



Architecture and Dynamics of Proteins and Aqueous Solvation Complexes
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by Stephan Lotze

This thesis concerns the study of conformational dynamics of (bio)molecular systems with the help of non-linear optical techniques. Hydrogenbonds are one of the most important types of intermolecular interactions in the context of molecular conformation. Molecular vibrations such as the OH-stretch vibration of water molecules and the amide vibrations of peptides and proteins are used as a marker for hydrogenbonding interactions. In this thesis, we use the technique of time-resolved mid-infrared pump-probe spectroscopy to study the dynamics of hydrogenbonded systems. In a pump-probe experiment, a first laser pulse labels a subset of molecules and a second, time delayed probe pulse interrogates the dynamics of the molecules that occur during a time-window ranging from the femtosecond to the hundreds of picoseconds time-scale, allowing us to follow molecular motions in real time. More specifically, we study the molecular motions of water in salt solutions and in highly non-ideal mixtures of water and amphiphiles. The fast, (sub-)picosecond conformational dynamics of a natural cryoprotectant and a model-system for salt bridges are investigated by adding a second spectral dimension to the pump-probe experiments.

Another aspect of molecular conformation, the absolute conformation of chiral molecules, is covered in a second part of this thesis. The specificity of second-order non-linear optical spectroscopy to non-centrosymmetric media is exploited to distinguish between molecules that are mirror images of each other with (near) monolayer sensitivity.