Foundations of Second-Principles DFT

How to run SCALE-UP

Pablo García-Fernández

Santander July 2017

Universidad de Cantabria

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Predict material properties using just fundamental constants

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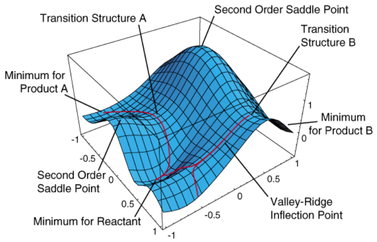
$$\hat{H}\Psi = E\Psi \quad \text{or DFT}$$



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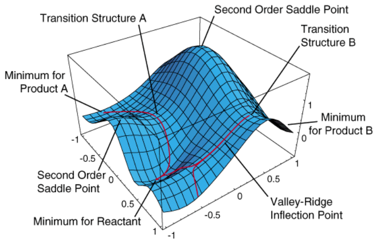

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- ✓ Accurate energies
- ✓ Equilibrium geometries

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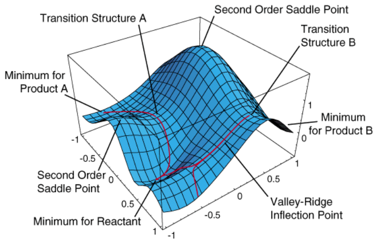

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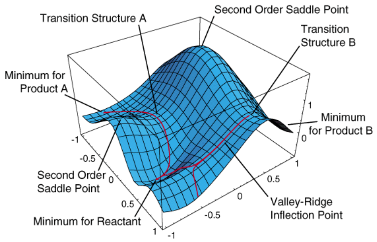

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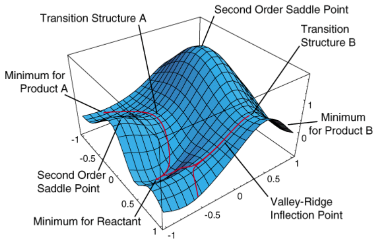

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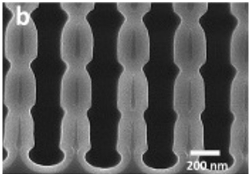
Wealth of information that could be difficult to obtain experimentally

The problem

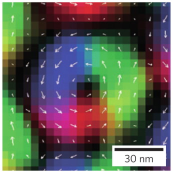
Scale of interest

✓ Nanoscale ($\approx 10 - 100 nm$)

✗ DFT $\approx$ 1 nm



Nanowires



skyrmion in MnSi

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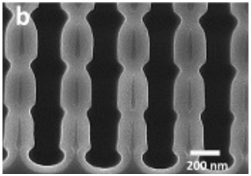

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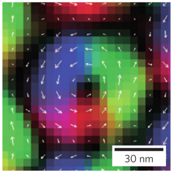

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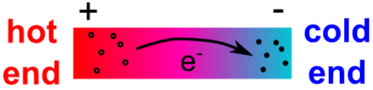

Thermoelectrics, polarons...

Non-equilibrium states

✓ Resistivity

✓ Charge diffusion

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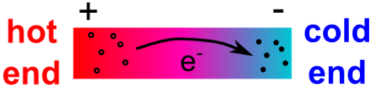

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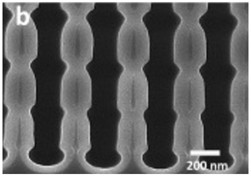
Disorder

✓ Domains

✓ Thermal

✓ Defects (polarons, impurities...)



