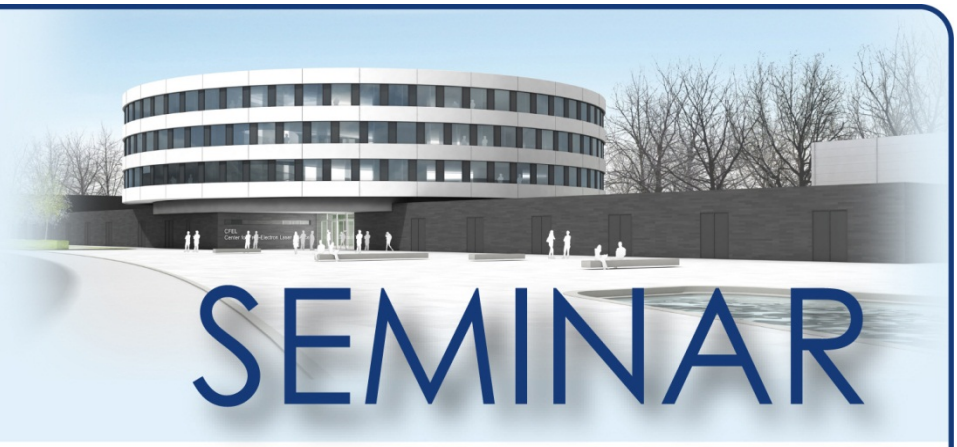
CFEL SCIENCE



SEMINAR

18 January 2024, 10:00–11:00h
CFEL (Bldg. 99) seminar room I+II

Graham Worth

University College London

Simulating Photo-Excited Processes using Grids and Gaussian Wavepackets

Simulating the time-evolution of molecular systems after photo-excitation is complicated by the need to include a manifold of excited electronic states. These states are coupled by so-called non-adiabatic coupling due to the interaction of the electronic and nuclear motion which makes he system highly quantum mechanical. As a result it is necessary to solve the time-dependent Schrödinger Equation (TDSE) for reliable results. To solve the TDSE, however, is a hard computational problem due to the exponential scaling of required resources with system size. The multi-configuration time-dependent Hartree (MCTDH) method provides a suitable algorithm that efficiently contracts the wavefunction to make it manageable for many degrees of freedom [1]. Such simulations can provide a full description of the evolving system, and it is possible to include the excitation by light pulses to obtain a spectroscopic signal that can be directly compared to experiments, such as REMPI spectra [2]. It is also possible to study the coherences induced in a system by a set of pulses [3]. These simulations, however, are constrained by the need for an analytic Hamiltonian, which is usually unable to describe long-range motions. For this, methods such as the direct dynamics variational multi-configuration Gaussian (DD-vMCG) method are required that calculate the potential surfaces on-the-fly and so allow a flexible description of the longe-time molecular response [4].

[1] Beck *et al.*, *Phys. Rep.* (00) 324: 1
[2] Dey *et al.*, *PCCP* (24), In Press
[3] Dey *et al.*, *PRL* (22) 129: 173203
[4] Spinlove *et al.*, *Farad. Discuss*. (18) 212: 191

Host: Jochen Küpper/ CFEL Molecular and Ultrafast Science Seminar