

Supporting Information

A valence balancing rule for design of bimetallic phosphides targeting high thermoelectric performance

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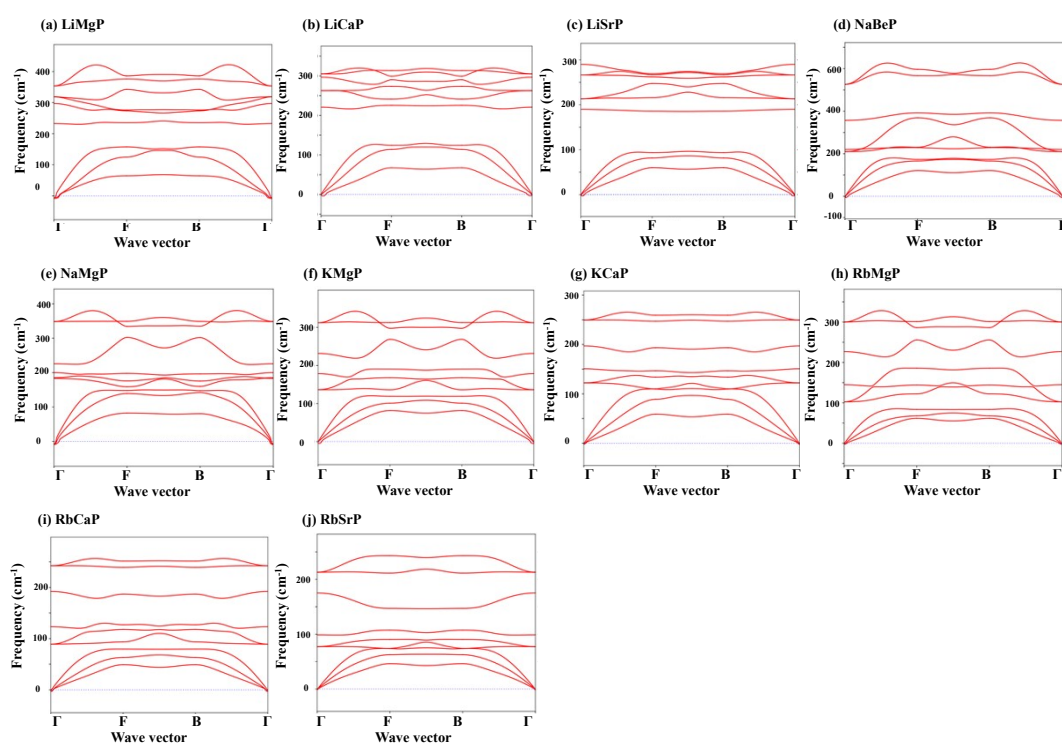


Fig. S1. Phonon dispersion relationships of the 2D ABP monolayers.

