ÉCOLE POLYTECHNIQUE
FÉDÉRALE DE LAUSANNE

Seminars of Condensed-Matter Physics

Monday, 4th of June 2012 – CE 3 – 12h15

Probing electronic processes in molecular complexes by coherent multidimensional visible, UV and X-Ray spectroscopy

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Energy- transfer and charge-separation pathways in the bacterial reaction center may be revealed by coherent two-dimensional optical spectroscopy. The excited state dynamics and relaxation of electrons and holes are simulated using a two-band tight-binding model. The dissipative exciton and charge carrier motions are calculated using a transport theory, which includes a strong coupling to a harmonic bath with experimentally determined spectral density, and reduces to the Redfield, the Förster, and the Marcus expressions in the proper parameter regimes. Direct spectroscopic signatures of the charge separation are identified.

Time-domain experiments that employ sequences of attosecond x-ray pulses in order to probe electronic and nuclear dynamics in molecules are made possible by newly developed bright coherent ultrafast sources of soft and hard x-rays. By creating multiple core holes at selected atoms and controlled times it should be possible to study the dynamics and correlations of valence electrons as they respond to these perturbations. The stimulated x-ray Raman spectra of *trans*-N-methylacetamide and Cysteine at the Nitrogen, Sulfur and the Oxygen K-edges in response to two soft x-ray pulses are calculated. The signals are interpreted in terms of the dynamics of valence electronic wave packets prepared and detected in the vicinity of (either the nitrogen or the oxygen) atom. The evolving electronic charge density and electronic coherences are visualized using a basis set of time-dependent natural orbitals. Effects of orbital relaxation upon core excitations are resolved. A two-dimensional extension of the technique that involves a sequence of three resonant Raman pulses will be presented.