

## O 12: Poster Session I: Electronic structure theory: General

Time: Monday 10:30–12:30

Location: P

O 12.1 Mon 10:30 P

**Electronic Structure of Cesium-based Photocathode Materials from Density Functional Theory** — ●HOLGER-DIETRICH SASSNICK<sup>1</sup> and CATERINA COCCHI<sup>1,2</sup> — <sup>1</sup>Institut für Physik - Carl von Ossietzky Universität Oldenburg, 26129 Oldenburg, Germany — <sup>2</sup>Institut für Physik - Humboldt-Universität zu Berlin und IRIS Adlershof, 12489 Berlin, Germany

The development of novel materials for vacuum electron sources is an active field of research that can greatly benefit from the results of *ab initio* calculations [1]. Density functional theory offers a good compromise between accuracy and computational feasibility, however, its results crucially depend on the choice of the exchange-correlation potential,  $v_{xc}$ . To address this point, we systematically analyze the performance of three popular approximations of  $v_{xc}$  (PBE, SCAN and HSE06) on the structural and electronic properties of bulk Cs<sub>3</sub>Sb and Cs<sub>2</sub>Te, two representative Cs-based semiconductors employed in photocathode applications. We find that PBE shows expectedly the largest discrepancies from the target, while both SCAN and HSE06 perform remarkably well in reproducing the materials properties. Additionally, we study the effect of spin-orbit coupling which mainly impacts the valence region of both materials inducing a band splitting of about 150 meV. Our results indicate SCAN as the best trade-off between accuracy and computational costs, outperforming the considerably more expensive HSE06 [2].

[1] Cocchi et al, *J. Phys.: Condens. Matter* **2019**, 31, 014002

[2] Saßnick & Cocchi, *submitted 2020*, arXiv:2101.04596

O 12.2 Mon 10:30 P

**Solvated Electrons in Alkali Metal Doped Zeolites: Insights from *Ab-initio* Atomistic Thermodynamics** — ●DEBALAYA SARKER<sup>1,2</sup>, MARIA TROPPEZ<sup>3</sup>, SANTIAGO RIGAMONTI<sup>3</sup>, CLAUDIA DRAXL<sup>3</sup>, SERGEY V LEVCHENKO<sup>1,2</sup>, and MATTHIAS SCHEFFLER<sup>2</sup> — <sup>1</sup>Skoltech, Moscow, RU — <sup>2</sup>Fritz-Haber-Institut der MPG, DE — <sup>3</sup>Humboldt-Universität zu Berlin, DE

Doping nanoporous aluminosilicate zeolites viz. faujasite Y, with alkali metal atoms (M) is a promising way of producing outstanding catalysts. The dopants, along with extra-framework M atoms, often form  $M_4^{3+}$  clusters inside zeolite pores, leaving the valence electron of the dopant solvated and available for catalysis. Despite extensive experimental efforts[1], the distribution of the dopants and solvated electrons remains debated to date. Combining a cluster-expansion model, parameterized with density-functional theory calculations, with *ab-initio* thermodynamics, we address this issue. The electronic structure for low-energy configurations is calculated with the hybrid functional HSE06. We find that even at room temperature, Na atoms in NaY zeolites with 2 extra-framework atoms/unit cell on average redistribute such that areas with lower and higher local concentrations emerge. The redistribution is driven by increased configurational disorder, mainly at higher concentrations. This explains why solvated electrons can be located inside both small and large cages in NaY, reconciling experiments that assign the solvated electrons to a particular pore type.

[1] A. R. Armstrong *et al.*, *J. Am. Chem. Soc.* **117**, 9087 (1995).

O 12.3 Mon 10:30 P

**Hole mobility response through gas adsorption at the inner surface of the iron (1,2,3)-triazolate metal-organic framework** — ●CHRISTOPH MUSCHIELOK<sup>1</sup>, ALEXANDER REINER<sup>2</sup>, RICHARD RÖSS-OHLENROTH<sup>2</sup>, ANDREAS KALYTTA-MEWES<sup>2</sup>, DIRK VOLKMER<sup>2</sup>, ACHIM WIXFORTH<sup>2</sup>, and HARALD OBERHOFER<sup>1</sup> — <sup>1</sup>Technische Universität München — <sup>2</sup>Universität Augsburg

Metal-organic frameworks (MOFs) are hybrid network materials, built from metal centers interconnected by organic linker molecules. Often they exhibit pores, into which atoms or small molecules may be absorbed, and a select few of them even display some electronic conductivity. Utilizing both, we investigate the response of the hole mobility in the iron (1,2,3)-triazolate MOF to guest atoms for a possible use as a trace gas sensor. To deepen our understanding of the material, we employ our adapted variant of Bardeen and Shockley's deformation potential theory to analyze the impact of the guest atoms' presence on the corresponding material properties: the effective mass, the bulk modulus, and the deformation potential of the MOF crystal. We find

significant influence of guest atom absorption on the hole mobility, consistent with first experimental results.

O 12.4 Mon 10:30 P

**First-Principles Study of Lead-free Halide Double Perovskites for Photovoltaic Applications** — ●ELISABETTA LANDINI<sup>1,2</sup>, HARALD OBERHOFER<sup>2</sup>, and KARSTEN REUTER<sup>1</sup> — <sup>1</sup>Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195, Berlin, Germany — <sup>2</sup>Chair for Theoretical Chemistry and Catalysis Research Center, Technische Universität München, Lichtenbergstr. 4, D-85747 Garching, Germany

Metal halide perovskites, such as methylammonium lead iodide [1], are promising materials in the field of optoelectronics. However, the toxicity of lead, hysteresis effects, and the sensitivity of the structures to moisture and heat prevent this technology to become technologically viable. To overcome these limitations, heterovalent substitution of Pb forming the double perovskite structure  $A_2BB'X_6$ , has been suggested.

