



Tracing Cr Migration in Sb_2Te_3 with Computational Simulation: Topological Materials Engineering for Next-Generation Thermoelectrics



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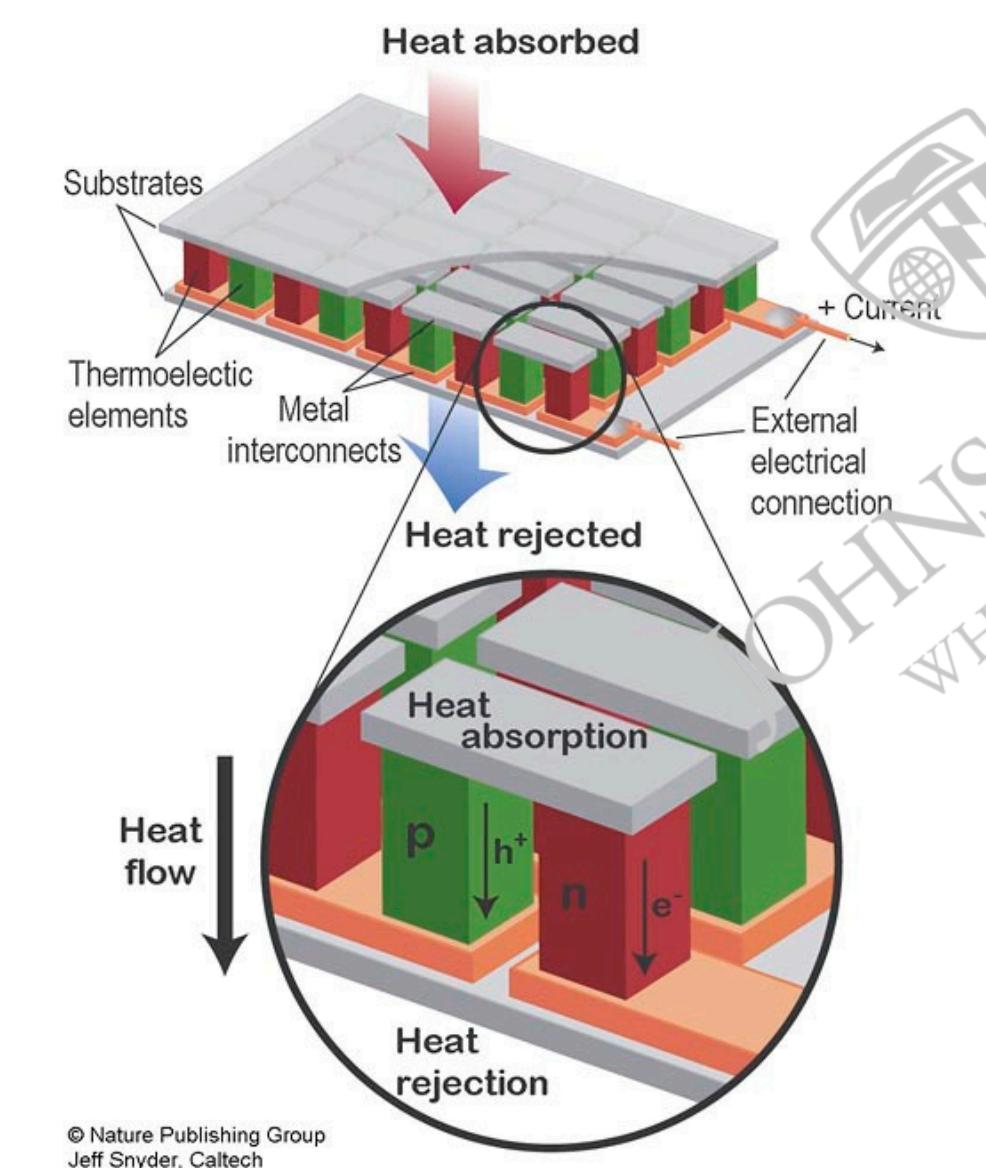
Introduction

Thermoelectrics: Conversion of waste heat to electrical energy—an approach to fight the energy crisis!

Why Sb_2Te_3 ? 2D material's quantum confinement and spin textures can largely modulate thermopower generation.

Why doping with Cr? Magnetically doped topological insulators enable energy-efficient electronics through the quantized anomalous Hall effect (QAHE).

