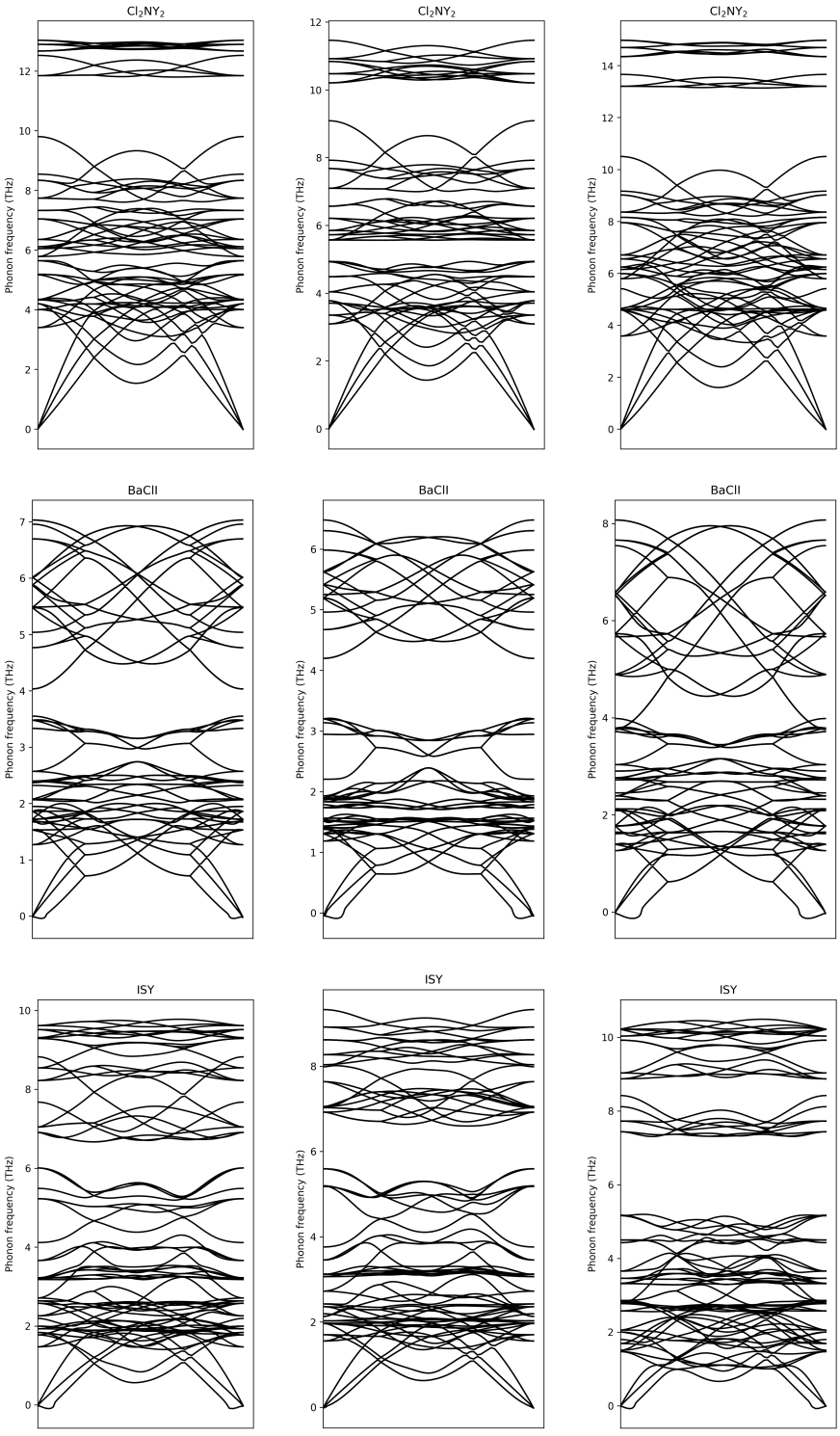
0 strain	**+5% strain**	**−5% strain**



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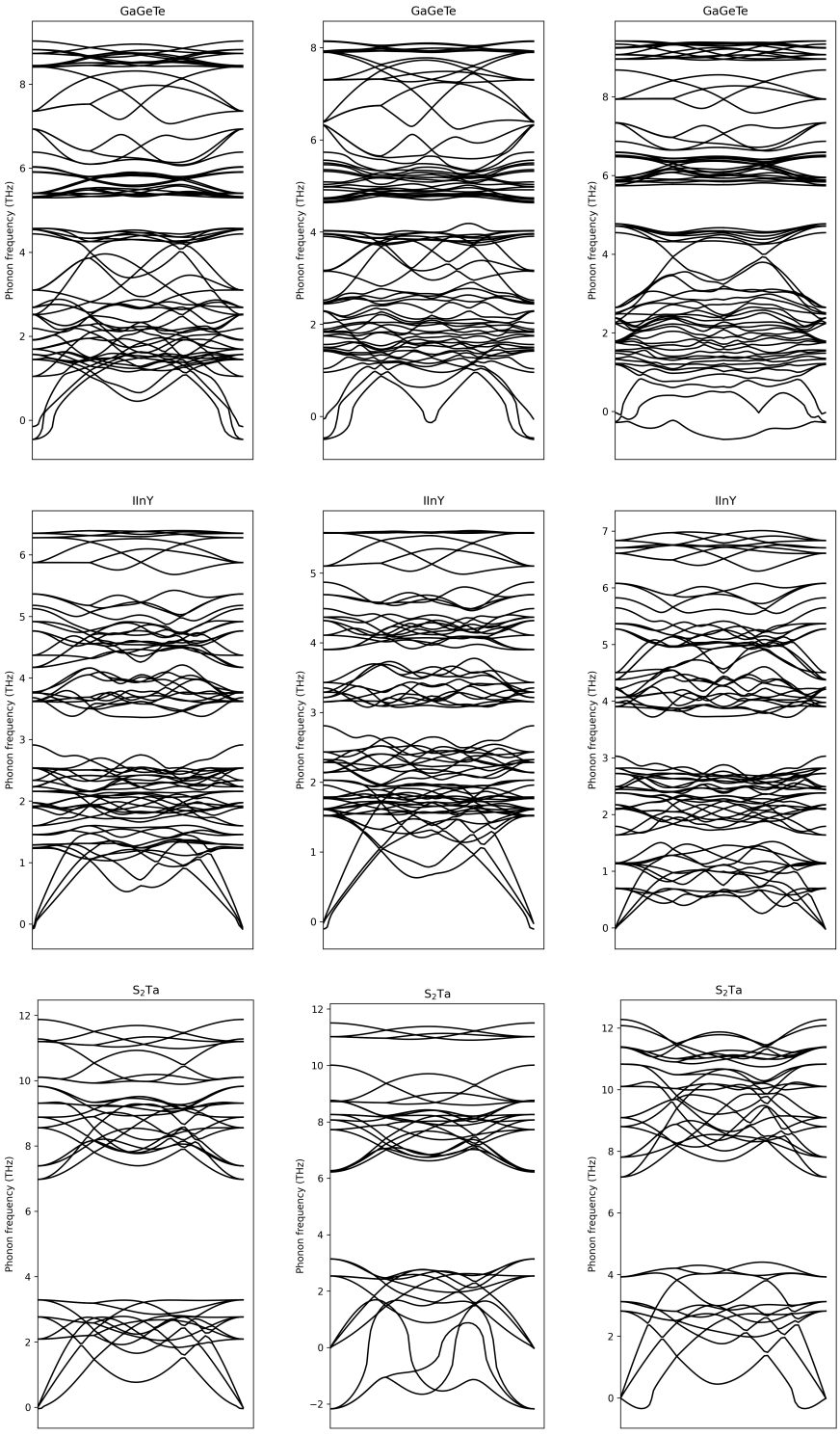

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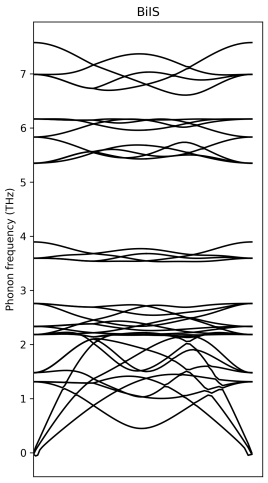	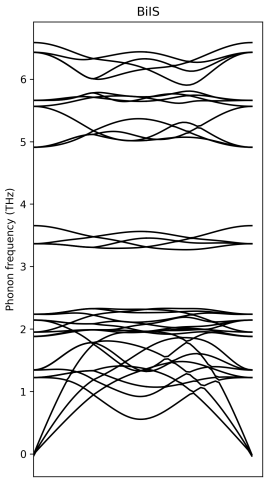	