Deep Learning of Electronic Structure: Challenges and Opportunities

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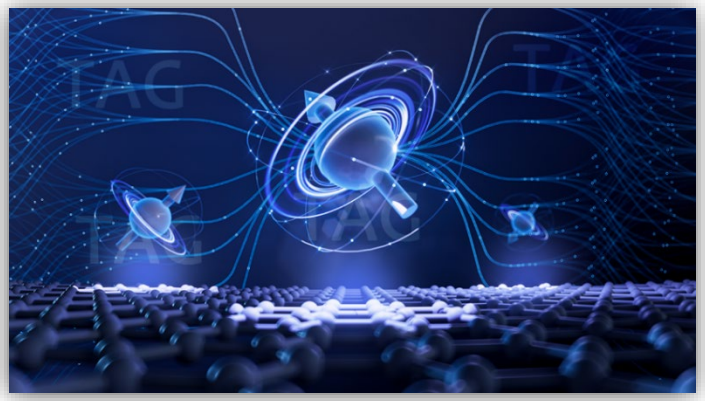
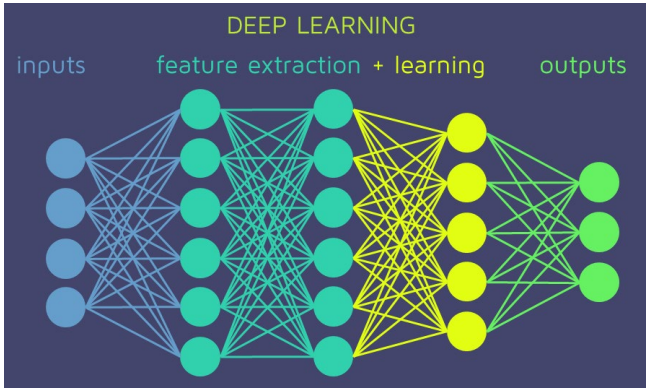


DeepH-pack



Acknowledgment

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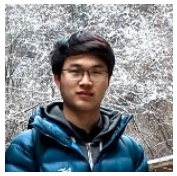

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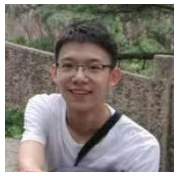

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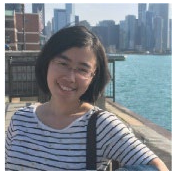

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Minghui Sun

Boheng Zhao

Zhiyuan Zhou

Ting Bao

Zhiming Xu

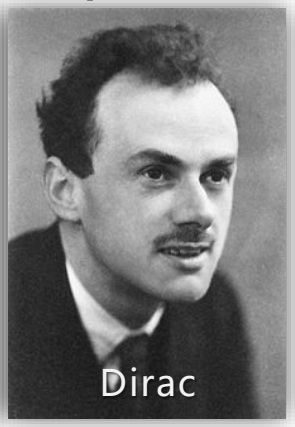
Contents

Quantum Mechanics Revolution

1925 **Matrix Mechanics**

1926 **Schrödinger Equation**

1928 **Dirac Equation**



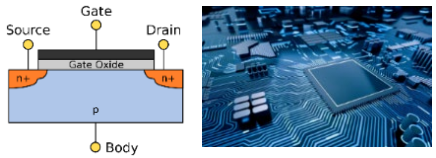
100-Year Development

2025

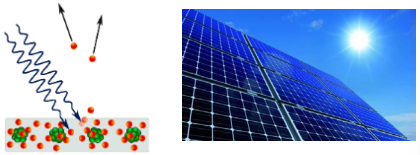
INTERNATIONAL YEAR OF
Quantum Science and Technology

100 years of quantum is just the beginning...

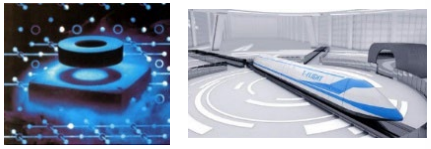
Quantum mechanics has fundamentally changed our understanding of matter and revolutionized modern science and technology!



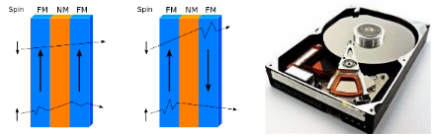
Transistor effect
→ Chip



Photoelectric effect
→ Solar cells



Superconductivity
→ Maglev train



Giant magnetoresistance
→ Hard disk

First-principles calculation

First-principles calculation: Based on the fundamental principles of quantum mechanics, it predicts the properties of matter by solving the Schrödinger equation without relying on empirical parameters.

Schrödinger equation of many-body interacting systems

$$\hat{H}|\Psi\rangle = E|\Psi\rangle$$

$$\hat{H} = -\sum_i \frac{1}{2}\nabla_i^2 + \frac{1}{2}\sum_{i\neq j}\frac{1}{|\mathbf{r}_i - \mathbf{r}_j|} - \sum_I \frac{1}{2M_I}\nabla_I^2 + \frac{1}{2}\sum_{I\neq J}\frac{Z_I Z_J}{|\mathbf{R}_I - \mathbf{R}_J|} - \sum_{i,I}\frac{Z_I}{|\mathbf{r}_i - \mathbf{R}_I|}$$

- One of the most important and challenging problems in science

Density functional theory (DFT)

- Solve the Schrödinger equation for **interacting electrons** of matter

$$\hat{H}|\Psi\rangle = E|\Psi\rangle$$

$$\hat{H} = -\sum_i \frac{1}{2}\nabla_i^2 + \frac{1}{2}\sum_{i\neq j}\frac{1}{|\mathbf{r}_i - \mathbf{r}_j|} - \sum_I \frac{1}{2M_I}\nabla_I^2 + \frac{1}{2}\sum_{I\neq J}\frac{Z_I Z_J}{|\mathbf{R}_I - \mathbf{R}_J|} - \sum_{i,I}\frac{Z_I}{|\mathbf{r}_i - \mathbf{R}_I|}$$

If you don't like the answer, change the question.

- **Kohn-Sham DFT:** map to an auxiliary problem of **non-interacting electrons** with interacting density

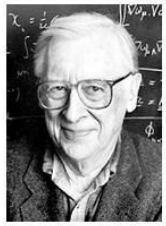
$$\hat{H}_{\mathrm{DFT}}\,|\psi\rangle = \epsilon\,|\psi\rangle$$

$$\hat{H}_{\mathrm{DFT}} = -\frac{\nabla^2}{2} + V_{\mathrm{ext}}(\mathbf{r}) + V_{\mathrm{H}}(\mathbf{r}) + V_{\mathrm{xc}}(\mathbf{r})$$

- Predict materials properties from first principles

The Nobel Prize in Chemistry 1998

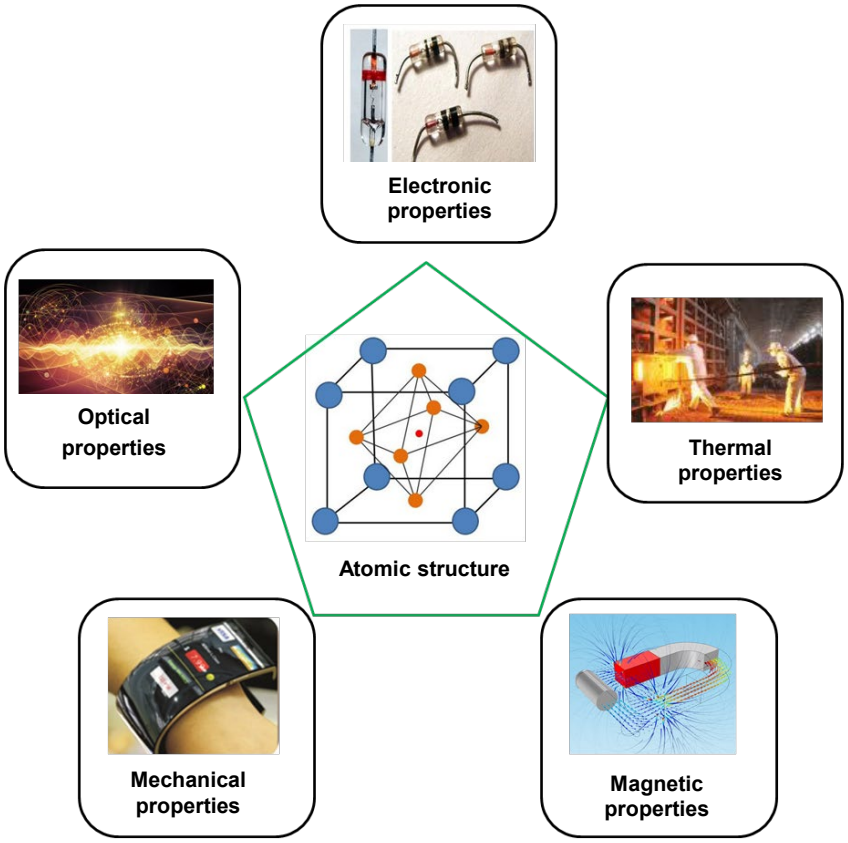
Walter Kohn
Prize share: 1/2



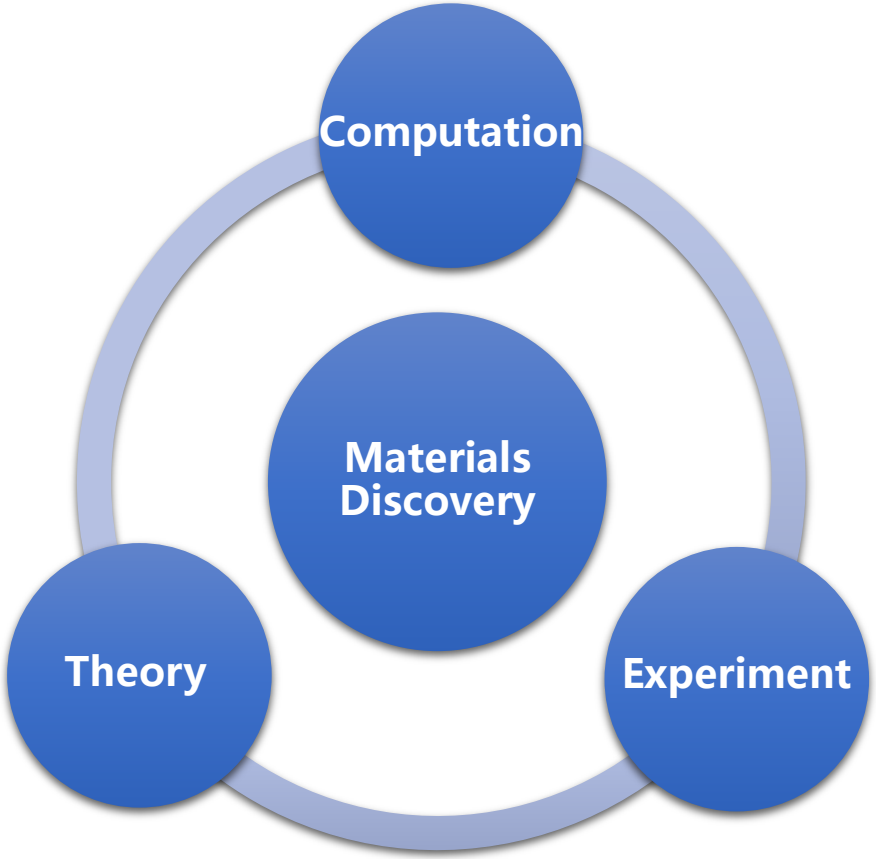
John A. Pople
Prize share: 1/2

From quantum mechanics to materials discovery

Material prediction and design

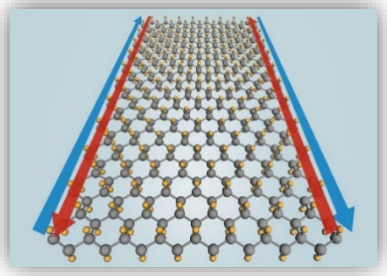


Theory, experiment and computation



From quantum mechanics to materials discovery

Quantum spin Hall insulators



Theory: PRL 111, 136804 (2013) Google citation: **1500**

Nat. Mater. 14, 1020 (2015)

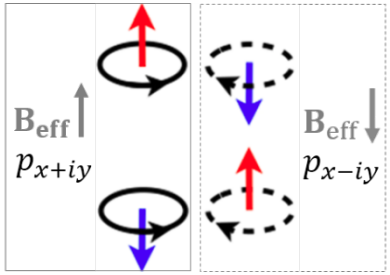
Nat. Mater. 16, 163 (2017)

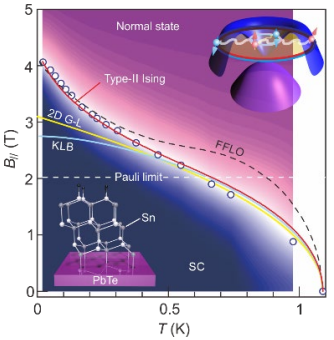
Nat. Mater. 17, 1081(2018)

Nat. Phys. 14, 344 (2018)

PRL 121, 126801 (2018)

New-type Ising superconductivity



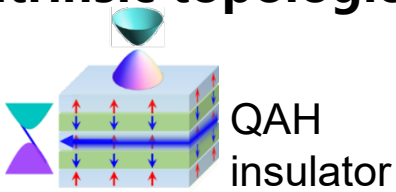


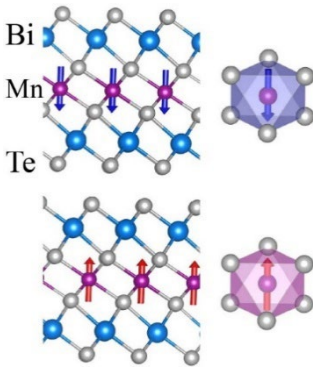
Theory: PRL 123, 126402 (2019)

Science 367, 1454 (2020)

Acc. Mater. Res. 2, 526 (2021)

Intrinsic topological magnetic materials





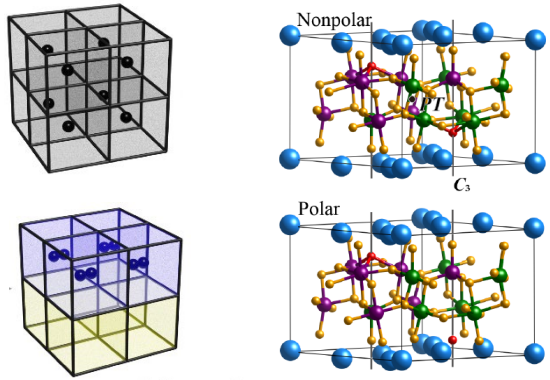
Google citation: **1200**

Theory: Science Advances 5, eaaw5685 (2019)

Chin. Phys. Lett. 36, 076801 (2019)

Nat. Mater. 19, 522 (2020) PRL 125, 086401 (2020)