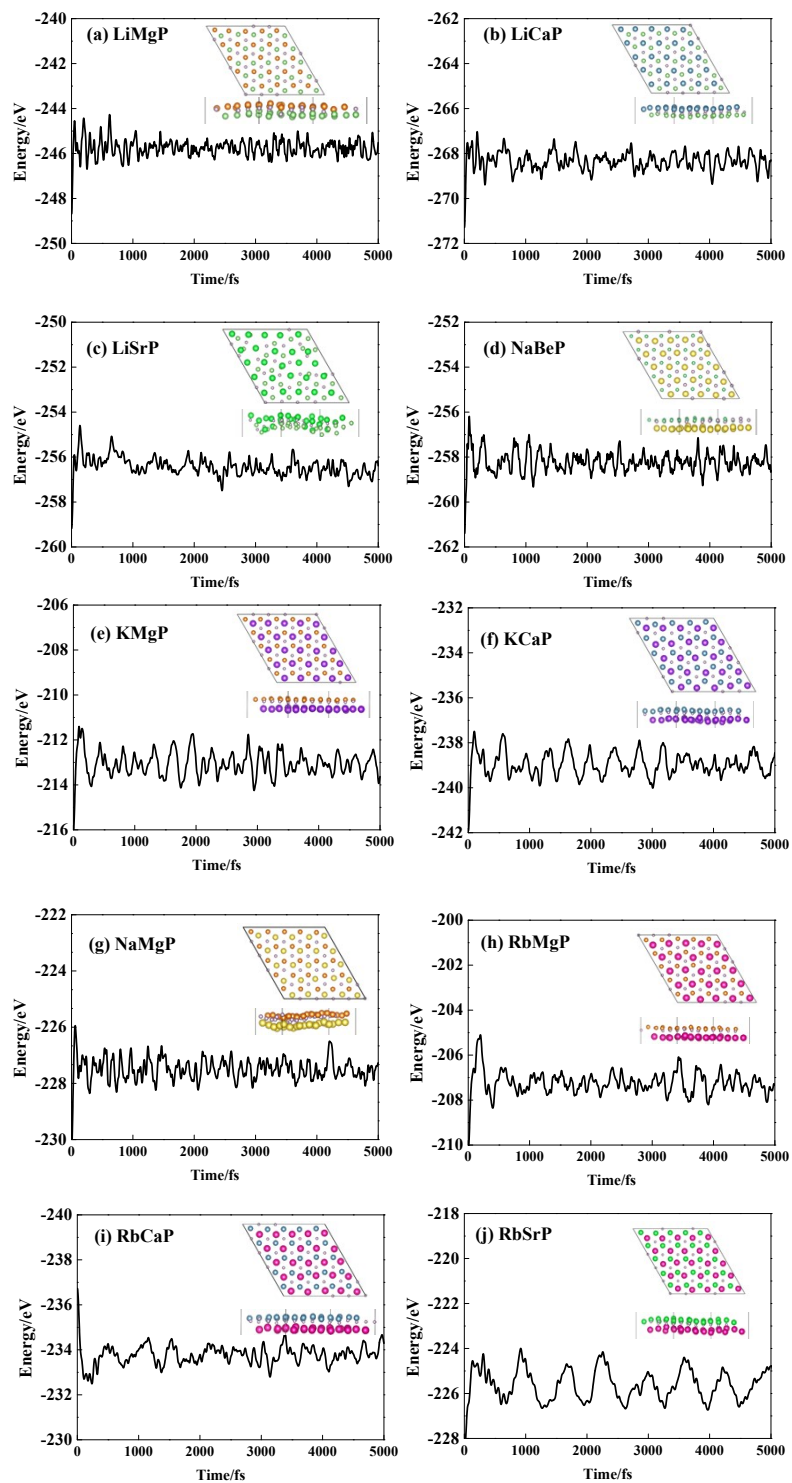


Fig. S2. Snapshots from MD simulations showing top and side views of the 2D ABP monolayers at 300 K. Also shown is the variation of the total energy over a 5 ps simulation time.

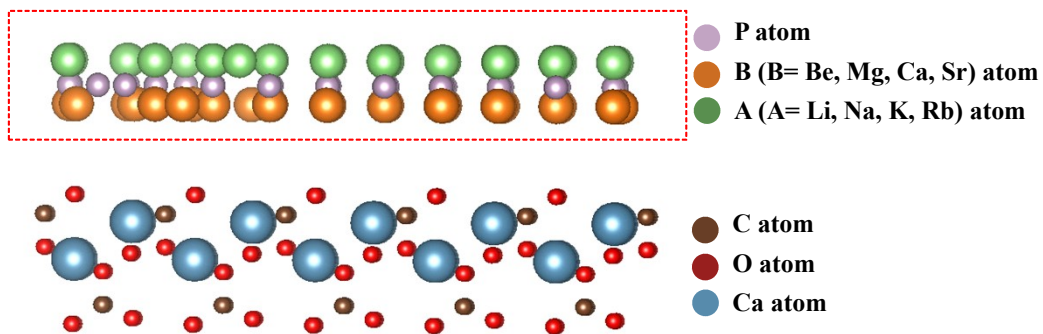


Fig. S3. Schematic representation of ABP grown on the surface of CaCO₃.

