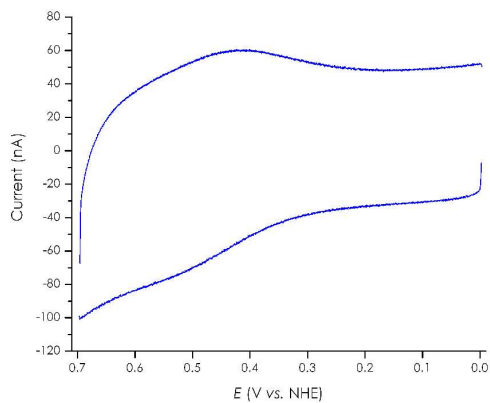
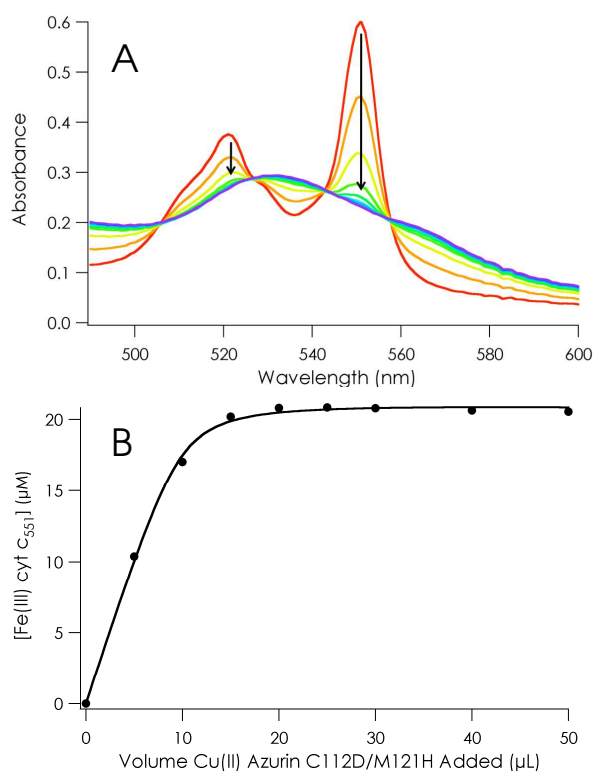


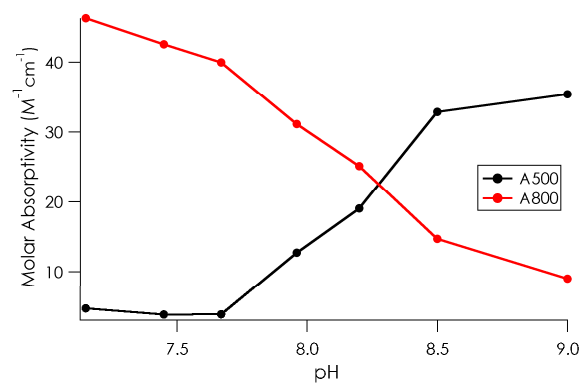
**Figure S1:** CV of azurin C112D/M121E on SAM-modified Au electrode in 10 mM sodium phosphate pH 7.0. 50 mV/s, referenced to Ag/AgCl (197 mV vs NHE). Apparent  $E^{\circ}_{1/2} = \sim 270$  mV, however the signal is broad and weak. SWV produced stronger signals, and as such was reported in the body of the text.



**Figure S2:** CV of azurin C112D/M121H on SAM-modified Au electrode in 10 mM sodium phosphate pH 7.0. 50 mV/s, referenced to Ag/AgCl (197 mV vs NHE). Apparent  $E^{\circ}_{1/2} = \sim 450$  mV, however the signal is broad and asymmetric, casting suspicion on this value.



**Figure S3:** Redox titration of 20  $\mu\text{M}$  Fe(II) *P. aeruginosa* cytochrome  $c_{551}$  ( $E^{\circ}_{1/2} = 255$  mV) with 5.2 mM Cu(II) azurin C112D/M121H in 100 mM sodium phosphate pH 7.0. Decrease in intensity and resolution of the heme Q-bands are commensurate with oxidation of Fe(II) to Fe(III) (A). Fe(III) concentration was fit to a volume-dependent expression for the Nernstian equilibrium constant (B). Extrema of fitting multiple trials yielded a reduction potential of  $305 \pm 10$  mV.



**Figure S4:** Optical pH titration of Cu(II) azurin C112D/M121H. The LF absorption maximum exhibits a blue shift with increasing pH, likely owing to a reorganization of coordination due to deprotonation of the H121 imidazole side chain.