SCALE-UP: An implementation of Second-Principles DFT

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Luxembourg Institute of Science and Technology

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Cantabria University Campus

Funding

RyC programme

What can we do with first-principles simulations?

Predict material properties using just fundamental constants

Little input from user necessary to obtain reliable information

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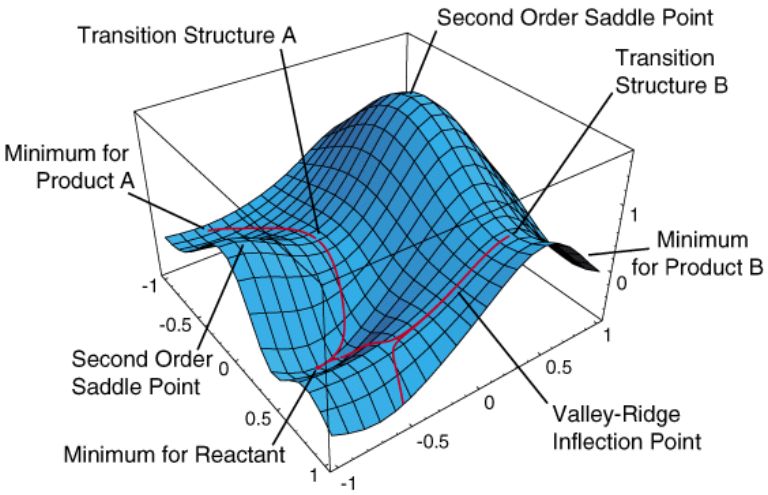

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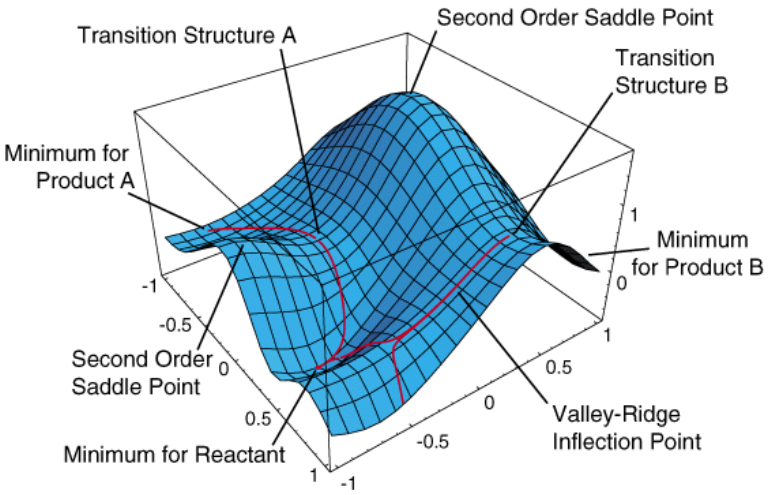

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- ✓ Accurate energies
- ✓ Equilibrium geometries

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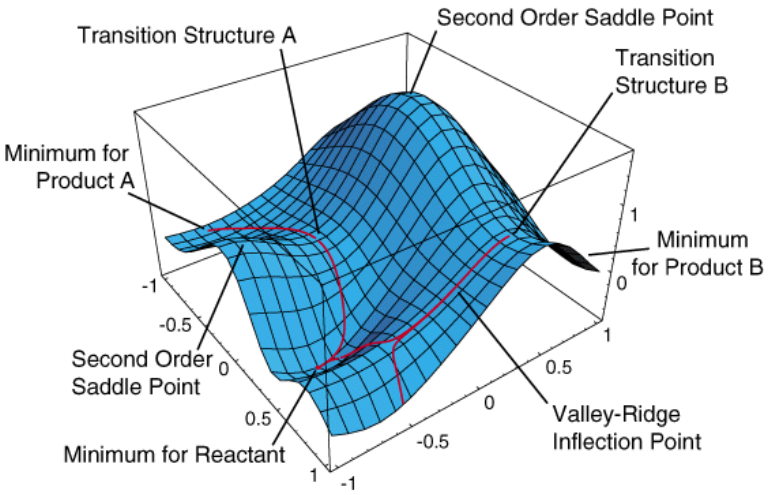

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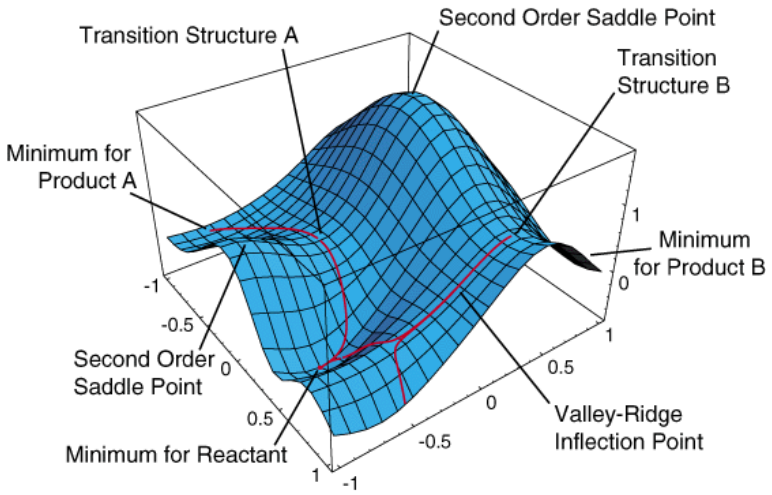

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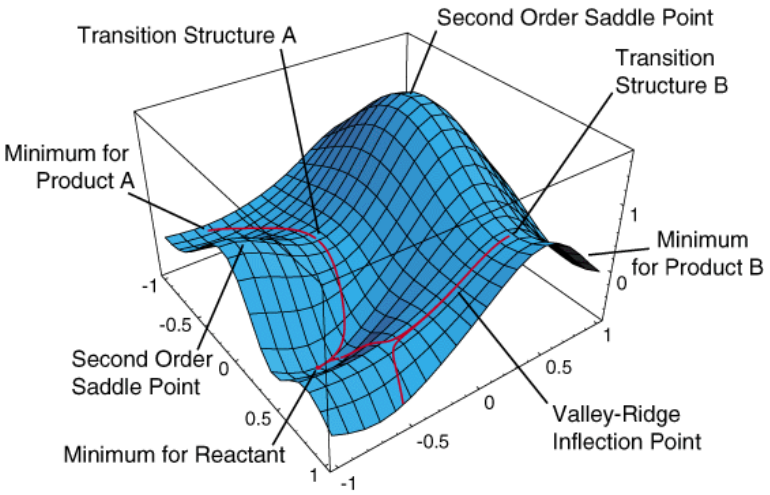

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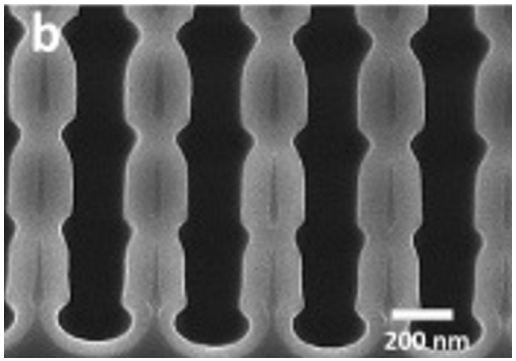
Wealth of information that could be difficult to obtain experimentally

The problem

Scale of interest

✓ Nanoscale ($\approx 10 - 100nm$)

✗ DFT ≈ 1 nm



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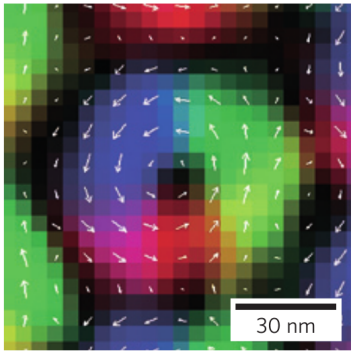

skyrmion in MnSi

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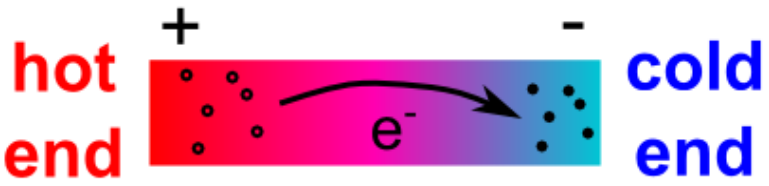
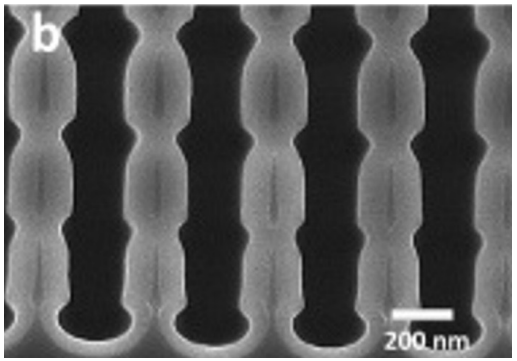


Thermoelectrics, polarons...