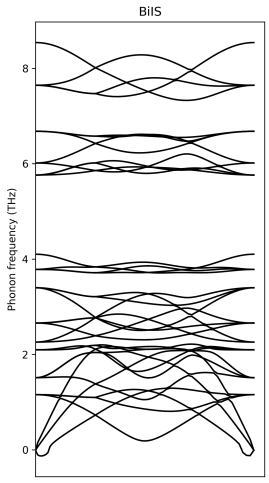
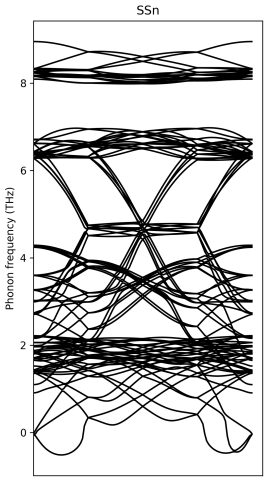	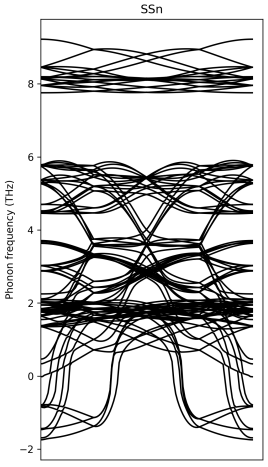	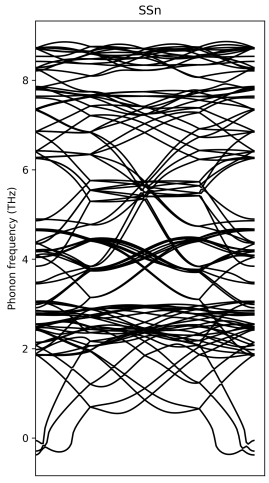
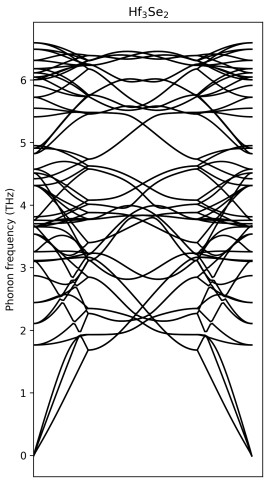	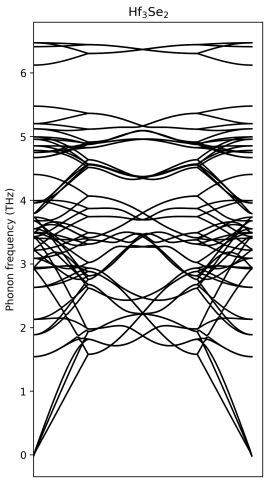	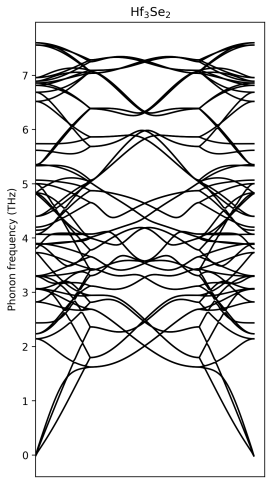