Negative Hubbard *U*



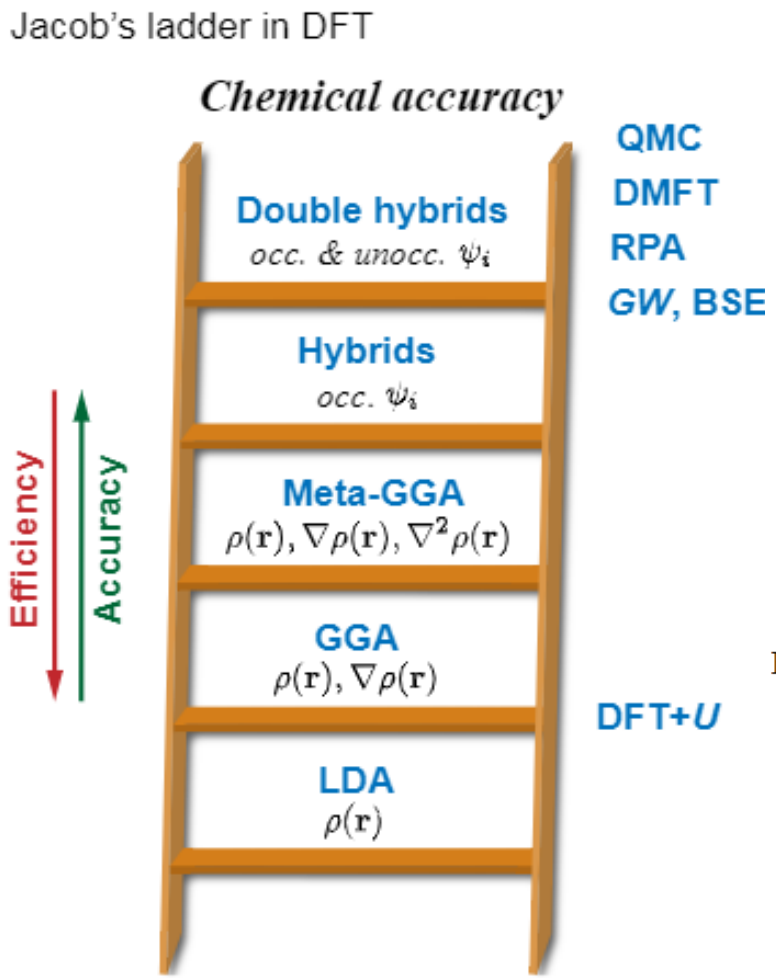
$U_{eff} < 0$ $Lu_7Fe_{14}O_{34}$

Theory: under review at PRL

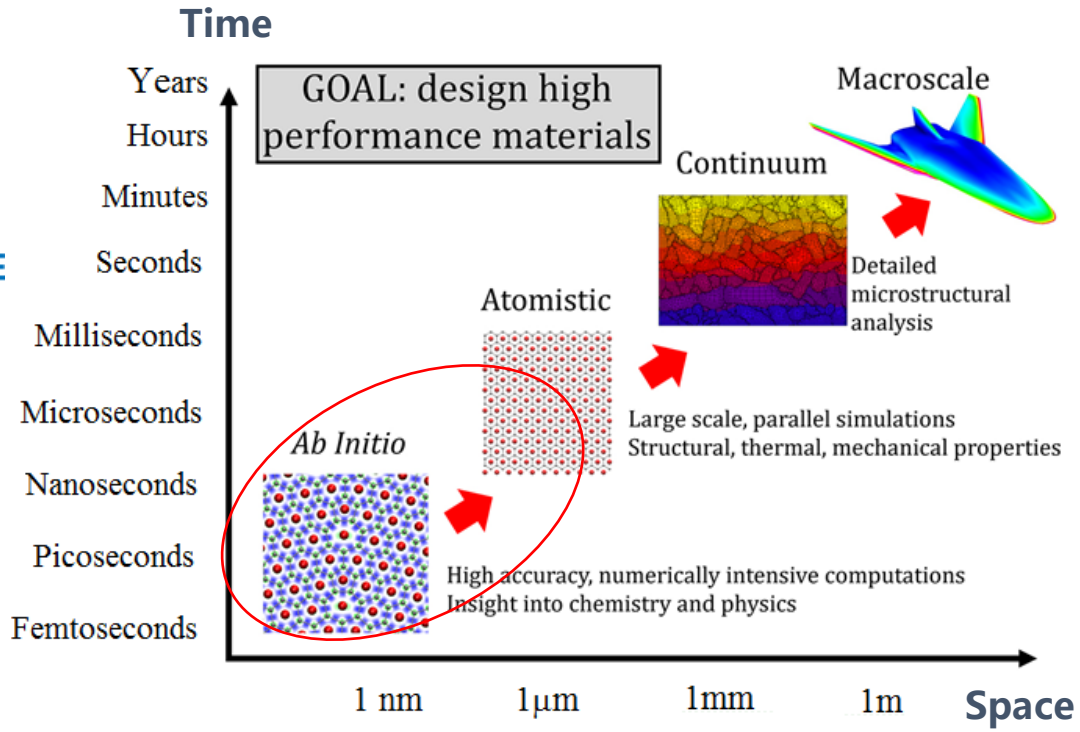
Exp.: Yang Zhang, Zhen Chen, Jing Zhu

Challenges of first-principles calculation

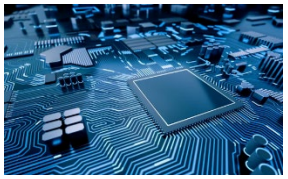
Improve accuracy



Enhance efficiency



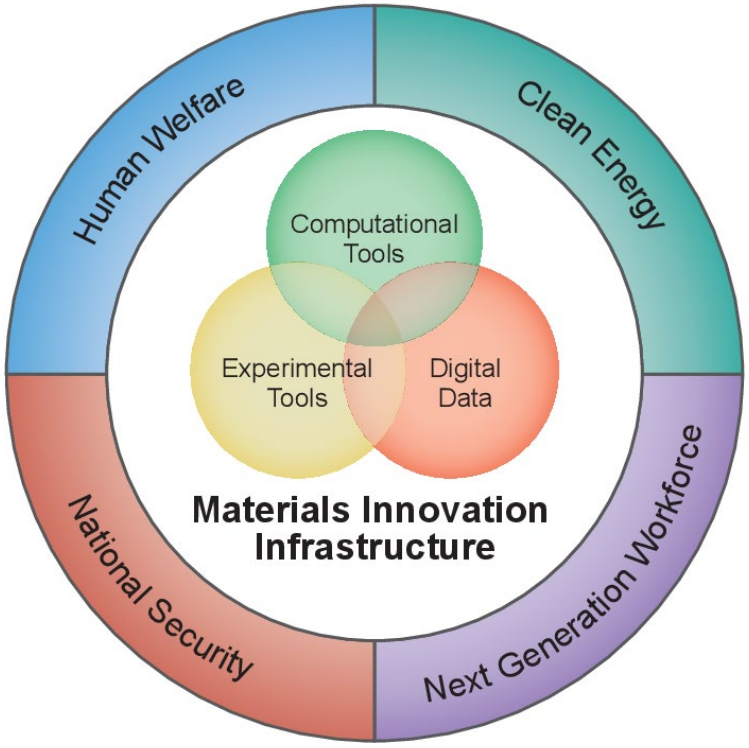
Chip design

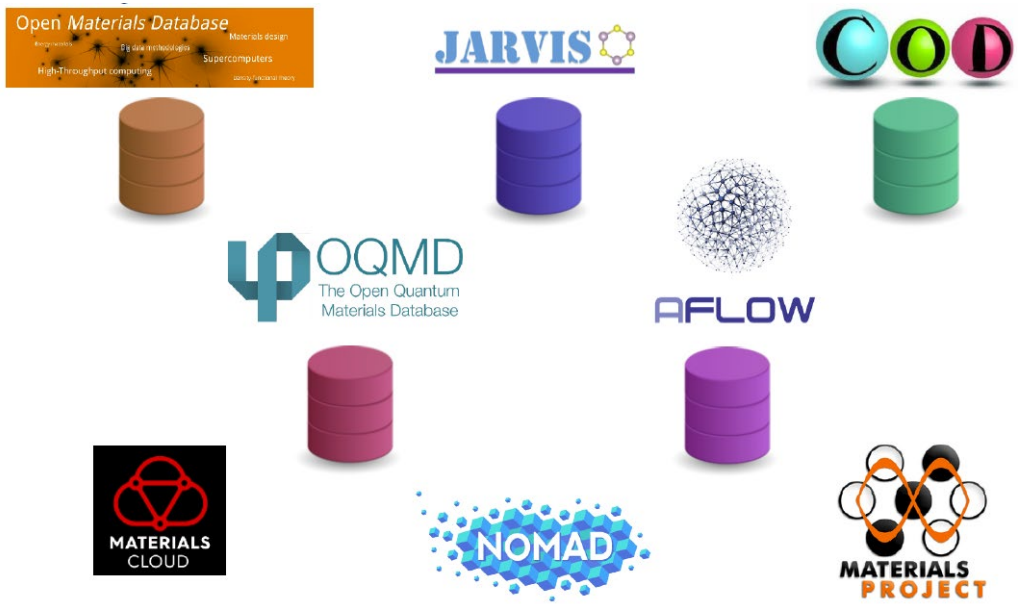


Bottlenecked by the accuracy-efficiency dilemma

Challenges of first-principles calculation

- **High throughput first-principles calculations are applied to build materials databases, but they are too expensive to create bigdata**



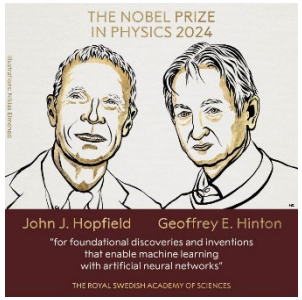


(118 elements in the periodic table)

Current databases: ~10^6 stable solid materials only

How to can we obtain materials big data?

AI-driven materials discovery: Critical challenges



2024 Nobel Prize in Physics:

Materials modeling and discovery are among the most important applications of neural networks.

First-principles calculation → Big materials database → AI-driven materials discovery

Low efficiency of traditional algorithms

Limited amount of materials data

Complex structure–property relationships

First principle + AI

AI + Physics

Contents

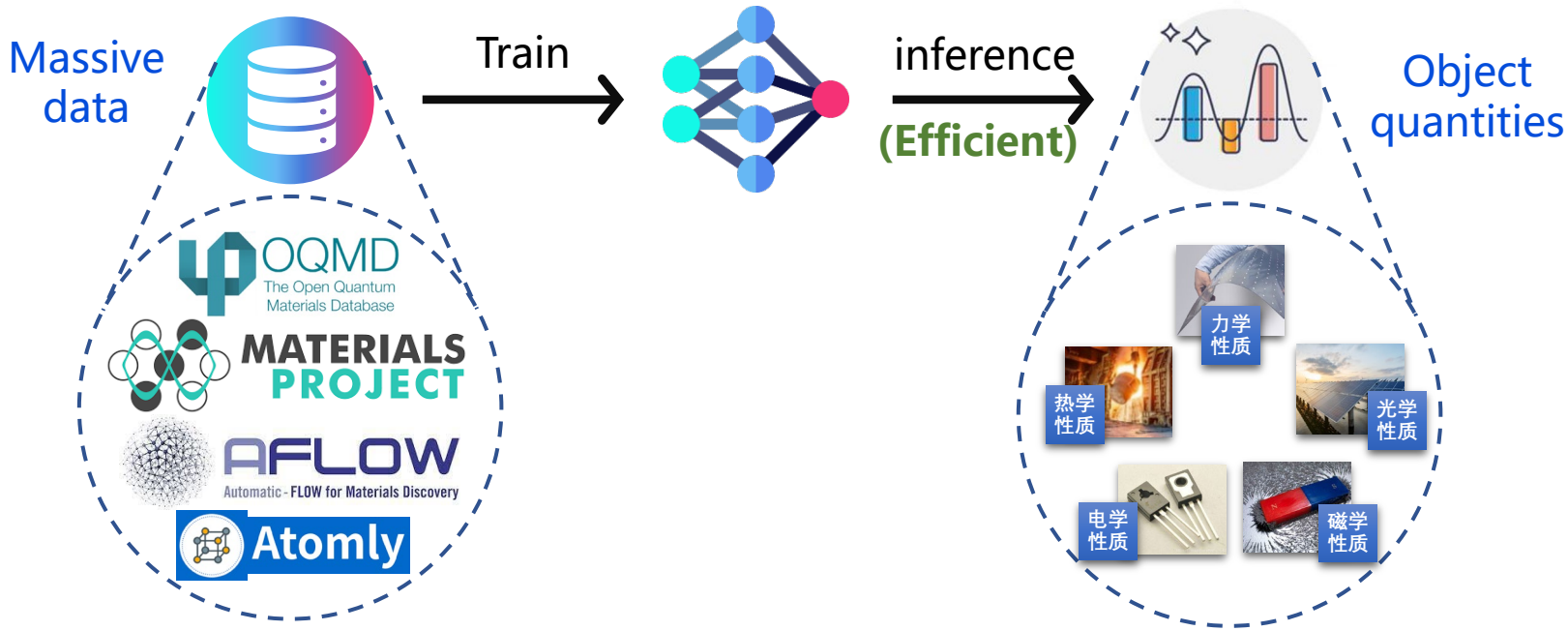
First-principles methods + AI

- **Traditional scientific computation**

(Time-consuming)

Physical principles → Mathematical formula → Numerical Simulation

- **Deep learning accelerated scientific computation**



Develop efficient and intelligent first-principles methods

AI + Physics

Physics: Based on theory and models

Concise, rigorous, and highly interpretable

Use fundamental principles and laws of physics

AI: Relying on data and algorithms

Complex, flexible, and poorly interpretable

Simplify AI models

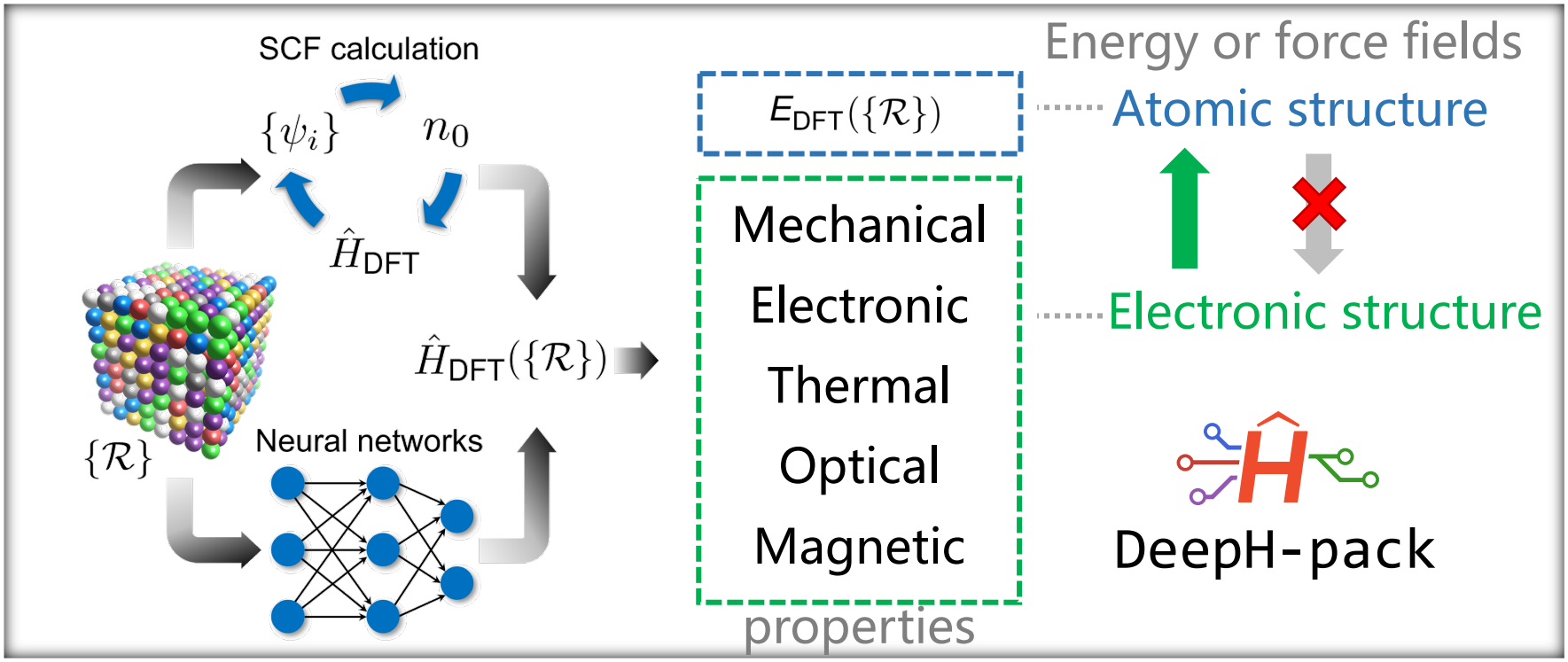
Physics-based AI

Improve accuracy and generalization ability

Key problem: How to properly incorporate prior knowledge into the design of neural networks?

