Table S1. Ni EXAFS Fitting Results for Model Compounds and for the

ACDS β Subunit Using Integer Coordination Numbers

Compound	C.N.	R (Å)	σ^2 (10^{-3} Å^2)	F
[Fe{N($CH_2CH_2S)_3$}(NO)-NiCl(dppe)]	4S	2.192(2)	6.47	522.4
	1Cl	2.484(10)	4.53	
	4S	2.183(2)	6.26	391.1
	1Cl	2.486(9)	4.34	
	1Fe	3.295(7)	3.04	
$Ni(tfd)_2$	4S	2.132(2)	2.55	776.9
	4C	3.086(13)	5.30	
ACDS β subunit As-isolated	4S	2.187(4)	6.35	558.2
	3S	2.187(3)	4.50	483.1
	3S	2.182(3)	4.23	425.2
	1N	1.887(7)	2.35	
	2S	2.190(2)	2.39	398.2
	2N	1.929(11)	6.55	
	3S	2.182(2)	4.27	
	1N	1.886(7)	2.35	374.9
	1Fe	2.963(7)	6.26	
	2S	2.191(3)	2.41	
	2N	1.932(10)	6.67	345.3
	1Fe	2.969(8)	6.62	

ACDS β subunit Ti^{3+}-reduced	3S	2.174(3)	3.73	618.2
	1N	1.846(7)	1.68	
	2S	2.176(3)	1.85	626.0
	2N	1.879(13)	7.50	
	3S	2.176(3)	3.71	607.2
	1N	1.847(7)	1.76	
	1Fe	2.982(10)	6.01	

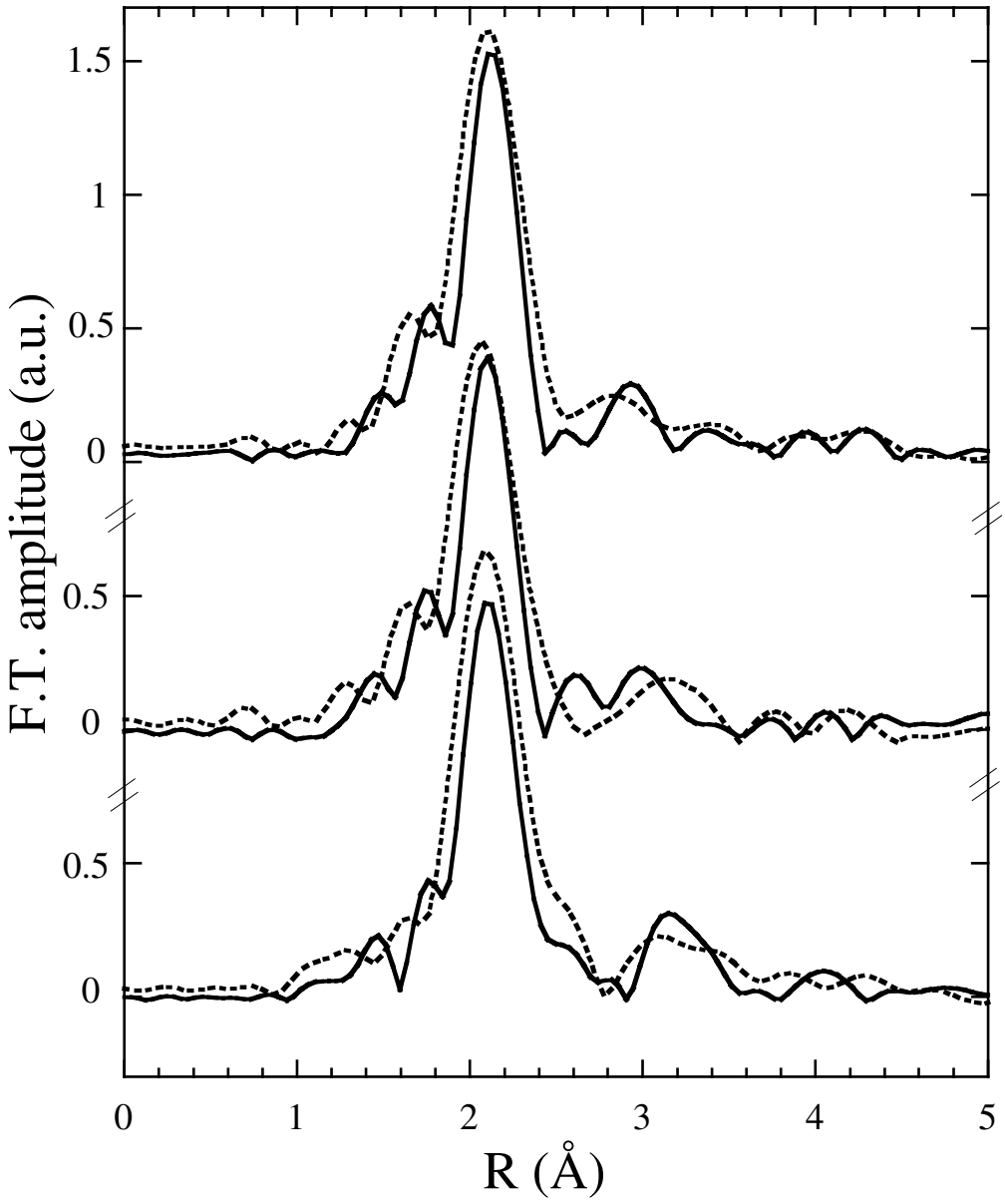

Figure S1. Long and short range Ni EXAFS Fourier transforms show persistence of the ~3.0 Å feature, most likely due to a Ni-Ni interaction. Upper, Ni FT EXAFS of as-isolated ACDS β subunit. Middle, Ti^{3+}-reduced β subunit. Lower, Ni FT EXAFS of the Ni-Fe compound [Fe{$N(CH_2CH_2S)_3$}(NO)NiCl(dppe)]. FT range is k = 1 to 12.5 $Å^{-1}$ (—) and k = 1 – 16.0 $Å^{-1}$ (----). The ~3.0 Å peak persists regardless of the k range over which the transform was

performed. This feature also shifts to a longer average distance upon reduction with Ti^{3+} in both cases, suggesting an increased Ni-Ni distance in the reduced enzyme.