In this context theoretical modeling is a valuable tool to develop a comprehensive picture of the electronic structure of different double perovskites. Density-functional theory calculations have been carried out using semi-local and screened hybrid functionals and including spin-orbit coupling corrections. Experimentally studied Cs<sub>2</sub>AgBiX<sub>6</sub> (X=Br,Cl) [2] are chosen as a starting point. Native point defects and incremental substitutions are then created to systematically study their effect on the band structure and charge carrier mobility.

[1] D. Zhao *et al.*, *ACS Eng. Lett.* **3**, 305 (2018).

[2] X. Zhao *et al.*, *J. Am. Chem. Soc.* **139**, 2630 (2017).

O 12.5 Mon 10:30 P

**Electronic Structure of Self-assembly Crystalline Perylene in Metal-Organic Frameworks Thin Film** — ●YOHANES PRAMUDYA, MARIANA KOZLOWSKA, SHAHRIAR HEIDRICH, MARIUS JAKOBY, LIUYANG PAN, BRYCE RICHARD, CHRISTOF WÖLL, RITESH HALDAR, and WOLFGANG WENZEL — Karlsruhe Institute of Technology, Eggenstein-Leopoldshafen, Germany

Metal organic frameworks (MOFs) are a very versatile platform for exploring nearly unlimited chemical space of organic components linkers connected by the metal centers or vertices in various topologies and their self-assembly for device optimization. Here, we demonstrated the impact of interchromophore interactions in the photophysical properties of a surface-anchored MOF (SURMOF) based on 3,9-perylenedicarboxylic acid linkers. We predicted the structural assembly of the perylene molecules in the MOF via robust periodic density functional theory calculations and described the impact of unit topology and  $\pi^*\pi$  interaction patterns on spectroscopic and semiconducting properties of the MOF films. We elaborated the dual nature of excited states in the perylene MOF, where strong temperature-modulated excimer emission, enhanced by the formation of perylene J-aggregates, and low stable monomer emission was observed. We used band transport and hopping transport mechanisms to predict the semiconducting properties of perylene SURMOF-2 films as a function of inter-linker interactions and the resulting both p-type and n-type electronic transport mechanisms.

O 12.6 Mon 10:30 P

**Optical and X-ray absorption spectra of oxides: Theoretical description including many-body effects** — ●VIJAYA BEGUM<sup>1</sup>, MARKUS E. GRUNER<sup>1</sup>, CHRISTIAN VORWERK<sup>2</sup>, CLAUDIA DRAXL<sup>2</sup>, and ROSSITZA PENTCHEVA<sup>1</sup> — <sup>1</sup>Faculty of Physics and Centre for Nanointegration, University of Duisburg-Essen, Duisburg. — <sup>2</sup>Institute für Physik and IRIS Adlershof, Humboldt-Universität zu Berlin, Germany.

We present a comprehensive study of the optical and x-ray absorption spectra (XAS) in two paradigmatic oxides – SrTiO<sub>3</sub> (STO) and MgO, from first-principles calculations. Many-body effects were taken into account by a single step  $G_0W_0$ , followed by the excitonic effects by solving the Bethe-Salpeter Equation (BSE). The results demonstrate that the inclusion of the excitonic/electron-(core)hole (e-h) interactions is integral to describe the spectra accurately. For the optical spectra, the influence of different starting exchange-correlation functionals [1,2] is discussed, and the best correspondence with experiment is achieved using a hybrid functional for MgO [2]. The simulated XAS

spectra for the O and Mg K-edge (MgO), and O K-edge (STO) are in excellent agreement with experiment w.r.t. the spectral shape and peak positions. We also analyze the origin of prominent peaks in the spectra and identify the orbital character of the relevant contributions by projecting the e-h coupling coefficients from the BSE eigenvectors on the band structure. The real-space projection of the respective wave functions in MgO shows a Wannier-Mott type lowest energy exciton for the optical spectrum, and a strong localization for the O K-edge. [1] Phys. Rev. Materials **3**, 065004 (2019), [2] arXiv:2012.08960v1

O 12.7 Mon 10:30 P

**Self-energy self-consistent density functional theory plus dynamical mean field theory** — •SUMANTA BHANDARY<sup>1</sup> and KARSTEN HELD<sup>2</sup> — <sup>1</sup>School of Physics and CRANN Institute, Trinity College Dublin, The University of Dublin, Dublin 2, Ireland — <sup>2</sup>Institute of Solid State Physics, TU Wien, 1040 Wien, Austria

We propose a hybrid approach which employs the dynamical mean-field

theory (DMFT) self-energy for the correlated, typically rather localized orbitals and a conventional density functional theory (DFT) exchange-correlation potential for the less correlated, less localized orbitals. We implement this self-energy self-consistent DFT+DMFT scheme[1], in addition to charge self-consistency[2], in a basis of maximally localized Wannier orbitals using Wien2K, wien2wannier, and the DMFT impurity solver w2dynamics. We apply the method to SrVO3 and report a significant improvement as compared to previous d+p calculations. In particular the position of the oxygen p bands is reproduced correctly, which has been a persistent problem in DFT+DMFT before, and has unwelcome consequences for the d-p hybridization as well as for the correlation strength. Taking the (linearized) DMFT self-energy also in the Kohn-Sham equation bypasses the uncertainty of the "double-counting" problem of DFT+DMFT and yields very similar quasiparticle renormalized bands on the "DFT" and "DMFT" side. [1] S. Bhandary and K. Held, arXiv:1904.02967 (2020). [2] S. Bhandary, E. Assmann, M. Aichhorn, and K. Held, Phys. Rev. B **94**, 155131 (2016).