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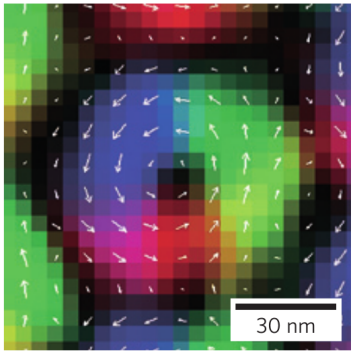

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✓ Charge diffusion

✓ Reaction yield

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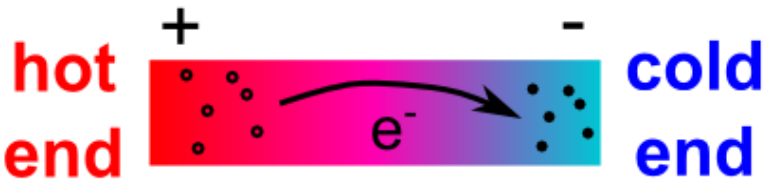

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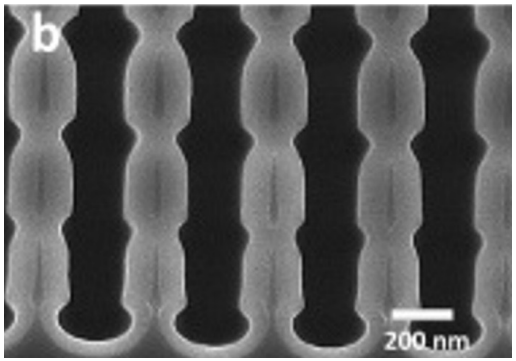
Disorder

✓ Domains

✓ Thermal

✓ Defects (polarons, impurities...)



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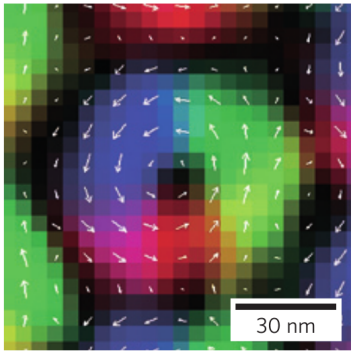

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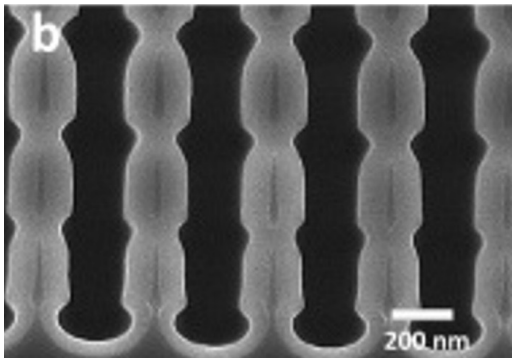

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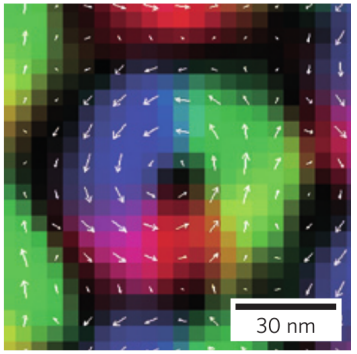

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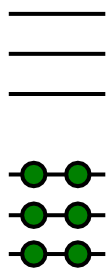
Ferroelectric domains in $BiFeO_3$

- Perturbations/disorder are key elements in experiments.
- Room temperature is fundamental for applications.

Are larger/faster FP simulations possible?

First principles simulations deal with all electrons in the system:



Number of electrons grows fast

Hamiltonian$\sim N^2$

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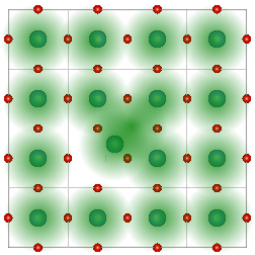

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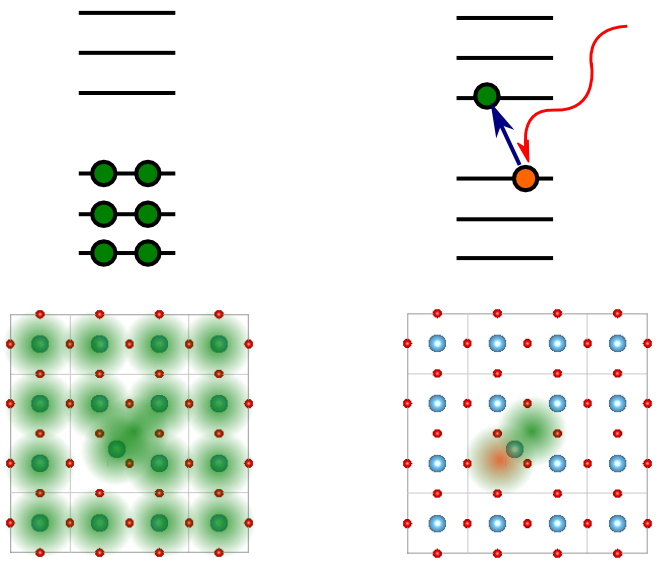

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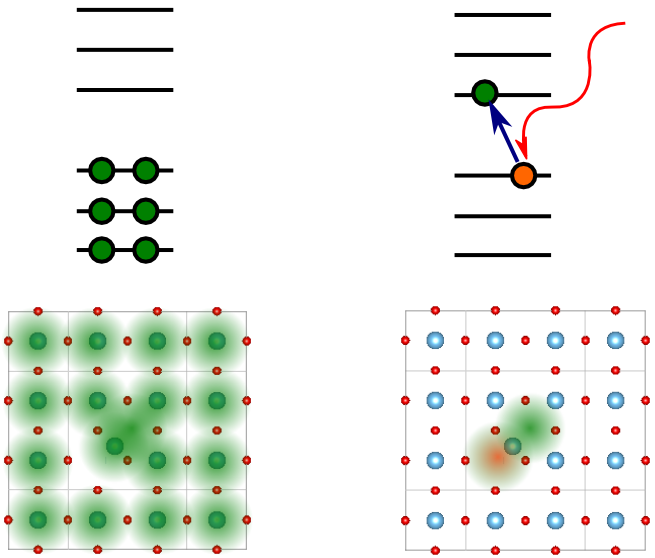

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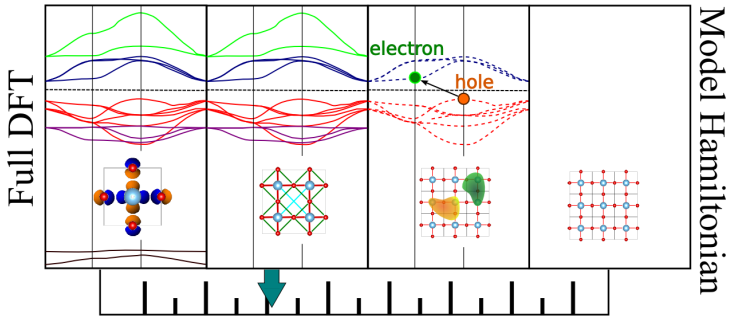

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Can we make it efficient?

Can we reliably parameterize it?

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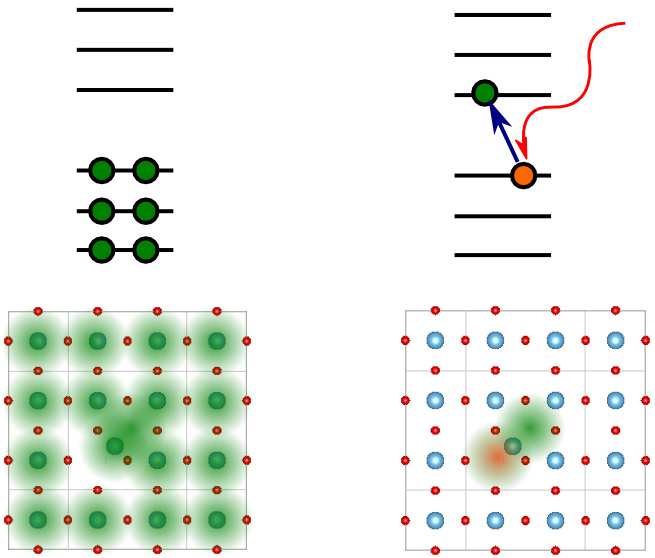

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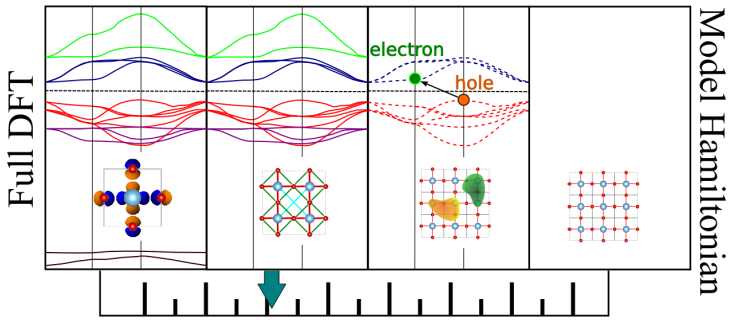

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Second-principles Density Functional Methods

Basic concepts

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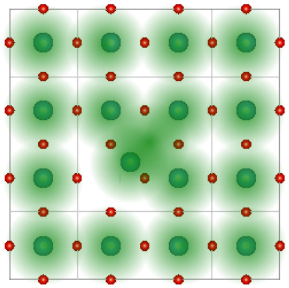

