Supporting Information:

Procedure for constrained synthesis of bimetallic nanoarrays: Two-dimensional crystals of TF55β chaperonins were prepared at a protein concentration of 1 mg/mL as described elsewhere.[3] To bind palladium ions to the engineered protein crystals, 10 microliters of 20 mM palladium(II) acetate, pH 7.5 was added to 100 microliters of a preparation of 2D crystals. After incubation at room temperature for one hour, unwanted Pd was rinsed from the crystallization suspension by microdialysis against 25 mM MES buffer, pH 6.0 overnight followed by 2 hrs against DDI-H_2O. The Pd-charged crystals were applied to either carbon stabilized formvar coated TEM grids, micromachined carbon ("Quantifoil") or SiN "window" substrates and floated on 100 microliters of a mixture of either 1mM $CoSO_4$ or 1 mM $NiSO_4$ and 1g/L dimethylamine borane complex (DMAB) for 1 minute. Samples were washed in 100 microliters of DDI-H_2O and dried under vacuum for analysis.

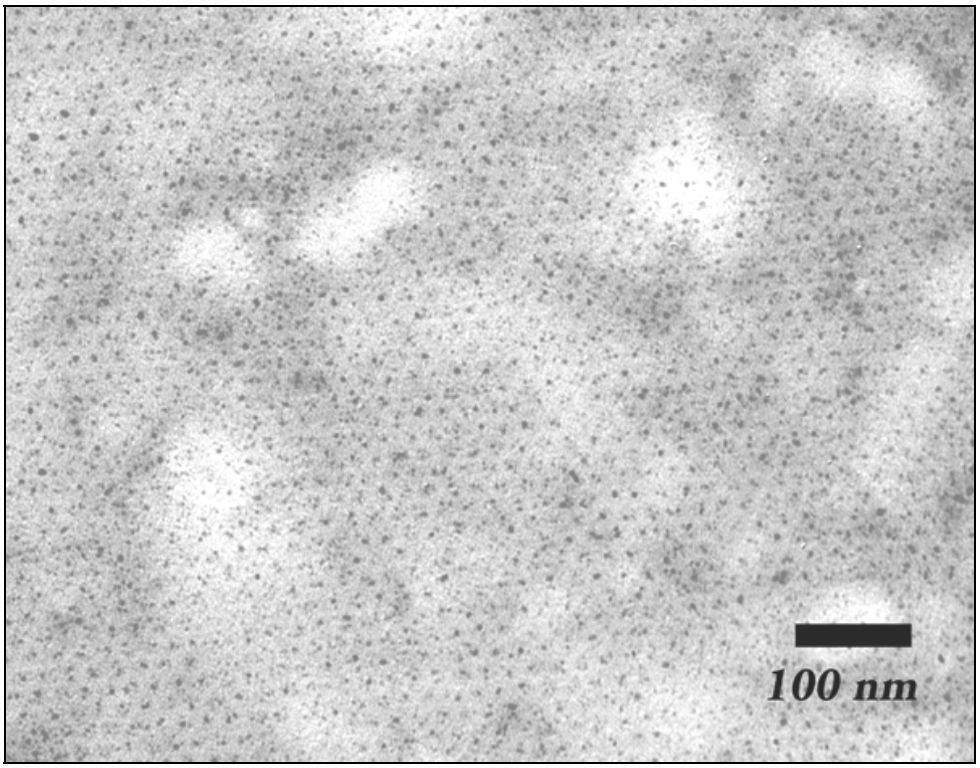

Figure S1. Unstained, conventional TEM image (60 kV) of a Pd-charged template after rinsing by dialysis. The periodic pattern of dark, electron dense regions suggests palladium acetate selectively accumulates on the hexagonally packed chaperonins within the 2D crystal. Control experiments using protein crystals of TF55β with the apical loop deletion and lacking the poly-histidine modification did not exhibit any apparent periodicity in electron density when incubated with Pd and washed (not shown).

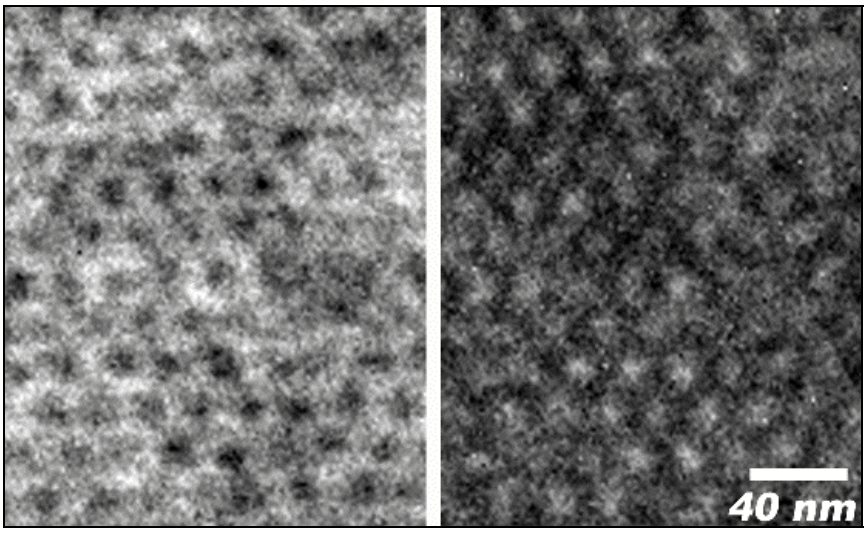

Figure S2. Elemental distribution (EF-TEM EELS map) of Ni-Pd array prior to annealing as in ***Figure 2*** (**C**). Data was collected from an area suspended over a hole on the micromachined Quantifoil substrate to enable accurate mapping of the distribution of carbon and nickel across the template. The left image was generated by filtering the loss signal (K edge) and maps the carbon distribution (lighter areas). The darker regions represent areas on the template devoid of carbon, suggesting possible areas of monolayer thickness on the chaperonin crystal. Similarly, the image on the right is a Ni map and reveals a hexagonal distribution of Ni that accumulates on the template.