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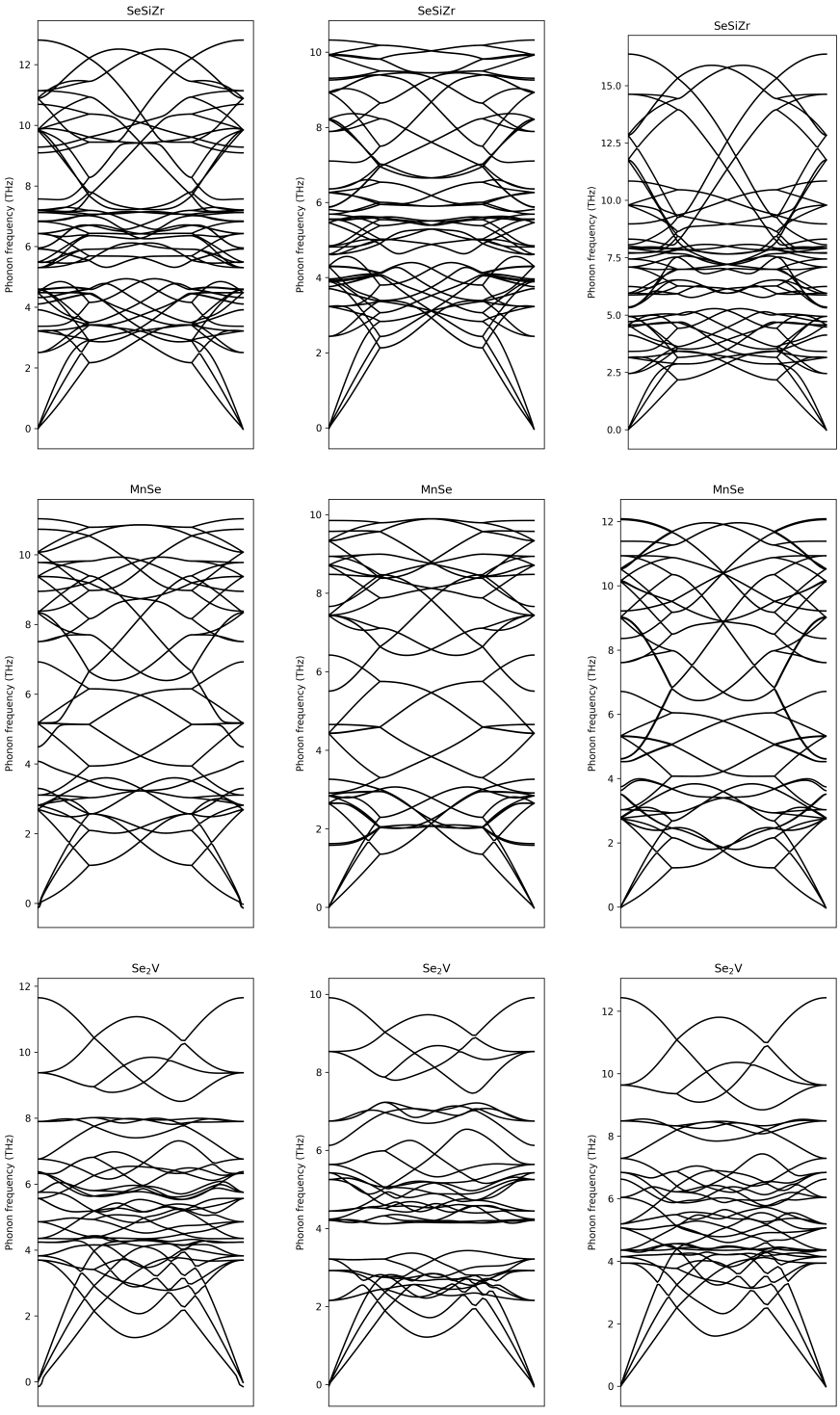

