

Supporting Information for

**Ultra-low thermal conductivity and high thermoelectric performance
of two-dimensional triphosphides (InP₃, GaP₃, SbP₃ and SnP₃):**

A comprehensive first-principles study

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1. The convergence test of energy with the variation of k -point.

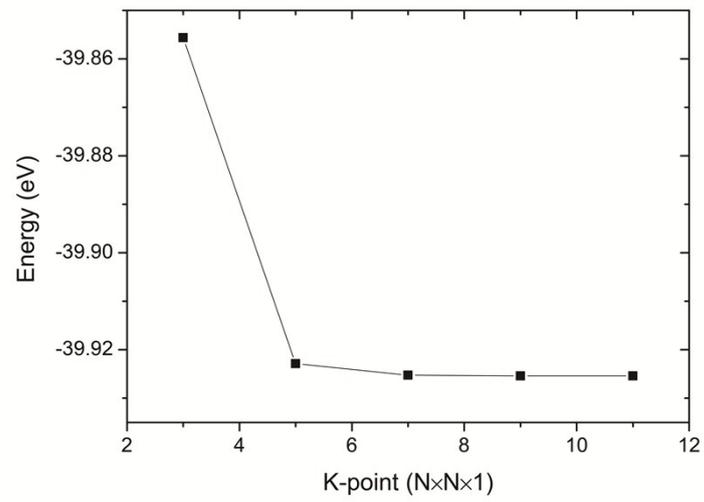


Fig. S1 The convergence test of energy for optimized structure of SbP_3 with the variation of k -point of the Brillouin zone.

2. The convergence test of lattice parameter for optimized structure of SbP_3 with the variation of the distance along the *out-of-plane* direction.

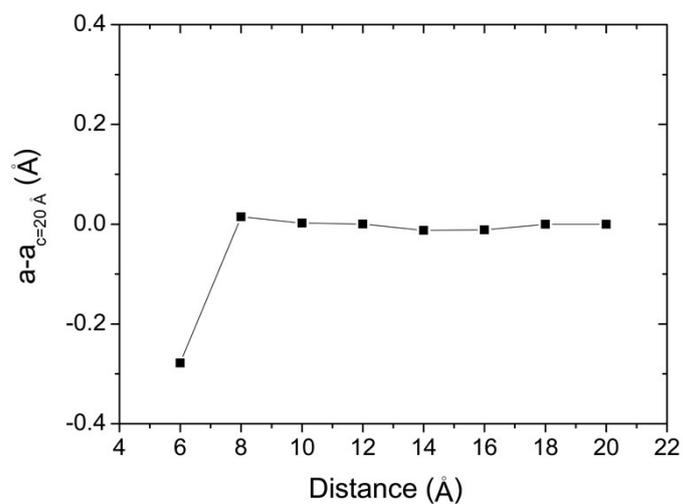


Fig. S2 The convergence test of lattice parameter for optimized structure of SbP_3 with the distance along the *out-of-plane* (c) direction, where a is optimized lattice parameter, $a_{c=20\text{Å}}$ means optimized lattice parameter when using 20 Å as the distance along the *out-of-plane* (c) direction for structural optimization.

3. The locally magnified phonon dispersion of flexural phonon branch.

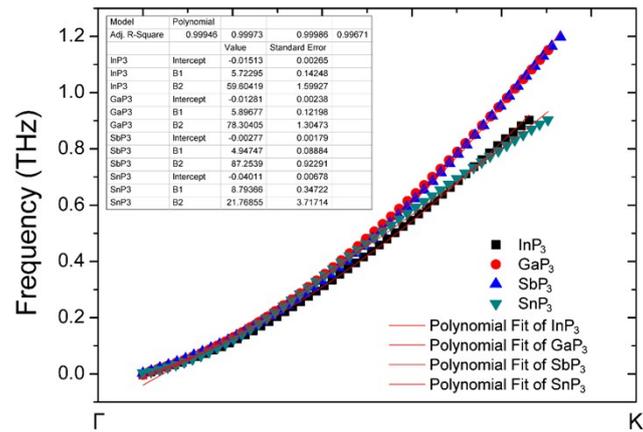


Fig. S3 The phonon dispersion of flexural phonon branch along the Γ -K direction.