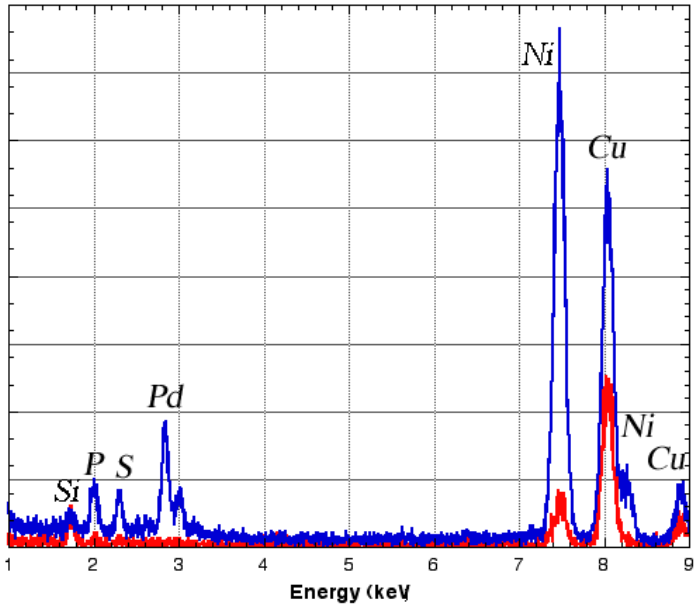

Figure S3. XEDS spectra collected from sample from ***Figure S2*** comparing Ni-Pd array suspended over the holes in the support film (blue trace) to a background area on the carbon support film (red trace).

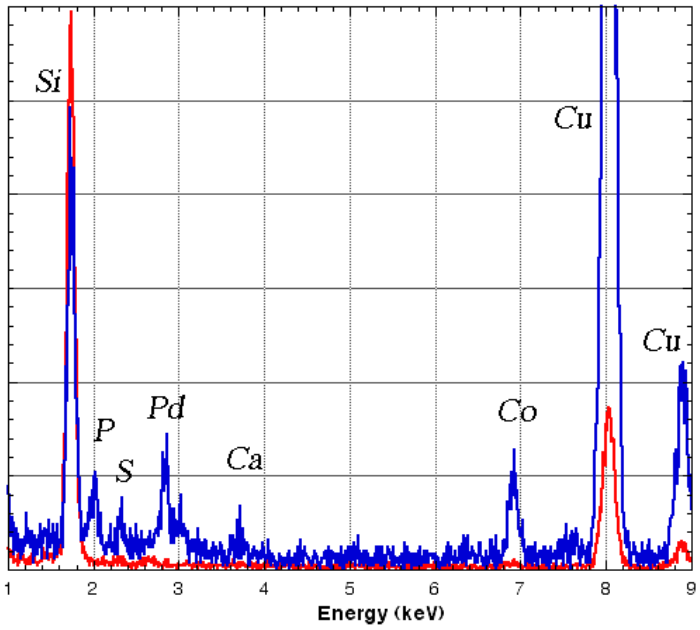

Figure S4. XEDS spectra similar to ***Figure S3*** collected from a Co-Pd array sample prepared on a SiN substrate confirming the presence of Co and Pd in the arrays (blue trace) verses the background SiN substrate (red trace).

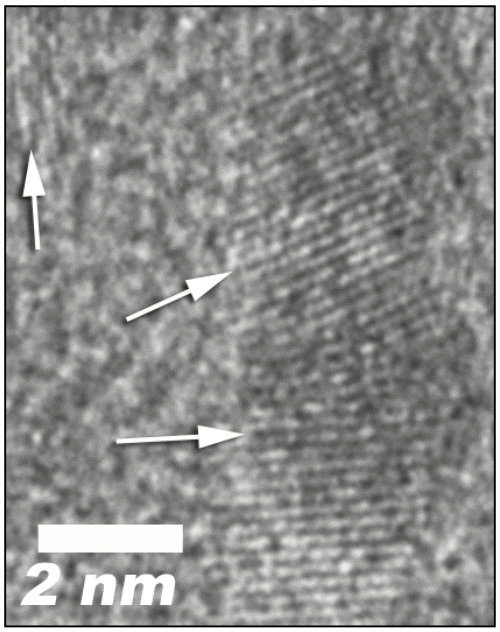

Figure S5. HR-TEM lattice image of three coalesced Co-Pd nanoparticles (arrows) similar to the Ni-Pd sample in ***Figure 2F*** from the text, but synthesized by substituting $CoSO_4$ for $NiSO_4$. The measured lattice spacing of 2.2 Å is consistent with the {111} *d*-spacing for fcc Co (2.15 Å, JCPDS, #15-806).