Deep-learning DFT Hamiltonian (DeepH)

➢ DFT Hamiltonian as a function of material structure $\{\mathcal{R}\}$



***"Compress* DFT" into neural networks**

➢ **Integrate important physical priors into deep learning**

Locality: Learn from small structures and generalize to large structures

Symmetry: Further enhance the generalization capability

H. Li, *et al.* **Nat. Comput. Sci.** 2, 367 (2022) arXiv: 2104.03786

Ab initio tight-binding Hamiltonian

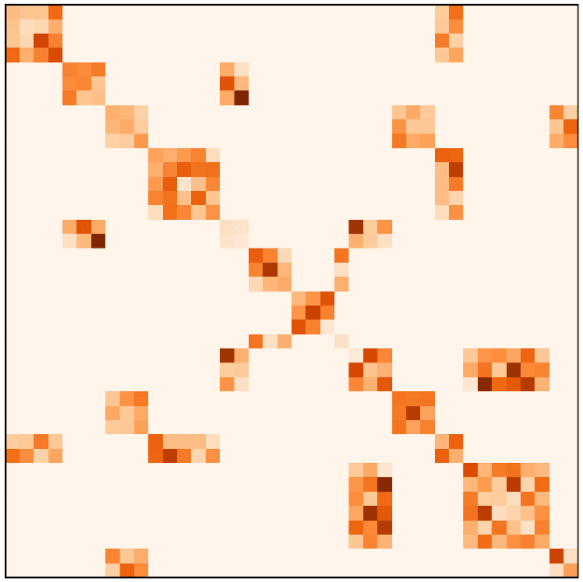
- Localized basis: **sparseness**

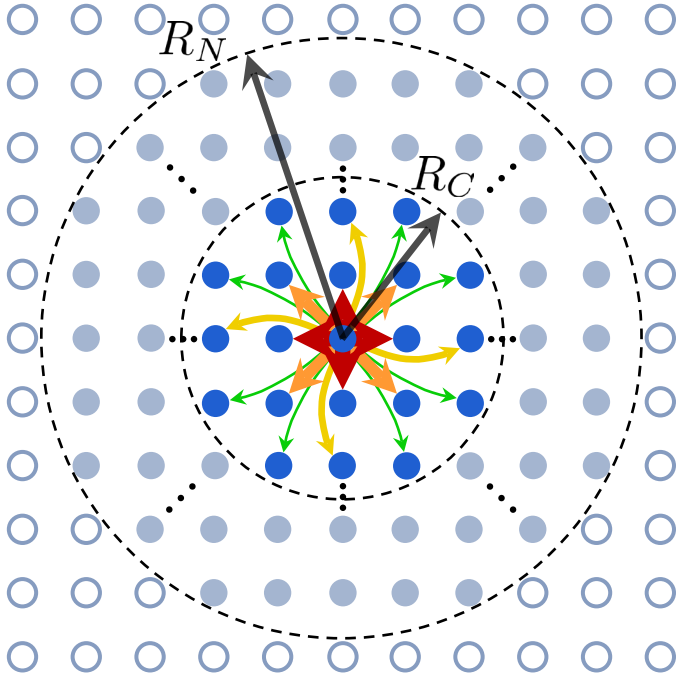
  Only H_{ij} between neighboring atom pairs (within R_C) are nonzero.

- **Nearsightedness (or locality)**

  Only information of neighborhood (within R_N) is relevant.

H_{DFT} for localized basis



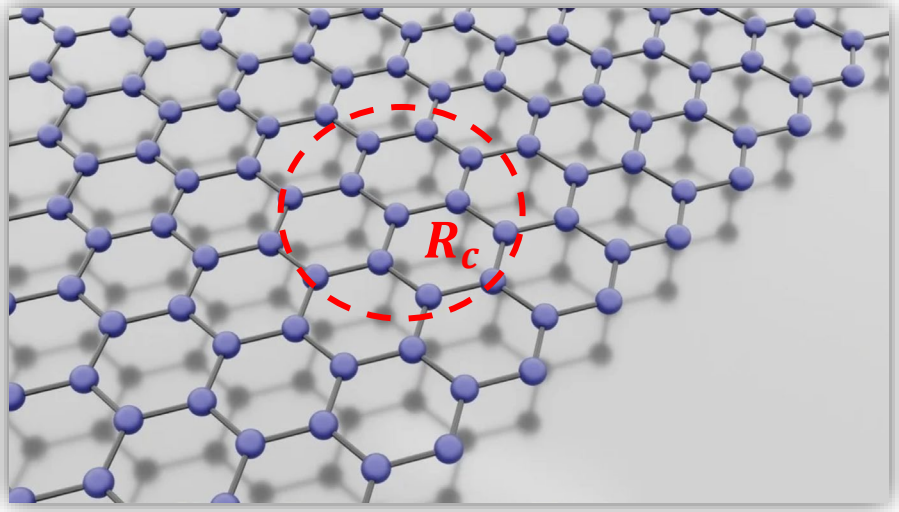


$$H\left(\{\mathcal{R}\}\right) \rightarrow H_{ij}\left(\{\mathcal{R}\}_N\right) \qquad r_{ij} < R_\mathrm{C}$$

DeepH: Enhance the performance by locality

A prior knowledge

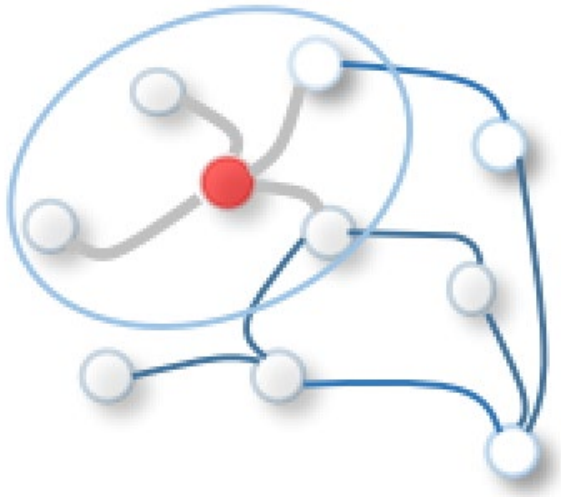
Local electronic properties only affected by neighboring chemical environment



- Short-range interatomic interactions
- **Learn from small structures, generalize to study large structures**
- $O(N)$ computation complexity

Technical insight

Integrate the locality principle into the design of neural networks



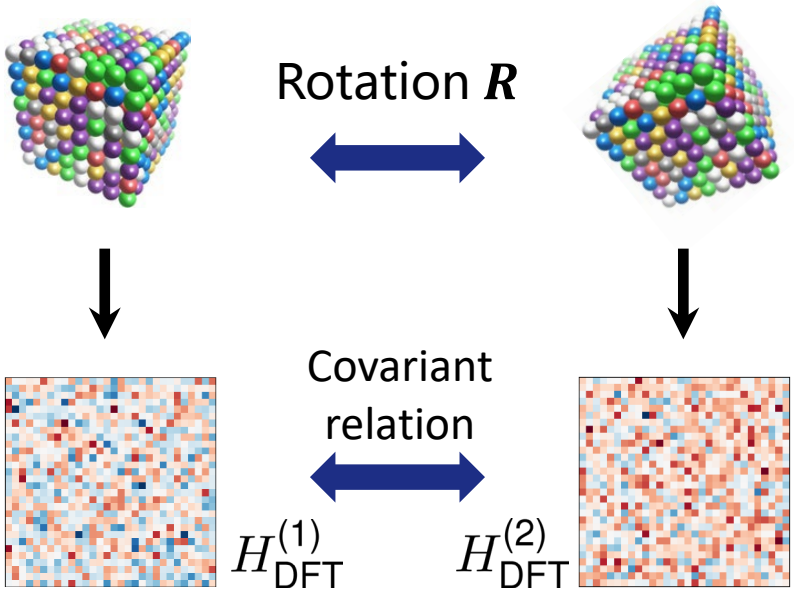
Graph neural networks:

- Crystal structure → crystal graph
- Atom → node; atom pair → edge
- Message passing among neighbors

DeepH: Enhance the performance by symmetry

A prior knowledge

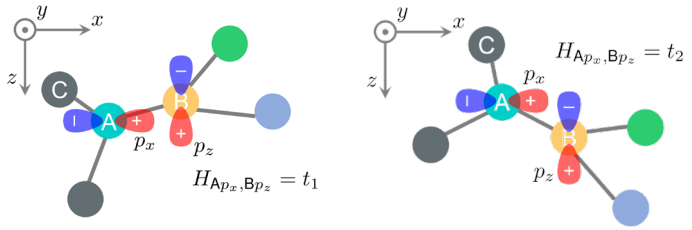
Rotation covariance of H



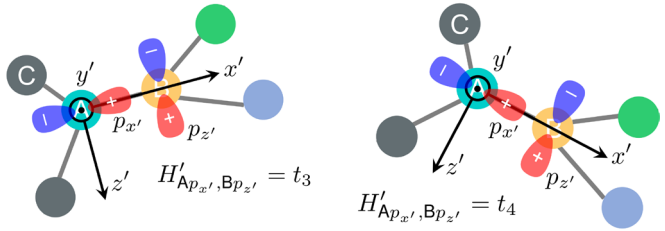
- Covariant transformation of H upon structural rotation
- Challenging for neural-network training and inference

Technical insight

Introduce local coordinates



Global coordinate: **rotation covariance**



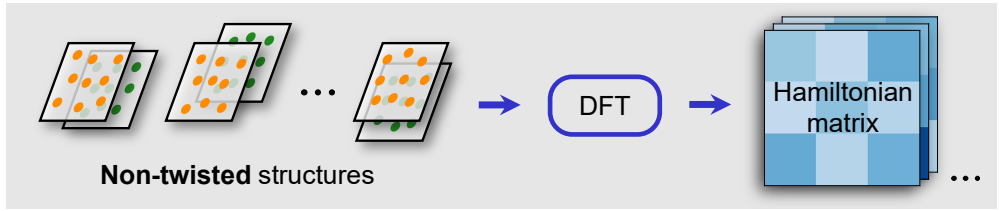
Local coordinate: **rotation invariance**

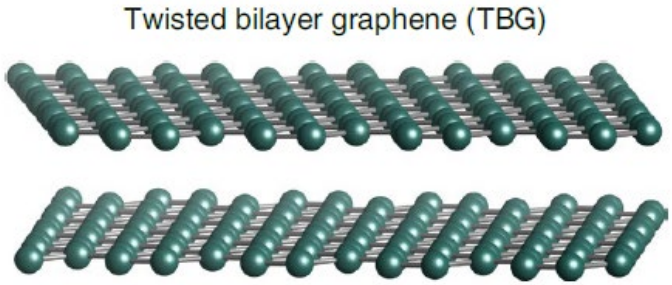
- Change rotation covariance to invariance by local coordinates
- Enhance the performance of DeepH

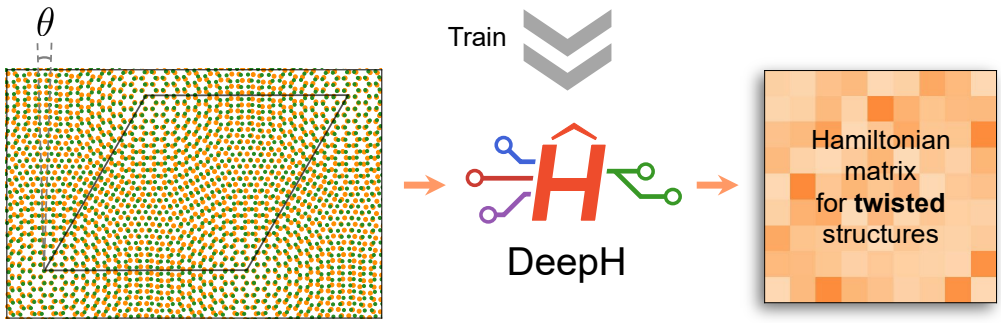
H. Li, *et al.*, **Nat. Comput. Sci.** 2, 367 (2022)

Application: twisted van der Waals materials

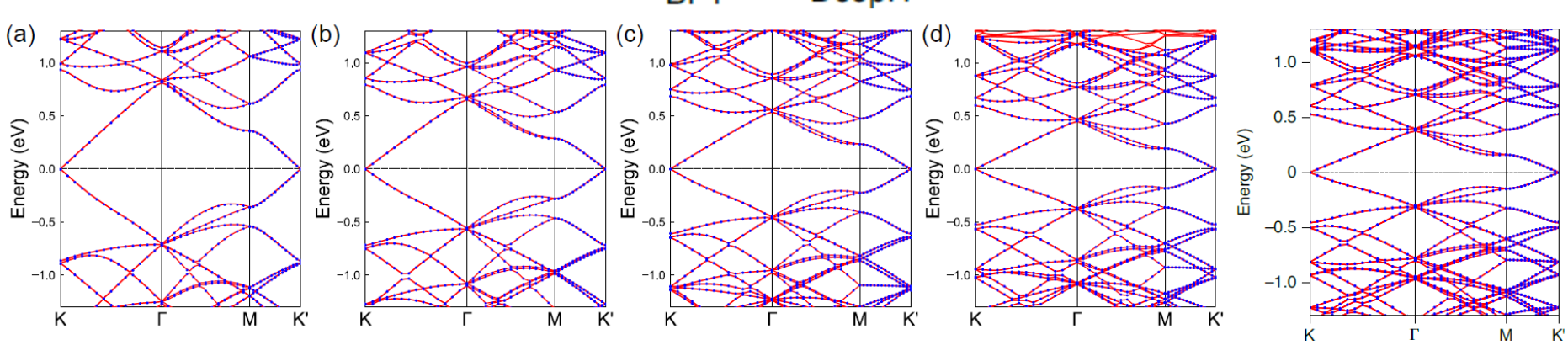
Learn from non-twisted systems → Study materials of arbitrary twist angles





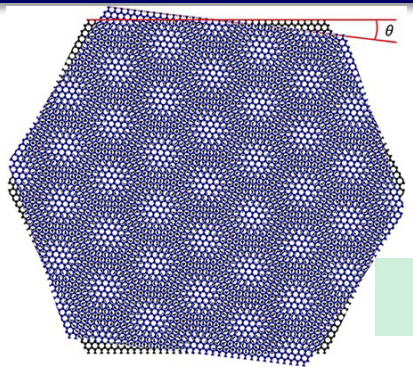


4×4 supercell, 300 random configurations for training



$\theta = 6.01^\circ$	$\theta = 5.09^\circ$	$\theta = 4.41^\circ$	$\theta = 3.89^\circ$	$\theta = 3.48^\circ$
$N_{\text{atom}} = 364$	$N_{\text{atom}} = 508$	$N_{\text{atom}} = 676$	$N_{\text{atom}} = 868$	$N_{\text{atom}} = 1,084$

Magic-angle twisted bilayer graphene



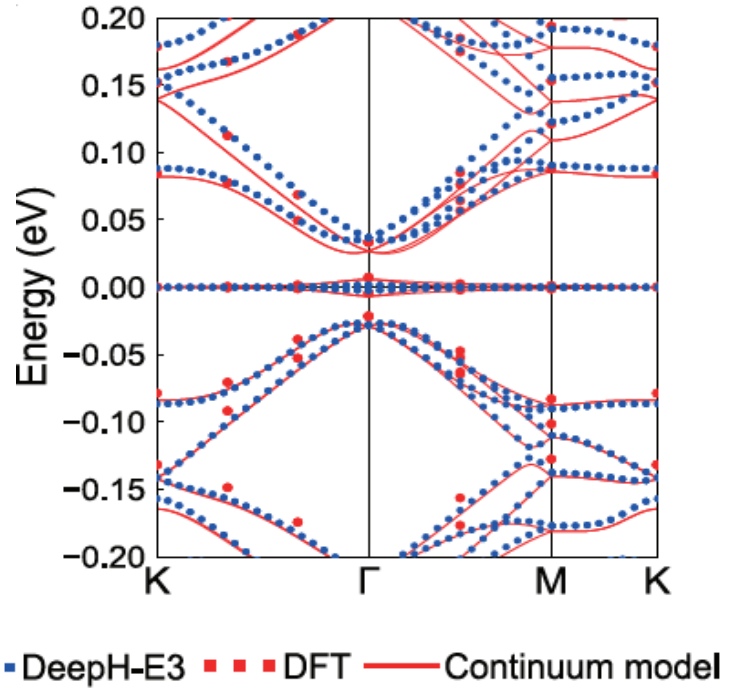
$$\theta = 1.08^\circ$$
$$N_{\text{atom}} = 11,164$$

DFT: VASP (plane-wave, PAW)

DeepH: learned from OpenMX

DFT (10^5~10^6 CPU hours) vs. DeepH (~10^2 CPU hours)

Yong Xu Wenhui Duan

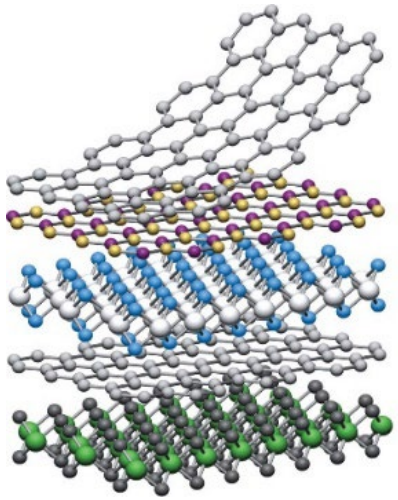


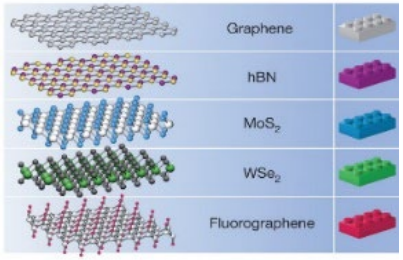
Maintain first-principles accuracy while improve the computational efficiency by several orders of magnitude

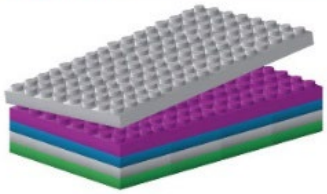
H. Li, *et al*. **Nat. Comput. Sci.** 2, 367 (2022) arXiv: 2104.03786