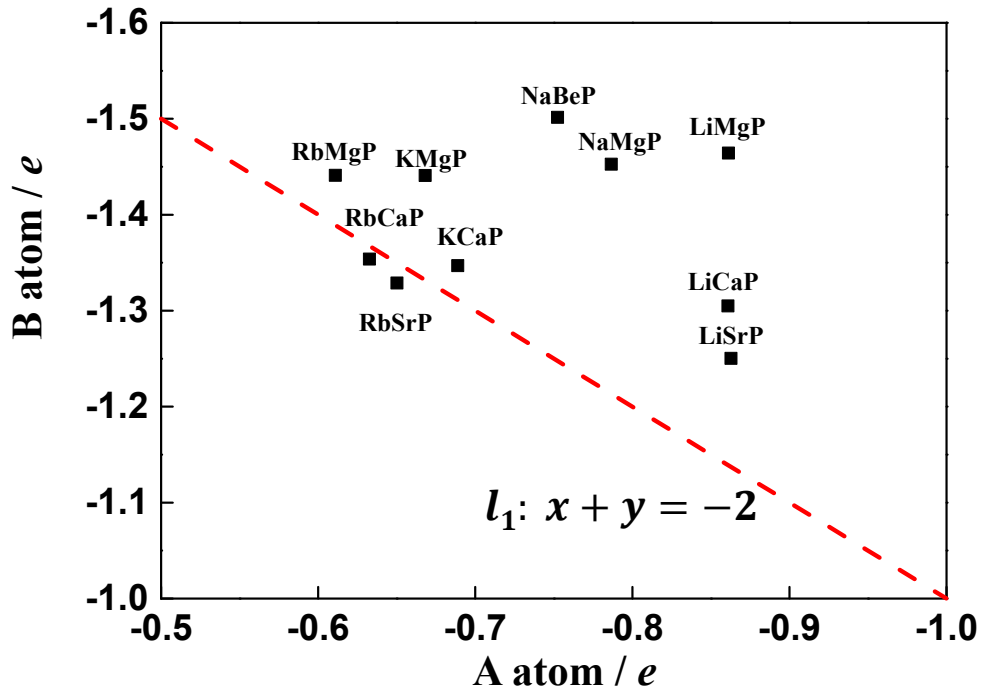


Fig. S4. Bader charge analysis for A (A = Li, Na, K, Rb) and B (B = Be, Mg, Ca, Sr) atoms in the 2D ABP monolayers. Each P atom gains about 2 electrons from the neighboring metal atoms. Among them, the A element contributes about 0.5 electrons more than the B atom, indicating that the B–P bond is more sensitive than the A–P bond to the electronegativity of the metal atom.

