

# NAMMA Psi-k 2023

## Abstract Book for Invited Talks



New Approaches and Machine learning Methods for  
*Ab initio* calculations: NAMMA Psi-k Workshop



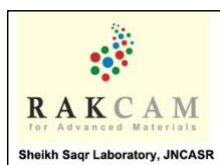
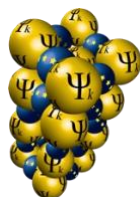
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## Table Of Contents

---

1. Abir De Sarkar.....	3
2. Aftab Alam .....	4
3. Ambesh Dixit.....	5
4. Amrita Bhattacharya.....	6
5. Ananth Govind Rajan .....	7
6. Anirudh Raju Natarajan.....	8
7. A. Jain.....	9
8. Anuj Goyal .....	10
9. Aparna Chakrabarti.....	11
10. Arti Kashyap.....	12
11. Awadhesh Narayan.....	13
12. Ayan Datta .....	14
13. Bahadur Singh .....	15
14. Biplab Sanyal.....	16
15. Bulumoni Kalita .....	17
16. Sharma S. R. K. C. Yamijala.....	18
17. D. D. Sarma .....	19
18. Debjani Karmakar.....	20
19. Dibyajyoti Ghosh.....	21
20. G Vaitheeswaran.....	22
21. Gour P. Das.....	23
22. Hemant Kumar.....	24
23. Indra Dasgupta.....	25
24. Jithin John Varghese.....	26
25. Joydeep Bhattacharjee .....	27
26. Kavita Joshi .....	28
27. Koushik Pal.....	29
28. Manoj K. Harbola .....	30
29. Michele Kotiuga .....	31
30. Mudit Dixit .....	32
31. Mukul Kabir .....	33
32. Munima B Sahariah .....	34
33. N.S. Vidhyadhiraja .....	35
34. Navaneetha K. Ravichandran .....	36

35. Nirmal Ganguli .....	37
36. Nisanth N. Nair .....	38
37. P. Ravindran .....	39
38. Phani Motamarri .....	40
39. Prafulla K Jha .....	41
40. Prasanjit Samal .....	42
41. Prasenjit Ghosh .....	43
42. Prasenjit Sen .....	44
43. Priya Johari .....	45
44. Priya Mahadevan .....	46
45. Rabeet Singh .....	47
46. Rajeev Ahuja .....	48
47. B. R. K. Nanda .....	49
48. Ranjit Thapa .....	50
49. Sundaram Balasubramanian .....	51
50. Sai Gautam Gopalakrishnan .....	52
51. Santanu Mahapatra .....	53
52. Saroj Kumar Nayak .....	54
53. Saswata Bhattacharya .....	55
54. Satadeep Bhattacharjee .....	56
55. Souvik Paul .....	57
56. S. Battaglia .....	58
57. Subhradip Ghosh .....	59
58. Dr. Sudip Chakraborty .....	60
59. Swapan K Pati .....	61
60. Swastika Chatterjee .....	62
61. Tanusri Saha-Dasgupta .....	63
62. Tej S. Choksi .....	64
63. Umesh V Waghmare .....	65
64. V Kanchana .....	66
65. Valerio Vitale .....	67
66. V. Srinivasan .....	68
67. Vikram Gavini .....	69

# DFT perspectives on valleytronics, piezoelectricity and spintronics in selected functional 2D materials

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## **Abstract**

Novel properties such as piezoelectricity and valley physics arise at the nanoscale which are usually non-existent in the bulk form of the materials. HfN<sub>2</sub> monolayers<sup>[1]</sup> exhibit valleytronic properties complementary to that in single-layer MoS<sub>2</sub>, while the merger of spintronic with valleytronic properties is observed in h-NbN and h-TaN monolayers<sup>[2]</sup>. The feasibility of the synthesis of Janus MoSSe monolayer has motivated us to investigate piezoelectricity in Janus structures in centrosymmetric 2D Group IV transition metal dichalcogenides and trichalcogenides. Ultrahigh out-of-plane piezoelectricity meets the giant Rashba effect in 2D Janus monolayers and bilayers of Group IV transition metal trichalcogenides<sup>[3]</sup>. Out-of-plane piezoelectricity is induced at the interfaces of 2D semiconducting planar monolayers, which show in-plane piezoelectricity individually and zero out-of-plane polarization/piezoelectricity, such as GaN and boron monophosphide (BP) monolayers. The understanding reached in GaN/BP van der Waals heterobilayers (vdWHs) has been reinforced on MoS<sub>2</sub>/BP and MoSSe/BP vdWHs. The origin of negative piezoelectricity at the interfaces of 2D dialkali oxide and chalcogenide monolayers has been elucidated together with the enhanced electrical conductivity arising from nearly free electron gas (NFEF) states<sup>[4]</sup>. The conflux of tunable Rashba effect and piezoelectricity observed in flexible MgTe, CdTe, and ZnTe monolayers signify its super high prospects for self-powered flexible-piezo-spintronics<sup>[5]</sup>. The nature of metal-semiconductor contacts plays a crucial role in determining device performance. Non-resistive/Ohmic contact is found to occur in graphene/MgS vdWH, rendering it ideal for charge injection<sup>[6]</sup>.

[1] M. K. Mohanta, A. De Sarkar, *Phys. Rev. B*, **2020**, *102*, 125414.

[2] R. Ahammed, A. De Sarkar, *Phys. Rev. B*, **2022**, *105*, 045426.

[3] R. Ahammed, N. Jena, A. Rawat, M. K. Mohanta, Dimple, A. De Sarkar, *J. Phys. Chem. C*, **2020**, *124*, 21250.

[4] A. Arora, A. Rawat, A. De Sarkar, *Phys. Rev. B*, **2023**, *107*, 085402.

[5] M. K. Mohanta, F. IS, A. Kishore, A. De Sarkar\*, *ACS Appl. Mater. Interfaces*, **2021**, *13*, 40872.

[6] M. K. Mohanta, A. Arora, A. De Sarkar, *Phys. Rev. B*, **2021**, *104*, 165421.

# Thermoelectric Properties of Topological Semimetal $\text{Cu}_2\text{ZnGeTe}_4$ : A New paradigm to Renewable Energy

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## **Abstract**

The study of topological quantum materials for enhanced thermoelectric (TE) energy conversion has received significant attention recently. Topological materials (including topological insulators and Dirac/Weyl/nodal-line semi-metals) with the unique combination of band structure involving linear and regular parabolic bands near Fermi level ( $E_F$ ) have the potential to show promising TE properties. In this talk, I will report the promising TE performance of a quaternary chalcogenide ( $\text{CuZnGeTe}_4$ ) having non-trivial topological phase. At ambient condition, the compound is a narrow band gap (0.067 eV) semiconductor, with a TE figure of merit (ZT)  $\sim 1.2$ . Application of 5% strain drives the system to a topologically non-trivial Weyl semi-metal with the right combination of linear and parabolic bands near  $E_F$ , giving rise to a reasonable ZT of 0.36. Apart from strain, alloy-engineering (Sn substituted at Ge) will also be shown to induce topological non-triviality. The present work demonstrates the potential of such unique semimetals for exceptional electronic transport properties and hence appreciable thermoelectric performance.

[1] Alam et al., Phys. Rev. B **2021**, *104*, 085113.

[1] Pan et al., Adv. Mater. **2021**, *33*, 2003168.

[1] Alam et al., Phys. Rev. B **2021**, *103*, 195143.

# Designing Lead-free Cs<sub>2</sub>BB'X<sub>6</sub> Double Perovskites for Energy Application(s): A Hybrid Computational Approach

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## **Abstract**

The perovskites, primarily the lead-based ones, unearthed their exciting promise for photovoltaics (PV). Yet the associated lead toxicity and instability with this contemporary class, the halide double perovskites (DPs), are gaining attention. The talk will cover the design of lead-free, highly stable Cs<sub>2</sub>BB'X<sub>6</sub> (B: Ag/Au/Cu, B': Bi/Sb/Tl and X: Br/Cl/I) halide DPs using different exchange-correlation approximations under density functional umbrella and will explore their stability using Goldschmidt tolerance factor (t), modified tolerance factor (t'), and thermodynamical investigations considering decomposition pathways. The spin-orbit coupling (SOC) is used to understand the impact of the heavy element as a replacement for Pb in these DPs. A wide range of band gaps (0.2 eV to 2.35 eV) is noticed for the 27 explored DPs and the trivalent thallium cation-based DP exhibits metallic characteristics. Further, the mechanism behind band gap variation will be discussed, correlating the different B/B'/X combinations. Further, in pursuit of the photovoltaic, the macroscopic simulation approach is employed to investigate Cs<sub>2</sub>CuBiCl<sub>6</sub> based inverted device with TiO<sub>2</sub> and Cu<sub>2</sub>O as electron and hole charge-transporting layers, resulting in 17.03% power conversion efficiency in a single junction configuration. The wide band gap spectra of these DPs allow them to be integrated into multiple junction solar cells with potential thermoelectric materials devices for a wide operating temperature range.<sup>[1]</sup>

[1] A. J. Kale, R. Chaurasiya, A. Dixit *ACS Applied Energy Materials*, **2022** 5, 10427-10445.

# Exploring the thermodynamic stability and spin orbit driven Rashba splitting in Perovskite oxides

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## **Abstract**

In material discovery, scanning the potential energy surface of any compositional space (i.e. perovskites, in this context) using formation energy calculations i.e. with  $E_{\text{hull}}$  analysis is not sufficient to comment on thermodynamic stability without taking vibrational free energy into consideration. The calculation of vibrational Helmholtz free energy through first principles is very expensive. Hence, machine learning can be used to predict the zero point energy (ZPE) and Helmholtz free energy variation with temperature. The temperature dependence is modelled using 3rd order polynomial fit. Our study gives highly accurate ML models using only elemental and simple compound descriptors for ZPE [1]. Once the thermodynamic stability of these Perovskites has been explored, they can be analysed for their technological applications such as in spintronics research i.e. as spin control devices. One such manifestation of spin control can be seen in Rashba-Dresselhaus effect in non-magnetic insulators with broken inversion symmetry. This phenomenon of spin splitting in the momentum space if it occurs in the band edges can help in realising the spin field effect transistor devices. To achieve the spin control via switching of gate voltage, existence of ferroelectric polarisation in the material is necessary. This has led to a lot of research on ferroelectric Rashba semiconductors (FERSC) and formulation of design principles necessary to search for such materials. Using first principles density functional theory based methods, we analyse the Iodate perovskites viz.  $\text{AIO}_3$ ,  $\text{A}=\text{K, Rb, Cs, and Tl}$  in non-centrosymmetric rhombohedral phases (R3m). More specifically, we study the effect of A cation in the Rashba spin splitting by taking into the account of atomic as well as geometric relaxation, spin polarization, and spin orbit coupling. Our study reveals that the Rashba splitting is inversely proportional while the band gap is directly proportional to the ionic radii of the A cation [2].

[1] Machine learning the vibrational free energy of single perovskites, Krishnaraj Kundavu, Suman Mondal, and Amrita Bhattacharya, *Materials Advances*, 2023 (Accepted).

[2] Momentum-space Spin Splitting in non-Centrosymmetric Iodate Perovskites, Krishnaraj Kundavu, Suman Mondal, and Amrita Bhattacharya (communicated).

# First-Principles Discovery of the Mechanisms Underlying the Synthesis of Nanoporous 2D Materials

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## **Abstract**

First-principles calculations based on electronic density functional theory (DFT) offer a convenient yet accurate framework to study the thermodynamics and kinetics of mechanisms that underlie several important chemical processes. Prominent among these processes are the synthesis of two-dimensional (2D) materials via chemical vapor deposition, the formation of nanopores in 2D materials, and the functionalization of nanopores by various chemical moieties in aqueous and atmospheric environments. I will discuss our group's recent work on using first-principles calculations within the framework of DFT to understand the synthesis of the 2D material molybdenum disulfide (MoS<sub>2</sub>) from metal-organic precursors. I will present an extensive set of DFT calculations that reveal the nucleation and growth mechanism of MoS<sub>2</sub> during chemical vapor deposition synthesis by capturing the previously unidentified elementary steps of MoS<sub>2</sub> growth. I will also show the use of reaction-rate theory to predict the temperature- and pressure-dependent transition of MoS<sub>2</sub> flakes from triangular to hexagonal shapes. Next, I will discuss the application of a theoretical framework based on Marcus theory for atom-transfer reactions to understand the mechanism of nanopore formation in MoS<sub>2</sub>. Finally, I will close with a DFT-based study of the thermodynamic and kinetic stability of functionalized nanopores in 2D materials. Overall, I will explain how the combined use of phenomenological theories and DFT calculations can allow for the development of models for complex atomic processes at the nanoscale.



