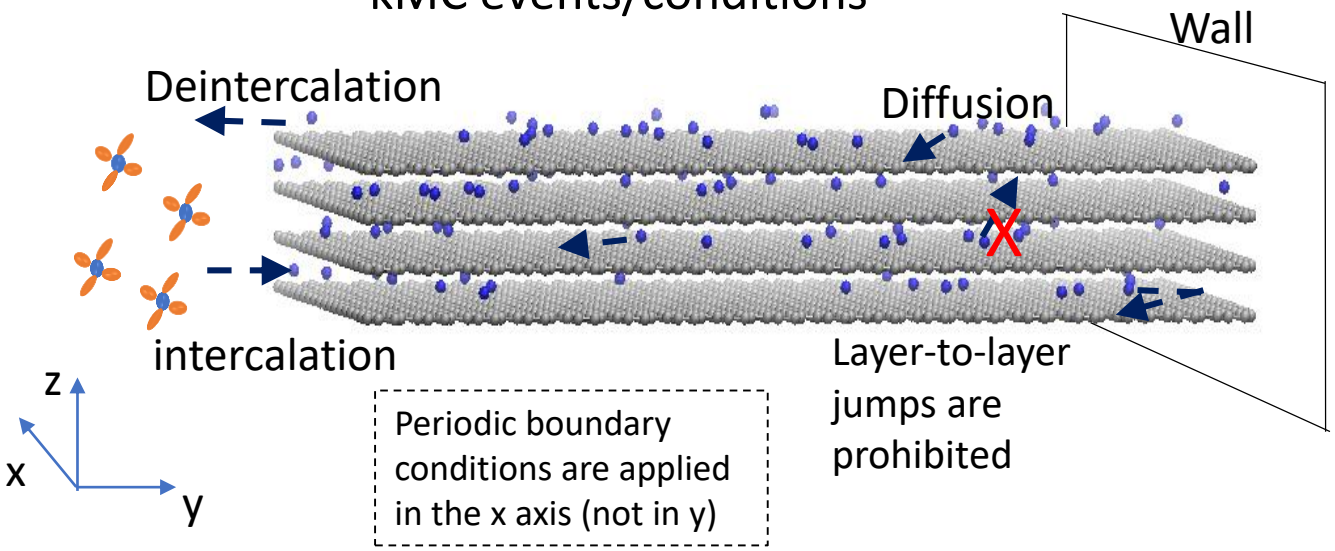
kMC model for Li ion/graphite

Workflow for autonomous Discovery of Battery Components

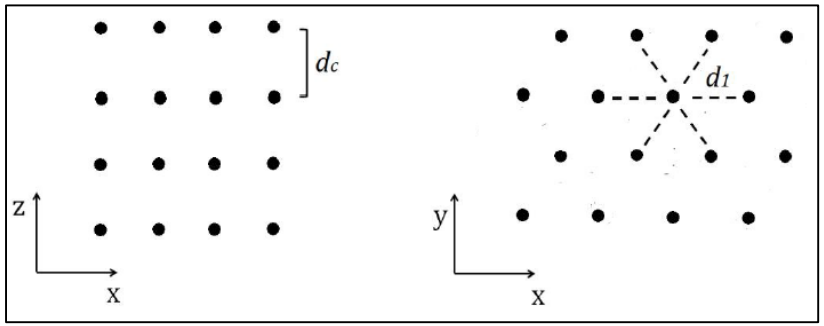
Maximiliano Gaviláns

kMC events/conditions



Periodic boundary conditions are applied in the x axis (not in y)

A perfect graphite particle is modelled with a lattice-gas, which is contructed with the sites where Li+ intercalate (black points). This forms a pile of layers, each of them with a triangular geometry of fixed intercalation points

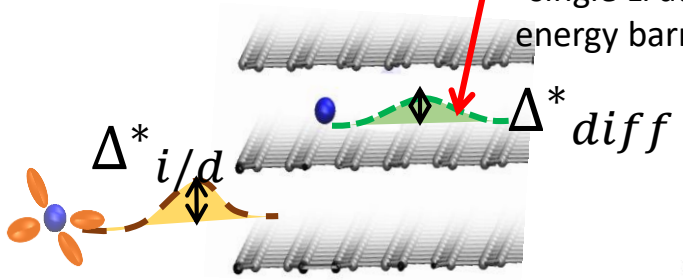


Rate equation [1]

$$\Gamma = \nu_0 \exp\left(-\frac{\Delta^*_\lambda}{k_B T}\right) \exp\left(-\frac{H_F - H_I}{2k_B T}\right)$$

Pre-exponential (vibrational) factor [3]

"single Li atom" energy barrier



Energy barriers to calculate with DFT!

