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A Multi-Nuclear Magnetic Resonance And Density Functional Theory Investigation Of Epitaxially Grown $InGaP_2$

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Table S1: Calculated and experimental lattice constants a in 10^{-10} m. [a]Experimental numbers from: http://www.ioffe.ru/SVA/NSM/Semicond/index.html. [b]Based on pure compounds, assuming linear scaling of a with composition.

	PBE	exp.
GaP	5.51	5.4505[a]
$InGaP_2$	5.73	5.6596[b]
InP	5.96	5.8687[a]

Table S2: Standard deviations of Ga and In isotropic chemical shifts in ppm calculated for the disordered model for four different lattice constants a. The numbers for $a = 5.6596 \times 10^{-10}$ m are reported in the main text in Table 4. The relaxed lattice constant is 5.73×10^{-10} m. See Table S4 for details.

In	25.4	20.3	19.2	16.9
Ga	10.1	8.9	8.6	7.8
a (10^{-10} m)	5.4505	5.6596	5.73	5.8687

Table S3: Calculated isotropic ^{31}P chemical shifts in ppm at the PBE optimized lattice constants (a[PBE]) and the experimental lattice constants (a[exp.]) from Table S1. The ^{31}P shifts are referenced such that $\delta_{\text{iso}} = -142$ ppm for bulk GaP at its experimental lattice constant.

	a[PBE]	a[exp.]
GaP	−113.6	−142.0
InP	−120.6	−160.9

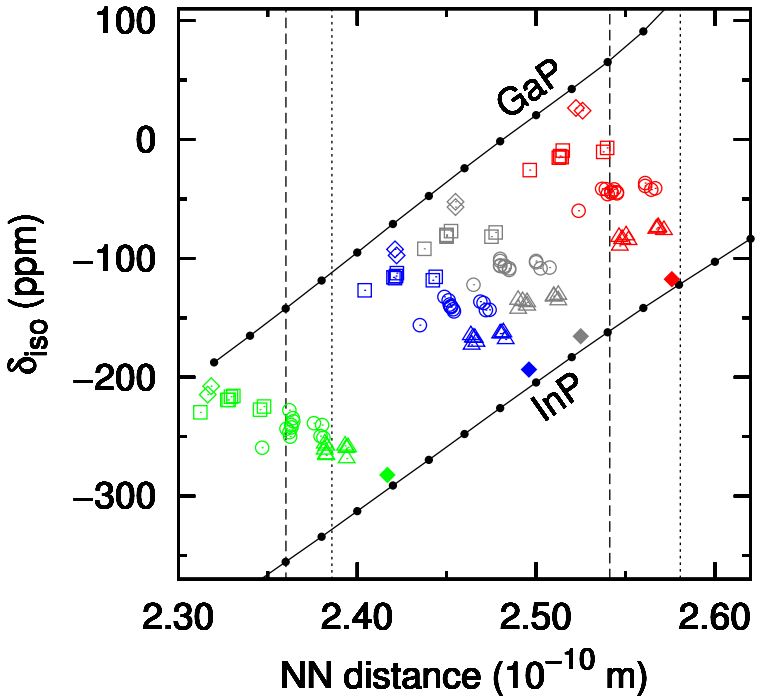

Figure S1: ^{31}P isotropic chemical shifts for bulk GaP and InP (black dots) as a function of the nearest neighbour distance. The lines are guides to the eye. Vertical dashed [dotted] lines indicate the experimental [calculated PBE] bulk GaP (left) and InP (right) lattice constants. Diamonds: ^{31}P isotropic chemical shifts versus average nearest neighbour distance for the 32 P nuclei in the $In_{0.5}Ga_{0.5}P$ disordered model, with $a = 5.4505 \times 10^{-10}$ m (green), $a = 5.6596 \times 10^{-10}$ m (blue, reported in Figure 9 in main text), $a = 5.73 \times 10^{-10}$ m (grey, relaxed lattice constant), and $a = 5.8687 \times 10^{-10}$ m (red). Coordinations P[Ga_nIn_{4-n}], diamonds: $n = 4$, squares: $n = 3$, circles: $n = 2$, triangles: $n = 1$, solid diamonds: $n = 0$. All ^{31}P shifts are referenced such that $\delta_{iso} = -142$ ppm for bulk GaP at its experimental lattice constant. The shifts of the random models are listed in Table S4.

Table S4: Calculated ^{31}P chemical shifts δ_{iso} in ppm for various coordinations P[$\mathrm{Ga}_n\mathrm{In}_{4-n}$] for the random model for three different lattice constants a. Data for all 32 nuclei are listed. The periodically repeated cell has dimensions $2a \times 2a \times 2a$. Atomic positions were relaxed. The cells were kept fixed. The ^{31}P shifts are referenced such that $\delta_{\mathrm{iso}} = -142$ ppm for bulk GaP at its experimental lattice constant.

P[Ga_4]	−214.67	−97.63	−56.85	24.32
	−207.62	−92.43	−52.37	26.55
P[$\mathrm{Ga}_3\mathrm{In}_1$]	−219.10	−114.90	−81.67	−10.52
	−224.75	−112.39	−92.01	−25.80
	−215.79	−116.02	−78.16	−7.21
	−219.09	−118.44	−77.08	−9.44
	−216.36	−116.72	−80.97	−13.98
	−227.29	−126.99	−81.68	−14.80
	−229.47	−115.45	−80.31	−15.24
P[$\mathrm{Ga}_2\mathrm{In}_2$]	−240.30	−144.63	−103.35	−39.22
	−250.87	−141.03	−107.67	−41.14
	−249.65	−137.65	−108.43	−42.30
	−259.23	−135.24	−122.21	−59.93
	−238.61	−132.54	−102.26	−36.64
	−234.43	−143.34	−102.26	−41.67
	−227.72	−136.42	−100.44	−41.61
	−243.37	−142.93	−106.13	−41.89
	−246.29	−143.58	−108.44	−44.55
	−240.18	−156.21	−106.34	−43.84
	−250.01	−140.14	−106.09	−45.90
	−242.16	−138.87	−109.88	−45.43
	−237.14	−140.58	−107.31	−44.79
P[$\mathrm{Ga}_1\mathrm{In}_3$]	−259.04	−172.91	−135.19	−82.29
	−257.79	−170.55	−136.16	−80.86
	−255.89	−166.87	−142.70	−89.48
	−261.12	−162.34	−139.91	−84.98
	−268.48	−163.38	−137.16	−83.98
	−265.75	−168.24	−130.99	−73.95
	−265.02	−163.43	−131.87	−74.39
	−257.82	−164.82	−135.65	−76.50
	−259.87	−167.17	−132.37	−75.24
P[In_4]	−282.15	−193.49	−165.47	−117.60
a (10^{-10} m)	5.4505	5.6596	5.73	5.8687

Table S5: ^{31}P isotropic chemical shifts calculated at the PBE optimized lattice constant (5.73×10^{-10} m) in ppm compared to experiment. ^{31}P is referenced such that its shift is -142 ppm for GaP at the PBE-optimized lattice constant $a = 5.51 \times 10^{-10}$ m.

		experiment		random supercell
	n	W20	W50	
P[Ga_4]	4	-86.6	-87.2	-83
P[Ga_3In_1]	3	-109.2	-110.2	-110
P[Ga_2In_2]	2	-135.3	-136.4	-135
P[Ga_1In_3]	1	-162.2	-164.7	-164
P[In_4]	0	-191.2	-194.7	-194