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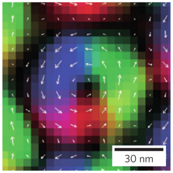

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Ferroelectric domains in $BiFeO_3$

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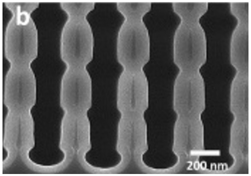

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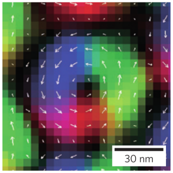

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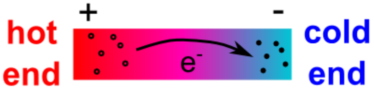

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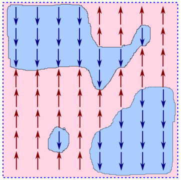
Ferroelectric domains in $BiFeO_3$

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

Model Hamiltonians

Simple Hamiltonians can be used to simulate large systems

$$E = -\sum_{\{i,j\}} J_{ij} \vec{S}_i \cdot \vec{S}_j$$



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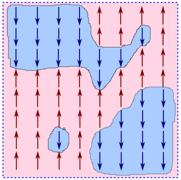

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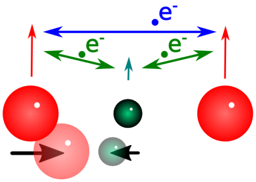

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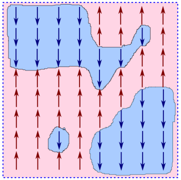

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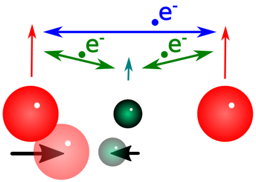
Electrons mediating magnetic interactions?

✗ Inaccurate

J_i constant with temperature?

Geometry dependence?



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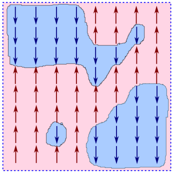
Predictive Model Hamiltonians $\rightarrow$ Based on fundamental equations
Well defined physics $\rightarrow$ Ab initio constants/good fitting procedures

Lattice Hamiltonians

Typically used in large-scale simulations of ferroelectrics

$$E = H_{\text{short}}(\{Q\}, \overleftrightarrow{\eta}) + H_{\text{elec}}(\{Q\})$$



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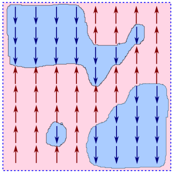

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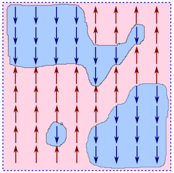

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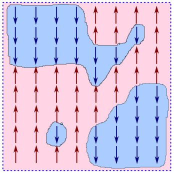

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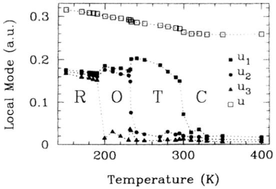
✓ Domains, superlattices...



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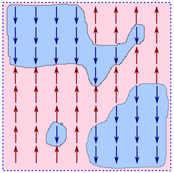

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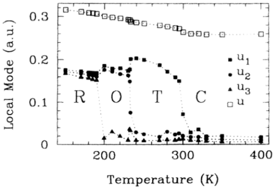
✗ Idealized degrees of freedom (not atomistic)

✗ No electrons!



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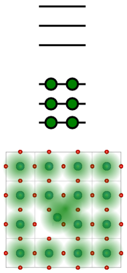
Are larger/faster FP simulations possible?

First principles simulations deal with all electrons in the system:



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Hamiltonian$\sim N^2$

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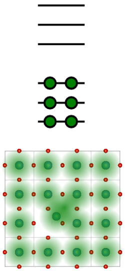

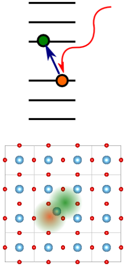

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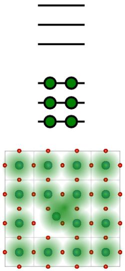

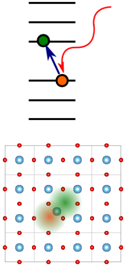

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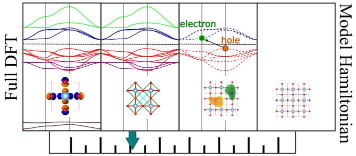

Can we select the level of fidelity of our calculations?

