Computational modeling and data science for new material

Session Chair: Ilya Grinberg

Keynote 1: Prof. Leeor Kronik (WIS)

Title: *Defects, defect tolerance, and self-healing in lead halide perovskites: a first principles perspective*

Abstract

Halide perovskites are a promising class of materials for photovoltaic energy conversion and other optoelectronic applications. Among many riddles posed by halide perovskites, the surprising apparent near-absence of harmful defects - in a material prepared by low-cost methods - stands out. This is usually ascribed to defect tolerance or to self-healing. Here, I will explain these perplexing phenomena and present insights into them, gained from first principles calculations. In particular, I will present studies that attempt to explain selected experimental results pertaining to these two phenomena, in terms of specific physical scenarios arising from the underlying chemical behavior. I will also discuss challenges associated with the calculations.

Keynote 2: Prof. Dan Major (BIU)

Title: *Computational studies of structure, composition, and electrochemical behavior of Li-ion battery components*

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Abstract

Considerable efforts have been invested in recent decades to further the improvement of lithium-ion battery components – electrodes, solutions, and separators. In this presentation we will focus on recent computational work in our group on cathodes, anodes, and electrolyte solutions. We will show how computations are an essential part of the analytical toolbox, allowing deeper understanding of physical phenomenon related to energy materials. First, we will discuss studies on layered Ni-rich cathode materials (i.e., Li[Ni-Co-Mn]O2, NCM, family) for lithium-ion batteries.1 Specifically, we will discuss lattice doping with a range of high-valent dopants in NCM materials, which is one of the most promising approaches to improve electrochemical properties, cycle life, and thermal behavior. Second, we will study cation and anion intercalation phenomenon in MXene materials.2 Third, we will show some recent simulations on Water-in-Salt electrolyte solutions.3

References:

1. Layered Cathode Materials for Lithium-Ion Batteries: Review of Computational Studies on LiNi1-x-yCoxMnyO2 and LiNi1-x-yCoxAlyO2. Chakraborty, A.; Kunnikuruvan, S.; Kumar, S.; Markovsky, B.; Aurbach, A.; Dixit, M.; Major, D. T. Chem. Mater. 32, 915-952 (2020).
2. Can anions be inserted into MXene? Shpigel, N.; Chakraborty, A.; Malchik, F.; Bergman, G.; Nimkar, A.; Gavriel, B.; Turgeman, M.; Hong, C.; Lukatskaya, M. R.; Levi, M.; Gogotsi, Y.; Major, D. T.; Aurbach, D. J. Am. Chem. Soc. 143, 12552-12559 (2021).
3. New Cost-effective Aqueous Electrolyte Enables Highly Stable Operation of 2.15V Lithium-Ion Battery. Turgeman, M.; Wineman-Fisher, V.; Malchick, F.; Saha, A.; Bergman, G.; Gavriel, B.; Penki, T. R.; Nimkar, A.; Baranauskaite, V.; Aviv, H.; Fan, T.; Tischler, Y. R.; Levi, M. D.; Noked, M.; Major, D. T.; Shpigel, N.; Aurbach, D. Cell Reports Physical Science 3, 100688 (2022).

Invited 1: Prof. Oswaldo Dieguez (TAU)
Title: Trends of the mean inner potential of solids from computations using density-functional theory
Abstract
The mean inner potential of a solid is the average over a unit cell of the Coulomb potential due to its charges. This magnitude is of relevance, for example, in transmission electron microscopy imaging and holography. It has been computed in the past for a few materials with density-functional theory, typically using all-electron codes. In this work we present a recipe to do so using files produced by a standard projected-augmented wave code (VASP). We have used this method to assess the agreement with experiment regarding the values of the mean inner potential of alumina [1] and graphite [2] in the past. In this talk I will report how we recently exploited its flexibility to understand how trends in the value of the mean inner potential change, for example, depending on composition and surface orientation of elemental crystals.
[1] A. Auslender, M. Halabi, G. Levi, O. Diéguez, and A. Kohn; Ultramicroscopy 198, 18 (2019)
[2] A. Auslender, G. Levi, V. Ezersky, S. Gorfman, O. Diéguez, and A. Kohn, Carbon 179, 288 (2021)

Invited 2: Assist. Prof. Eli Kraisler (HUJI)
Title: *Potentials in density functional theory (DFT): exact properties and advanced approximations*
Abstract
Density functional theory (DFT) is the leading theoretical framework used to describe materials. The most common approach in DFT is that of Kohn and Sham: it describes a material, namely, a system of N interacting electrons, via a fictitious system of N non-interacting electrons subject to an effective potential termed the Kohn-Sham (KS) potential. The KS potential – a central quantity DFT – is not known exactly, and we have to approximate it, accurately and efficiently. We have, however, some 'hints' as to the potential's form. In particular, we know that it exhibits sharp steps in various scenarios, such as dissociation, ionization, excitation and charge transfer. However, these properties are rarely modelled in common approximations.

