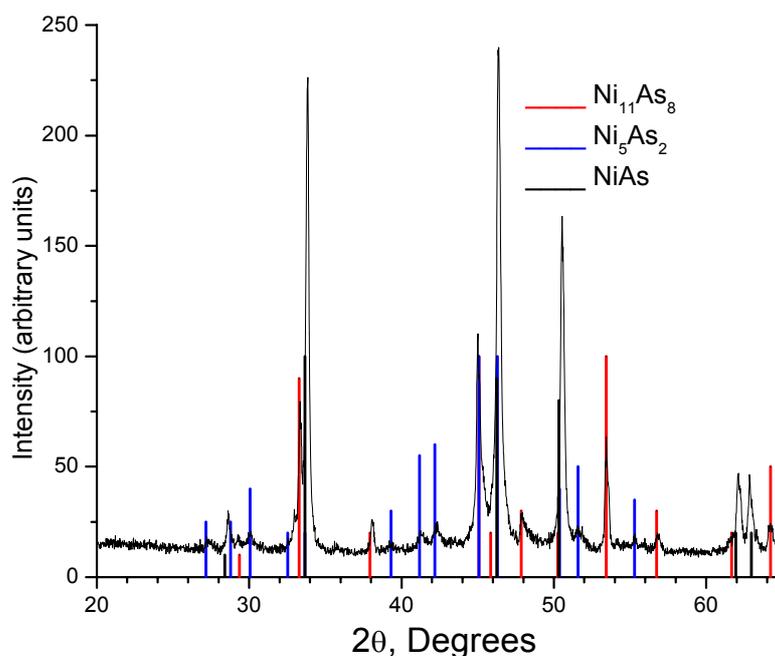


SUPPORTING INFORMATION

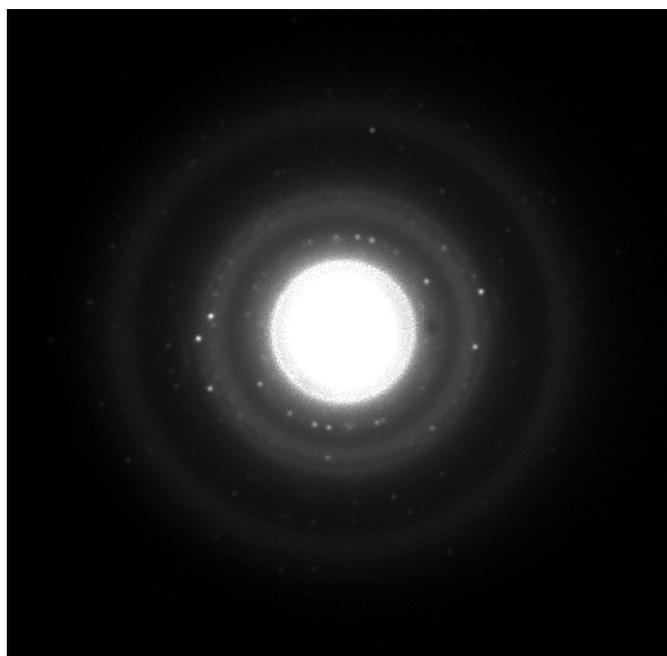
## From Ribbons to Nanodot Arrays: Nanopattern Design Through Reductive Annealing