Can we make it efficient?

Can we reliably parameterize it?

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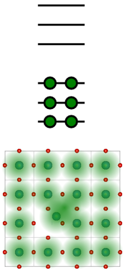

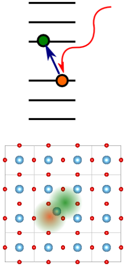

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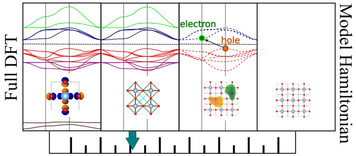

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Second-principles Density Functional Methods

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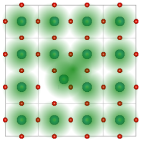

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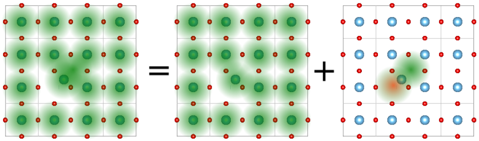

The total density is separated in reference and deformation densities:

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

n_0 = reference density
δn = deformation density

Approximating the DFT energy

Our starting point is the DFT energy

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

We want to write the energy in terms of the reference and deformation densities.

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(see e. g. M. Elstner et al.,*Phys. Rev. B*, 58, 7260 (1998)):

$$E_{\mathrm{xc}}[n] = E_{\mathrm{xc}}[n_0] + \int \left. \frac{\delta E_{\mathrm{xc}}}{\delta n(\vec{r})} \right|_{n_0} \delta n(\vec{r}) d^3r + \frac{1}{2} \iint \left. \frac{\delta^2 E_{\mathrm{xc}}}{\delta n(\vec{r})\delta n(\vec{r}')} \right|_{n_0} \delta n(\vec{r})\delta n(\vec{r}') d^3r d^3r' + \cdots$$

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(see e. g. M. Elstner et al.,*Phys. Rev. B*, 58, 7260 (1998)):

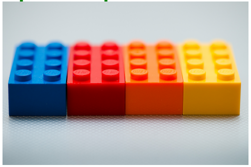
$$E_{\mathrm{xc}}[n] = E_{\mathrm{xc}}[n_0] + \int \left. \frac{\delta E_{\mathrm{xc}}}{\delta n(\vec{r})} \right|_{n_0} \delta n(\vec{r}) d^3r + \frac{1}{2} \iint \left. \frac{\delta^2 E_{\mathrm{xc}}}{\delta n(\vec{r}) \delta n(\vec{r}')} \right|_{n_0} \delta n(\vec{r}) \delta n(\vec{r}') d^3r d^3r' + \cdots$$

As usual in 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.

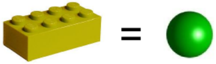
Second-principles DFT approach



Material simulations
allow for various approaches

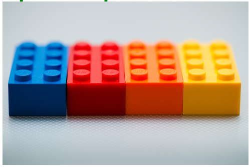
First principles methods are atomistic with flexible detailed bonding

FP or TB-DFT



Based on atoms

Second-principles DFT approach



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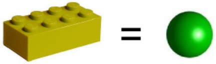

Based on atoms

$$E_{\mathrm{DFT}} \approx \underbrace{E_0}_{\text{atomic cores}} + \underbrace{E_1}_{\text{full 1e energy}} + \underbrace{E_2}_{\text{full 2e energy}} + \dots$$

Atoms $\Longrightarrow$ FP $\Longrightarrow$ Materials

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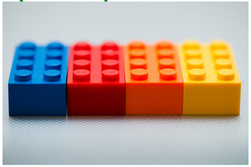

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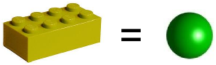

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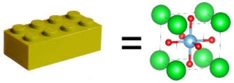


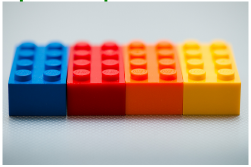
Based on materials

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

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

Accurate properties do not require bond-breaking!