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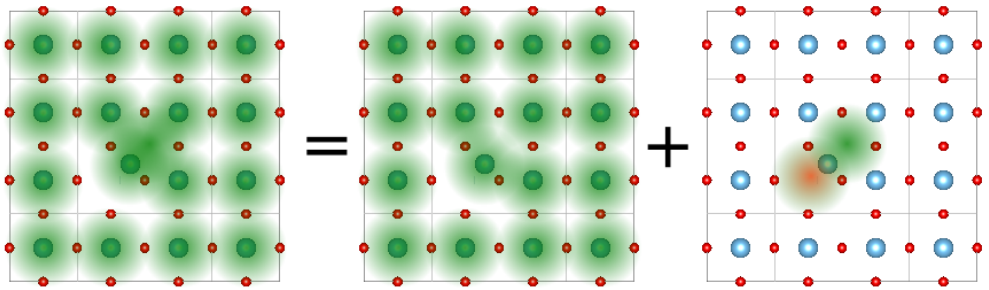

The total density is separated in reference and deformation densities:

$$n(\vec{r}) = n_0(\vec{r}) + \delta n(\vec{r})$$

n_0 = reference density
δn = deformation density

Approximating the DFT energy

Our starting point is the DFT energy

$$E_{\mathrm{DFT}} = \sum_{j\vec{k}} o_{j\vec{k}} \left\langle \psi_{j\vec{k}} \right| \hat{t} + v_{\mathrm{ext}} \left| \psi_{j\vec{k}} \right\rangle + \frac{1}{2} \iint \frac{n(\vec{r}) n'(\vec{r}')}{|\vec{r} - \vec{r}'|} d^3 r d^3 r' + E_{\mathrm{xc}}[n] + E_{\mathrm{nn}}$$

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The only difficulty is the exchange-correlation energy that we expand in terms of δn (see e. g. M. Elstner et al., *Phys. Rev. B*, 58, 7260 (1998)):

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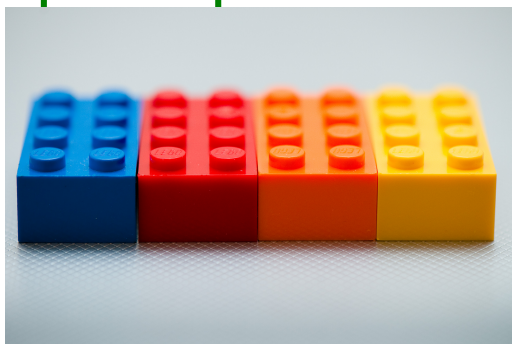
As in usual TB-DFT approximations, we cut at second-order

$$E_{\mathrm{DFT}} \approx E = E^{(0)} + E^{(1)} + E^{(2)}$$

However, we group the terms in a different way to TB-DFT.

Second-principles DFT approach

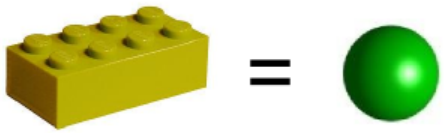


Material simulations

allow for various approaches

First principles methods are atomistic with flexible detailed bonding

FP or TB-DFT



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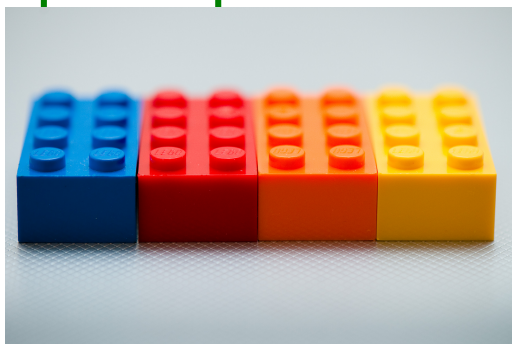

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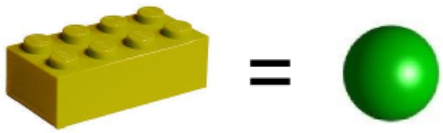

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$$E_{\mathrm{DFT}} \approx \underbrace{E^{(0)}}_{\text{atomic cores}} + \underbrace{E^{(1)}}_{\text{full 1e energy}} + \underbrace{E^{(2)}}_{\text{full 2e energy}} + \ldots$$

Atoms $\Longrightarrow$ FP $\Longrightarrow$ Materials

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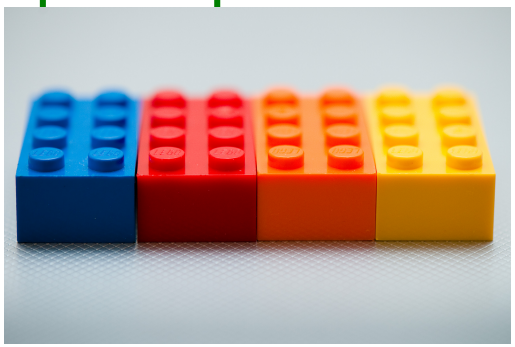

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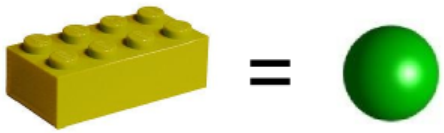

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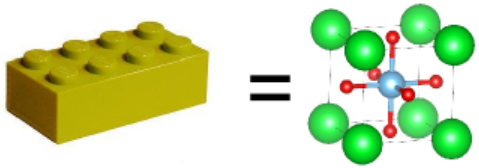

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$$E_{DFT} \approx \underbrace{E^{(0)}}_{\text{atomic cores}} + \underbrace{E^{(1)}}_{\text{full 1e energy}} + \underbrace{E^{(2)}}_{\text{full 2e energy}} + \dots$$

$$E_{DFT} \approx \underbrace{E^{(0)}}_{\text{lattice}} + \underbrace{E^{(1)} + E^{(2)} + \dots}_{\text{electron excitations}}$$

Atoms $\Longrightarrow$ FP $\Longrightarrow$ Materials $\Longrightarrow$ SP $\Longrightarrow$ Large-scale

Accurate properties do not require bond-breaking!

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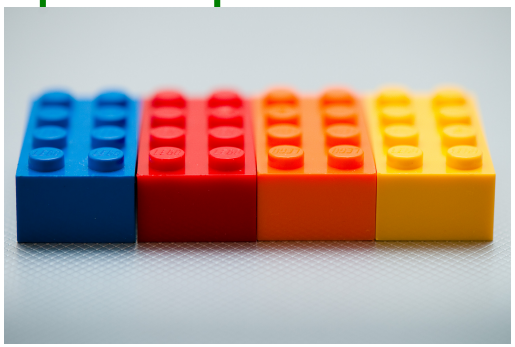

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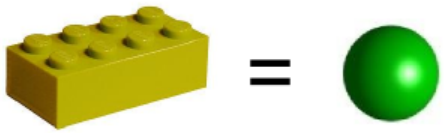

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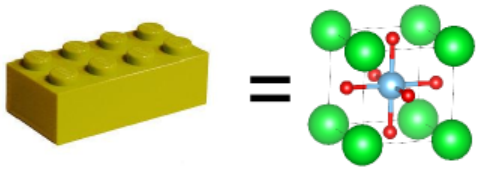

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Precise, small, material-adapted basis $\rightarrow$ Wannier-like functions

I. Souza et al., *Phys. Rev. B*, 65, 035109 (2001)

Energy terms: $E = E^{(0)} + E^{(1)} + E^{(2)}$ Reference

This term is the full DFT energy for the reference state:

$$E^{(0)} = \sum_{j\vec{k}} o^0_{j\vec{k}} \left\langle \psi^0_{j\vec{k}} \right| \hat{t} + v_{\text{ext}} \left| \psi^0_{j\vec{k}} \right\rangle + \frac{1}{2} \iint \frac{n_0(\vec{r}) n'_0(\vec{r}')}{|\vec{r} - \vec{r}'|} d^3 r d^3 r' + E_{\text{xc}}[n_0] + E_{nn}$$

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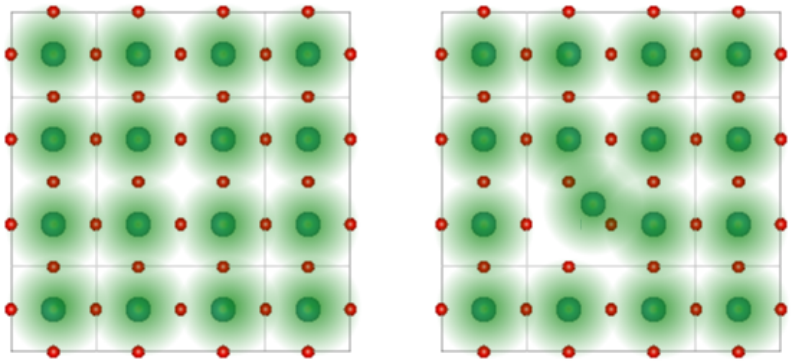
No approximations

At difference with usual TB-DFT this term is very large and contains most of the total energy. It can be made really accurate.



The reference state is defined for all geometries

$E_0(\eta, \{\vec{u}\})$ is the energy surface for the reference state

It can be represented by a high-quality model potential.

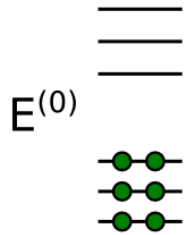
J. Wojdeł et al., *JPCM*, 25, 305401 (2013)

This lattice Hamiltonian is implemented in Multibinit!