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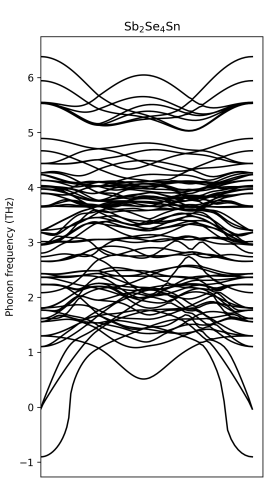	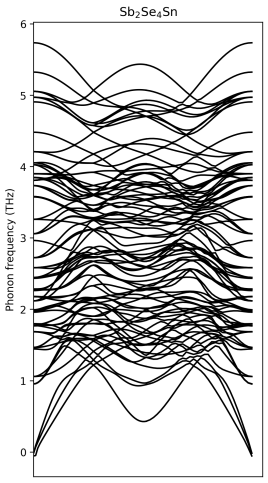	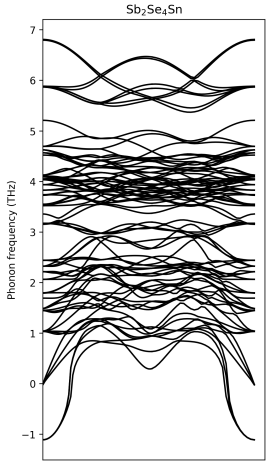
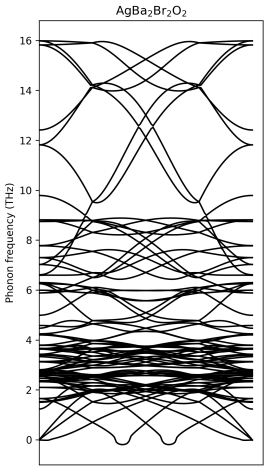	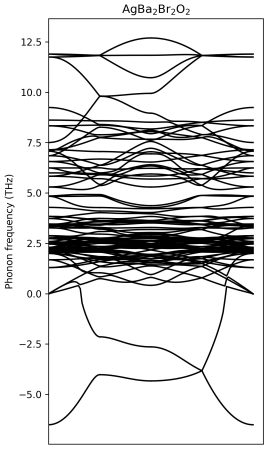	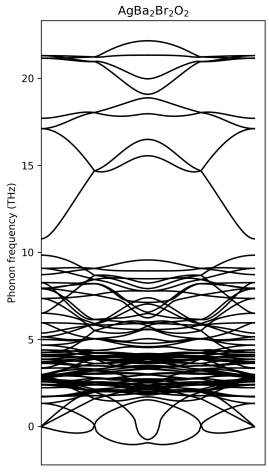
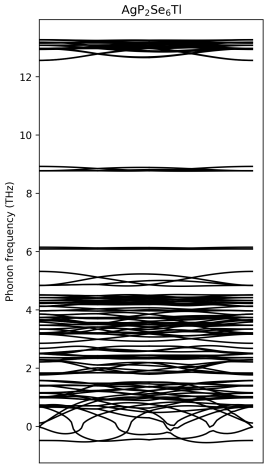	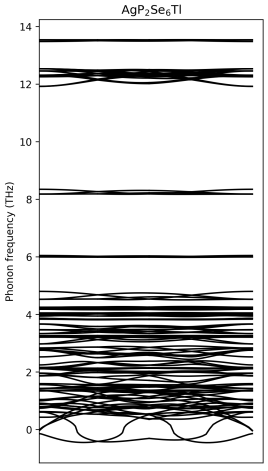	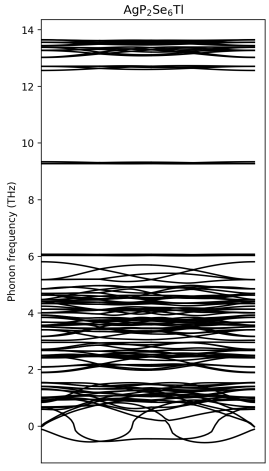

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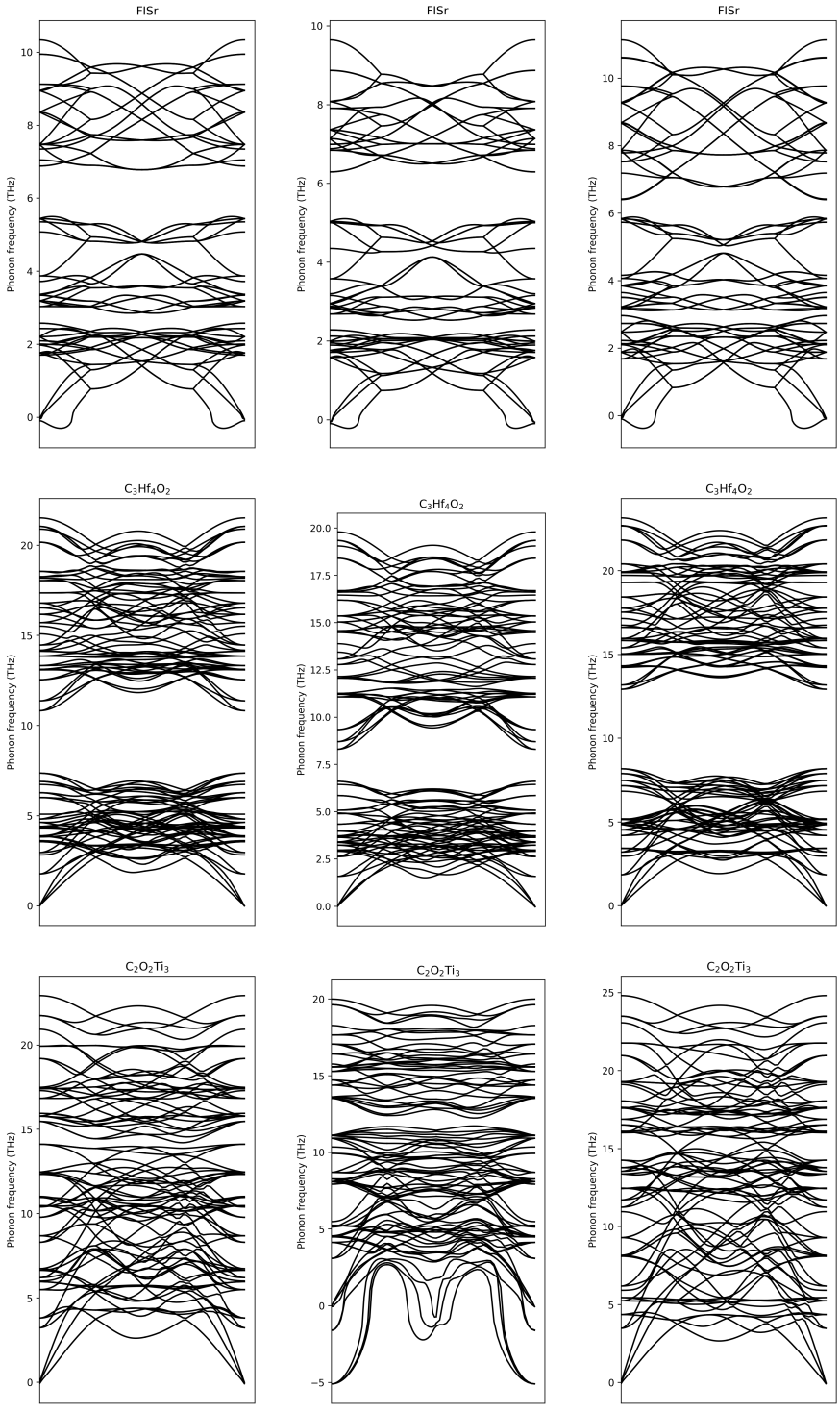