4. Detailed information about two-dimensional *in-plane* elastic modulus.

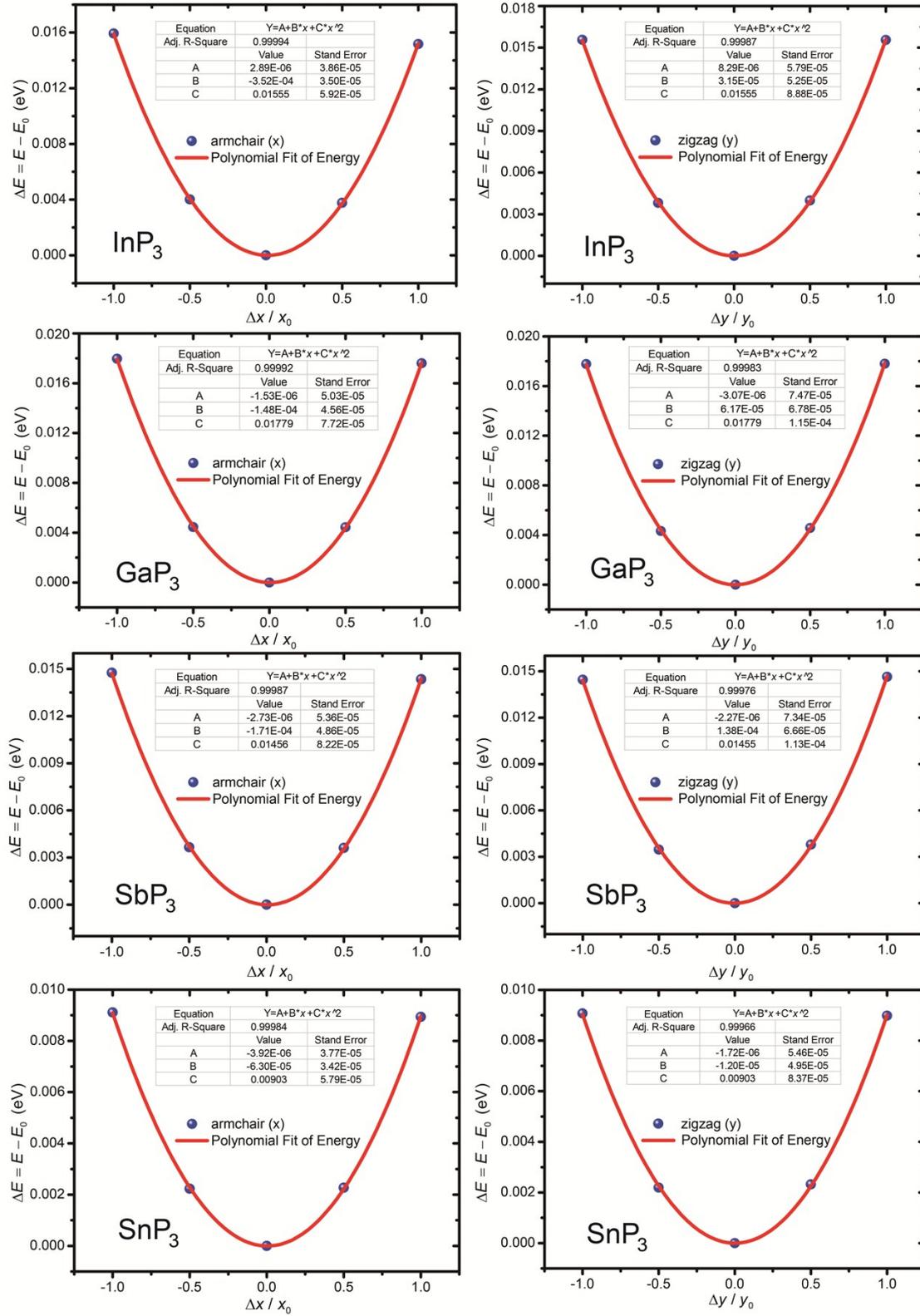


Fig. S4 The variation of energy under strain along the armchair and zigzag directions.