Energy terms: $E = E^{(0)} + E^{(1)} + E^{(2)}$ One electron

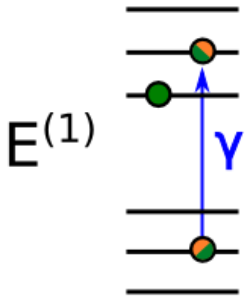
Reference

- Full DFT energy for n_0
- Force field
- Multibinit



$E^{(1)}$ contains differences in one-electron energies

$$E^{(1)} = \sum_{j\vec{k}} \left[o_{j\vec{k}} \left\langle \psi_{j\vec{k}} \right| \hat{h}_0 \left| \psi_{j\vec{k}} \right\rangle - o^0_{j\vec{k}} \left\langle \psi^0_{j\vec{k}} \right| \hat{h}_0 \left| \psi^0_{j\vec{k}} \right\rangle \right]$$



where $\hat{h}_0$ is the Kohn-Sham Hamiltonian for the reference density:

$$\hat{h}_0 = \hat{t} + v_{\text{ext}} + v_{\text{H}}(n_0) + v_{\text{xc}}[n_0]$$

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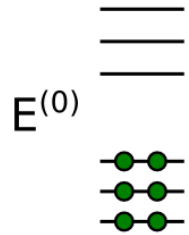

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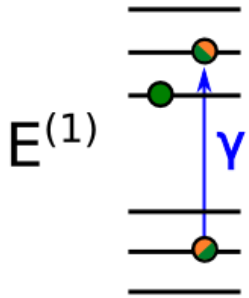
$$= \sum_{ab} D_{ab}\gamma_{ab} \quad \text{(Wannier basis, } \chi_a\text{)}$$

$$\delta n(\vec{r}) = \sum_{ab} D_{ab}\chi^*_a(\vec{r})\chi_b(\vec{r})$$



γ_{ab} takes the role of the hopping constant in TB schemes.

$$\gamma_{ab} = \int d^3 r \chi^*_a(\vec{r}) \hat{h}_0 \chi_b(\vec{r})$$

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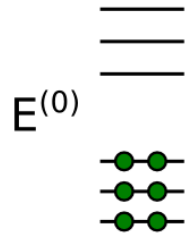

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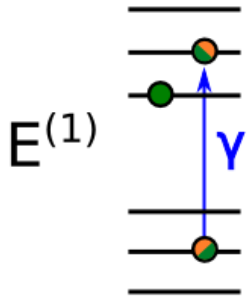

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Only depends on difference density!

Energy terms: $E = E^{(0)} + E^{(1)} + E^{(2)}$ Two electron

Reference

- Full DFT energy for n_0
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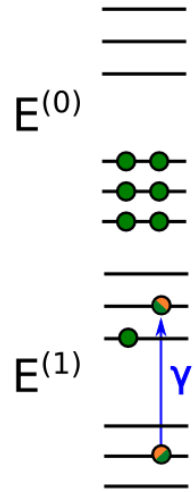
One-electron

- Depends only on difference density
- Tight-binding like



$E^{(2)}$ are interactions between 2 electrons ($E^{(3)}$ 3-electron, etc.):

$$E^{(2)} = \frac{1}{2} \int d^3r \int d^3r' g(\vec{r}, \vec{r}', s, s') \delta n(\vec{r}, s) \delta n(\vec{r}', s')$$

where g is a screened electron-electron interaction operator.

Energy terms: $E = E^{(0)} + E^{(1)} + E^{(2)}$ Two electron

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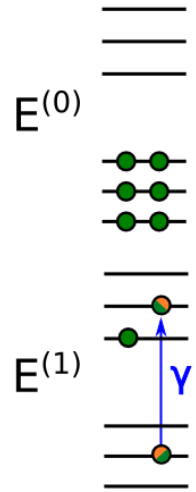

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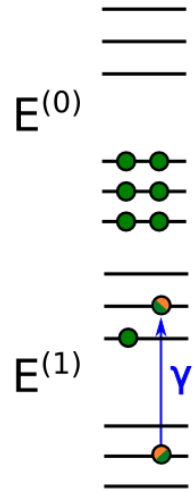

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Energy terms: $E = E^{(0)} + E^{(1)} + E^{(2)}$ Calculation

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One-electron

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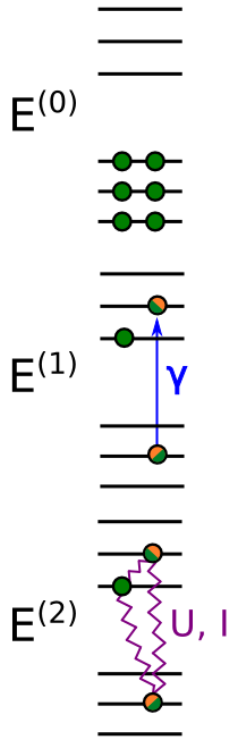
Two-electron

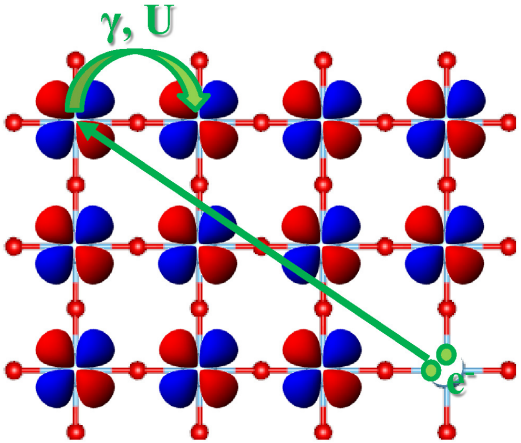
- Depends only on difference density
- Screened mean-field interactions



- Accurate
- Fast
- Valid for all kind of systems (magnetic, metallic, ...)

Electrostatics/Electron-lattice interactions

All interactions occur between localized objects:



At long-range (far-field regime) shape of source density is unimportant

Multipolar expansion

γ and U contain electrostatic (long-range) contributions

Hartree/electron-nucleus

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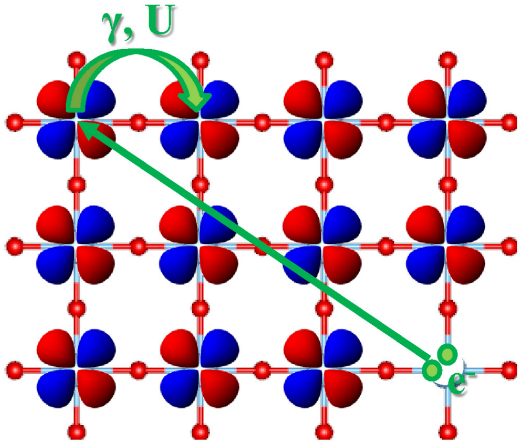

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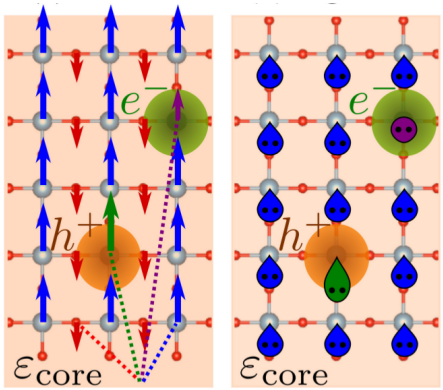
- Hybridizations $\rightarrow \langle \chi_a | \vec{r} | \chi_b \rangle$



Potential approximated by field of point charges and dipoles localized at the reference geometry

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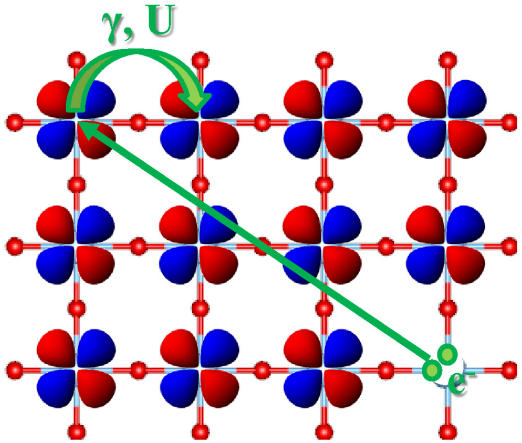

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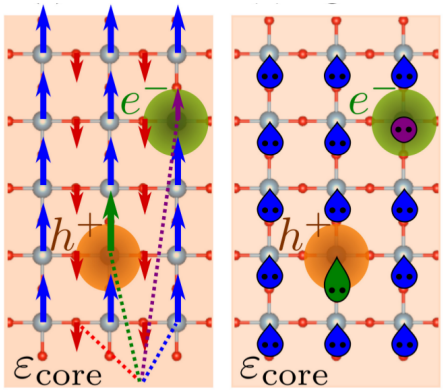
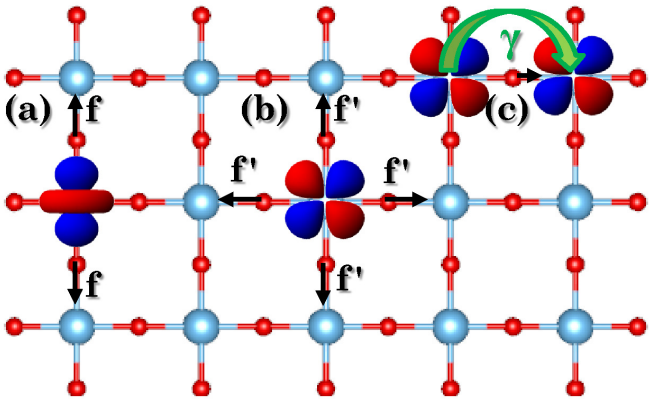


Potential approximated by field of point charges and dipoles localized at the reference geometry

Model parameters $\longrightarrow$ long and short range contributions.