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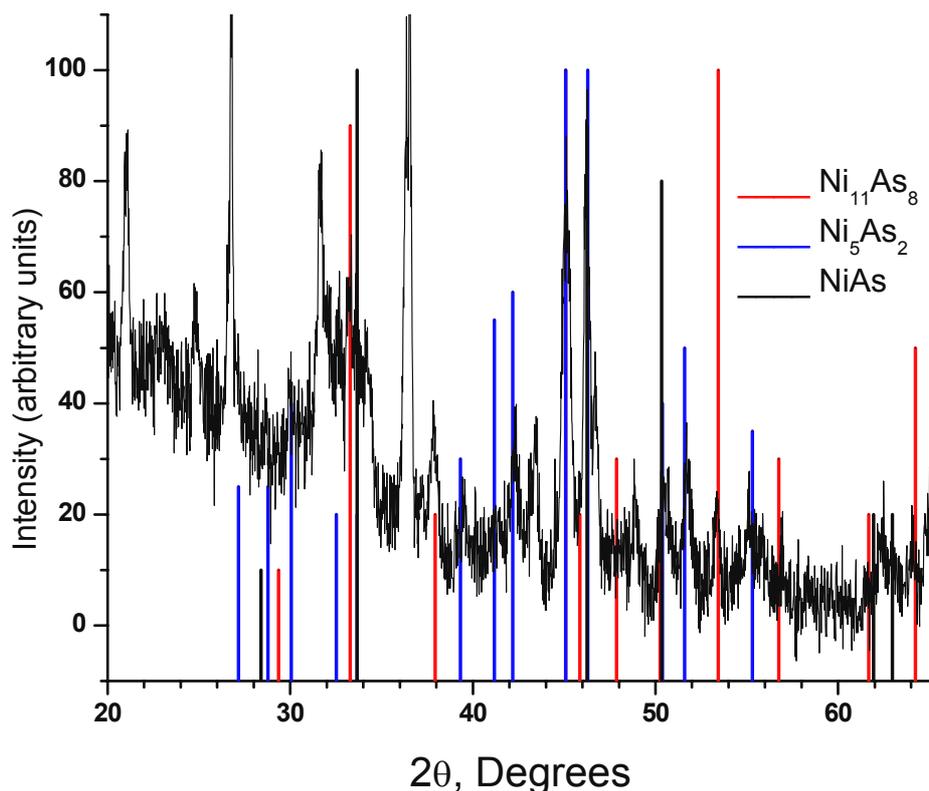
**Figure S1.** The X-ray diffraction pattern of the product from hydrogen annealing of  $\text{Ni}_3(\text{AsO}_4)_2 \cdot \text{H}_2\text{O}$  precursor conducted at 425 °C for an hour using an unsupported media (alumina boat). The solid black line indicates the NiAs phase, the red line indicates the  $\text{Ni}_{11}\text{As}_8$  phase, and the blue line indicates the  $\text{Ni}_5\text{As}_2$  phase.



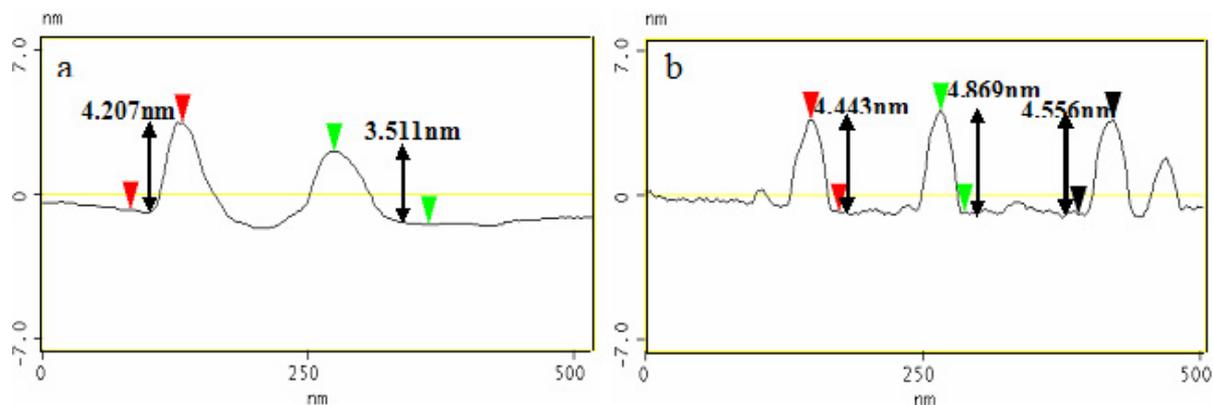
**Figure S2.** Selected area diffraction pattern of the product of *in situ* carbothermal reduction of the  $\text{Ni}_3(\text{AsO}_4)_2 \cdot \text{H}_2\text{O}$  precursor conducted at 420 °C.