Hamiltonian of final and initial configurations of the lattice [2]

We can also model a Hamiltonian with DFT (second step?)

[1] Mitchell et al. Journal of Electroanalytical Chemistry 493 (2000) 68–74

[2] E.M. Perassi and E.P.M. Leiva, Electrochemistry Communications 65 (2016) 48–52

[3] K. Toyoura et al. Phys. Rev. B 78 (2008)

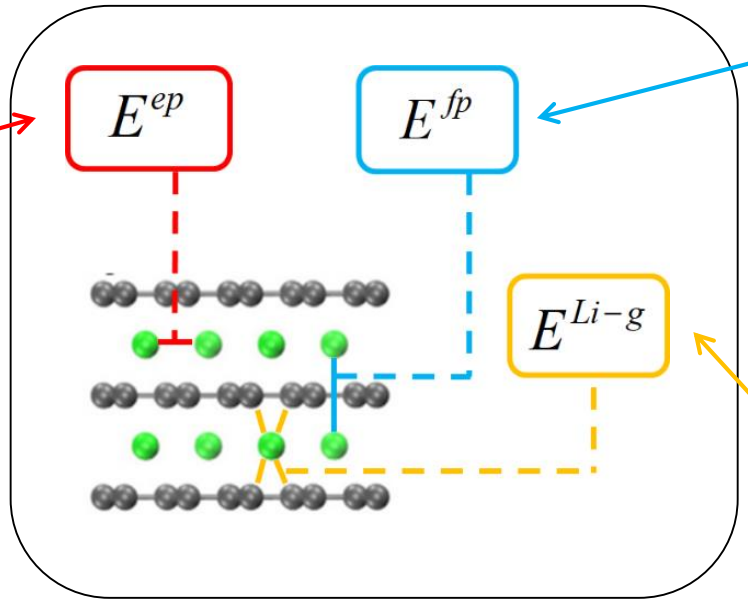
Hamiltonian details

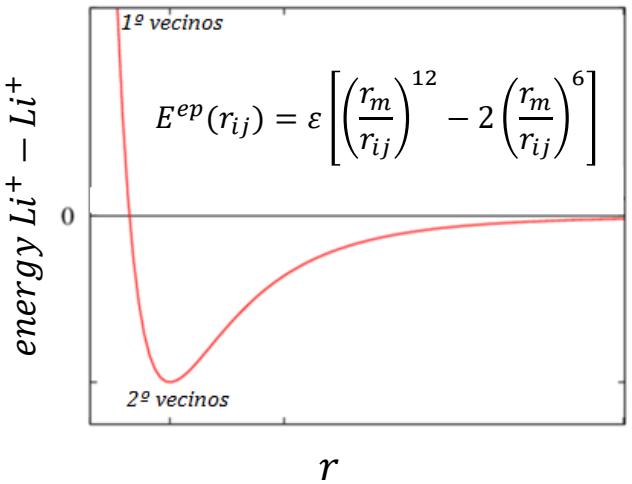
Three energy terms:
- interaction Li-Li in the same layer (red)
- interaction Li-Li in ions different layers (blue)
- interaction Li-C (yellow)

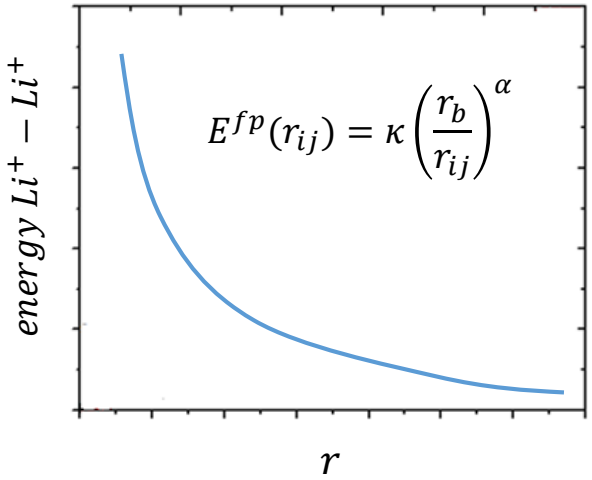
$$H = \sum_{i}^{M}\sum_{j\neq i}^{N^{ep}} \frac{c_i c_j}{2} E^{ep}(r_{ij}) + \sum_{i}^{M}\sum_{j}^{N^{fp}} \frac{c_i c_j}{2} E^{fp}(r_{ij}) + \sum_{i}^{M} c_i E^{Li-g} \quad [2]$$

Repulsion term for Li-Li interactions in different layers

Lennard-Jones potential for Li-Li interactions in the same layer







Constant value

[2] E.M. Perassi and E.P.M. Leiva, Electrochemistry Communications 65 (2016) 48–52