Ab Initio Simulations of Cathode Materials — Efficient Configurational Screening to Discover Novel and Stable Compositions

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Introduction to the GOAC code

- **Partial site occupations** result in configurational disorder
- Configurations can be evaluated by **electrostatics** in ionic crystals
- Global optimization of Atomistic Configurations by Coulomb (**GOAC**) finds lowest energy configuration
- Utilizes advanced **optimization** algorithms based on **Monte Carlo** and **Genetic Algorithms**
- **Open-Source Code**

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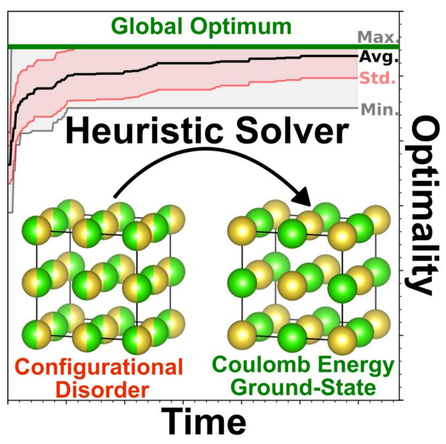
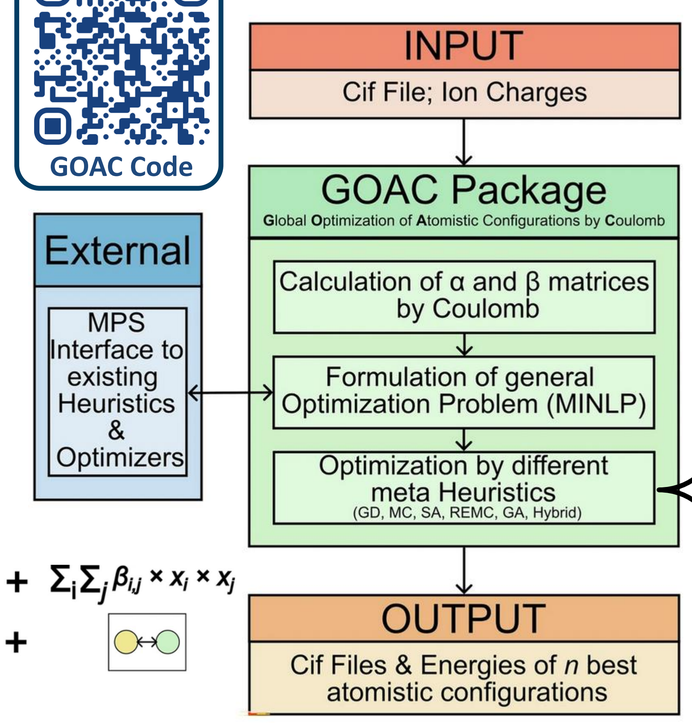


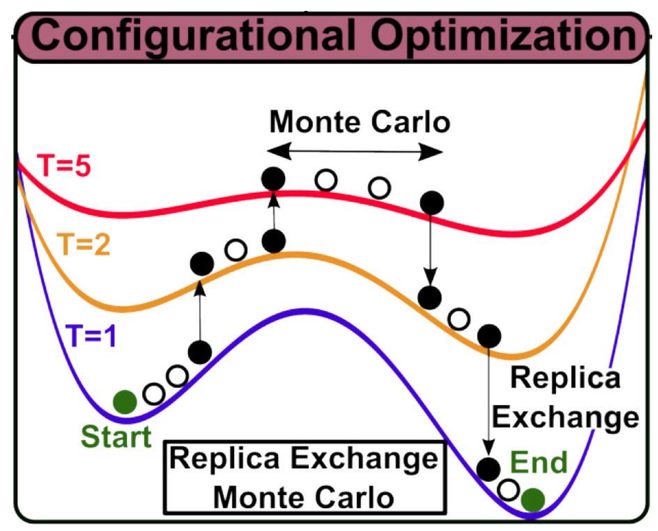
Energy Calculation:

$$E_{tot} = E_{const} + \sum_i \alpha_i \times x_i + \sum_i \sum_j \beta_{i,j} \times x_i \times x_j$$

[1,0,0,1]

