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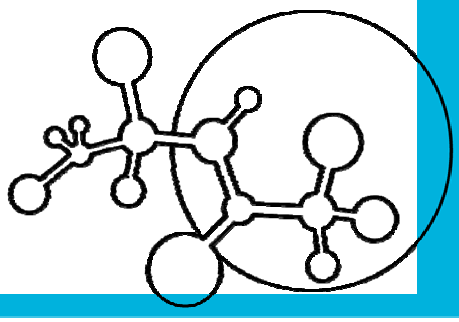

Quantum Efficiency
Seminar and Colloquium

Harald Kauffmann
Faculty of Physics, University at Vienna

Best light at the pulse of molecular quantum dynamics - The beauty of two-dimensional coherent electronic spectroscopy

With the recent advance of two-dimensional Fourier-Transform (FT) electronic spectroscopy (2D-ES), cutting edge experiments have emerged where recording the diffracted molecular Four-Wave-Mixing - electrical field under controlled phase at the amplitude level probes dipole-correlations of molecular resonances along two frequency axes on the 500 THz scale of the optical cycle oscillation. While diagonal signals peaks report on evolutions in population space, off-diagonal signals from cross-peaks are directly sensitive to the presence of (mesoscopic) quantum-interference effects such as electronic coupling, electronic wave-packets and inter-band electronic quantum beating.

After an introductory excurse into the world of coupled electronic oscillators in biological and synthetic light harvesting aggregates, novel results of 2D-ES research are presented in this talk for complex molecular systems where nuclear motional modes couple to electronic transitions, with special emphasis placed on the use of bio-inspired single (monomer) and double (dimeric) Porphyrine - type compounds. In the non-excitonic monomer Zinc-Phthalocyanine (Zn-Pc), long waves modulate electronic diagonal peaks through oscillatory back-&-forth tilt motion along the anti-diagonal axis, while high-frequency nuclear motions modulating the electronic oscillator give rise to vibronic couplings appearing as off-diagonal cross peaks in ultra-broad-band 2D-FT-projections. Thus, off-diagonal couplings, immediately, enable to look into the mystery of FC-vibronic progressions in 1D-linear absorption spectra and reveal the fundamental underpinnings of molecular motion in the fore-field of photo-chemical dynamics.

Furthermore, very recent 2D-ES measurements on the dimeric excitonic Porphyrine system Lu (phthalcyanine) 2 are discussed where the dimer provides a powerful test-system for fundamental research on inter-molecular quantum-dynamics in the presence electronic and vibrational wave-packets, additionally, modulated by charge-transfer resonance splitting. Primarily, expected to sense both electronic and vibronic quantum-packets in a nice choreography between delocalization and partial location by functionally important nuclear modes, the results, however, yield strong deviations from the “usual”. The strange behavior is currently under vivid debate, but may have considerable impact on the perception of long-wave vibrational quantum-coating function claimed in long-lived oscillatory electronic quantum transport at physiological temperatures for the FMO-complex and related light-harvesters. Very recent quantum-coating strategies as refined concepts replacing the generalized electron-phonon approaches by novel physics and super-operators will be addressed.

Date: **Tuesday, July 5th, 2011 4:15 pm**

Location: **FRIAS Seminar Room, Albertstr. 19, Freiburg**

Contact: Andreas Buchleitner, Institute of Physics, Quantum Optics and Statistics,
T +49 761 203 5929 F +49 761 203 5967 E beate.spingler@frias.uni-freiburg.de
www.physik.uni-freiburg.de

Physikalisches Institut
Albert-Ludwigs-
Universität Freiburg