

<u>Figure 1S</u>. Nitrogen ENDOR is shown for Fe(OEP)(NO) (**A**) and ¹⁴NO-Cyt c' (**B**) at amagnetic field of 1.178 T. Adiabatic rapid passage; T = 2 K; microwave power = 0.22 nW, 100 kHz field modulation = 2 G ptp (**A**) and 5 G (**B**), a system time constant = 160 ms, radio frequency power ~ 20 W, radio frequency sweep rate = 2 MHz/s, overall signal averaging time = 500 s, v_{EPR} = 34.10 GHz. RF was pulsed with a 100/900 µs duty cycle.



Figure 2S. Nitrogen ENDOR is shown for 6-coordinated ¹⁴NO-Cyt *c'* (**A**) and ¹⁴NO-Mb (**B**) at magnetic field of 1.227 T (g = 1.985). Adiabatic rapid passage; T = 2 K; microwave power = 0.22 nW, 100 kHz field modulation = 5 G p.t.p., a system time constant = 160 ms, radio frequency power ~ 20 W, radio frequency sweep rate = 2 MHz/s, overall signal averaging time = 500 s, v_{EPR} = 34.10 GHz. RF was pulsed with a 100/900 µs duty cycle.