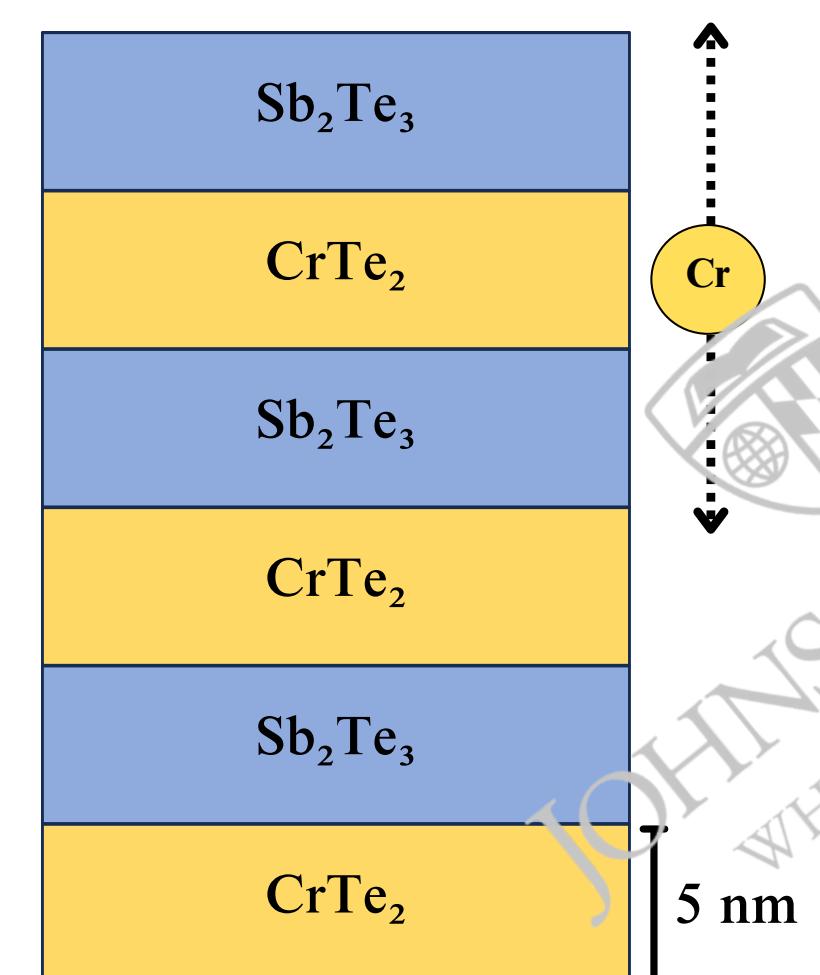
Focus: Sb_2Te_3 doped with Cr



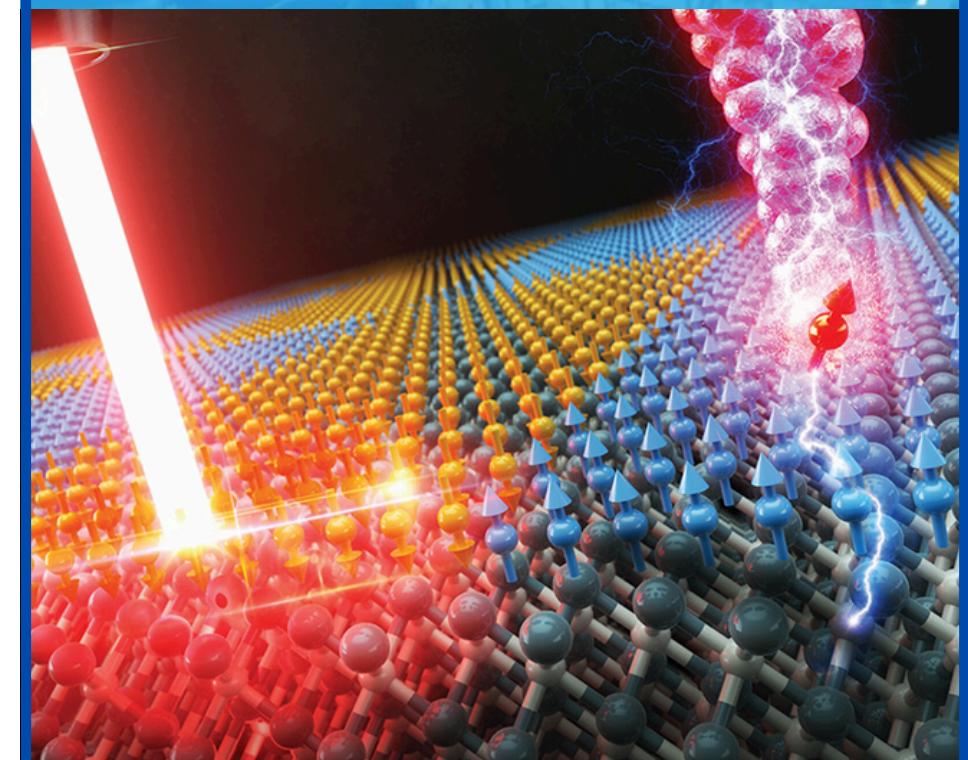
Objectives & Impact

Combine computational and experimental investigation on Cr dopant diffusion in vdW layered Sb_2Te_3

- How will the Cr doping evolve? — energetic profile analysis
- How do different Cr doping sites affect thermoelectric behavior? — electronic structure and molecular dynamics simulation



