The IOP-FHI workshop on the frontiers
of electronic-structure theory
and materials genomics

# BOOK OF ABSTRACTS

October 16 - 19 2023 | Beijing · China

**Sponsors:**

Chinese Academy of Sciences
Institute of Physics, CAS
Fritz Haber Institute, MPG
ShangHai University
National Natural Science Foundation of China
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# Organized by

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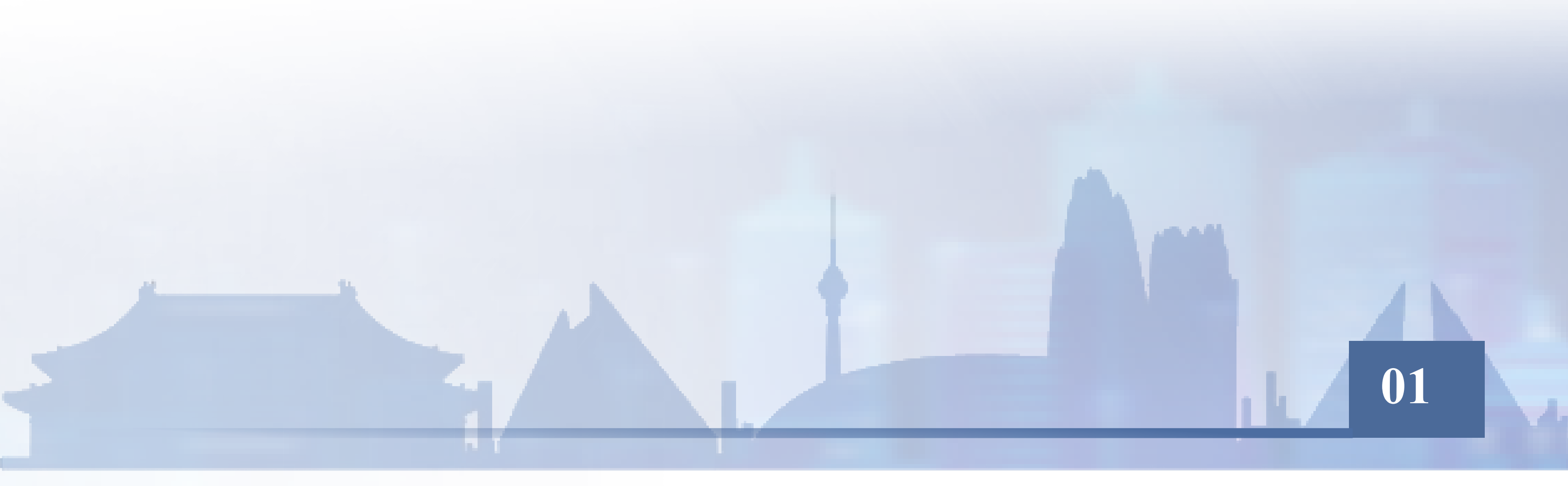
http://iop-fhi-workshop.cpsjournals.cn



# Welcome

Welcome to the *IOP-FHI workshop on the frontiers of electronic-structure theory and materials genomics*. The present workshop intends to bring together world-leading experts in different areas of electronic-structure theory, including developers of density-functional approximations, quantum chemists, many-body physicists, as well as developers of computational software packages, to present their state-of-the-art research work, and discuss open issues of the field. Such an interdisciplinary (albeit with common interest) meeting will provide an excellent platform for exchanging knowledges, stimulating new ideas, and identifying important new research directions.

This workshop is the second edition of the electron structure theory workshops co-organized by the Max Planck Partner Group on Advanced Electronic Structure Methods and the Fritz Haber Institute (FHI). (For more information about the first workshop, see https://nomad.fhi.mpg.de/meetings/faesm2016/index.php). This time, the workshop will also feature sessions on the big-data driven approach to materials science, which has become an actively developed field in recent years. The merger of new developments of electronic-structure methods and big-data/artificial intelligence (AI) approaches will be an important theme of the workshop.

The IOP-FHI workshop is sponsored by the Institute of Physics, Chinese Academy of Sciences, the Duke University, and the Shanghai University.

Enjoy the workshop!

The organizers

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**John Perdew**

**Weitao Yang**

**Igor Ying Zhang**

**Andreas Grüneis**

**Hardy Gross**

**Sheng Meng**

# Program

## Sunday, October 15

16:00 – 20:00: Registration

18:00 – 19:30: Dinner ( 爱丽舍餐厅 )

18:00 – 20:00: Reception

**Metting Place**:
Beijing Grand Skylight Catic Hotel, Beijing Yun-Hai-Ting ( 云海厅 )

## Monday, October 16

## Session I:

08:35 – 08:50
Xinguo Ren & Volker Blum
Opening remarks

08:50 – 09:00
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Chair: Matthias Scheffler (Fritz Haber Institute)

09:00 – 09:35
John Perdew (Tulane University, online)
Unconventional Error Cancellation Explains the Success of Hartree-Fock Density Functional Theory for Barrier Heights

09:35 – 10:10
Weitao Yang (Duke University, online)
Overcoming Delocalization Error for Finite and Bulk Systems with the Localized Orbital Scaling Correction

**10:10 – 10:40 Coffee Break**
Chair: Hong Jiang (Peking University)

10:40 – 11:15
Igor Ying Zhang (Fudan University)
A Cross-Entropy Corrected Hybrid Multiconfiguration Pair-Density Functional Theory for Complex Molecular Systems

11:15 – 11:50
Andreas Grüneis (Vienna University of Technology)
Towards Chemical Accuracy for Surface Chemistry Using Coupled-Cluster Theory

12:00 – 14:00 Lunch (Elysee Western restaurant ( 爱丽舍餐厅)

**Session II:**

Chair: Xingao Gong (Fudan University)

14:00 – 14:35

Hardy Gross (The Hebrew University of Jerusalem, online)

Mechanisms of Decoherence, and Other Lessons from the Exact Factorization

14:35 – 15:10

Sheng Meng (Institute of Physics, CAS)

Laser Control of Transport and Phase Transition

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Lucas Foppa (Fritz Haber Insititute)

Identifying Rules and Materials Genes via Artificial Intelligence

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Rossitza Pentcheva (University of Duisburg-Essen, online)

Designing Quantum Materials for Energy Conversion Applications

16:50 – 17:25

Yi Liu (Shanghai University)

Development and Applications of ReaxFF Reactive Force Fields: From Carbon to Alloy

18:00 – 19:30: Dinner ( 爱丽舍餐厅 )

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## Tuesday, October 17

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Roberto Car (Princeton University, online)

A Study of the Water Self-Ionization Reaction with the Deep Potential Method

09:05 – 09:40

Xinzheng Li (Peking University)

Complex Phase Diagram and Supercritical Matter

09:40 – 10:15

Ding Pan (The Hong Kong University of Science and Technology)

First-Principles and Machine Learning Studies of Water and Carbon in A Large P-T Range

**10:15 – 10:45 Coffee Break, Group Photo**

Chair: Gian-Marco Rignanese (Université Catholique de Louvain)

10:45 – 11:20

Claudia Draxl (Berlin Humboldt University)

From Many-Body Theory to Nonlinear Modeling and Classification

11:20 – 11:55

Miao Liu (Institute of Physics, CAS)

Atomly.net: Advancing Materials Science via High-Performance Computing, Data, and AI

12:00 – 14:00 Lunch ( 爱丽舍餐厅 )

## Session IV:

Chair: Xinzheng Li (Peking University)

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15:10 – 15: 45
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Deep Uncertainties

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16:50 – 17:25
Honghui Shang (University of Science and Technology of China)
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Peter Kratzer (University of Duisburg-Essen)

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Hua Wu (Fudan University)

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Adrienn Ruzsinszky (Tulane University, online)

Linear Response in Space and Time, with Application to Symmetry Breaking

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Sergey Levchenko (Skolkovo Institute of Science and Technology)

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## Session VI:

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14:35 – 15:10
Gian-Marco Rignanese (Université Catholique de Louvain)
Combining the Power of High-Throughput Ab Initio Calculations and Machine Learning towards Materials Informatics

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Volker Blum (Duke University, online)
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18:00 – 19:30
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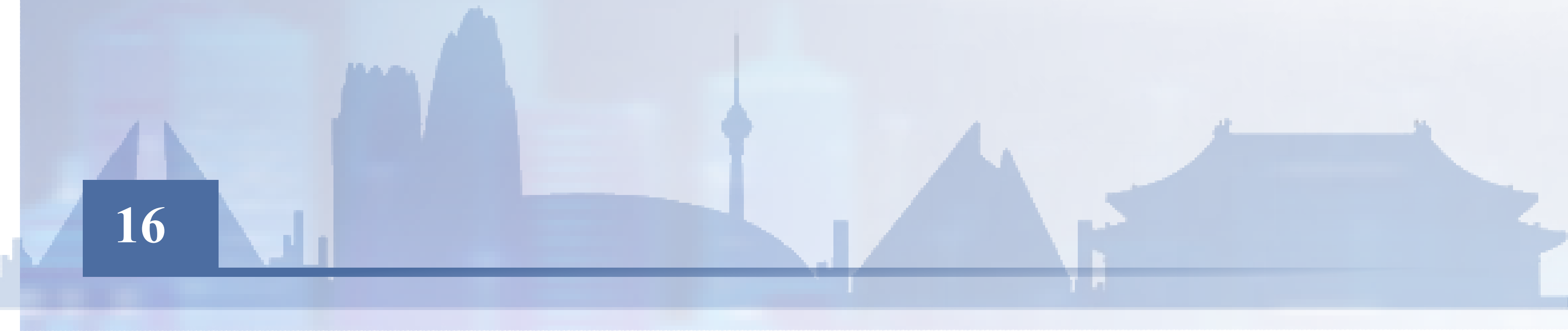

## Special Session:

### 19:30 – 21:30

**Special session dedicated to Prof. Matthias Scheffler**

Chair: Xinzheng Li (Peking University)

(Speakers: Enge Wang, Zhenyu Zhang, Weixue Li, Peter Kratzer, Claudia Draxll.)

## Thursday, October 19

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Non-Fermi-Liquid Metallic Thermoelectrics

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Runhai Ouyang (Shanghai University)

Development of the SISSO Methods for Symbolic Regression in Materials Science

09:40 – 10:15

Hongming Weng (Institute of Physics, CAS)

Quantum Anomalous Hall Insulator from First-principles Calculation

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Chair: Peter Kratzer (University of Duisburg-Essen)

10:45 – 11:20

Weixue Li (University of Science and Technology of China)

Metal-Support Interaction for Supported Nanocatalysts

11:20 – 11:55

Xiaodong Wen (Institute of Coal Chemistry, CAS)

Rational Design of Fe-based Catalysts for Fischer-Tropsch Synthesis from Theoretical Prediction to Experimental Confirmation

11:55 – 12:10 Closing remarks

12:10 – 14:00 Lunch ( 爱丽舍餐厅 )

End of the workshop

2023 The IOP-FHI Workshop

# Abstracts of Speakers

October 16-19, 2023

Beijing·China

Monday, October 16, 09:00-09:35

# Unconventional Error Cancellation Explains the Success of Hartree-Fock Density Functional Theory for Barrier Heights

Bikash Kanungo,[1] Aaron D. Kaplan,[2] Chandra Shahi,[3] Vikram Gavini,[1]
and John P. Perdew,[3]

[1] *Department of Mechanical Engineering, University of Michigan, Ann Arbor, MI, USA*

[2] *Lawrence Berkeley National Laboratory, Berkeley, CA. USA*

[3] *Department of Physics and Engineering Physics, Tulane University, New Orleans, LA, USA*

The energy barrier controls the rate of a chemical reaction. The accuracy of a semi-local density functional approximation has long been known to be boosted strongly for reaction barrier heights by evaluating that approximation non-self-consistently on Hartree-Fock electron densities. The conventional explanation is that Hartree-Fock theory yields the more accurate density. This article presents a benchmark Kohn-Sham inversion of accurate coupled-cluster densities for the reaction $H_2 + F \rightarrow HHF \rightarrow H + HF$, and finds a strong, understandable cancellation between positive (excessively over-corrected) density-driven and large negative functional-driven errors within this Hartree-Fock density functional theory. This work supports the conclusions reached earlier [1] for 76 barrier heights using three less reliable proxies for the exact density. Those proxies, which achieve reasonable electron transfers and reasonably accurate self-consistent barrier heights, are the global hybrid of SCAN with 50% exact exchange, the range-separated LCωPBE hybrid of PBE with exact exchange, and the Fermi-Loewdin-orbital self-interaction correction to SCAN.

## References

[1] A.D. Kaplan, C. Shahi, R.K. Sah, P. Bhetwal, and J.P. Perdew, Understanding Density-Driven Errors for Reaction Barrier Heights, Journal of Chemical Theory and Computation_19, 532 (2023).