GOAC Code

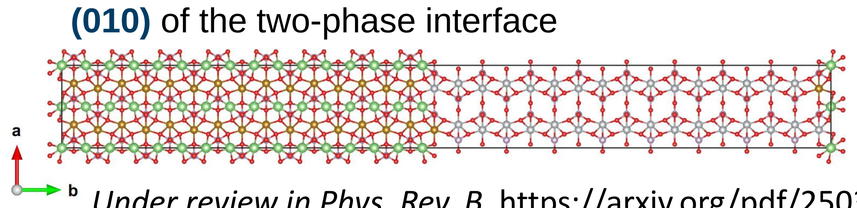




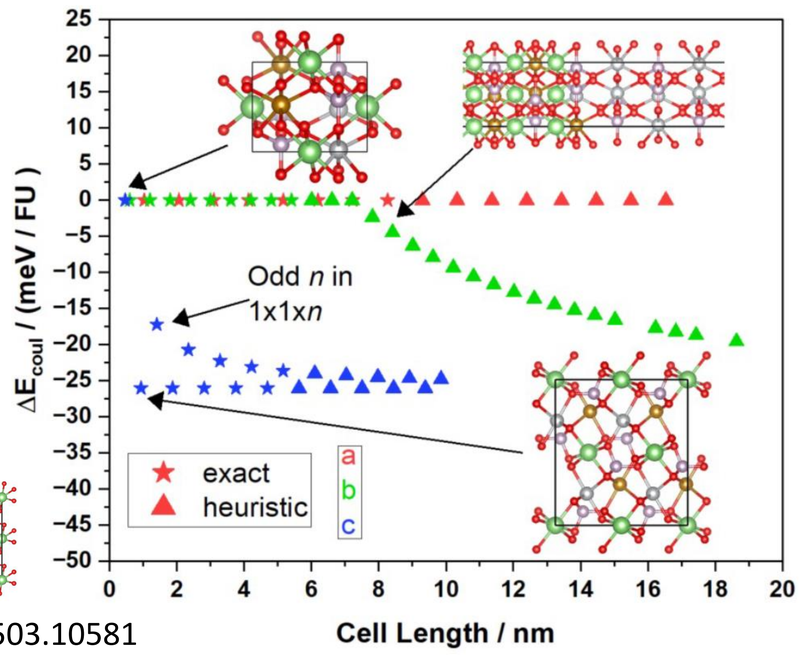
Gradient Descent (GD), Metropolis Monte Carlo (MC) , Simulated Annealing MC (SA), Replica Exchange MC (REMC), Genetic Algorithm (GA)

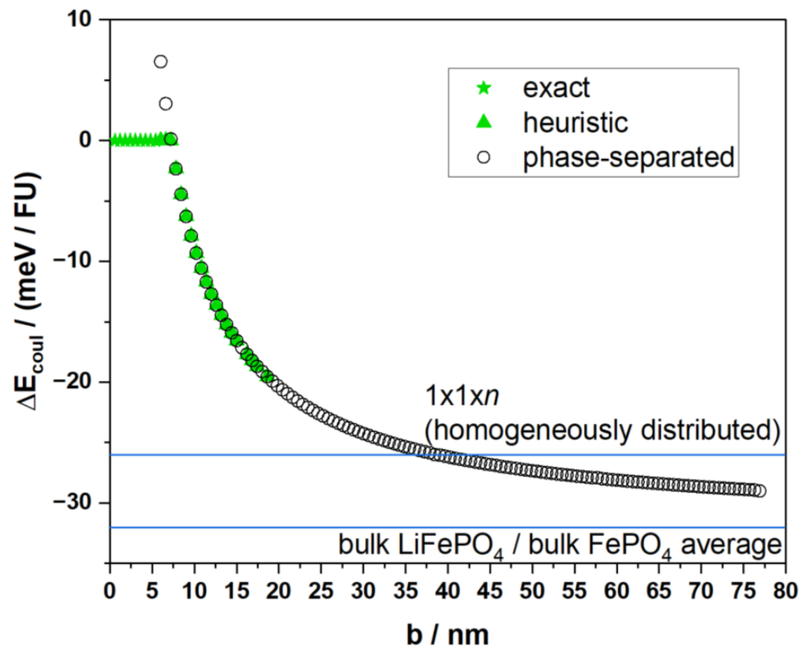
Particle-Size Dependence of Single-Phase – Two-Phase Charging Characteristics in LFP

- Lithium-Iron Phosphate (LFP) is known to show a **two-phase charging mechanism**
- Optimizations with GOAC in various supercell sizes yield a **single-phase up to** particle-sizes of approx**. 40 nm**
- **Experimentally** it is also shown that LFP has a **single-phase** charging mechanism at the **nanoparticle** regime
- Moreover, electrostatic optimizations with GOAC match the reported **surface orientation (010)** of the two-phase interface



