

DS 13: Frontiers of Electronic-Structure Theory: Focus on the Interface Challenge IV (joint session O/ CPP/ DS/ TT)

Time: Tuesday 14:00–16:45

Location: H9

Topical Talk

DS 13.1 Tue 14:00 H9
The Data Revolution in Materials Science, Through the Lens of the Materials Project — ●KRISTIN PERSSON — University of California, Berkeley, USA

Advanced materials are essential to economic and societal development, with applications in multiple industries, from clean energy, to national security, and human welfare. Historically, novel materials exploration has been slow and expensive, taking on average 18 years from concept to commercialization. Due to the tremendous improvements in computational resources, coupled with software development during the last decades, real materials properties can now be calculated from quantum mechanics much faster than they can be measured. A new era of computational materials prediction and design has been born. A result of this paradigm change are databases like the Materials Project which is harnessing the power of supercomputing together with state of the art quantum mechanical theory to compute the properties of all known inorganic materials and beyond, design novel materials and offer the data for free to the community together with online analysis and design algorithms. We leverage the rich data from the Materials Project for machine learning; accelerating materials design, characterization and finally synthesis of materials. This talk will survey this rapidly evolving and exciting paradigm in science, showcasing the possibilities and iteration between ideas, computations, insight and new materials development.

DS 13.2 Tue 14:30 H9

High-throughput simulations of complex band structure — ●EMANUELE BOSONI and STEFANO SANVITO — School of Physics and CRANN, Trinity College Dublin, College Green, Dublin 2

The Complex Band Structure (CBS) generalizes the conventional band structure of a material by considering wave-vectors with complex components. The CBS extends the description of the allowed states of a material beyond the bulk propagating states, including in the picture the evanescent wave-functions that grow or decay from one unit cell to the next. Even though these latter states are forbidden by translational symmetry, they become important when this is broken via, for example, an interface. In the past, many studies made use of CBS calculations in order to confirm or motivate experimental findings, but only recently we see some efforts to give an unified prospective to the study of this quantity [1]. Moreover, the growth in recent years of high-performance computational resources available at relatively low cost opens the possibility to make a systematic, high-throughput, study of the CBS within the Density Functional Theory (DFT) framework.

In this contribution we will present our implementation of the Transfer Matrix Method [1] for the calculation of the CBS within the DFT code Siesta [2] and we will explain challenges and benefits of the high-throughput approach. We will present a proof of concept example in which we calculated the CBS for a set of materials and we will conclude explaining the relevance of our project in the field of spintronics.

- [1] Reuter M. G., J. Phys.: Condens. Matt. 29, 053001 (2017)
- [2] J. M. Soler et al., J. Phys.: Condens. Matt. 14, 2745 (2002)

DS 13.3 Tue 14:45 H9

Computational Screening of 2D Materials for Solar Cells Application — ●ANDERS CHRISTIAN RUIJ-JENSEN and KRISTIAN SOMMER THYGESEN — Institute for Physics, Technical University of Denmark

Two-dimensional (2D) materials have attracted great attention in recent years, not least due to their extremely strong coupling to light and pronounced excitonic effects. This makes 2D materials an ideal playground for studying light-matter interaction in nano-scale materials for applications within e.g. solar cells and photo-detectors. In this work, we present a large-scale computational study of a wide variety of 2D materials with the aim of identifying novel candidates with strong light-matter interaction. Specifically, we calculate the absorption spectrum of almost 2000 materials at the level of the Random Phase Approximation (RPA) and/or by solving the Bethe-Salpeter Equation (BSE). This enables us of calculating the Power Conversion Efficiency (PCE) for all these materials fully ab-initio. Based on this we identify specific 2D semiconductors for both single- and tandem solar cells. The best candidates present power densities (PCE per unit mass) of several or-

ders of magnitude larger than both Si and GaAs. Lastly, we also show a detailed analysis of a few 2D materials, in which we find exciton states with exceptionally strong coupling to light and large exciton binding energies.

