

## MM 7: Computational Materials Modelling - Methods

Time: Monday 11:45–13:00

Location: H24

MM 7.1 Mon 11:45 H24

**Quantum electrodynamics of nanostructures** — ●WALTER TARANTINO, J. K. DEWHURST, S. SHARMA, and E. K. U. GROSS — Max-Planck-Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany

The project is aimed to develop a time-dependent functional theory for quantum electrodynamics. We want to establish a Kohn-Sham system for relativistic electrons, as well as positrons and photons. Apart from representing the natural generalization of the usual non-relativistic many-body problem, the approach will provide us with a novel instrument for computational investigations of the interaction between light and matter. Details about preliminary results on KS systems and the effective (exchange-correlation) potentials will be presented.

MM 7.2 Mon 12:00 H24

**An Approach to Coarse-Grained Molecular Dynamics** — ●DAVID EDMUNDS, PAUL TANGNEY, DIMITRI VVEDENSKY, and MATTHEW FOULKES — Imperial College London, London, UK

Traditional molecular dynamics (MD) simulations provide an effective method for simulating the evolution of a system at the atomic level, without the associated computational cost of ab-initio methods such as density-functional theory. However, many physical and biological processes occur on length scales of millimetres and time scales of seconds, which remain out of reach to fully atomistic simulations on even the most powerful modern supercomputers.

Fortunately, a family of techniques known as coarse-grained molecular dynamics (CGMD) exists, which can bridge the gap between atomistic simulation and the mesoscale. These approaches allow for a larger time step to be used, as the fast degrees of freedom in the system have effectively been integrated out. They can also produce CG potentials which are much less computationally intensive than their all-atom counterparts. This results in CG simulations which are several orders of magnitude faster than all-atom MD, allowing the simulation of billion atom systems on standard desktop computers.

We present two strategies for deriving a coarse-grained pairwise interaction potential, based on the internal energy and free energy of a pair of constrained molecules. We benchmark these methods against a fully atomistic MD simulation of interacting C60 buckyballs, chosen since they are trivially represented by a single CG site located at their centre of mass.

MM 7.3 Mon 12:15 H24

**New Methods for Calculating the Free Energy of Charged Defects in Solid Electrolytes.** — ●ROBERT HORTON, ANDREW HASLAM, AMPARO GALINDO, GEORGE JACKSON, and MICHAEL FINNIS — Imperial College London, London, United Kingdom

Solid electrolytes are utilised in a myriad of technological applications ranging from fuel cells to gas sensors. It is the presence of charged defects in their lattice structure that is primarily responsible for their useful electrolytic behaviour [1] and, accordingly, a detailed understanding of the thermodynamic behaviour of these defects is highly desirable. Analytic models for describing the thermodynamic contribution of defects break down at concentrations of a few percent, due to the long-ranged nature of the Coulombic defect-defect interactions. We have developed and tested an alternative scheme, using Wang-Landau sampling [2], a temperature-independent Monte Carlo method, to statistically calculate the free energy of the system at the atomistic level. It is interesting to see that this calculated free energy can be fitted

by traditional functional forms, such as regular-solution and Redlich-Kister, with which the thermodynamic properties of the system can be calculated.

[1] S. C. Singhal (2000). Science and Technology of Solid-Oxide Fuel Cells. MRS Bulletin, 25, pp 16-21 doi:10.1557/mrs2000.13

[2] F. Wang and D. P. Landau, (2001). Physical Review E. 64, 056101-1-16.

MM 7.4 Mon 12:30 H24

**A polarizable potential for the Al<sub>2</sub>O<sub>3</sub>-Y<sub>2</sub>O<sub>3</sub> system** — ●HANNES GUHL, MATTHEW FOULKES, MICHAEL FINNIS, and PAUL TANGNEY — Department of Physics and Materials, Imperial College London, SW7 2AZ, UK

The high-temperature oxidation rate of alloys containing aluminium is much reduced in the presence of rare earth (RE) elements such as yttrium. Apparently, the grain boundaries acting as the primary channel via which oxygen and metal ions cross the oxide film are effectively blocked by the segregating RE elements, yet it is controversial whether or not this “reactive element effect” is due to the alteration of the electronic- or atomic structure.

We apply density functional theory (DFT) as the best available tool for addressing the subtle interplay of defect formation energies and mobilities, including yttrium in grain boundaries of alumina. However, due to their complexity, realistic structural models of grain boundaries cannot be established with DFT alone. Therefore, we present here an empirical, computationally cheap, polarizable ion potential, which matches the DFT interatomic forces in Y doped alumina. We make a direct comparison between computed and experimentally measured grain boundary structures and energies. In addition, the potential captures the essential energetic and structural properties of pure Y<sub>2</sub>O<sub>3</sub> as well as of mixed phases such as Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>, suggesting that it will be useful for atomistic studies of the complex thermodynamics across the Al<sub>2</sub>O<sub>3</sub>-Y<sub>2</sub>O<sub>3</sub> system.

MM 7.5 Mon 12:45 H24

**Exploring the exohedral reactivity of endohedral metallofullerenes (EMFs): the encapsulation effect and the fullerene strain energy** — ●SILVIA OSUNA<sup>1,2</sup>, MARC GARCIA-BORRÀS<sup>2</sup>, JOSEF M. LUIS<sup>2</sup>, MARCEL SWART<sup>2,3</sup>, and MIQUEL SOLÀ<sup>2</sup> — <sup>1</sup>University of California, Los Angeles (UCLA), Los Angeles, USA — <sup>2</sup>Universitat de Girona, Girona, Spain — <sup>3</sup>Institució Catalana de Recerca i Estudis Avançats (ICREA), Barcelona, Spain

Endohedral metallofullerenes (EMFs) have attracted increasing attention in the last years due to their potential applications in the field of biology and medicine as a result of the magnetic, spectroscopic, and nuclear properties of the metal ions encapsulated inside. [1] The third most abundant fullerene is the trimetallic nitride template (TNT) EMF, Sc<sub>3</sub>N@C<sub>80</sub>. In these compounds, there exists a formal electronic transfer of six electrons from the M<sub>3</sub>N unit to the fullerene structure (M<sub>3</sub>N<sup>+6</sup>: C<sub>x</sub><sup>6-</sup>). Consequently, the metal cluster encapsulated inside has a huge influence on the reactivity of these compounds, which is, in general, reduced in TNT EMFs as compared to free fullerenes. In the present work, we perform a theoretical study of the thermodynamics and the kinetics of the [4+2] Diels-Alder (DA) reaction of s-cis-1,3-butadiene with several isomers of free fullerene cages (i.e. I<sub>h</sub>-C<sub>80</sub>, D<sub>5h</sub>-C<sub>80</sub>, D<sub>3h</sub>-C<sub>78</sub>) and their respective TNT-EMF compounds encapsulating differently sized cluster metals (i.e. Sc<sub>3</sub>N, Y<sub>3</sub>N, Lu<sub>3</sub>N, Gd<sub>3</sub>N, Ti<sub>2</sub>C<sub>2</sub>). (1) Osuna, S. et al. Phys. Chem. Chem. Phys. 2011, 13, 2486-2488.