5. Detailed information about deformation potential energy.

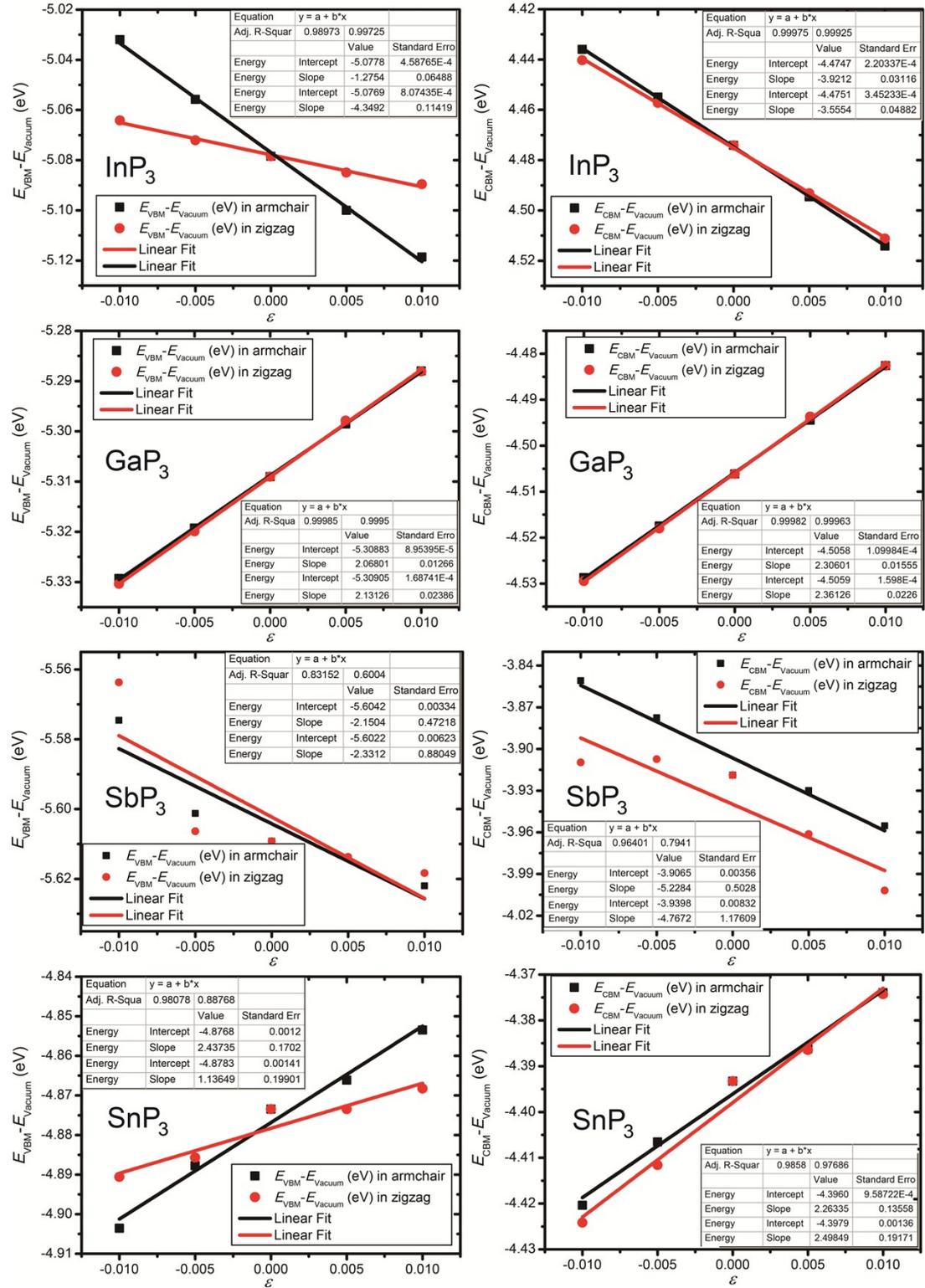


Fig. S5 The CBM and VBM along the armchair and zigzag directions as a function of deformation proportion.