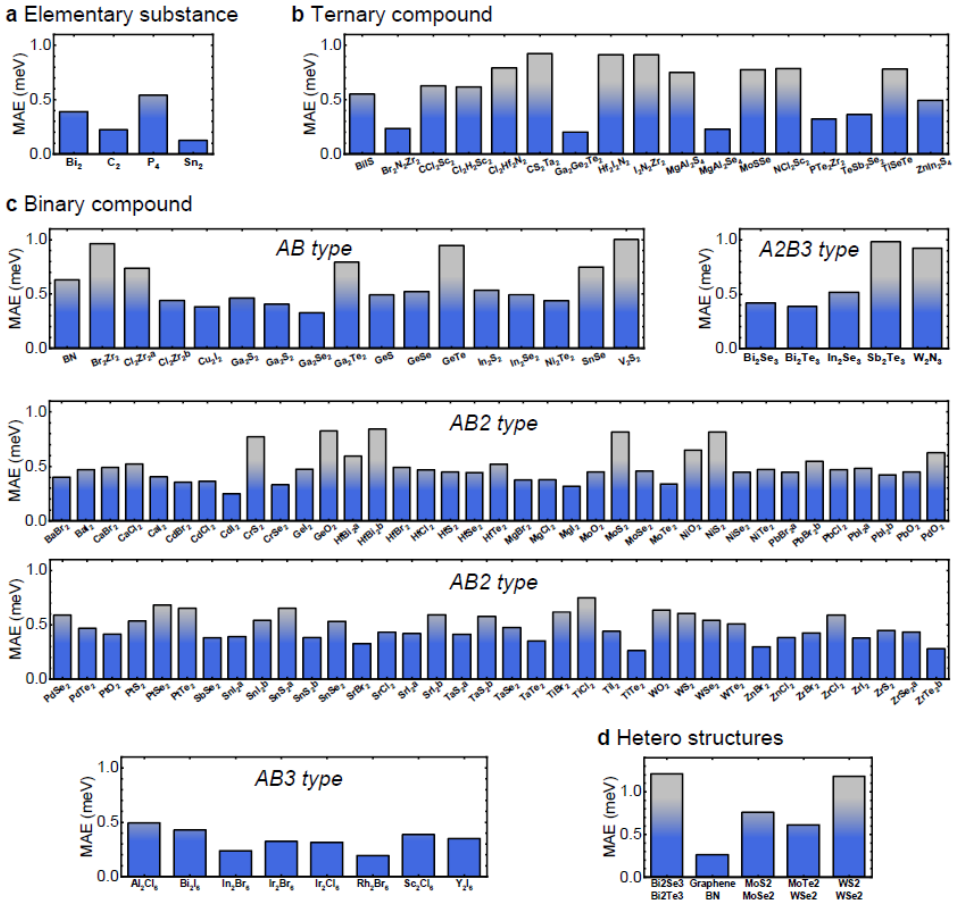
Deep-learning database of twisted materials

DeepH models for twisted bilayer vdW materials:
Over 100 materials, all with sub-meV accuracy,
showcasing the method's robustness









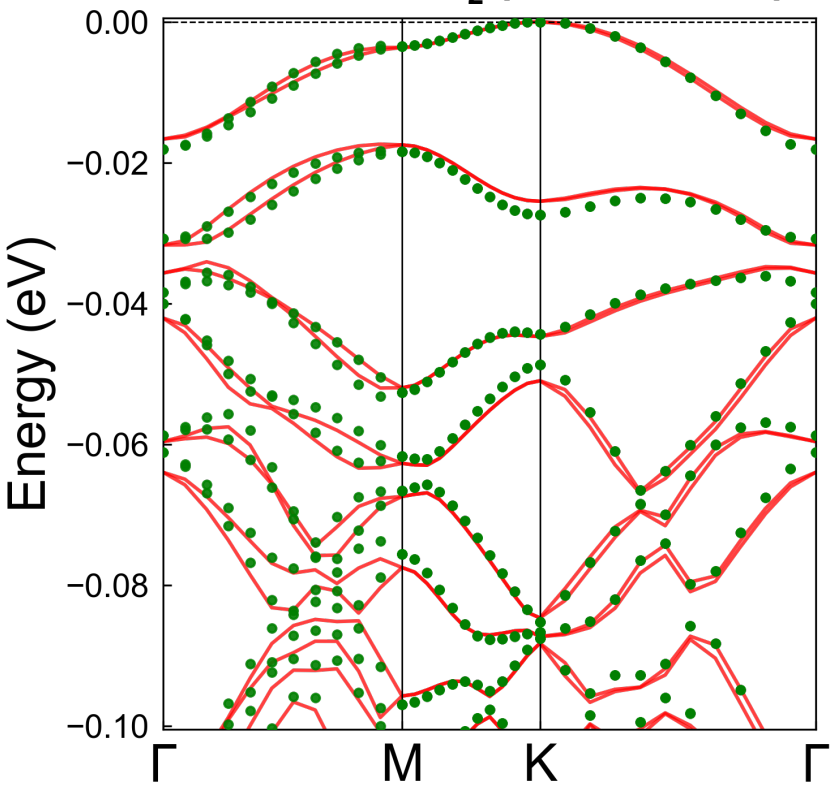
Yong Xu Wenhui Duan

T. Bao, R. Xu, *et al.*, **arXiv**:2404.06449

Application example: Twisted bilayer $MoTe_2$

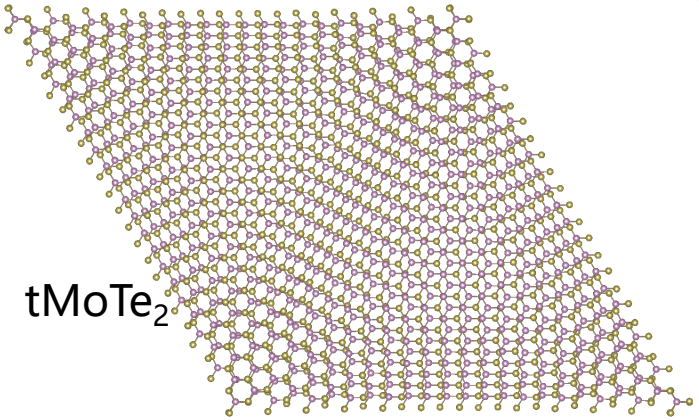
DeepH vs. OpenMX: MAE 0.09 meV

3.89° $tMoTe_2$ (1,302 atoms)

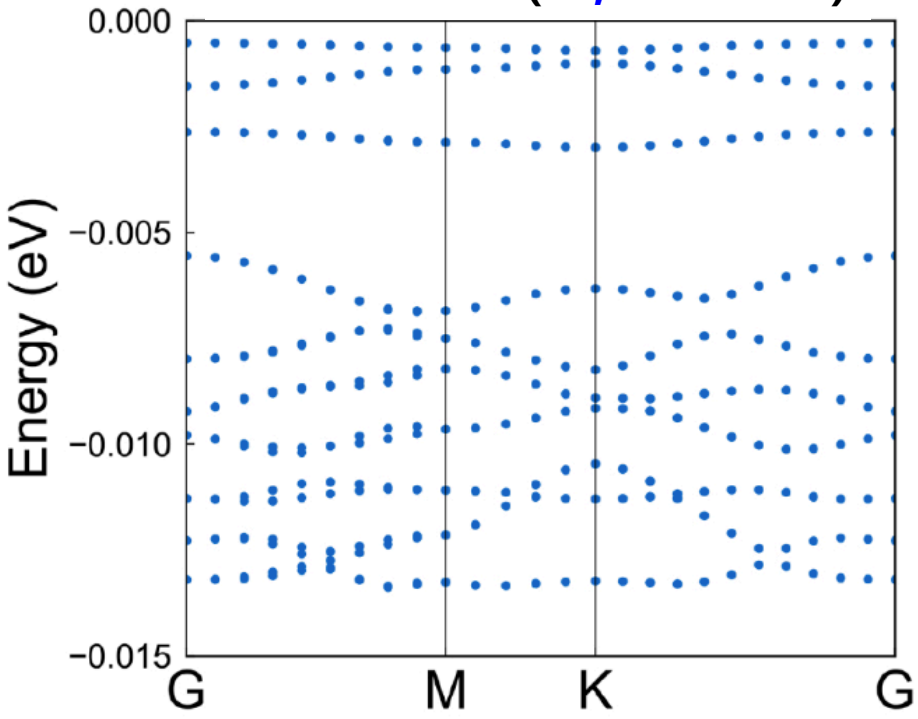


Chern number	1st	2nd	3rd
VASP	1	1	-2
DeepH	1	1	-2

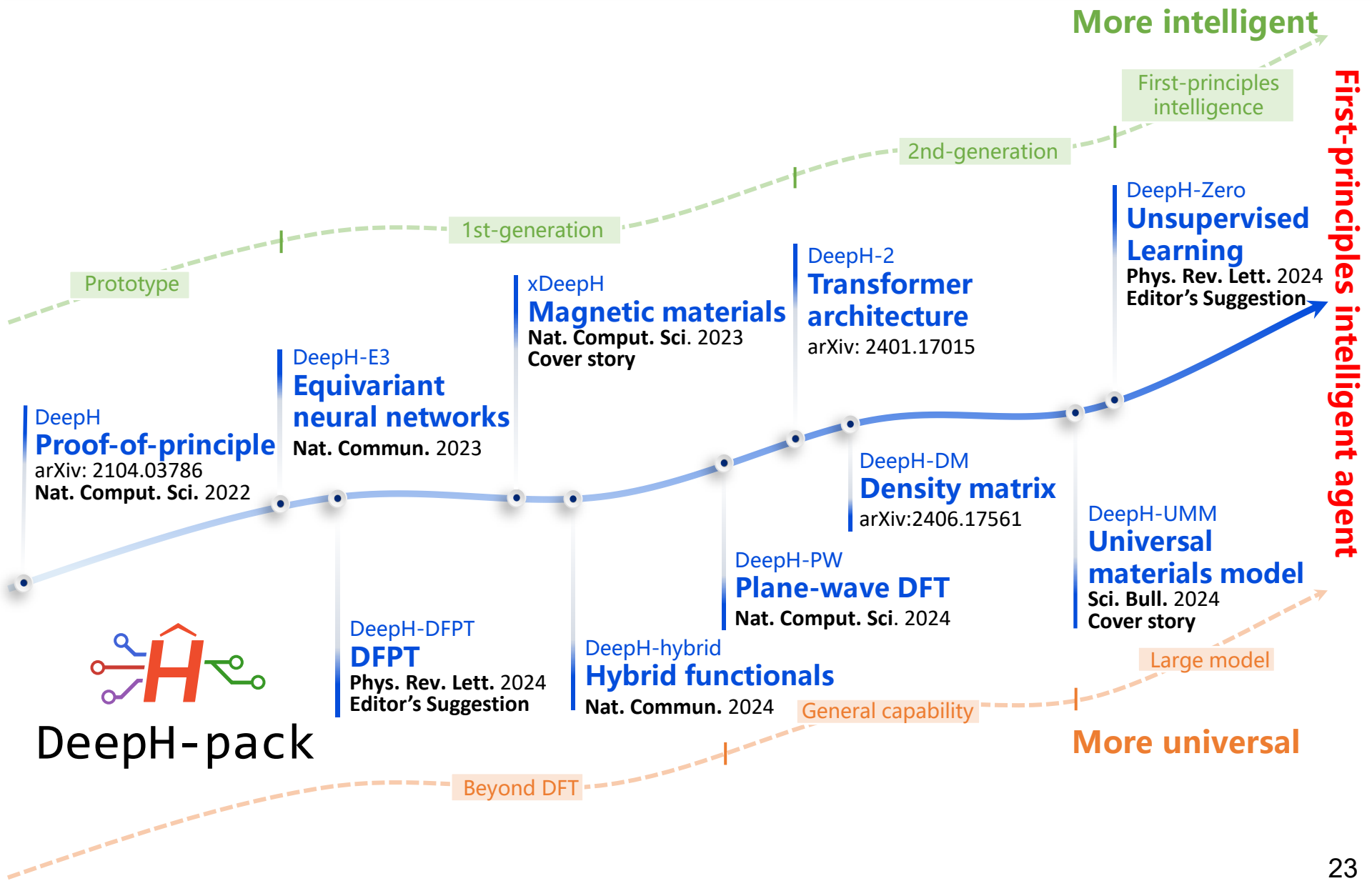
Commun. Phys. 7, 262 (2024)



0.88° tMoTe2 (25,314 atoms)



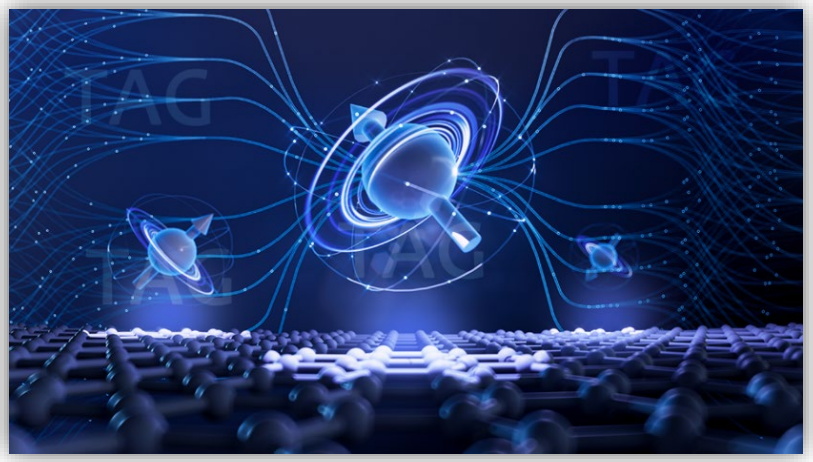
Recent developments of DeepH

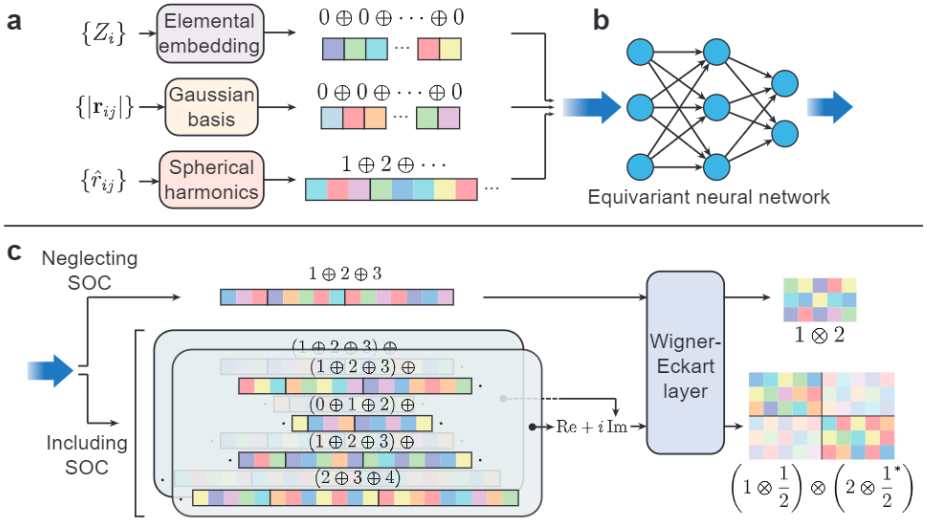


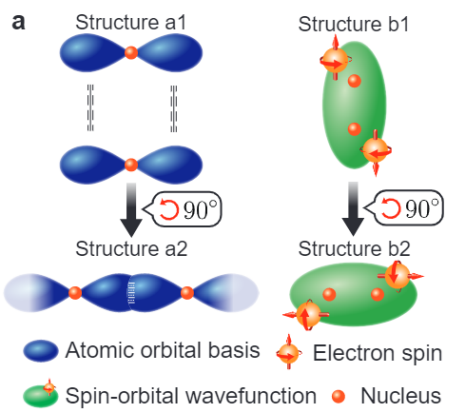
DeepH-E3: Equivariant neural networks

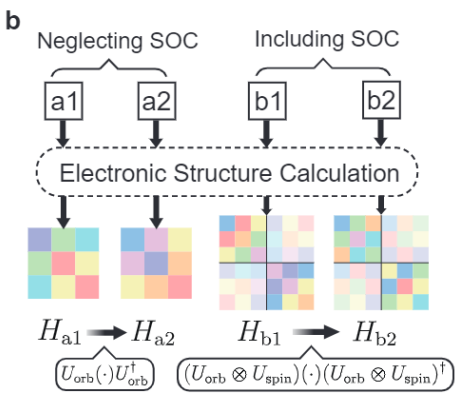
General equivariant framework:

