

Supplementary information

A list of supplementary information, which may help readers to understand the manuscript better.

1. A table of the atomic positions of the $A_{0.50}B_{0.50}C$ 32-atom SQS supercell.
2. A table of the atomic positions of the $A_{0.25}B_{0.75}C$ 32-atom SQS supercell.
3. A table of the computed band bowing parameters for the five semiconducting alloys compared with experiment
4. A figure showing the computed direct, Γ , indirect, X, and indirect, L, gap energies for the full composition range of four semiconducting alloys compared with experimental fits given by Vurgaftman *et al.*

TABLE I. Atomic positions of the $A_{0.50}B_{0.50}C$ 32-atom SQS supercell. The lattice vectors are defined as $\vec{a}_1 = \frac{a}{2}(4, 2, 2)$, $\vec{a}_2 = \frac{a}{2}(-4, 2, 2)$, and $\vec{a}_3 = \frac{a}{2}(0, -1, 1)$. The following atomic positions are listed as $\vec{R} = x_1\vec{a}_1 + x_2\vec{a}_2 + x_3\vec{a}_3$. The anion sublattice (C-atoms) is offset from the cation sublattice with atomic positions described by $(x_1 + \frac{3}{16}, x_2 + \frac{1}{16}, x_3)$.

Atom	(x_1, x_2, x_3)	(x_1, x_2, x_3)	(x_1, x_2, x_3)
A	$(0, 0, 0)$	$(\frac{1}{4}, 0, 0)$	$(\frac{1}{2}, 0, 0)$
	$(\frac{3}{4}, \frac{1}{4}, 0)$	$(0, \frac{3}{4}, \frac{1}{2})$	$(\frac{1}{2}, \frac{3}{4}, \frac{1}{2})$
	$(\frac{3}{4}, 0, \frac{1}{2})$	$(\frac{3}{4}, \frac{1}{2}, \frac{1}{2})$	
B	$(0, \frac{1}{2}, 0)$	$(\frac{1}{4}, \frac{3}{4}, 0)$	$(\frac{1}{2}, \frac{1}{2}, 0)$
	$(\frac{3}{4}, \frac{3}{4}, 0)$	$(0, \frac{1}{4}, \frac{1}{2})$	$(\frac{1}{4}, 0, \frac{1}{2})$
	$(\frac{1}{4}, \frac{1}{2}, \frac{1}{2})$	$(\frac{1}{2}, \frac{1}{4}, \frac{1}{2})$	

TABLE II. Atomic positions of the $A_{0.25}B_{0.75}C$ 32-atom SQS supercell. The lattice vectors are defined as $\vec{a}_1 = \frac{a}{2}(1, 1, -2)$, $\vec{a}_2 = \frac{a}{2}(0, -3, -1)$, and $\vec{a}_3 = \frac{a}{2}(-4, 1, -1)$. The following atomic positions are listed as $\vec{R} = x_1\vec{a}_1 + x_2\vec{a}_2 + x_3\vec{a}_3$. The anion sublattice (C-atoms) is offset from the cation sublattice with atomic positions described by $(x_1 + \frac{11}{16}, x_2 + \frac{5}{64}, x_3 + \frac{3}{64})$.

Atom	(x_1, x_2, x_3)	(x_1, x_2, x_3)	(x_1, x_2, x_3)
A	$(0, \frac{1}{4}, \frac{3}{4})$	$(\frac{1}{4}, \frac{15}{16}, \frac{9}{16})$	$(\frac{1}{4}, \frac{11}{16}, \frac{13}{16})$
	$(\frac{3}{4}, \frac{9}{16}, \frac{15}{16})$		
B	$(0, 0, 0)$	$(\frac{1}{4}, \frac{7}{16}, \frac{1}{16})$	$(\frac{1}{2}, \frac{7}{8}, \frac{1}{8})$
	$(\frac{3}{4}, \frac{5}{16}, \frac{3}{16})$	$(0, \frac{3}{4}, \frac{1}{4})$	$(\frac{1}{4}, \frac{3}{16}, \frac{5}{16})$
	$(\frac{1}{2}, \frac{5}{8}, \frac{3}{8})$	$(\frac{3}{4}, \frac{1}{16}, \frac{7}{16})$	$(0, \frac{1}{2}, \frac{1}{2})$
	$(\frac{1}{2}, \frac{3}{8}, \frac{5}{8})$	$(\frac{3}{4}, \frac{13}{16}, \frac{11}{16})$	$(\frac{1}{2}, \frac{1}{8}, \frac{7}{8})$

TABLE III. Band bowing parameters for five important semiconducting alloys obtained from HSE and PBE compared with experimental measurements. A composition dependent bowing parameter is used for the direct gap of AlGaAs. Experimental values taken from Vurgaftman *et al.*, J. Appl. Phys. **89**, 5815 (2001) and references therein. Due to the large disparity in experimental bowing parameters seen in the above reference, no direct comparison with experiment is made in the paper.

