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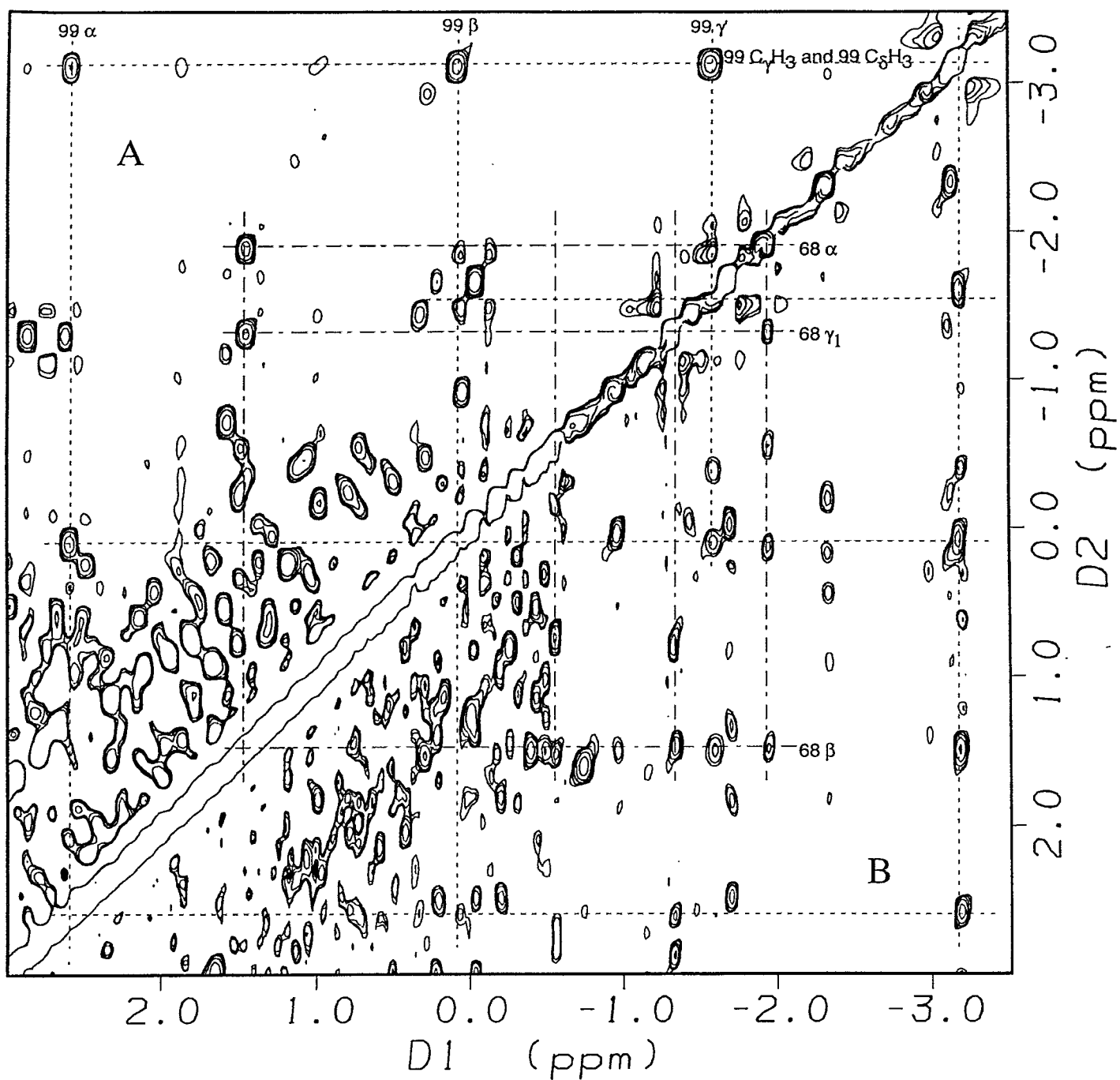


Figure 1S. The up-field portions of the 500 MHz ¹H NMR (A) TOCSY and (B) NOESY spectra of S92A-metMbCN in H₂O, pH 7, at 25°C.