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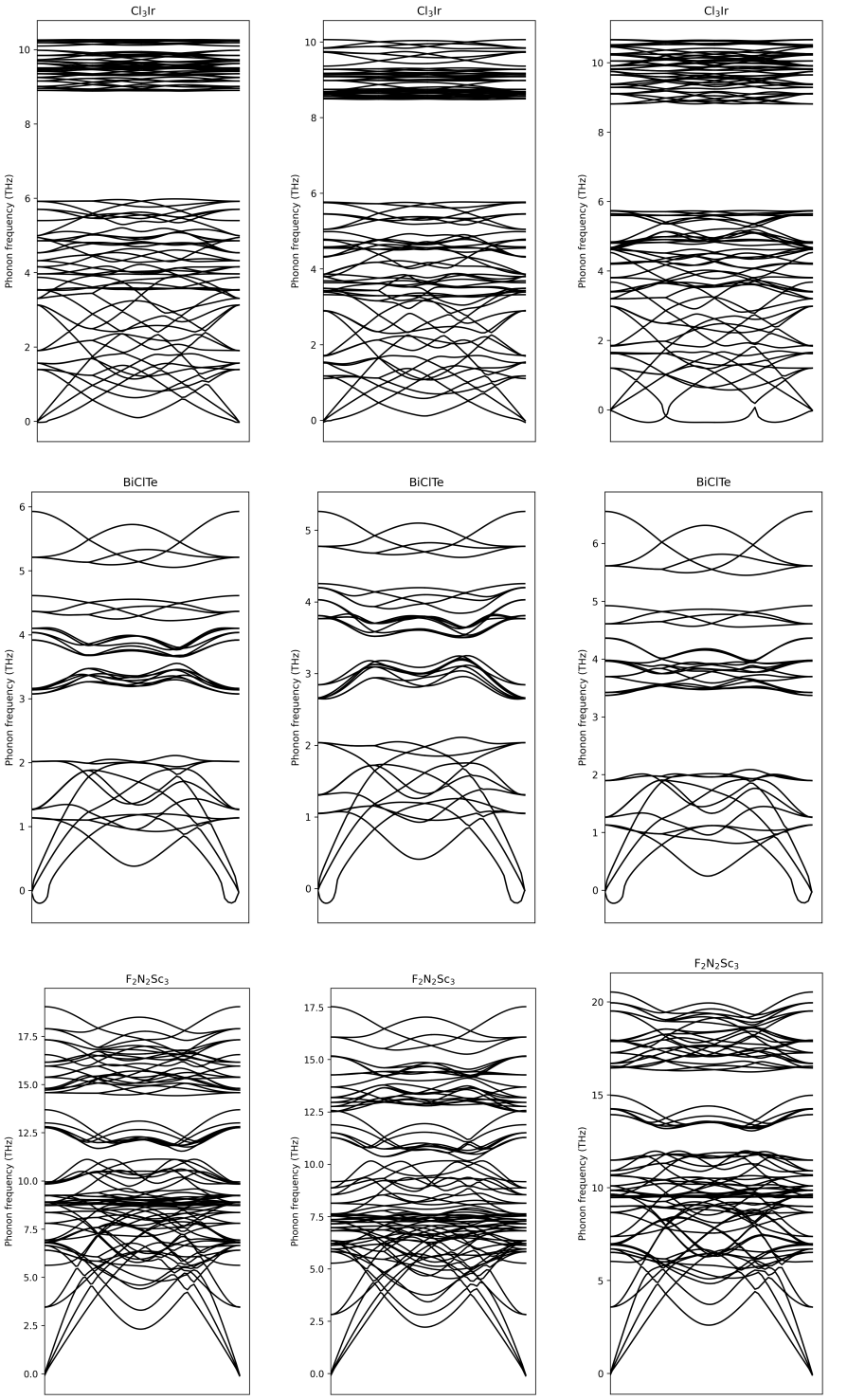

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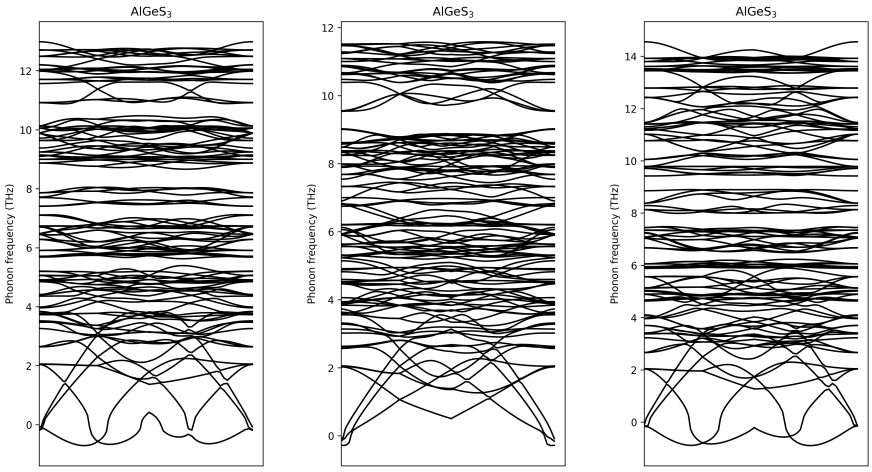


