Supporting Information

Composition and Phase Tuned InGaAs Alloy Nanowires

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Table S1: Length (nm) of twin segments as a function of diameter and composition.

x	NW Diameter (nm)				
	40	**60**	**80**	**100**	**120**
0.3		50	100		
0.4	15	20	25	35	40
0.5	2	2.5	2.5	2.5	
0.6		50		100	

Table S2: Composition and area fraction of two resolved XRD peaks (GaAs- and InAs-like components), and average mole fraction.

x	InAs-like			GaAs-like			x_{avg} [1]
	2θ	x_1	f_1	2θ	x_2	f_2	
0.2	26.8	0.28	0.23	27.0	0.17	0.77	0.17
0.3	26.7	0.34	0.36	26.9	0.23	0.64	0.27
0.4	26.5	0.45	0.48	26.6	0.40	0.52	0.42
0.5	26.0	0.74	0.32	26.5	0.45	0.68	0.54
0.6	26.2	0.62	0.64	26.4	0.51	0.36	0.58
0.8	25.8	0.85	0.82	26.0	0.74	0.18	0.83
0.9	25.7	0.91	0.87	25.9	0.80	0.13	0.89

[1] $x_{avg} = x_1 f_1 + x_2 f_2$

Figure S1: Diameter distribution of the $In_xGa_{1-x}As$ NWs with x = 0 (GaAs), 0.3, 0.4, 0.5, 0.6, and 0.9.

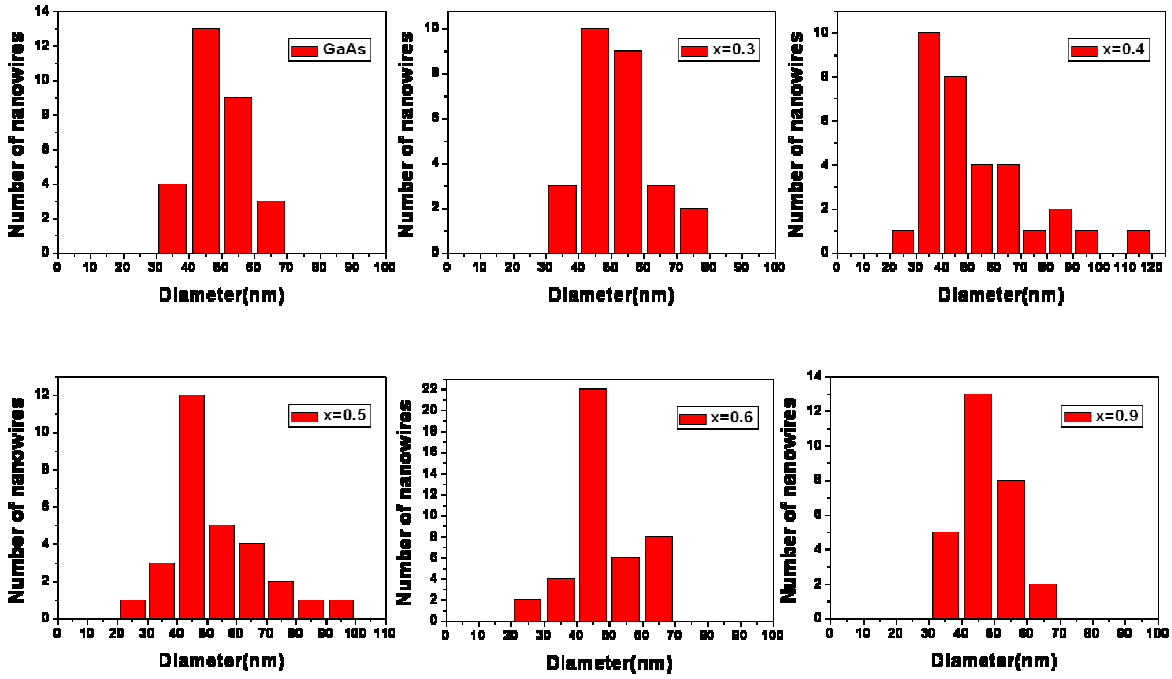

Figure S2: EDX data of the $In_xGa_{1-x}As$ NWs with (a) x = 0.05, (b) 0.1, (c) 0.2, (d) 0.3, (e) 0.4, (f) 0.5, (g) 0.6, (h) 0.7, (i) 0.8, and (j) 0.9. The HAADF STEM images are shown in the insets.