Geometry dependence - Forces

Geometry is involved by expanding γ on the atomic positions:



$$\gamma^{\mathrm{sr}}_{\boldsymbol{ab}} = \gamma^{0}_{\boldsymbol{ab}} + \sum_{\boldsymbol{\lambda\upsilon}} \left[\vec{f}_{\boldsymbol{ab},\boldsymbol{\lambda\upsilon}} \cdot \delta\vec{r}_{\boldsymbol{\lambda\upsilon}} + \delta\vec{r}_{\boldsymbol{\lambda\upsilon}} \cdot \overleftrightarrow{g}_{\boldsymbol{ab},\boldsymbol{\lambda\upsilon}} \cdot \delta\vec{r}_{\boldsymbol{\lambda\upsilon}} + \ldots \right]$$

U,I should also depend on $\delta\vec{r} \rightarrow$ neglected

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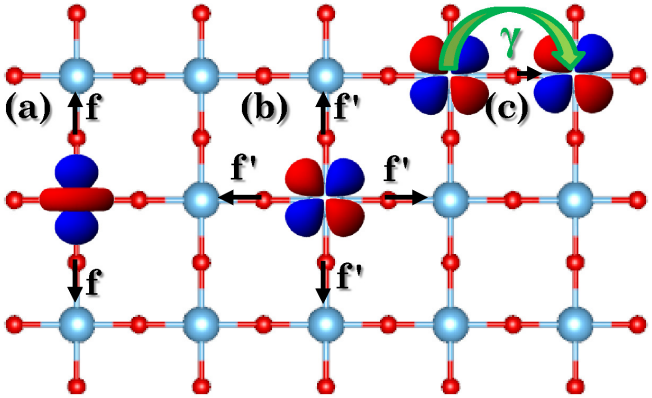

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U,I should also depend on $\delta\vec{r} \rightarrow$ neglected

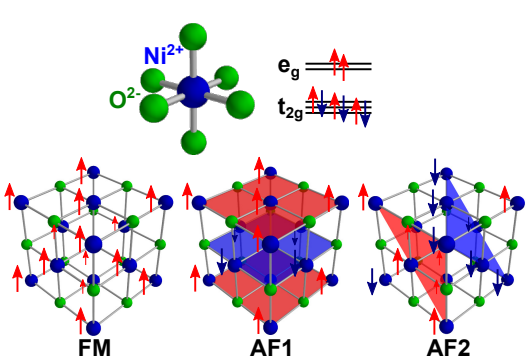
The forces are then obtained:

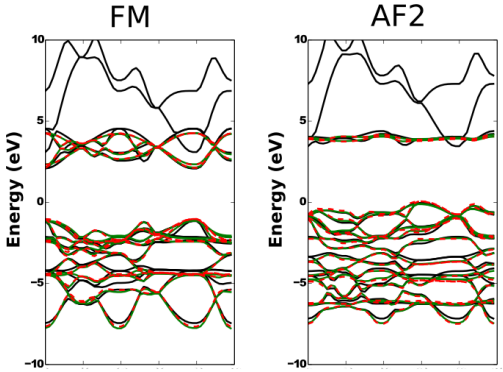
$$\vec{F}_\lambda = -\vec{\nabla}_\lambda E = -\vec{\nabla}_\lambda E^{(0)} - \sum_{ab} D_{ab} \vec{\nabla}_\lambda \gamma_{ab}.$$

Electronic contribution corrects the force field

Applications of SPDFT

NiO - Insulator with highly correlated electrons:





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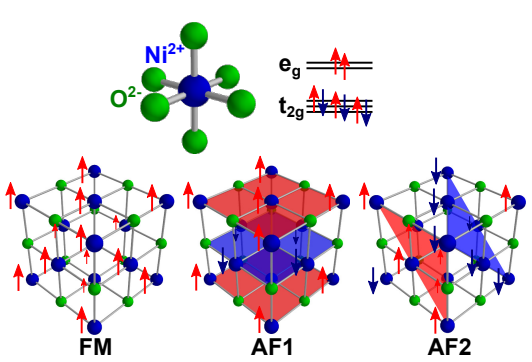

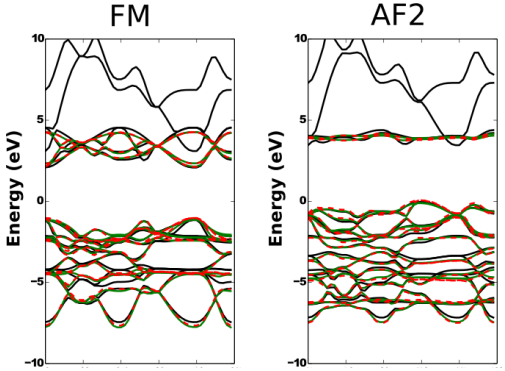

Magnetic Properties

Method	J_1 (meV)	J_2 (meV)
neutron	1.4	-19.0
LDA+U	2.6	-17.5
SP-$Ni(3d)$ + $O(2p)$	3.3	-17.6

Electronic/magnetic properties predicted at DFT level

2000 atoms single point 1 hour in 1 Desktop cpu (full diag)

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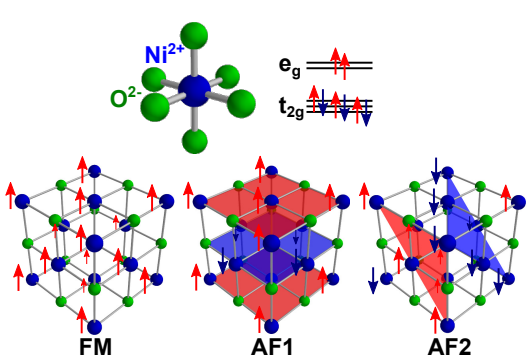

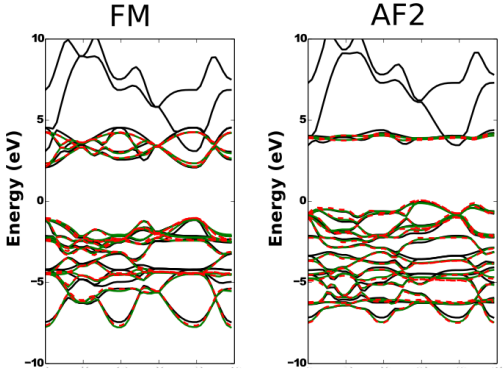

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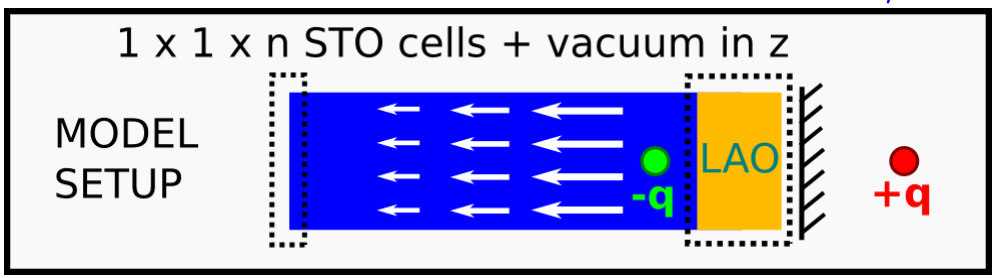
2DEG at $SrTiO_3/LaAlO_3$ Interface



FP $\rightarrow$ M. Stengel, *PRL*, 106, 136803 (2011)

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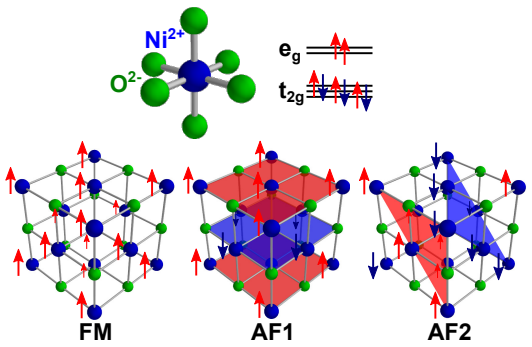

