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Experimental Studies of Structure, Function, and Coherent Oscillations in Biomolecules[1]

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Femtosecond coherence spectroscopy can be used to prepare and monitor coherent states of biological samples such as heme proteins. Following laser pulse induced ligand photolysis of myoglobin, the (initially planar) heme group is left far from its final product state equilibrium geometry. This leads to coherent oscillations of those modes composing the reaction coordinate for diatomic ligand binding and dissociation. Coherence studies, along with "white light" continuum measurements of the spectral dynamics, show that the timescale for diatomic ligand dissociation is much shorter than the 150fs period of the Fe-histidine vibration (the Fe-histidine bond constitutes the sole covalent linkage between the heme and protein material). Recent measurements of the effects of temperature and sample condition on the coherent motions of the heme and on the ultrafast geminate rebinding of various diatomic ligands are also reported. Investigations of heme model compounds, in the absence of the protein material, show that the spectrum of low frequency heme modes can be altered by the choice of sample conditions. The studies of the heme model compounds also allow the diatomic ligand rebinding barrier to be separated into "proximal" and "distal" contributions that can be separately determined.

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