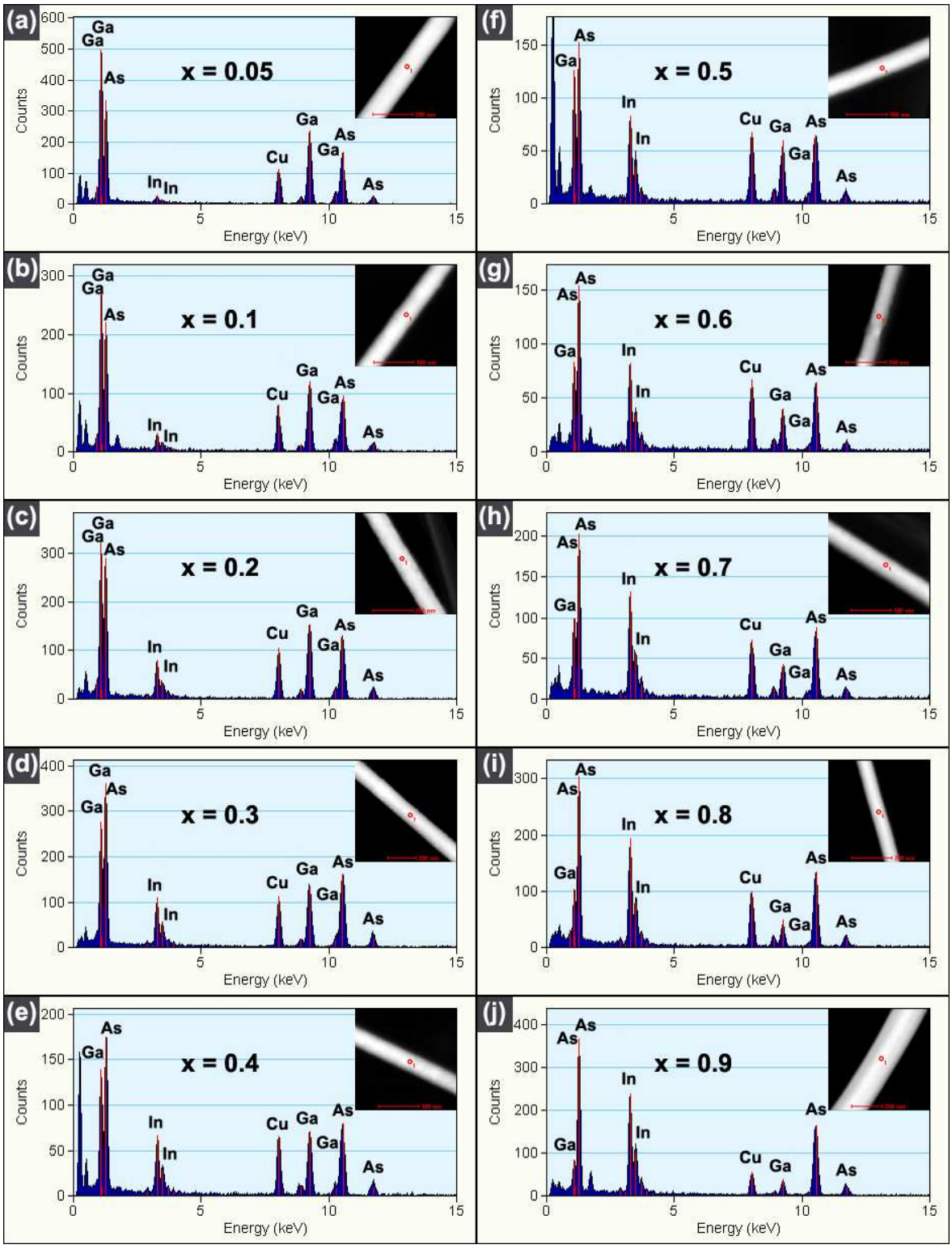

Figure S3: (a) Fine-scanned Ga $2p_{3/2}$ peak of the GaAs and InGaAs alloy NWs.. The peak can thus be resolved into two bands at 1117 (PG1) and 1118 (PG2) eV, using a Voigt function. The binding energy of the Ga atoms bonded to the As atoms would be expected to appear at a lower energy compared to that of the Ga atoms bonded to the more electronegative O atoms. Therefore, the PG1 and PG2 bands can be assigned to the Ga-As and Ga-O bonding structures, respectively. The large fraction (> 70 %) of the PG2 band is due to the GaO_x outerlayers. (b) Finely scanned In $3d_{5/2}$ peak of the InAs and InGaAs alloy NWs. Their peak was resolved into two bands at 444 (PI1) and 445 (PI2) eV, which were assigned to the In-As and In-O bonding structures, respectively. Compared to the Ga $2p_{3/2}$ peak, the fraction of the oxide bonding structures is lower. This indicates that the outerlayers mainly consisted of GaO_x composition, which is consistent with the EDX data.

