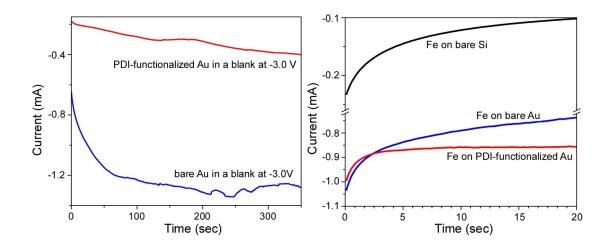
## **Supporting Information**

Nanoscale Architecture of Bimetallic Hybrid Fe-Au Nanostructures with and without 1,4-Phenylene Diisocyanide Pre-Functionalization

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**Figure S1**: I-t curves of bare and PDI-functionalized Au NITs in a blank deionized water at a potential of -3.0 V (Figure 1, Samples A1 and A2, respectively), and Fe electrodeposition on bare Si, bare Au NIT, and PDI-functionalized Au NIT.

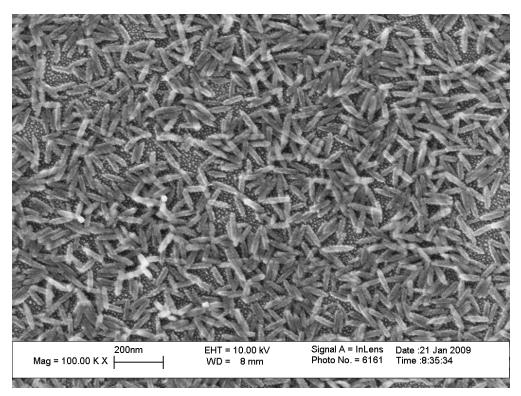


Figure S2a: SEM image of Fe NrPs electrodeposited on PDI-functionalized Au NIs.

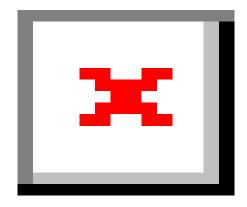


Figure S2b: Back-scattered electron image of Fe NrPs electrodeposited on bare Au NIs.

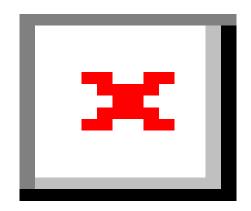


Figure S3a: SEM image of Fe NrPs electrodeposited on a bare Si substrate.

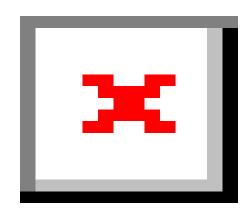


Figure S3b: SEM image of Fe NrPs electrodeposited on a bare Au NI template.

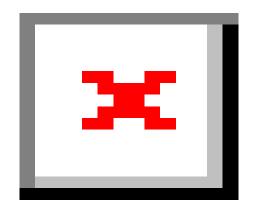


Figure S3c: SEM image of Fe NrP electrodeposited on PDI-functionalized Au NIs.

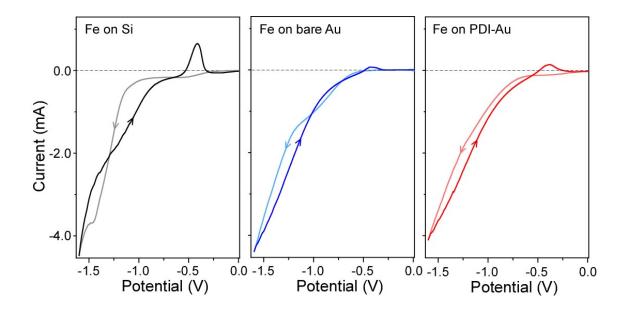
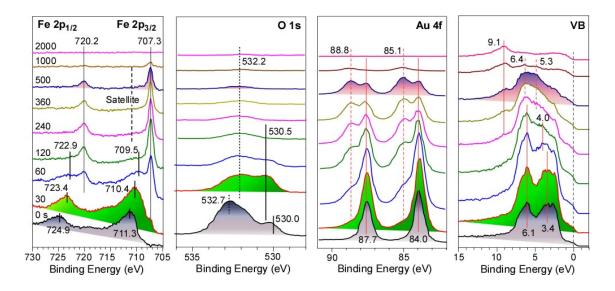


Figure S3d: Cyclic voltammograms for samples shown in Figure S3a, S3b, and S3c.



**Figure S4**: XPS spectra of Fe 2p, O 1s, Au 4f and valence band (VB) regions of Fe electrodeposited on large Au NIs, shown in Figure 2, Sample F1.

 Table S1:
 Review of Au 4f XPS of Fe-Au hybrid materials.

Preparation method	System	Au Species	Ref.
		(4f <sub>7/2</sub> Binding Energy, eV)	
Co-precipitation,	Au/ FeO(OH),	Au <sup>0</sup> (84.3)	
1wt% Au	Fe <sub>2</sub> O <sub>3</sub>	Au <sup>0</sup> , electron-rich state (83.8)	51
	Au/ Fe <sub>3</sub> O <sub>4</sub> , FeO		
Deposition-	Au/ Fe <sub>x</sub> O <sub>y</sub>	Au⁰ (≤ 84.1)	52
precipitation on	anneal(150-300°C)		
Fe <sub>x</sub> O <sub>y</sub> , 3wt%			
Impregnation of	Au/Fe(OH) <sub>3</sub> , and	Au <sup>I</sup> 84.9: strongly bound	
Au(PPh <sub>3</sub> )(NO <sub>3</sub> ) on	Au/ Fe <sub>2</sub> O <sub>3</sub>	[Au(PPh₃)]⁺	53
Fe oxides		Au <sup>0</sup> 84.0: upon annealing	
Au reduction on	Fe <sub>3</sub> O₄@Au core-	Au <sup>0</sup> (84.2)	
Fe <sub>3</sub> O <sub>4</sub>	shell		54
5nm core/1nm shell			
Reduction of Au into			
Porous alpha-Fe <sub>2</sub> O <sub>3</sub>	Au into porous	Au <sup>0</sup> (83.2) negative state	55
Nanorods	alpha-Fe <sub>2</sub> O <sub>3</sub>		
0.5 wt%			
Co-precipitation and	Au/Fe <sub>2</sub> O <sub>3</sub>	Au <sup>0</sup> (84.0):70-85%	56
deposition-		AuO <sub>x</sub> (84.8)30-15%	
precipitation			
Co-precipitation,	Au/Fe <sub>2</sub> O <sub>3</sub>	As-prepared: mixed Au <sup>0</sup>	
5wt% Au		(84.3) and AuO <sub>x</sub> (85.1)	57
		Upon 673K calcination:	
		mainly Au <sup>0</sup> (84.3)	
Sputter deposition of	Au/ Fe <sub>x</sub> O <sub>y</sub>	Au <sup>0</sup> (84.1)	This work
Au on Fe <sub>x</sub> O <sub>y</sub>		Au-O (84.5) at the interface	
Electrodeposition of	Au/ Fe <sub>x</sub> O <sub>y</sub>	Au <sup>0</sup> (84.1)	This work
Au on Fe <sub>x</sub> O <sub>y</sub>		Au-O (84.5) at the interface	
Electrodeposition of	Fe <sub>x</sub> O <sub>y</sub> /Au	Au <sup>0</sup> (84.1)	This work
Fe oxides on Au/Si			