# Computational tools for the ab-initio design of high-temperature structural alloys

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## **Abstract**

Materials used in high temperature applications are carefully engineered to achieve optimal mechanical properties and chemical durability. Refractory alloys comprising elements from groups four, five and six of the periodic table are an emerging class of high-temperature materials that promise high strength and thermal stability. However, relationships between alloy chemistry, processing strategies and material properties remain unclear. In this talk I will highlight recent advances in computational materials science that leverage machine-learning techniques to enable the rigorous and rational design of high-performance engineering alloys. First-principles calculations will be used with machine-learning and statistical mechanics techniques to rigorously describe the thermodynamic and kinetic properties of multicomponent refractory alloys. High-throughput calculations will be employed to discern design rules for multi-component refractory alloys with desirable properties. Diffusion mechanisms and phase transformations in multicomponent refractory alloys will also be highlighted. The techniques and results presented in this talk provide important insights for the design of advanced structural alloys.

# Phonon thermal transport in low- and high-thermal conductivity solids

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## **Abstract**

Understanding phonon thermal transport physics in semiconductors is crucial for applications such as thermoelectricity, nuclear reactors, heat dissipation, data storage, etc. Conventionally, the thermal transport in crystalline semiconductors is described using Boltzmann Transport Equation with three-phonon intrinsic phonon-phonon scattering. Recently, this description of phonons needs to be revised to explain thermal transport in technologically relevant low- and high-thermal conductivity solids. In this talk, I will present some of our group efforts on methodology development to account for the relevant higher-order thermal transport physics for these materials. I will show results for two material systems, one with ultra-low thermal conductivity where temperature-dependent sampling of the potential energy surface, phonon renormalization due to anharmonicity, four-phonon scattering, and multi-channel thermal transport are all necessary, and the second system where all available methodologies fail, and the obtained thermal conductivity from state-of-the-art calculations is off from the experimental measurements by a factor of 3-5. <sup>[1-2]</sup>

[1] A. Jain, Physical Review B 102, 201201 (R) (2020)

[2] S.Alam, AG Gokhale, and A. Jain, Journal of Applied Physics 133, 215102 (2023)

# A computational framework to accelerate defect energy prediction and materials discovery for clean energy applications

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## **Abstract**

In this talk, I will present an open-source Python framework to automate point defect calculations using density functional theory (DFT). The framework provides an efficient method for defect generation and the creation of simple yet customizable workflows to analyze defect calculations<sup>[1]</sup>. Using the DFT reference data for vacancy defects in oxides, we develop a defect graph neural network (dGNN) model that fully automates the prediction of defect formation enthalpies of any crystallographic site from its relaxed host crystal structure and replaces the DFT supercell relaxations otherwise required for each symmetrically unique crystal site<sup>[2]</sup>. The approach is applicable to arbitrary structures with an accuracy limited principally by the amount/diversity of the data on which it is trained. Interfaced with thermodynamic modeling of reduction entropies and associated free energies allows us to connect the predicted defect energies to high-temperature process conditions relevant to the different application areas. For example, by high-throughput screening metal oxides in the materials databases, we rapidly "re-discover" and identify potential candidate materials for hydrogen generation via solar thermochemical water splitting. The computational framework provides an effective screening and discovery capability for applications where vacancy defects are the primary driver of a material's utility. The approach is generalizable to other defect types and will aid in tackling future materials discovery problems in clean energy and beyond.

- [1] Goyal, A., Gorai, P., Peng, H., Lany, S. & Stevanović, V. A computational framework for automation of point defect calculations. *Comput Mater Sci* **130**, 1–9 (2017).
- [2] Witman, M., Goyal, A., Ogitsu, T., McDaniel, A. & Lany, S. Materials discovery for high-temperature, clean-energy applications using graph neural network models of vacancy defects and free-energy calculations. *ChemRxiv* (2023).

# Exploring the Prospect of Technological Applications of Quantum Materials using DFT

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## **Abstract**

Research on novel quantum materials is quite important in today's world – from points of view of fundamental science and technological applications. For several years, we are working on these materials with potential in the field of applications.

One interesting field of research is the magneto-tunneling junction(MTJ), as these systems are crucial for spintronic devices, like memory devices, spin transfer torque devices, etc. MTJ devices comprise two ferromagnetic electrodes separated by a non-magnetic insulator or semiconductor spacer. MgO has become the most important spacer in this quest. However, achieving high conductance and oxide layer formation at the interface during annealing remains a challenge for MgO-based MTJs. Here comes the need of all-Heusler alloy MTJ materials (AHA-MTJs). Therefore, researchers have started searching for promising electrodes with unique properties, like high spin polarization, low Gilbert damping, and high Curie temperature and spacers with a narrower band gap that can have similar symmetry-filtering transport properties as MgO.

Very recently from ab-initio density functional theory based electronic structure calculations, we have predicted a few combinations of AHA-MTJs with a half-metallic(HM) electrode layer.<sup>[1]</sup> Our calculations suggest technologically desired high quality electrode/spacer interface and find that the HM properties at the interface are robust. These materials exhibit diverse electronic and transport properties, with efficient spin conductance and strain-sensitive transmission under both compressive and tensile strain. These also show very low resistance-area product, which is important for highly sensitive magnetic sensors. Our on-going work searches for more coherent  $\Gamma$ -centric tunneling, as observed in case of MgO.

Another of our technologically relevant works aims towards augmenting the experimental results on ion batteries. Search for new anode/cathode materials has gained enormous attention of the researchers. Two different phases of 2D MoTe<sub>2</sub> have been established as good anode materials for Li-ion battery, with support from our theoretical calculations<sup>[2]</sup>

[1] J. Bhattacharya et al,(i) *Comp. Mat. Sci.* **2023**, 216, 111852; (ii) Manuscript in preparation.

[2] M. R. Panda et al, (i) *Small* **2020**, 16, 2002669; (ii) *ACS Appl. Energy Mater.* **2022**, 5, 9625.

# Magnetic MXene: A Machine learning model with small data

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## **Abstract**

Two-dimensional (2D) nitrides and carbides of transition metals called MXene are gaining popularity due to their tremendous applications in energy storage [1],[2], as catalysts [3], [4] photonics [5] and many more fields. The MXenes are hexagonal layered nitrides, carbides or carbonitrides of transition metals with the chemical formula  $M_{n+1}X_nT_x$  where n range from 1 to 3 and M represents the early transition metal, X is C or N,  $T_x$  is the surface terminations like OH, F, O, or Cl. These 2D materials are produced by etching out the A layers from the MAX phase, which is a family of hexagonal layered ternary transition metal nitrides, carbides, and carbonitrides with a composition of  $M_{n+1}AX_n$  where A stands for a group A element (Al, Si, Sn, In etc.). Therefore, a large number of chemical combinations are possible, and after the discovery of the first MXene ( $Ti_3C_2T_x$ ) in 2011, more than 30 compositions were published.[6] Interestingly, the presence of 3d transition metals makes them a potential candidate for low-dimensional magnetism.[7] This can be of interest to various applications such as data storage, electromagnetic interference shielding, and spintronic devices.

Here, we focus on the magnetic MXenes, which are only a few in number out of known MXenes. We propose machine learning models to predict the magnetic moments of the MXenes and to classify the MXenes based on their stability.

- [1] B. Anasori, M. R. Lukatskaya, and Y. Gogotsi, “2D metal carbides and nitrides (MXenes) for energy storage,” *Nature Reviews Materials*, vol. 2, no. 2. 2017, doi: 10.1038/natrevmats.2016.98.
- [2] J. Pang *et al.*, “Applications of 2D MXenes in energy conversion and storage systems,” *Chemical Society Reviews*, vol. 48, no. 1. 2019, doi: 10.1039/c8cs00324f.
- [3] Z. W. Seh *et al.*, “Two-Dimensional Molybdenum Carbide (MXene) as an Efficient Electrocatalyst for Hydrogen Evolution,” *ACS Energy Lett.*, vol. 1, no. 3, 2016, doi: 10.1021/acsenerylett.6b00247.
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- [5] H. Kim, Z. Wang, and H. N. Alshareef, “MXetronics: Electronic and photonic applications of MXenes,” *Nano Energy*, vol. 60. Elsevier Ltd, pp. 179–197, 2019, doi: 10.1016/j.nanoen.2019.03.020.
- [6] Y. Gogotsi and B. Anasori, “The Rise of MXenes,” *ACS Nano*, vol. 13, no. 8, pp. 8491–8494, 2019, doi: 10.1021/acsnano.9b06394.
- [7] Q. Yao *et al.*, “A Promising Intrinsic Half-Metallic MXene Nanosheet  $Sc_2Li_2N_3$ : the First-Principles Study,” *J. Supercond. Nov. Magn.*, vol. 35, no. 12, pp. 3727–3734, 2022, doi: 10.1007/s10948-022-06436-w.

# Berry curvature dipole and non-linear Hall effect in two-dimensional materials

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## **Abstract**

In recent years, it has been discovered that inversion symmetry broken quantum systems can exhibit non-linear Hall effect even under time-reversal symmetric conditions.<sup>[1]</sup> The underlying quantum object leading to this effect is the first order moment of the Berry curvature, termed the Berry curvature dipole. Such non-linear Hall effects open up different possibilities for direct applications, such as non-linear photocurrents and terahertz radiation detection.<sup>[2]</sup> However, despite such promise for exciting physics and applications, the Berry curvature dipole induced non-linear Hall effect has been experimentally realized only in a handful of materials. It is, therefore, of vital importance to find materials with large and controllable Berry curvature dipole.

In this talk, I will give examples from our work where such a controllable Berry curvature dipole has been predicted. First, we propose Janus monolayers of transition metal dichalcogenides as a promising materials platform to explore the non-linear Hall effect and Berry curvature dipole physics.<sup>[3]</sup> Here the topology and the Berry curvature dipole are tunable by chemical composition. Second, we discover a giant non-linear Hall effect in the elemental buckled honeycomb lattices -- silicene, germanene, and stanine.<sup>[4]</sup> In this case, the Berry curvature dipole is tunable by a transverse electric field which breaks inversion symmetry. We demonstrate that the electric field induced topological phase transitions are associated with a giant Berry curvature dipole near the critical field.

[1] I. Sodemann and L. Fu, Phys. Rev. Lett. 115, 216806 (2015)

[2] Z. Du, H.-Z. Lu, and X. Xie, Nature Reviews Physics 3, 744 (2021)

[3] N. B. Joseph, S. Roy, and A. Narayan, Materials Research Express 8, 124001 (2021)

[4] A. Bandyopadhyay, N. B. Joseph, and A. Narayan, 2D Materials 9, 035013 (2022)

# Buckling and Defects in Two - Dimensional Atomically Thin Monolayers

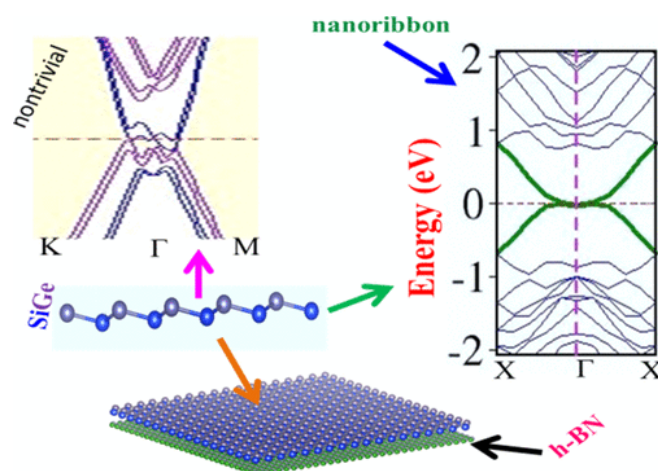
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## **Abstract**

Graphene has emerged as one of the most fascinating areas of research in condensed matter and materials science. Apart from graphene, several other 2D analogues like silicene, phosphorene, MXenes, MoS<sub>2</sub>, germanene have been recently isolated and characterized. One unifying theme among these new materials is that unlike graphene, they are non-planar. The mode and measure of the buckling (puckering) from planarity depends on the local electronic structure. The effects of such ripples are manifold particularly in its local reactivity to halogens and hydrogens along the more  $\sim sp^3$  atoms. Pseudo Jahn-Teller (PJT) distortions is shown as a central unifying concept that explains the overall structural preferences of these systems. The extent of buckling also leads to interesting and emerging phenomenon like phase-transitions into Topological Insulators from normal semi-metal due to spin-orbital coupling (SOC). The effect of PJT in bestowing structural diversity and novel electronic and spin phases in 2D-monoatomic layers will be discussed in this lecture.