S2 AIMD simulation of the bilayers

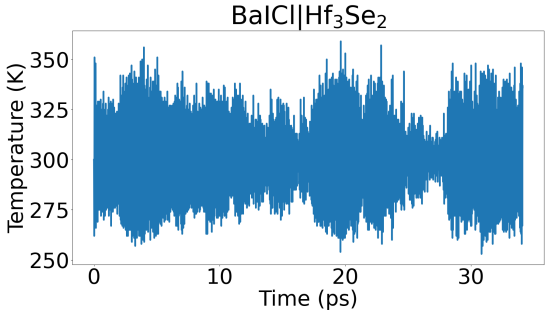
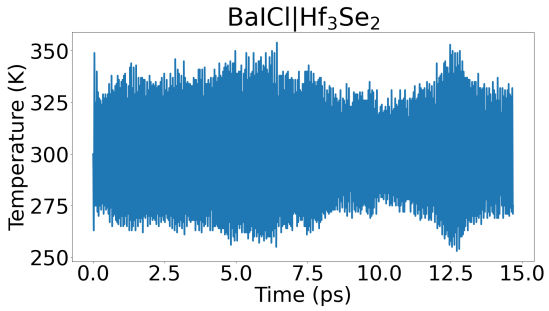
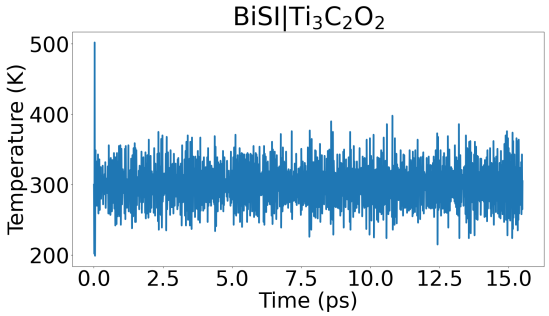
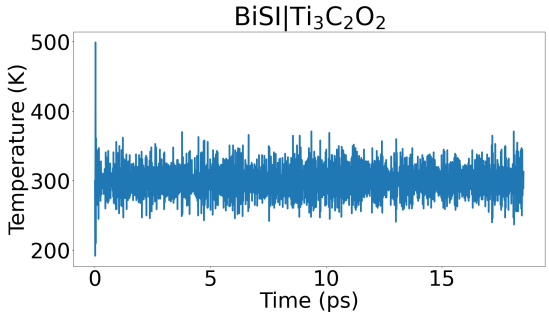
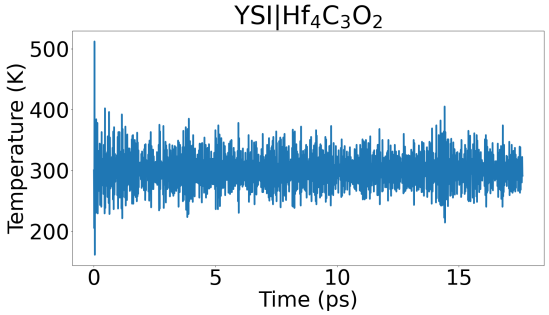
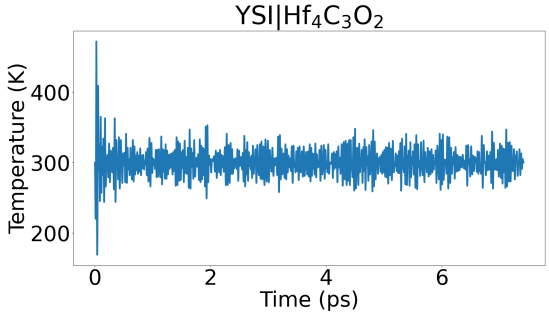
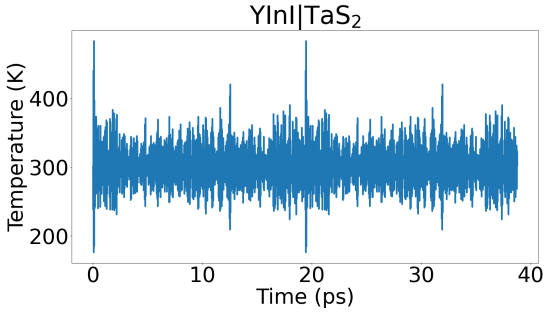
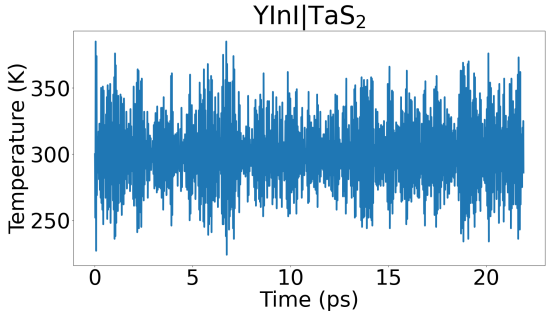
Frustrated	Non-frustrated
	
	
	
	

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TABLE S2: The evolution of temperature as a function of time.

