Supplementary Information

for

Stable Multifunctional Aluminum Phosphides at High Pressures

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Supplementary Figures

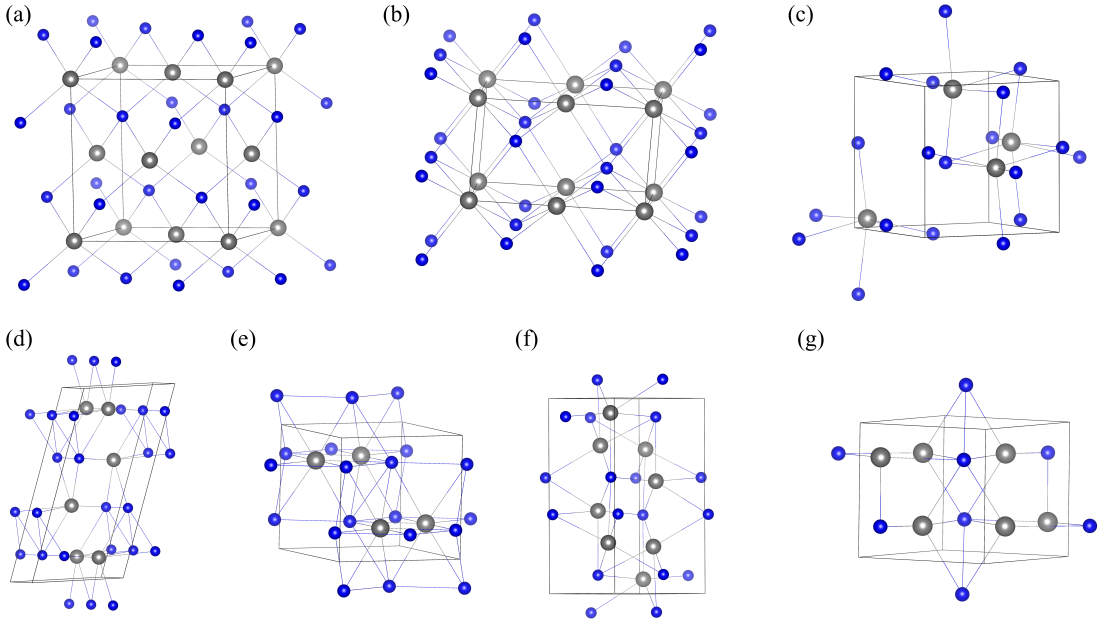

Figure S1. Crystal structures of stable Al-P compounds. (a) *F*-43*m* phase of AlP at ambient pressure. (b) $P6_3/mmc$ phase of AlP at 20 GPa. (c) $P2_13$ phase of AlP at 50 GPa. (d) *C*2/*m* phase of AlP at 100 GPa. (e) *Pnma* phase of AlP at 100 GPa. (f) *Pnma* phase of Al_2P at 150 GPa. (g) *P*1 phase of Al_3P at 150 GPa. The small blue and large grey represent P and Al atoms, respectively.

