



NWO CW Study group meeting
Chemistry in Relation to
Physics and Materials Sciences
4-5 March 2013



H																			He
Li	Be											B	C	N	O	F	Ne		
Na	Mg											Al	Si	P	S	Cl	Ar		
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr		
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe		
	Ba		Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn		
	Ra		Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Uub								
		La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb				
		Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	N				

Netherlands Organisation for Scientific Research

Dynamic energy flow in a hybrid organic-inorganic crystal

Author: **Alexey Polyakov**

Co-authors: *Antonio Caretta, Remco Havenith, Thomas Palstra and Paul van Loosdrecht*
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Organic-Inorganic Hybrid (OIH) materials represent a new class of multiferroics, with ferroelectric order predominantly arising from the organic constituents, and magnetic order from the inorganic counterpart. A Raman spectroscopic study, combined with DFT calculations shows that the polar nature of the OIH di-phenylethylaminecoppertrichloride $[(C_6H_5CH_2CH_2NH_3)_2CuCl_4]$ originates from a tilting of the dipolar organic cations and that the phase transition has a weakly first order character. In order to investigate the interplay between the organic and the inorganic moieties we performed ultrafast spectroscopy experiments aimed at unraveling the thermal coupling between both constituents. Apart from giving an estimate for the coupling constant, the data shows that the critical phenomena near the phase transition are primarily occurring in the organic part as evidenced by a diverging term of the heat capacity. We interpret these effects in terms of rotational and/or librational motions of the organic molecules. Our investigation brings microscopic insight in the origin of polar moment in this unusual ferroelectric material, and presents a novel method to measure the heat capacity of parts of a compound system as well as the coupling between these parts.

Session 12 – Theory: method development (I)

Differential polarization effects with wave function/DFT embedding for excited states

Author: **Csaba Daday**

Co-authors: *Omar Valsson, Carolin Koenig, Johannes Neugebauer and Claudia Filippi*
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Embedding potentials are frequently used to describe the effect of an environment on the electronic structure of molecules in larger systems, including their excited states. If the excitation is accompanied by significant rearrangements in the electron density of the embedded molecule, the environment is expected to strongly respond to this perturbation. These differential polarization effects must therefore be accounted for and state-specific embedding potentials are required for an accurate theoretical description of the excitation. We show that simple strategies to include environmental effects in terms of effective one-electron operators break down if state-specific embedding potentials are used, and propose a simple correction that leads to significantly improved excitation energies.

We demonstrate these concepts within a wave function in density functional theory (WF/DFT) hybrid scheme, where the active subsystem is described by multireference perturbation theory (CASPT2) or quantum Monte Carlo (QMC) methods. The effect of the environment is included via density-based state-specific embedding potentials obtained with the aid of orbital-free DFT. We illustrate the good performance of our embedding scheme in describing the excitations of a set of small molecules in various solvents.

The physical superiority of the LDA over the B3LYP functional and how we can do better

Author: **Klaas Giesbertz**

Co-authors: *Robert van Leeuwen and Ulf von Barth*

The past few decades density functional theory (DFT) has become one of the most used methods to deal with the quantum nature of electrons. However, the correct description of strongly correlated electrons (multiple determinants are important) remains elusive. A typical example is the breaking of a chemical bond. Using exchange-correlation holes I will demonstrate how the exact DFT functional handles the dissociation of a chemical bond. Although B3LYP is the most popular DFT functional in chemistry, it turns out to be worse than the simple local density approximation (LDA) from a physical point of view. I will also show a possible route to incorporate the correct physics in a fully non-local DFT functional.

Session 13 – Nano & quantum dots

In Vivo Near Infrared FRET Imaging of Nanoparticle Accumulation and Dissociation Kinetics in Tumour-bearing Mice

Author: **Yiming Zhao**

Co-authors: *Inge van Rooy, Sjoerd Hak, Francois Fay, Jun Tang, Aurelian Radu, Zahi. A. Fayad, Celso de Mello Donegá, Andries Meijerink and Willem Mulder*
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In the last decade self-assembled lipidic nanoparticles have been increasingly explored as intravenously injectable agents for biomedical purposes. They can serve as delivery vehicles for a wide variety of drugs, ranging from cytostatic agents to RNA interference and proteins, and as molecular imaging probes. Despite their widespread application in in vivo studies, most studies have not addressed where and how fast such self-assembled lipid nanoparticles lose their original composition after administration, or subsequent trafficking upon accumulation at the targeted site.