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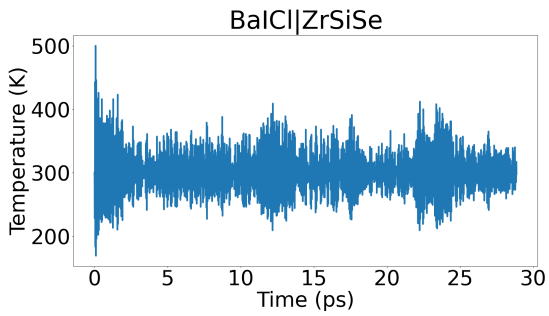	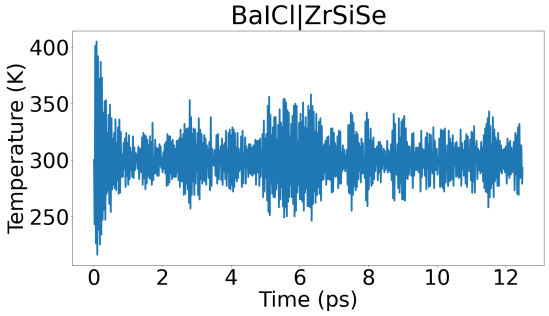
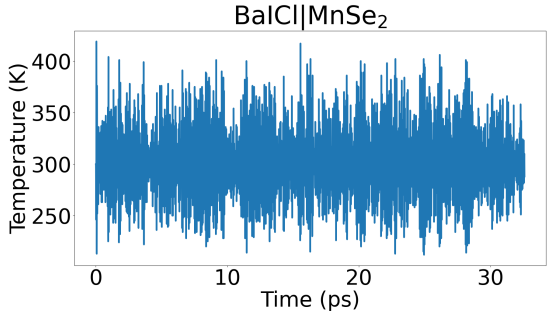	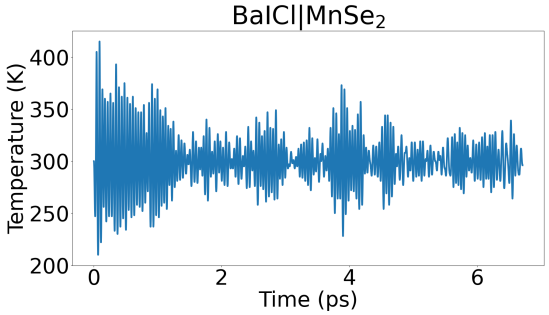
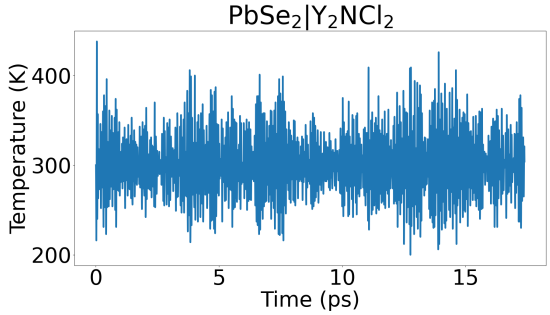	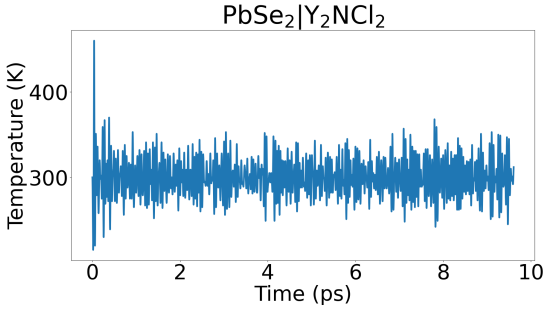
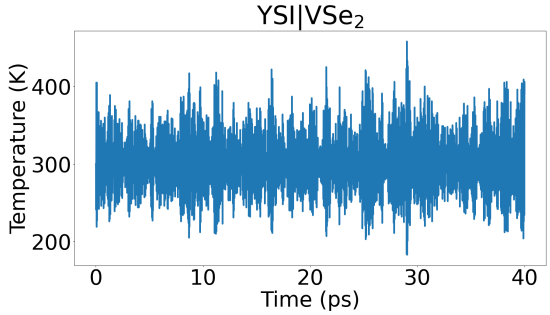	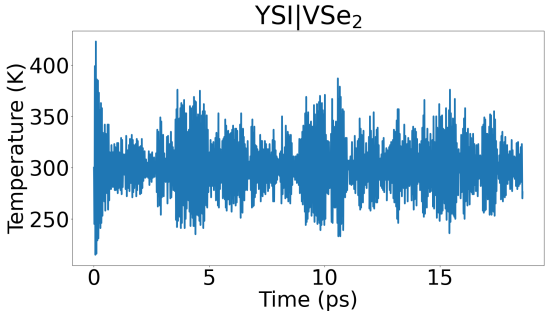

S3 Bandgap tuning

Frustration can have a significant impact on the electronic structure of the material. It leads to bandgap narrowing in GaGeTe | $AlGeS_3$ and $SnSb_2Se_4$ | $IrCl_3$, and bandgap widening in SnS | BiTeCl and SnS | SrIF. We display the density of states of the four bilayers in Figure S1. Importantly, the change in the bandgap of GaGeTe | $AlGeS_3$ would not require the sustenance of strain, a technically challenging and an energy-costly process, such as in the cases reviewed by Qi et al.[1] The presence of strain in both layers yields a thermodynamically stable combination that can then be incorporated in devices. Of the two layers in GaGeTe | $AlGeS_3$, GaGeTe can be cleaved from the bulk structure with ICSD ID `35386`, and has a C2DB ID `2GaGeTe-1`. This semiconductor, with HSE06-computed nearly direct bandgap of 1.165 eV (C2DB), a p-type material, has previously attracted attention for the tunability of its bandgap as well as its high carrier mobility, making it a candidate material for photovoltain applications.[2] $AlGeS_3$, with C2DB ID `2AlGeS3-1`, is a hypothetical structure that has not yet been explored in the literature. Its HSE06-computed indirect bandgap is 2.909 eV (C2DB) and is also a p-type material. The frustration in this bilayer was generated by straining GaGeTe 5% and compressing $AlGeS_3$ 5%, and the bandgap of GaGeTe rapidly drops due to positive biaxial strain.[2] This explains the drop of the bandgap in the frustrated bilayer.