Hot & Cold Reactions to Produce Electricity



Significance

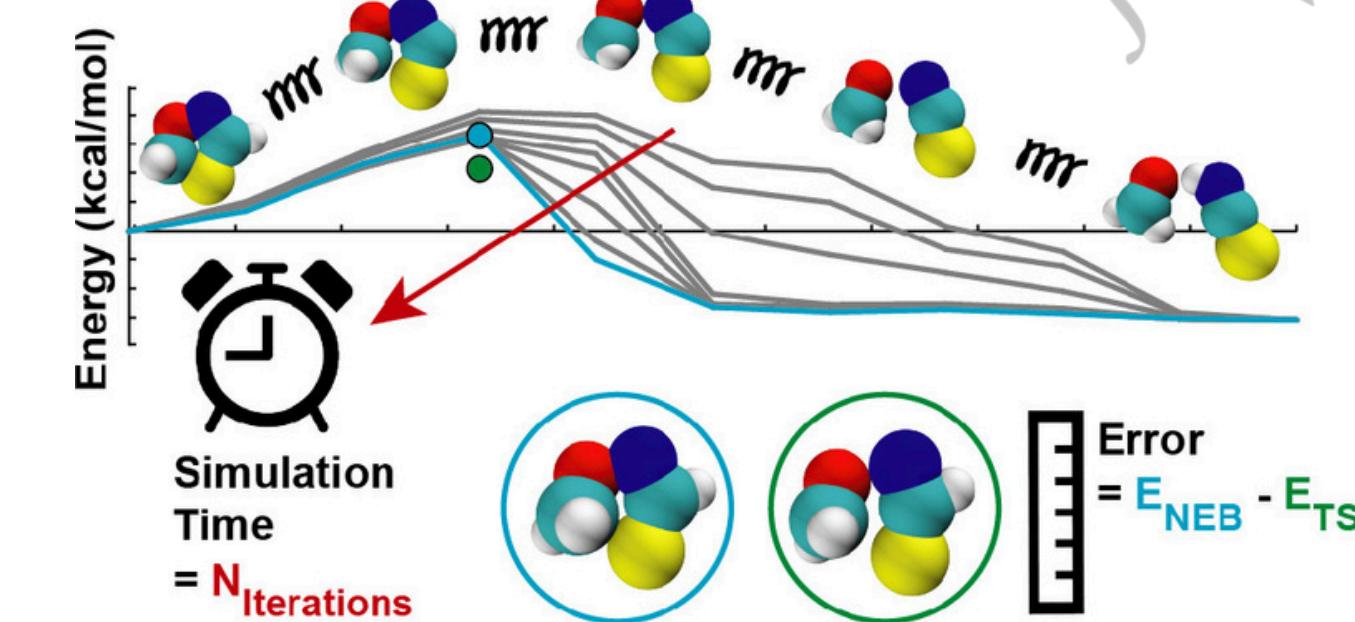
Temperature differences are harnessed by thermoelectrics to generate electricity, turning waste heat into power.

Investigating the microscopic nature of thermopower in quantum materials provides insights for the design of next-generation thermoelectrics!

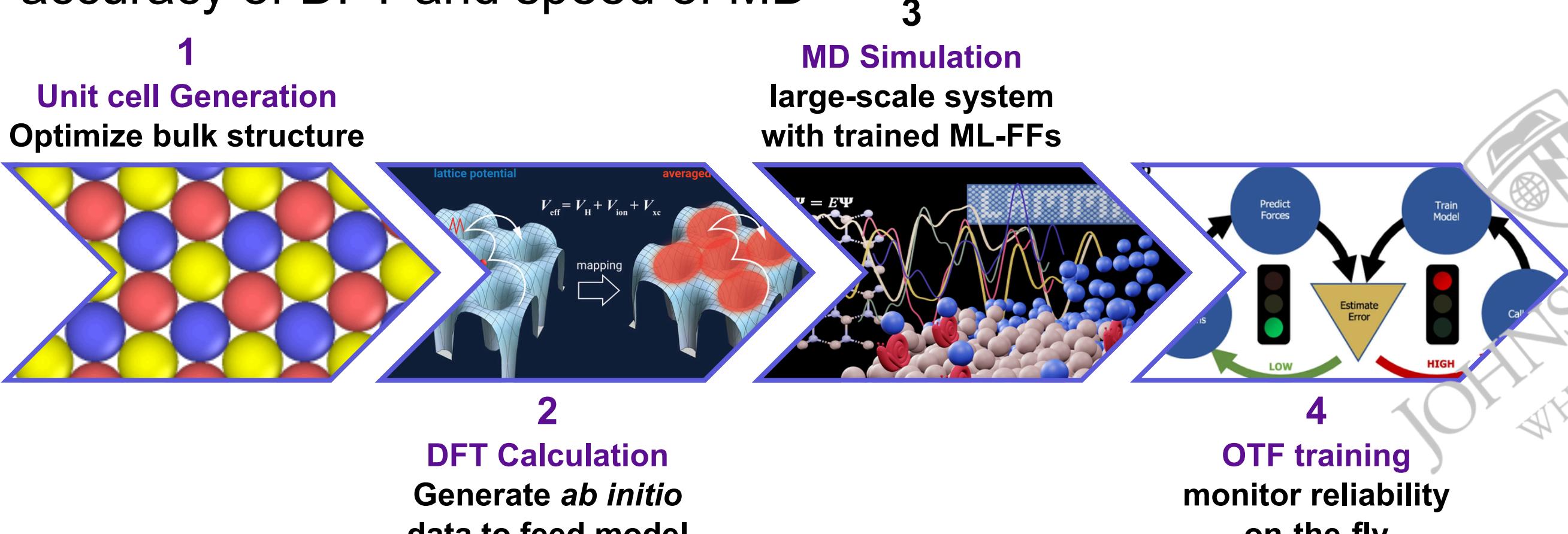
Methods

Density Functional Theory (DFT) & Nudged Elastic Band (NEB)

Ab initio Molecular Dynamics (AIMD): "ground truth" of Cr diffusion behavior



Machine learned force field (MLFF) model: combines accuracy of DFT and speed of MD