Monday, October 16, 09:35-10:10

# Overcoming Delocalization Error for Finite and Bulk Systems with the Localized Orbital Scaling Correction

Weitao Yang

*Department of Chemistry and Department of Physics*

*Duke University, USA*

We will report our development of the localized orbital scaling correction (LOSC) in overcoming systematic delocalization errors in commonly used density functional approximations (DFAs).LOSC is capable of correcting system energy, energy derivative and electron density in a size-consistent manner for all commonly used density functional approximations (DFAs). LOSC leads to systematically improved results, including the dissociation of ionic species, the band gaps of molecules, polymer chains and bulk systems, the energy and density changes upon electron addition and removal, and photoemission spectra, and energy-level alignments for interfaces. The inclusion of orbital relaxation, or screening, improve LOSC description of quasiparticle energies and band gaps for finite molecules, and is critical for bulk systems. It thus provides a single approximate functional that corrects the systematic errors of DFAs in total energy, electron density, and quasiparticle energies and band gaps, for finite and bulk systems.

The LOSC DFA orbital energies are excellent approximations to quasiparticle energies, comparable to or better than GW. This leads to the QE-DFT (quasiparticle energies from DFT) approach: the calculations of excitation energies of the N-electron systems from the ground state DFA calculations of the (N - 1) electron systems. Results show good performance with accuracy similar to TDDFT for valence excitations with commonly used DFAs with or without LOSC. For charge transfer and Rydberg states, good accuracy was obtained only with the use of LOSC DFA. The QE-DFT method has been further developed to describe excited-state potential energy surfaces (PESs), conical intersections, and the analytical gradients of excited-state PESs. We have also made the LOSC software available for the community.

## References

[1]J. Cohen, P. Mori-Sanchez, and W. Yang. Insights into current limitations

of density functional theory. Science, 321:792, 2008.

[2]P. Mori-Sánchez, A. J. Cohen, and W. Yang, “Localization and Delocalization Errors in Density Functional Theory and Implications for Band-Gap Prediction,” Physical Review Letters, 100: 146401, 2008.

[3]P. Mori-Sanchez, A. J. Cohen, and W. Yang. Discontinuous Nature of the Exchange-Correlation Functional in Strongly Correlated Systems, Physical Review Letters, 102:066403, 2009.

[4]J. Cohen, P. Mori-Sanchez, and W. Yang. Challenges for Density Functional Theory. Chem. Rev. 112:289, 2012

[5]Zheng, X.; Cohen, A. J.; Mori-Sánchez, P.; Hu, X.; Yang, W. Improving Band Gap Prediction in Density Functional Theory from Molecules to Solids. Physical Review Letters 2011, 107 (2).

[6]Zhang, D.; Zheng, X.; Li, C.; Yang, W. Orbital Relaxation Effects on Kohn– Sham Frontier Orbital Energies in Density Functional Theory. The Journal of Chemical Physics 2015, 142 (15), 154113.

[7]C. Li, X. Zheng, N. Q. Su, and W. Yang, “Localized orbital scaling correction for systematic elimination of delocalization error in density functional approximations,” National Science Review, 5: 203–215, 2018.

[8]Y. Mei, C. Li, N. Q. Su, and W. Yang, “Approximating Quasiparticle and Excitation Energies from Ground State Generalized Kohn-Sham Calculations,” arXiv:1810.09906 2018; J. Phys. Chem. A, 123(3), 666–673, 2019

[9]Y. Mei and W. Yang, “Charge transfer excitation energies from ground state density functional theory calculations,” J. Chem. Phys., 150, 144109, 2019.

[10]Y. Mei and W. Yang, “Excited-State Potential Energy Surfaces, Conical Intersections, and Analytical Gradients from Ground-State Density Functional Theory,” J. Phys. Chem. Lett. 10, 2538–2545, 2019.

[11]Mei,Y.; Chen, Z.; Yang, W. Self-Consistent Calculation of the Localized Orbital Scaling Correction for Correct Electron Densities and Energy-Level Alignments in Density Functional Theory. J. Phys. Chem. Lett., 11 (23), 10269, 2020

[12]Mei, Y.; Yu, J.; Chen, Z.; Su, N. Q.; Yang, W. LibSC: Library for Scaling Correction Methods in Density Functional Theory. J. Chem. Theory Comput. 2022, 18 (2), 840–850.

Monday, October 16, 10:40-11:15

# A Cross-Entropy Corrected Hybrid Multiconfiguration Pair-Density Functional Theory for Complex Molecular Systems

Igor Ying Zhang

*Fudan University*

Hybrid density functionals, such as B3LYP and PBE0, have achieved remarkable success by substantially improving over their parent methods, namely Hartree-Fock and the generalized gradient approximation, and generally outperforming the more expensive second-order Møller-Plesset perturbation theory (MP2). Here, we extend the linear scheme of hybrid multiconfiguration pair-density functional theory (HMC-PDFT) by incorporating a cross-entropy ingredient to balance the description of static and dynamic correlation effects, leading to a consistent improvement on both exchange and correlation energies. The B3LYP-like translated on-top functional (tB4LYP) developed along this line not only surpasses the accuracy of its parent methods, the complete active space self-consistent field (CASSCF) and the original MC-PDFT functionals (tBLYP and tB3LYP), but also outperforms the widely used complete active space second-order perturbation theory (CASPT2). Remarkably, while remaining satisfactory for general purpose, tB4LYP shows superior performance for challenging cases like the Cr2 dissociation and the associated low-lying vibrational energies, the ethylene torsional rotation and the ethyne diabatic colinear dissociations. With the significantly lower computational cost than CASPT2, tB4LYP promises an accurate yet efficient treatment for complex molecular systems where dynamic and static correlation effects are equally significant.

Monday, October 16, 11:15-11:50

# Towards Chemical Accuracy for Surface Chemistry Using Coupled-Cluster Theory

A .Grüneis[1], A. Irmler[1], T. Schäfer[1], F. Hummel[1], N. Masios[1] and A. Gallo[1]
*[1]TU Wien, Institut für Theoretische Physik*
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Coupled cluster theory is widely used in high-accuracy ab initio calculations in the field of molecular quantum chemistry to produce reliable benchmark energies and related properties. Reliable benchmark results can be used to resolve existing discrepancies between theory and experiment or to improve more approximate methods, such as density functional theory using approximate functionals. Recent methodological developments have significantly expanded the scope of periodic coupled cluster theory to study solids and surfaces. This talk will review recent methodological developments, such as its extension to the study of metallic systems[1]. The discussed applications will cover a wide range of materials science problems including the study of defects in alkaline earth oxides[2], CO adsorption on MgO[3] and $CO_2$ adsorption in porous materials[4]. These calculations and the comparison to experiment and other theories including quantum Monte Carlo and approximate density functionals often enables to reach consensus for important materials properties.]

**References:**

[1]N. Masios, A.Irmler, T. Schäfer, A. Grüneis. https://arxiv.org/abs/2303.16957 , accepted in PRL (2023)
[2]A. Gallo, F. Hummel, A. Irmler, A. Grüneis, Journal of Chemical Physics 154 (2021) 064106
[3]B. X. Shi, A. Zen, V. Kapil, P. R. Nagy, A. Grüneis and A. Michaelides, https://doi.org/10.26434/chemrxiv-2023-h4czl-v2 (2023)
[4]B. Herzog, A. Gallo, F. Hummel, M. Badawi,T. Bučko, S. Lebègue, A. Grüneis and D. Rocca, https://doi.org/10.26434/chemrxiv-2023-mvsxn (2023)

Monday, October 16, 14:00-14:35

# Mechanisms of Decoherence, and Other Lessons from the Exact Factorization

Hardy Gross

*Fritz Haber Center for Molecular Dynamics, The Hebrew University of Jerusalem, Israel*

A prominent goal of present-day condensed-matter physics is the design of electronic devices with ever faster switching times. As an example, the optically induced spin transfer between magnetic sublattices, the so-called OISTR effect, will be presented. This effect allows the switching of magnetic textures on the scale of a femto-second or less. It was first predicted with real-time TDDFT and later confirmed in several experiments. To create from this effect a real-world device, one has to face the problem of decoherence, i.e. the phenomenon that quantum systems tend to lose their “quantumness” due to interactions with other degrees of freedom. Decoherence is responsible for preventing genuine quantum computing at useful scales to this day. For electrons, the principal source of decoherence is the non-adiabatic interaction with nuclear degrees of freedom, i.e. with an “environment” that cannot be removed or reduced. In fact, the paradigm of electronic-structure theory where electrons move in the static Coulomb potential of clamped nuclei is an idealization hardly ever satisfied in nature when systems are manipulated/excited/switched. Non-adiabaticity, i.e. effects of the coupled motion of electrons and nuclei beyond the Born-Oppenheimer approximation are found everywhere. In this lecture, the exact factorization will be presented as a universal approach to understand and, ultimately, control non-adiabatic effects, in particular decoherence, from an ab-initio perspective.

Monday, October 16, 14:35-15:10

# Laser Control of Charge Transport and Phase Transitions

Sheng Meng
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Photoexcitation is a powerful means in distinguishing different interactions and manipulating the properties of matter, especially for charge transport and phase transitions in complex quantum systems.

Here we show that laser-controlled coherent phonon excitation enables orders of magnitude enhancement of carrier mobility via accelerating polaron transport in a prototypical material, lithium peroxide (Li2O2). The selective excitation of specific phonon modes, whose vibrational pattern directly overlap with the polaronic lattice deformation, can remarkably reduce the energy barrier for polaron hopping. The strong nonadiabatic couplings between the electronic and ionic subsystem play a key role in triggering charge transport. These results extend our understanding of charge transport dynamics to the nonequilibrium regime and allow for optoelectronic devices with ultrahigh on-off ratio and ultrafast responsibility competitive with those of state-of-the-art devices. We also investigate photoexcitation induced ultrafast phase-transition dynamics in two-dimensional materials, where we identify a laser-induced collective pathway from 2H phase to 1T phase in MoTe2 that is significantly different from thermal phase transitions. Our results provide insights from a new perspective on the coherent electron and lattice quantum dynamics in materials upon photoexcitation.

**References:**

[1]H.M. Wang, X.B. Liu, S.Q. Hu, D.Q. Chen, Q. Chen, C. Zhang, M.X. Guan, S. Meng. Giant acceleration of polaron transport by ultrafast laser-induced coherent phonons. Science Advances 9, eadg3833 (2023).

[2]R.J. Zhao, P.W. You, S. Meng, Ring polymer molecular dynamics with electronic transitions, Phys. Rev. Lett. (2023).

[3]C.Y. Wang, X.B. Liu, Q. Chen, D.Q. Chen, Y.X. Wang, S. Meng. Coherent-Phonon-Driven Intervalley Scattering and Rabi Oscillation in Multivalley 2D Materials. Phys. Rev. Lett. 131, 066401 (2023).

[4]M.X. Guan, X.B. Liu, D.Q. Chen, X.Y. Li, Y.P. Qi, Q. Yang, P.W. You, S.

Meng. Optical control of multi-stage phase transition via phonon couplings inMoTe2. Phys. Rev. Lett. 128, 015702 (2022).

[5]J.Y. Xu, D.Q. Chen, S. Meng. Decoupled ultrafast electronic and structural phase transitions in photoexcited monoclinic VO2. Science Advances 8, eadd2392 (2022).

Monday, October 16, 15:10-15:45

# Identifying Rules and Materials Genes via Artificial Intelligence

Lucas Foppa
*Institute of Physics, Chinese Academy of Sciences, Beijing 100190*
Email: smeng@iphy.ac.cn

The performance in heterogeneous catalysis is an example of a complex materials function, governed by an intricate interplay of several processes such as the surface chemical reactions, the dynamic restructuring of the catalyst material, and the transport of reactants and products. Modelling the full catalytic progression via first-principles statistical mechanics is impractical, if not impossible. Instead, we show here how artificial intelligence can be applied to model catalysis and determine the key descriptive parameters ("materials genes") reflecting the processes that trigger, facilitate, or hinder catalyst performance.[1] This data-centric approach is based on consistent experimental and computational data characterizing the catalyst materials, possible underlying processes, as well as the performance, and it can be applied even to the small number of materials that are accessible by experiment.[1,2,3]

**References:**

[1] L. Foppa, L. M. Ghiringhelli, F. Girgsdies, M. Hashagen, P. Kube, M. Hävecker, S. Carey, A. Tarasov, P. Kraus, F. Rosowski, R. Schlögl, A. Trunschke, and M. Scheffler, MRS Bull. 46, 1 (2021).
[2] L. Foppa, F. Rüther, M. Geske, G. Koch, F. Girgsdies, P. Kube, S. Carey, M. Hävecker, O. Timpe, A. Tarasov, M. Scheffler, F. Rosowski, R. Schlögl, and A. Trunschke, J. Am. Chem. Soc. 145, 3427 (2023).
[3] L. Foppa, C. Sutton, L. M. Ghiringhelli, S. De, P. Löser, S.A. Schunk, A. Schäfer, and M. Scheffler, ACS Catal. 12, 2223 (2022).