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# Local structural motifs driven topological electronic states in quantum materials

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## **Abstract**

Recent materials classification efforts based on the topology of electronic states and crystalline lattice symmetries predicted many topological materials in high-throughput first-principles calculations. Such symmetry-to-topology characterizations, however, ignore the effects of various crystallographic or nanoscopic motifs and their associated wavefunction properties even though they are essential for describing the numbers, energy-momentum relations, and geometries of the nontrivial states. Here we will discuss how spatial arrangements of atoms dictate energy dispersions of topological electronic states in materials. Specifically, we will show the role of lattice geometry-driven effective mass anisotropies in forming hybrid nodal-line fermion states by considering transition metal tetraphosphides  $\text{TmP}_4$  (Tm = Transition metal) as exemplary materials.<sup>[1]</sup> We will also demonstrate how the passivation of dangling bond states and symmetry-lowering structural distortions can result in designer states with tunable topological properties and enhanced spin-Berry curvature in quantum materials.<sup>[2]</sup>

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# Complexities in electronic structure and magnetism of 2D $\text{Fe}_n\text{GeTe}_2$ ( $n=3, 4, 5$ ) magnets

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## **Abstract**

In recent years, the realization of magnetic long-range order in atomically thin 2D materials has shown a big potential in spintronic applications in ultrathin magnets due to the possibility of manipulation of magnetism by external fields, strain or proximity effects in heterostructures. In this regard, the family of metallic magnets  $\text{Fe}_n\text{GeTe}_2$  ( $n=3, 4, 5$ ) has attracted huge attention due to their high Curie temperatures and intriguing properties. Some recent results will be shown obtained by ab initio density functional theory, calculations of interatomic exchange interaction parameters and Monte Carlo simulations. A systematic study of the electronic structure and magnetism of these magnets will be presented along with some critical discussions on the importance of electron correlation<sup>[1]</sup> and spin-orbit coupling. Moreover, structural reconstruction and peculiar magnetism in  $\text{Fe}_5\text{GeTe}_2$ <sup>[2]</sup> will be discussed. Finally, the effects of Ni doping in  $\text{Fe}_5\text{GeTe}_2$ <sup>[3]</sup> in controlling the structural, electronic and magnetic properties will be shown.

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# Effect of Structural Point Defects on Electronic Properties of MgO Monolayers

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## **Abstract**

Two-dimensional (2D) graphene-like hexagonal materials emerging during the last two decades are of interest for their fascinating electronic, optical, mechanical properties.<sup>[1]</sup> Very recently, point defects such as vacancies and non-metallic substitutional doping have been observed to strongly affect the electronic band structure in graphene-like MgO monolayers, increasing their applicability in optoelectronics and spintronics.<sup>[2-4]</sup> We have performed hybrid-HSE level of DFT calculations using Quantum ESPRESSO package to study the effect of various point defects on the structural and electronic properties of MgO (111) monolayers (MLs). In this study, concentrations as well as position of oxygen vacancy defects are found to significantly alter the electronic energy band structure of MgO MLs [**Error! Bookmark not defined.**]. Further, substitutional doping and co-doping of 2*p* elements such as B, C, N in both Mg and O sites have been extensively investigated. Such type of defects introduces huge alteration in the electronic structure of ML systems. Single doping converts the non-magnetic monolayers into magnetic while homo- and hetero-type of co-doping results in both magnetic and non-magnetic states along with metallic and semiconducting nature of electronic structures. The semiconducting monolayers exhibit a large variation in their energy band gap from IR to Visible range of the electromagnetic spectrum.

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# Exploring the Formation and Evolution of Solid Electrolyte Interphase at Calcium Surfaces

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## **Abstract**

Energy storage devices with high energy densities (Wh/kg) are required for long-range electric vehicles. Multivalent ion batteries such as magnesium and calcium ion batteries (CIBs) have the potential to exhibit high specific charge capacities (Ah/kg) as they can hold/deliver more than one electron per ion. Among the multivalent ions, Ca is the most electropositive element ( $E^0_{\text{Ca}^{2+}/\text{Ca}} = -2.87$  V vs. SHE) and it is the fifth most abundant element in the earth's crust. Since its reduction potential is just 170 mV lower than Li, a Ca anode can deliver high cell voltages.<sup>[1]</sup> Together with high specific capacities (Ah/kg) and voltages (V), CIBs are expected to deliver high energy densities (Wh/kg). Despite these advantages, the major impediment in the growth of CIBs is the sluggish kinetics of calcium ions through the solid-electrolyte interphase (SEI). Due to the high reduction potential of Ca, electrolytes spontaneously decompose on the Ca surface and form an SEI. Interestingly, experiments show that the solvent and salt molecules have a huge impact in the formation and structure of SEI, and thereby, affecting the mobility of Ca-ions. For example, the SEI formed in the presence of  $\text{Ca}(\text{BF}_4)_2$  is permeable to Ca ions, whereas the SEI formed with  $\text{Ca}(\text{TFSI})_2$  is not.<sup>[2]</sup> Apparently, it is crucial to understand the nature of SEI in the presence of different salts and solvents, and to modify the SEI structure to realize efficient CIBs.<sup>[3]</sup> In this work, using *ab initio* molecular dynamics, we investigated the formation and evolution of SEI on a Ca(001) surface when it is in contact with a EC: PC solvent mixture both in the absence and presence of 0.45 M  $\text{Ca}(\text{TFSI})_2$  salt or  $\text{Ca}(\text{BF}_4)_2$  salt. The simulation results will be presented in my talk.

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# Putting a spin on battery electrodes - the importance of strong correlation physics

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## **Abstract**

There have been recent experimental suggestions that higher energy can be achieved by utilizing anionic redox processes in cathode materials of lithium ion batteries. We investigate the origin of this effect using first-principles electronic structure calculations for Li-rich layered sulphides. We find that conventional approaches do not capture the energetic contribution arising from intra-atomic Hund's exchange coupling, hindering a reliable description of the processes involved. This drawback is overcome by developing a particularly efficient flavor of charge-selfconsistent combined density functional + dynamical mean-field theory (DFT+DMFT) calculations for investigating such battery materials. This scheme allows us to describe the spin ground states of the transition metal d shell, the electronic structure of the materials, and its energetics accurately, capturing a spin-state transition between the charged and the discharged state of the electrode. As a result of this high-spin to low-spin transition the average intercalation voltage shows a non-monotonic behavior, emphasising the importance of correlation effects.

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# Spin-dynamical ground-state: An answer to the inherent symmetry breaking in magnetic superconductors

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## **Abstract**

In the superconducting series  $AV_3Sb_5$  ( $A = Cs, Rb$  and  $K$ ), an intricate interplay of strong-correlations, non-trivial electronic topology and intriguing fermiology result into mutually dependent exotic phases like charge-order or superconductivity. For this series, the interconnection of magnetism and complex collective phenomena and their combined impact on the underlying exotic phases are not known. Using first-principles based DFT+DMFT analysis; we demonstrate that electron correlations and non-trivial magnetism in conjunction with the spin-orbit coupling have strong influences on their electronic structure, complex fermiology and phonon dispersions. Ab-initio computation of inter-site magnetic exchanges along with the analysis of  $q$ -dependence of the response functions and electron-phonon coupling provide an answer to the unresolved experimental questions like the occurrence of giant anomalous Hall effect, magnetic frustration and its inter-dependence with superconductivity, charge-order and multiple Q-nesting. With *ab-initio* calculated inter-site exchanges as a function of the nearest neighbor distances, the magnetocrystalline anisotropy and the site projected magnetic moments, we have solved the full-fledged atomistic spin-dynamical calculation to arrive at the dynamical magnetic ground state. The calculations reveal a complex magnetic chiral ground state with significant DM-interactions of their non-collinear spin moments for this system. This analysis has the potential to provide a possible explanation for the experimentally observed inherent  $C_6$  rotational and time-reversal symmetry-breaking in their exotic phases.

# Energy Materials: Atomic-scale Insights from ab initio modelling

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## **Abstract**

On a global scale, clean energy generation and its efficient use are important aspects to meet rising energy demands with sustainability. However, complex and interdependent structure-property-performance relations strongly limit the directed search and discovery of efficient materials for targeted energy applications. To tackle these challenges, I apply cutting-edge computational simulations that are invaluable for understanding and manipulating the functionalities and performances of a wide range of energy materials. In this talk, first I will discuss the details of charge carrier dynamics and structural stability of metal halide perovskites that are leading contenders for next-generation optoelectronic devices. Using non-adiabatic molecular dynamics, I will illustrate the complex influences of dynamic structures on the excited-state carrier dynamics that strongly impact the optoelectronic performances of these materials.<sup>[1-4]</sup> Following that, my talk will focus on defect formations and their impact on the functional properties of materials. We will discuss our findings on the substantial influences of surface defects on the optoelectronics of semiconductor materials.<sup>[5]</sup> The defect-assisted ion migration and its suppression through compositional engineering will also be explored through examples.<sup>[6,7]</sup> In conclusion, I will briefly outline our multidisciplinary present and future research plans that concentrate on building up reliable computational simulation schemes for fast-tracking systematic designing, optimization, and discovery of materials, highly desirable for various energy technologies.

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# Metallization of Solid Iodanil ( $C_6I_4O_2$ ) under pressure

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## **Abstract**

A comprehensive *ab-initio* investigation has been carried out to explore the structural, lattice dynamics, and pressure-induced metallization of solid iodanil ( $C_6I_4O_2$ ). Our calculated ground state structural properties highlight the importance of including van der Waals correction for an accurate determination of these properties. The computed phonon dispersion confirm the dynamical stability of solid iodanil in the  $P21/c$  symmetry. Upon increasing pressure, we observed a softening of the transverse acoustic (TA) phonon mode in the D-B direction. Notably, at 22 GPa, corresponding to the insulator-metal transition, the TA branch along the D-B direction exhibited a hardening effect, in which the optical phonon mode ( $A_u$ ) play a prominent role. To further assess the mechanical stability, we calculated the elastic constants of solid iodanil. Additionally, we employed the quasiparticle  $G_0W_0$  approximation to determine the electronic band structure, resulting in a computed band gap of 2.49 eV. This value significantly exceeded the value obtained using the generalized gradient approximation, establishing the significance of quasiparticle correction in accurately describing solid iodanil. Our study demonstrated a reduction in the band gap under pressure, leading to a band overlap that ultimately triggered the metallization of iodanil at approximately 22 GPa. Furthermore, we analyzed the pressure-dependent variations in inter- and intramolecular bond lengths, as well as the charge density plots, explaining the significant role of intermolecular I-I distances in understanding the metallization mechanism of solid iodanil.

# Computational design of materials for energy storage

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## **Abstract**

The energy storage community is gainfully utilizing the symbiosis of first-principles DFT simulation with experiments and data-driven machine learning paradigms. We have been working on metal-ion batteries and hydrogen storage, the two most well studied and complementary candidates for energy storage. After giving a brief overview of the role of DFT in understanding and improving their performance, I shall discuss two specific examples viz. (a)  $\text{Na}_x\text{MnO}_2$  a candidate materials for Na-ion battery and (b) Si-C alloy nanocluster for hydrogen storage. Notwithstanding the clear advantages of Li-ion batteries, there is ongoing research to replace Li-ion by other earth abundant materials such as Na-ion [1]. Layered P2 type different TM-doped  $\text{Na}_x\text{MnO}_2$  cathodes have been investigated for their electronic structure and voltage profile [2]. The inclusion of more Jahn-Teller active  $\text{Mn}^{3+}$ -ions by desodiation induces sub-lattice formation in the matrix and concomitant increment of c axis. Better cyclability is observed for decreasing 'x' in  $\text{Na}_x\text{MnO}_2$  cathodes. Upon desodiation till  $x=0.67$  the density of O atoms increases near the Fermi level, indicating the higher probability of O reduction. In the second part of the talk, we shall discuss hydrogenated Si-C alloy nanocluster as a candidate material for Hydrogen storage [3]. We conducted a projected population analysis to demonstrate a correlation of Si-Si and Si-H bonding interactions with the nanoparticle size, and argued the ease of Si-Si dimerization in presence of C-core, thereby favoring the release of  $\text{H}_2$ . Here we deploy real-space DFT calculations using finite-element basis, in order to extract information in large realistic systems via a newly developed approach [3], which is an alternative route to the well known COHP analysis. The latter approach has already been used to study Li-ion diffusivity in  $\text{Li}_3\text{YCl}_6$  solid electrolyte [4].