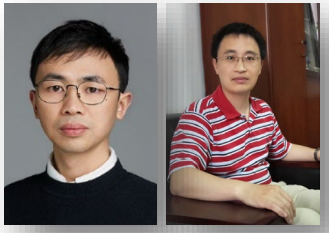
- **Equivariance to the E(3) group (Euclidean group in 3D)**
- **Introduce the degree of spin and spin-orbit coupling**
- **Achieve sub-meV accuracy**





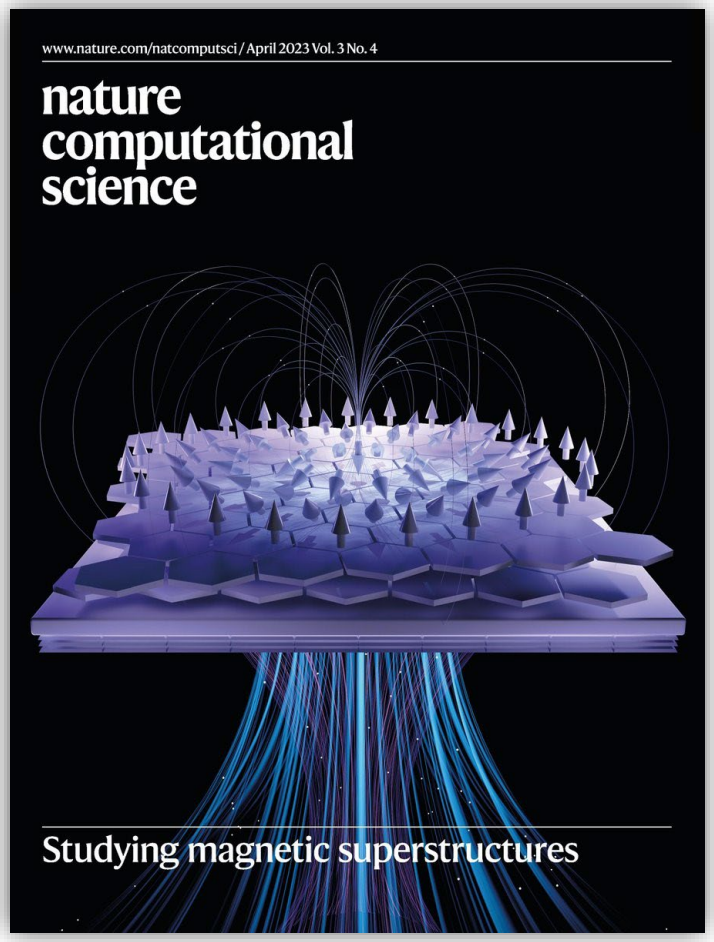


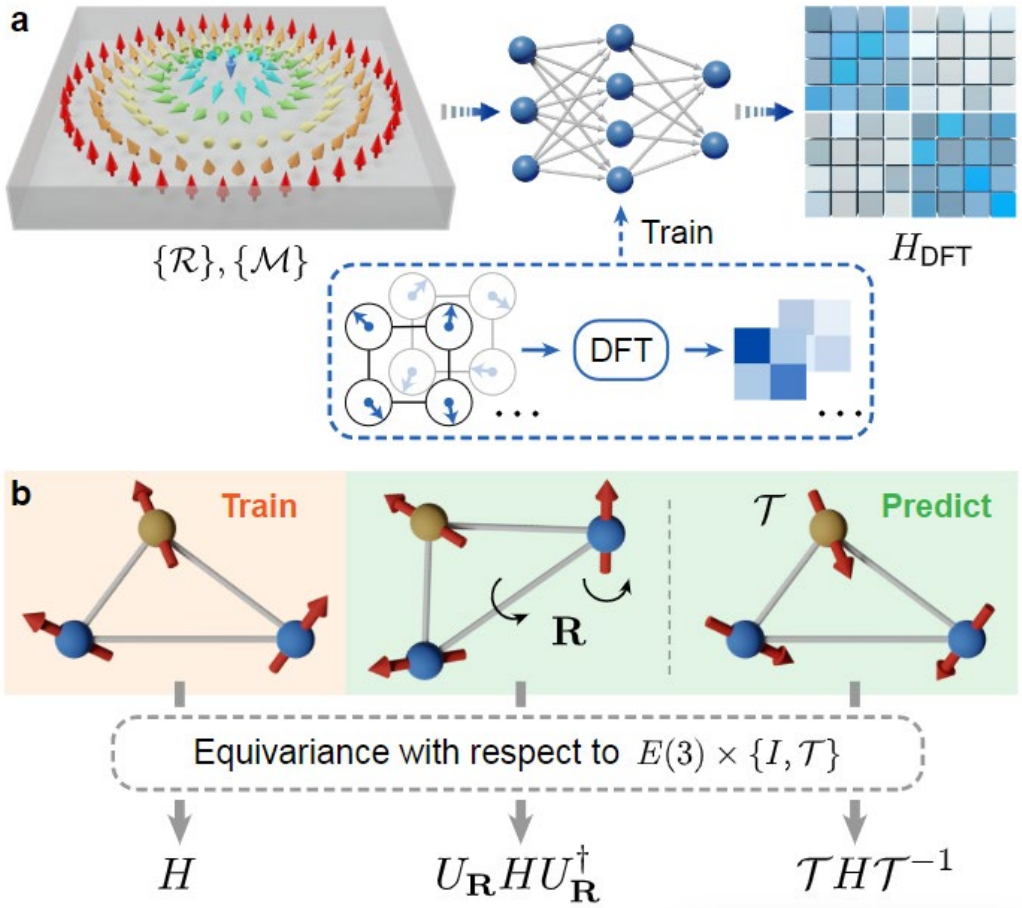




X. Gong, H. Li, *et al.*, **Nat. Commun.** 14, 2848 (2023)

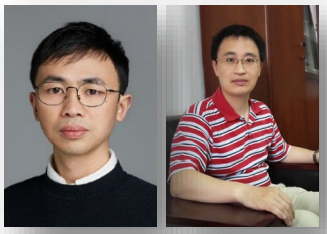
xDeepH: For studying magnetic materials



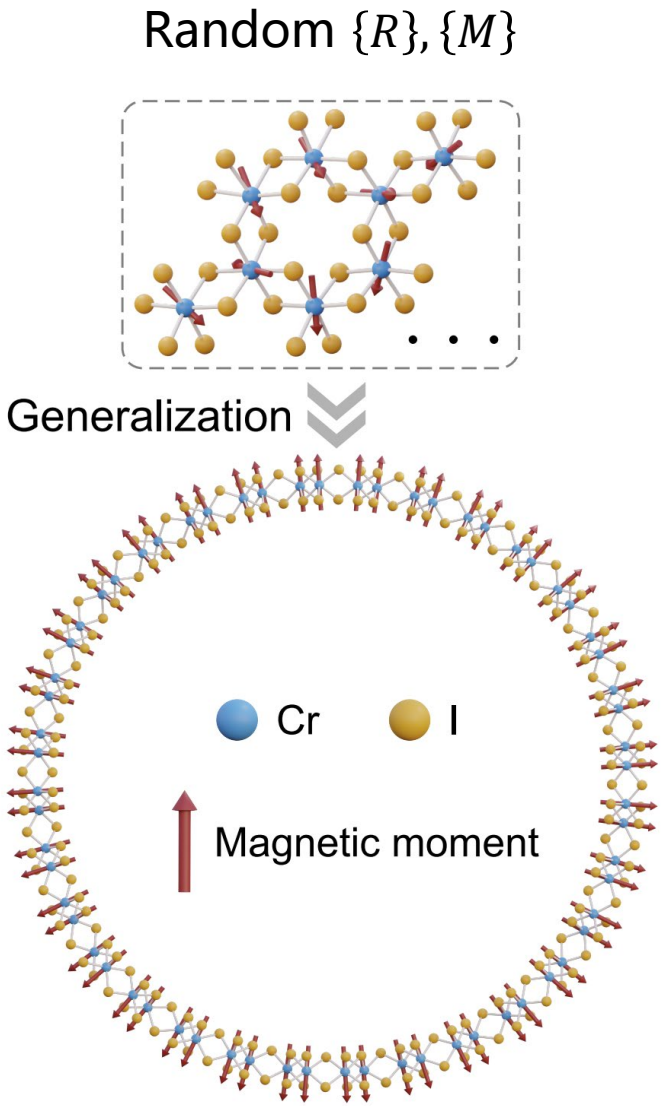


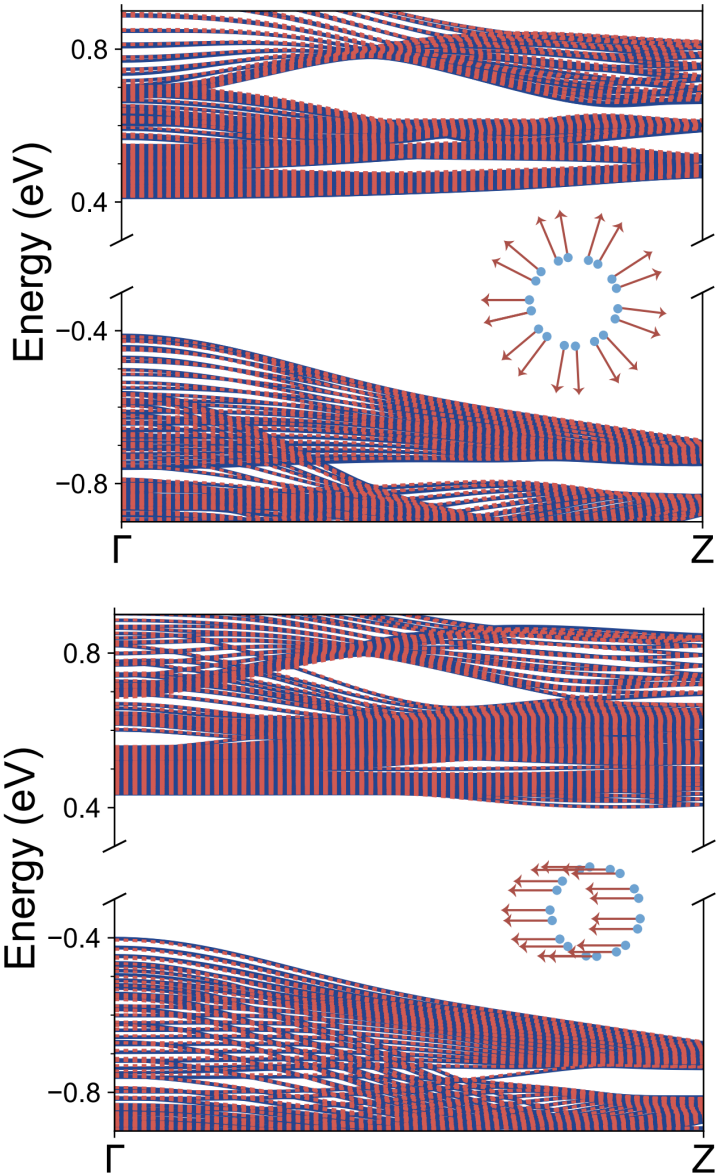
H. Li, Z. Tang, et al., **Nat. Comput. Sci.** 3, 321 (2023) **Cover story**

Research Briefing: https://www.nature.com/articles/s43588-023-00425-2
Editorial: https://www.nature.com/articles/s43588-023-00451-0
News & Views: https://www.nature.com/articles/s43588-023-00434-1



Curved magnetism in CrI_3 nanotube





DFT

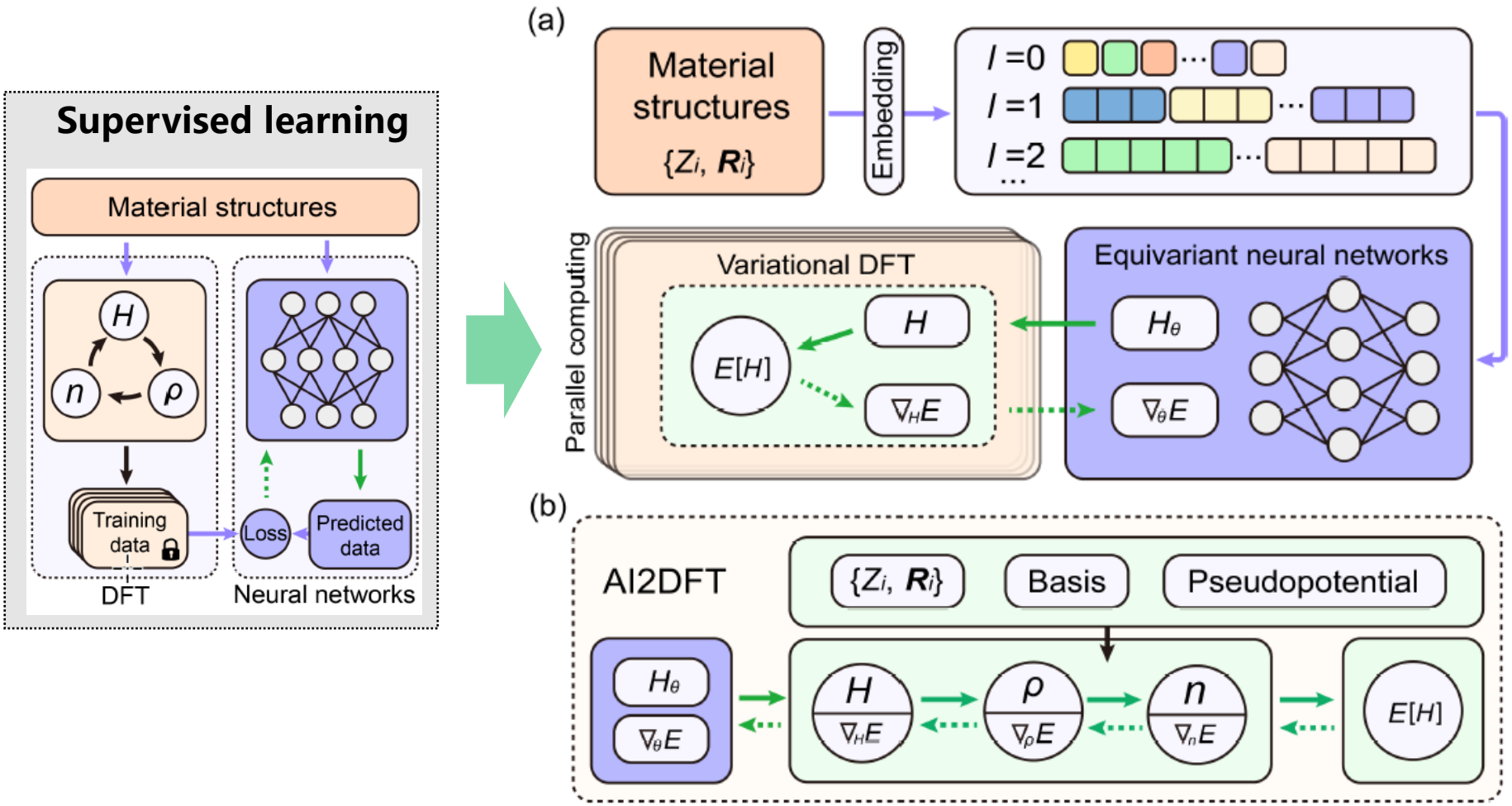
xDeepH

H. Li, Z. Tang, et al., **Nat. Comput. Sci.** 3, 321 (2023) **Cover story**

Neural-network density functional theory

Physics-informed unsupervised learning (DeepH-Zero):

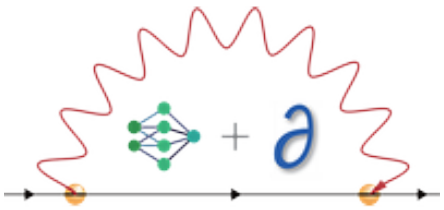
Coherently integrate DFT algorithms into neural networks