The C2DB bandgaps mentioned above have for the individual layers have been retrieved them from the C2DB database, which have been computed using the HSE06 method.[3]

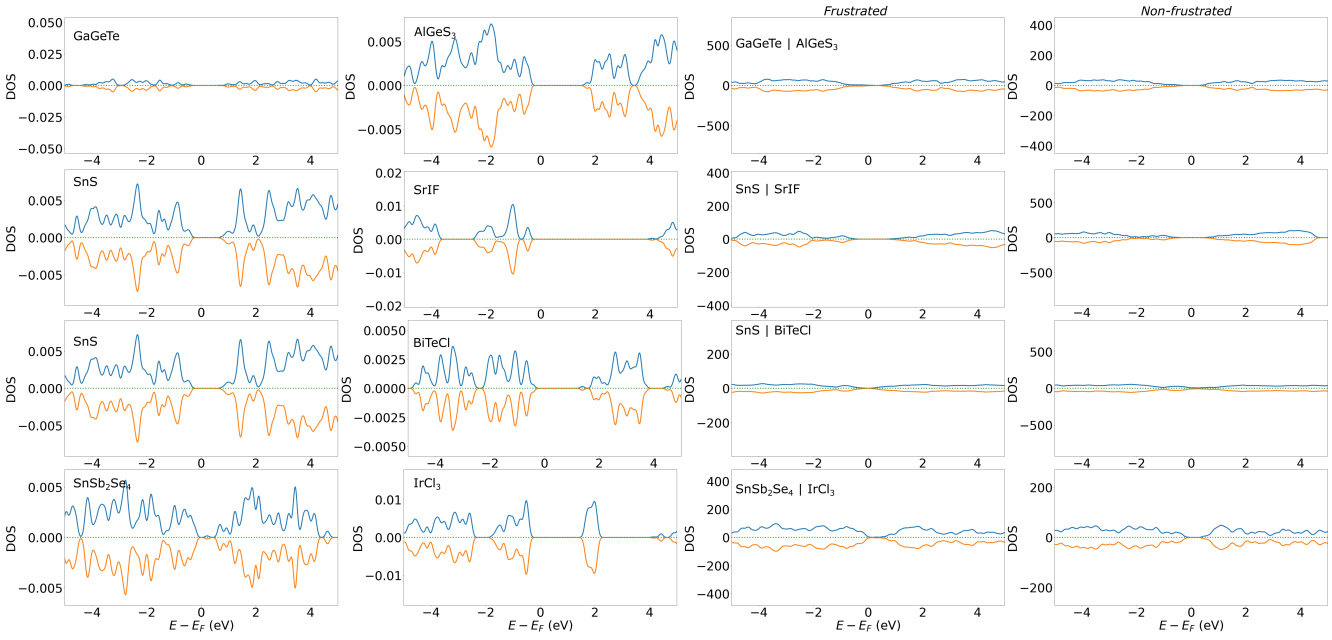

FIG. S1. The electronic density of states (DOS) of the materials GaGeTe | $AlGeS_3$, $SnSb_2Se_4$ | $IrCl_3$, SnS | BiTeCl and SnS | SrIF, as well as the DOS of the individual layers.

TABLE S3. The difference in total energy/atom, ΔE_{f-n} (meV/atom), between the frustrated and the non-frustrated supercells; the binding energy between the two layers in frustrated, E_b^f (meV/atom), and the non-frustrated, E_b^n (meV/atom); the electronic band gaps in the frustrated, E_g^f (eV), the non-frustrated, E_g^n (eV), supercells; the optimised lattice constants in Layer 1, Layer 1 a_{opt} and Layer 1 b_{opt}, and in Layer 2, Layer 2 a_{opt} and Layer 2 b_{opt}; and the qualification of whether structural integrity is maintained after the AIMD simulation.

Layer 1 ($+5\%$)	**Layer 2** (-5%)	ΔE_{f-n}	E_b^f	E_b^n	E_g^f	E_g^n	**Layer 1** a_{opt}	**Layer 1** b_{opt}	**Layer 2** a_{opt}	**Layer 2** b_{opt}	**AIMD**
SnS	BiTeCl	$+38$	-143	-129	174	23	3.6	0.8	-7.0	-9.6	No
GaGeTe	$AlGeS_3$	-58	-16	-78	112	733	6.8	6.8	-4.2	-4.2	Yes
$SnSb_2Se_4$	$IrCl_3$	$+39$	-25	-63	120	293	4.8	4.8	-5.9	-5.9	Yes
SnS	SrIF	$+16$	-67	-99	1159	952	7.1	7.1	-3.9	-3.9	No

References

[1] Y. Qi, M. A. Sadi, D. Hu, M. Zheng, Z. Wu, Y. Jiang, and Y. P. Chen, Advanced Materials **35**, 2205714 (2023), ISSN 0935-9648, 1521-4095, URL https://onlinelibrary.wiley.com/doi/10.1002/adma.202205714.

[2] J. Zhang, S.-s. Li, W.-x. Ji, C.-w. Zhang, P. Li, S.-f. Zhang, P.-j. Wang, and S.-s. Yan, J. Mater. Chem. C **5**, 8847 (2017), ISSN 2050-7526, 2050-7534, URL https://xlink.rsc.org/?DOI=C7TC03001K.

[3] A. V. Krukau, O. A. Vydrov, A. F. Izmaylov, and G. E. Scuseria, The Journal of Chemical Physics **125**, 224106 (2006), ISSN 0021-9606, 1089-7690, URL https://pubs.aip.org/jcp/article/125/22/224106/953719/Influence-of-the-exchange-screening-parameter-on.