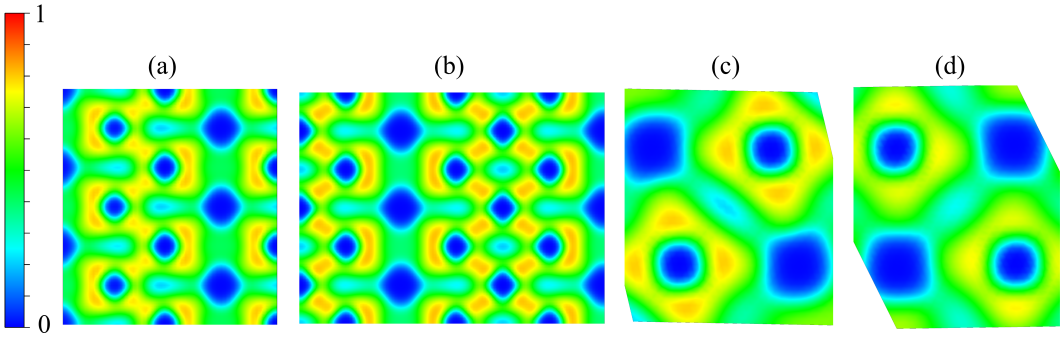

Figure S2. (a) Calculated ELF plots in (-1, 0, -3) section of AlP_2 in the *I*4/*mmm* phase of 100 GPa. (b) Calculated ELF plots in (0, 1, 0) section of AlP_3 in the *Immm* phase of 150 GPa. (c) Calculated ELF plots in (1, -3, 1) section of Al_2P in the *I*4/*mmm* phase of 100 GPa. (d) Calculated ELF plots in (1, 1, 2) section of Al_3P in the *P*1 phase of 150 GPa. The large atom is Al, and the small atom is P

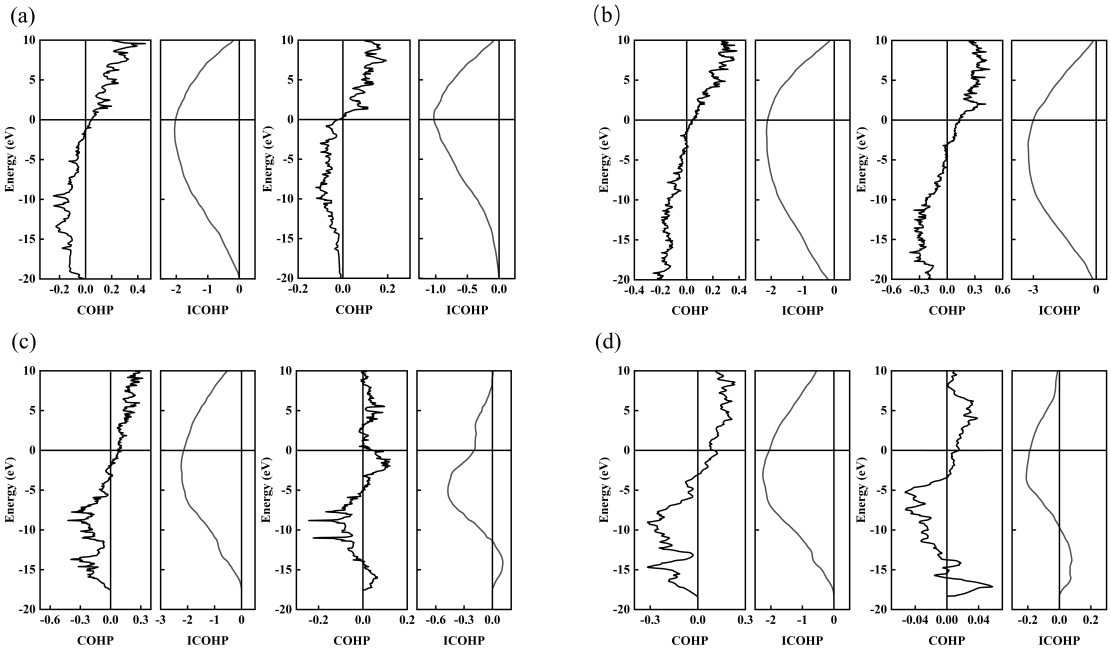

Figure S3. Plots of the COHP and ICOHP of Al-P and P-P, respectively. (a) AlP_2 for *I*4/*mmm* phase at 100 GPa. (b) AlP_3 for *Immm* phase at 150 GPa. (c) Al_2P for *I*4/*mmm* phase at 100 GPa. (d) Al_3P for *P*1 phase at 150 GPa.

