

**MM 41: Computational Materials Modelling - Solids and Molecules (joint session MM/ CPP)**

Time: Wednesday 15:45–17:00

Location: IFW B

MM 41.1 Wed 15:45 IFW B

**Molecular Adsorption Potential Energy Surfaces and their Reproducibility** — •LUKAS HÖRMANN, ANDREAS JEINDL, and OLIVER T. HOFMANN — Institute of Solid State Physics, NAWI Graz, Graz University of Technology, Petersgasse 16, 8010 Graz, Austria

Molecular adsorption on surfaces depends on a range of mechanisms: covalent bonds, charge transfer and van-der-Waals (vdW) interactions shape the potential energy surface (PES), making the PES key to understanding molecule-substrate interfaces. Describing these interfaces with density functional theory requires a wise selection of the exchange correlation functional and vdW correction scheme. To explore the robustness of the PES with respect to the choice of method, we present a benchmark of common local, semi-local and non-local functionals in combination with various vdW corrections. We investigate these methods using perylenetetracarboxylic dianhydride (PTCDA) on Ag(111), one of the most frequently studied systems.

We use an in-house developed Gaussian process regression algorithm, which requires only about 50 DFT calculations as input to generate a PES with DFT accuracy. This allows a detailed analysis of the PES's features, such as positions and energies of minima and saddle points. Comparing the results from different exchange correlation functionals enables us to identify trends and differences between the approaches. Finally, we compare key features, such as local minima, with experimental data to determine a "quality seal" for the different functionals and vdW corrections.

MM 41.2 Wed 16:00 IFW B

**The role of structural symmetry for proton tautomerization in aromatic molecules** — •ANTONIOS RAPTAKIS<sup>1,2</sup>, ALEXANDER CROY<sup>2</sup>, RAFAEL GUTIERREZ<sup>2</sup>, AREZOO DIANAT<sup>2</sup>, and GIANAURELIO CUNIBERTI<sup>2</sup> — <sup>1</sup>Max Planck Institute for the Physics of Complex Systems, 01187 Dresden, Germany — <sup>2</sup>Institute for Materials Science and Max Bergmann Center of Biomaterials, TU Dresden, 01062 Dresden, Germany

Proton tautomerization is a prototype reaction and controllable on a single molecule level. In particular, studies of aromatic molecules, such as porphyrins and phthalocyanines, show the potential of use in molecular electronics, sensor devices and solar cells. Here, we simulate the reaction paths using well-tempered Metadynamics simulations, in molecular dynamic engine of DFTB+. We study the influence of symmetric and asymmetric functionalization on the tautomerization rates and metastable states of porphyrin and porphyrazine cores.

MM 41.3 Wed 16:15 IFW B

**First-principles description of phase transitions in solids with rotating molecules and diffusing atoms** — •SERGEI SIMAK — IFM, Linköping University, Sweden

Solids with rotating molecules and diffusing atoms are examples of dynamically disordered phases of materials with great potential for applications, from fuel cells to optoelectronics. Description of thermodynamics of these materials is a challenge, as standard methods, like static phonon calculations fail due to the large displacements of atoms or the absence of well-defined equilibrium atomic positions. We briefly outline a method that offers a solution to the problem based on a stress-strain thermodynamic integration [1]. An example of lithium carbide, a fascinating material that combines strong covalent and weak

ionic bonding resulting in a wide range of unusual properties, is considered [2]. The mechanism of its phase transition from the ground-state orthorhombic to the high-temperature cubic crystal, which is subject to both rotations of C-C dumbbells and Li self-diffusion, is revealed, and the thermodynamics is described.

[1] J. Klarbring and S. I. Simak, Phys. Rev. Lett. 121, 225702 (2018).

[2] S. Filippov, J. Klarbring, U. Häussermann, and S. I. Simak, Phys. Rev. Materials 3, 023602 (2019).

MM 41.4 Wed 16:30 IFW B

**Development of a Neural Network Potential for Metal-Organic Frameworks** — •MARIUS HERBOLD, MARCO ECKHOFF, and JÖRG BEHLER — Georg-August Universität Göttingen, Institut für Physikalische Chemie, Theoretische Chemie, Tammannstraße 6, 37077 Göttingen, Germany

Metal-organic frameworks (MOFs) are crystalline porous materials with many applications in chemistry and materials science, from gas separation to heterogeneous catalysis. Computer simulations of chemical processes in MOFs are severely limited by the use of classical force fields (FFs), because most FFs are unable to describe bond formation and breaking. In principle, electronic structure methods, like density-functional theory (DFT), can overcome this problem, but often the required systems are too large for routine applications of DFT. Here a high-dimensional neural network potential (NNP) is presented for a series of MOFs, which combines the advantages of both worlds - the accuracy of first principle methods with the efficiency of simple empirical potentials. We demonstrate the possibility to obtain a reliable description of the potential-energy surface of bulk MOFs based on reference calculations of molecular fragments only.

MM 41.5 Wed 16:45 IFW B

**Analysis of organic-inorganic thermal interfaces in Metal-Organic Frameworks** — •SANDRO WIESER<sup>1</sup>, TOMAS KAMENCEK<sup>1</sup>, ROCHUS SCHMID<sup>2</sup>, JOHANNES PETER DÜRHOFT<sup>2</sup>, NATALIA BEDOYA MARTÍNEZ<sup>3</sup>, and EGBERT ZOJER<sup>1</sup> — <sup>1</sup>Institute of Solid State Physics, NAWI Graz, Graz University of Technology, Austria — <sup>2</sup>Chair of Inorganic Chemistry 2, CMC Group, Ruhr-Universität Bochum, Germany — <sup>3</sup>Materials Center Leoben, Austria

Metal-Organic Frameworks (MOFs) represent a type of porous materials that show promising properties for many applications including gas-storage and catalysis, which in most of the cases rely on the effectiveness of dissipating heat. Due to the complex structure of these materials, an in depth understanding of how the fundamental structural characteristics of individual building blocks, as well as their interconnection, impact the system's properties is necessary. To that aim, we investigate isoreticular and tetragonal MOFs utilizing classical non-equilibrium molecular dynamics simulations. The interatomic interactions are described by specifically tailored MOF-FF potentials fitted to density-functional-theory reference data. From the obtained temperature profiles one can conclude that the factor primarily limiting thermal transport is a weak thermal coupling across the interface between the inorganic nodes and the organic linkers combined with a poor conduction within the nodes. We additionally investigate, how the thermal coupling across the heterointerface is impacted by parameters like the mass and extent of the individual components and the docking chemistry.