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High accuracy simulation methods for realistic materials modelling

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14. ABSTRACT
The main objective of this short study is to demonstrate the feasibility of using highly accurate methods, based on the quantum Monte Carlo (QMC) technique, to provide benchmarks energies to aid the construction of machine learning (ML) approaches to study complex and realistic material science systems. The properties of materials and how they interact with the environment is often modified by the adsorption of water and other molecules on their surfaces. In particular, the interaction of water with various surfaces, and the interaction of water with other molecules on surfaces is ubiquitous in several chemical and biological processes, not to mention that every surface in contact with Earth's atmosphere is exposed to water vapour. Interaction energies between water and other molecules on carbidic and other complex surfaces is important to an almost endless list of practical applications such as corrosion, lubrication, friction, catalysis, coatings, energy storage, and sensors to name a few. Given that water interacts weakly with graphitic surfaces and that there are many competing structures with similar energies, this is one of the most challenging systems for any modelling method, and for this reason an accurate reference atomic level approach is needed to make progress with understanding these systems. If the approach can be validated on such a complex system, it would provide a firm ground to build on for the study other difficult systems, such as 2 dimensional layered materials. To demonstrate the feasibility of our approach, our initial focus will be on water and its interaction with graphene, as this is a system for which we have considerable experience and made most of the ground work, demonstrating the high accuracy of the QMC method. We will generate large sets of configurations of water clusters interacting with a graphene layer and compute the QMC energies of these configurations. We will then use these benchmark energies to train an appropriately chosen ML potential. This ML potential will then be available to study coarse grained systems, such as the shape and behaviour of water droplets on graphitic surfaces, with possible applications to lubrication, catalysis and water filtering. Publications from the grant are:
Publications
1. Brandenburg, J. G., Zen, A., Alfè, D. & Michaelides, A. Interaction between water and carbon nanostructures: How good are current density functional approximations? J. Chem. Phys. 151, 164702 (2019). .DOI:10.1063/1.5121370
2. Zen, A., Brandenburg, J. G., Michaelides, A. & Alfè, D. A new scheme for fixed node diffusion quantum Monte Carlo with pseudopotentials: Improving reproducibility and reducing the trialwave- function bias. J. Chem. Phys. 151, 134105 (2019).,.DOI:10.1063/1.5119729
3. Nakano, K. et al. TurboRVB: a many-body toolkit for ab initio electronic simulations by quantum Monte Carlo. (2020). J. Chem. Phys. (in press) .arxiv.org/abs/2002.07401

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Slide 1: Project Summary

- Project Title: High Accuracy simulation methods for realistic materials modelling
- Grant Number: FA9550-19-1-7007
- Keywords: material modeling, electronic structure, quantum Monte Carlo, water, graphene
- Objective: To investigate and establish the feasibility of developing a machine learning potential for molecular adsorption on two-dimensional layered systems. To develop and improve the accuracy of the quantum Monte Carlo method, in order to provide accurate benchmark energies for the training set of the machine learning procedure.
- Progress: Graphene finds an exceptional range of potential applications in industry and technology; it is exploited for drug delivery, water desalination, nerve tissue engineering to probe and augment cell behavior, and it is used as filler in tier materials to strengthen reinforcement properties. Many of these applications find water interfaced with graphitic surfaces, for instance in desalination and drug delivery. Understanding the interaction between water and graphene at the molecular lever can be of fundamental importance to foster technological developments and promote new applications. However, this kind of understanding is hard to achieve, because the traditional computational techniques do not have the required accuracy to describe these systems. We have used an advanced and highly accurate computational approach to investigate precisely the water graphene interaction: the diffusion quantum Monte Carlo (DMC) method. DMC is a stochastic approach to solve the Schrödinger equation and obtain an understanding of the physics underlying these systems from first principles. Moreover, DMC is exceptionally efficient on modern supercomputer facilities. This kind of investigations leads material modeling to new horizons, yielding a description of complex interface interactions with unprecedented accuracy. In the last year we assembled a benchmark set for the water-carbon interaction, for which we provide accurate evaluations of the binding energy based on DMC. Moreover, we worked on an improved DMC algorithm. In particular, developed and implemented a new algorithm to employ pseudopotentials on DMC, yielding more accurate and reproducible evaluations.

Slide 2: Project Team

- PI: Dario Alfè
- Co-PI: Angelos Michaelides
- Key Researchers: Andrea Zen
- Students: none