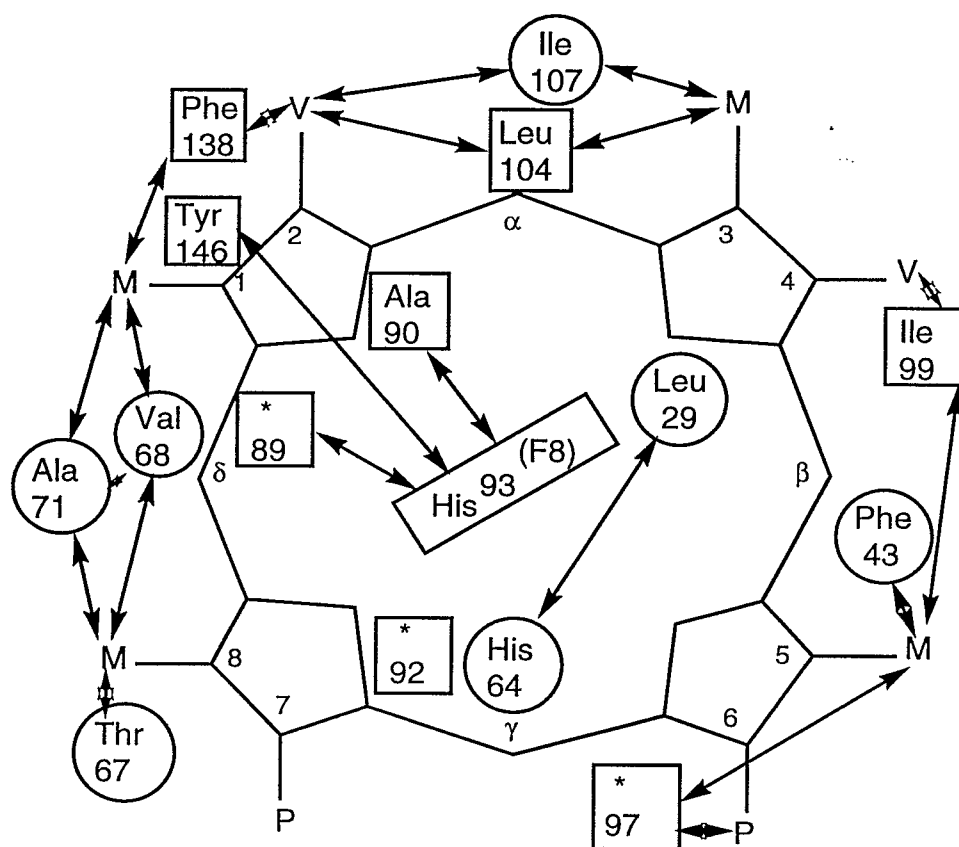


Figure 2S. Schematic representation of heme cavity dipolar connections of S92A-metMBCN

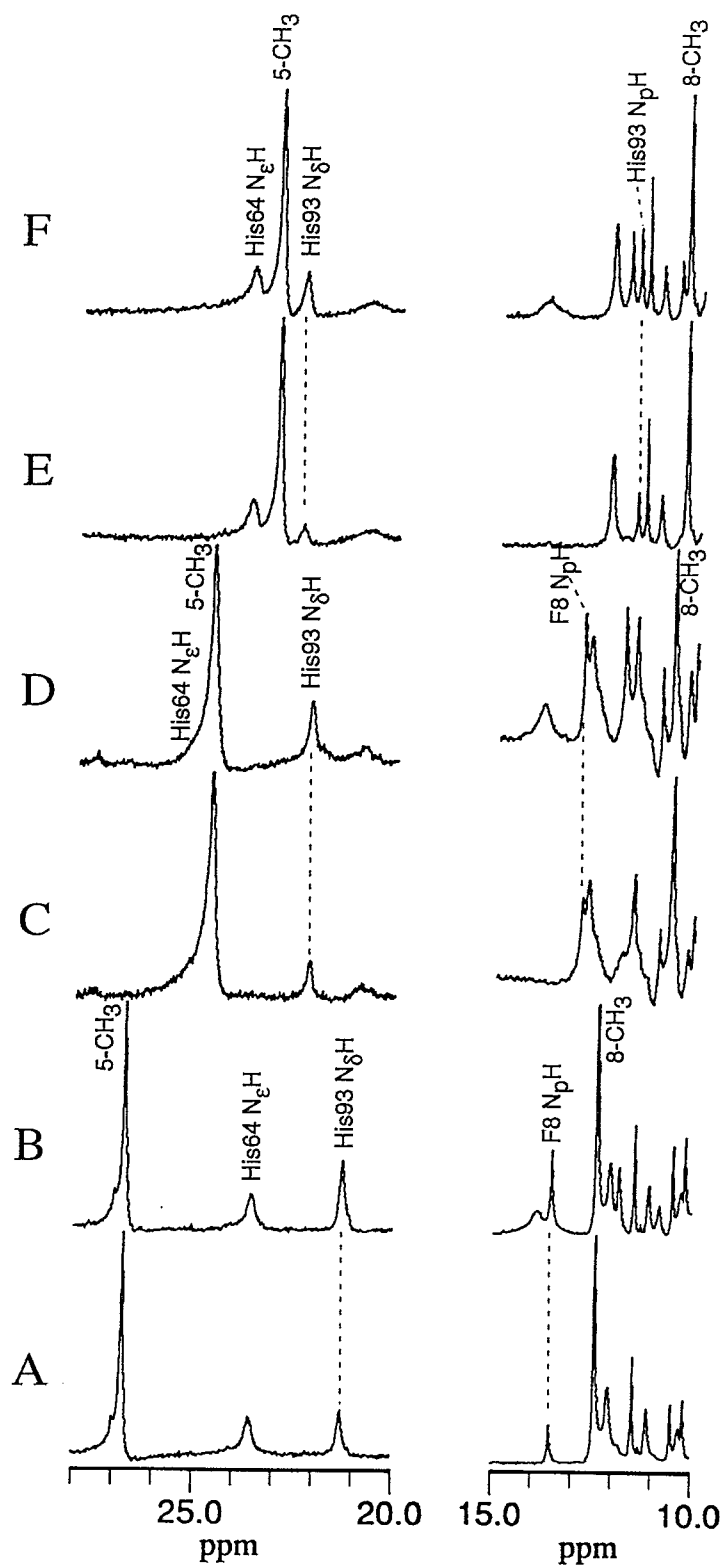


Figure 3S. Portions of the 500 MHz 1:1 ^1H NMR spectra from sperm whale WT metMbCN (A, B); S92A-metMbCN (C, D), and S92P-metMbCN (E, F), in $^1\text{H}_2\text{O}$, pH 10.2 at 25°C (left column) and 35°C (right column), without (A, C, E) and with (B, D, F) saturation of the bulk solvent signal.