6. The electronic band structures of monolayer SnP₃.

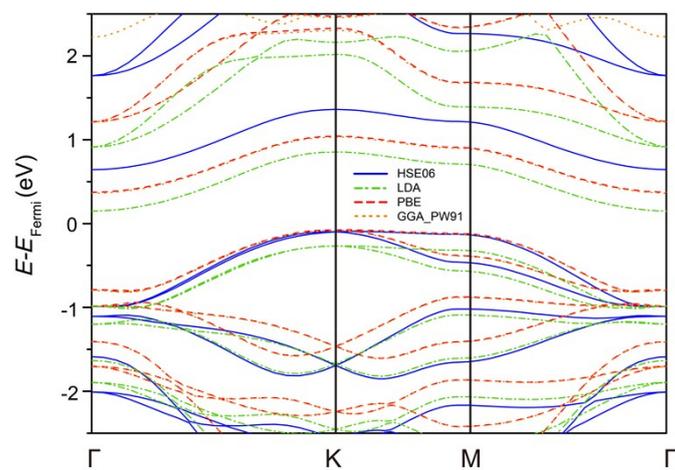


Fig. S6 Electronic band structures of monolayer SnP₃ calculated by performing HSE06, LDA, PBE and GGA functionals.

7. The effect of considering spin-orbit coupling (SOC) on the band structures of monolayer SnP₃.

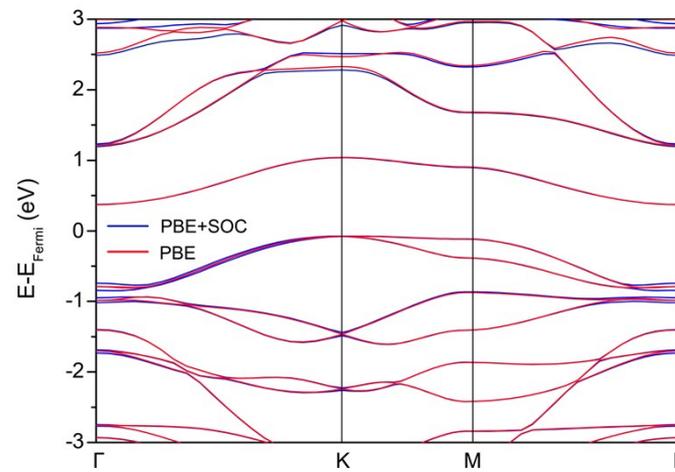


Fig. S7 Electronic band structures of monolayer SnP₃ calculated by PBE functional with SOC and without SOC.

8. The thermoelectric performance of triphosphides for not considering the computational uncertainty of electronic relaxation times.