Monday, October 16, 16:50-17:25

# Development and Applications of ReaxFF Reactive Force Fields: From Carbon to Alloy

Yi Liu

*Materials Genome Institute, Shanghai University*

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Reactive force field can describe large-scale chemical reactions in condensed phase or material deformation and fracture efficiently. We introduce the development of new generation of ReaxFF reactive force fields for carbon allotropies (ReaxFFC-S22) and Ni-Al alloys (ReaxFFNiAl-S22). ReaxFFC-S22 can describe various carbon allotropies with better accuracy than the existing force fields in literatures. ReaxFFNiAl-S22 performs better than the previous ReaxFF and EAM potentials. More importantly, ReaxFF alloy potentials explicitly include electrostatic interactions into molecular simulations of alloys. Several case studies are introduced as validation of the force fields developed in this work.

Tuesday, October 17,08:30-09:05

# A study of the Water Self-Ionization Reaction with the Deep Potential Method

Roberto Car

*Princeton University, Princeton NJ08544, USA*

Water self-ionization, a prototypical reaction in solution, is the most fundamental acid-base reaction in aqueous solutions. Computer simulations of the process are challenging because bond breaking requires quantum mechanics, the event is rare, and the ionic products convert frequently into solvating molecules. To date, the roles of energy and entropy in the process have not been quantitatively established. A deep potential model combined with state-of-the-art enhanced sampling methods can address these issues when electrostatic interactions are properly included. Calculations of the dissociation free energy requires relatively large simulation boxes due to restructuring of the hydrogen bond network and increased local order around the ions extending over three solvation shells. Entropy sets the ionic products free when their separation exceeds 8 A.

Tuesday, October 17,09:05-09:40

# Complex Phase Diagram and Supercritical Matter

Xiaoyu Ouyang, Qijun Ye, and Xin-Zheng Li (speaker)
*School of Physics, Peking University, Beijing 100871, P. R. China*
Email:xzli@pku.edu.cn

A theorical framework for descriptions of supercritical phenomena is provided, accessible to experiments and numerical simulations, using an extension of the Lee-Yang (LY) theory. We found that the Widom line is determined by the complex zeros of partition function, i.e., LY zeros, whilst the conventional phase diagram just manifests a tip of the iceberg underlying the complex phase diagram formed by these LY zeros. The complex phase diagram offers a unified picture of phase transition and crossover phenomena, which is supported by theoretical calculations and molecular simulations on two prototype supercritical model/matters, i.e., vdW fluid and water. When the complex values of the LY zeros are projected onto the physical (real) plane, response properties such as heat capacity and compression coefficient reach their maximum [1]. If time allows, I will also explain some details of a related theory for descriptions of the dynamical states in condensed matter [2-4].

**References:**

[1]X. Y. Ouyang, Q. J. Ye, and X. Z. Li, arXiv:2303.16784
[2]L. Zhuang, Q.-J. Ye, D. Pan, and X.-Z. Li, Chin. Phys. Lett. 37, 043101 (2020).
[3]Q.-J. Ye, L. Zhuang, and X.-Z. Li, Phys. Rev. Lett. 126, 185501 (2021).
[4]Q.-J. Ye and X.-Z. Li, SCPMA 66, 227212 (2023).

Tuesday, October 17, 09:40-10:50

# First-Principles and Machine Learning Studies of Water and Carbon in A Large P-T Range

Ding Pan

*Hong Kong University of Science and Technology*

In this talk, we will introduce a series of interesting carbon reactions in the bulk and nanoconfined water solutions studied by first principles and machine learning methods. To keep a strong connection with experiments, we calculated the Raman and infrared spectra of aqueous solutions in a large P-T range. Our results can be used to interpret the related experimental data, and our studies have great implications for carbon mineralization and deep carbon cycle.

Tuesday, October 17, 10:45 – 11:20

# From Many-Body Theory to Nonlinear Modeling and Classification

Claudia Draxl

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Light-matter interaction is an exciting playground to explore a wide variety of physical phenomena. Here, many-body perturbation theory (MBPT) is the state of-the-art approach to describe charged and neutral excitations in crystalline materials. With selected examples, I will show how these methods can be used to understand the interplay between electron-electron, electron-hole, and electron-phonon interactions in the excitation spectra of wide-gap materials. Here, a most promising candidate for optoelectronic applications is the oxide perovskite BaSnO3. While we can achieve full understanding and excellent agreement with experiment for the ideal crystal, the situation is much more complicated for “real” materials that are prone to defects like oxygen vacancies. Understanding their behavior from first principles alone, is hampered by the numerous configurations as a function of defect concentration and the huge unit cells required to describe them. In this work, we combine a novel approach to cluster expansion (CE) with machine-learning techniques toward nonlinear modeling and classification such to overcome these problems. We demonstrate our approach by addressing the stability and electronic structure of BaSnO3.

Tuesday, October 17, 14:35 – 15:10

# Electron-Phonon Interactions in Thermoelectric Half-Heusler Compounds

Jinyang Xi, Jiong Yan*

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Electron-phonon interaction (EPI) is a commonly existed intrinsic process for materials, and have strong influence on the multiple properties, such as electrical and thermal transport properties. EPI can not only scatter the transport of electrons or holes which is fully understood by the thermoelectric community, but also scatter the transport of phonons at high carrier concentrations. Furthermore, EPI can also alter the band structures, known as the electron-phonon renormalization, and in turn, affect the transport of electrons. In this talk, we will present the aforementioned effects of EPI, exemplified in the rmoelectric half-Heusler compounds. We systematically study the reduction of lattice thermal conductivity of 18 half-Heusler compounds by EPI. The largest reduction at 300 K under the carrier concentration $10^{21}$ $cm^{-3}$ is 32%, found in p-type TiCoSb. The high carrier concentration, large effective mass or density of states at the Fermi level, and large electron-phonon coupling matrix are favorable to a high reduction rate by EPI.[1] Further calculations show that, in order to have a better agreement with experiments, the phonon alterations by the dopants should be included.[2] The band renormalization in half-Heusler compounds by EPI is also studied in a high-throughput manner.[3] Most of the entries show the Varshni effect, i.e., the band gap decreasing with temperature increase, while 4 entries violate this rule. A favorable band renormalization--band convergence by the temperature--is also identified.

**References:**

[1] Materials Today Physics, 31, 100993, 2023
[2] Nature Physics, DOI: 10.1038/s41567-023-02188-z
[3] Scientific Data, 8, 236, 2021

Tuesday, October 17, 15:10 – 15: 45

# Deep Uncertainties

Luca Ghiringhelli
*Fritz Haber Institute*

Data-centric models provide statistical predictions on the basis of the empirical data they are trained on. Due to the uncertainties, of various origins, present in the data and the very likely incompleteness of the input descriptors, such models have statistical uncertainties, which are often not easy to quantify in a reliable fashion. The amount of reliability needed from the uncertainty quantification (UQ) is obviously related to the use one wants to make of it.

Here, I show an example of a deep Bayesian classifier trained on perfect crystal structures, where its UQ is successfully exploited to identify and characterize grain boundaries.

In contrast, I show the example of a deep graph network trained on atomistic environments to predict energies and forces, where its ensemble-based UQ is found to be not accurate enough for performing automatic global potential-energy searches.

Tuesday, October 17, 16:15 – 16:50

# Ab Initio Tight Binding Model and Its Applications in Non-Linear ptics

Lixin He
*School of Physics, University of science and technology of china*

In this talk, I will introduce the PYATB code developed by our team. PYATB is designed to study the wide range of properties of materials based on ab initio tight-binding Hamiltonian. PYATB can directly generate tight-binding models by interfacing with ABACUS, a first-principles calculation software based on numerical atomic orbital basis set, which avoids the tedious process of generating maximally localized Wannier functions. PYATB can efficiently calculate band structures and related properties, including the topological and linear and nonlinear optical properties. We studied the shift current effects in the ferroelectric SnTe monolayer using the code. We found that there is a giant nonlinear shift current effect in SnTe monolayer, but its linear optical response is extremely weak, resulting in an enormous Glass coefficient for this material. We analyzed the mechanism behind this giant shift current effect and found that it is caused by singularities in the Riemann space composed of the optical transition matrix, which lead to divergence of the shift vector. The singularity in the excited state space can be analogous to the Wyle points in the ground-state band structure.

Tuesday, October 17, 16:50 – 17:25

# Direct Solving The Many-Electron Schrödinger‘s Equation with a Language Model

Honghui Shang
*University of Science and Technology of China*

Accurately solving the equation for intricate systems remains a prominent challenge in physical sciences. A paradigm-shifting approach to address this challenge involves the application of artificial intelligence techniques. In this study, we introduce a machine-learning model named QiankunNet, based on the transformer architecture employed in language models. By incorporating the attention mechanism, QiankunNet adeptly captures intricate quantum correlations, which enhances its expressive power. The autoregressive attribute of QiankunNet allows for the adoption of an exceedingly efficient sampling technique to estimate the total energy, facilitating the model training process. Additionally, performance of QiankunNet can be further improved via a pre-training process. This work not only demonstrates the power of artificial intelligence in quantum mechanics but also signifies a pivotal advancement in extending the boundary of systems which can be studied with a full-configuration-interaction accuracy.

Tuesday, October 17, 17:25 – 18:00

# Unlocking The Power of The Variational Free-Energy Principle with Deep Generative Models

Lei Wang
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At finite temperatures, Nature tries to balance energy and entropy to achieve minimal free energy. Ironically, it was rather difficult to turn such a free energy minimization principle into a practical algorithm, especially for quantum matter consisting of fermions. Such a difficulty is mainly due to the prohibitive calculation of the entropy term in the free energy, also known as the "intractable" partition functions problem. One can harness the free energy minimization principle by employing tractable deep generative models such as the normalizing flow and autoregressive network as variational ansatz. For classical statistical mechanics problems, this is a new development that "inherits the glorious tradition"; while for the quantum many-body problems, it is expected to become a unique technique that is "one of a kind". I shall illustrate these developments with concrete examples ranging from spin glasses to uniform electron gases. Finally, the latest application of the deep variational free energy approach to dense hydrogen may provide new insights into this fundamental state of matter.

Wednesday, October 18, 08:30 – 09:05

# Computational Study of Magnetically Ordered Two-Dimensional Materials

Peter Kratzer

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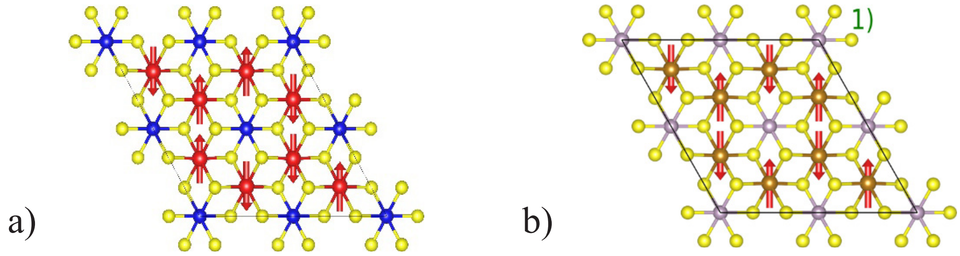
In recent years, the discovery of two-dimensional semiconducting materials and semi-metals has revolutionised semiconductor physics. Some of the most interesting representatives of this materials class combine semiconducting properties with magnetic order down to atomically thin layers. These materials ideally lend themselves to utilization in future spintronics devices.

In my talk, I will present computational studies of two such materials classes. We carry out first-principles calculations using density functional theory (DFT) including a 'Hubbard U' term to account for the on-site Coulomb repulsion between electrons. The semiconductors CrI3 and CrBr3 display ferromagnetic coupling of the Cr magnetic moments within one atomic layer. The coupling from one layer to the next is much weaker and can be either ferromagnetic or antiferromagnetic, depending on details of the atomic structure and the number of stacked layers [1,2].

The second class of materials, mixed phosphorus sulphides with the formula MPS3 (M= Mn, Fe, Co, Ni) display antiferromagnetic coupling between the transition metal atoms within a layer. The magnetic order can be either of the Neel type, such as in MnPS3, or display zig-zag chains of alternating spins, coupled ferromagnetically to each other [3], such as in $FePS_3$, $CoPS_3$ and $NiPS_3$. In my research group, we use DFT+U calculations to obtain the electronic band structure, the magnetic exchange couplings, critical temperatures and magnon spectra.



**Antiferromagnetic order a)** as zig-zag chains in $FePS_3$, **b)** as Neel order in $MnPS_3$

**References:**

[1]S. Sarkar and P. Kratzer, Phys. Rev. Mater. 4, 104006 (2020).

[2]S. Sarkar and P. Kratzer, Phys. Rev. B 103, 224421 (2021).

[3]M. Amirabbasi and P. Kratzer, Phys. Rev. B 107, 024401 (2023).

Wednesday, October 18, 09:05 – 09:40

# Giant Magnetic Anisotropy of Surface Adatoms and 2D Materials

Hua Wu

*Department of Physics, Fudan University, Shanghai, China*

Magnetic anisotropy (MA) is crucial for low-dimensional magnetism and nanoscale magnetic devices. We start from our previous prediction of a giant MA for Co, Ru, and Os adatoms on MgO (001) surface, which is due to a joint effect of the special ligand field, orbital multiplet, and significant spin-orbit interaction [1]. For the emergent two-dimensional (2D) magnetic materials, strong magnetic coupling and anisotropy are both highly desirable. Here we provide an example showing that a strong single ion anisotropy (SIA) makes $VI_3$ a possible first 2D Ising ferromagnetic (FM) semiconductor [2], which is supported by several experiments. Then, by referring to a set of specific spin-orbital states which could favor the Ising magnetism, we figure out $FeS_2$ monolayer is in an unusual high valence but low-spin state, both of which yield strong FM exchange and huge SIA and then make $FeS_2$ a high $T_C$ 2D Ising ferromagnet [3]. Our discussion is based on the pictures of spin-orbital physics, DFT calculations, Monte Carlo simulations, and renormalized spin-wave theory.