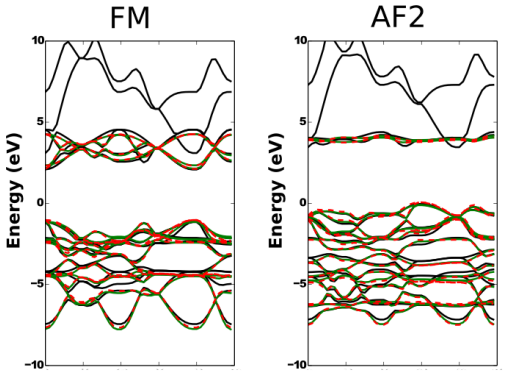

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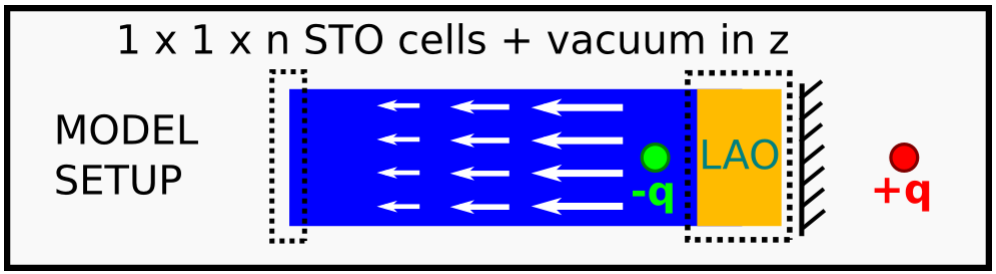
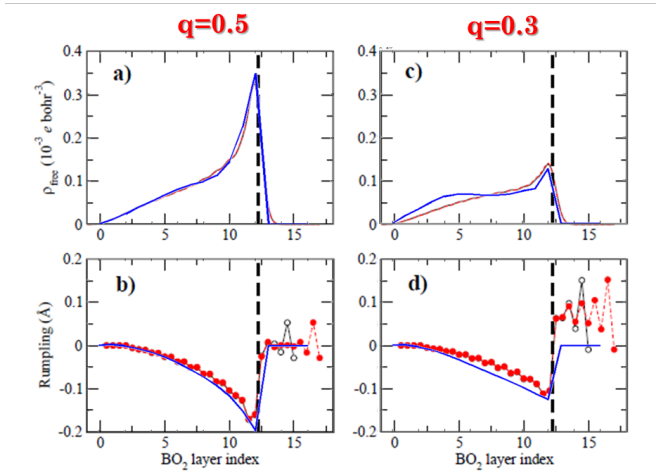


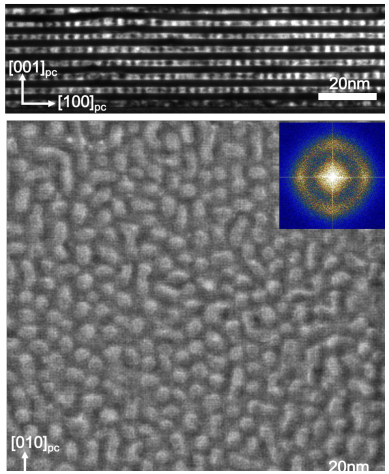


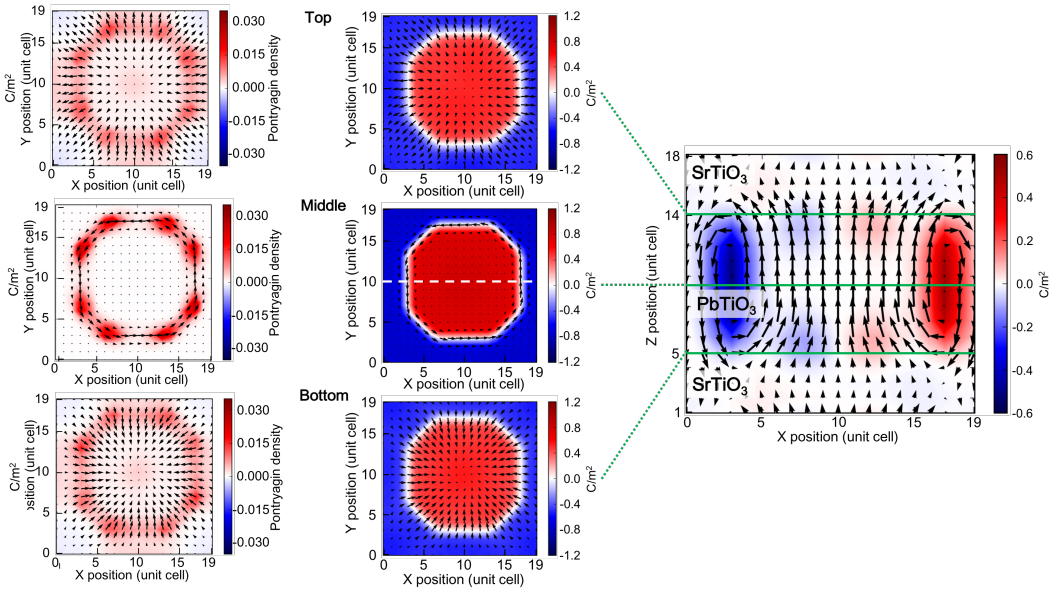
FP → M. Stengel, *PRL*, 106, 136803 (2011)

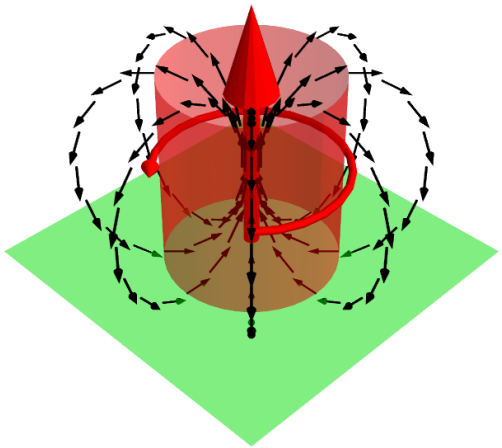
SPDFT captures doping and lattice screening!

Ferroelectric bubbles

SP can simulate ferroelectric bubble skyrmions in $PbTiO_3/SrTiO_3$



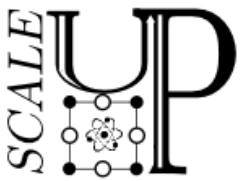




- $\sim$ 40000 atoms
- Tangential polarization to bubble
  Makes bubbles chiral
  Explains XCD signal
- Bubble shows non-trivial topology
  Planes $\rightarrow$ same topological charge

S. Das et al.,*Nature*, 568, 368 (2019)

The implementation of SP-DFT: SCALE-UP



P. Garcia-Fernandez, J. Wojdeł, J. Iñiguez and J. Junquera
Phys. Rev. B, **93**, 195137 (2016)

- Fully integrated electron+lattice models
- Single-points, Dynamics (isokinetic, Langevin), Montecarlo
- SCF with convergence acelerators
- TDDFT: Real-time density propagation, $\vec{E}(t)$ fields

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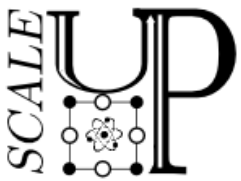

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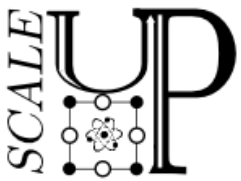

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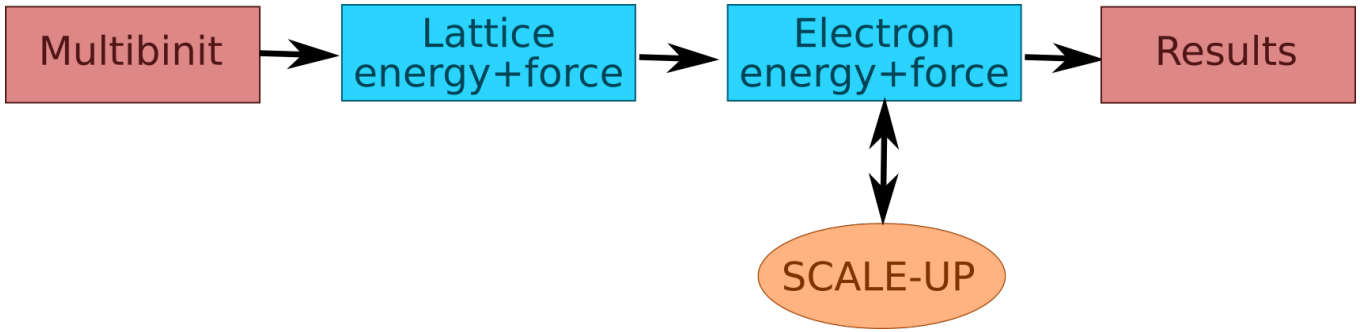
- Future: Spin-orbit, defects (surfaces, interfaces, impurities...)
- Future: Pure density-matrix implementation

The connection between SCALE-UP and MULTIBINIT

SCALE-UP can be run as a library by other codes



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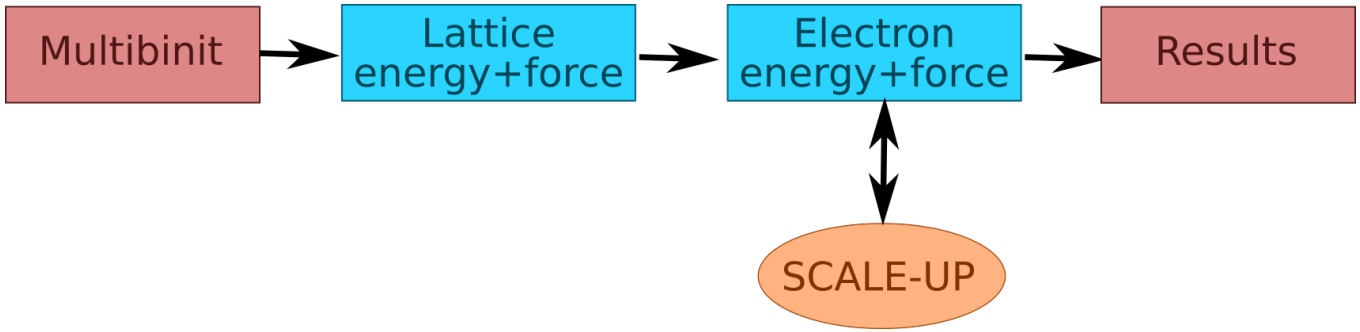