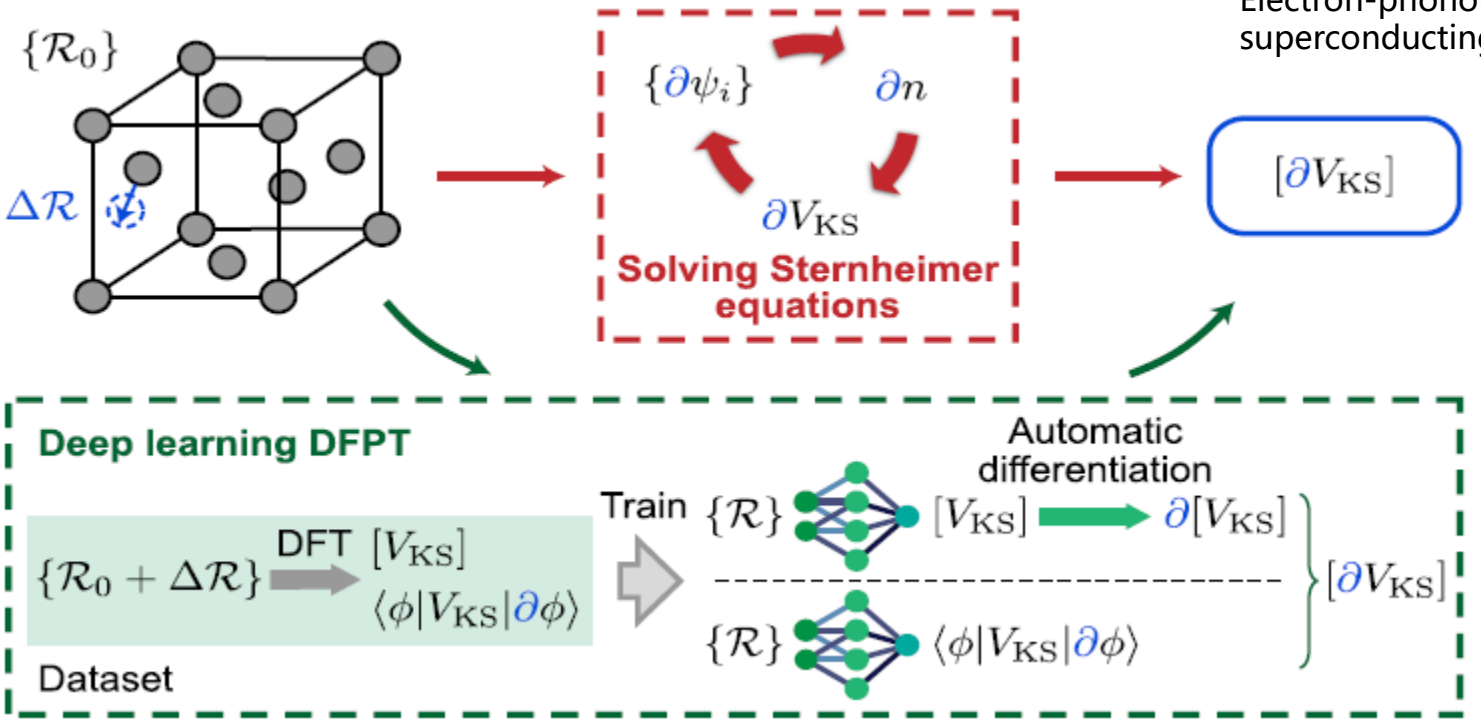
Y. Li, Z. Tang, Z. Chen, *et al.*, **Phys. Rev. Lett.** 133, 076401 (2024) **Editors' suggestion**

Density functional perturbation theory (DFPT)

DeepH-DFPT: Bring DFT and DFPT into a unified deep-learning framework → Efficient calculation of e-p coupling, BCS superconductivity, etc.



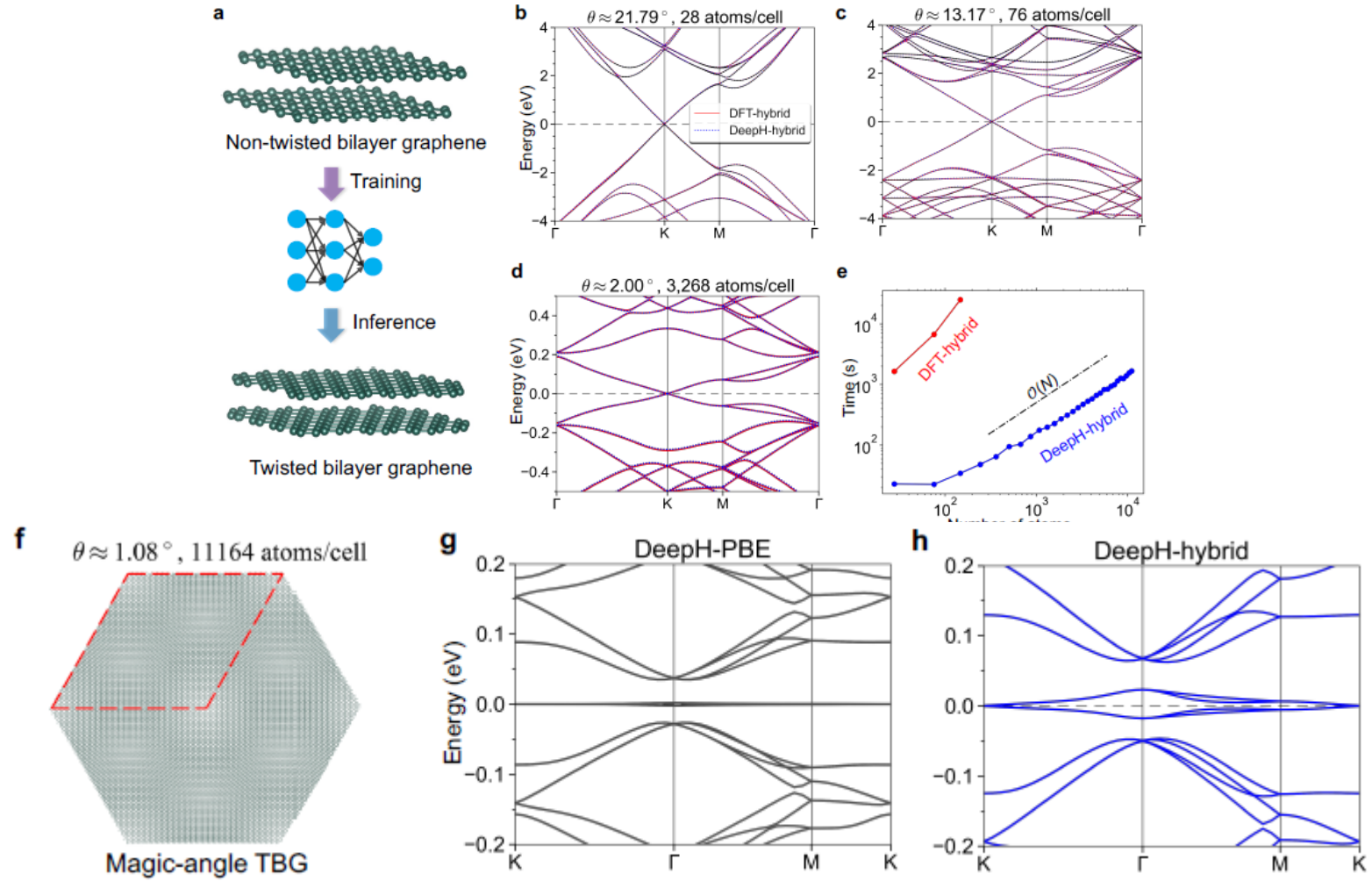
Electron-phonon coupling, superconducting properties



H. Li, Z. Tang, *et al.,* **Phys. Rev. Lett.** 132, 096401 (2024) **Editors' suggestion**

DeepH-hybrid: For hybrid-functional calculations

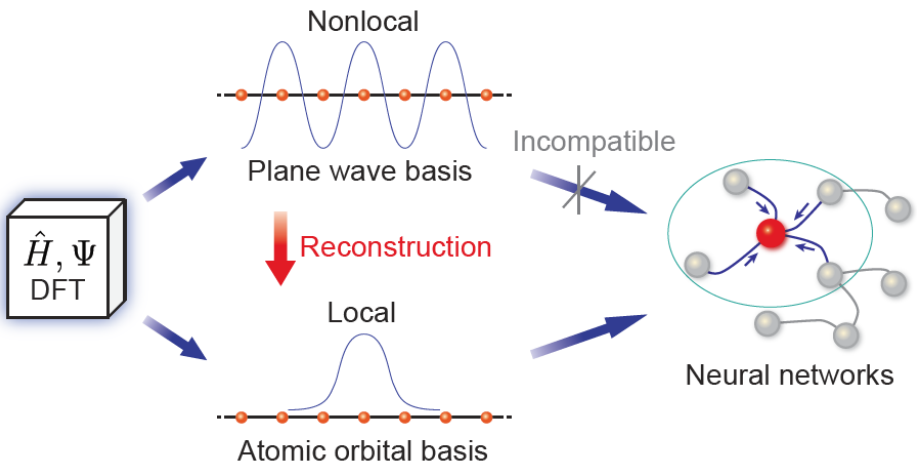
Generalize from Kohn-Sham DFT to generalized Kohn Sham schemes



Z. Tang, H Li, P, Lin, et al., **Nat. Commun.** 15, 8815 (2024)

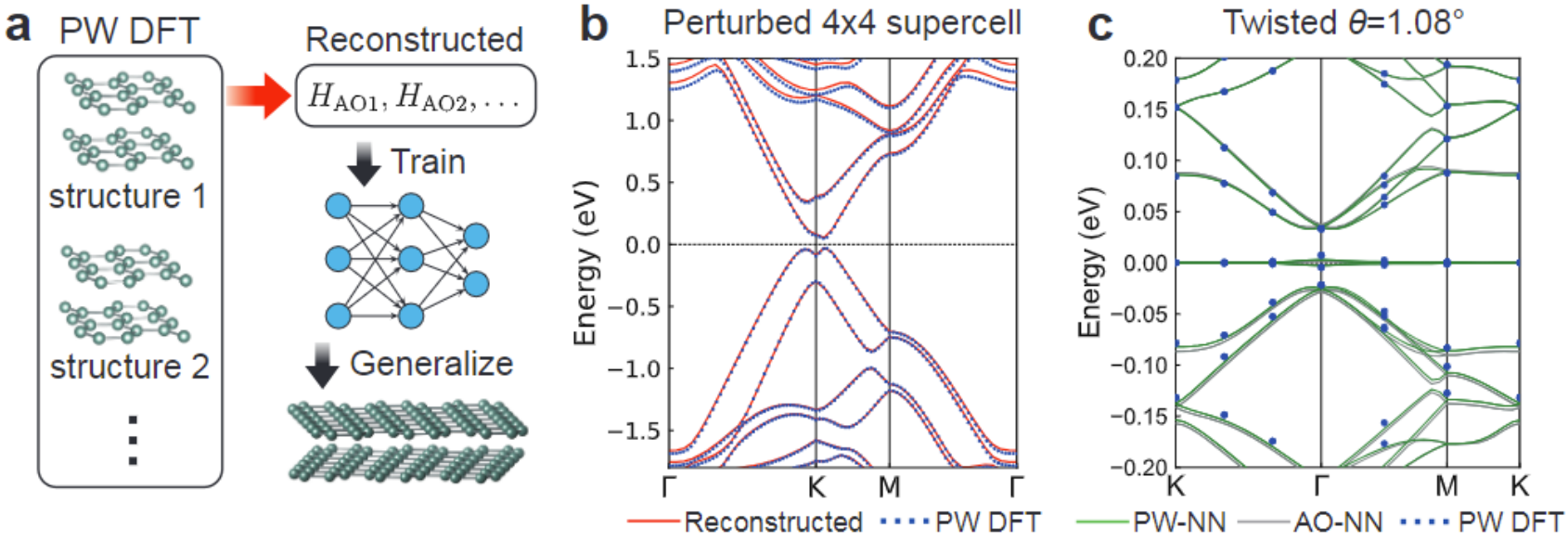
Collaborators: Dr. Peize Lin, Prof. Xinguo Ren, Prof. Hong Jiang , Prof. Lixin He

DeepH-PW: Generalize to plane-wave DFT



DeepH: Access to all DFT codes

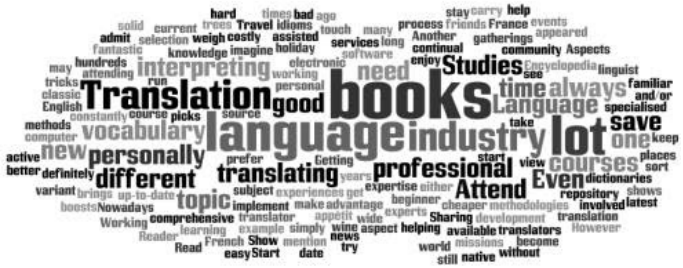
- **Big materials data of DFT**
- **Large materials model**



X. Gong, *et al.*, **Nat. Comput. Sci.** 4, 752 (2024) Collaborate with Steven G. Louie

From large language model to large materials model

Language agent



Language database

Large language model

Q: What is large language model?

A: Large language model is a type of AI designed to understand and generate human-like language.

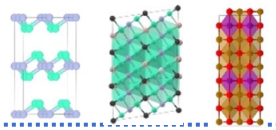
Materials agent

Materials database

Large materials model

Q: High-Tc superconductors?

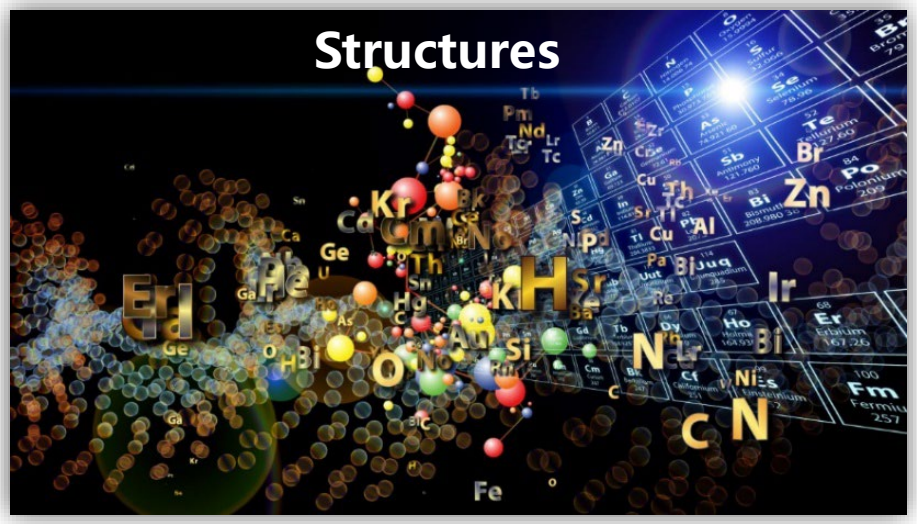
A: Here are some predicted materials:

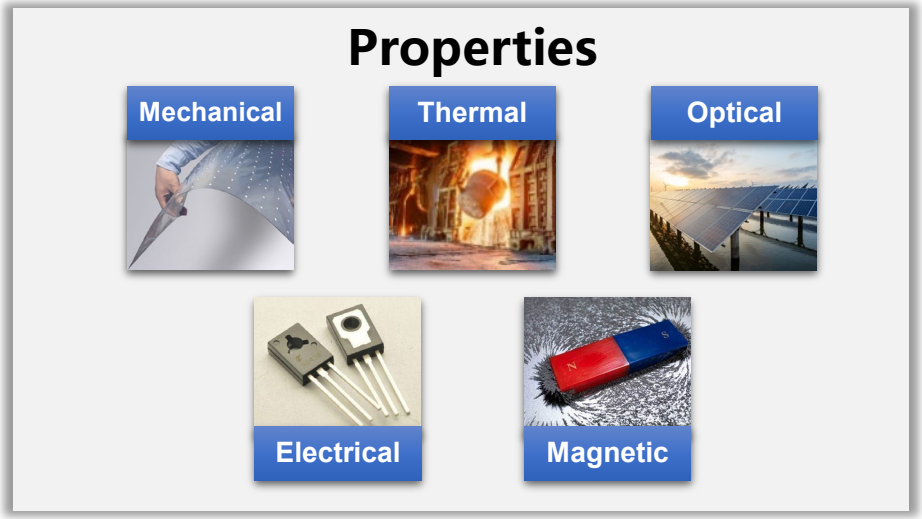


Large Materials Model

Describe the universal structure-property relationship

Large materials model





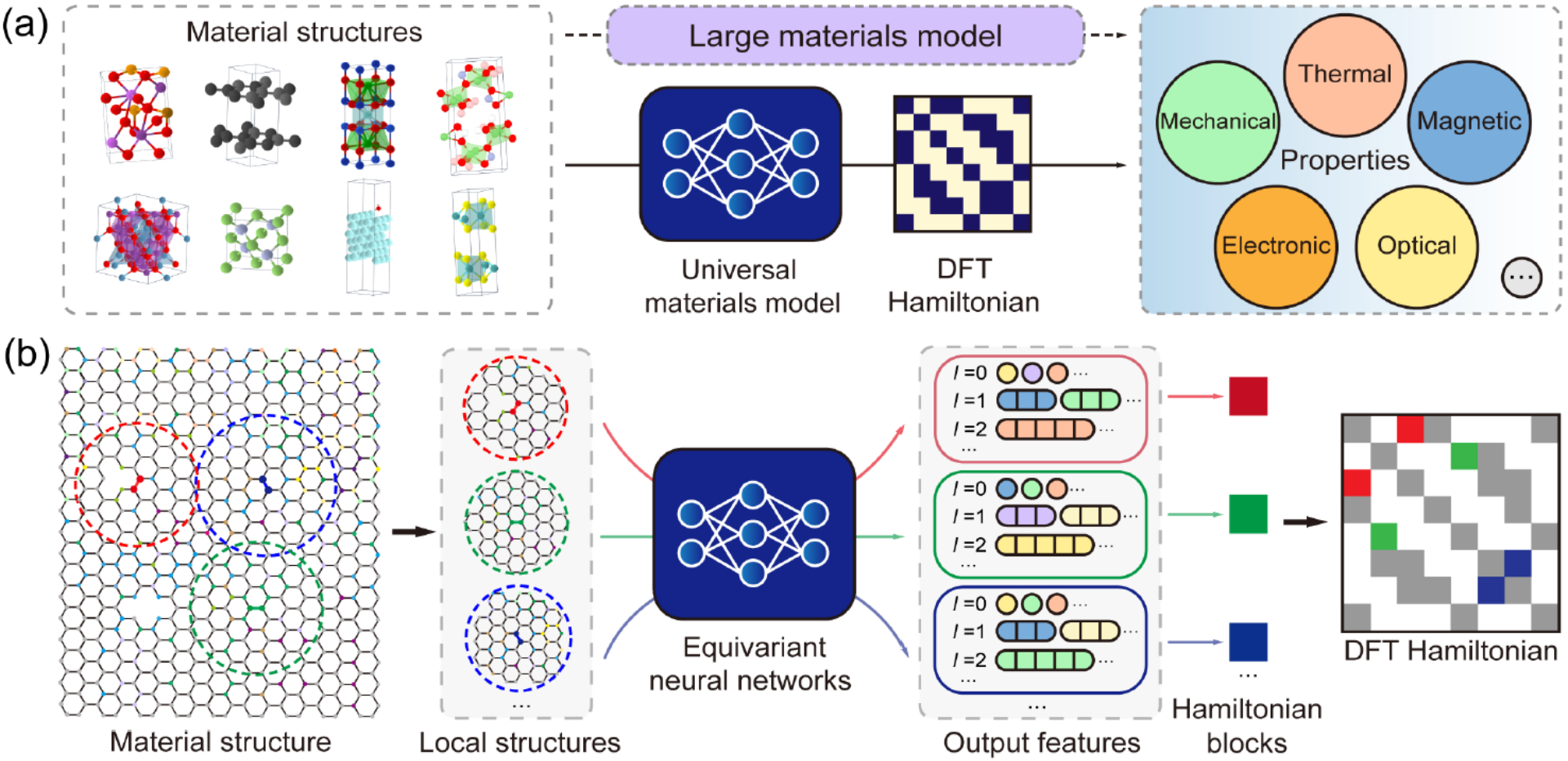
Varying atomic compositions/structures
Countless potential candidates

Rich physical properties
Complicated dependence on structure

AI-driven material discovery ← Big materials data

Universal materials model of DeepH

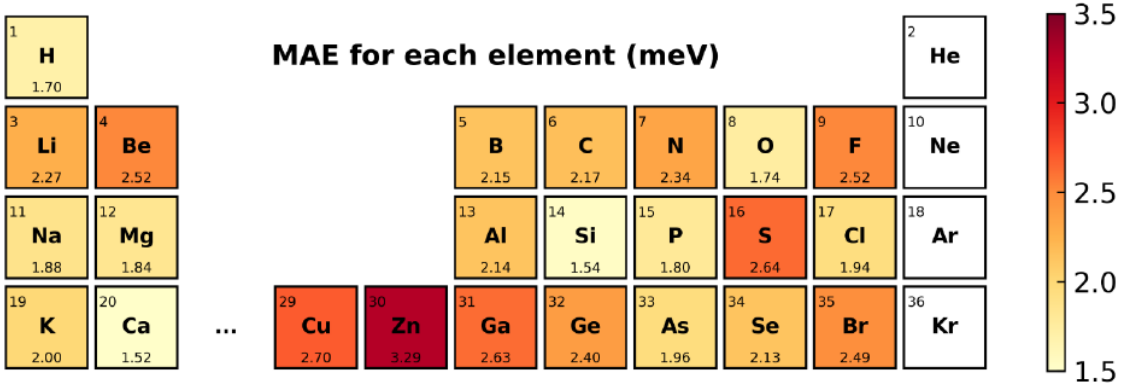
Large materials model: AI-driven materials discovery



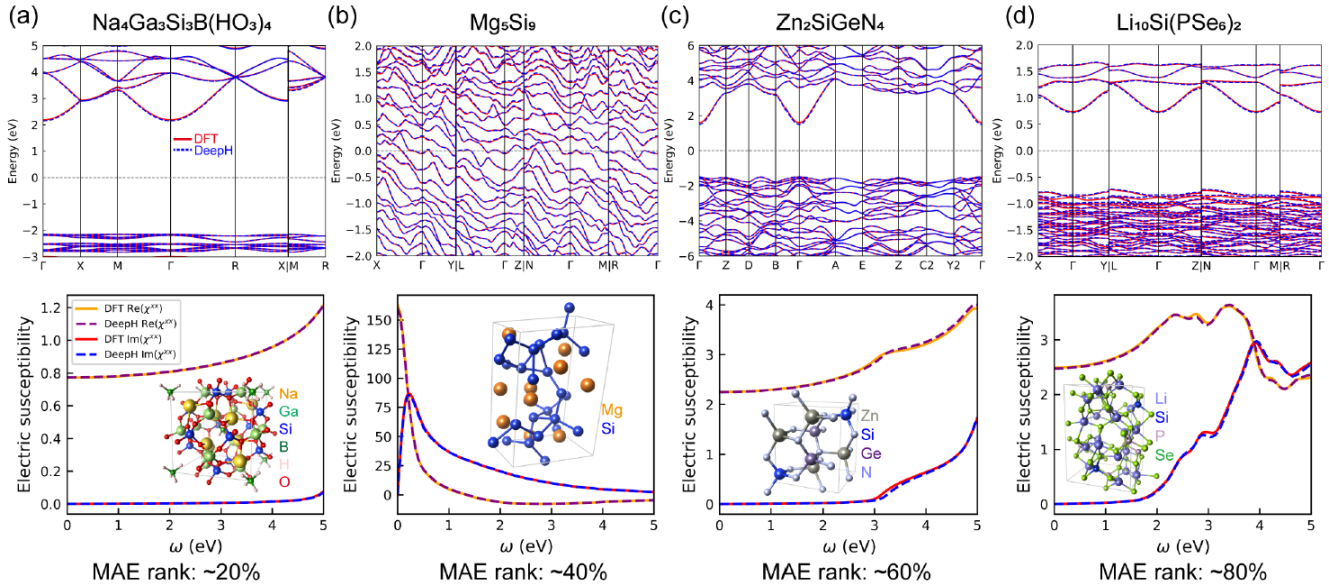
Y. Wang, Y. Li, Z. Tang, et al. **Sci. Bull.** 69, 2514 (2024) **Cover story**

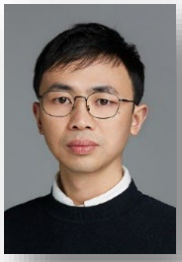
Universal materials model of DeepH

~10^4 materials (20% for test), Averaged MAE 2.2 meV



Outstanding generalization ability



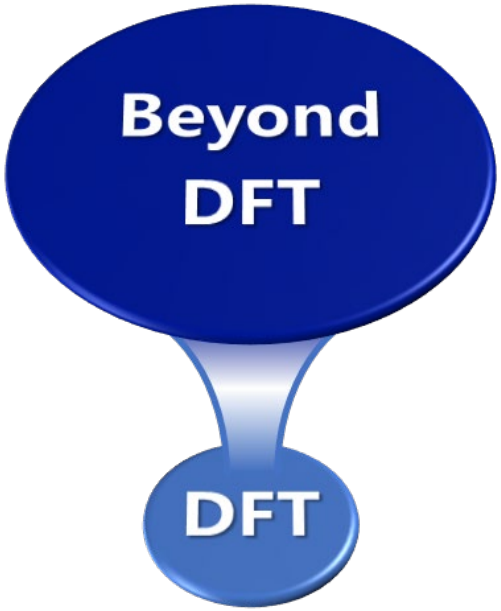


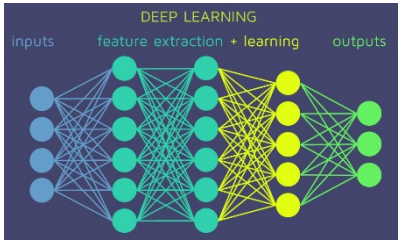
Y. Wang, Y. Li, Z. Tang, et al. **Sci. Bull.** 69, 2514 (2024) **Cover story**

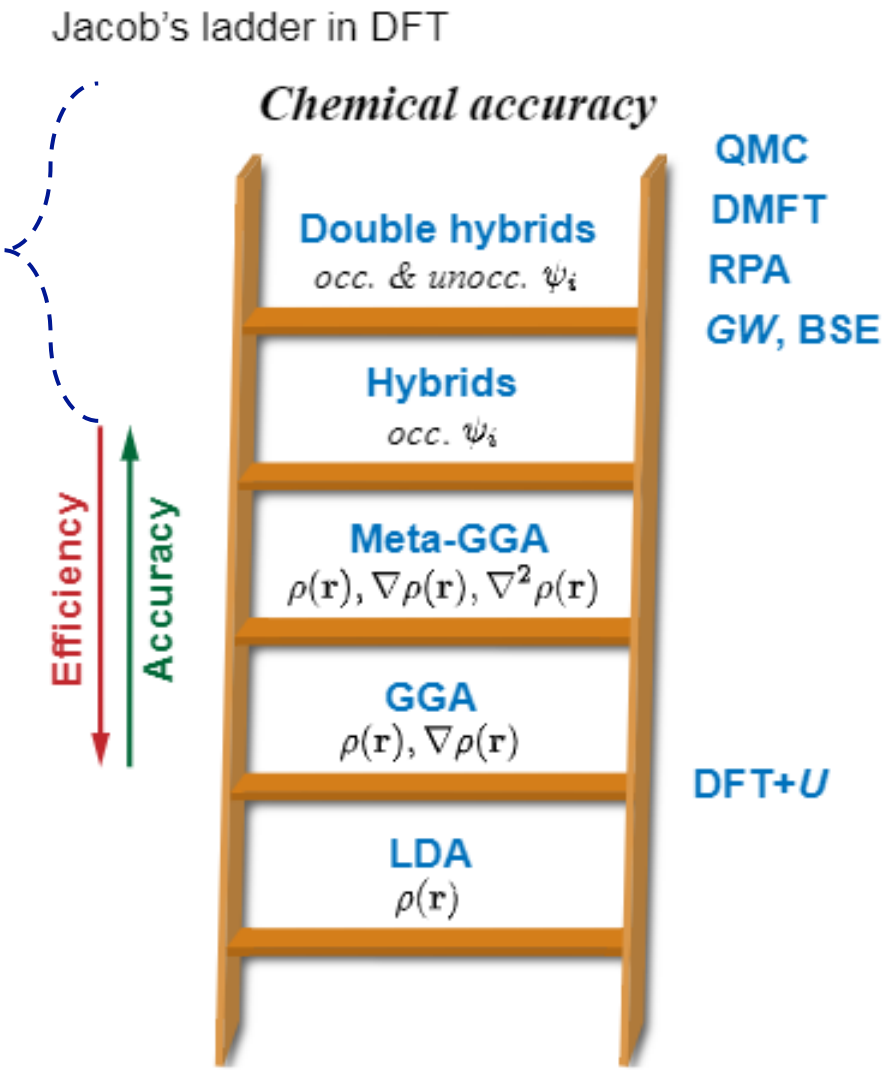
Contents

Outlook (1): Method development

- **Extend the DeepH method from DFT to Beyond DFT**



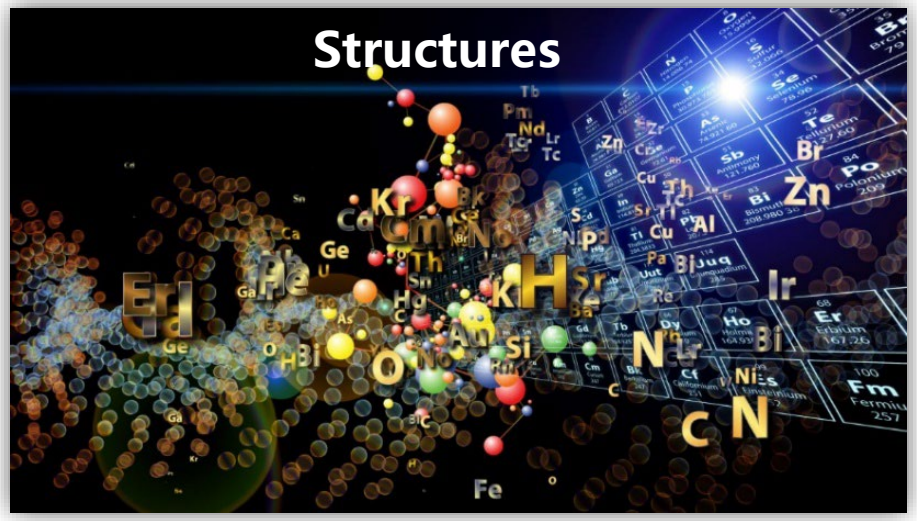


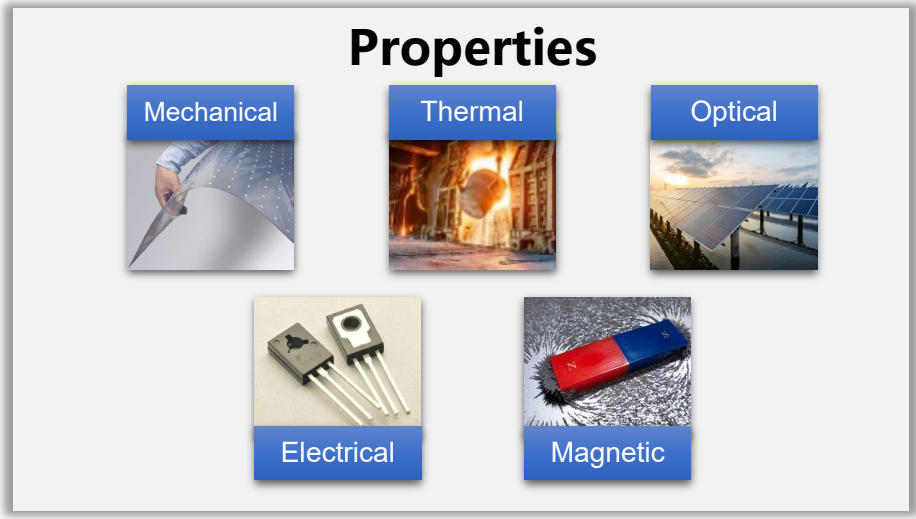


Outlook (2): Foundation models

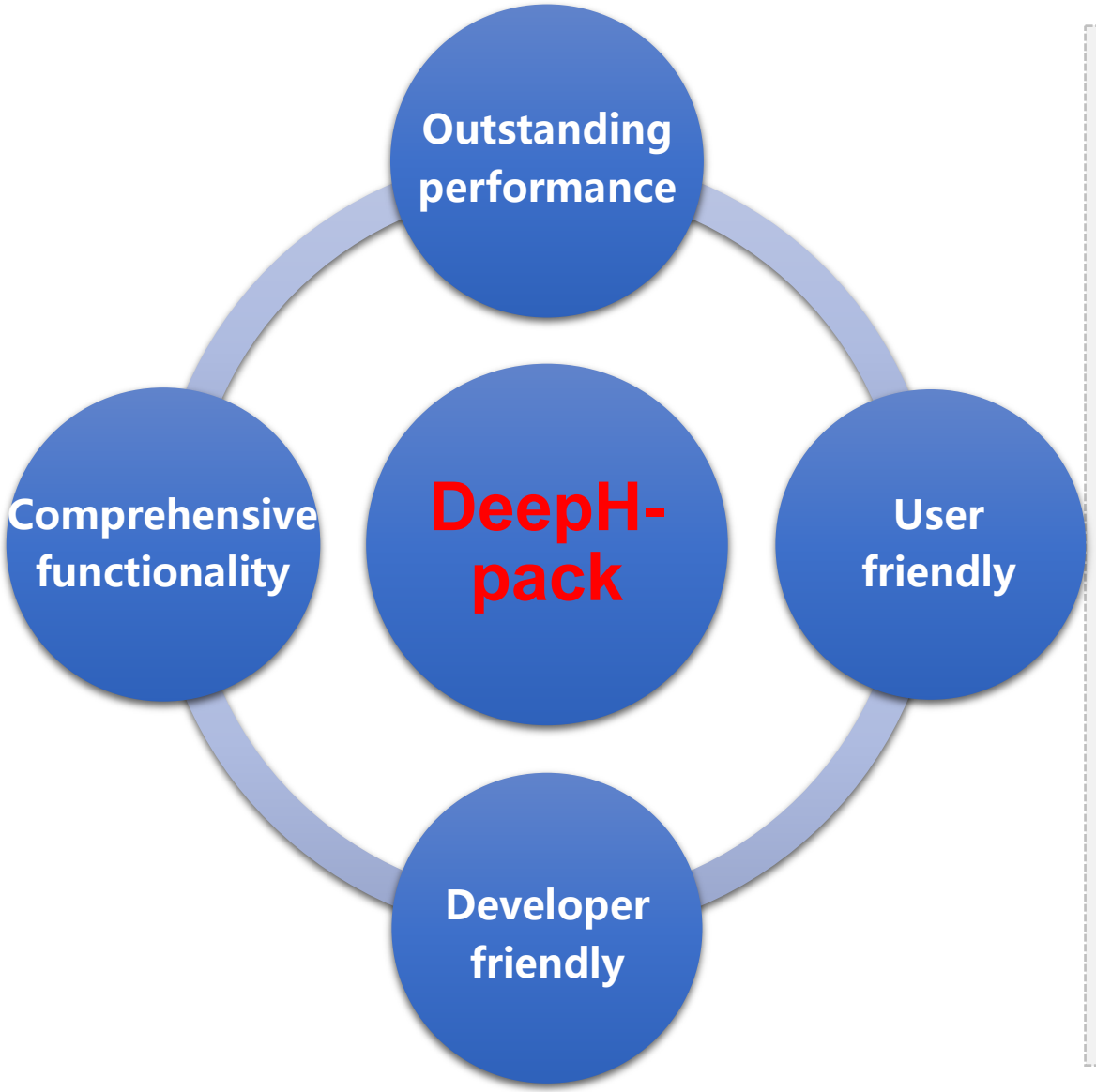
- **Develop foundation models of electronic structure**