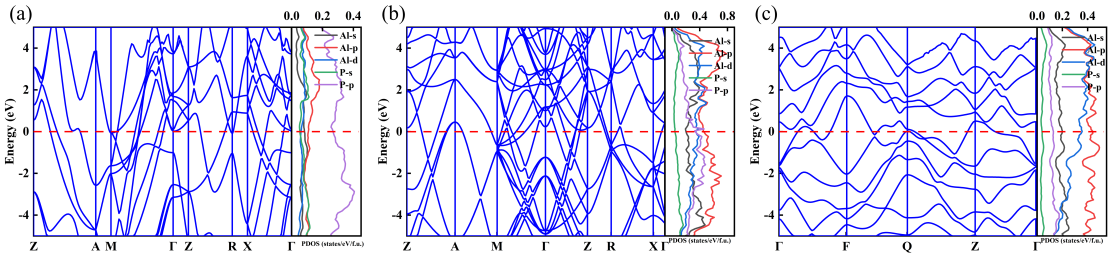

Figure S4. Energy band structure and density of states for Al-P phases. (a), (b) and (c) represent *I*4/*mmm* phase of AlP_2 at 100 GPa, *I*4/*mmm* phase of Al_2P at 100 GPa, *P*1 phase of Al_3P at 150 GPa, respectively.

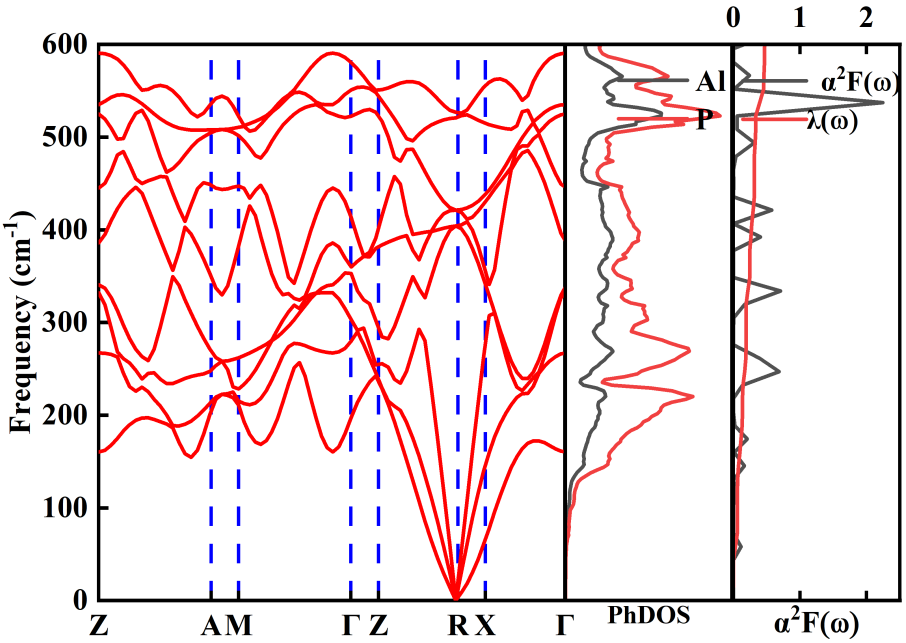

Figure S5. Calculated phonon spectra, and phonon density of states (PhDOS), Eliashberg spectral function $\alpha^2F(\omega)$, and electron−phonon coupling integral $\lambda(\omega)$ of AlP_2 at 100 GPa.