† Work done in collaboration with Tanmoy Paul, Phani Motamarri, Satadeep Bhattacharjee

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# Improving the Design of Solid-State Electrolytes with Machine Learning

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## **Abstract**

The demand for affordable, safe, and high-performance energy storage systems is increasing rapidly in the world of portable electronics and electric vehicles. Solid-state batteries have the potential to meet these requirements while outperforming their liquid-electrolyte-based counterparts. However, despite the continuous quest to discover suitable solid-state electrolytes (SSE), the current findings fall short in terms of ideal ionic conductivity and contact with electrode.

The traditional method of screening SSE involves relying on human knowledge, which frequently introduces biases, as well as a time-consuming and resource-intensive trial-and-error approach. As a result, a wide range of promising Li-containing structures remain unexplored. To accelerate the search for optimal SSE materials, it is crucial to understand the chemical and structural factors that govern ion transport within a crystalline lattice.

In this seminar, we will present a new approach to the design of SSEs that combines machine learning (ML) with density functional theory (DFT). ML techniques can be used to identify the key features that influence ion mobility in solid-state materials, while DFT can be used to calculate the energetics and dynamics of ion transport. This approach has been used to design new NASICON electrolytes with significantly improved migration barriers and ionic conductivity.

We will also discuss the application of ML to the study of grain boundaries in garnet-type solid electrolytes, such as LLZO. Grain boundaries are important defects in polycrystalline materials that can significantly affect the electrical, chemical, and mechanical properties of the material. By using ML to predict the behavior of ions at grain boundaries, we can design materials with improved performance.

This work demonstrates the potential of ML to accelerate the discovery of new SSE materials with enhanced properties. We believe that this approach will be a valuable tool for the development of next-generation solid-state batteries.

# Spin-Orbit Coupling Induced Emergent Phases in Quantum Materials

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## **Abstract**

Spin-orbit coupling (SOC) is a relativistic effect, which may be thought of as an interaction between the intrinsic spin moment of an electron and the magnetic field generated in the rest frame of the electron due to its orbital motion around the positively charged nucleus. We shall illustrate that the combined effect of electron correlations and SOC leads to novel  $J_{\text{eff}}=1/2$  Mott Insulators, bond-directional dependent exchange interactions important for the realization of Kitaev spin liquids and the possibility of Magnon Chern Insulators in honeycomb systems. <sup>[1]</sup>, <sup>[2]</sup>, <sup>[3]</sup>.

In addition, we shall show emergence of Rashba and Dresselhaus spin-orbit interactions due to a gradient of electrostatic potential in non-centrosymmetric systems. Rashba-Dresselhaus systems exhibit characteristic spin textures important for spintronics research <sup>[4]</sup>, <sup>[5]</sup> <sup>[6]</sup>.

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# Computational spectroscopy and first principles microkinetic modelling in computational catalysis

Anoop P Pushkar, Pushkala Venkatesh, Balaji C Dharmalingam, Ajay Koushik V, Niket S Kaisare, Preeti Aghalayam, **Jithin John Varghese**<sup>1\*</sup>

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## **Abstract**

Density Functional Theory (DFT) simulations have become the backbone of computational investigations of structure of catalysts, analyses of reaction mechanisms and reaction pathways on identified catalyst surfaces. Workflows integrating these with insights from experiments are crucial to ascertain validity and predictability of the computational approaches.<sup>1</sup> *Ab initio* thermodynamics calculations enable assessment of the stability of different surfaces of the catalysts and determination of near operando surface structure of catalysts in equilibrium with reaction environments. Computational Raman and Infrared (IR) spectroscopic analyses are powerful tools to further characterize and validate *ab initio* thermodynamics predicted surface states and active site motifs. These will be demonstrated with examples on ceria and vanadia deposited ceria catalysts which are explored as catalysts for oxidative dehydrogenation of propane.<sup>2</sup> Computational IR analyses with appropriate probe species such as CO and CH<sub>4</sub> can also be used to identify potential surface planes of metal oxide catalysts relevant for catalytic reactions. This will be shown with the example methane partial oxidation to methanol on Rh-ZrO<sub>2</sub> catalyst. DFT reaction pathways analysis and analyses of the underlying energy landscapes enable qualitative interpretations and predictions of reaction mechanisms. IR spectroscopy of the key intermediates enable validation of the identified reaction mechanisms and assigning molecular identities to species whose signatures are observed in the experiments.<sup>2</sup> DFT based microkinetic modelling bridges the gap between surface science-like DFT predictions and global kinetic modelling approaches by enabling predictions of macroscopic observables in catalytic reactor systems at typical operating conditions while incorporating the complexity of the reaction networks and identities of the catalyst active sites. This will be demonstrated with an example of CO<sub>2</sub> reduction to methanol on Cu/ZnO<sub>x</sub>/ZrO<sub>x</sub>/Al<sub>2</sub>O<sub>3</sub> catalyst.<sup>3</sup> Approaches in closing the loop a iteratively refining the computational predictions will be discussed.<sup>1</sup>

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# Estimation of quasi-particle band-gap and optical absorption threshold of large systems in a minimal tight-binding basis

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## **Abstract**

In this talk I will discuss efforts we have undertaken over the last few years towards developing inexpensive computational strategies for workably accurate estimation of quasi-particle band-gap and optical gap in large systems comprising of hundreds to thousands of atoms, as typical of experimentally realizable nano-structures, for which it is impossibly expensive to compute those measurable quantities explicitly using standard methodologies.

Central to our approach is a minimal tight-binding basis constituted by a set of optimally directed orthonormal hybrid atomic Wannier orbitals[1] constructed from the Kohn-Sham(KS) states of a given system. Directions of the basis orbitals are optimized[2] to incorporate maximum valency as quantifiable by the Mayer's bond order, with minimum number of orbitals, which also naturally reveal the bent nature of covalent bonds for systems with non- $\dot{D}FT + \dot{G}_0W_0$ es.

Self-energy corrected tight-binding(SEC-TB) parameters computed in the directed basis at the level are found to effectively transfer self-energy correction(SEC) from smaller reference system to much large isomorphic systems through mapping of neighborhood beyond the nearest neighbors. Transferred SEC-TB parameters typically up to second nearest neighbor are found[3] to almost completely account for self-energy correction to the single particle levels, implying the inherently local nature of self-energy correction in covalent systems.

We have further proposed[4] a systematically parametrizable anti-causally corrected real time dynamics scheme with transferred SEC-TB parameters in directed basis, to make workably accurate estimation of the optical-gap of large systems.

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# Solid-state hydrogen storage: Decoding the path through machine learning

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## **Abstract**

The globe has reached a point where switching to renewable or green energy sources is one of the few viable options for a sustainable environment. In this milieu of sustainability, hydrogen as an energy carrier is the linchpin in achieving energy security due to its high energy density (142 MJ/Kg). One of the major roadblocks to realizing the hydrogen economy is its economic storage. While compressed and liquefied hydrogen is utilized widely in industries, operational conditions, such as elevated pressure and cryogenic temperature, often restrict its usage at a wider scale. Storing hydrogen in a metal or alloy via chemical absorption offers higher volumetric energy densities than compressed gas or liquid hydrogen at ambient conditions.

In this talk, I will present an ML-based framework HEART (**H**ydrog**E**n stor**A**ge prope**R**ty predic**T**or) that we recently developed for identifying suitable families of metal alloys for hydrogen storage under ambient conditions. The framework consists of two ML models that predict the hydrogen storage capacity and the enthalpy of hydride formation of multicomponent metal alloys. Our model includes absorption temperature as a feature which improved H<sub>2</sub>wt% prediction significantly. For out-of-the-bag samples, it predicted H<sub>2</sub>wt% with an R<sup>2</sup> score of 0.81 and mean absolute error (MAE) of 0.45 wt%, whereas the R<sup>2</sup> score is 0.89 and MAE is 4.53 kJ/molH<sub>2</sub> for ΔH. These models are further employed to predict H<sub>2</sub>wt% and ΔH for ~6.4 million multicomponent metal alloys. Taking a clue from our predictions, we have also synthesized ternary alloys, which show promising results with H<sub>2</sub>wt%~6.2 and appreciable reversibility. If time permits, I will discuss our plans to improve the existing model.

# Leveraging Data-driven Approaches to Accelerate Novel Materials Discovery

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## **Abstract**

Technological advancement often relies on the development of advanced materials. Therefore, the discovery of new materials with desired properties is an important but challenging task as trial-and-error methods of synthesizing novel compounds in the laboratory can be extremely labor, time, and cost-intensive. Computational methods combined with autonomous design strategy have the potential to reduce all three factors, significantly. In this talk, I will focus on the rapid discovery of a class of energy materials having low lattice thermal conductivities and promising thermoelectric performance using advanced computational methodologies. First, I will show how the rational design of an automatic workflow can accelerate the discovery of thermodynamically stable compounds based on high-throughput density functional theory calculations <sup>[1]</sup>. Then, I will demonstrate how a machine-learning model based on the crystal graph convolutional neural network can help discover the unsuspected compounds in the same family of materials, that would otherwise be difficult to do in conventional ways <sup>[2]</sup>. Recent experimental validation of some our predictions underpins the success and usefulness of computationally guided materials design approach. I will also show how data-driven discovery unravels novel physical insights behind the unusual thermal transport in some predicted compounds, providing new avenues for research and exploration. Lastly, I will briefly discuss efficient materials innovation strategy that can be achieved through the design of a closed-loop autonomous infrastructure capable of performing discovery, synthesis, and characterization of novel materials.

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# Inversion problem in density functional theory

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## **Abstract**

Density functional theory (DFT) is the most widely used theory for calculating electronic structure of materials. It is based on the profound theorem of Hohenberg and Kohn (HK) stating that the ground-state density of a many-electron system determines the Hamiltonian and hence the ground-state (and excited-state) wavefunction of the system. The DFT equation can be either the Euler equation purely in terms of the density or a Schrödinger-like equation for non-interacting electrons. The latter is known as the Kohn-Sham (KS) equation and is the mainstay of DFT calculations.

While the HK theorem proves the existence of a map from the ground-state density to the corresponding wavefunction, the inverse problem in DFT addresses whether it is possible to construct the Hamiltonian, or the many-electron wavefunction, or the KS equation from a given exact density, when those are available by solving the Schrödinger equation directly. This falls under the general inverse problems in physics such as finding the interaction potential from differential scattering cross section. The inverse problem is important to study the exact fundamental results of DFT presented in the first part. We present solution of inverse problem starting from constrained search formulation of DFT and how these are applied to in different situations such as adiabatic connection in DFT or to get the KS Hamiltonian for the given density. We then discuss inversions methods developed over the years using different functions of the density to obtain the KS Hamiltonian. Following this, we present our recent work where all these methods are shown to be arising from a single algorithm thereby demonstrating their universal nature. This then provides a general scheme of developing a variety of methods to get the KS Hamiltonian from a given density and we demonstrate this through examples. Interestingly, our work leads to use of random numbers for inversion which is also discussed. Our work also allows us to develop a general penalty method to carry out the density-to-KS system inversion and we show how this is done. We conclude by discussing possible future directions for research.

# A symmetry-based approach to identify structural prototypes and its application to perovskites

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## **Abstract**

We have exploited a methodology using group-subgroup relations to construct representative structural models for a given phase that satisfy a desired point symmetry<sup>[1]</sup> consisting of supercells built from combinations of lower-symmetry primitive cells. Using first-principles calculations we identify from these the structural prototypes: the smallest supercells that are both energetically and dynamically stable. This has allowed us to systematically explore and identify two structural prototypes for paraelectric BaTiO<sub>3</sub> which possess titanium displacements while maintaining a cubic point symmetry and, furthermore, correspond to phonon instabilities of the 5-atom-primitive cell.<sup>[2]</sup> We extend this work to lower symmetry phases as well as several other perovskites and further establish a connection between the phonon instabilities and the structural prototypes. This approach is general and can be used in ordered and disordered crystalline systems to find candidate templates and efficiently search for prototypes for use in the modelling and computational engineering of functional materials.

