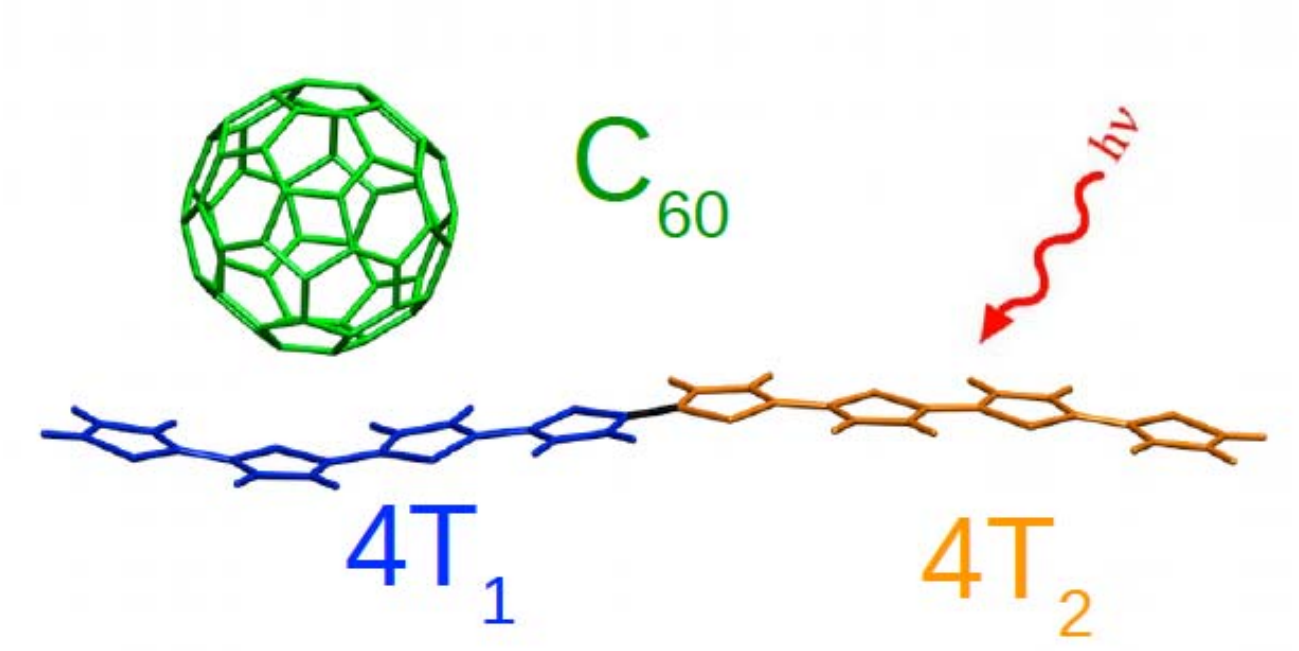
Atomistic modeling of quantum processes in nanoscale devices

Thomas Frauenheim, University of Bremen
***B**remen **C**enter for **C**omputational **M**aterials **S**cience*
http//:www.bccms.uni-bremen.de

The new release of DFTB+ as a density-functional (DFT)-based approach, combining DFT-accuracy and Tight-Binding (TB) efficiency, is reported; http//:www.dftb.org. Methodological details and recent extensions to improve reliability and accuracy will be described. Advanced functions include spin degrees of freedom, time dependent methods for excited states, non-adiabatic electron-ion dynamics and quantum transport calculations under open boundary conditions using non-equilibrium Green´s function methods.

The major focus of the talk will be on the time-dependent DFTB extensions. I am going to present the first real-time atomistic simulation on the quantum dynamics of plasmon excitations in icosahedral silver nanoparticles under strong laser pulses. We identify the emergence of sub-picosecond breathing-like radial oscillations starting immediately after laser pulse excitation, with increasing amplitude as the field intensity increases. The ultrafast dynamic response of nanoparticles to laser excitation points to a new plasmon assisted mechanism rather than longer time-scale equilibrium electron-phonon scattering previously assumed.

The TD-DFTB implementation in real time domain also allow to simulate transient absorption spectra (TAS) fully atomistically. When this technique is applied to the study of ultrafast dynamics of Soret-excited zinc(II)-tetraphenylporphyrin in the sub-picosecond time scale, quantum beats in the transient absorption caused by impulsively excited molecular vibrations are observed. As last examople ultrafast electron transfer in P3HT-PBCM organic blends will be demonstrated to become enhanced by the nuclear motion and under open boundary conditions with applied bias, see ***example below:***



Thomas Frauenheim has made his PhD in 1976 and his Habilitation in 1982 at Technical University Dresden. As Postdoc he has worked 5 years in Joint Institute for Nuclear Research in Dubna until 1982. Later, in 1998 he accepted an offer as chair professor in Computational Materials Science from University of Paderborn and in 2006 he moved to University of Bremen to become the founder of the Bremen Center for Computational Materials Science. Since the mid-90th he pioneered the development of the density-functional based tight-binding method (DFTB) which combines the high efficiency of semi-empirical methods with the accuracy of ab initio density-functional theory (DFT). The new implementation of the method, DFTB+ http://www.dftb-plus.info/overview/ is distributed as open source code.

As director of the German CECAM-Node ***Multi-scale modelling from 1-st principles*** since 2009 he has initiated noumerous International CECAM-Workshops and hands-on-tutorials on atomistic simulations bringing together world leading experts and young researchers from computational solid state and materials physics, theoretical chemistry and molecular biology, promoting a new quality of scientific networking and exchange of knowledge and software developments.. http://www.bccms.uni-bremen.de/events/