**References:**

[1] Xuedong Ou, Hongbo Wang et al., PRL 115, 257201 (2015).

[2] Ke Yang et al., PRB Rapid 101, 100402 (2020).

[3] Ke Yang, Yaozhenghang Ma et al., submitted.

Wednesday, October 18, 09:40 – 10:15

# Linear Response in Space and Time, with Application to Symmetry Breaking

Adrienn Ruzsinszky
*Department of Physics and Engineering Physics*
*Tulane University*

In the framework of linear response time-dependent density functional theory (LR-TDDFT), the exact exchange-correlation (xc) kernel $f_{xc}(q, \omega)$ determines the ground-state energy, excited-state energies, lifetimes, and the time-dependent linear density response of any homogeneous many-electron system. The recently developed dynamic MCP07 exchange-correlation kernel delivers excellent quality correlation energies for the ground state, it predicts a plausible finite plasmon lifetime at all wavevectors for the first time within TDDFT [1-3].

Away from and the correlation contribution to the kernel becomes more relevant and dominant over exchange. The dielectric function in principle can encompass xc effects relevant to describe low-density physics in the homogeneous electron gas. Furthermore, beside collective plasmon excitations, the dielectric function can reveal collective electron-hole excitations, often dubbed “ghost excitons” [4]. Beside collective excitons, the physics in the low-density regime is rich, as exemplified by a static charge-density wave that was recently found for $r_s$>69 with the MCP07 kernel and was shown to be associated with softening of the plasmon mode.

In this talk I will give a detailed view of xc model kernels for excitations of various kinds in the low-density homogeneous electron gas [4] as preliminary results toward the extension of our work to real materials with broken symmetry [5].

Work was supported by DOE-BES under Award Number DE-SC0021263 and by Tulane University’s startup funds.

**References:**

[1] A. Ruzsinszky, N.K. Nepal, J.M. Pitarke, J.P. Perdew, Phys. Rev. B 101, 245135 (2020)

[2] J.P. Perdew, A. Ruzsinszky, J. Sun, N. K. Nepal, and A. D. Kaplan, Proceedings of the National Academy of Sciences of the United States of America, 118 (4) (2021)

[3] A.D. Kaplan, N.K. Nepal, A. Ruzsinszky, P. Ballone, J.P. Perdew, Phys. Rev. B 105, 035123 (2022)

[4] A.D. Kaplan, A. Ruzsinszky, J. Chem. Theo. Comp. submitted, arXiv preprint arXiv:2308.15392.

[5] H. Tang A. Ruzsinszky, in preparation.

Wednesday, October 18, 10:45 – 11:20

# Closest Wannier Functions to A Given Set of Localized Orbitals

Taisuke Ozaki

*ISSP, the Univ. of Tokyo*

Wannier functions (WFs) play a pivotal role in analyzing the electronic structures of real materials and in furthering electronic structure methods, alongside density functional theory (DFT) and other electronic structure theories [1]. In this talk, I will present a novel method to calculate the closest Wannier functions (CWFs) to a given set of localized guiding functions, such as atomic orbitals, hybrid atomic orbitals, and molecular orbitals, based on minimization of a distance measure function [2]. It is shown that the minimization is directly achieved by a polar decomposition [3] of a projection matrix via singular value decomposition, making iterative calculations and complications arising from the choice of the gauge irrelevant. The disentanglement of bands is inherently addressed by introducing a smoothly varying window function and a greater number of Bloch functions, even for isolated bands. In addition to atomic and hybrid atomic orbitals, we introduce embedded molecular orbitals in molecules and bulks as guiding functions, and demonstrate that the Wannier interpolated bands accurately reproduce the targeted conventional bands of a wide variety of systems including Si, Cu, the TTF-TCNQ molecular crystal, and a topological insulator of $Bi_2Se_3$ [4]. We further show the usefulness of the proposed method in calculating effective atomic charges. These numerical results not only establish our proposed method as an efficient alternative for calculating WFs, but also suggest that the concept of CWF can serve as a foundation for developing novel methods to analyze electronic structures and calculate physical properties.

**References:**

[1] N. Marzari and D. Vanderbilt, Phys. Rev. B 56, 12847 (1997).
[2] T. Ozaki, arXiv:2306.15296.
[3] K. Fan and A. J. Hoffman, Proc. Amer. Math. Soc. 6, 111 (1955).
[4] https://www.openmx-square.org/cwf/

Wednesday, October 18, 14:00 – 14:35

# Predictive Design of Two-Dimensional Topological Superconductors

Zhenyu Zhang
*International Center for Quantum Design of Functional Materials (ICQD)*
*University of Science and Technology of China*
Email: zhangzy@ustc.edu.cn)

Discovery of new superconductors with high transition temperatures has been a perpetual drive of condensed matter physics. The recent marriage of superconductivity and topology in the emerging field of topological superconductivity has further expanded the scope to multiple disciplines with conceptual new challenges and technological incentives. In this talk, I will attempt to give an overview on recent advances in this vibrant area, with some of own stories squeezed in. We start from predictive designs of freestanding or supported superconducting monolayers that exhibit high-$T_c$ superconductivity, as well as monolayered systems that display intrinsic *p-wave* superconductivity or coexistence of superconductivity and nontrivial band topology. On the mechanistic side, we demonstrate how plasmonic excitations or pronounced correlation effects can enhance the superconductivity of iron-based superconductors and beyond. We will also highlight three dominant routes to achieving topological superconductivity, and explore ways to gain their tunability.

Wednesday, October 18, 14:35 – 15:10

# Combining the Power of High-Throughput Ab Initio Calculations and Machine Learning towards Materials Informatics

Gian-Marco Rignanese

The progress in first-principles simulation codes and supercomputing capabilities have given birth to the so-called high-throughput (HT) ab initio approach, thus allowing for the identification of many new compounds for a variety of applications (e.g., lithium battery and photovoltaic). As a result, a number of databases have also become available online, providing access to various properties of materials, mainly ground-state though. Indeed, for more complex properties (e.g., linear or higher-order responses), the HT approach is still out of reach because of the required CPU time. To overcome this limitation, machine learning approaches have recently attracted much attention in the framework of materials design.

In this talk, I will review recent progress in the emerging field of materials informatics. I will briefly introduce the OPTIMADE API [1] that was developed for searching the leading materials databases (such as AFLOW, the Materials Cloud, the Materials Project, NOMAD, OQMD, ...) using the same queries. I will introduce the MODNet framework and its recent developments for predicting materials properties [2-5]. This approach, which is particularly well suited for limited datasets, relies on a feedforward neural network and the selection of physically meaningful features. Finally, I will illustrate the power of materials informatics, combining high-throughput ab initio calculations and machine learning, through a few recent examples.

**References**:

[1] C.W. Andersen et al., Sci. Data 8, 217 (2021).

[2] P.-P. De Breuck, G. Hautier, and G.-M. Rignanese, npj Comput. Mater. 7, 83 (2021).

[3] P.-P. De Breuck, M.L. Evans, and G.-M. Rignanese, J. Phys.: Condens. Matter 33, 404002 (2021).
[4] P.-P. De Breuck, G. Heymans, and G.-M. Rignanese, J. Mater. Inf. 2, 10 (2022).
[5] X. Liu, P.-P. De Breuck, L. Wang, and G.-M. Rignanese, npj Comput. Mater. 8, 233 (2022).

Wednesday, October 18, 16:50 – 17:25

# Density-Matrix Embedding Theory Based Multi-Configurational Wavefunction Approach to Strongly Correlated Single-Impurity Systems

Hong Jiang

*College of Chemistry and Molecular Engineering, Peking University, Beijing, 100871*

Electronic structure theory for strongly correlated systems (SCSs) poses a long-standing challenge in molecular quantum chemistry and condensed matter theory and has attracted tremendous efforts in recent decades. Among various theoretical methods that have been developed for strongly correlated molecular systems, multi-configurational (MC) self-consistent field (SCF) theory has played a particularly important role as it provides a systematic approach to static correlation that is the most challenging part of SCSs. However, due to the exponential scaling of MCSCF-type methods with respect to the size of the system under study, a direct application of those approaches to complex systems is computationally demanding and becomes even prohibitive for extended systems like solids and surfaces. There are continuous efforts in developing various quantum embedding methods that combine low- and high-level quantum solvers to treat different parts of a system in a consistent manner. Density matrix embedding theory (DMET) provides a systematic framework to combine low-level (e.g., Hartree−Fock approximation) and high-level correlated quantum chemistry methods to treat strongly correlated systems. In this work, we present an efficient quantum embedding approach that combines DMET with the complete active space self-consistent field and subsequent state interaction treatment of spin−orbit coupling (CASSI-SO) and its applications to single-ion magnets (SIMs) [1]. We have developed a novel regularized direct inversion of iterative subspace (R-DIIS) technique that ensures restricted open-shell Hartree-Fock, chosen as the low-level solver, converging to a physically correct ground state, which is found to be crucial for the efficacy of subsequent DMET treatment.

The DMET+CASSI-SO approach can predict accurately zero-field splitting parameters in typical 3d- and 4f-SIMs with dramatically reduced computational cost compared to its all-electron counterpart. This work therefore demonstrates the great potential of DMET-based multi-configurational approaches for efficient ab initio study of strongly correlated single-impurity systems.

**References**:

[1] Ai, Y.; Sun, Q.; Jiang, H. J. Phys. Chem. Lett. 2022, 13(4): 10627.

Thursday, October 19, 08:30 – 09:05

# Non-Fermi-liquid Metallic Thermoelectrics

Wenqing Zhang
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Thermoelectrics (TE) of narrow-gap semiconductor has been dominating the world-wide TE research. Seeking novel thermoelectrics especially metallic TEs also ran into difficulties due to the reported figure-of-merit *zTs* much less than the unity. This talk report on the discovery of non-Fermi-liquid (NFL) metallic thermoelectrics in off-stoichiometry Fe-based Heusler-like compounds (e.g. TiFe$_x$M$_y$Sb with non-integer x+y>1). The metallic TiFe$_x$Cu$_{2x-1}$Sb show the excellent Seebeck coefficients, reasonable electrical conductivity and a high *zTs* $\sim$ 0.75 or even higher, comparable with the state-of-the-art semiconductor thermoelectric (TE) materials. A nearly linear-in-temperature dependence of metallic electrical resistivity in 2K-700K and the logarithmic *T*-dependent electronic specific heat at low temperature are observed to coexist with the excellent thermopower, implying the strong coupling of the non-Fermi-liquid quantum critical behavior of electrons with TE transports. Those novel TE materials also possess weak magnetism, spin compensation, strong disorder, and wide composition tunability, all pointing different topics from the traditional thermoelectrics. We propose a d-orbital-determined compensation rule to reveal the abundant phase space of the specific off-stoichiometry Heusler-like compounds, such as TiFe$_x$Co$_y$Sb with non-integer x+y>1, being the potential candidates of the NFL thermoelectrics. This talk will also discuss the requirements for effective *ab initio* screening of this kind of new materials.

Thursday, October 19, 09:40 – 10:15

# Quantum Anomalous Hall Insulator from First-Principles Calculation

Hongming Weng

*Institute of Physics, CAS*

The materials having quantum anomalous Hall effect possess robust topological insulating bulk state and chiral conducting edge state. They can be used as ideal conducting wires for building exotic spintronic device, topological superconductor and Majorana zero mode. Presently, people have realized such quantum anomalous Hall insulator in several ways, such as the magnetic ion doped topological insulator thin film, a few layers of intrinsic magnetic topological insulator, and twisted two-dimensional systems (including twisted graphene and transition-metal dichalcogenides). Nevertheless, these realization schemes have their own limitations, which hinders the potential in practical applications. An ideal quantum anomalous Hall insulator should be stable, easy to be synthesized and exhibit the quantized conducting edge effect in relatively higher temperature. After comparison, we find that two possible ways. One is the quantum topological Hall effect induced by scalar spin chirality in non-coplanar antiferromagnets. The other one is the quantum anomalous Hall effect induced by the staggered bond loop current state due to strong electron correlation in two-dimensional system. We proposed several materials to realize these two schemes through first-principles calculation and analyzed the phase diagram and possible experimental condition to realize a better quantum anomalous Hall insulator.