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# Designing Improved Cathode Materials for High-Capacity Sodium-Ion Batteries through Electronic Structure Tuning

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## **Abstract**

The design of cost-effective materials with improved properties is critical for addressing some of the most important problems in our society, ranging from energy storage to the reduction of fossil fuel-related harmful emissions. The activity, stability, and efficiency of materials for energy conversion and storage processes can be modulated by tuning their electronic structure as it dictates the majority of material properties.<sup>[1]</sup> Our results demonstrate that the electronic structure tuning method could be utilized to discover improved positive electrode materials for high-capacity sodium-ion batteries. Specifically, based on our detailed analysis of electronic structure properties (density of states), magnetic properties, chemical bond covalency, and molecular orbital diagrams, we propose key factors that regulate reversible oxygen oxidation (anionic redox) in Na-rich cathodes. Additionally, this approach can guide experiments toward identifying promising material candidates, thereby decreasing the use of human resources and efforts.

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# Manipulating Magnetism in Two-Dimension

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## **Abstract**

High-temperature ferromagnetism and its control in ultrathin materials are critical in disruptive quantum technologies. However, due to fundamental spin fluctuation in reduced dimensions, intrinsic ferromagnetic ordering in recently discovered two-dimensional materials is limited to cryogenic temperatures. This talk will discuss an experimentally feasible way to manipulate magnetism and elevate ordering to room temperature.<sup>[1,2]</sup> Next, we will discuss magnetism in a new platform, ultrathin non-vdW flakes derived from a quantum spin-liquid candidate, where both spin-orbit coupling and electron correlation are strong. The interplay between Heisenberg and Kitaev interactions will be explored, including the phase transitions observed in the charge-doped flakes.<sup>[3]</sup>

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# Stoichiometry dependent properties in Mn-based Heusler alloys: An *ab initio* study

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## **Abstract**

The electronic and magnetic properties of Heusler alloys are very sensitive to their crystal symmetry and the number of valence electrons. This opens up the possibility of large scale tuning of the properties facilitating diverse range of applications in fields like spintronics, shape memory effect, magnetic cooling effect, etc. The partially filled d-orbitals of the transition metal elements, which are the major constituents of the ternary intermetallic Heusler alloys, are responsible for many of their interesting properties. Lattice compatibility with the common major substrates enhances the possibility of device applications. Of particular interest are Mn-based Heusler alloys as prospective materials to be used in contemporary data storage devices.<sup>[1, 3]</sup> The efficiency of data storage devices requires a proper balance of many parameters, such as magnetic anisotropy, magnetization, spin polarization, and current density. Stoichiometry plays a very crucial role in balancing the parameters and in achieving the targetted characteristics.<sup>[2]</sup> *Ab initio* density functional theory gives a precise estimate of the parameters for a particular configuration of the system. However, screening for the suitable composition with optimum parameters giving efficient output from a large number of possibilities through *ab initio* calculations, is a computationally highly demanding task. One-time generation of a large database and application of optimized machine learning techniques for property-based screening followed by *ab initio* calculation on selected candidates for precise data is the present-day demand in this field of research.

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# Material-specific investigations of strongly correlated electron systems through DFT+DMFT

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## **Abstract**

Strongly correlated electron systems (SCES) have traditionally been investigated through effective, low-energy many-body Hamiltonians such as the Hubbard and the periodic Anderson model. While such toy models offer great insight into phenomena such as the Mott metal-insulator transition and heavy fermion physics, these types of investigations are not material-specific. The advent of dynamical mean field theory (DMFT) three decades ago followed by quantum cluster theories, opened possibilities of predictive modeling of SCES through a combination with density functional theory. In this talk, I will begin by reviewing some of the progress made through the DFT+DMFT approach. Subsequently, I will discuss our work on the development and application of a fast and inexpensive, albeit perturbative, impurity solver that yields results directly on the real frequency axis and scales polynomially with the number of orbitals<sup>[1]</sup>. I will conclude by discussing the utility of such a solver in large unit cell systems such as multi-orbital materials, disordered systems, and Moire lattices.

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# Controlling heat flow by manipulating phonons and their interactions: a bottom-up approach

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## **Abstract**

Microscopic interactions among quantized lattice vibrations called phonons govern the macroscopic thermal properties of semiconductors. In this talk, I will describe how our newly developed first-principles computational framework to predict these microscopic interactions of phonons unveils a new paradigm for heat conduction in these materials. As an example, I will first describe a curious case of heat conduction in boron arsenide (BAs), where the lowest order interactions involving three phonons are unusually weak and higher-order scattering among four phonons affects the thermal conductivity significantly, in stark contrast with commonly-used semiconductors such as silicon, germanium and diamond [1]. I will demonstrate that, in fact, this large effect of four-phonon scattering on thermal conductivity is fairly common among compound semiconductors, driven largely by selection rules for phonon scattering processes [2]. Next, I will show that these selection rules cause intricate competition between three and four phonon scattering processes, which can be exquisitely tuned to drive an unusual non-monotonic pressure dependence of the thermal conductivity in BAs and BP, unlike in most other materials [3, 4]. Finally, I will demonstrate the capability of our unified framework in capturing other phonon scattering processes, such as those caused by isotopic impurities, which resulted in the experimental realization of a giant isotope effect on the thermal conductivity of cubic Boron Nitride [5].

Our group's ongoing work is supported by a Young Investigator Award from the Infosys Foundation and the DST-SERB Core Research Grant (CRG/2020/006166)

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# Asymmetry-Driven Spin-Splitting for Spintronics and Topological Quantum Technology

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## **Abstract**

Asymmetry in the crystal structure or magnetic arrangement of material, combined with a sizable spin-orbit interaction, often leads to a host of interesting physical phenomena, including spin-splitting, spin-orbit torque, topologically protected states, and spin-Hall effect. Understanding the inversion and time-reversal asymmetry in the system leading to such phenomena becomes imperative for developing quantum technologies based on them. Taking examples of BiInO<sub>3</sub> surface and MnTe [1], we will demonstrate how the inversion and time-reversal asymmetry leads to spin-splitting assisted by spin-orbit interaction or even without spin-orbit interaction. We will also demonstrate the topologically nontrivial states arising from broken symmetry in the system. A comprehensive symmetry analysis based on group theory would help systematically understand these features and their applications for quantum technology.

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# Molecular Dynamics Simulations at the Fourth Rung of DFT Functionals

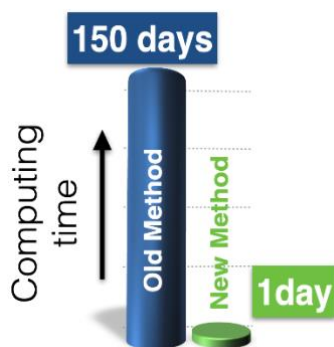
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## **Abstract**

Molecular dynamics (MD) simulations employing density functional theory (DFT) and plane waves are routinely carried out at the level of generalized gradient approximation (GGA). On the other hand, hybrid density functionals are more accurate and reliable than GGA functionals for predicting energetics and structural and dynamic properties. However, the computational cost for carrying out MD simulations using hybrid functionals and plane-wave basis sets is at least two orders of magnitude higher than that using GGA functionals for a system of ~100 atoms. Here we propose a method to reduce the computational cost of MD simulations with hybrid functionals to the extent that the computational cost is the same as that with GGA. This development has made it possible to study the mechanism and free energetics of complex chemical reactions in condensed matter systems through very long MD simulations at the level of hybrid density functionals.



# Machine learning driven DFT approach to understand and design new functional materials for energy technology

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## **Abstract**

This talk explores the synergistic combination of density functional theory (DFT) calculations and data-driven methods to understand and design materials with exceptional properties for a range of energy technologies. Specifically, we focus on materials for high-efficiency solar cells, photocatalysts for hydrogen production, multinary thermoelectrics with high Z, low cost and high capacity cathodes for rechargeable batteries alternative to lithium ion batteries, magnetic multicalorics, and hydrogen storage materials from non-hydrides. By harnessing the capabilities of DFT calculations, which provide atomistic insights into electronic structure and properties, along with data-driven methodologies, researchers can accelerate the discovery and optimization of materials for renewable energy technologies. Machine learning algorithms, statistical analysis, and high-throughput screening techniques play crucial roles in identifying important relationships between material properties and structural characteristics<sup>1</sup>.

This talk will highlight recent advancements and case studies that demonstrate the effectiveness of the combined DFT and data-driven approach in developing efficient and environmental friendly materials for sustainable energy technologies. It showcases how this integrated methodology has revolutionized the design and development of materials with enhanced performance, efficiency, and functionality. Examples include the discovery of novel solar cell materials with improved light absorption and charge transport, the identification of efficient photocatalysts for sustainable energy production, the optimization of thermoelectric materials for waste heat recovery, the exploration of alternative battery chemistries for energy storage, the design of hydrides for hydrogen storage from nonhydrides with high capacity, and the development of high-entropy alloys with exceptional mechanical and functional properties.

By bridging the gap between theory and experiment, this lecture provides valuable insights into the underlying principles governing the behavior of materials in diverse energy applications. It also discusses the challenges and future prospects of utilizing the combined DFT and data-driven approach, emphasizing the need for interdisciplinary collaboration and further research to unlock the full potential of advanced materials for energy technologies to have sustainable future. The lecture ultimately aims to inspire and encourage the audience to explore this integrated methodology for addressing critical energy challenges and advancing sustainable energy solutions.

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# Fast and accurate real-space finite-element based methodologies for Projector-Augmented Wave (PAW) formalism in density functional theory calculations

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## **Abstract**

The predictive capability offered by quantum modelling of materials, especially using density functional theory (DFT), has opened up a gateway for gaining crucial insights into materials' behavior, leading to an accurate prediction of mechanical, transport, chemical, electronic, magnetic and optical properties of materials. However, the stringent accuracy requirements required to compute meaningful material properties and the asymptotic cubic-scaling computational complexity of the underlying DFT problem demand enormous computational resources. Thus, these calculations are routinely limited to periodic simulation domains with material systems containing a few hundred atoms. Additionally, these DFT calculations occupy a sizeable fraction of the world's computational resources today but mostly remain in the high throughput calculation mode as the widely used DFT implementations struggle to keep up with evolving heterogeneous architectures in today's exascale era. To this end, the recently proposed finite-element-based DFT [1][2] methods -via- the DFT-FE code provide a systematically convergent, computationally efficient and scalable hybrid CPU-GPU framework for large-scale norm-conserving pseudopotential DFT calculations that overcomes these limitations with no restrictions on the boundary conditions that can be applied. However, these norm-conserving pseudopotentials for most material systems are known to be harder and require higher degrees of freedom (DoFs) to achieve the required chemical accuracy, leading to computationally expensive DFT calculations.

In this talk, we will discuss our group's ongoing work on developing a local real-space formulation of the projector augmented wave (PAW) method [3] that is amenable to finite-element (FE) discretization. Efficient and scalable computational methodologies for solving the underlying FE discretized PAW generalized eigenvalue problem will be discussed, along with accuracy and performance benchmarks on representative material systems. We demonstrate that our framework facilitates a substantial reduction in the degrees of freedom to achieve the required chemical accuracy, thereby enabling finite-element-based DFT simulations at significantly larger length scales and longer time scales, pivotal for addressing intricate challenges in complex materials problems.

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# Exploring the Quantum Frontier: Topological Insulators and Their Extraordinary Properties.

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## **Abstract**

In 2016, Nobel prize was awarded for discovery of topological phenomena in matter accounting for non-trivial phases and phase transitions. At the foundation we have the Kosterlitz-Thouless phase transitions (observed at ultra-cold temperatures) and the Haldane model of spin chains with non-trivial edges. The non-trivial phenomena in these cases were explained using the mathematical concepts of Topology. The former and latter have led to the development of topological states of matter; redefining our understanding of condensed matter. Ever since, topological phases such as, topological insulators, topological semi-metals, topological superconductors, topological Kondo insulators etc. have been at the forefront of quantum materials unravelling unconventional physics. These materials find applications in numerous field such as, spintronics, nano-electronics, quantum computations, valleytronics, sensors, photodetectors, quantum catalysis etc. Especially, the topological insulators fall under the category wherein the material can host multi-functional properties making them even more promising. The topological insulators are characterized by non-trivial phenomena such as, insulation in  $N$  dimensions and conduction across  $N-1$  dimensions. Such phenomena have been predicted and observed experimentally in bulk as well as low dimensional materials. Such non-trivial phase of matter can be observed either intrinsically or by employing various techniques such as, band engineering using strain/pressure, electric fields, functionalization (partial/complete), dimensional engineering etc. Here, we present, non-trivial topological quantum phases of matter such as topological insulators and different mechanisms by which we can realize such systems in bulk and low dimensional materials. We then discuss the future prospects in terms of applications of non-trivial topological quantum phases of matter in different fields such as, quantum catalysis and superconductivity

# Challenges ahead of ML-DFT: Additions within and beyond the Jacob's Ladder of DFT from Semilocal through Dielectric Dependent Hybrids

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## **Abstract**

Density functional theory (DFT) is often the go-to computational method for solving the electronic structure problems of molecules, solids and materials. Although it is an exact theory, one must approximate the exchange-correlation (XC) part, which includes all the many-body interactions. The development of new XC approximations having insightful physical content and which are also accurate as well as efficient for solids is always desirable. Nevertheless, accurate density functional approximations are always the starting point of machine-learning-based functional developments, automated, and high-throughput computational work flow for quantum simulations and material modeling. In our work, we have shown how our developed density functional approximations eliminate some of the drawbacks of earlier proposed XC functionals and one can improve step-by-step the density functional approximations starting from the kinetic energy density functional (KEDF), semilocal DFT, hybrid density functionals to dielectric-dependent hybrids<sup>1-5</sup>. To summarize, our work will pave way for the future directions of further developments and challenges faced along with new horizons.

