

SELECTED PUBLICATIONS

Beck, W. F., "Ultrafast Spectroscopy" in *Encyclopedia of Chemical Physics and Physical Chemistry*; Moore, J. H., Spencer, N. D., Eds.; Institute of Physics Publishing: Bristol, England; Vol. II: Methods, pp 1743-1772 (2001).

Vibrational Coherence from the Dipyridine Complex of Bacteriochlorophyll a Intramolecular Modes in the 10–220 cm⁻¹ Regime, Intermolecular Solvent Modes, and Relevance to Photosynthesis, Shelly, K. R.; Carson, E. A.; Beck, W. F., *J. Am. Chem. Soc.* **2003**, 125, 11810-11811.

Dynamic-absorption Spectral Contours: Vibrational Phase-dependent Resolution of Low-frequency Coherent Wave-packet Motion of IR144 on the Ground and Excited Surfaces, Carson, E. A.; Diffey, W. M.; Shelly, K. R.; Lampa-Pastirk, S.; Dillman, K. L.; Schleicher, J. M.; Beck, W. F., *J. Phys. Chem. A* **2004**, 108, 1489-1500.

Excited-state Axial-Ligand Photodissociation and Non-polar Protein-matrix Reorganization in Zn(II)-substituted Cytochrome c, Lampa-Pastirk, S., Lafuente, R. C., Beck, W. F., *J. Phys. Chem. B* **2004**, in press.

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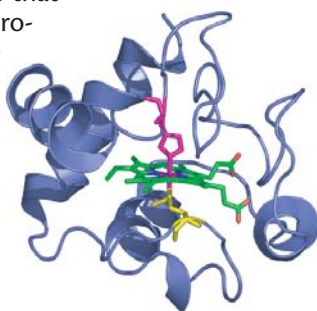
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The Beck group uses ultrafast laser spectroscopy to study the dynamics of biological molecules. At present the group focuses on solvation dynamics in proteins, the reorganizational motions that the polypeptide and surrounding solvent water molecules make in response to a change in the electronic structure of an imbedded chromophore. We are especially interested in the role that solvent water molecules play in protein unfolding and misfolding reactions.

We use femtosecond dynamic absorption spectroscopy with very short laser pulses (10–60 fs) to observe the coherent vibrational motions by a chromophore and its surrounding solvent. We recently observed for the first time that solvent molecules vibrate coherently in response to $\pi \rightarrow \pi^*$ transitions in bacteriochlorophyll *a* solutions and in bacteriochlorophyll proteins from photosynthetic bacteria. The solvent molecules form weak bond-like interactions with the π -electron density of the bacteriochlorophyll molecule. Solvent-like interactions between electronic chromophores and their protein-derived surroundings may



Structure of horse-heart ferricytochrome *c*, as rendered from the pdb file 1HRC. Left: ribbon representation showing the polypeptide's secondary and tertiary structure, with the heme and axial ligands to the Fe(III) ion, Met 80 and His 18, rendered as tube structures. Right: solvent-contact surface, with the heme rendered as a space-filling representation.

play an important role in allowing the protein to tune the reactivity of the imbedded chromophore.

We use femtosecond photon-echo spectroscopy and picosecond time-resolved fluorescence spectroscopy to observe the structural fluctuations of a protein and its surrounding solvent. In this work, an imbedded electronic chromophore is used as the probe, a molecule that senses the motions of its surroundings. We are applying this approach to the study of protein-folding and protein-misfolding reactions. This work pertains to the search for the molecular mechanism behind misfolding-related diseases such as Alzheimer's disease and the spongiform encephalopathies, of which the Creutzfeldt-Jakob and "mad cow" diseases are notable examples. We are interested especially in the motions of partially folded states such as the molten globule. These intermediate structures lie along the reaction coordinate to the fully folded, native state but prior to the transition state, the special structure at the top of the Gibbs free-energy barrier that separates the reactant and product structures.

