Table S1. Summary of crystallographic statistics

Data Sets	*Native 1*
Space group	$P2_1$
Unit cell parameters (Å)	
a	39.55
b	37.98
c	43.76
β	102.68
Temperature	100 K
Resolution limits (Å)	19.3 - 1.35
Last shell limits (Å)	1.37 - 1.35
Completeness (%) [1]	94.8 (89.3)
Reflections	51168
Unique reflections	24978
I/s	11.7 (2.1)
R_{sym} (%) [2]	5.0 (45.5)

Final refinement statistics	*Native model*
Resolution limits (Å)	19.3 - 1.35
Refined number of reflections	24978
Number of reflections for R_{free}	1567
Overall R_{cryst} (%) [3]	18.5
Overall R_{free} (%) [4]	21.4

Number of protein residues	**139**
Mean bond distance (RMS deviation)	1.427 (0.018)
Mean angle distance (RMS deviation)	2.427 (0.030)
Average B-factor (Å^2)	15.34
Total refined atoms	1567
Protein Atoms	1132
Solvent Atoms	109
Fe^{+3} Ions	1

[1] Number in parentheses indicate values for the highest resolution shell.
[2] $R_{sym} = \Sigma[|I_i - \langle I_i \rangle|] / \Sigma[\langle I_i \rangle]$
[3] $R_{cryst} = \Sigma||F_{obs}| - |F_{calc}||/\Sigma|F_{obs}|$
[4] R_{free} is the same as R_{cryst} for a random subset not included in the refinement of 6 % of total reflection.

Table S2. Redox mediators and their midpoint potentials

Mediator	**E_m (*vs.* NHE) mV**	**E_m (*vs.* Ag/AgCl) mV**
$Fe(NOTA)^{0/-}$	195	-1
$[Fe(tacn)_2]^{3+/2+}$	146	-50
$[Co((NMe_3)_2sar)]^{5+/4+}$	10	-186
$[Co(CLME\text{-}N_4S_5\text{-}sar)]^{3+/2+}$	-136	-332
$[Co(AMME\text{-}N_5S\text{-}sar)]^{3+/2+}$	-220	-416
$[Co(sep)]^{3+/2+}$	-296	-500
$[Co(AMMEsar)]^{3+/2+}$	-380	-576
$[Co(cis\text{-}diammac)]^{3+/2+}$	-503	-699

Fig. S1. Absorption spectra of purified Cgb. *A.* The absorption spectra for CN-bound crystallized Cgb (blue), CN-bound Cgb in solution (black), and oxyferrous Cgb in solution (red). *B.* The absorption spectra for as-purified $G5_{YF}$ (blue) and $H23_{EA}$ (black) Cgb.

