P3: Stacking-dependent binding energy of bilayer graphene from quantum Monte Carlo

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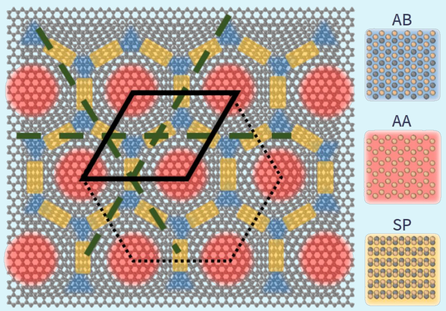
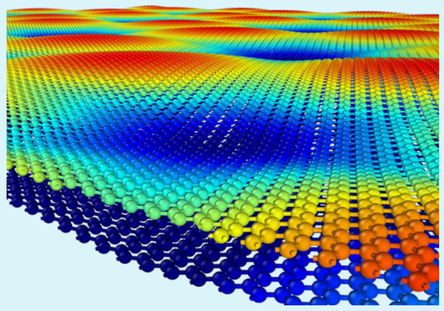
qmc-hamm.github.io

Regions of twisted bilayer graphene with different stacking types



This results in out-of-plane corrugations, which are influenced by the interlayer interactions

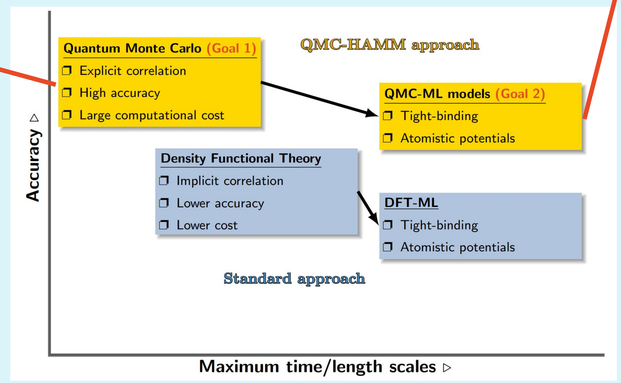
Geometry relaxations require an accurate van der Waals description between bilayers



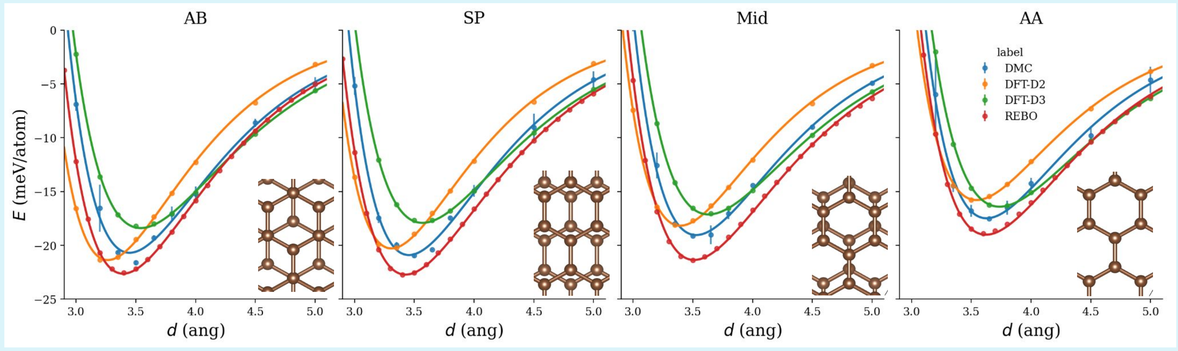
Problem: Not enough accurate data to fully parameterize the stacking dependence

Goal 1: Diffusion Monte Carlo (DMC) to calculate the accurate interlayer interactions of bilayer graphene

- DMC is a stochastic method for finding the ground state energy
- Computes the van der Waals energy accurately [1]
- QMCPACK code package scales efficiently on the Summit supercomputer [2]



Diffusion Monte Carlo binding energy data



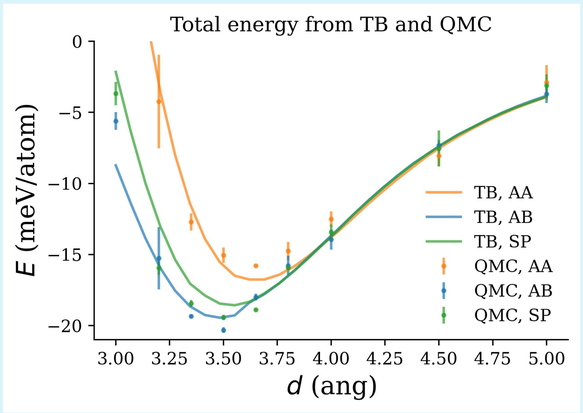
Shortcomings of DFT van der Waals functionals and an empirical potential

- DFT-D2 functional mispredicts the minimum
- DFT-D3 functional underestimates the difference between AA and AB stacking
- REBO empirical potential overestimates the binding energies

DMC energy data product: https://github.com/qmc-hamm/interlayer_energies

Goal 2: Total energy tight-binding parameterization by fitting to the DMC data

- Twisted bilayer graphene is not tractable by first principles calculations
- **Problem:** Existing parameterizations are not transferable to twisted and corrugated geometries.



Software product: parameterization integrated tools for large scale calculations in LATTE-LAMMPS

Conclusions

- **Accurate** binding energy data set is generated using diffusion Monte Carlo
- **Fast**: The data set is used to develop the total energy correction to the tight-binding parameterization
- **General**: Enables a low-cost study of twisted bilayer graphene

References

[1] Mostaani, Drummond, and Fal'Ko. Physical Review Letters 115.11 (2015): 115501.
[2] Kim et al. Journal of Physics: Condensed Matter 30.19 (2018): 195901.

Acknowledgements

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