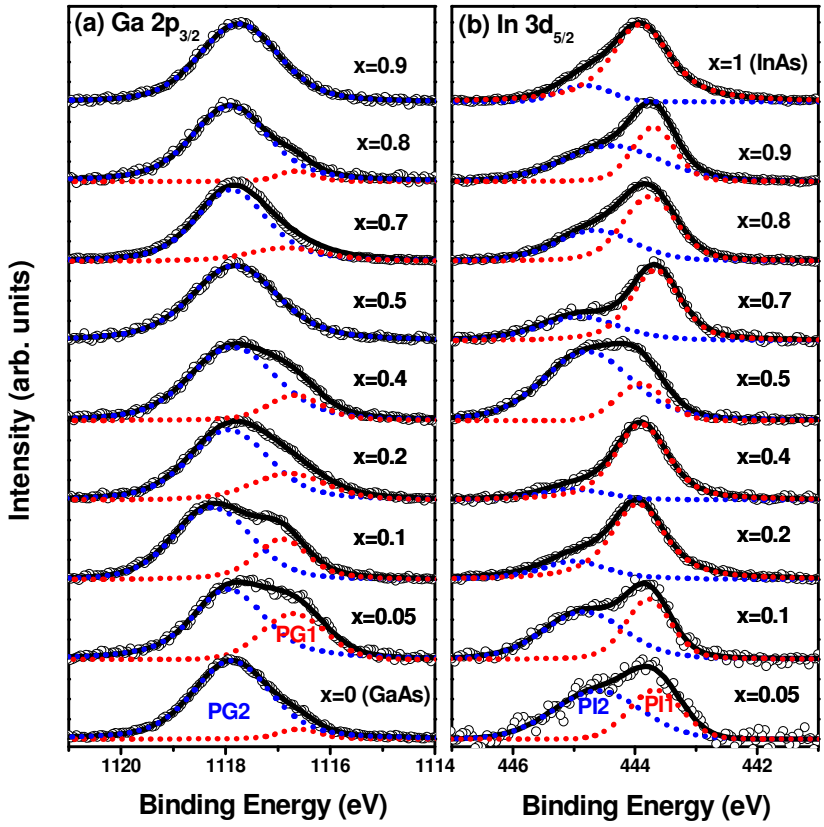

Figure S4. (a) Raman spectra of $In_xGa_{1-x}As$ (x = 0, 0.1, 0.2, 0.3, 0.5, 0.7, 0.9, and 1) NWs. The excitation wavelength is 514.5 nm. The GaAs (x = 0) NWs exhibit TO (transverse optical) and LO (longitudinal optical) phonon modes at 270 and 291 cm^{-1}, respectively. The InAs (x = 1) NWs exhibit TO and LO peaks at 216 and 236 cm^{-1}, respectively. The peak of the InGaAs alloy NWs was resolved into four bands using a Voigt function, *viz.* GaAs-like TO (blue) and LO (cyan) and InAs-like TO (red) and LO(magenta), clearly showing two-mode behavior throughout the composition range x = 0.1-0.8. The black dotted line represents the sum of the resolved bands. The GaAs-like modes is dominant in the range of $0 \leq x \leq 0.5$, whereas the InAs-like mode is dominant in the range of $0.5 \leq x < 1$. At x = 0.5, two modes takes the nearly same fraction. As x approaches 0.5, the FWHM of GaAs-like TO mode increases from 12 (x = 0) to max. 26 cm^{-1}, while the InAs-like TO modes increases from 17 (x = 1) to max. 27 cm^{-1}. The largest broadening around at x = 0.5 indicates constantly the maximized crystalline disorder. (b) The optical phonon frequencies as a function of x. As x increases from 0 to 0.7, the GaAs-like TO and LO modes show a large red shift, 27 and 19 cm^{-1}, respectively. In the case of the InAs-like modes, as x increases from 0.2 to 1, they show 10 and 15 cm^{-1} red shift for LO and TO modes, respectively. The two-mode behavior of Raman scattering is quite consistent with previous works on the bulk film.[1-3] In these previous works, similar to our result, as x increases, the optical phonon frequencies of both the GaAs- and InAs-like phases decrease almost linearly, with a more significant change for the GaAs-like phase. For comparison, we plotted the work of Groenen *et al*. in the figure.[2]

References:

(1) Estrera, J. P.; Stevens, P. D., Glosser, R.; Duncan, W. M.; Kao, Y. C.; Liu, H. Y.; Beam III, E. A. *Appl. Phys. Lett*. **1992**, *61*, 1927-1929.

(2) Groenen, J.; Carles, R.; Landa, G.; Guerret-Piécourt, C.; Fontaine, C.; Gendry, M. *Phys. Rev. B*. **1998**, *58*, 10452-10462.

(3) Pagès, O.; Postnikov, A. V.; Kassem, M.; Chafi, A.; Nassour, A.; Doyen, S. *Phys. Rev. B* **2008**, *77*, 125208 1-9.