PHYSIKALISCHES KOLLOQUIUM

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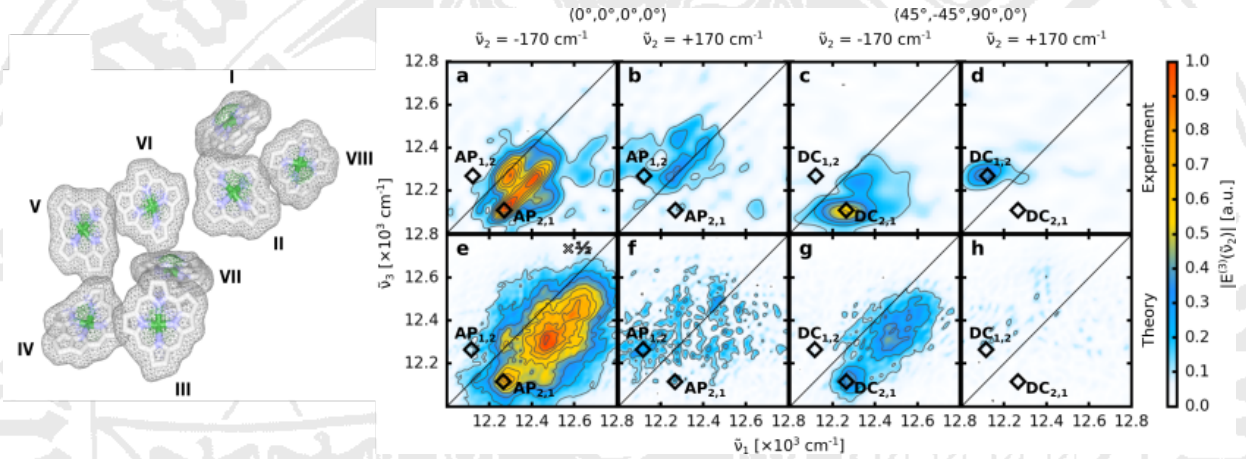

EXPLORING COHERENCES IN MOLECULES AND PHOTOSYNTHETIC COMPLEXES

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Long-lasting coherence signals have been observed in time-resolved spectroscopy measurements of various photosynthetic complexes, and were often attributed to the electronic coherences, which were claimed to be a signature of the coherent energy transfer.
We have carried out high-fidelity polarization-controlled 2DES measurements and carried out quantum beats analysis on a range of systems to clarify the origin of coherences. Even a study of non-interacting chlorophyll molecules in a solution reveals photoexcitation of multiple coherences with vibrational/vibronic origin. The results enable us to establish that the two lower states in chlorophyll molecules are vibronically mixed, which involves several vibrational modes.
Analysis of the coherence dynamics in the Fenna-Matthews-Olson complex, which for a few decades served as a prototypical light-harvesting system for theoretical and experimental studies, allows us to map a very complex picture of coherence signals. We determine that all long-lived coherences have clearly vibrational origin. While electronic coherences are also observed, they, contrary to the earlier claims, dephase on the ~100 fs time scale. We further observe that specific vibrational coherences are excited via vibronically coupled excited states, which indicate the importance of vibronic mixing for energy transfer process in this complex. No signatures of coherent energy transfer are observed, nor such type of transport is required for the efficient function of the light-harvesting complex.