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# C-vacancy Mediated Methane Activation and C-C Coupling on Titanium Carbide Surface

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## **Abstract**

Methane, the main component of natural gas, is one of the major greenhouse gases contributing to global warming. Therefore, capturing methane and converting it to other useful products is highly desirable. Methane activation is challenging due to the high energy of the C-H bonds and the non-polar, non-reactive nature of the molecule. In this work, using density functional theory based calculations and ab initio thermodynamic analysis, we have studied the role of C-vacancies in TiC(001) surface towards methane activation and its non-oxidative coupling to form C<sub>2</sub> hydrocarbons. Our C-vacancy concentration dependent study of CH<sub>4</sub> activation shows that (i) the first C-H bond cleavage is facile and less sensitive to the concentration of C-vacancy and (ii) the dissociation of the subsequent ones strongly depend on the vacancy concentration and becomes arduous in presence of fewer vacancies. Amongst the two vacancy concentrations considered in this study, namely, 12.5% and 25%, we find that on the former though the first C-H bond cleavage is facile, the barriers for the subsequent C-H bonds are high suggesting that this might be a good candidate for further C-C coupling studies. Our C-C coupling studies show this catalyst will yield acetylene at around 800 K. However, the rate limiting step is the formation of H<sub>2</sub> from the H atoms occupying the C-vacancies, which might block the vacancies thereby deactivating the catalyst.

# Machine Learning aided exploration of materials space and inverse design of materials

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## **Abstract**

The traditional trial-and-error experimental approach to materials design is time consuming, expensive and uncertain. Over the past couple of decades, rapid progress in first principles calculation methods, and computational hardware and software have helped accelerate materials design and discovery. High throughput computing strategy has been particularly helpful in these efforts. Yet, a comprehensive search of the materials space to identify materials with a desired set of properties requires large computational resources, and is time consuming.

In recent years, Machine Learning methods have emerged as an alternative to expensive first principles calculations. Models can be trained on available materials data generated either from experiments, via high throughput computing or by crowd sourcing from the community of researchers, to predict one or more properties.

We have devised a strategy to use available materials data for efficient exploration of the materials space. In this approach, a series of Machine Learning models are trained to either predict some material property (e.g, heat of formation) or to classify materials according to some property (e.g., magnetic vs. non-magnetic). These models are used to hierarchically screen a set of new materials, which can be either existing ones, or virtually designed using our domain knowledge, to identify the ones that satisfy all the desired properties. The small set of materials identified via this screening procedure is tested in DFT. Materials passing the DFT test are proposed as new candidate materials with specific functionalities. We have used this strategy to design new rare earth free magnets with large magnetization and anisotropy energy.

In a second strategy, we design materials with a set of target properties using a constrained variational auto-encoder (cVAE), which is a generative deep learning model. We again attempt to generate new magnetic materials with large anisotropy.

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# Computationally Predicted Efficient Energy Materials

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## **Abstract**

Fast diminishing fossil fuel reservoir, environmental concerns and the requirement to meet the ever growing energy demand, prompt for a rigorous search for efficient energy materials that may lead to the development of economical, effective, and competent devices for renewable energy generation and storage. Traditionally experimental studies have led to the discovery of new promising materials but most of these studies are based on trial-and-error and serendipity. On the other hand, in current time development of computational resources and methods have taken a lead in reliable prediction of promising materials in a cost and time effective manner, providing an efficient route to materials discovery. Using such methods (evolutionary algorithms and machine learning) with the first-principles density functional theory based approach as the core, we comprehensively study a variety of materials and predict efficient materials for rechargeable batteries and solar cells. While one of them has covered the journey from prediction to experimental realization, others may guide experimentalists to realize the next-generation efficient energy devices.

# Why do twisted bilayers behave differently from their untwisted counterparts?

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## **Abstract**

The Mo and W based transition metal dichalcogenides have been known for several decades as examples of semiconductors whose electronic structure is well described by band theory. It is only recently that one finds unusual phenomena arising in them on doping holes via gating into twisted bilayers [1], an aspect that we would associate with correlated materials. We have recently examined the electronic structure of twisted bilayers of Mo [2] and W [3] based transition metal dichalcogenides. In contrast to graphene, we find the emergence of flat bands for several angles of rotation. The origin of this can be linked to patches of various types of stackings which include an atom-on-atom as well as a staggered stacking. The former lead to larger inter layer separations because of the larger repulsion between the electrons in the two layers in contrast to the latter. This leads to larger perturbations in some regions of the moire cell. Building on the fact that these materials represent van der Waals structures, and so the perturbation induced by one layer on the other should be small, we explore different twist angles and quantify the perturbation in each instance from the untwisted limit. Surprisingly, at large twist angles we find that we recover the low energy electronic structure of the untwisted limit, while at small angles we find flat band formation as well as other unusual aspects of the electronic structure.

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# Confined two-electron atomic systems without using any cut-off factor in the wavefunction explicitly

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## **Abstract**

We propose a new method of studying confined two-electron systems using an explicit form of the wavefunction. We solve the Schrödinger equation directly for calculating the part of wavefunction needed for satisfying the required boundary conditions, and, thus, avoiding the use of any cut-off factor in the wavefunction explicitly. We demonstrate our method by comparing the results for kinetic energies, electron-nuclear interaction, electron-electron interaction, total energies, densities, pressures, and moments of the confined  $H^-$ , and He atom with the most accurate results available in the literature. The confinement potential we consider in this study are the spherical well and harmonic potential.



# Computational materials science and its applications in the area of materials for energy

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## **Abstract**

Energy storage has been a theme for scientists for two hundred years. The Lead acid battery research on batteries occupied some of the best minds of the 19th century. Gaston Plante in 1859 invented the lead acid battery which starts your car and ignites the internal combustion which takes over the propulsion. Although the lead battery is over 150 years old but the origin of its open circuit voltage (OCV) of 2.1 V is still known. In present talk, I will show how one can explain the origin of OCV of 2.1 V based on foundations of relativistic quantum mechanics. Surprisingly, seems to be the first time its chemistry has been theoretically modeled from the first principles of quantum mechanics. The main message of this work is that most of the electromotoric force (1.7-1.8 Volts out of 2.1 V) of the common lead battery comes from relativistic effects. While the importance of relativistic effects in heavy-element chemistry is well-known since over two decades, this is a striking example on "everyday relativity". We believe that the fact that "cars start due to relativity."

The purpose of this talk is to provide an overview of the most recent studies in the field of hydrogen storage materials & rechargeable battery research with the focus how computational material science can play an important role in search and design of new hydrogen storage materials & next generation battery materials. On specific examples, the application of density functional theory calculations and molecular dynamics simulations will be illustrated to show how these computational methods can be of great use in the effort to reach a better understanding of materials and to guide the search for new promising candidates.

# A theoretical framework to analyze and tailor electronic and optoelectronic properties of halide perovskites

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## **Abstract**

Single and double halide perovskites (HP) have truly emerged as efficient optoelectronic materials since they display superior stability and are free of toxicity. However, challenges still exist due to either wide and indirect bandgaps or parity-forbidden transitions in many of them. The lack of understanding in chemical bonding and the formation of parity-driven valence and conduction band edge states have hindered the design of optoelectronically efficient HPs. In this talk, we will develop a theoretical framework using a multi-integrated approach involving ab-initio density functional theory (DFT) calculations, model Hamiltonian studies, and molecular orbital picture leading to momentum matrix element (MME) estimation. This framework gives us a detailed insight into chemical bonding and parity-driven optical transition between edge states. We will talk about a band-projected molecular orbital picture (B-MOP) connecting free atomic orbital states obtained at the Hartree-Fock level and orbital-resolved DFT bands. From the B-MOP, we will show that the nearest neighbor cation-anion interaction determines the position of atom-resolved band states, while the second neighbor cation-cation interactions determine the shape and width of band dispersion and, thereby, MME. The latter is critical to quantify the optical absorption coefficient. Considering both B-MOP and MME, we will demonstrate a mechanism of tailoring bandgap and optical absorptions through chemical doping at the cation sites. Finally, the cause of bandgap bowing, a common occurrence in doped HPs, will be explained by ascribing it to chemical effect and structural distortion.

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# Electronic Descriptor then Predictive model using QM/ML Approach then Experimental Validation

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## **Abstract**

Energy storage and conversion devices including metal–air batteries and fuel cells are highly dependent on cathodic Oxygen Reduction Reaction (ORR) and Oxygen Evolution Reaction (OER). The carbon-based materials are identified as potential catalysts for ORR/OER with ideal electronic properties but the random approach using an experimental method delays the finding of the best carbon catalysts in large material space. To overcome this challenge, we proposed a predictive model equation based on QM/ML approach that can find the site-specific activity of the graphene-based system. Here, we demonstrate the key role of  $\pi$  orbital descriptors ( $D\pi(EF)$ ,  $R-O\pi$ ) to influence the adsorption property of carbon sites (1-3). By using these descriptors as features, we employed various ML models and observed the optimal predictive performance of the SVR method. We applied SVR based predictive model to estimate the ORR/OER activity of unknown graphene systems and the accuracy of the results is validated by DFT calculations.

I will discuss the validation of the prediction with experimental results (collaborative work). 66 different possible carbon active sites of three models of doped and defective graphene. To find the theoretical overpotential on 66 sites, **we need to do 198 DFT calculations** (three intermediate for each site). Which needs huge resources and is also time-consuming. But by applying our SVR model we can predict the  $\Delta G_{OH}$  values and overpotential for these active sites by doing **three DFT calculations** of the host surface only (4). I will discuss the journey from fundamental to experimental proof during my talk.

Note: The work is in collaboration with multiple groups as seen from the references

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- [3] Sakshi Bhardwaj, Samadhan Kapse, Soirik Dan, **Ranjit Thapa** and Ramendra Sundar Dey, **Journal of Materials Chemistry A**, In Press 2023. <https://doi.org/10.1039/D3TA00871A>

# Machine Learning Models and Potentials for Shear Viscosity of Fluids

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## **Abstract**

The shear viscosity of fluids is computationally expensive to calculate from equilibrium molecular dynamics simulations. However, Machine Learning (ML) methods to estimate it face significant challenges such as overfitting due to the small size of data sets. In this work, we train seven ML models to predict the shear viscosity of a Lennard–Jones fluid, with particular emphasis on addressing issues arising from a small data set. Specifically, the issues related to model selection, performance estimation, and uncertainty quantification were investigated. We find that the k-fold cross validation (CV) based procedure shows a lower variance of error estimates. Finally, two methods for uncertainty quantification—Gaussian process regression (GPR) and ensemble method—were used to estimate the uncertainty of individual predictions. The uncertainty estimates from GPR were also used to construct an applicability domain using which the ML models provided even more reliable predictions on an independent viscosity data set generated in this work.<sup>[1]</sup>

In the second part of the talk, a neural network potential (NNP) to model aqueous alkali halide solutions based on density functional theory data is presented. Molecular dynamics simulations using this NNP reproduce the retardation and acceleration of water dynamics ('inverse-viscosity') in sodium chloride solutions and cesium iodide solutions with increasing salt concentration respectively. The microscopic underpinning of these varied behaviors will be presented.<sup>[2]</sup>

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# Modelling complex configurational spaces with machine learned interatomic potentials

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## **Abstract**

Most commercial high-energy-density cathodes for rechargeable lithium batteries are made of ordered, layered materials that have specific arrangements of lithium and redox-active transition metals. The potential of cation-disordered materials, specifically disordered rocksalt cathodes that can have a broad variety of compositions, has been established in recent years, including their ability to transport lithium via percolating channels resulting in high capacities, and activation of anion redox resulting in higher voltages. Thus, disordered rocksalts can yield storage devices with high energy densities. However, disordered materials are challenging to model using conventional density functional theory (DFT) based calculations because of their scale and configurational complexity. Machine learned interatomic potentials (MLIPs), which can exhibit computational speeds similar to classical force fields and accuracies similar to DFT calculations, exhibit the advantage of modelling large-scale systems over long time scales. Hence, MLIPs can be used to model configurationally complex systems such as disordered cathodes, but have to be carefully constructed and rigorously benchmarked. Here, we comprehensively evaluate the performance of four different MLIP frameworks, including neural network potentials, tensor-based potentials, and Gaussian, and spectral analysis potentials. To train the MLIPs, we used a diverse DFT-calculated data set comprising LiTMO<sub>2</sub> ordered and disordered rocksalts, where TM can be any combination of Sc, Ti, V, Cr, Mn, Fe, Co, Ni, and Cu. We demonstrate that the neural network potentials train and predict total energies accurately while tensor-based potentials are better for force predictions. Also, we quantify the accuracy-computational cost trade-off for each MLIP. The MLIPs developed in our work can pave the way for the study of cation-disordered systems, besides rocksalts, which can aid in designing energy-dense storage systems.