Second-principles DFT approach



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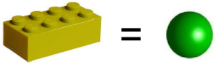

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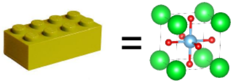

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Atoms $\Longrightarrow$ FP $\Longrightarrow$ Materials $\Longrightarrow$ SP $\Longrightarrow$ Large-scale

Accurate properties do not require bond-breaking!

Main approximation: Fixed bond topology
A perovskite stays a perovskite, no surface reactivity, etc.

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

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

$$E_0 = \sum_{j\vec{k}} o^0_{j\vec{k}} \left\langle \psi^0_{j\vec{k}} \right| \hat{t} + v_{\text{ext}} \left| \psi^0_{j\vec{k}} \right\rangle + \frac{1}{2} \iint \frac{n_0(\vec{r}) n_0'(\vec{r}')}{|\vec{r} - \vec{r}'|} d^3r d^3r' + E_{\text{xc}}[n_0] + E_{nn}$$

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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.

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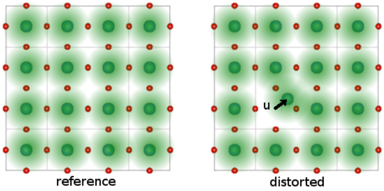
Energy is defined as difference from the reference geometry



The reference state is defined for all geometries

$E^{(0)}(\eta, \{\vec{u}\})$ is the energy surface for the reference state

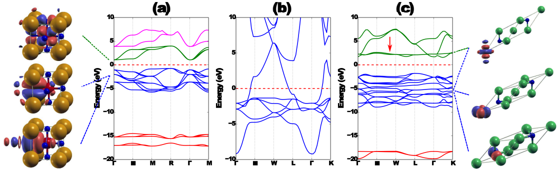
It can be represented by a high-quality model potential.

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

$E^{(1)} + E^{(2)}$ electron excitation basis

In order to express electron excitations we need basis:

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



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

$$|\chi_{\mathbf{a}}\rangle = \frac{V}{(2\pi)^3}\int_{\mathrm{BZ}} d\vec{k} e^{-i\vec{k}\cdot\vec{R}_A}\sum_{m}^{M} B_{a,m\vec{k}}|\psi_{m\vec{k}}\rangle$$

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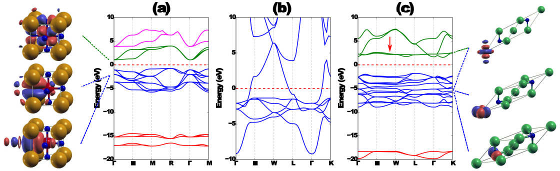

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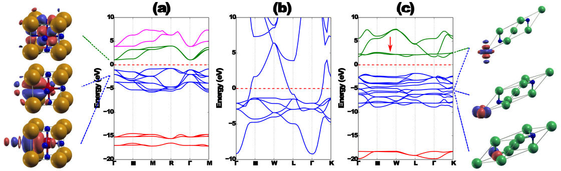

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2. allow disentanglement of electron degrees of freedom
   Reduced, meaningful physical models

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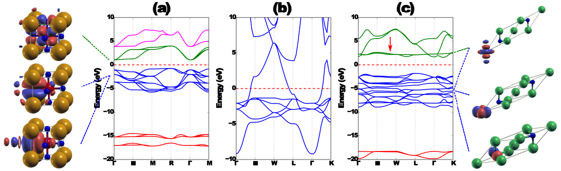

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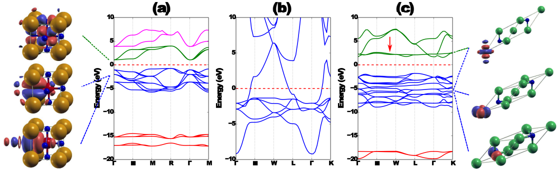

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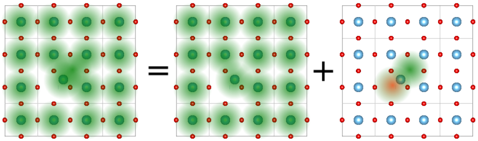
Wannier functions...