**Table S1.** Measured d-spacing values from the electron diffraction pattern (Figure S2), as well as d-spacing values for the metal-rich nickel arsenide phases and their respective (hkl) values

| Measured<br>d-spacing<br>(Å) | $\text{Ni}_{11}\text{As}_8$ |          | $\text{Ni}_5\text{As}_2$ |          | $\text{NiAs}$    |         |
|------------------------------|-----------------------------|----------|--------------------------|----------|------------------|---------|
|                              | d-spacing<br>(Å)            | (h k l)  | d-spacing<br>(Å)         | (h k l)  | d-spacing<br>(Å) | (h k l) |
| 2.189                        | 2.370                       | (1 1 8)  | 2.190                    | (2 1 1)  | 2.520            | (0 0 2) |
| 1.922                        | 1.899                       | (1 0 11) | 1.960                    | (1 0 6)  | 1.961            | (1 0 2) |
| 1.602                        | 1.621                       | (3 2 7)  | 1.660                    | (2 1 5)  | 1.568            | (2 0 0) |
| 1.441                        | 1.449                       | (4 0 8)  | 1.403                    | (2 2 5)  | 1.471            | (1 1 2) |
| 1.017                        | 1.023                       | (6 2 7)  | 1.054                    | (3 0 10) | 1.033            | (1 1 4) |



**Figure S3.** The X-ray diffraction pattern of the product from carbothermal reduction of the  $\text{Ni}_3(\text{AsO}_4)_2 \cdot \text{H}_2\text{O}$  precursor conducted at 550 °C for three hours in a sealed fused silica ampoule. The precursor arsenate was mixed with charcoal and ground before carrying out the reduction. The solid black line indicates the NiAs phase, the red line indicates the  $\text{Ni}_{11}\text{As}_8$  phase, and the blue line indicates the  $\text{Ni}_5\text{As}_2$  phase. The powder pattern shows several other peaks that have yet to be identified, but may correspond to products of incomplete reduction. Attempts to perform the reduction at 420 °C, as in the TEM, did not result in arsenide product formation. The *in situ* reduction in the TEM may be aided by localized electron beam heating, carbon sputtering, or the higher surface area present in the dispersed sample.



**Figure S4.** Sectional analysis of AFM images presented in Figure 5: a) height profile measured along the line in Fig 5a; the average height is ca. 3.9 nm, b) height profile measured along the line in Fig 5b; the average height is found to be ca. 4.6 nm. Since the measured lateral dimensions are a consequence of the tip shape, heights are used as a more reliable measure of particle size.

**Notes on particle coalescence:** The melting point of bulk NiAs is 967 °C (CRC, 76<sup>th</sup> edition), far greater than the temperature at which we observe coalescence. However, it is well demonstrated that nanoparticles can undergo melting at far lower temperatures than bulk phases (A. N. Goldstein, C. M. Echer, A. P. Alivisatos, *Science*, 1992, **256**, 1425-1427), which may explain the behavior observed here. On the other hand, island movement is a well-demonstrated phenomenon often attributed to the mobility of the underlying phase (W. B. Phillips, E. A. Desloge, J. G. Skofronick, *J. Appl. Phys.* 1968, **39**, 3210-3218; M. S. Kunz, K. R. Shull, A. J. Kellock, *J. Appl. Phys.* 1992, **72**, 4458-4460). Given the high temperatures employed and the likelihood of carbon sputtering, this is also a reasonable explanation for the observed coalescence.