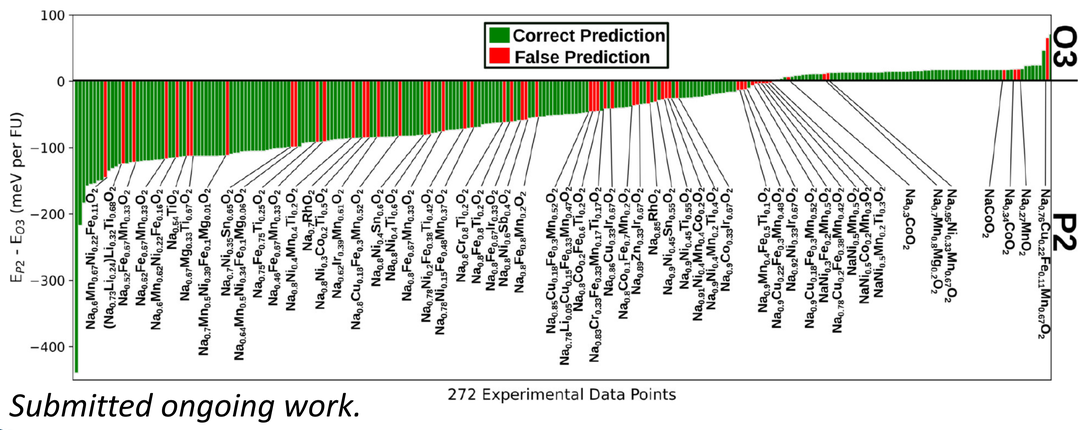
Under review in Phys. Rev. B, https://arxiv.org/pdf/2503.10581



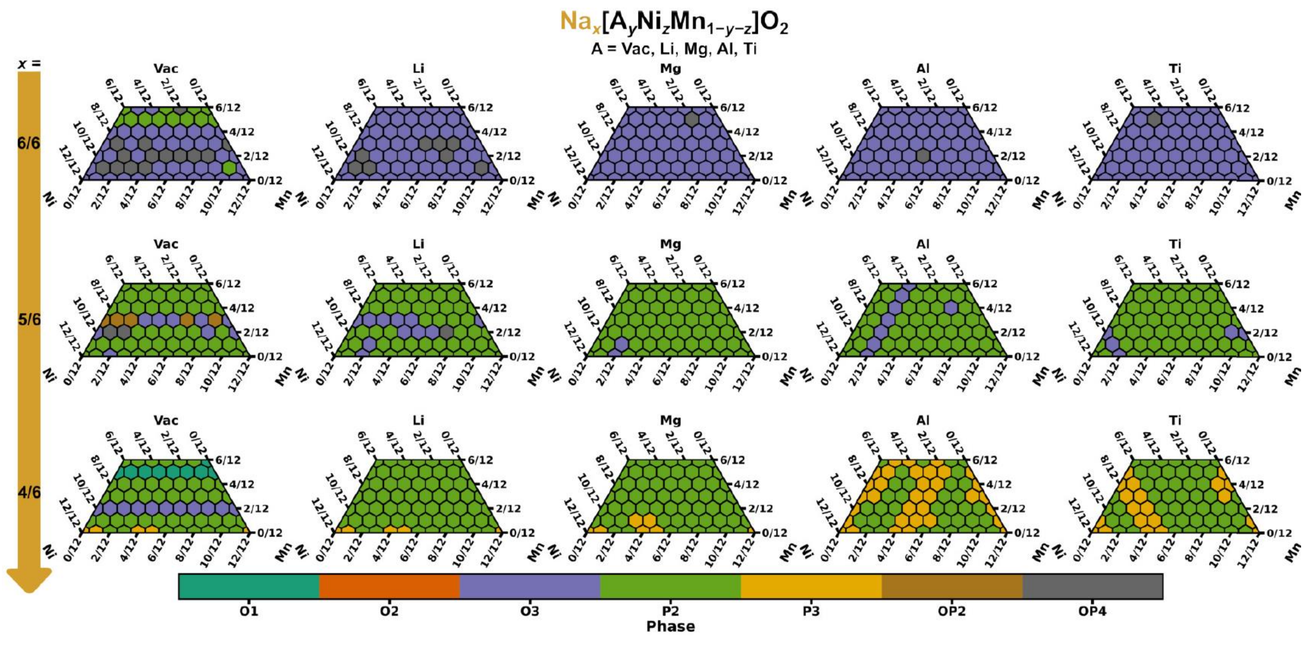


High-Throughput Predictions of Phase-Stabilities of Layered Oxide Sodium-Ion Cathodes

- **Quantitative, *ab initio*** prediction of most stable phases (**O1, O2, O3, P2, P3, OP2, OP4**) in layered oxide sodium-ion cathodes
- Energy evaluation of phases at **lowest energy configurations**
- Predictions are in **80% of cases in agreement to experiment**
- **High-Throughput studies** are possible employing the GOAC code



Submitted ongoing work.



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Conclusions and Outlook

- **GOAC** is a tool to optimize configurational disorders
- Configurational optimization of **battery materials** can give insights into **charging mechanisms** and **phase stabilities**
- **GOAC** offers **fast optimizations** in **huge configurational spaces** (10^{300} configurations)

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