Thursday, October 19, 10:45 – 11:20

# Metal-Support Interaction for Supported Nanocatalysts

Wei-Xue Li
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The stability of supported nanocatalysts is crucial to meeting environmental and energy challenges and necessitates fundamental theory to relieve trial-and-error experimentation and accelerate lab-to-fab translation. Among others, metal-support interaction is thought as one of the most fundamental concepts and vital to the overall stability of supported nanocatalysts. In particularly, strong metal-support interaction received long standing attention since it was proposed in 1969, though the description was limited to the phenomenal and qualitative aspects. Here we report a Sabatier principle of metal-support interaction for stabilizing metal nanocatalysts against sintering based on the kinetic simulations of 323 metal-support pairs using scaling relations from 1252 energetics data (Hu and Li (2021)). Too strong of an interaction is shown to trigger Ostwald ripening (Hu and Li (2018)) whereas too weak of an interaction stimulates particle migration and coalescence (Hu and Li (2019)). High-throughput screening of supports enables the sintering resistance of nanocatalysts to reach the Tammann temperature on homogeneous supports and far beyond it on heteroenergetic supports. This theory is substantiated by first-principles neural network molecular dynamics simulations, enable to quantify when strong metal-support interaction is strong metal-support interaction and paves the way for the design of stable nanocatalysts. Finally, I will report the latest progress on data-driven discovery of the interfacial interaction equation between metals and oxides from SISSO, which allows us to identify the origin of the strong metal support

interaction, one of the most importance concepts in heterogenous catalysis proposed in fifty years ago.

**References:**

[1]Hu, SL,Li, WX, Theoretical Investigation of Metal Support Interaction on Ripening Kinetics of Supported Particles, ChemNanoMat 4 (2018) 510

[2]Hu, SL,Li, WX, Metal Support Interaction Controlled Migration and Coalescence of Supported Particles, Sci. China-Technol. Sci. 62 (2019) 762-772

[3]Hu, SL,Li, WX, Sabatier principle of metal-support interaction for design of ultrastable metal nanocatalysts, Science 374 (2021) 1360-1365

Thursday, October 19, 11:20 – 11:55

# Rational Design of Fe-based Catalysts for Fischer-Tropsch Synthesis from Theoretical Prediction to Experimental Confirmation

Xiaodong Wen[1*],Yong Yang[1], Yongwang Li[1]

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Fischer-Tropsch process coverts syngas (CO and H2 which can be derived from nature gas, coal and biomass, etc.) into clean liquid fuels and high valuable chemicals. In China, the so-called Coal To Liquid (CTL) via Fischer-Tropsch synthesis (FTS) is not only to achieve the clean/efficient usage of coal, but also to alleviate China's excessive dependence on oil imports with important strategic significance to national security. We have been performing systematical studies on FTS for more than 30 years across from the fundamental research to R&D to industrial practice. The FTS in two 160,000 tons/year demo plants has passed the long-term operation and technical verification (more than 7 years), and achieved a remarkable successful application of the world’s largest single-set of Coal-to-Oil industrial plant in Shenhua Ningmei with a production capacity of 4 million tons/year

In the talk, the corresponding fundamental researches on FTS will be focused. The rational design of Fe-based catalysts for FTS will be presented via combining experimental and theoretical insights. In details, starting with theoretical studies, the quantum mechanics approach is extensively used to study the active phases of Fe-based catalyst for FTS (across from detailed benchmarks on method to predication on morphology/surface to design new generation Fe-based industrial catalysts), as well as FT reaction mechanism. The purpose of theoretical study is to provide theoretical insights for experimental designs, and to further guide the development of new generation FTS catalysts. Finally, the new generation Fe-based catalysts are designed rationally from theoretical prediction to experiment confirmation. In the meantime, the new challenge on designing Fe-based catalysts will be discussed

2023 The IOP-FHI Workshop

# Abstracts of Contributed Poster

October 16-19, 2023

Beijing·China

# Variance Extrapolation Method for Neural-Network Variational Monte Carlo

Weizhong Fu, Weiluo Ren, Ji Chen

*School of Physics, Peking University; Bytedance Research*

Constructing more expressive ansatz has been a primary focus for quantum Monte Carlo, aimed at more accurate calculations. However, with more powerful ansatz, e.g. various recent developed models based on neural-network architectures, the training becomes more difficult and expensive, which may have a counterproductive effect on the accuracy of calculation. In this work, we propose to make use of the training data to perform variance extrapolation when using neural-network ansatz in variational Monte Carlo. We show that this approach can speed up the convergence and surpass the ansatz limitation to obtain an improved estimation of the energy. Moreover, variance extrapolation greatly enhances the error cancellation capability, resulting in significantly improved relative energy outcomes, which are the keys to chemistry and physics problems.

# Quantum Tunneling Driven H2 Formation on Graphene

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It was widely accepted that the diffusion rates of H atoms adsorbed on graphene surface are very low at low temperatures (lower than 100 K), so two H atoms that are far away from each other cannot meet through diffusion and only recombine into H2 by colliding with other free H atoms in the environment [1]. But there are actually very few free H atoms in space, so this classical mechanism often fails to explain the higher abundance of H2 in space. *Ab initio* instanton is an accurate method to process quantum tunneling effect, search for the optimal high-dimensional tunneling paths and calculate the quantum tunneling rate, and has been widely used in H atom containing systems in recent years [2-3]. Using this method, we find that the deep tunneling of H atoms at low temperature dominates, while the desorption reaction is inhibited. At the same time, the ortho reaction rate, which is prohibited in the classical case, is greatly improved. Furthermore, we find that there is a significant difference between the high-dimensional quantum reaction path and the classical reaction path at low temperatures, namely the Quantum corner cutting effect. This has not been observed in previous studies based on the classical reaction pathways. This work shows that the recombination of H atoms at low temperatures is a fast process, which cannot be ignored in the interaction of graphene, graphite and other carbon-based materials with the environment [4].

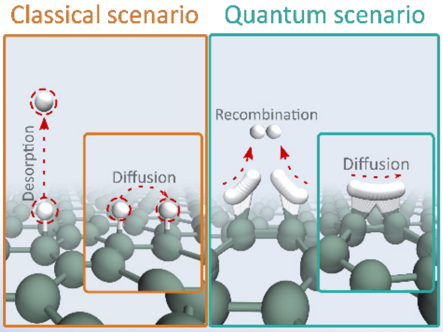

Fig. 1 Quantum and classical H2 formation reactions

**Keywords:** Quantum tunneling effect; Graphene; H2 formation; Quantum corner cutting effect

**References:**

[1] Wakelam, V.; Bron, E.; Cazaux, S.; Dulieu, F.; Gry, C.; Guillard, P.; Habart, E.; Hornekaer, L.; Morisset, S.; Nyman, G.; Pirronello, V.; Price, S. D.; Valdivia, V.; Vidali, G.; Watanabe, N., Mol. Astrophys. 9 (2017): 1-36.

[2] Fang, W.; Chen, J.; Pedevilla, P.; Li, X.-Z.; Richardson, J. O.; Michaelides, A., Nat. Commun. 11 (2022): 1689

[3] Litman, Y.; Rossi, M., Phys. Rev. Lett. 125 (2020): 216001

[4] Han, E., Fang, W., Stamatakis, M., Richardson, J. O. & Chen, J. J. Phys. Chem. Lett. 13 (2022): 3173–3181

# Spin-Triplet Topological Excitonic Insulators in Two-Dimensional Materials

Huaiyuan Yang, Jiaxi Zeng, Yuelin Shao, Yuanfeng Xu, Xi Dai, Xin-Zheng Li

*State Key Laboratory for Artificial Microstructure and Mesoscopic Physics*

Quantum spin-hall insulator (QSHI) processes non-trivial topology. Using first-principles many-body perturbation theory (GW+BSE) and model, we show that topological spin-triplet excitonic insulators (EIs) can exist in such QSHIs with non-vanishing band gaps. Spin-triplet type EI phase induced by strong electron-hole interaction preserves time-reversal symmetry and topological characteristics. A novel optical selection rule exists. Upon going through the phase transition from the normal QSHIs to the topological EIs, absorption spectroscopy shows pronounced temperature-dependent changes, and discussions for more experimental verications are provided.

# Graphic Characterization and Clustering Configuration Descriptors of Determinant Space for Molecules

Lei Sun,Zixi Zhang, Tonghuan Jiang, Yilin Chen, Ji Chen

*Peking University*

Quantum Monte Carlo approaches based on stochastic sampling of determinant space have evolved to be powerful methods to compute the electronic states of molecules. These methods not only calculate the correlation energy at an unprecedented accuracy but also provide insightful information on the electronic structures of computed states, for example, the population, connection, and clustering of determinants, which have not been fully explored. In this work, we devise a configuration graph for visualizing determinant space, revealing the nature of the molecule’s electronic structure. In addition, we propose two analytical descriptors to quantify the extent of configuration clustering of multideterminant wave functions. The graph and descriptors provide us with a fresh perspective of the electronic structures of molecules and can assist with further development of configuration interaction-based electronic structure methods.

# Implementation of Plane-Wave-Based Stochastic-Deterministic Density Functional Theory for Extended Systems in ABACUS

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Traditional Kohn-Sham density functional theory (KSDFT) is one of the most popular quantum-mechanics-based methods in modeling materials since it balances accuracy and efficiency well. However, traditional KSDFT based on the diagonalization method (DG) must solve all occupied bands. The number of bands rises with the increase of temperature and sizes, and the CPU time of KSDFT calculation scales as $O(N^3T^3)$. Thus, it is inefficient to simulate large-size and high-temperature systems. The evaluation of the ground-state density in KSDFT can be replaced by the Chebyshev trace (CT) method. Recently, stochastic density functional theory[1,2] (SDFT) and its improved theory, mixed stochastic-deterministic density functional theory[3] (MDFT) are developed based on the CT method and stochastic orbitals, which makes it possible to simulate large-size and high-temperature systems more efficiently. We have implemented the four methods based on the plane-wave basis set within the first-principles package ABACUS. In addition, all methods are adapted to the norm-conserving pseudopotentials and periodic boundary conditions using k-point sampling in the Brillouin zone. By testing the Si and C systems from the DG method as benchmarks, we systematically evaluate the accuracy and efficiency of the SDFT and MDFT methods by examining a series of physical properties, which include electron density, free energy, atomic forces, stress, and density of states. We conclude that they not only reproduce the DG results with sufficient accuracy but also exhibit several advantages over the DG method. We expect these methods can be of great help in studying both large-size systems and high-temperature systems.

# Machine Learning Based Non-Local Kinetic Energy Density Functional for Metals

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The development of an accurate kinetic energy density functional (KEDF) remains a major hurdle in the orbital-free density functional theory (OFDFT) field. We propose a non-local machine learning (ML) based KEDF for simple metals and their alloys, which adhere to three exact physical constraints: the scaling law, the free electron gas (FEG) limit, and the non-negativity of Pauli energy density. We conclude that incorporating the non-local information and obeying exact physical constraints are essential to improve the accuracy, generalization ability, and stability of ML-based KEDF. These results shed new light on the construction of ML-based functionals. (manuscript in preparation)

## References:

[1] R. Baer, D. Neuhauser, and E. Rabani, “Self-Averaging Stochastic Kohn-Sham Density-Functional Theory,” Phys. Rev. Lett. 111, 106402 (2013).

[2] Y. Cytter, E. Rabani, D. Neuhauser, and R. Baer, “Stochastic density functional theory at finite temperatures,” Phys. Rev. B 97, 115207 (2018).

[3] A. J. White and L. A. Collins, “Fast and Universal Kohn-Sham Density Functional Theory Algorithm for Warm Dense Matter to Hot Dense Plasma,” Phys. Rev. Lett. 125, 055002 (2020)

# Electric Polarization and Interatomic Force Fromneural Network-Based Quantum Monte Carlo

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Neural networks have been applied to quantum Monte Carlo (QMC) calculations and can achieve great accuracy for energy in materials. Here we show that the superiority lies not only in energy, but also in many other observables, such as electric polarization and interatomic force, which are difficult to calculate accurately in traditional QMC. We show that the dielectric response results for several systems outperform conventional density functional theory and are in agreement with the most accurate calculations and experimental data. We also show that the improvement of the wavefunction by the neural network ansatz can also benefit the force calculation. Our method demonstrates a promising future for the application of neural networks in condensed matter physics, achieving a higher level of accuracy and properly accounting for electron correlations.

**References:**

[1] X. Li, Y. Qian, and J. Chen, Electric Polarization from Many-Body Neural Net-work Ansatz, arXiv:2307.02212.

[2] Y. Qian, W. Fu, W. Ren, and J. Chen, Interatomic Force from Neural Network Based Variational Quantum Monte Carlo, The Journal of Chemical Physics 157,164104 (2022).

# Ab initio Simulation of Spin Relaxation Time of Single Ion Magnets Made

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Single ion magnets (SIMs) are a special kind of single molecule magnets (SMMs), in which magnetic anisotropy is contributed from a single d or f metal ion. The relaxation time of magnetic moment is a key property of a SMM for its application in information storage. A lot of recent experiments have shown that the coordination compounds of trivalent lanthanide ions have a long relaxation time, but its physical chemical mechanism is still not fully understood.

In recent years, the fully ab initio simulation of spin relaxation process has explained the physical mechanism of this phenomenon to some extent, which has found that the rate-determining step of relaxation is often a transition from the ground state to the first or second excited state. The energy level of the low excited state has a great influence on the relaxation time. At present, the ab initio research in this field is just getting started, and the demanding computational cost involved in such studies makes it infeasible to predict the spin relaxation properties of SMMs with complicated structures.