Dario Alfè

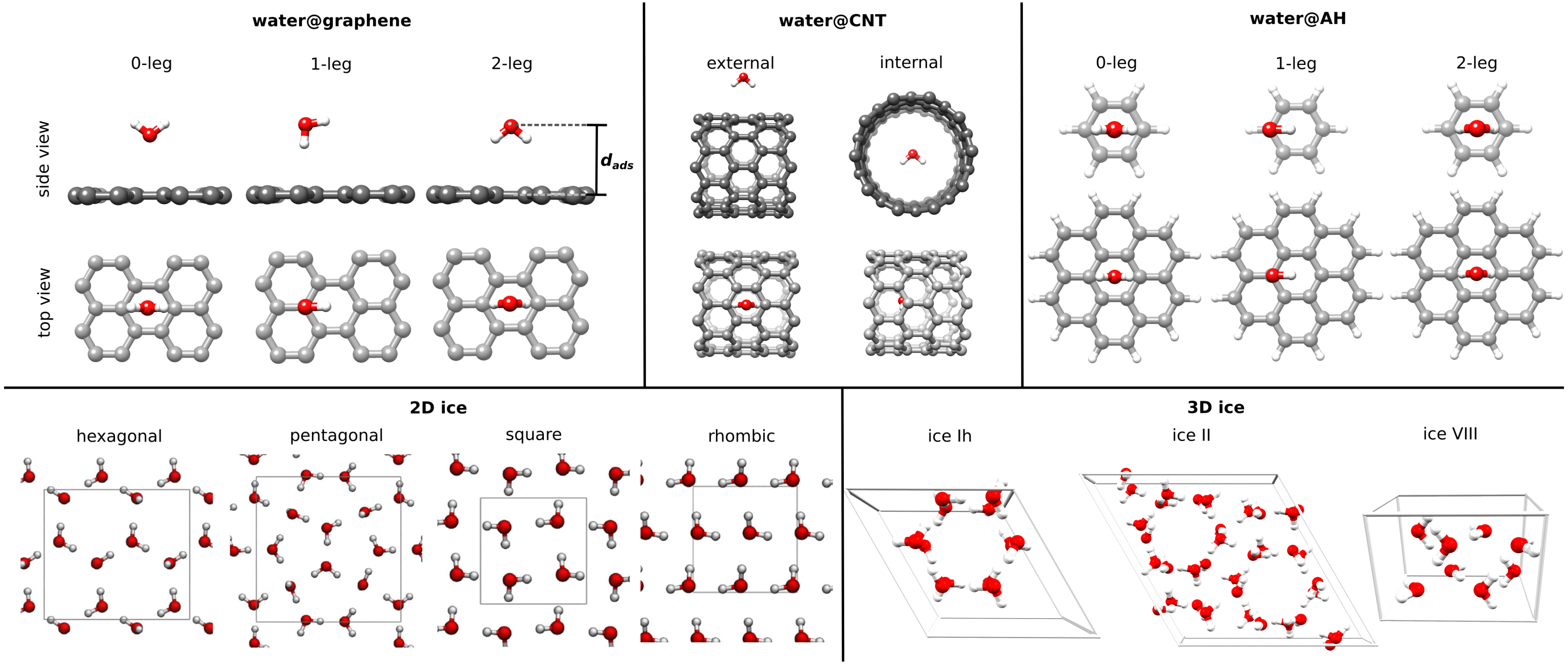
Angelos Michaelides

Andrea Zen

Slide 3: Activities and Accomplishments

- We published three papers in peer-review international journals (see slide 5)
- Our publication "*Interaction between water and carbon nanostructures: How good are current density functional approximations?*", J. Chem. Phys., has been selected as an Editor's pick.
- Our improvement to the diffusion Monte Carlo algorithm, described in our publication "*A new scheme for fixed node diffusion quantum Monte Carlo with pseudopotentials: improving reproducibility and reducing the trial-wave-function bias"* [J. Chem. Phys., **151**, 134105], has been implemented in three different packages for QMC simulations: QMCPACK, CASINO and TurboRVB.
- We disseminated the results of our research:
  - Andrea Zen, invited talk at the conference "MACHINE LEARNING FOR QUANTUM DESIGN" at the Perimeter Institute, Waterloo, Canada (July 10th , 2019)
  - Andrea Zen, presentation at the Max Planck Institute for Solid State Research, Stuttgart, Germany (May 15th , 2019)
  - Dario Alfè, invited talk, APS March meeting "Chemical equilibrium at the Earth's core mantle boundary", Boston (March 2019)
  - Dario Alfè, invited talk, State Key Laboratory of Lunar and Planetary Science (MUST) "Chemical equilibrium at the Earth's core mantle boundary", Macau, China (June 2019)
  - Dario Alfè, invited talk, Workshop The interior of Jupiter and Saturn, MUST "The inner core nucleation paradox", Macau, China (November 2019)
- We obtained an OLCF Director's Discretion Project for 2020, one for 2021 and one for 2022, which granted us the access to some of the most powerful high-performance computing facilities on the planet, namely the Summit and Rhea supercomputers, to pursue our research.
- We profited from collaboration with (late) Prof. Sandro Sorella (SISSA, Italy) and his group.

Slide 4: Technical Highlights



The figure shows the **WaC18** benchmark set, that we have assembled and reported in our paper [J. Chem. Phys., 151, 134105] to test and train the interaction between water and graphitic surfaces.

Slide 5: Publications

<u>Publications</u>

1. Brandenburg, J. G., Zen, A., Alfè, D. & Michaelides, A. Interaction between water and carbon nanostructures: How good are current density functional approximations? *J. Chem. Phys.* **151**, 164702 (2019). DOI:10.1063/1.5121370
2. Zen, A., Brandenburg, J. G., Michaelides, A. & Alfè, D. A new scheme for fixed node diffusion quantum Monte Carlo with pseudopotentials: Improving reproducibility and reducing the trial-wave-function bias. *J. Chem. Phys.* **151**, 134105 (2019). DOI:10.1063/1.5119729
3. Nakano, K. *et al.* TurboRVB: a many-body toolkit for *ab initio* electronic simulations by quantum Monte Carlo. (2020). *J. Chem. Phys. (in press)* arxiv.org/abs/2002.07401