Results

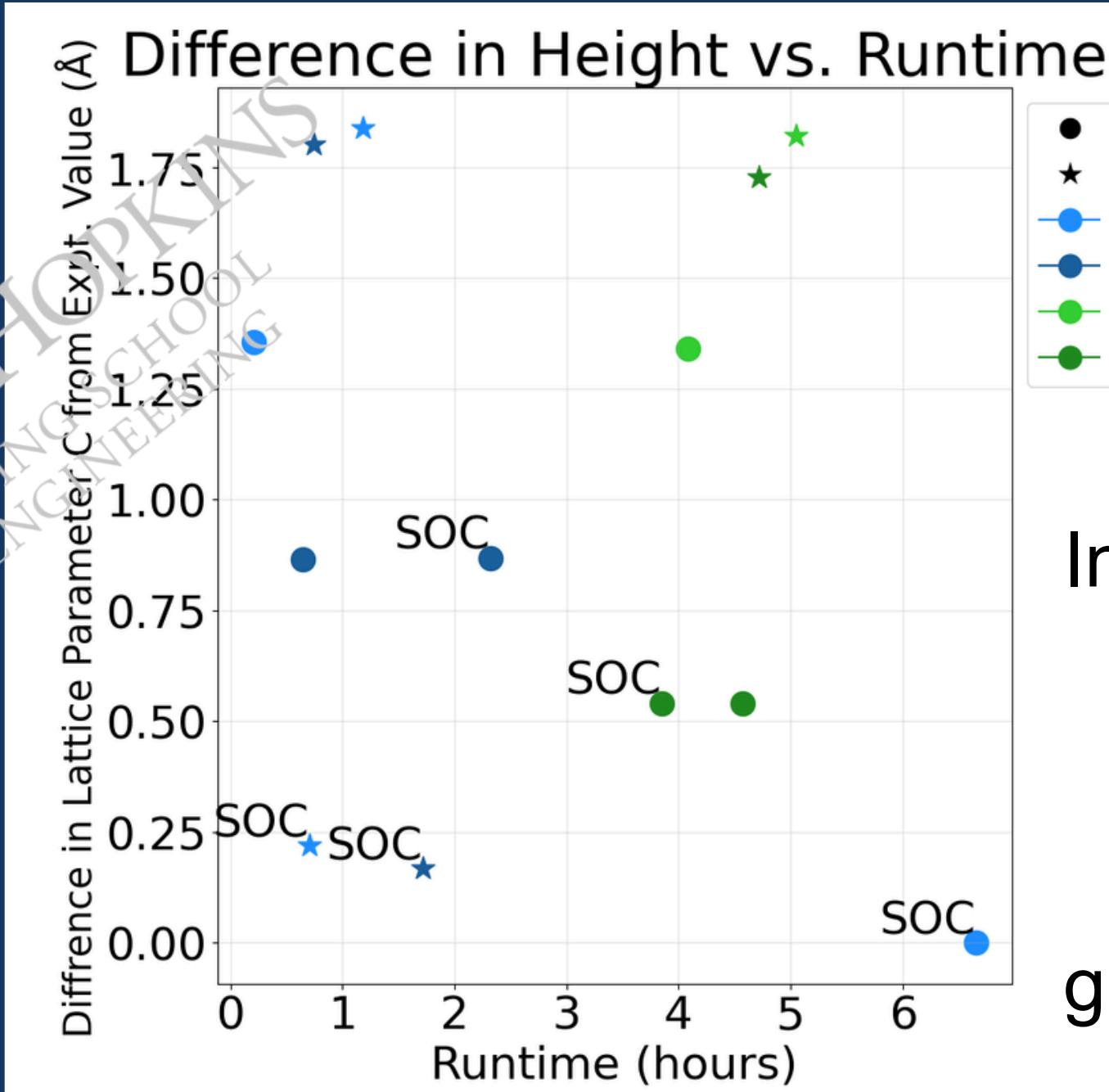
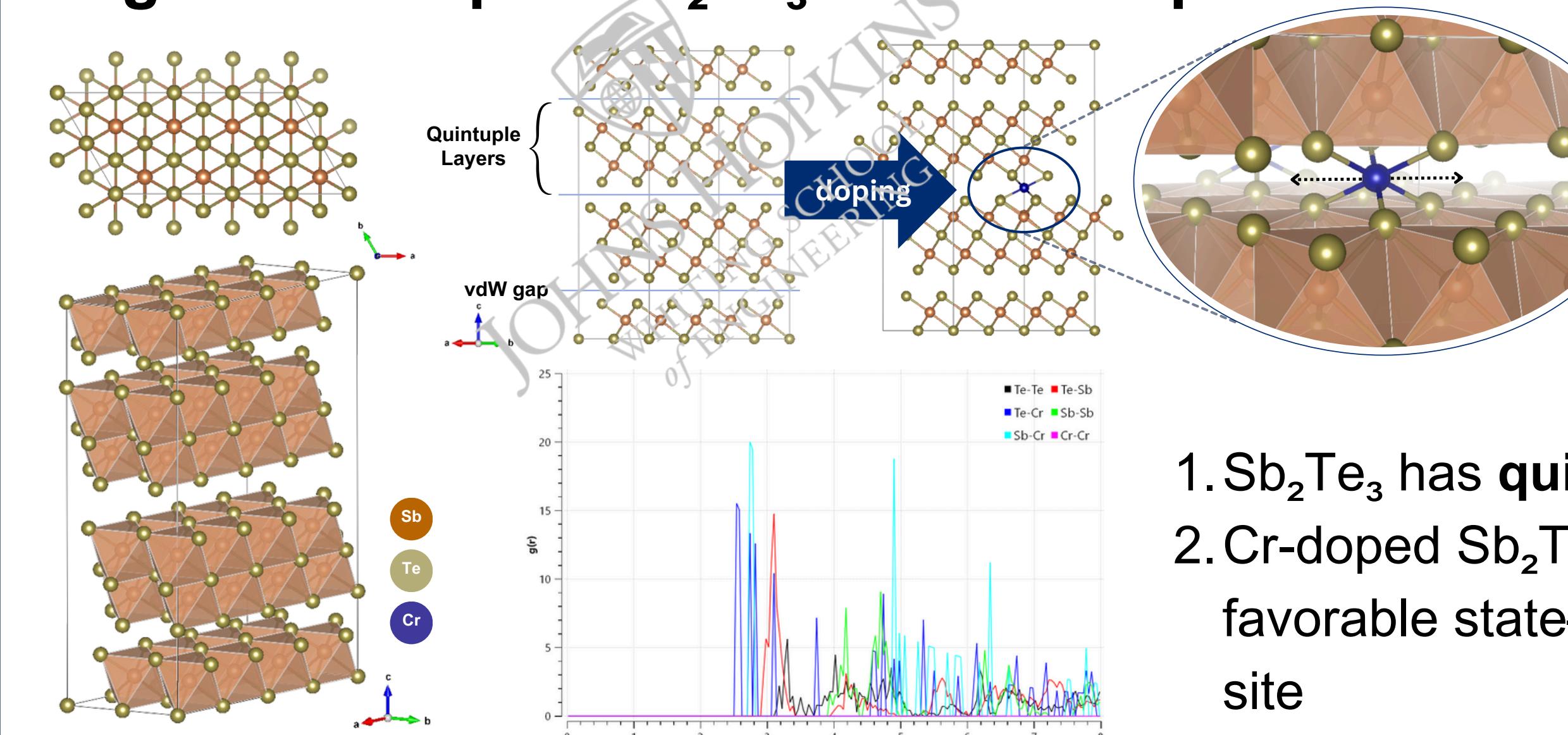