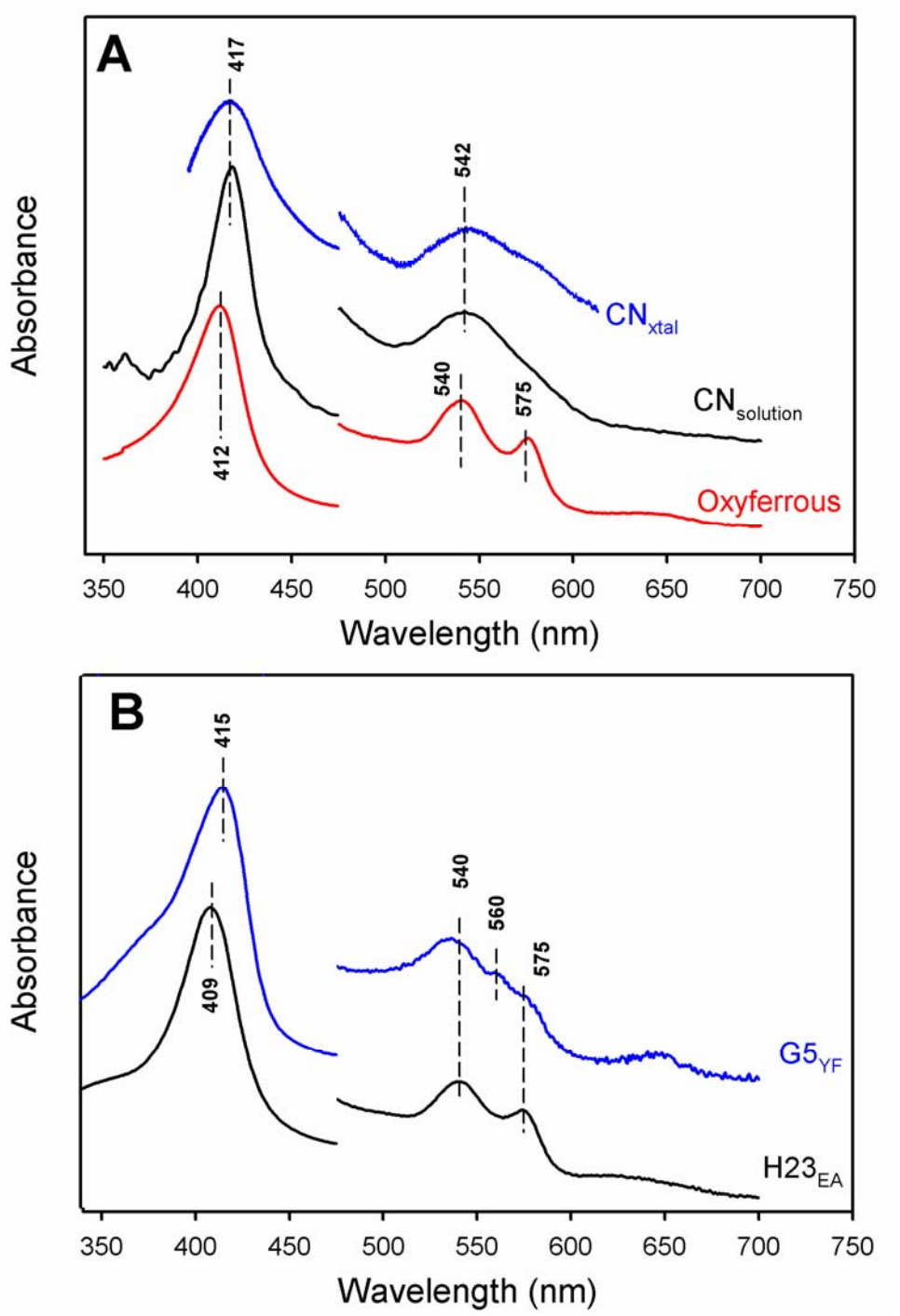

Fig. S2. Multiple sequence alignment of bacterial globins and myoglobin. Cgb shares 42%, 33%, and 10%, sequence identity with Vgb, the globin domain of Hmp, and swMb, respectively. The B10, CD1, E7, G5, and H23 labels correspond to residues of the bacterial globins, but do not align with these residues in sperm whale myoglobin (swMb).

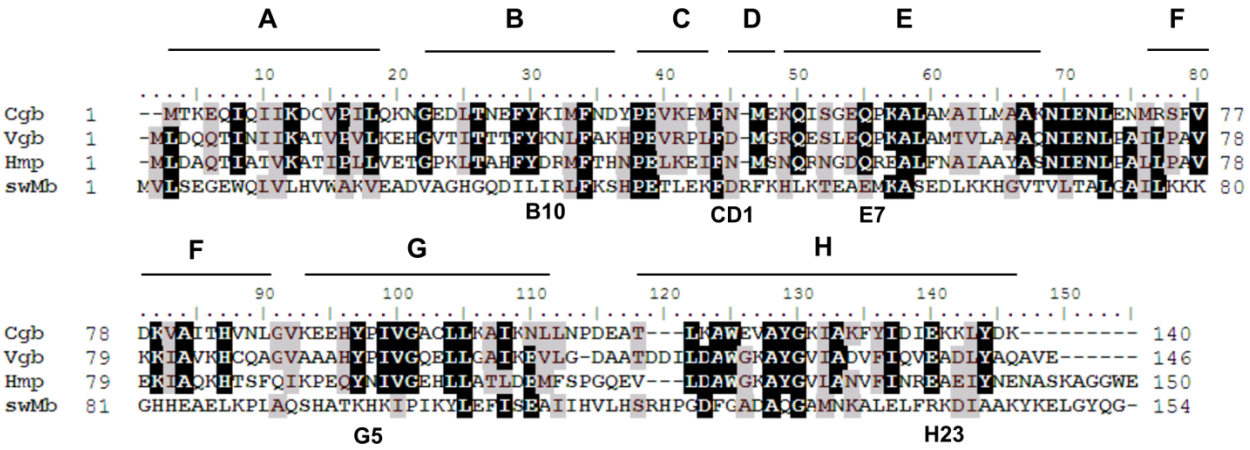

Fig. S3. 2Fo-Fc electron density map for residues 43-46 of the D-helix of Cgb. The map is contoured at 1.70 σ.