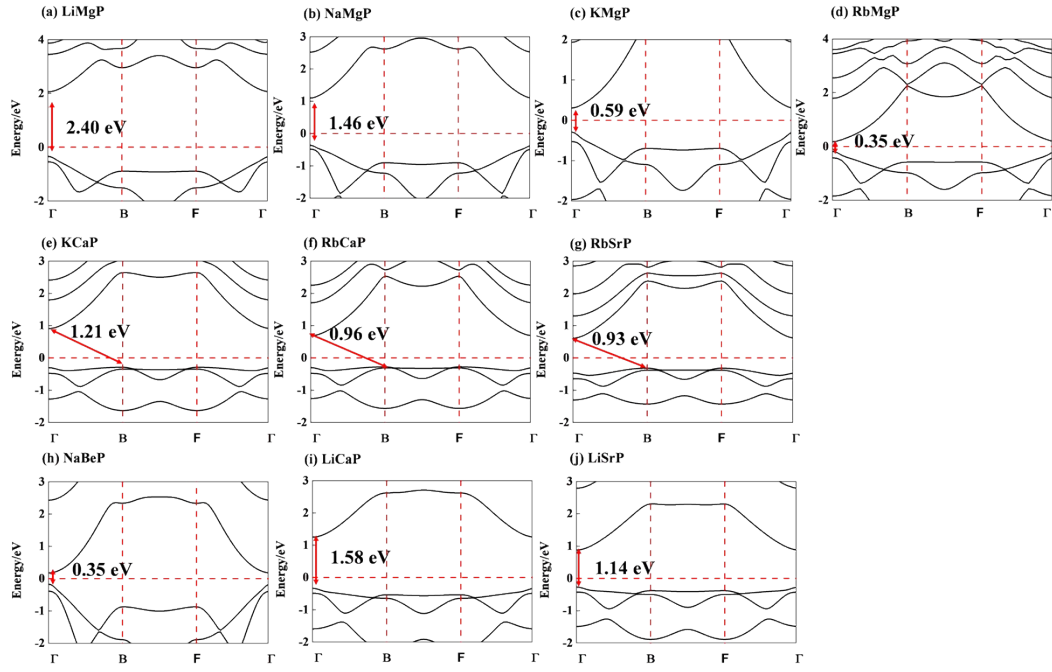


Fig. S5. Electronic band structures of the 2D ABP monolayers calculated using the HSE06 function.

The Fermi level is set to zero.