In this talk I discuss the step structure of the KS potential, highlight the common origin of steps that appear in different situations and underscore the significance of these features, serving the bridge between the real, many-electron energy differences and the fictitious Kohn-Sham energies. I further show that the Pauli potential – a central quantity in orbital-free DFT (OF-DFT) and in the emerging exact electron factorization (EEF) method – exhibits steps, as well. Surprisingly, detailed analytic characterization of the Pauli steps opens the door to accurately approximate also the KS potential. I suggest ways to incorporate potential steps in new approximations to exchange and correlation in DFT.

Contributed 1: Long Nguyen and Guy Makov, Department of Materials Engineering, Ben-Gurion University of the Negev, Beer-Sheva, Israel

Title: *Structural prediction of group IV monochalcogenides and their phase transitions mechanism: Evolutionary algorithm and ab-initio calculations*

Abstract

Monochalcogenides Sn-X and Ge-X (X = O, S, Se, Te) are candidates for future functional materials in semiconductor technologies. Previously, multiple meta-stable states of SnS and SnSe were identified, characterized and synthesized by experimental and theoretical means, indicating a rich phase space beyond their ground state. It was found SnS and SnSe could manifest as a stable nanometric cubic phase or form a new layered structure at ambient conditions [1,2]. Our present study focuses on predicting novel undiscovered structures of GeS, SnS, SnSe, and SnO and determining their optical and mechanical properties. Using a combination of density functional theory (DFT) modelling and a genetic algorithm, we systematically explored the structural diversity and phase stability of these materials under ambient and high-pressure conditions. Multiple structural transitions of Sn-X and Ge-X upon compression were found to be consistent with experimental observations [3,4]. Several novel, stable and yet to be synthesized, phases of Sn-X and Ge-X with attractive electronic and optical properties are predicted. Lone pair delocalization and elastic instability are the main drivers for the pressure-induced phase transitions.

[1] R.E. Abutbul, E. Segev, U. Argaman, G. Makov, and Y. Golan, Adv. Mater., 30(41), 1870310 (2018).

[2] N. Zakay, A. Schlesinger, U. Argaman, L. Nguyen, N. Maman, B. Koren, M. Ozeri, G. Makov, Y. Golan, D. Azulay. ACS Appl. Mater. Interfaces (2023). (submitted).

[3] L.T. Nguyen, G. Makov. Materials 14(21), 6552 (2021).

[4] L.T. Nguyen, G. Makov. ACS Cryst. Growth Des. 22(8), 4956-69 (2022

Contributed 2: H. Kugler (PhD BIU)
Title: *Network-Based Biocomputation Circuits*

Abstract

Engineering biological devices to perform computation is of major interest due to the potential of utilizing inherent parallelism in biological components to speed computation, construct low energy consuming devices and interface with biological material, opening up potential diagnostic and medical applications. Network-Based Biocomputation Circuits (NBCs) offer a new paradigm for solving complex computational problems by utilizing biological agents that operate in parallel to explore manufactured planar devices [1,2]. NBC solves a given combinatorial problem by encoding it into a graphical, molecular network that is embedded in a nanofabricated planar device. These devices then run biological agents through the network, exploring it in parallel, thus solving a given combinatorial problem, including NP-complete problems such as the Subset Sum Problem (Fig. 1) [3]. In addition, due to the utilization of biological agents, this technology could potentially be used to carry cells through the devices and perform complex computational processing with medical and diagnostic applications. An integrated toolset was developed for simulation and verification to enable analysis of circuit correctness and effects of agent stochastic dynamics on circuit behavior [3]. We will discuss challenges still needed to be overcome before NBC can be scaled up to fill a technological niche and reach an industrial manufacturing level[4].

Fig. 1(a) SSP network for set S={2 5 9}. (b) Pass (left) and split (right) junctions.

[1] D.V. Nicolau Jr. et al. National Academy of Sciences, 113(10):2591–2596 (2016).

[2] T. Korten et al. New Journal of Physics, 23(8):085004 (2021).

[3] M. Aluf-Medina et al. VMCAI, volume 12597: 464–485, (2021).

[4] F. van Delft et al. Nano Futures 6 032002, (2022).

Contributed 3: Dr. M. Camarasa-Gómez (Postdoc, WIS)
Title: *Transferability of Screened Range-Separated Hybrid Functionals for Layered Materials*

Abstract

The accurate description of electronic and optical properties is a long-standing challenge in density functional theory. Screened range-separated hybrid (SRSH) functionals have been successfully employed for the determination of gap energies and optical spectra in solids [1, 2]. Generally, the range-separation parameter and fraction of short-range exact exchange that define the SRSH are material- and structure-dependent, raising the question of transferability between 2D and bulk phases of layered materials. Following [3], we explore 2D and bulk phases of various layered materials. We show that systematic determination of SRSH parameters that simultaneously describe different structural phases of the material with quantitative accuracy is possible.

[1] L. Kronik and J. B. Neaton, Ann. Rev. Phys. Chem., 67, 587 (2016).

[2] D. Wing et al., PNAS, 118, e2104556118 (2021).

[3] A. Ramasubramaniam et al., Phys. Rev. Materials, 3, 084007