

Fig. S1: (a-b) 2D-IR spectra of NO bound to ferric Cytochrome C recorded at various waiting times,  $T$ . The linear contour scales run from red (negative) to blue (positive).

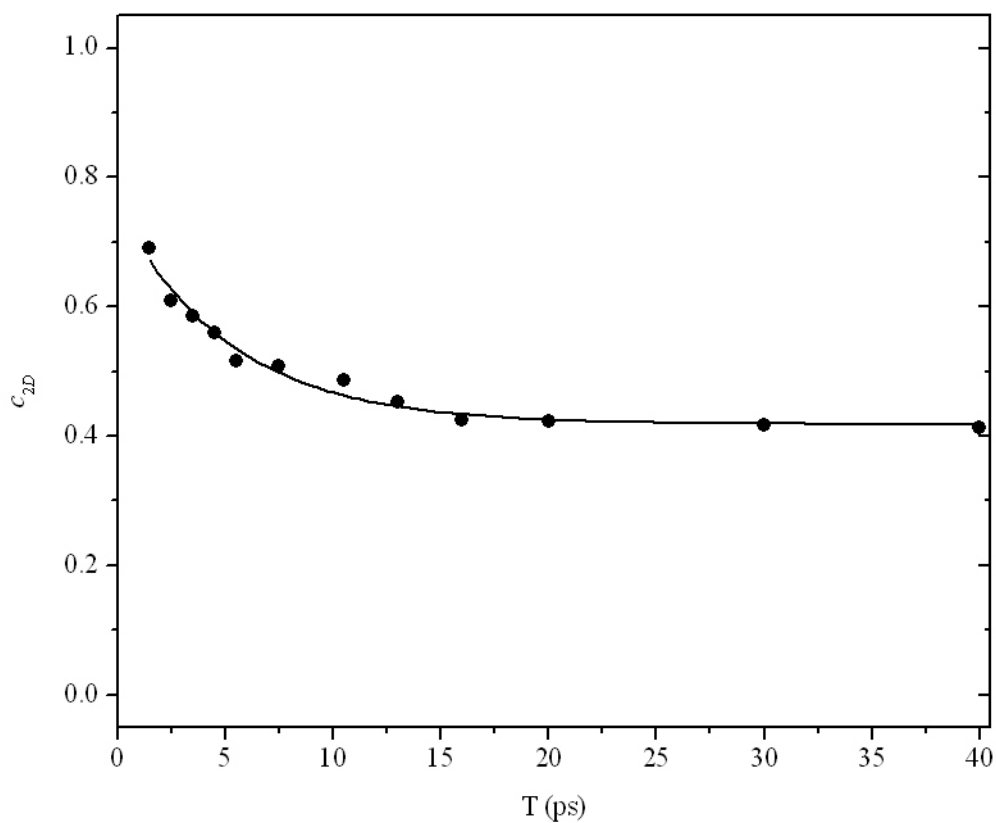


Fig. S2: Temporal evolution of cross-correlation parameter  $c_{2D}$  for the Cytochrome C-NO complex (black circles) determined in same manner as for wt-Mb and mutant Mb in main text. The results of fitting the data to a function consisting of a single exponential decay and a constant parameter are shown as a black line:  $\alpha_1$  (amplitude) = 0.34,  $\tau$  (decay time) = 5.2 ps and  $\alpha_2$  (constant term) = 0.42.