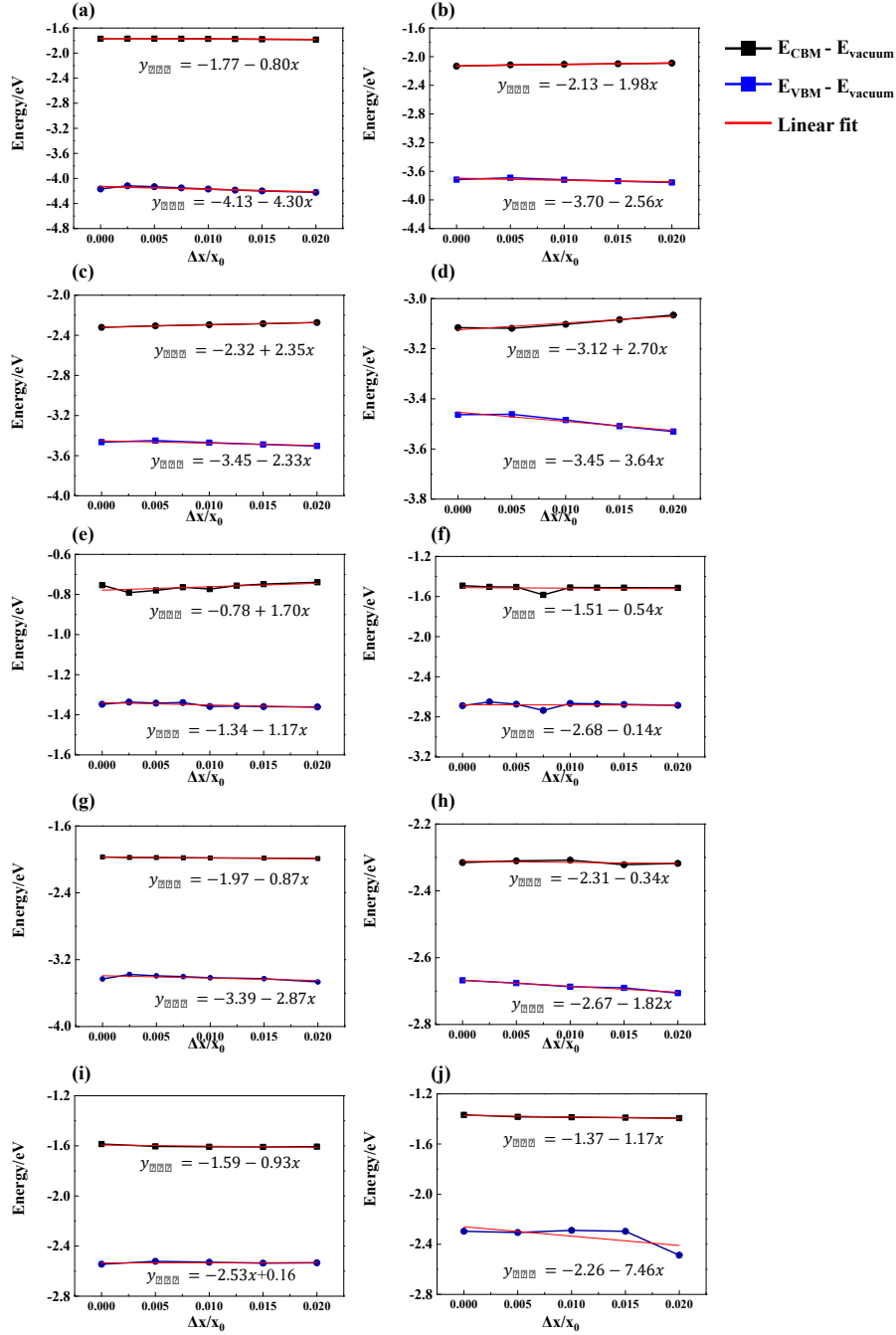


Fig. S6. Energies of the CBM and VBM of the 2D (a) LiMgP, (b) LiCaP, (c) LiSrP, (d) NaBeP, (e) KMgP, (f) KCaP, (g) NaMgP, (h) RbMgP, (i) RbCaP and (j) RbSrP, respectively. The band energies were calculated using the HSE06 function. The solid red lines are fitting curves.

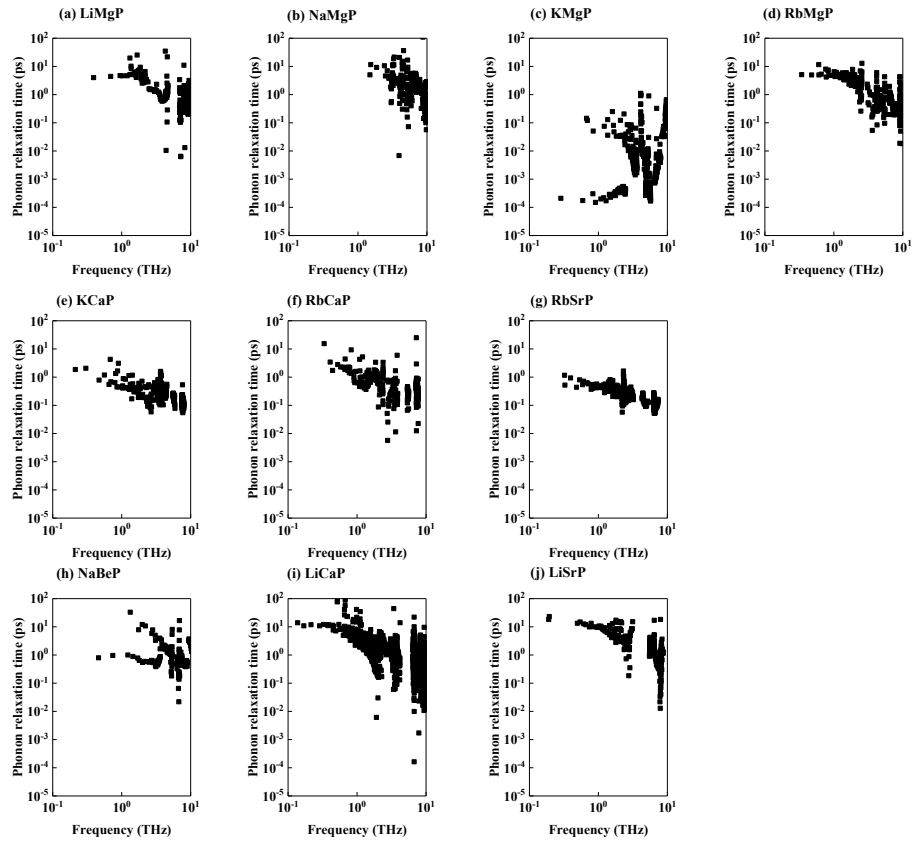


Fig. S7. Phonon relaxation times of the ABP monolayers.

Table S1. The in-plane lattice parameter a (Å), and lengths of the A–P and B–P bonds, d_{A-P} and d_{B-P} , respectively, for the 2D ABP monolayers.

		d_{A-P} (Å)	d_{B-P} (Å)	a (Å)
Type i	LiMgP	2.56	2.49	4.15
	NaMgP	2.85	2.53	4.27
	KMgP	3.15	2.57	4.36
	RbMgP	3.28	2.58	4.37
Type ii	KCaP	3.22	2.82	4.36
	RbCaP	3.35	2.83	4.70
	RbSrP	3.39	2.98	4.89
Type iii	LiCaP	2.65	2.74	4.36
	LiSrP	2.71	2.88	4.49
	NaBeP	2.77	2.16	3.73