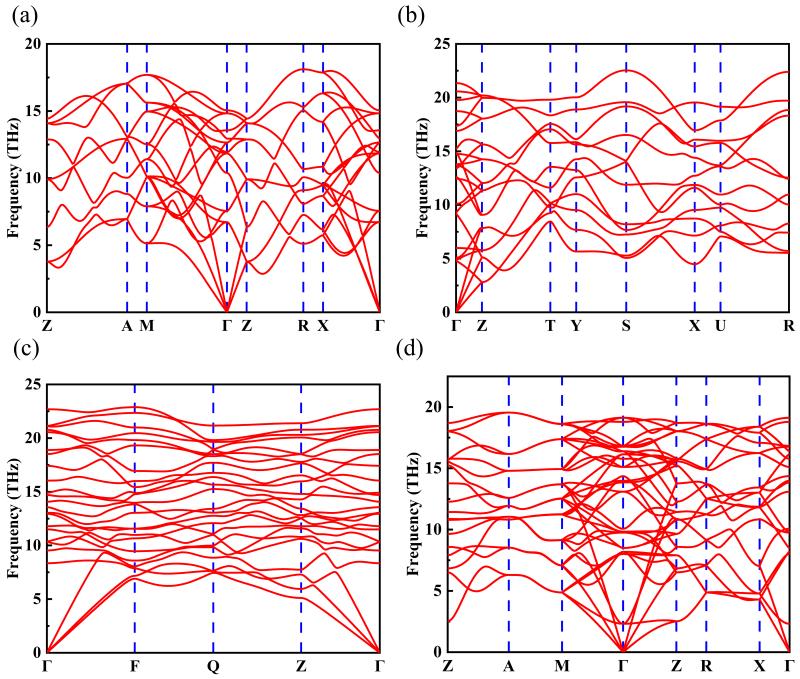

Figure S6. (a) Phonon dispersion curves of AlP_2 in the *I*4/*mmm* phase at 100 GPa. (b) Phonon dispersion curves of AlP_3 in the *Immm* phase at 150 GPa. (c) Phonon dispersion curves of Al_3P in the *P*1 phase at 100 GPa. (d) Phonon dispersion curves of Al_2P in the *I*4/*mmm* phase at 100 GPa.

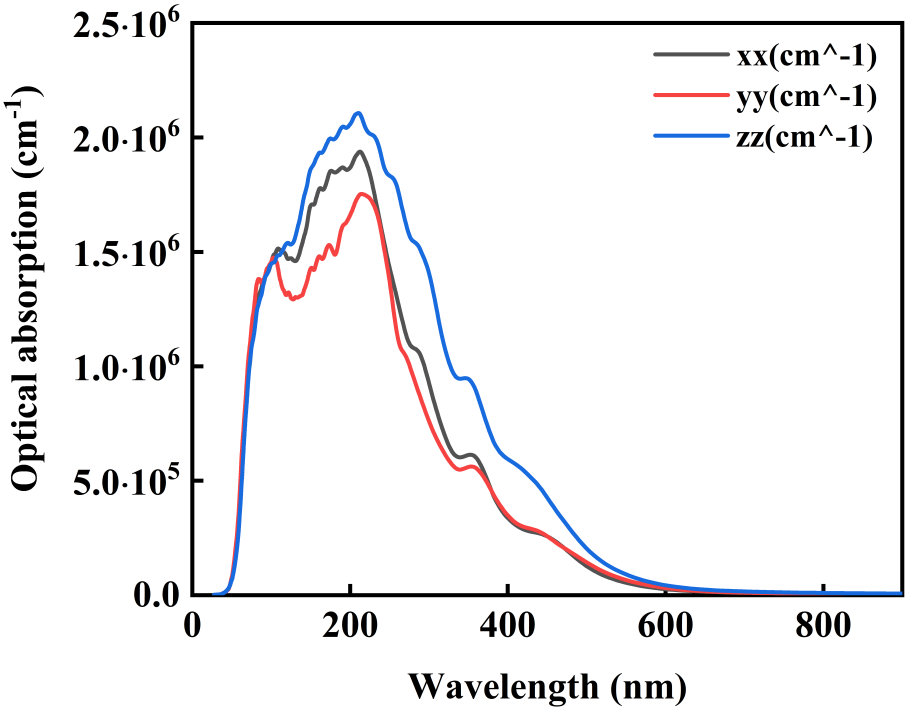

Figure S7. The optical absorption coefficient of *P*21-AlP_2 at 0 GPa.