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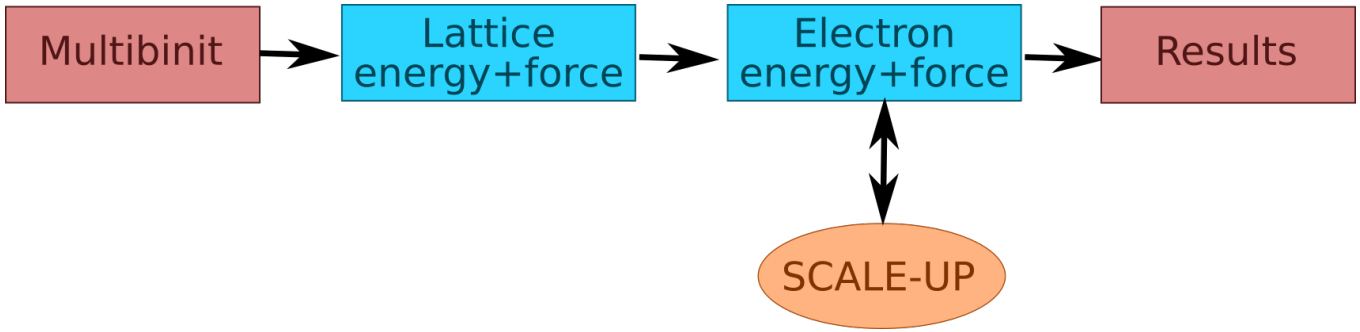

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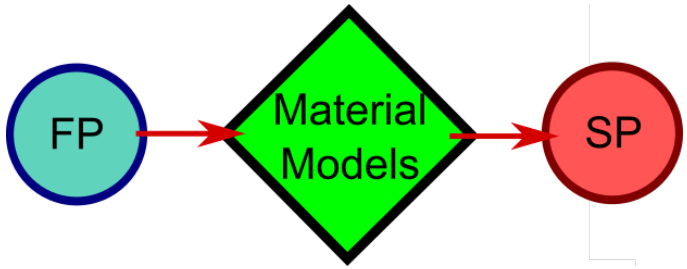
Distribution SCALE-UP $\longrightarrow$ Attendance to workshop

Rationale: Forming community, second-principles are not blackbox

Model construction

Second Principles depend on First Principles to create models



FP-SP Interface

Very delicate process, we need:

- Accuracy: SP model needs to reproduce FP
- Lightweight: SP model needs to be efficient

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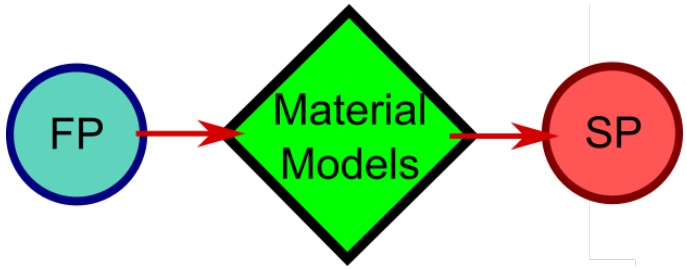

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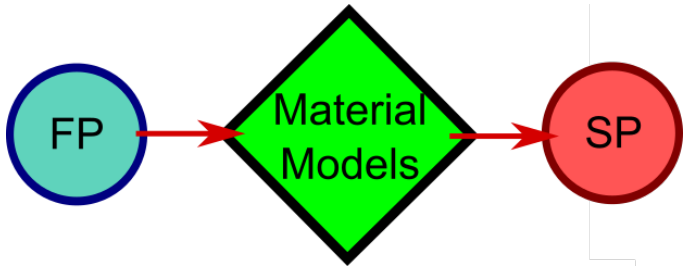

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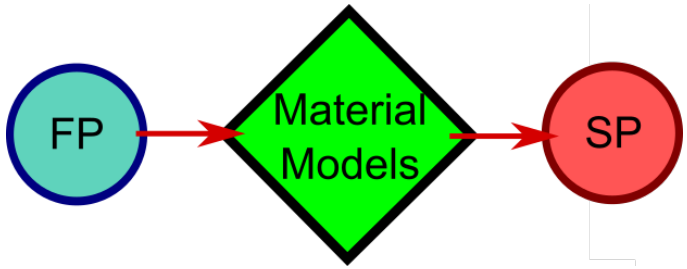

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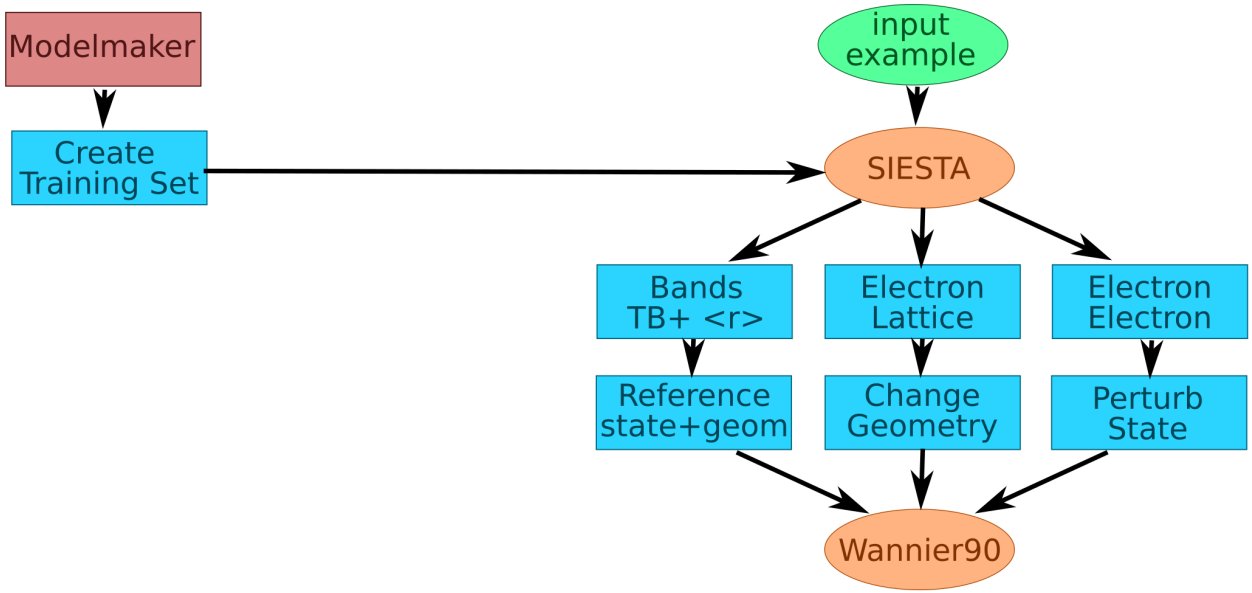
  3. Systematically improvable models
  4. Produces hopping, electron-lattice, electron-electron-parameters

Currently we have a SCALE-UP - SIESTA - WANNIER90 Interface

Modelmaker

The first step to create a model is to create a FP training set



$$h^s_{ab} = \gamma^{\text{RAG,sr}}_{ab} + \delta\gamma^{\text{el-lat,sr}}_{ab}(\{\vec{u}_\lambda\}) + \sum_{a'b'} \left(D^U_{a'b'} U_{aba'b'} \pm D^I_{a'b'} I_{aba'b'} \right) + \gamma^{\text{lr}}_{ab}$$

Model Hamiltonian $\longrightarrow$ capture FP physics

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- Electron-lattice terms: Change geometry on a supercell
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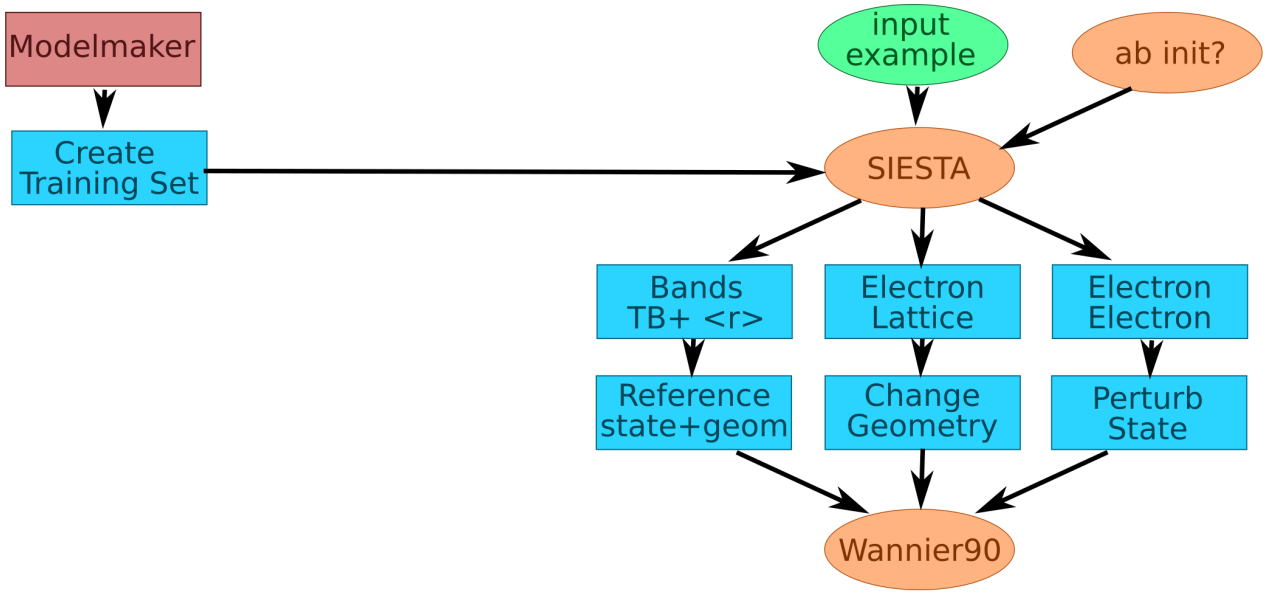