1. are localized and so are ideal to build range-limited models
2. allow disentanglement of electron degrees of freedom
   Reduced, meaningful physical models
3. are very accurate even with a small basis
4. are orthogonal
5. are defined for all geometries

$E^{(1)} + E^{(2)}$: Density matrix in Wannier basis

In SP-DFT there are three kinds of densities:



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

How is the density expressed in the WF basis?

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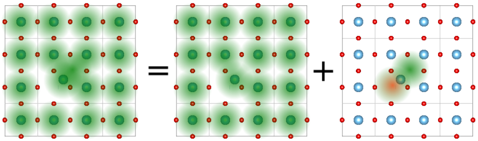

$$n(\vec{r}) = n_0(\vec{r}) + \delta n(\vec{r})$$

How is the density expressed in the WF basis?

Total	$n(\vec{r}) = \sum_{\mathbf{ab}} d_{\mathbf{ab}} \chi^*_{\mathbf{a}}(\vec{r}) \chi_{\mathbf{b}}(\vec{r})$	$d_{\mathbf{ab}} = \sum_{j\vec{k}} o_{j\vec{k}} e^{i\vec{k}(\vec{R}_B - \vec{R}_A)} c^*_{ja\vec{k}} c_{jb\vec{k}}$
Reference	$n_0(\vec{r}) = \sum_{\mathbf{ab}} d^0_{\mathbf{ab}} \chi^*_{\mathbf{a}}(\vec{r}) \chi_{\mathbf{b}}(\vec{r})$	$d^0_{ab} = \sum_{j\vec{k}} o^0_{j\vec{k}} e^{i\vec{k}(\vec{R}_B - \vec{R}_A)} \left(c^0_{ja\vec{k}}\right)^* c^0_{jb\vec{k}}$
Difference	$\delta n(\vec{r}) = \sum_{\mathbf{ab}} D_{\mathbf{ab}} \chi^*_{\mathbf{a}}(\vec{r}) \chi_{\mathbf{b}}(\vec{r})$	$D_{\mathbf{ab}} = d_{\mathbf{ab}} - d^0_{\mathbf{ab}}$

where $d_{\mathbf{ab}}$, $d^{(0)}_{\mathbf{ab}}$, $D_{\mathbf{ab}}$ are density matrixes (occupations)

$E^{(1)} + E^{(2)}$: Density matrix in Wannier basis

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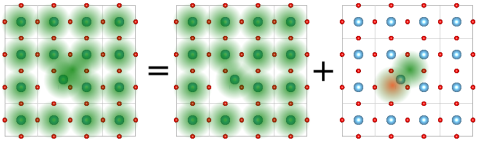

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where $d_{\mathbf{ab}}$, $d^{(0)}_{\mathbf{ab}}$, $D_{\mathbf{ab}}$ are density matrixes (occupations)

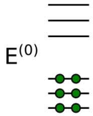
The main variable of the electron part is the Difference density

- $D_{\mathbf{ab}}$ is positive for excited electrons
- $D_{\mathbf{ab}}$ is negative for excited holes

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

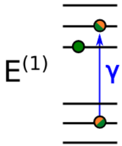
Reference

- Full DFT energy for n_0
- Force field



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

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



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

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

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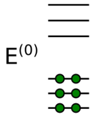

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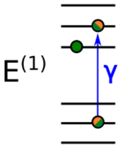
$$= \sum_{\mathbf{ab}} D_{\mathbf{ab}} \gamma_{\mathbf{ab}} \quad (\text{Wannier basis, } \chi_{\mathbf{a}})$$

$$\delta n(\vec{r}) = \sum_{\mathbf{ab}} D_{\mathbf{ab}} \chi^*_{\mathbf{a}}(\vec{r}) \chi_{\mathbf{b}}(\vec{r})$$



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$\gamma_{\mathbf{ab}} = \int d^3 r \chi^*_{\mathbf{a}}(\vec{r}) \hat{h}_0 \chi_{\mathbf{b}}(\vec{r})$ similar to hopping constant in TB

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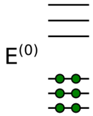

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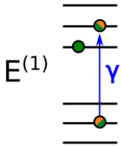

Only depends on difference density!