Fig. 1 - DFT Benchmarking analysis:
Which terms are important?
Incorporation of:
1. Spin Orbit Coupling (SOC)
2. vdW Correction Term
3. Relativistic Effects
gives accurate and fast results.

Fig. 2 - Cr-doped Sb_2Te_3 structure optimization



1. Sb_2Te_3 has quintuple layers with vdW gaps.
2. Cr-doped Sb_2Te_3 : relaxes for energetically favorable state—Cr "trapped" in octahedral site

Fig. 3 - Cr Doping site matters!

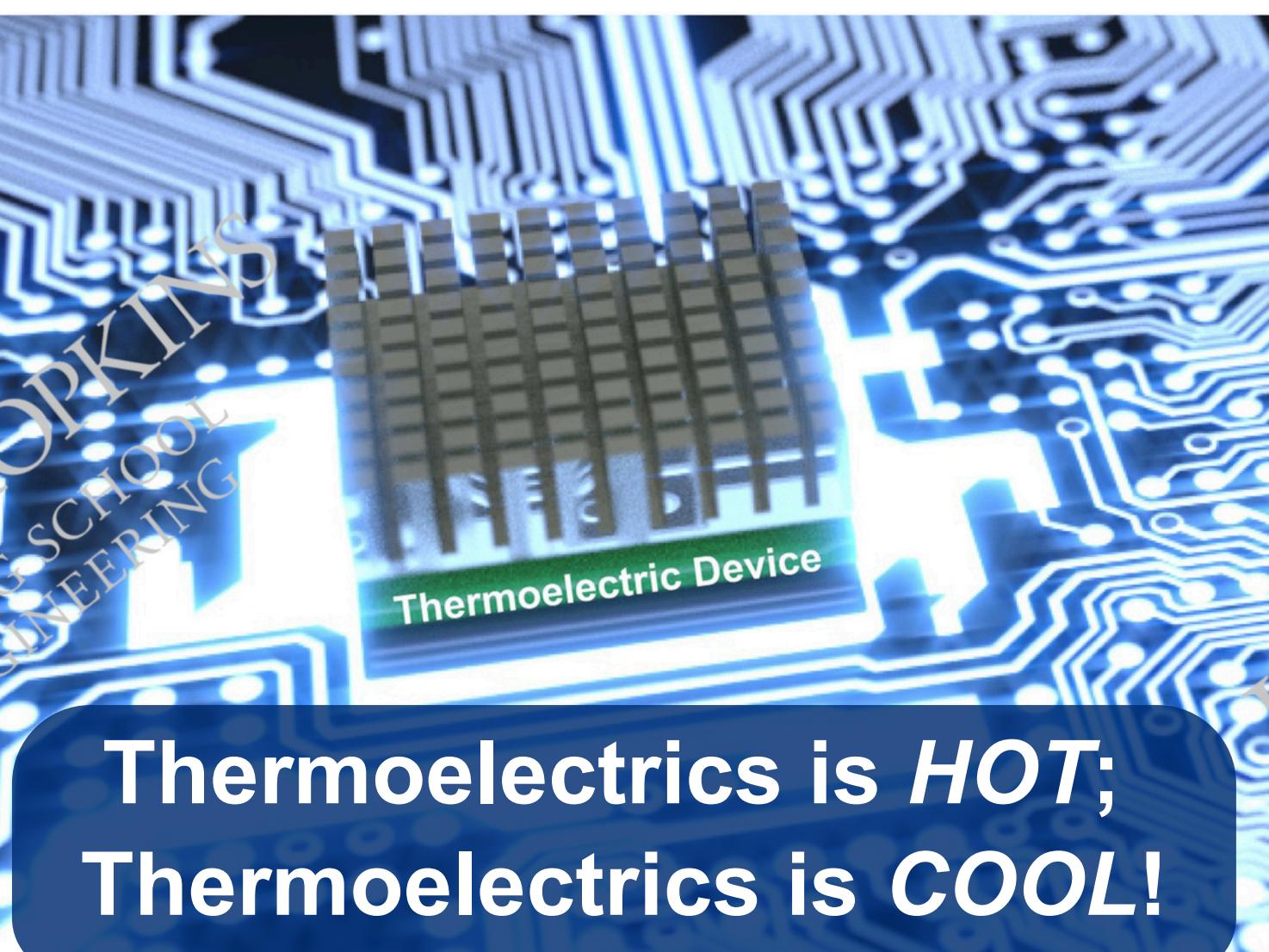
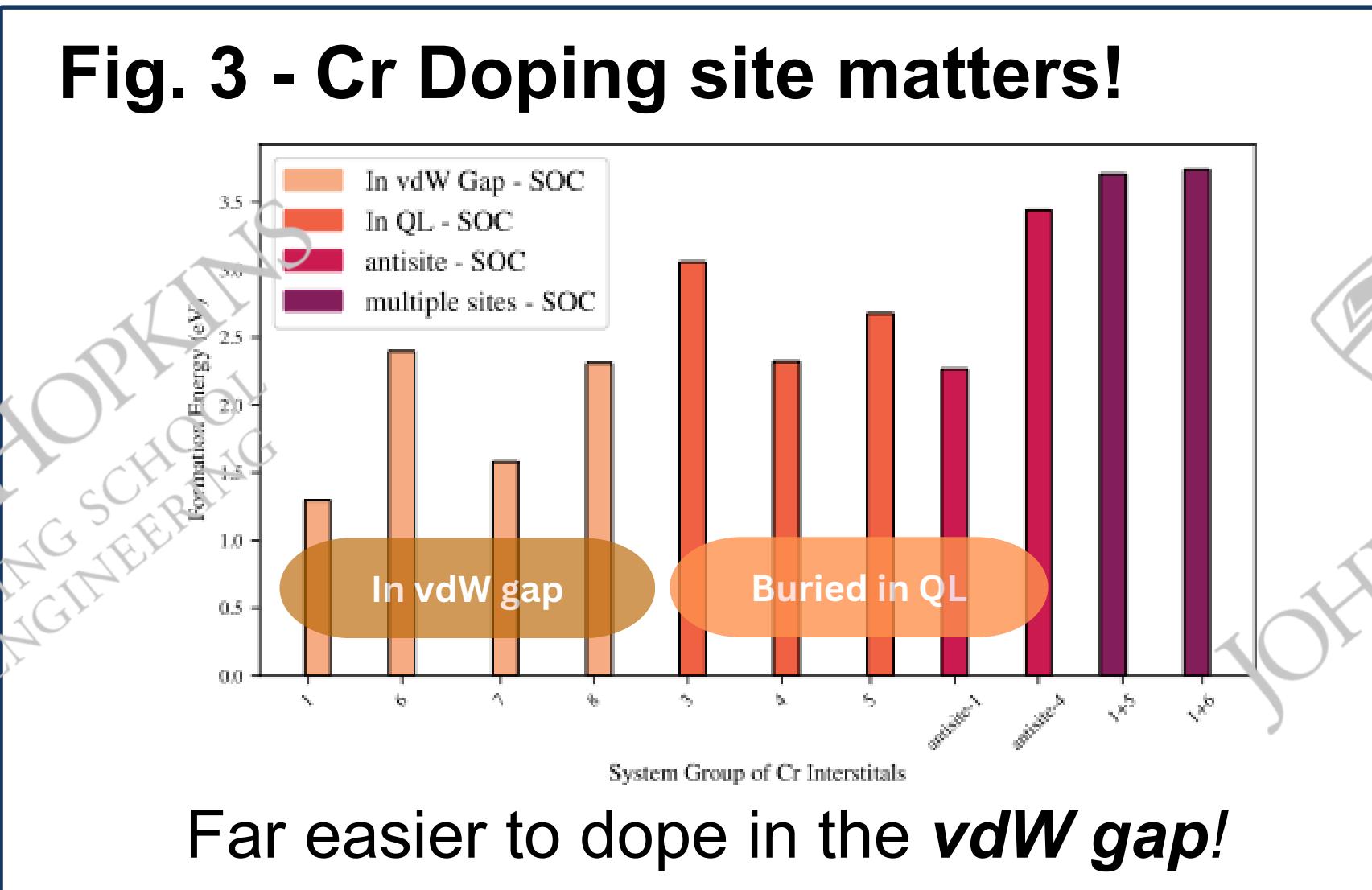
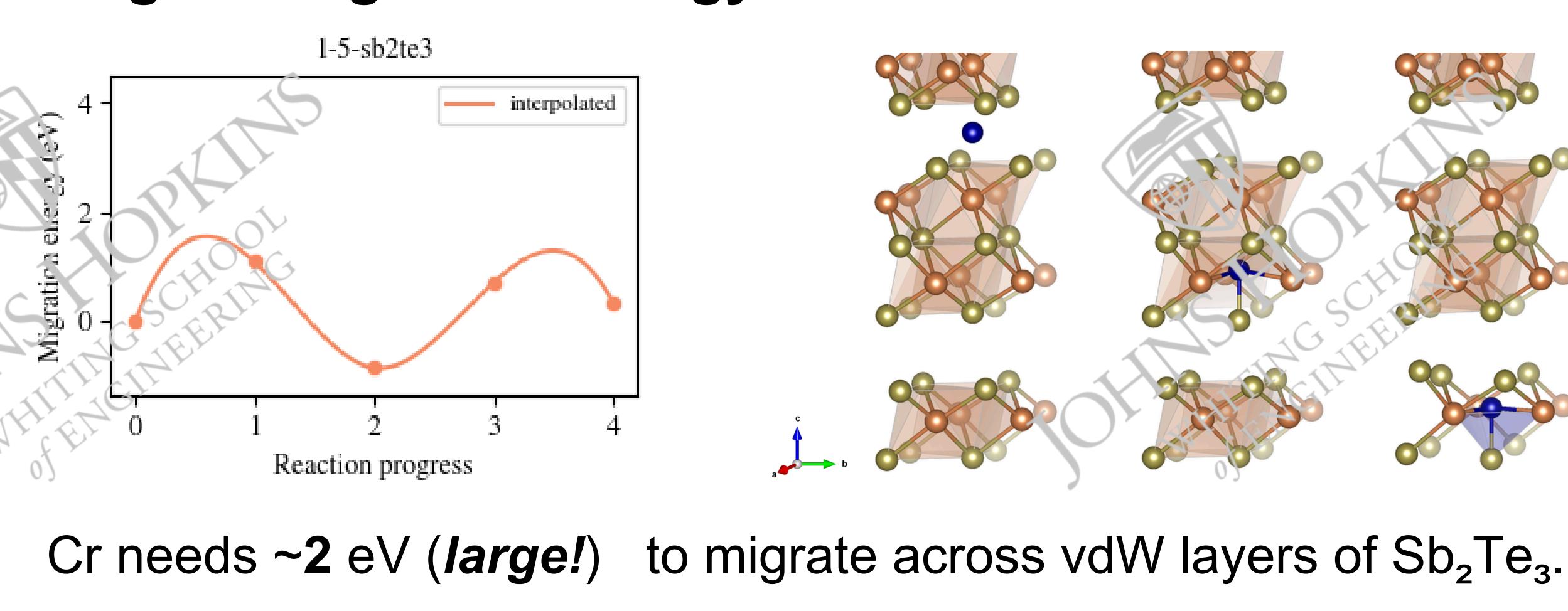