	HSE	PBE	Exp.
$\text{Al}_x\text{Ga}_{1-x}\text{As}$			
b_Γ	$0.168+0.992x$	$0.023+0.825x$	$-0.127+1.310x$
b_X	0.109	0.047	0.055
$\text{Al}_x\text{In}_{1-x}\text{As}$			
b_Γ	0.735	0.601	0.70
b_X	0.194	0.132	0
$\text{Al}_x\text{In}_{1-x}\text{P}$			
b_Γ	0.918	0.763	-0.48
b_X	0.219	0.162	0.38
$\text{Ga}_x\text{In}_{1-x}\text{P}$			
b_Γ	0.826	0.844	0.65
b_X	0.044	0.017	0.20
b_L	0.864	0.600	1.03
$\text{GaAs}_{1-y}\text{P}_y$			
b_Γ	0.298	0.165	0.19
b_X	0.050	-0.038	0.24
b_L	0.180	0.053	0.16

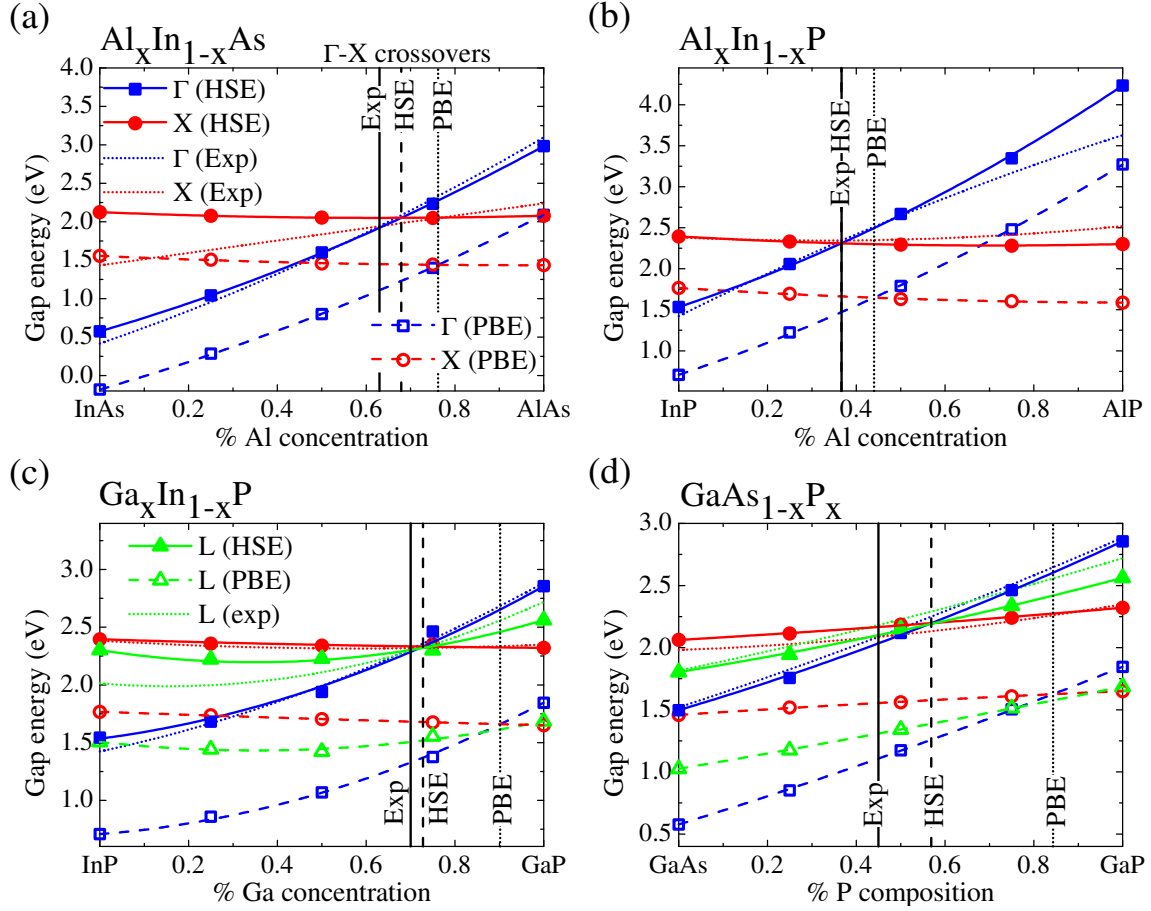


FIG. 1. (Color online) Direct, $\Gamma_c \rightarrow \Gamma_v$ (blue), indirect, $X_c \rightarrow \Gamma_v$ (red), and indirect, $L_c \rightarrow \Gamma_v$ (green), conduction to valence gap energies for the entire alloy range of (a) $\text{Al}_x\text{In}_{1-x}\text{As}$, (b) $\text{Al}_x\text{In}_{1-x}\text{P}$, (c) $\text{Ga}_x\text{In}_{1-x}\text{P}$, and (d) $\text{GaAs}_{1-x}\text{P}_x$. The computed HSE (solid line) and PBE (dashed line) results are shown alongside the experimental fits taken from Vurgaftman *et al.*, J. Appl. Phys. **89**, 5815 (2001) and references therein. Only the Γ -X crossovers are displayed and denoted by vertical lines labeled appropriately.