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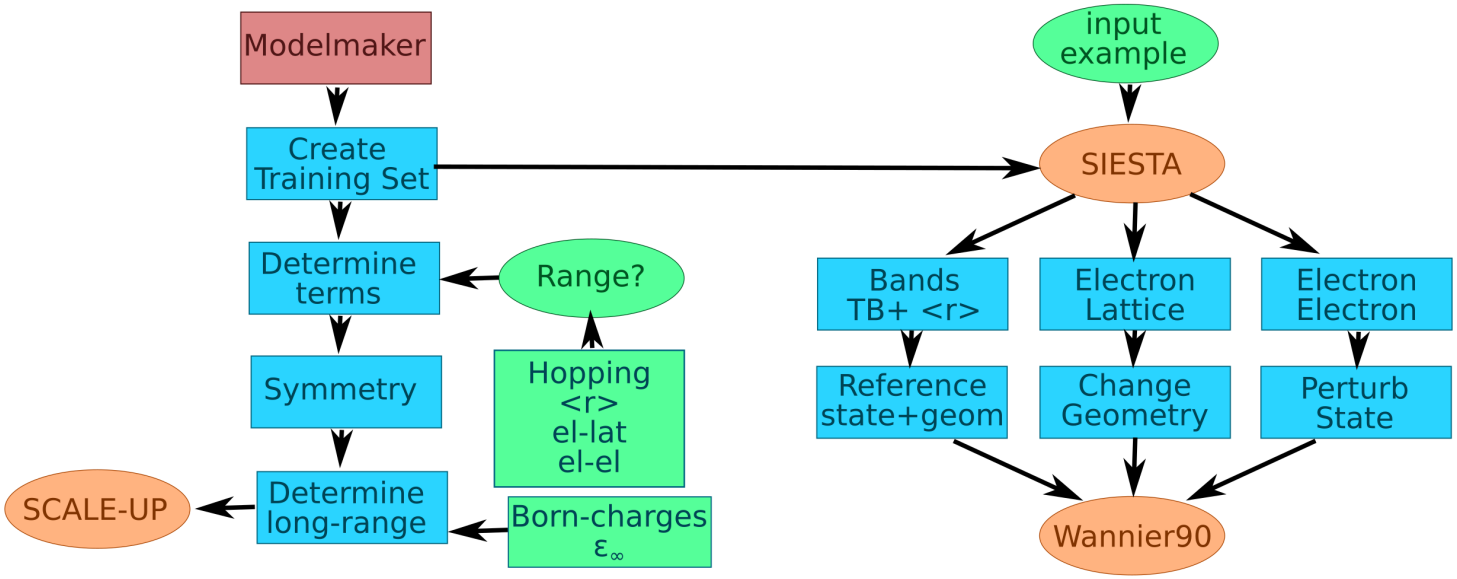
The SIESTA-WANNIER90 interface...

Javier Junquera

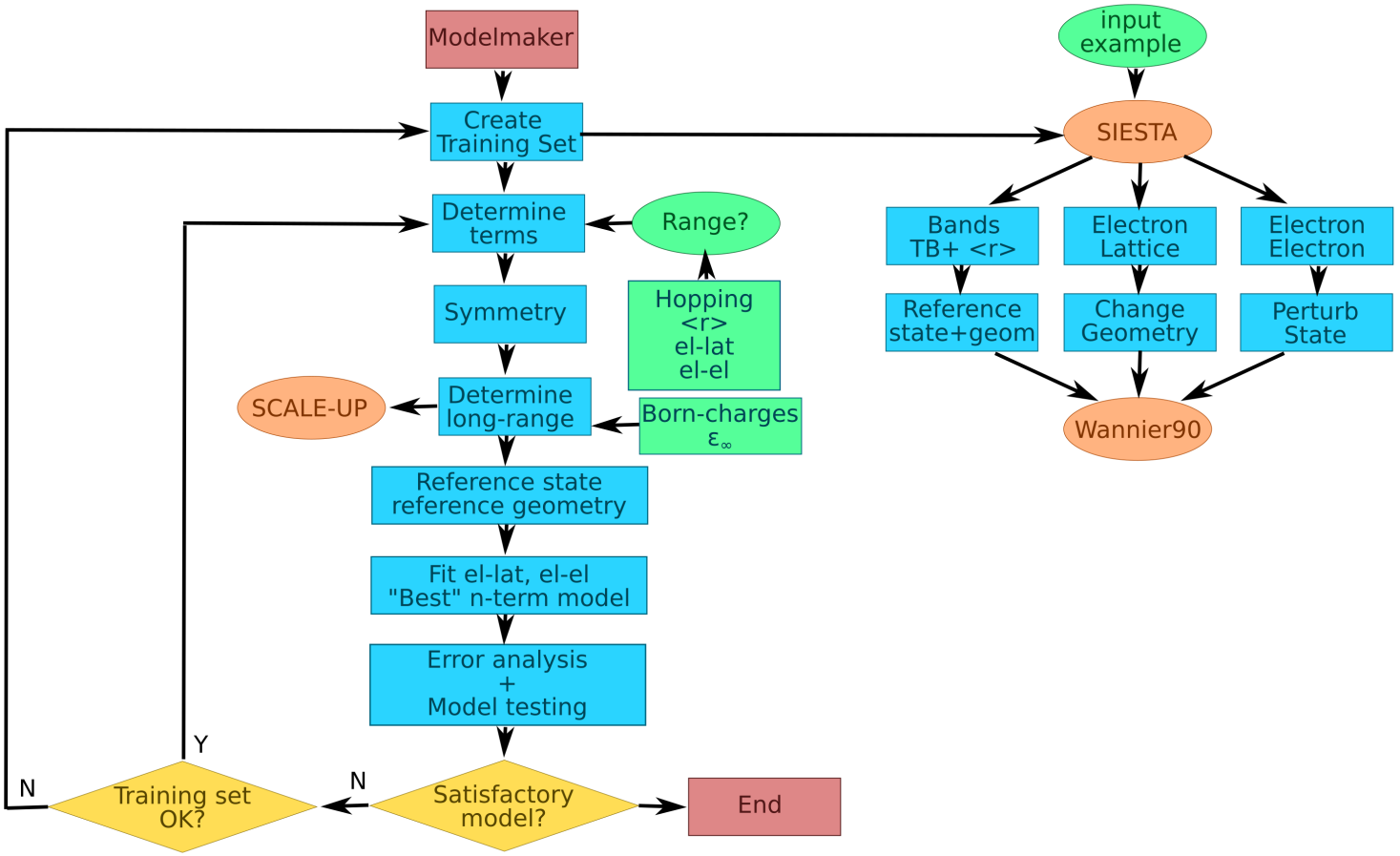
Modelmaker

Next we need to determine the terms that form the model



- The input is just 4 distances that determine the range of hopping, position, electron-lattice and electron-electron matrix elements.
- The code then filters the created terms using symmetry
- Finally, modelmaker calls SCALE-UP to determine the long-range corrections

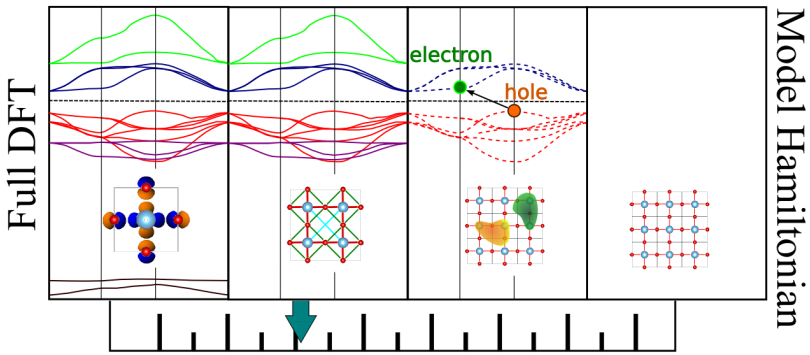
Modelmaker



- The code determines iteratively "best n-terms" models
- It provides with tools to check accuracy of the model and systematic error detection
- The user decides if model is good enough and how to improve

Summary

- Second-Principles DFT bridges the gap between first-principles and model Hamiltonians
- The goal is getting closer to "Computational experiments"
  P. Garcia-Fernandez et al.,*PRB*, 93, 195137 (2016)
  J. Wojdeł et al.,*JPCM*, 25, 305401 (2013)



- Current applications show the accuracy and versatility of method
  S. Das et al.,*Nature*, 568, 368 (2019)
- Their use requires still a lot of user input
  Strong interactions with FP codes

Thank you for your attention!