Supplementary Tables

Table S1. Structural information of predicted Al-P phases.

Space group Pressure	Lattice parameters (Å, °)	Atomic coordinates (fractional)				Sites
F-43*m*-AlP	a = 5.506	Al1	0.000	0.000	0.000	4a
0 GPa	b =5.506	P1	0.250	0.250	0.250	4c
	c =5.506					
	α = β = γ = 90					
*P*63/*mmc*-AlP	a = 3.424	Al1	0.000	0.000	0.000	2a
20 GPa	b = 3.424	P1	0.667	0.333	0.250	2d
	c = 5.539					
	α = β = 90					
	γ = 120					
$P2_13$ - AlP	a = 4.510	Al1	0.062	0.062	0.062	4a
60 GPa	b =4.510	P1	0.445	0.445	0.445	4a
	c =4.510					
	α = β = γ = 90					
*C*2/*m* - AlP	a = 7.918	Al1	0.886	-0.000	0.667	4i
100 GPa	b = 2.737	P1	0.136	0.500	0.845	4i
	c = 3.775					
	α = γ = 90					
	β = 108.368					

Pnma - AlP	a = 4.052	Al1	-0.103	0.250	0.875	4c
150 GPa	b = 3.799	P1	0.105	0.750	0.627	4c
	c = 4.511					
	α = β = γ = 90					
$P2_1$ – AlP_2	a = 6.799	Al1	0.000	0.000	0.000	2a
0 GPa	b = 6.692	Al2	0.500	0.500	0.500	2d
	c = 5.098	P1	0.297	0.250	0.138	2e
	α = γ = 90	P3	0.524	0.750	0.144	2e
	β = 111.285	P5	0.197	0.750	0.413	2e
		P7	0.810	0.250	0.165	2e
I4/mmm – AlP_2	a = 2.709	Al1	0.500	0.500	0.500	2a
100 GPa	b = 2.709	P1	0.000	0.000	0.324	4e
	c = 7.804					
	α = β = γ = 90					
Immm - AlP_3	a = 2.666	Al1	-0.000	0.500	0.000	2d
150 GPa	b = 3.650	P1	0.500	-0.000	0.000	2b
	c = 7.055	P2	0.500	0.500	0.268	4i
	α = β = γ = 90					
I4/mmm - Al_2P	a = 2.867	Al1	-1.500	0.500	0.162	4e
100 GPa	b = 2.867	P1	-2.000	0.000	0.000	2a
	c = 7.210					
	α = β = γ = 90					
Pnma - Al_2P	a = 4.661	Al1	0.140	0.750	-0.925	4c
150 GPa	b = 3.254	Al3	0.389	0.250	-0.759	4c
	c = 6.985	P1	0.358	0.250	-0.095	4c
	α = β= γ = 90					
*P*1 - Al_3P	a = 4.021	Al1	0.758	0.623	0.806	1a
150 GPa	b = 4.038	Al2	0.753	0.130	0.559	1a
	c = 4.630	Al3	0.254	0.875	0.939	1a
	α = 89.753	Al4	0.242	0.377	0.194	1a
	β = 105.829	Al5	0.247	0.870	0.441	1a
	γ = 81.222	Al6	0.746	0.125	0.061	1a
		P1	0.738	0.626	0.304	1a
		P2	0.262	0.374	0.696	1a

Table S2. Calculated elastic constants (GPa) of AlP and AlP_2 at atmospheric pressure.

Structure	C_{11}	C_{22}	C_{33}	C_{44}	C_{55}	C_{66}	C_{12}	C_{13}	C_{14}	C_{15}	C_{25}	C_{35}	C_{46}
$P2_1$	175.68	184.97	228.94	37.41	59.31	52.27	29.24	36.48	39.72	-2.99	-5.99	-12.5	7.6
F-43*m*	126.56			60.51			61.38						