Fig. 4 - Migration energy barrier for Cr diffusion



Findings & Conclusions

Cr doping site matters:

The location of the Cr interstitial strongly affects the energy that it needs to form and the band structure, potentially enabling band engineering by doping.

Migrating Cr dopants needs energy:

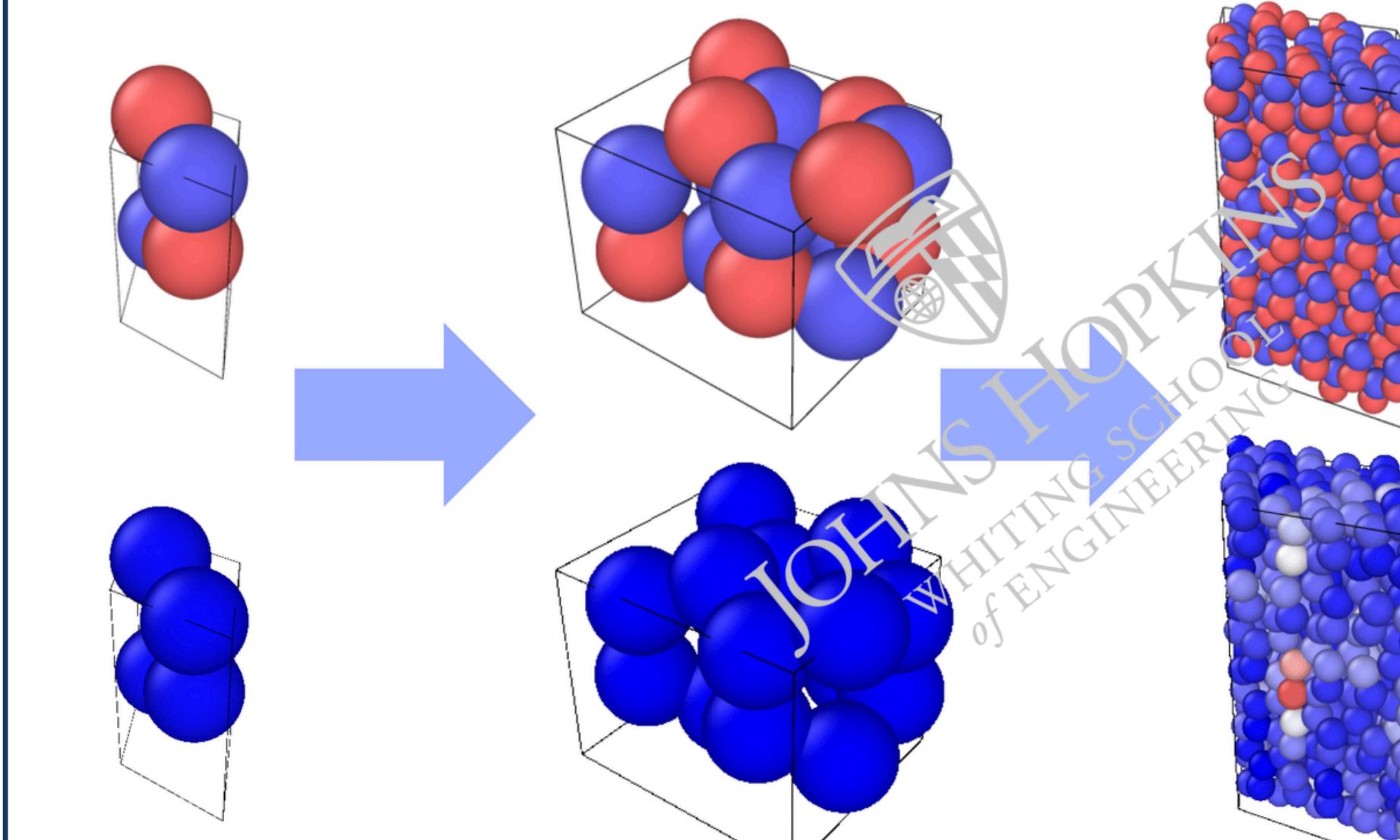
It is easier to form Cr interstitials in Sb_2Te_3 's vdW gap than to form buried in the quintuple layer. As a result, Cr atoms are likely to be trapped in the vdW gaps

