Supporting Information for

Nonlinear Optical ASnX (A = Na, H; X = N, P) Nanosheets with Divalent Tin Lone Electron Pair Effect by First-Principles Design

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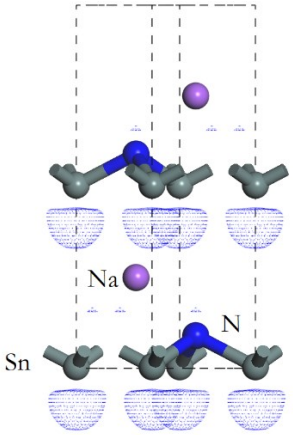
1. **Table S1** Experimental and calculated crystallographic data of $ZnGeP_2$ and NaSnX (X = N, P). It can be found that the LDA results are sufficiently accurate for the calculations of lattice constants.

			a=b (Å) (relative error)	c (Å) (relative error)
$ZnGeP_2$	Exp.		5.465	10.711
	Cal.	by LDA	5.387 (-1.4%)	10.496 (-2.0%)
	Cal.	by sX-LDA	5.270 (-3.6%)	10.342 (-3.4%)
NaSnN	Exp.		3.331	10.892
	Cal.	by LDA	3.201 (-3.9%)	10.773 (-1.1%)
	Cal.	by sX-LDA	3.176 (-4.7%)	10.774 (-1.1%)
NaSnP	Exp.		3.880	11.667
	Cal.	by LDA	3.775 (-2.7%)	11.602 (-0.6%)
	Cal.	by sX-LDA	3.795 (-2.2%)	11.664 (-0.0%)

2. **Figure S1.** First-principles analysis of electron localization function (ELF) for NaSnN.



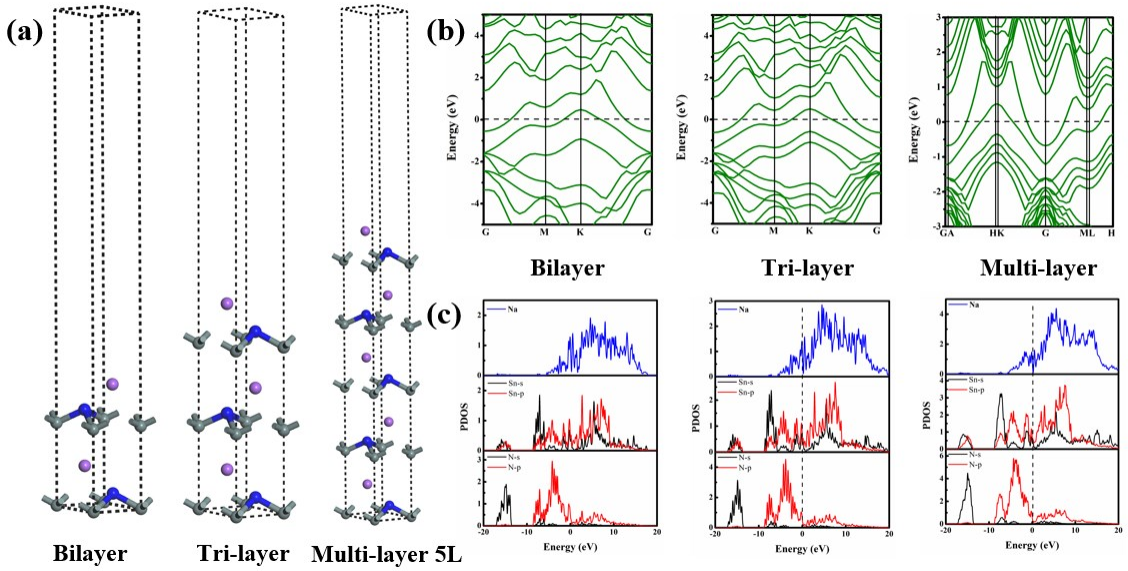
3. **Table S2.** Detailed VE and VH contribution to the total SHG coefficient d_{ijk} as shown in Figure 3.