# Unsupervised Learning Driven Discovery of 2D Charge-Density-Wave Materials

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## **Abstract**

Progress in artificial intelligence and machine learning and related examples of superhuman performance have fueled expectations that AI/ML will revolutionize scientific discoveries in general and materials science in particular. In this talk, we discuss how unsupervised machine learning techniques may lead to discovery of 2D charge-density-wave (CDW) materials, which has potential application in electronic switches. We first discuss ‘mean-shift-algorithm’ - a non-parametric, density-based clustering algorithm that can be used to identify clusters in a dataset. Next, we discuss Ab initio Random Structure Searching (AIRSS) technique for predicting atomic structures near local or global energy minima. We then see how these two techniques could be combined to develop a fully automated high-throughput computational framework, which identifies CDW phases from a unit cell with inherited Kohn anomaly. We will see that the proposed methodology not only rediscovers the known CDW phases but also predicts a host of easily exfoliable CDW materials (30 materials and 114 phases) along with associated electronic structures. We will pay special attention to ZrTiSe<sub>4</sub> and conduct a comprehensive analysis to gain insight into the Fermi surface nesting, which causes significant semiconducting gap opening in its CDW phase.<sup>[1]</sup>

[1] A. Kabiraj and S. Mahapatra, *J. Phys. Chem. Lett.* 2020, 11, 6291–6298

# Computational Design of Sustainable Materials: From Super Capacitor to Solar Energy

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## **Abstract**

One of the sustainable development goals (SDG-7) by the United Nations General Assembly is to ‘Ensure access to affordable, reliable, sustainable and modern energy for all’. Energy from the sun (solar energy) is clean energy and due to its abundance, it is highly scalable. Energy storage is a big part of the total energy solution. Emerging materials such as graphene and other 2-dimensional materials provide advanced storage options such as through batteries, supercapacitors, etc. This talk will present our recent computational study of the development of such materials in collaboration with experiments. For example, a novel approach was applied for the development of Aluminum graphene (AlGra) composite materials for the fabrication of energy storage devices such as supercapacitors, and batteries that are affordable and environmentally friendly. In collaboration with MIT, USA, and through our start we introduced the world’s first lightweight and rollable solar panel for mobile irrigation that could provide clean energy solutions for livelihoods around the world. This talk will provide an overview of our energy research and its translations for societal impact.

# Accelerated Neural Network Training through Dimensionality Reduction for High Throughput Screening of Quantum Materials

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## **Abstract**

In the first part of the talk I will introduce a mechanism that consists of machine learning methods<sup>[1]</sup> to discover novel quantum materials. Neural Networks are currently limited by the size of data provided and the huge computational cost of training a model, especially when dealing with a large set of features without much prior knowledge of how good or bad an individual feature is. We tackle this problem using Dimensionality Reduction algorithms to construct more meaningful features. A study is performed to compare the accuracy and training times of raw features and transformed features to deduce a sufficient number of dimensions without sacrificing accuracy. This is accomplished using AdaBoost, as it trains faster than Neural Networks. Finally, Neural Networks are trained for different systems of compounds using the transformed features and predictions arrived at. The accuracy of this prediction are thoroughly benchmarked and duly validated followed by proposing promising candidates for further research.

In the second part of the talk I will present a discovery of persistent spin texture (PST) that maintain a consistent spin configuration in momentum-space. Our findings reveal the emergence of full-plane PSTs in the  $k_z=0$  and  $k_z=\pi/c$  planes, distinct from the previously observed locally existing PSTs in bulk materials<sup>[2]</sup>. These full-plane PSTs are protected by the in-plane mirror and three-fold rotation symmetry. The purely cubic nature of split bands offers distinct advantages in spin transport. Through high-throughput calculations based on the DFT and k.p models, we identify a significant class of materials that exhibit the aforementioned properties. We demonstrate the achievement of PST throughout the full-zone by inducing a dispersionless band along the z-direction using a layered prototype material  $WSi_2N_4$ <sup>[3]</sup>. The observed PSTs provide a pathway to non-dephasing spin transport with enhanced spin-Hall conductivity, presenting a promising platform for future spintronics.

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# Unlocking materials properties: Exploring approaches from symbolic regression to deep learning

**Satadeep Bhattacharjee**<sup>1\*</sup>, Kishalay Das<sup>3</sup>, Swetareakha Ram<sup>1</sup>, Pritam Das<sup>1</sup>, Pawan Goyal<sup>3</sup>, Seung-Cheol Lee<sup>1,2</sup>, Niloy Ganguly<sup>3</sup>

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## **Abstract**

This presentation highlights the application of machine learning in computational materials science and provides an introduction to its techniques and their importance in accelerating materials discovery and design. The concept of so-called transparent and less transparent approaches to modeling and prediction is explored, and the benefits and trade-offs of each are highlighted. Particular emphasis is placed on the transparent approach, which focuses on symbolic regression and its historical relation to physics, as well as its relevance in modern materials science. Our research results are presented, demonstrating the effectiveness of such transparent, interpretable models in predicting magnetic moments of Heusler alloys, establishing adsorption energy scaling relationships in 3d transition-metal-based catalysts, estimating overpotentials, and making general predictions about adsorption energies. Moving to less transparent approaches, methods based on deep neural networks are introduced, focusing on pre-training and its impact on prediction. Our recent research results with such approaches are presented.

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# New routes to control skyrmions in ultrathin transition-metal films

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## **Abstract**

Magnetic skyrmions have drawn immense attention in recent times as they promise to be the building blocks of future storage devices, neuromorphic and quantum computers due to their nanoscale size and low energy consumption. The protection arising from topological nature of the spin structure gives them an extra stability against external perturbations. The associated topological charge can be used to represent the classical bits “0” and “1”. Recently, various schemes to design skyrmion qubits have been proposed and one and two qubit gate operations have also been demonstrated theoretically. However, the key challenge to realize a skyrmion based data processing and storage device is the robustness of the information bits, i.e., stability of skyrmions against thermal fluctuations.

Skyrmions are stabilized in ultrathin transition-metal films due to the interplay among the pairwise Heisenberg exchange, dzyaloshinskii-moriya and magnetic anisotropy interactions. The higher-order multi-spin exchange interactions (HOI) beyond the Heisenberg pair-wise interaction, were seen to stabilize magnetic ground state in ultrathin transition-metal films. However, they are not believed to be important for a metastable structure such as skyrmions, since they arise from fourth order perturbation theory.

Using a multiscale approach via combining the atomistic spin dynamics and density functional theory, I would show that aforementioned HOI have a significant impact on the stability of skyrmions in transition-metals films. Thus, the higher-order terms are extremely important for skyrmions stability, opposite to earlier thought, and would provide a new route to control these structures. Next, I would apply an external electric-field, which would create and annihilate skyrmions, avoiding the Joule heating effect- as required for storage devices and quantum application. I will highlight the microscopic mechanism of this creation and annihilation processes.

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# Hybrid quantum-classical periodic embedding

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## **Abstract**

In this talk, I will delve into the details of our ongoing efforts to implement a hybrid quantum-classical periodic embedding scheme based on the long-range wave function in short-range DFT approach, as discussed in [1, 2]. To give you a better understanding of the subject, I will provide a comprehensive overview of the theoretical foundations of the methodology and discuss the interface between CP2K [3] and the IBM quantum device API, qiskit-nature. The challenges associated with such a hybrid quantum-classical interface that we encountered during the implementation process will also be presented, along with the issues associated to integrate a periodic description of the embedding. Finally, I will present the first results obtained for small benchmark systems that validate the implementation and provide an overview of the future directions that we are aiming at.

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# Computational modeling of electrochemical capacitance of Nitrogen doped $Ti_3C_2T_x$ supercapacitor electrode in acidic electrolyte

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## **Abstract**

Using Density functional theory in conjunction with a solvation model we have investigated the phenomenon of electrode-electrolyte interaction at the electrode surface and its consequences on the electrochemical properties like the charge storage and total capacitance of nitrogen doped  $Ti_3C_2T_x$  supercapacitor electrode. in acidic electrolyte  $H_2SO_4$  solution. By considering nitrogen doping at different sites, we explored the dominant mechanism behind the changes in the capacitances. Our results agree well with the available experiments and provided necessary insights to understand the effects of nitrogen doping at different sites on the electrochemical properties of this MXene electrode.

# Computational Roadmap of Hybrid Perovskites Materials: Insight from Rashba Effect and Piezochromism

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## **Abstract**

In this talk, I would start with the brief introduction of first principles electronic structure calculations within the framework of Density Functional Theory (DFT) in hybrid perovskite materials, and how it could be connected to the Computational Screening for achieving highly efficient and stable solar cell materials [1, 2]. Next, I will be talking about the fundamentals and possible implications of Rashba phenomena in hybrid perovskite materials [3, 4]. The rest of the talk would be devoted to the theoretical understanding of piezochromism in lead free metal chalcogenide perovskites. Hydrostatic pressure is an effective tool, which can give rise to novel crystal structures and physical properties, while it has proven to be an alternative to chemical pressure. Therefore, new functional materials with intriguing properties can be designed by exerting external pressure. We have recently [5,6] envisaged the structural, electronic and optical properties under the influence of hydrostatic pressure along with the effective mass evolution of the charge carriers.

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# Computational Modeling of Materials for Energy Conversion

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## **Abstract**

In this talk, I will mainly focus on three of our works on modeling thermoelectric and catalytic materials for energy conversion. Two of these are on finding efficient materials for oxygen evolution reaction (OER) & oxygen reduction reactions (ORR) and hydrogen evolution reactions (HER). While the former two are involved in oxygen-air batteries and metal-air batteries (chemical energy storage), the latter is a water splitting process.

To compute thermal conductivity and thermoelectric efficiency, we have developed computational codes based on Boltzmann transport equations. We have computed the thermoelectric efficiency of a few interesting materials, namely, p-type SnO-PbO super lattice, and n-type ternary Ag<sub>2</sub>Se systems with S and Te doping<sup>[1]</sup>. I shall compare and contrast results on thermal conductivity values of two sets of half-Heusler alloys, namely, TiRhBi with TiCoBi and NbFeSb with NbFeIn, computed by considering 2 and 3 phonon scatterings<sup>[2]</sup>.

We have modeled catalytic materials using ab-initio DFT methods. We have considered tetrazine based covalent organic framework and charge transfer modulated hetero-interfaces for HER<sup>[3]</sup>. We also have worked on transition metal-nitrogen embedded graphene as bifunctional electrocatalyst for OER and ORR. Working on single atom catalysts on graphitic carbon nitride, we have modeled multifunctional (OER, ORR, HER) behavior of a few efficient electrocatalysts<sup>[4]</sup>.

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# Detecting water deep inside the earth's mantle

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## **Abstract**

The earth's mantle transition zone (MTZ) which lies at a depth of 410-660KM is potentially hydrated. Laboratory experiments suggest that the two dominant mineral phases in the region, namely, wadsleyite ( $\beta$ -(Fe,Mg)<sub>2</sub>SiO<sub>4</sub>) and ringwoodite ( $\gamma$ -(Fe,Mg)<sub>2</sub>SiO<sub>4</sub>) can store upto 3.3 wt% water in their crystal structure in the form of hydroxyl ions. Using first principles density functional theory we study the water incorporation mechanism in wadsleyite and ringwoodite under MTZ conditions and evaluate the effect of Fe and H<sub>2</sub>O on the elastic properties (and hence on P and S wave velocities) of the two minerals.