DS 13.4 Tue 15:00 H9

First-principles Modelling of Solid-Solid Interfaces in all Solid-State Batteries — ●BORA KARASULU¹, JAMES P. DARBY¹, CLARE P. GREY², and ANDREW J. MORRIS³ — ¹Dept. of Physics, Univ. of Cambridge, UK — ²Dept. of Chemistry, Univ. of Cambridge, UK — ³School of Metallurgy and Materials, Univ. of Birmingham, UK

All solid-state batteries (ASSBs) can potentially mitigate the safety issues known for conventional Li-ion batteries, and provide enhanced energy densities, by replacing the organic electrolyte solutions with solid inorganic equivalents. Mechanical and (electro)chemical incompatibilities between the ASSB solid components, however, lead to high resistances, curtailing the Li-ion transport at their interfaces. In this talk, we introduce a high-throughput ab initio modelling approach towards the rational design of electrolyte/electrode interfaces in ASSBs. First, we obtain phase diagrams of sulfide-based electrolytes with diverse compositions, phases, vacancies and doping using the Ab Initio Random Structure Searching (AIRSS) method. Next, the stable and low-lying metastable phases are screened for their ionic conductivity using ab initio molecular dynamics simulations. Finally, diverse surfaces of the selected electrolyte phases are interfaced with the surfaces of electrodes or other interfacial (e.g. solid electrolyte interphase, SEI) layers to determine the stable combinations using an automated procedure (INTERFACER). The devised approach will be demonstrated in action for a collection of interfaces, e.g. Li-P-S electrolytes with LiCo₂, Li-metal electrodes and alike.

DS 13.5 Tue 15:15 H9

New Insights into Amorphous Materials and their Surfaces by Combining Machine Learning and DFT — ●VOLKER DE-RINGER — University of Cambridge, Cambridge, UK

Understanding links between atomic structure, chemical reactivity, and physical properties in amorphous solids is a long-standing challenge. DFT-based atomistic simulations have played important roles in this, but come at high computational cost. Novel interatomic potentials based on machine learning (ML) achieve close-to DFT accuracy, but require only a small fraction of the cost. In this talk, I will argue that such ML-based potentials are particularly useful for studying amorphous solids.

First, I will describe a Gaussian Approximation Potential (GAP) for amorphous carbon, which we recently used to simulate the deposition of tetrahedral amorphous carbon (*ta*-C) films, one atom at a time. These simulations reproduced the experimentally observed count of sp³ atoms and gave new insight into the microscopic growth mechanism. I will then discuss how ML-based potentials can be combined with density-functional methods to yield new insights into surface functionalization (specifically, hydrogenation and oxidation) of *ta*-C. Finally, I will present recent work on amorphous silicon, another prototypical non-crystalline material that ML-driven simulations can describe with high accuracy. Looking ahead, these studies suggest that ML-based potentials may become more widespread tools for the realistic modelling and understanding of the amorphous state.

DS 13.6 Tue 15:30 H9

Harvesting from unbiased sampling of open systems: phase diagrams and property maps of surfaces and clusters in reactive atmosphere — ●YUANYUAN ZHOU, MATTHIAS SCHEFFLER, and LUCA M. GHIRINGHELLI — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany

Numerous processes that occur at surfaces of materials play a critical role in the manufacture and performance of functional materials, e.g., electronic, magnetic, and optical devices, sensors, catalysts, and coatings. A prerequisite for analyzing and understanding the electronic properties and the function of surfaces is detailed knowledge of the atomic structure, i.e., the surface composition and geometry under realistic condition. We introduce a Replica-Exchange (RE) Grand-Canonical (GC) Monte-Carlo algorithm. By means of the coupled sam-

pling at several chemical potentials (i.e., partial pressures) of a reactive gas atmosphere and temperatures, the REGC scheme enables the unbiased calculation of (p, T) phase diagrams of surfaces, nanoparticles, or clusters in contact with reactive atmosphere, where all anharmonic contributions are included. Moreover, the multi-canonical sampling yields the temperature-pressure dependence (map) of all equilibrium observables that can be measured within the given model Hamiltonian. For instance, structural parameters such as the radial distribution function, or the fundamental electronic gap. This allows for rational design, where *operando* conditions are taken fully into account. We demonstrate the approach for model Lennard-Jones surfaces as well as Si clusters and surfaces in a hydrogen atmosphere.

DS 13.7 Tue 15:45 H9

Crystal structure prediction for high capacity battery materials — ●ANGELA F HARPER¹ and ANDREW J MORRIS² — ¹Department of Physics, University of Cambridge, JJ Thomson Ave, Cambridge CB3 0HE, UK — ²School of Metallurgy and Materials, University of Birmingham, Edgbaston, Birmingham, UK

The future of large-scale energy storage relies heavily on the ability of Li-ion batteries to have high capacity and long-term stability. At present, graphite anodes limit the overall capacity of Li-ion batteries to a theoretical maximum of 372 mAh/g, and thus there is a need for higher capacity anodes such as phosphorus. We have studied lithiation in phosphorus using a combination of *ab initio* random structure searching (AIRSS) and density-functional theory calculations. In the Li-P system we found a novel phase of $P2_12_12_1$ Li_4P_3 , which showed stable 0K phonon modes [1]. We further showed, using defect AIRSS searches, that doping the known Li-P phases with aluminium improved their electronic density of states at the Fermi level, and could improve conductivity in phosphorus anodes. To improve the cyclability of these phosphorus anodes, which break down after several cycles due to volume expansion of over 200%, in this talk we will investigate the phase diagrams and voltage profiles of several ternary compounds of Li-P-M where M is a metallic element which maintains the conductivity of aluminium doping and adds stability to the anode.