In this work, the fully ab initio simulation of spin relaxation process is carried out for several lanthanide SIMs. In order to analyze the detailed mechanism of the Orbach process, we construct a Markov process based on the master equation theory and find out the main factors that affect the barrier and relaxation time of the relaxation process, which provides a theoretical basis for some phenomenological explanations in the experiment. Moreover, the influence of the error of the spin-phonon Hamiltonian on the results is analyzed. We find that the accuracy of the perturbation cannot be transferred to the calculation result and the greatest influence of the perturbative CFPs on the result is its distribution rather than the exact value, which means that the large number of perturbative CFPs calculations do not make the relaxation time more accurate. Then we develop a simplified approach to the relaxation time by using the random perturbative Hamiltonian with a Gaussian distribution. The accuracy of the proposed method is verified to be comparable to that of the direct ab initio treatment but the computational cost is dramatically reduced, which makes it possible to predict the relaxation time of complicated SIMs quickly, and is therefore highly promising to provide helpful guidance for rational design of new SMMs

# Giant Acceleration of Polaron Transport by Ultrafast Laser-Induced Coherent Phonons

Huimin Wang

*Chinese Academy of Sciences Institute of Physics*

Polaron formation is ubiquitous in polarized materials, but severely hampers carrier transport for which effective controlling methods are urgently needed. Here, we show that laser-controlled coherent phonon excitation enables orders of magnitude enhancement of carrier mobility via accelerating polaron transport in a prototypical material, lithium peroxide ($Li_2O_2$). The selective excitation of specific phonon modes, whose vibrational pattern directly overlap with the polaronic lattice deformation, can remarkably reduce the energy barrier for polaron hopping. The strong nonadiabatic couplings between the electronic and ionic subsystem play a key role in triggering the migration of polaron, via promoting phonon-phonon scattering in q space within sub-picoseconds. These results extend our understanding of polaron transport dynamics to the nonequilibrium regime and allow for optoelectronic devices with ultrahigh on-off ratio and ultrafast responsibility, competitive with those of state-of-the-art devices fabricated based on free electron transport.

# Semiclassical Vibrational Spectroscopy of Real Molecular Systems by Means of Cross-Correlation Filter Diagonalization

Jia-Xi Zeng, Shuo Yang, Yu-Cheng Zhu, Wei Fang, Ling Jiang, En-Ge Wang,
Dong H. Zhang, and Xin-Zheng Li

*School of Physics, Peking University*

We applied the harmonic inversion technique to extract vibrational eigenvalues from the semiclassical initial value representation (SC-IVR) propagator of molecular systems described by explicit potential surfaces. The cross-correlation filter-diagonalization (CCFD) method is used for the inversion problem instead of the Fourier transformation, which allows much shorter propagation time and is thus capable of avoiding numerical divergence issues while getting rid of approximations like the separable one to the pre-exponential factor. We also used the “Divide-and-Conquer” technique to control the total dimensions under consideration, which helps to further enhance the numerical behavior of SC-IVR calculations and the stability of harmonic inversion methods. The technique is tested on small molecules and water trimer to justify its applicability and reliability. Results show that the CCFD method can effectively extract the vibrational eigenvalues from short trajectories and reproduce the original spectra conventionally obtained from long-time ones, with no loss on accuracy while the numerical behavior is much better. This work demonstrates the possibility to apply the combined method of CCFD and SC-IVR to real molecular potential surfaces, which might be a new way to overcome the numerical instabilities caused by the increase of dimensions.

# Co-Existence of Independent Charge Density Wave and Anti-Ferromagnetic Coupling in the Spin-Chain Compound $Ba_6Cr_2S_{10}$

Jianhua Zhu[12] ,Jianfeng Zhang[3], Yilin Zhang[1], Ji Chen1, and Wei Wu[2]

[1] *School of Physics, Peking University,* [2] *Department of Physics and Astronomy, University College London,* [3] *Institute of Physics, Chinese Academy of Sciences*

Quantum fluctuations become much more important than thermal ones at sufficiently low temperature, which can lead to fascinating physics such as Luttinger liquid [1]. Quasi-one-dimensional spin chains are important for studying the magnetic properties in low dimension. Here we theoretically studied the electronic structure and magnetic properties of the spin chain compound $Ba_6Cr_2S_{10}$ (BCS), which is newly synthesized as shown in [2]. In this work we have performed first principles calculations based on density-functional theory + U+ spin-orbit interaction methods implemented in VASP [3]. Based on our results, we have found the co-existence of independent charge density wave (CDW, labeled in red) phase and anti-ferromagnetic (AFM, labeled in blue) spin along separate chains (Fig.1). The dimerization of sulfur atoms loosely bonded with Ba atoms drives the system into an insulating state owing to the formation of charge density wave. Meanwhile, the small size of the effective Hubbard U parameter (~0.5 eV) for Cr atoms due to electrostatic screening mainly accounts for the anti-ferromagnetic ground state along the other chain. This co-existence equips us with a platform to tune the charge and spin degrees of freedom independently, shedding light on novel diluted magnetic semiconductors.

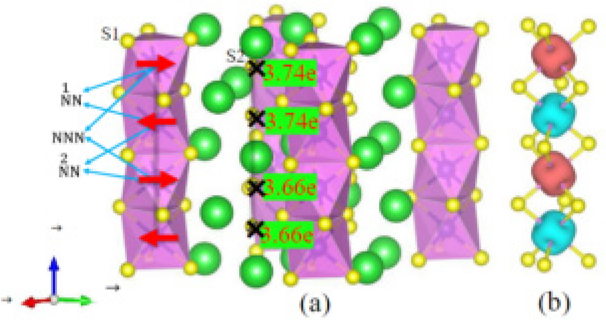

Figure 1: (a) The crystal structure of BCS is shown. Ba is in green, Cr is in blue, and S is in yellow. Cr atoms, bonded by S atoms, fom octahedron-face-sharing chains along the *c*-axis (illustrated by a blue line). S1 and s2 label the sulfur atoms for those forming ligands with Cr and those loosely bonded with Ba

(labeled by "x" ; the red line illustrates the S chain). respectively. We have also provided the atomic charges for the S atoms loosely bonded with Ba. The nearest-neighboring exchange interactions $J^{1}_{\mathrm{NN}}$ and $J^{2}_{\mathrm{NN}}$, and the next-nearest-neighboring exchange interaction $J_{\mathrm{NNN}}$ have been indicated as well.

(b) Spin densities on Cr suggest the orbital ordering on Cr along the chain. Due to the zig-zag ligand, the orientations of Cr orbitals will alternate along the c-axis.

**References:**

[1] J. M. Luttinger, J. Math. Phys. 4, 1154–1162 (1963).
[2] J. Zhang, et al. Adv. Mat. 34, 2106728 (2022).
[3] G. Kresse and J. Furthm ¨uller, Comput. Mater. Sci 6, 15 (1996).

# Crystal-Structure Matches in Solid-Solid Phase Transitions

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The exploration of solid-solid phase transition (SSPT) suffers from the uncertainty of how atoms in two crystal structures match. We devised a theoretical framework to describe and classify crystal-structure matches (CSM). Such description fully exploits the translational and rotational symmetries and is independent of the choice of supercells. This is enabled by the use of the Hermite normal form, an analog of reduced echelon form for integer matrices. With its help, exhausting all CSMs is made possible, which goes beyond the conventional optimization schemes. In an example study of the martensitic transformation of steel, our enumeration algorithm finds many candidate CSMs with lower strains than known mechanisms. Two long-sought CSMs accounting for the most commonly observed Kurdjumov-Sachs orientation relationship (OR) and the Nishiyama-Wassermann OR are unveiled. Given the comprehensiveness and efficiency, our enumeration scheme provide a promising strategy for SSPT mechanism research.

# Database Construction of Two-Dimensional Charged Building Blocks for Functional-Oriented Material Design

Jinbo Pan, Jun Deng, Yan-Fang Zhang, and Shixuan Du

Databases for charge-neutral two-dimensional (2D) building blocks (BBs), i.e., 2D materials, have been built for years due to their applications in nanoelectronics. Though lots of solids are constructed from charged 2DBBs, a database for them is still missing. Here, I would like to share our recent progress on the identification of over 1000 charged 2DBBs from Materials Project database using a topological-scaling algorithm. These BBs host versatile functionalities including superconductivity, magnetism, and topological properties. We construct layered materials by assembling these BBs considering valence state and lattice mismatch and predict 353 stable layered materials by high-throughput density functional theory calculations. These materials can not only inherit their functionalities but also show enhanced/emergent properties compared with their parent materials: CaAlSiF displays superconducting transition temperature higher than NaAlSi; $Na_2CuIO_6$ shows bipolar ferromagnetic semiconductivity and anomalous valley Hall effect that are absent in $KCuIO_6$; LaRhGeO possesses nontrivial band topology. This database expands the design space of functional materials for fundamental research and potential applications.

**Reference:**

[1] Deng, J. et al. Nano Lett. 2023, 23: 4634.

# Onset of Metallic Transition in Molecular Liquid Hydrogen

Jianqing Guo, Qingyuan Zhang, Bingqing Cheng, Limei Xu, Enge Wang, and Ji Chen

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Liquid-liquid phase transition of hydrogen is at the center of hydrogen phase diagram as a promising route towards emergent properties such as the Wigner-Huntington metallization, superconductivity, and superfluidity. Here we report a study on the liquid-liquid phase transition of hydrogen using the state-of-the-art diffusion quantum Monte Carlo and density functional theory calculations. Our results suggest that the metallization process happens at lower pressures and temperatures compared to the structural phase transition of molecular to atomic hydrogen. The consequence is that metallized molecular hydrogen is stable at a wide range of pressures and temperatures. Our study breaks the conventional assumption that metallization coinciding with dissociation of hydrogen molecule, and the molecular metallic hydrogen liquid phase is likely to become the frontier of studying hydrogen phase transitions.

# Basis-Set-Error-Free RPA Correlation Energies for Atoms Based on the Sternheimer Equation

Hao Peng

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Traditional Kohn-Sham density functional theory (KSDFT) is one of the most popular quantum-mechanics-based methods in modeling materials since it balances accuracy and efficiency well. However, traditional KSDFT based on the diagonalization method (DG) must solve all occupied bands. The number of bands rises with the increase of temperature and sizes, and the CPU time of KSDFT calculation scales as O(N3T3). Thus, it is inefficient to simulate large-size and high-temperature systems. The evaluation of the ground-state density in KSDFT can be replaced by the Chebyshev trace (CT) method. Recently, stochastic density functional theory1,2 (SDFT) and its improved theory, mixed stochastic-deterministic density functional theory3 (MDFT) are developed based on the CT method and stochastic orbitals, which makes it possible to simulate large-size and high-temperature systems more efficiently. We have implemented the four methods based on the plane-wave basis set within the first-principles package ABACUS. In addition, all methods are adapted to the norm-conserving pseudopotentials and periodic boundary conditions using k-point sampling in the Brillouin zone. By testing the Si and C systems from the DG method as benchmarks, we systematically evaluate the accuracy and efficiency of the SDFT and MDFT methods by examining a series of physical properties, which include electron density, free energy, atomic forces, stress, and density of states. We conclude that they not only reproduce the DG results with sufficient accuracy but also exhibit several advantages over the DG method. We expect these methods can be of great help in studying both large-size systems and high-temperature systems.

**Reference:**

[1] R. Baer, D. Neuhauser, and E. Rabani, “Self-Averaging Stochastic Kohn-Sham Density-Functional Theory,” Phys. Rev. Lett. 111, 106402 (2013).
[2] Y. Cytter, E. Rabani, D. Neuhauser, and R. Baer, “Stochastic density functional theory at finite temperatures,” Phys. Rev. B 97, 115207 (2018).
[3] A. J. White and L. A. Collins, “Fast and Universal Kohn-Sham Density Functional Theory Algorithm for Warm Dense Matter to Hot Dense Plasma,” Phys. Rev. Lett. 125, 055002 (2020)

# An Electron-Vibration Renormalization Method for Molecular Systems Based

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The electronic structure is influenced by the interaction between electrons and the vibration of nucleus. To calculate this effect, a many-body perturbation theory was proposed by Lars Hedin and Gordon Baym in the 1960s and further developed by H. Y. Fan et al. This theory has been used to calculate electron-phonon renormalization in DFT and GW calculations. However, it does not reveal the effects on the Green's functions, because it was always used as a post-processing method. To investigate properties affected by electron-vibration interaction, such as the spectral function, we derived a many-body perturbation theory suitable for molecular systems. The theory starts from the Hedin-Baym equations and corrects the GW approximation by introducing an electron-vibration coupling term in the self-energy. This method can calculate the effect of vibrations on the Green's function, and thus obtain the spectral function of molecular systems, which is validated by experimental photoelectron spectroscopy. In this study, we present the first-order Fan-Midgal term and second-order Debye-Waller term for molecular systems, demonstrating the influence of terms with different orders. Our work may provide the foundation for future electron-phonon interaction theories in periodic systems.