FLARE Model Improvement:

Model accuracy decreases at higher temperatures. Better sampling strategy will help model larger system!

Fig. 6 - Preliminary FLARE Training Results

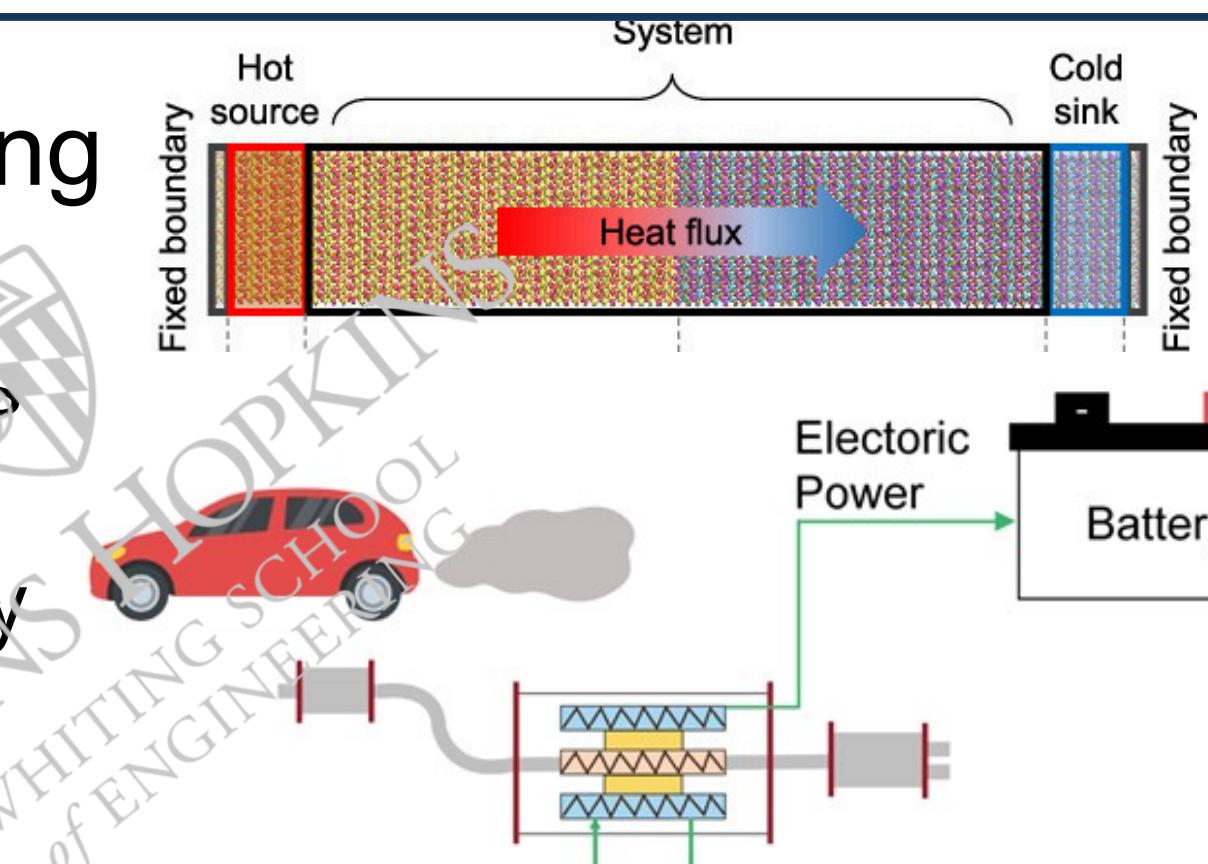
Trained on small Sb_2Te_3 system and tested on larger system for MD: complete model benchmark



Future Directions

Next step towards optimizing Cr doping in Sb_2Te_3 :

1. Larger-scale modeling: MD simulations for bulk material property
2. Bridge micro- and macro- properties by combining DFT and machine learning



References & Acknowledgements

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- [2] J. Chem. Theory Comput. 2022, 18, 2993–3005
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- [4] Phys. Rev. B 2024, 109, 075421