System			d_{ijk} (pm/V)	VE (pm/V)	VH (pm/V)
NaSnN	Bulk	Cal.	d_{333}=27	d_{333}=18	d_{333}=9.4
HSnN	ML	Cal.	d_{111}=147	d_{111}=256	d_{111}=-108
NaSnN	ML [a)]	Cal.	d_{113}=301; d_{333}=277	d_{113}=259; d_{333}=395	d_{113}=42; d_{333}=-118
NaSnP	Bulk	Cal.	d_{333}=16	d_{333}=6.7	d_{333}=9.7
NaSnP	ML	Cal.	d_{113}=618; d_{333}=533	d_{113}=565; d_{333}=662	d_{113}=54; d_{333}=-130
HSnP	ML	Cal.	d_{111}=12; d_{333}=-14	d_{111}=43; d_{333}=-9.4	d_{111}=-31; d_{333}=-4.8

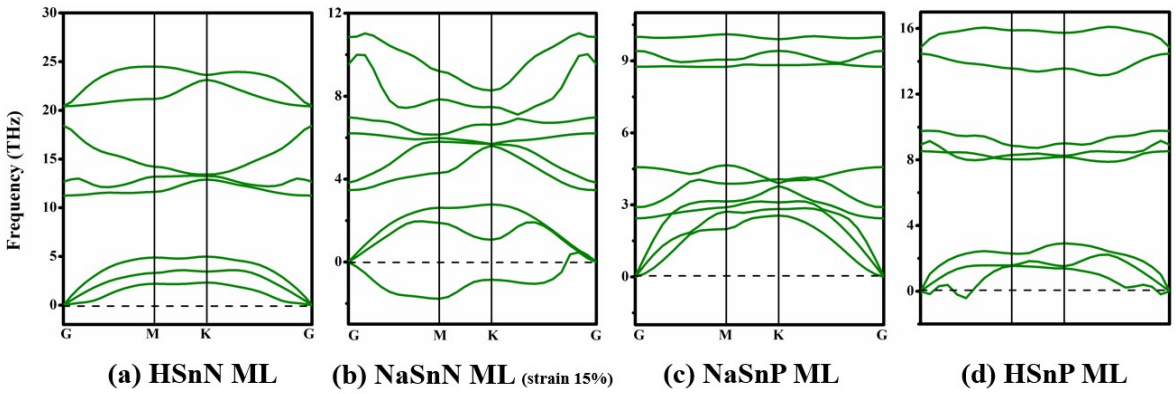
[a)] strain is 15%

4. **Figure S2.** Crystal structures (a), band structures (b) and PDOS (c) of the NaSnN bilayer, tri-layer and

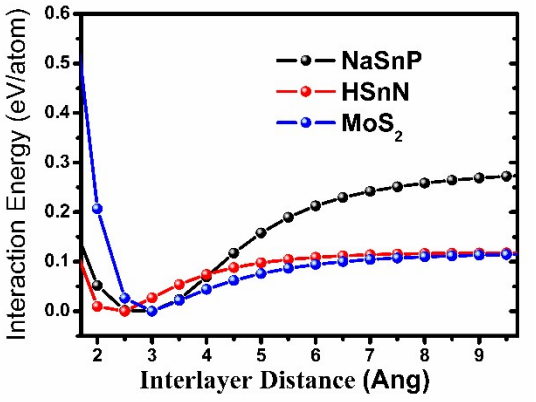
multi-layer (e.g., 5L)



5. **Figure S3.** Phonon vibration spectra of HSnN monolayer (ML) (a), 15%-strained NaSnN ML (b), NaSnP ML (c) and HSnP ML (d).



6. **Figure S4.** Interaction energies of HSnN, NaSnP and MoS_2 with respect to the interlayer distance.


Interaction Energy (eV/atom)
Interlayer Distance (Ang)
NaSnP
HSnN
MoS2
0.0
0.1
0.2
0.3
0.4
0.5
0.6
2
3
4
5
6
7
8
9