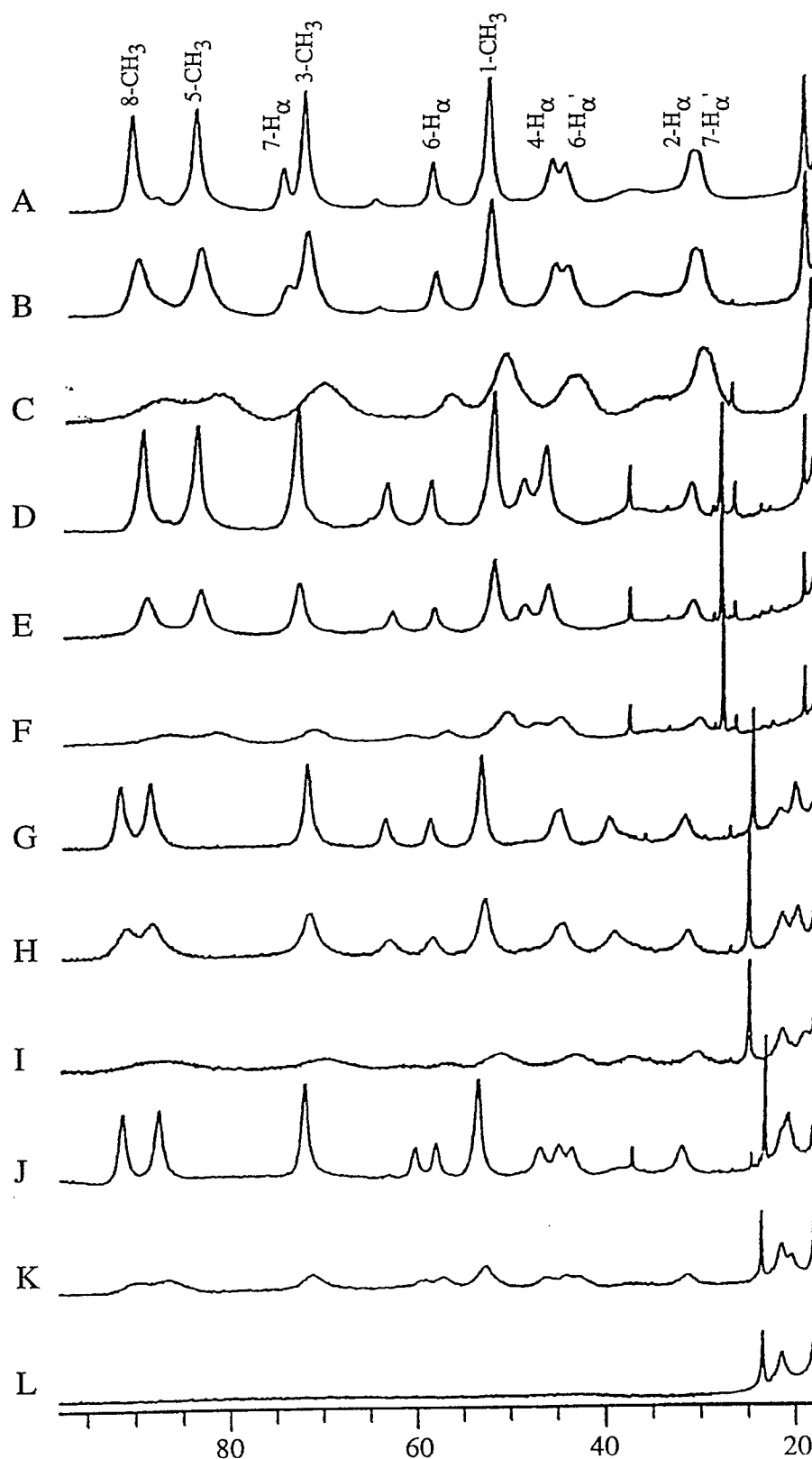


Figure 4S. Low-field resolved portions of the 500 MHz ^1H NMR spectra of: (A-C) WT metMb; (D-F) H97F-metMb; (G-I) S92A-metMb, and (J-L) S92P-metMb at pH 6.4 (A, D, G, J), 7.5 (B, E, H, K), and 8.2 (C, F, I, L) illustrating the similar thermodynamics and dynamics for the acid $\rightarrow$ alkaline transition. The very narrow peaks in the 25-30 ppm region arise from $\sim 3\%$ metMbCN present.