**Key Words:** molecular systems; many-body perturbation theory; electron-vibration interaction; spectral function

# Dynamic Phase Transition Theory

Qi-Jun Ye, Xin-Zheng Li

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Thermodynamic conventions suffer from describing dynamical distinctions, especially when the structural and energetic changes induced by rare events are insignificant. By using the ensemble theory in the trajectory space, we present a statistical approach to address this problem. Rather than spatial particle-particle interaction which dominates thermodynamics, the temporal correlation of events dominates the dynamics. The zeros of dynamic partition function mark phase transitions in the space-time, i.e., dynamic phase transition (DPT), as Yang and Lee formulate traditional phase transitions, and hence determine dynamic phases on both sides of the zeros. Analogous to the role of temperature (pressure) as thermal (mechanical) potential, we interpret the controlling variable of DPT, i.e., dynamic field, as the dynamical potential. These findings offer possibility towards a unified picture of phase and phase transition.

# Multiscale Theoretical Study of Carbon Dioxide Adsorption on Single-Atom Catalysts Embedded in Two-Dimensional Materials

Zhang Yi-Lin , Chen Ji

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For the sustainable and healthy development of human society, the catalytic reduction of carbon dioxide ($CO_2$) gas has been widely concerned by the scientific community in recent years. Due to the complexity of the $CO_2$ catalytic conversion process, understanding the physicochemical process of $CO_2$ adsorption on the catalyst surface at the atomic electron level is helpful to design a highly active and selective single-atom catalyst system. We will use a variety of theoretical calculation methods, including density functional theory and quantum Monte Carlo method, to study the key states and reaction paths of $CO_2$ on the surface of single-atom catalysts embedded in two-dimensional materials, calculate the basic physical quantities in each reaction process, and extract the descriptors of reaction kinetic characteristics. The structure-activity relationship between catalytic material system and catalytic performance was elucidated, and a more efficient and selective single atom catalyst system was designed.

# Combining Stochastic Density Functional Theory with Deep Potential Molecular Dynamics to Study Warm Dense Matter

Tao Chen, Qianrui Liu, Yu Liu, Liang Sun, Mohan Chen*

*HEDPS, CAPT, Peking University, Beijing*

In traditional finite-temperature Kohn-Sham density functional theory (KSDFT), the partial occupation of a large number of high-energy KS eigenstates restricts the use of first-principles molecular dynamics methods at extremely high temperatures. However, stochastic density functional theory (SDFT) can overcome the limitation. Recently, SDFT and its related mixed stochastic-deterministic density functional theory, based on the plane-wave basis set, have been implemented in the first-principles electronic structure software ABACUS [Phys. Rev. B 106, 125132 (2022)]. In this study, we combine SDFT with the Born-Oppenheimer molecular dynamics (BOMD) method to investigate systems with temperatures ranging from a few tens of eV to 1000 eV. Importantly, we train machine-learning-based interatomic models using the SDFT data and employ these deep potential models to simulate large-scale systems with long trajectories. Consequently, we compute and analyze the structural properties, dynamic properties, and transport coefficients of warm dense matter.

# Characterization of the Hydrogen-Bond Network in High-Pressure Water by Deep Potential Molecular Dynamics

Renxi Liu and Mohan Chen

*HEDPS, CAPT, College of Engineering, Peking University; Academy for Advanced Interdisciplinary Studies, Peking University*

The hydrogen-bond (H-bond) network of high-pressure water is investigated by neural-network-based molecular dynamics (MD) simulations with first-principles accuracy. The static structure factors (SSFs) of water at three densities, i.e., 1, 1.115, and 1.24 g/cm3, are directly evaluated from 512 water MD trajectories, which are in quantitative agreement with the experiments. We propose a new method to decompose the computed SSF and identify the changes in the SSF with respect to the changes in H-bond structures. We find that a larger water density results in a higher probability for one or two non-H-bonded water molecules to be inserted into the inner shell, explaining the changes in the tetrahedrality of water under pressure. We predict that the structure of the accepting end of water molecules is more easily influenced by the pressure than by the donating end. Our work sheds new light on explaining the SSF and H-bond properties in related fields.

# Ionic Transport Properties of in $Li_6PS_5Cl$ in system

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All-solid-state batteries (ASSBs) stand out as highly promising advancement to the traditional Li-ion battery technology. ASSBs employing solid electrolytes exhibit the potential for offering enhanced security, greater energy capacity, and improved power density [1]. The conductivity of solid electrolytes can be adjusted by atomic structures and compositions, allowing them to achieve ionic conductivity levels surpassing those of liquid electrolytes. Currently, various oxides and sulfide solid electrolytes (SSEs) with high $Li^+$ conductivity have been documented [2]. Sulfide-based superionic conductors are considered hopeful candidate materials for ASSBs due to their higher ionic conductivity, ease of synthesis at room temperature, and mechanical properties that facilitate close contact with electrodes [3]. We have selected synthetic Li-argyrodites, specifically $Li_6PS_5Cl$, as our research focus. We employed a deep potential generator to automatically generate interatomic potentials for $Li_6PS_5Cl$[4-6]. With these potentials, the accuracy of the evaluation for ionic conductivity in bulk materials can be significantly improved from three aspects. We extended the simulation of the diffusion process across a wide temperature range (200 K–1200 K). This allowed us to directly simulate the diffusion phenomena at room temperature, eliminating the need for traditional extrapolation methods. Additionally, by expanding the simulation size using 2×2×2 supercells, we increase the simulation system to thousands of atoms and mitigate size effect errors effectively. Moreover, we considered the influence of thermal expansion effects at different temperatures in the simulation, resulting in more accurate transport properties of the bulk $Li_6PS_5Cl$.

Building upon this foundation, we constructed an interface model to simulate the transport effects resulting from stress differences by varying the thickness of the vacuum layer. The simulation results indicate that the interfaces play a crucial role for ionic transport at low temperatures, including as low as 200 K.

**Reference:**

[1].Z. Gao, H. Sun, L. Fu, F. Ye, Y. Zhang, W. Luo, and Y. Huang, Advanced

materials, vol. 30, no. 17, p. 1705702
[2].A. Manthiram, X. Yu, and S. Wang, Nature Reviews Materials, vol. 2, no. 4, pp. 1
[3].Z. Deng, Z. Wang, I.-H. Chu, J. Luo, and S. P. Ong, Journal of The Electrochemical Society, vol. 163, no. 2,p. A67, 2
[4].H. Wang, L. Zhang, J. Han, and W. E, Comput. Phys. Commun. 228, 178–184 (2018).
[5].Y. Zhang, H. Wang, W. Chen, J. Zeng, L. Zhang, H. Wang, and W. E, Comput. Phys. Commun. 253, 107206 (2020).
[6].Huang, J., Zhang, L., Wang, H., Zhao, J., & Cheng, J. (2021). The Journal of Chemical Physics, 154(9).

# Simulating Dynamics of Electrochemical Interfaces under Realistic Conditions

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Outer layers typically is a key part for functionalities of nano-materials, while liquid-solid interfaces in electrochemistry is one of the most interesting yet challenging topics to study. The complexity and enrichment come from several elements: constant potential leads to different charge state upon different adsorptions; dynamical hydrogen bonds may accelerate/hinder reaction steps; electron transfer and proton transfer maybe coupled etc. In this talk, we will report our recent work on first-principles “constant potential-hybrid solvation-dynamic model”(CP-HS-DM) to simulate dynamics of electrochemical interfaces under realistic conditions. Particularly, we will use $CO_2$ reduction, $O_2$ reduction, and degradation/regeneration of Cu electrocatalyst as examples to discuss the importance of charges on electrode, explicit hydrogen bonds and kinetic free energy barriers based on ab-initio molecular dynamics.

# Periodic Coupled-Cluster Theory with Atom-Centered Basis Functions in FHI-Aims

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In recent years, the periodic formulation of coupled-cluster (CC) theory has proven itself repeatedly as a reliable and highly accurate benchmark electronic structure method in materials science [1-3]. The all-electron code FHI-aims [4], which employs numeric atom-centered orbitals, has recently been interfaced to the CC theory for solids (CC4S) code [5] via the CC-aims library [6], making CC theory for both the ground state and for excited states in the equation-of-motion coupled-cluster (EOM-CC) theory accessible to FHI-aims. In particular, the EOM-CC framework allows for the systematic inclusion of electronic correlation in the calculation of band gaps and band structures [7] and has been shown to predict quasi-particle energies more accurately and reliably than the GW approximation in the molecular case [8].

To fully capitalize on the utility of the CC and EOM-CC method, however, two sources of error have to be addressed: Firstly, periodic coupled-cluster theory– like most correlated wave function methods– exhibit excessively slow convergence to the thermodynamic limit. Even though a very powerful finite-size correction technique based on the transition structure factor has been formulated for plane waves [9], an equally effective treatment for atom-centered basis sets does not exist yet.

Secondly, wave function methods suffer from equally slow convergence to the complete basis set (CBS) limit. While this can be effectively addressed by employing basis sets that can be extrapolated to the CBS, convergence with respect to the auxiliary basis of the localized resolution-of-identity (RI-LVL) scheme employed in FHI-aims needs to be ensured.

As an all-electron code, FHI-aims treats core- and valence electrons on equal footing without resorting to approximations as they are encountered in pseudopotentials or the projector augmented wave method, which potentially can contribute to an erroneous description of the electronic structure. We present the current state of the coupled-cluster theory framework available in FHI-aims and possible avenues to address the auxiliary basis- and finite-size error.

**Reference:**

[1] G. Booth et al., Nature 493, 365–370 (2013)
[2] T. Gruber, A. Grüneis, Physical Review B, 98, 134108 (2018)
[3] X. Wang et al., Journal of Chemical Theory and Computation, 16, 3095-3103 (2020)
[4] The FHI-aims web page, https://fhi-aims.org, Accessed: 11-05-2022
[5] F. Hummel et al., Journal of Computational Physics, 146, 124105 (2017)
[6] E. Moerman et al., The Journal of Open Source Software 7, 4 (2022)
[7] J. McClain et al., Journal of Chemical Theory and Computation 13, 1209-1218 (2017)
[8] M. Lange et al., Journal of chemical theory and Computation, 14, 4224-4236 (2018)
[9] K. Liao, A. Grüneis, Journal of Computational Physics 145, 141102 (2016)

# X-ray Photoelectron Spectroscopy Study of Iron-Based Catalyst Model System

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X-ray photoelectron spectroscopy (XPS) is a widespread used technique in material characterization, which provides the electronic structure information and the chemical environment of the target element by measuring the core level electron binding energy. However, the lack of standard reference data increases the difficulty of spectrum analysis and linking up the signals to the structural and electronic information of the sample. Theoretical simulation of spectroscopy, starting from the atomic structure followed by quantum mechanism calculation, provides another important method to material characterization. In this paper, the Fe 2p core level binding energy shifts for various iron compounds were calculated in order to develop and implement first principle based XPS simulation method. The reliability of the theoretical method was comprehensively evaluated, including an all-electron extension of the delta self-consistent field (ΔSCF) method and core level orbital energy approximation methods for pseudopotential. The suitable method is selected by combining the calculation accuracy and efficiency, and then used in XPS simulation of iron based material model system. This study aims to provide a bridge between the XPS signal and the microstructure, and to increase the XPS reference from the theoretical dimension.

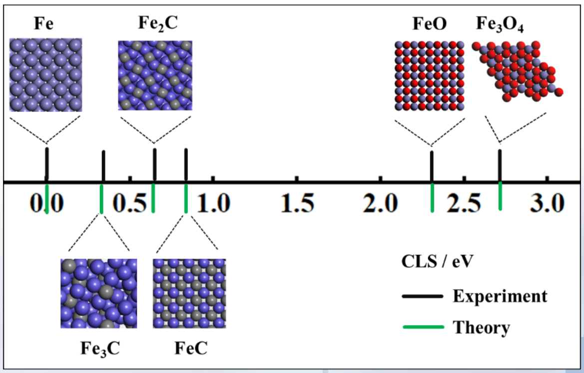

Figure 1. The ensemble analysis of Fe 2p core level shift for orbital energy approximation method based on PBE functional and compare the calcula1ted results with the experimental results.

# LibRI: An Open-Source Library for Advanced Electronic Structure Calculations Based on the Resolution of Identity

Peize Lin[12,] Rong Shi[34], Minye Zhang[156], Lixin He[34], and Xinguo Ren[1]

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The resolution of identity (RI) is a key numerical technique that enables efficient electronic structure calculations based on atomic-orbital basis sets.
Here, we propose a unified mathematical model for RI, where the RI-based formulation of several advanced electronic-structure methods, such as hybrid functionals, random-phase approximation, and GW can be considered as special cases of the model.

Based on this mathematical model, we develop an open-source library called LibRI.

The core of LibRI lies in the unified model, with the implementations of various advanced electronic-structure methods being simple callers.
Consequently, in LibRI all optimization algorithms and acceleration techniques implemented for the model are automatically applicable to all RI-based electronic-structure methods.

LibRI is massively parallel and can be called as a library by first-principles software with local atomic bases. The source code of LibRI is accessible via GitHub[1].