One of the most effective and hence prevalent methods for determining the state of hydration of the mantle is through seismic studies. It is now understood that hydration in general leads to a reduction in P and S wave velocities, a property which is used to gauge the amount of water deep inside the earth. However, it has been indicated that increased pressure may lead to a "suppression" of the effect of hydration on the sound wave velocity. This has raised doubts on the ability of seismology to detect water deep inside the MTZ, which is under very high pressure (~13-24 GPa) and temperature (~1500<sup>0</sup>C to 2000<sup>0</sup>C). Our calculations indicate that under MTZ pressure and temperature conditions there is indeed a "suppression" in the P and S wave velocities. However, in the case of wadsleyite, the suppression is not very significant, and seismic waves may be able to detect water content which is as low as 1.56 wt%. On the contrary, in the case of ringwoodite, the suppression is found to be significantly large. Our calculations indicate that it may not be possible to make a distinction between anhydrous ringwoodite and 1.56 wt% water containing ringwoodite using seismic tools. However, when the water content is significantly large (~3.3 wt%), seismic waves are found to be robust enough to detect the state of hydration of the MTZ that corresponds to the stability field of ringwoodite. We conclude that one can easily identify regions of high water concentration (>1.56 wt% in the case of wadsleyite and >3.3 wt% in the case of ringwoodite) in the MTZ using the seismic lens.

# Machine Learning Approach of Design of New Materials with Targeted Properties

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## **Abstract**

One of the strong pillars in advancement of designed materials, with targeted properties like magnetism, superconductivity, topological characters is computation of materials. The synthesis and optimization of properties of real materials in experiment is both time-consuming and costly, being mostly based on trial and error. Computational approach in this connection is of natural interest to screen materials, before they can be suggested and tested in the laboratory. For the prediction of new materials, a powerful tool is the machine-learning assisted high throughput computation. In this approach new materials have been computationally predicted by combining electronic-structure methods with intelligent machine learning technique based on data mining and database construction. In this talk we discuss application of this method for prediction of new magnetic double perovskites [1], low cost rare earth based permanent magnets [2] and semiconductor heterostructures,[3] binary nanoalloys.[4]

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# On the Data-Driven Design of Stable, Active, Selective, and Cost-Effective Nanocatalysts for the Oxygen Reduction Reaction

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## **Abstract**

Designing nanoparticles (NPs) with density functional theory (DFT) is typically performed using periodic slab approximations, as DFT can handle around  $N=200$  atoms with computational effort scaling as  $O(N^4)$ . Hence DFT-based approaches are too computationally demanding to screen NPs of arbitrary shape, size, and composition. We employ the alpha scheme, a surrogate model parametrized by DFT, to design stable and active NPs that catalyze the oxygen reduction reaction (ORR).

By screening across NPs of arbitrary structures, our surrogate model can identify those NPs with specific structures that meet operational constraints for ORR. We first determine the cohesive energy of the nanoparticle to evaluate its overall stability. We then calculate the stability of individual metal atoms on the nanoparticle, on-the-fly. These atomic site stabilities serve as descriptors for reactivity metrics like the free energy of adsorption of hydroxyl ( $\Delta_{\text{ads}}G_{\text{OH}^*}$ ). Scaling relations between the oxygenates  $\text{O}^*$ ,  $\text{OH}^*$  and  $\text{OOH}^*$  are then capitalized to compute the free energy of adsorption of other oxygenates. These free energies are, in turn, used to calculate the limiting potential for oxygen reduction and estimate the current density at each active site. The microscopic reactivity metrics are averaged over the nanoparticle, yielding macroscopic metrics like the mass-averaged current density.

Using this data-driven approach, we screen across  $> 5000$  Pt-based nanoparticles with diameters between 2 (300 atoms) to 8 nm (10000 atom) and in diverse shapes (cubooctahedrons, octahedrons, decahedrons, icosahedrons, and Wulff constructions). Considered compositions include random alloys, core-shell, and edge-decorated architectures. We identify architectures that yield mass specific activity higher than the Pt (111) benchmark. Our workflow provides an intrinsic link between the atomistic properties of each NP and macroscopic observables, thus enabling the high-throughput screening of an ensemble of NPs with near-DFT accuracy and significantly lower computational effort.

# Metavalent Bonding Origins of Unusual Properties of Group IV Chalcogenides

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## **Abstract**

A distinct type of metavalent bonding (MVB) was recently proposed to explain an unusual combination of anomalous functional properties of group IV chalcogenide crystals, whose electronic mechanisms and origin remain mysterious. Through theoretical analysis of evolution of bonding along continuous paths in structural and chemical composition space, we demonstrate emergence of MVB in rocksalt chalcogenides as a consequence of weakly broken symmetry of parent metallic simple-cubic crystals of Group V metalloids. High electronic degeneracy at the nested Fermi surface of the parent metal drives spontaneous breaking of its translational symmetry with structural and chemical fields, which open up a small energy gap and mediate strong coupling between conduction and valence bands making metavalent crystals highly polarizable, conductive, and sensitive to bond-lengths. Stronger symmetry breaking structural and chemical fields, however, transform them discontinuously to covalent and ionic semiconducting states respectively. MVB involves bonding and antibonding pairwise interactions alternating along linear chains of at least five atoms, which facilitate long range electron transfer in response to polar fields and cause unusual properties. Our precise picture of MVB predicts anomalous second order Raman scattering as an addition to set of their unusual finger-printing properties, and will guide in design of new metavalent materials with improved thermoelectric, ferroelectric and nontrivial electronic topological properties of relevance to quantum technologies. <sup>[1]</sup>

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# Exploring Quantum Emergence: Unveiling the Phenomena and Properties of Quantum Materials

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## **Abstract**

Quantum materials, with their intriguing properties and potential applications, have emerged as a captivating field of study in condensed matter physics. These materials display remarkable quantum mechanical phenomena resulting from the intricate interactions between electrons, giving rise to extraordinary electronic, magnetic, and topological characteristics. Understanding and harnessing the unique characteristics of quantum materials hold great promise for revolutionizing various technological domains, including energy, computing, and information storage. The investigation of electronic structure, precisely the topological properties of quantum materials have received a significant attention in current research. The unusual physical phenomena of perovskite materials' due to presence of complex structural characteristics and the interplay of charge, spin, and orbital degrees of freedom, encouraged us to study the structural, magnetic, electronic properties, orbital ordering, and spin-phonon coupling phenomena in the layered perovskite-type  $AMnF_4$  ( $A=K, Rb, Cs$ )<sup>[1,2]</sup> from first principles. Topological metals/semimetals have received a lot of attention recently due to their unusual properties both under normal and high pressure, although they have not been extensively studied. One such compound with the presence of non-trivial topological properties robust to high pressure is  $MnRhP$ <sup>[3]</sup>. We would like to discuss the topological features present in both electronic and phononic band structures of the  $ZGeSb$  ( $Z=Hf, Ti, Zr$ ) class of compounds<sup>[4]</sup>, which is quite uncommon in materials. In addition, we would like to focus on the Anomalous transverse effects of a few Heusler compounds<sup>[5-7]</sup>. The study of electronic structures, including the investigation of topological characteristics, provides insights into the fundamental behavior of quantum materials and paves the way for the development of advanced devices with unprecedented functionalities.

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# Tuning the electronic structure of a twisted transition metal dichalcogenide heterotrilinear with a vertical electric field

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## **Abstract**

Recently, moiré superlattices generated by twisting two or more layers of semiconducting transition metal dichalcogenides (TMDs) have attracted great attention due to their ability to host strongly correlated states, such as Mott insulators at half-filling (one hole per moiré cell) and generalised Wigner crystals at fractional fillings[1-6]. The highest lying narrow valence bands in these systems are derived either from the monolayer states in the K/K' valley or alternatively from monolayer states in the G valley.

Here, we demonstrate using first-principles density-functional theory calculations that the natural order of K/K' – derived and G – derived valence bands in a heterotrilinear consisting of a natural 2H – WSe<sub>2</sub> bilayer stacked on a twisted MoSe<sub>2</sub> monolayer ( $q = 3.1^\circ$ ) can be controlled by a perpendicular electric field. Interestingly, the wavefunctions of the top two valence bands (G – derived bands) are delocalized over the two WSe<sub>2</sub> layers and realize a honeycomb lattice at the moiré scale with inequivalent A and B sites, i.e., a moiré equivalent of hexagonal boron nitride; whereas K/K' – derived bands give rise to a triangular lattice at the moiré scale and are fully localized on specific layers. From these *ab initio* insights, we construct an effective Hamiltonian to study the correlated hole states that arise when the system is doped. Our results show that twisted heterotrilinear TMDs are an ideal platform to investigate Hubbard models on both triangular and honeycomb lattices in the same system and are in agreement with experimental measurements [7].

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# Probing the role of compression rates in the pressure-induced polymerization of crystalline acrylamide through *ab initio* molecular dynamics

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## **Abstract**

Pressure-induced polymerisation in molecular crystals can be viewed both as a chemical reaction as well as a solid-to-solid phase transition. Hence, both thermodynamic stability of the phases as well as kinetics of the transformation are equally important to understand the phenomenon. This is particularly so for crystals with weak interactions where many metastable structures, corresponding to local minima in configuration, can arise under pressure. Intrigued by various experimental reports<sup>1-5</sup> of dependence of polymorph formed on the compression rate in pressure-induced structural transformation, we have investigated the effect of pressure increase rate on the response of acrylamide crystal to hydrostatic pressures up to ~100 GPa at room temperature, employing *ab initio* molecular dynamics (AIMD) based on van der Waals corrected density functional theory. Our study<sup>6</sup> not only reveals the possibility of multiple polymeric polymorphs under rapid compression, but also details the mechanism of their formation along with their electronic and spectroscopic properties. Interestingly, the polymerization is facilitated by a topochemical alignment of the linking atoms at lower pressures (0-6 GPa)<sup>7</sup>. However, rapid compression to intermediate pressures (30-50 GPa) results in disordered polymers due to large non-equilibrium fluctuations. The topochemical polymer only results at higher pressures (> 67 GPa) where the fluctuations are contained to allow only the most stable polymeric linkages. Reducing the rate of compression, on the other hand, avoids any disordered intermediates and results in the stable polymer above 64 GPa. In either case, short chains form first through a hierarchical process followed by their inter-linking, resembling a nucleation and growth mechanism. We anticipate that other hydrogen-bonded crystals, with multiple polymerizing centres as in acrylamide, might also show such a strong compression rate dependence in transformations under pressure. Hence, compression rate offers a new experimental handle to explore different phases of a crystal.

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# Towards Large-scale Quantum Accuracy Materials Simulations

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## **Abstract**

Electronic structure calculations, especially those using density functional theory (DFT), have been very useful in understanding and predicting a wide range of materials properties. The importance of DFT calculations to engineering and physical sciences is evident from the fact that ~20% of computational resources on public supercomputers are devoted to DFT calculations. Despite the wide adoption of DFT, and the tremendous progress in theory and numerical methods over the decades, the following challenges remain. Firstly, the state-of-the-art implementations of DFT suffer from cell-size and geometry limitations, with the widely used codes in solid state physics being limited to periodic geometries and typical simulation domains containing a few hundred atoms. This limits the complexity of materials systems that can be treated using DFT calculations. Secondly, there are many materials systems (such as strongly-correlated systems) where the widely used model exchange-correlation functionals in DFT, which account for the many-body quantum mechanical interactions between electrons, are inaccurate. Addressing these challenges will enable large-scale quantum-accuracy DFT calculations, and will significantly advance our *ab initio* modelling capabilities to treat complex materials systems.

This talk will discuss our recent advances towards addressing the aforementioned challenges. In particular, the development of computational methods and numerical algorithms for conducting fast and accurate large-scale DFT calculations using adaptive finite-element discretization will be presented, which form the basis for the recently released DFT-FE open-source code.<sup>[1,2]</sup> The computational efficiency, scalability and performance of DFT-FE will be presented, which demonstrates a significant outperformance of widely used plane-wave DFT codes. In addressing the second challenge, our recent breakthrough in accurately solving the inverse DFT problem will be presented,<sup>[3]</sup> which has enabled the computation of exact exchange-correlation potentials for polyatomic systems. Ongoing efforts on using the exact exchange-correlation potentials to improve the exchange-correlation functional description in DFT will be discussed.

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