[1] Mayo, M. et al. Chem. Mater. 2016, 28, 2011*2021

DS 13.8 Tue 16:00 H9

Constructing Accurate Machine Learning Force Fields for Flexible Molecules — ●VALENTIN VASSILEV-GALINDO, IGOR POLTAVSKY, and ALEXANDRE TKATCHENKO — Physics and Material Science Research Unit, University of Luxembourg, Luxembourg

State-of-the-art machine learning (ML) models can reproduce potential energy surfaces (PES) for molecules containing up to a few tens of atoms with the accuracy comparable to the most exact *ab initio* methods. This provides a unique tool for computing different thermodynamic properties that would require millions of CPU years otherwise. For instance, a recently developed sGDML[1,2] model predicts forces and energy with CCSD(T) accuracy using just a few hundreds of configurations for training. However, up to now ML has been mainly applied to rather rigid molecules. In this regard, our objective is to test ML for flexible molecules and out-of-equilibrium configurations along transition paths. For this, we select molecules (e.g. azobenzene, stilbene) with relatively complex transition paths, which result from an interplay between long- and short-range interactions. Then, different paths connecting PES minima are tested using sGDML. This allows

us to define optimal descriptors and the most appropriate strategies for choosing the training sets, which is crucial for ML models relying on a limited number of training points. Our results open an avenue for efficiently calculating transport pathways, transition rates and other out-of-equilibrium properties with previously unattended accuracy.

[1] Chmiela, S. et al., Sci. Adv. 3, e1603015 (2017).

[2] Chmiela, S. et al., Nat. Commun. 9, 3887 (2018).

DS 13.9 Tue 16:15 H9

Calculating critical temperatures for magnetic order in two-dimensional materials — ●DANIELE TORELLI — CAMD, Department of Physics, Technical University of Denmark, 2820 Kgs. Lyngby, Denmark

Recent observation of ferromagnetic out-of-plane order in two-dimensional (2D) CrI_3 highlights the importance of a microscopic understanding of magnetic anisotropy (MA) in ground state magnetic systems. Single-ion anisotropy and anisotropic exchange coupling comprise crucial ingredients to escape the Mermin-Wagner theorem which implies that rotational symmetry cannot be spontaneously broken at any finite temperature in 2D and thus prevents magnetic order.

In the present work we investigate the variation of critical temperature in Heisenberg model systems using classical Metropolis Monte Carlo simulations. A fit for square, hexagonal and honeycomb lattices leads to a simple expression for the critical temperatures as a function of MA and exchange coupling constants.

Based on a new developed computational 2D materials database, we predict 2D structures with high critical temperatures and high thermodynamic and dynamic stability for future experimental investigations. As testing system, relevant Heisenberg exchange couplings and MA energies in mono-layer CrI_3 are obtained from first principle calculations and energy mapping analysis, yielding to an estimation of Curie temperature in good agreement with experimental results.

DS 13.10 Tue 16:30 H9

Amino-acids on metallic surfaces: searching conformational space — ●DMITRII MAKSIMOV, CARSTEN BALDAUF, and MARIANA ROSSI — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany

Organic-inorganic interfaces are challenging for computational modeling, in particular regarding the prediction of stable configurations at the interface, which determine the electronic properties of the system as a whole. The amino acid arginine is a flexible molecule in the gas phase and experiments show its self-assembly into dimers, rings, and chains on Ag(111) and Au(111) surfaces. For two protonation states (neutral Arg and charged $ArgH^+$), we perform systematic structure searches by placing known gas-phase minima in different orientations on top of the surfaces, followed by full relaxation within long-range dispersion corrected density-functional theory (DFT). In the analysis, we aim at understanding the alterations of the conformational space from the gas phase to surface adsorption by means of a dimensionality-reduced representation based on a combination of the Smooth Overlap of Atomic Positions (SOAP) and the Sketchmap techniques [1]. The favorable interaction with the metallic surface reduces the number of accessible conformations for neutral Arg. For the adsorption of charged $ArgH^+$, the number of local minima increases due to surface-dependent partial charge screening. [1] S. De et al., *J. Cheminform.*, **9:6** (2017)