**Reference:**

[1] https://github.com/abacusmodeling/LibRI

# Sub-Quadratic Scaling Real-Space Random-Phase Approximation Correlation Energy Calculations for Periodic Systems with Numerical Atomic Orbitals

Rong Shi, Peize Lin, Min-Ye Zhang, Lixin He, Xinguo Ren

*CAS Key Laboratory of Quantum Information, University of Science and Technology of China, Hefei 230026, Anhui, China4*

The random phase approximation (RPA) as formulated as an orbital-dependent, fifth-rung functional within the density functional theory (DFT) framework offers a promising approach for calculating the ground-state energies and the derived properties of real materials. Its widespread use to large-size, complex materials is however impeded by the significantly increased computational cost, compared to lower-rung functionals. The standard implementation exhibits an O(N4)-scaling behavior with respect to system size N. In this work, we develop a low-scaling RPA algorithm for periodic systems, based on the numerical atomic orbital (NAO) basis-set framework and a localized variant of the resolution of identity (RI) approximation. The rate-determining step for RPA calculations -- the evaluation of non-interacting response function matrix, is reduced from O(N4) to O(N2) by just exploiting the sparsity of the RI expansion coefficients, resultant from localized RI (LRI) scheme and the strict locality of NAOs. The computational cost of this step can be further reduced to linear scaling if the decay behavior of the Green's function in real space can be further taken into account. Benchmark calculations against existing k-space based implementation confirms the validity and high numerical precision of the present algorithm and implementation. The new RPA algorithm allows us to readily handle three-dimensional, closely-packed solid state materials with over 1000 atoms. The algorithm and numerical techniques developed in this work also have implications for developing low-scaling algorithms for other correlated methods to be applicable to large-scale extended materials.

# A Compact Tensor Regression Model for Accelerating First-Principle Molecular Dynamics Simulations

Li Hengyu,[1] Taisuke Ozaki,[1,2,*]

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The desired system size of ab initio molecular dynamics (AIMD) simulation has been ever-increasing in recent years. In combination with density functional theory (DFT), machine learning (ML) techniques have been proven to reproduce the potential energy surface (PES) in a DFT level accuracy [1]. However, existing ML potentials still face challenges for large complex systems with MD simulation. In this study we present a compact and robust atomic energy model for fitting local PES and expect it applied with on-the-fly algorithm for accelerating AIMD simulations for large-scale systems.

Our atomic energy model is defined by a linear combination of different dimension element inside the descriptor vector and their high-order couplings which represents neighboring chemical environment. The undetermined parameters for each term form a n-rank tensor. To construct the descriptor vector, we consider the atomic energy is contributed by 2-body and 3-body interactions in a finite range. Such features are stored in a descriptor vector that each dimension is a summation of all neighboring pairs for centra atom transformed by designed orthogonal polynomial. We also include the atomic number in all many-body pairs for multi-element systems. Our model can be interpreted as an approximation to a multilayer neural network model with nonlinear activation function. The fitting process is done by minimizing the quadratic loss function of energy and force and oriented to solving a linear equation problem. In the implementation, the fitting and predicting can be fully parallelized for each atom on different processor and leads to overall O(N) time complexity.

The benchmark validation calculations are conducted with OpenMX [2] for various type systems including isolated molecule (Methane, Nitrobenzene, Sialic Acid, Adenosine Triphosphate), bulk system (2D Graphene, Amorphous silicon, FCC Aluminum). For 2-rank model we report energy MAE from 0.83 meV/atom (Methane) to 4.21 meV/atom (Adenosine Triphosphate) with atomic force MAE from 23 meV/Å to 263 meV/Å. The accuracy level is comparable to reported ML potential (energy: ~5 meV/atom, force: ~300 meV/Å) with about 2 times less parameters (66 with 5th order descriptor function). To ensure the stability of our model for conducting MD simulation, we further present the long-time (2ps 2000 step) MD simulation for relaxed amorphous silicon system under 300K. Also, for indicating the ability of on-the-fly training, model is updated every 5 steps with 1 DFT calculation. The radial distribution function of Si-Si pair with 2 characteristic peaks (~2.4, 3.8 Å) are well reproduced.

**Reference:**

[1] K. T. Butler et al., Nature 559, 547 (2018).
[2] OpenMX: https://www.openmx-square.org/

# Preferential Decomposition of the Major Anion in a Dual-Salt Electrolyte Facilitates the Formation of Organic-Inorganic Composite Solid Electrolyte Interphase

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The performance of a lithium metal battery (LMB) with liquid electrolytes depends on the realization of a stable solid electrolyte interphase (SEI) on the Li anode surface. According to a recent experiment, a high-concentrated (HC) dual-salt electrolyte is effective in modulating the SEI formation and improving the battery performance. However, the underlying reaction mechanism between this HC dual-salt electrolyte and the lithium metal anode surface remains unknown. To understand the SEI formation mechanism, we first performed 95 ps ab initio Molecular Dynamics (AIMD) simulation and then extend this AIMD simulation to another 1 ns by using Hybrid ab Initio and Reactive Molecular Dynamics (HAIR) to investigate the deep reactions of such dual-salt electrolytes consists of lithium difluorophosphate (LiDFP) and lithium bis(trifluoromethanesulfonyl) imide (LiTFSI) in dimethoxyethane (DME) solvent at lithium metal anode surface. We observed the detailed reductive decomposition processes of DFP- and TFSI-, which include the formation pathway of $CF_3$ fragments, LiF, and $Li_xPOF_y$, the three main SEI components observed experimentally. Furthermore, after extending the simulation to 1.1 ns via the HAIR scheme, the decomposition reactions of DME solvent molecules were also observed, producing $LiOCH_3$, $C_2H_4$, and precursors of organic oligomers. These microscopic insights provide important guidance in designing the advanced dual-salt electrolytes for developing high-performance LMB.

# A Compact Tensor Regression Model for Accelerating First-Principle Molecular Dynamics Simulations

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**Reference:**

[1] K. T. Butler et al., Nature 559, 547 (2018).
[2] OpenMX: https://www.openmx-square.org/

# Solid-State High Harmonic Spectroscopy: A Potential All-Optical Route for Band Structure Probing of High-Pressure Quantum States

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High-pressure has triggered various novel states/properties in condensed matters, one of the most presentive examples is the high-pressure induced room temperature superconductivity in hydrogen sulfide ($H_3S$). The mechanism of superconductivity is still not confirmed in experiment, due to the lacking of effective approach for directly probing the electronic band structure. The angle-resolved photoemission spectroscopy (ARPES) loss its power under such high pressure.

Acting as a highly nonlinear response to the strong laser field, solid-state high harmonic generation (sHHG) naturally contains the fingerprints of electronic structure of materials [1]. This quality inspires us to investigate sHHG spectroscopy as a potential approach for band probing of high-pressure quantum states, for example the superconductors H3S.

The strategy is validated by the state-of-art first principle time-dependent density-functional theory, where the dispersion of multiple bands in high-pressure $H_3S$ near Fermi level along different momentum directions are effectively retrieved, under different laser conditions.

The setup of sHHG process in high-pressure H3S and corresponding sHHG spectra shown in Figure. 1. The laser pulses with photon energy ~ 0.4 eV is applied. By fitting the sHHG intensity as a function of field strength [2,3], the band structures can be measured

This work provides a promising alternative route for electronic structure characterization complementary to ARPES. Since band probing by sHHG is not restricted to electronic occupation, we propose this approach is also suitable for nonequilibrium electronic structure measurement far away from Fermi level, which may provide insights for the mechanism of light-induced potential superconductors such as $YBa_2Cu_3O_{6.5}$ and $K_3C_{60}$.

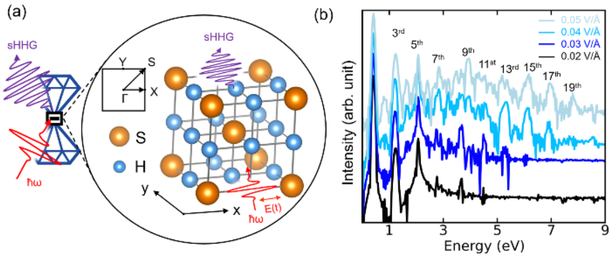

Figure 1. (a) Schematics showing the sHHG process of high-pressure H3S. (b) The sHHG spectra in H3S along Γ-X.

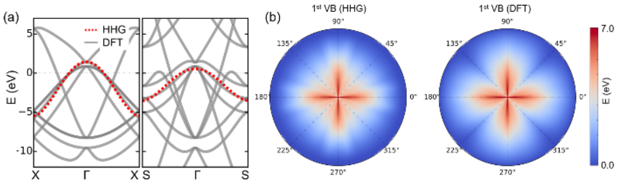

Figure 2. The band structure of 1st valence band retrieved from the sHHG.

## References:

[1]G. Vampa et al 2015 Phys. Rev. Lett.115 193603.
[2] T. T. Luu et al 2015 Nature 521 498.
[3] A. A. Lanin et al 2017 Optica 4 516.

# NOTES

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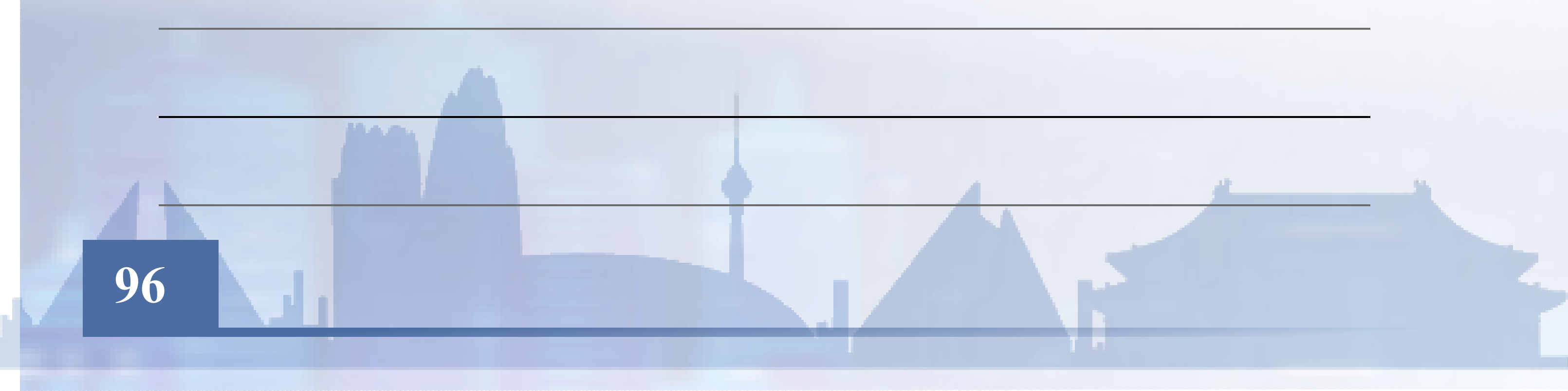

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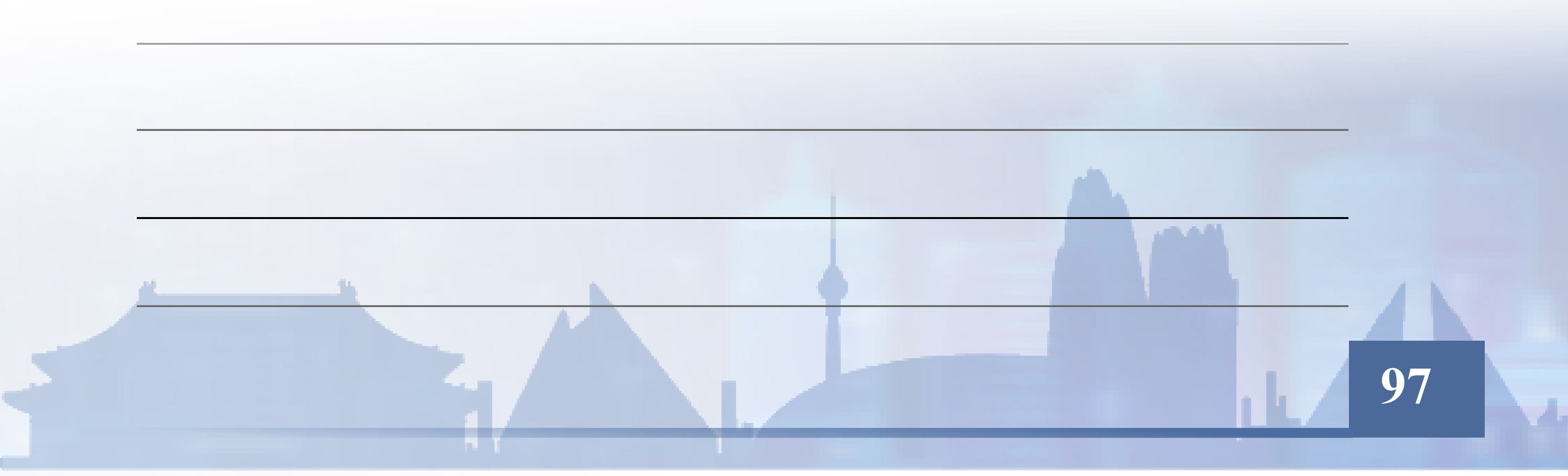

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