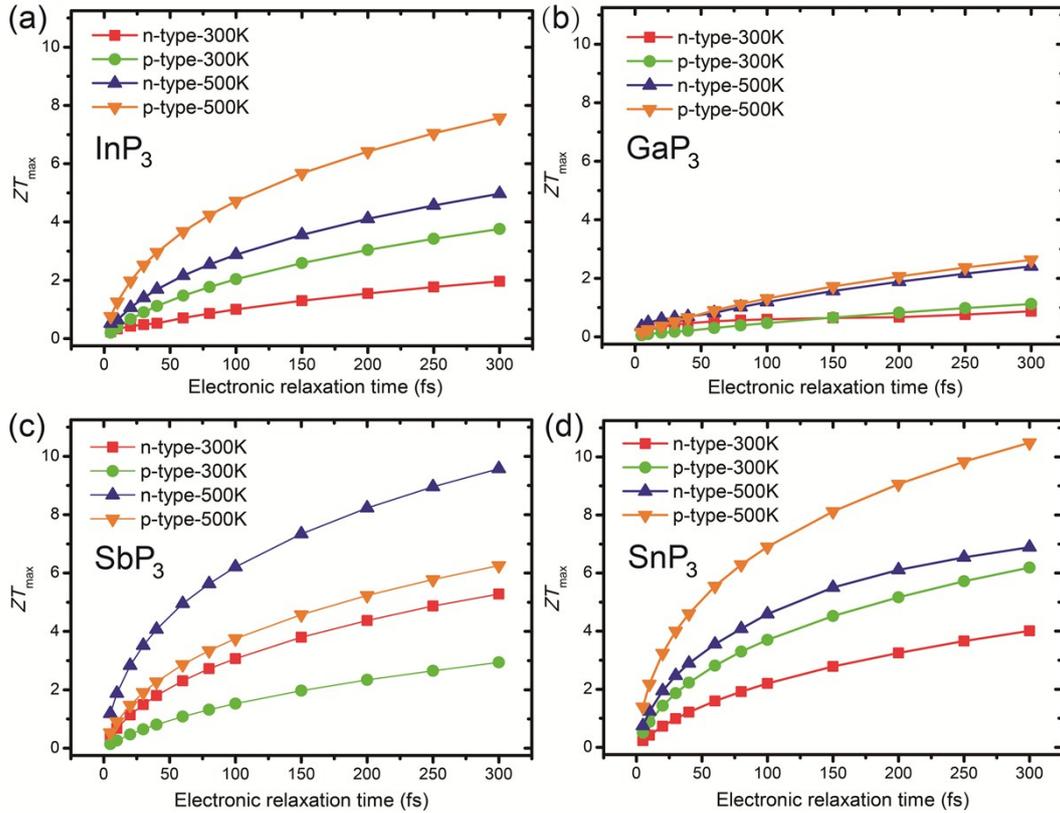


Fig. S8 The maximum values of thermoelectric figure of merit (ZT) of monolayer triphosphides with n-type and p-type doping versus electronic relaxation time at 300 and 500 K for (a) InP_3 (b) GaP_3 (c) SbP_3 (d) SnP_3 .

Although we have calculated the electronic relaxation times of triphosphides (Table 1), the electronic relaxation times acquired from deformation potential theory have large difference, even across the scale. Our main preoccupation is exploring the candidate of thermoelectric materials rather than discussion the accuracy of electronic relaxation times. Thus, we use Fig. 8 to account for the thermoelectric performance of triphosphides for not considering the computational uncertainty of electronic relaxation times. Fig. 8 represent the ZT_{max} of p-type doping of InP_3 and SnP_3 are higher than n-type doping under the same electronic relaxation time, while that of p-type doping of SbP_3 is lower than n-type doping, which also can be reflected from the Seebeck coefficient (Fig. 5). Obviously, the higher thermal conductivity

hinders the thermoelectric performance of GaP₃. We observe a rapid increase of ZT_{\max} in electronic relaxation time up to 300 fs for InP₃, SbP₃ and SnP₃. The calculated evolution of ZT_{\max} shows that monolayer InP₃, SbP₃ and SnP₃ is quite suitable for the future application as thermoelectric materials. Although the thermoelectric performance of GaP₃ is ultimately limited by the high lattice thermal conductivity, there are tunable methods that reduce the lattice thermal conductivity for promoting the thermoelectric performance.

9. **Table S1** A comparison from different work for carrier mobility of SnP₃ acquired from deformation potential theory.

μ^{2D} (cm ² /Vs) in zigzag direction	μ^{2D} (cm ² /Vs) in armchair direction	
132	105	Our work
302	703	<i>Nanoscale</i> , 11 , 19923-19932 (2019)
3890	4360	<i>J. Phys.: Condens. Matter.</i> , 30 , 475702 (2018)
172	359	<i>J. Mater. Chem. A</i> , 6 , 11890-11897 (2018)

10. **Table S2** A comparison of the different relaxation time under different temperature calculated by deformation potential theory.

	Carrier type	τ at 300 K (10^{-15} s)	τ at 500 K (10^{-15} s)
InP ₃	Electron (zigzag)	157.96	94.77
	Electron (armchair)	183.31	109.99
	Hole (zigzag)	154.88	92.93
	Hole (armchair)	13.19	7.91
GaP ₃	Electron (zigzag)	122.64	73.58
	Electron (armchair)	192.86	115.72
	Hole (zigzag)	190.36	114.21
	Hole (armchair)	223.47	134.08
SbP ₃	Electron (zigzag)	23.40	14.04
	Electron (armchair)	24.38	14.63
	Hole (zigzag)	89.80	53.88
	Hole (armchair)	157.65	94.59
SnP ₃	Electron (zigzag)	62.38	37.43
	Electron (armchair)	37.94	22.76
	Hole (zigzag)	123.43	74.06
	Hole (armchair)	51.66	30.99