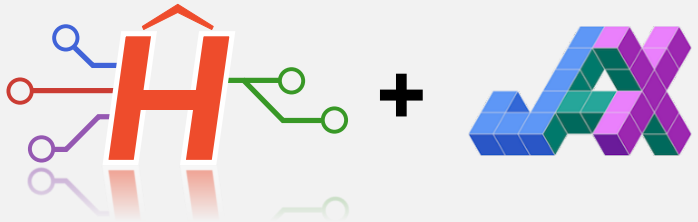
Large materials model





Next-generation computing platform



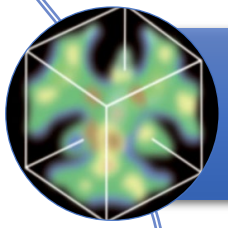


- Training and inference accelerated by 300%, GPU memory usage reduced by over 100%
- Multiple types of neural networks to learn various target physical quantities
- Well-structured software modules
- Comprehensive user manual, video tutorials

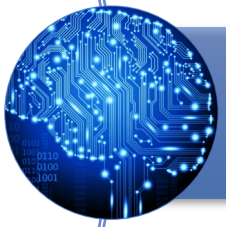
Large database of electronic structures

MIND
Materials INtelligent Database

AI-integrated computational database
(**2,000,000** crystalline materials)



Extensive physical property data



Compatible with deep learning

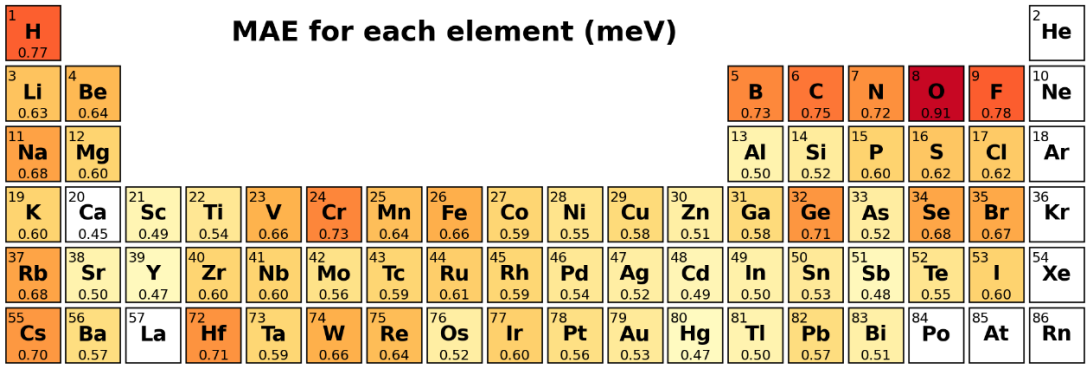
Various methods and software

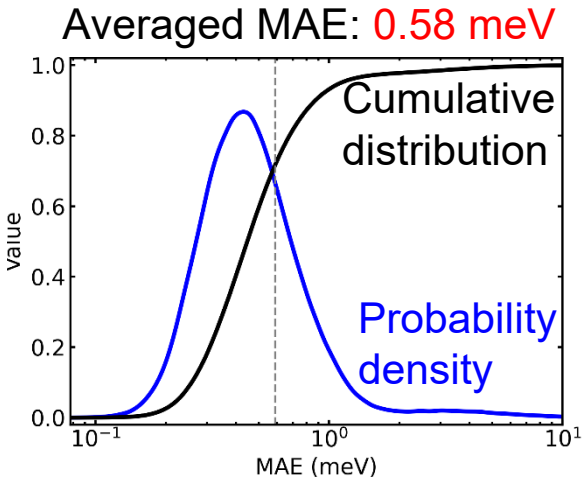
DeepH-pack

MIND
Materials INtelligent Database

Universal materials model of electronic structure

- **World's largest training dataset (2,000,000 crystalline materials)**
- **First-ever model training on millions of material structures**
- **First universal materials model achieving sub-meV accuracy**

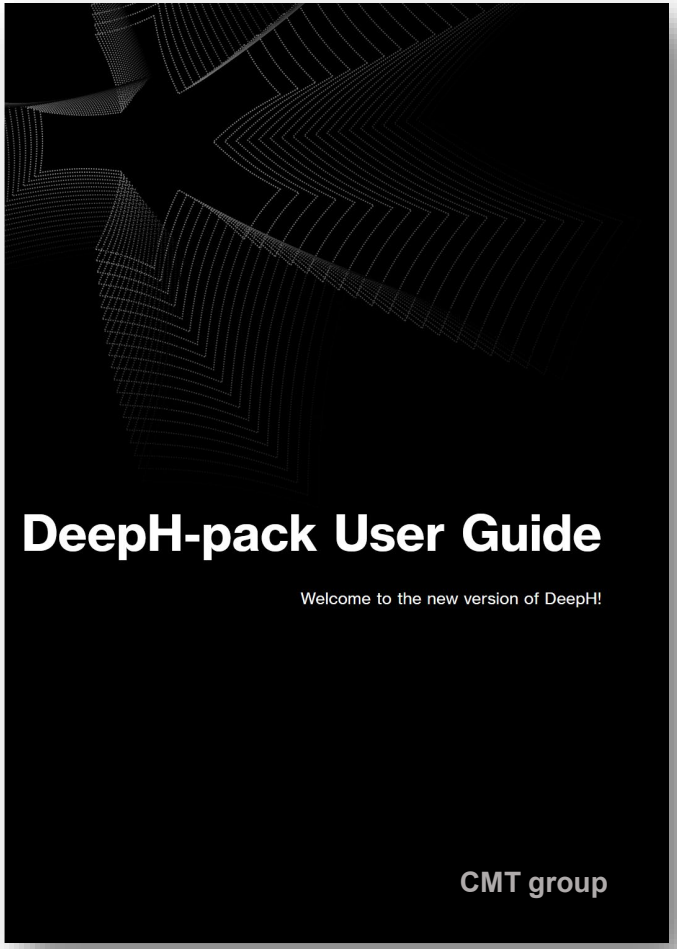


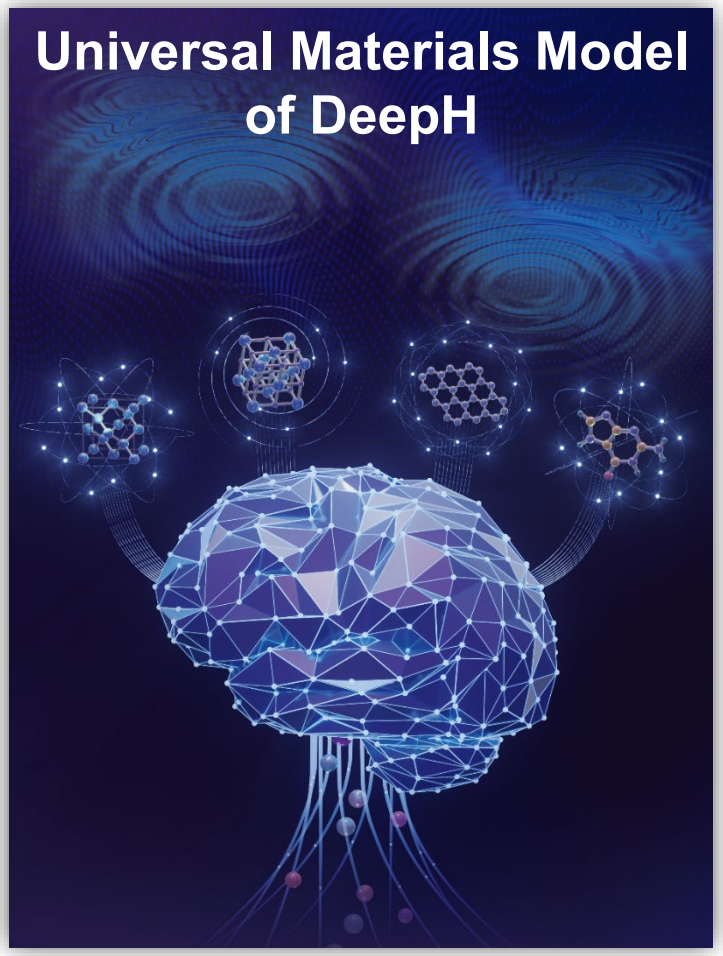


Massive training data	Advanced architecture	Superior performance
Millions of materials	Transformer based	Sub-meV accuracy
10TiB data	Explainable AI	O(*N*) scaling

DeepH Group (to be published)

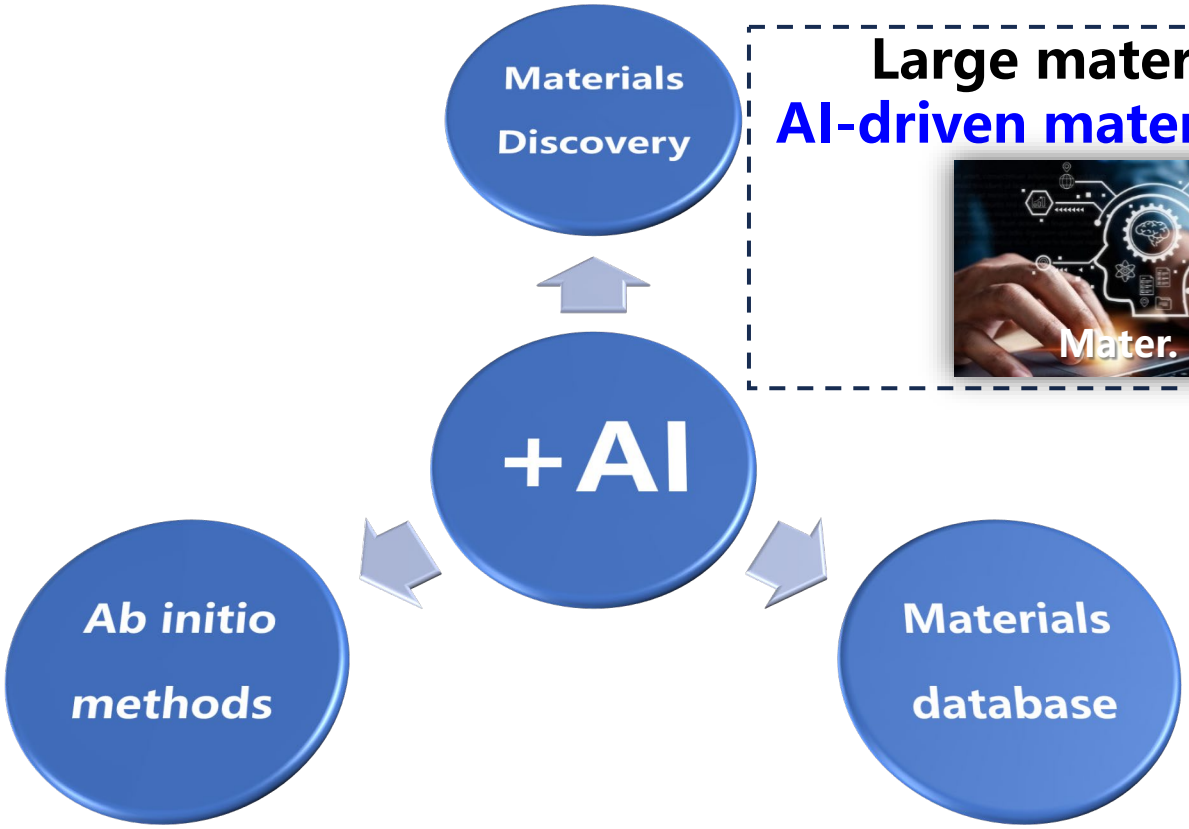
DeepH-pack & Universal materials model





The new version of DeepH-pack, along with universal materials model, will be released soon.

Outlook (3): AI-driven materials discovery



Large materials model

AI-driven materials discovery

Neural-network DFT method

Improving with growing data

DeepH-pack

Big materials database

AI-accelerated data generation

MIND
Materials INtelligent Database

DeepH: References and open-source codes

Develop deep-learning first-principles methods

[1] **DeepH**: H. Li, *et al*. **Nat. Comput. Sci.** 2, 367 (2022) arXiv: 2104.03786
[2] **DeepH-E3**: X. Gong, *et al*. **Nat. Commun.** 14, 2848 (2023)
[3] **xDeepH**: H. Li, *et al*. **Nat. Comput. Sci.** 3, 321 (2023) **Cover story**
[4] **DeepH-DFPT**: H. Li, *et al*. **PRL** 132, 096401 (2024) **Editors' suggestion**
[5] **MagNet**: Z. Yuan, *et al*. **Quantum Front.** 3, 8 (2024)
[6] **DeepH-hybrid**: Z. Tang, *et al*. **Nat. Commun.** 15, 8815 (2024)
[7] **DeepH-2**: Y. Wang, *et al*. arXiv:2401.17015
[8] **DeepH-PW**: X. Gong, *et al*. **Nat. Comput. Sci.** 4, 752 (2024)
[9] **DeepH-UMM**: Y. Wang, *et al*. **Sci. Bull.** 69, 2514 (2024) **Cover story**
[10] **DeepH-Zero**: Y. Li, *et al*. **PRL** 133, 076401 (2024) **Editors' suggestion**
Ab initio artificial intelligence, H. Li, *et al.* **MGE Advances** e16 (2023)
Deep-learning electronic structure calculations, Z. Tang, et al. **Nat. Comput. Sci.** (in press)

DeepH-pack

Tutorial: https://www.bilibili.com/video/BV1Tv4y1H7TD

DeepH: https://github.com/mzjb/DeepH-pack
https://deeph-pack.readthedocs.io

DeepH-E3: https://github.com/Xiaoxun-Gong/DeepH-E3

xDeepH: https://github.com/mzjb/xDeepH

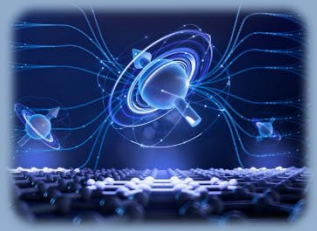
Welcome to 2025 DeepH Workshop!

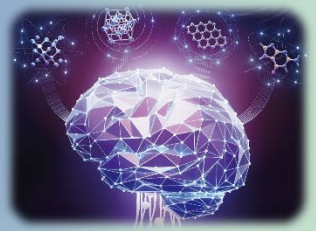
DeepH-pack

2025年DeepH研讨会

第一性原理遇见人工智能

想洞悉AI+物理最前沿?
想掌握DeepH新方法?
让我们相约清华物理系!





「时间」2025年11月21-24日
「地点」清华大学物理楼

活动安排

- AI物理前沿报告
- DeepH上机实践
- DeepH应用挑战赛

会议注册

主办单位: 清华大学物理系、AI物理研究中心
会议召集人: 徐勇、段文晖
会议联系人: 崔莹 **联系电话:** 010-62798591
联系邮箱: cuiying0121@tsinghua.edu.cn
报名截止时间: 2025年11月16日