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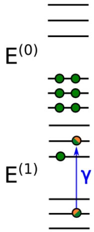
Reference

- Full DFT energy for n_0
- Force field

One-electron

- Depends only on difference density
- Tight-binding like



$E^{(2)}$ are interactions between 2 electrons (E_3 three-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.

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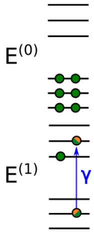

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$E^{(2)}$ only depends the difference density

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

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One-electron

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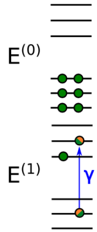

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$$
\begin{aligned}
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& + \left[D^{\uparrow}_{\mathbf{ab}} - D^{\downarrow}_{\mathbf{ab}}\right]\left[D^{\uparrow}_{\mathbf{a'b'}} - D^{\downarrow}_{\mathbf{a'b'}}\right]I_{\mathbf{aba'b'}}\Big\}
\end{aligned}
$$

where g is a screened electron-electron interaction operator.

$E^{(2)}$ only depends the difference density

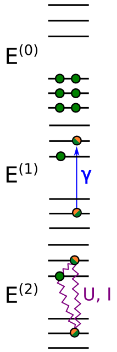
The term in I depends on the spin polarization

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

Reference
- Full DFT energy for n_0
- Force field

One-electron
- Depends only on difference density
- Tight-binding like

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



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

Efficiency of solid-state methods

Hartree-Fock

$$\hat{h} = \hat{t} + v[\{\psi_i\}](\vec{r})$$

Non-local exchange

N^5 scaling

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$$\hat{h} = \hat{t} + v[n](\vec{r})$$

Local xc functionals

N^3 scaling

Efficiency of solid-state methods

Hartree-Fock	DFT	SP-DFT
$\hat{h} = \hat{t} + v[\{\psi_i\}](\vec{r})$	$\hat{h} = \hat{t} + v[n](\vec{r})$	$h_{\mathbf{ab}} = \gamma_{\mathbf{ab}} + \sum_{\mathbf{a'b'}} D_{\mathbf{a'b'}} U_{\mathbf{aba'b'}}$
Non-local exchange	Local xc functionals	Equations non-local
N^5 scaling	N^3 scaling	N^4 scaling??

SP-DFT is not intrinsically fast unless models are range-limited

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SP-DFT

$$h_{\mathbf{ab}} = \gamma_{\mathbf{ab}} + \sum_{\mathbf{a'b'}} D_{\mathbf{a'b'}} U_{\mathbf{aba'b'}}$$

Equations non-local

N^4 scaling??

SP-DFT is not intrinsically fast unless models are range-limited

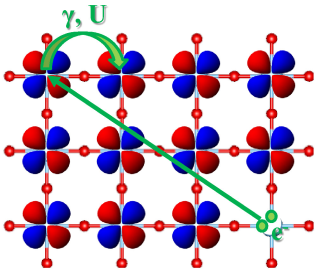
1. Accuracy is given by including many terms ($\gamma_{\mathbf{ab}}$, $U_{\mathbf{aba'b'}}$)
2. Speed is dictated by having range-limited interactions

Model building is a key step in the use of SP-DFT!

Electrostatics allows reducing number of parameters in simulation

Electrostatics

All interactions in the model are between localized objects:



Atomic displacement $\rightarrow$ local dipole

γ and U contain electrostatic (long-range) contributions

Hartree/electron-nucleus

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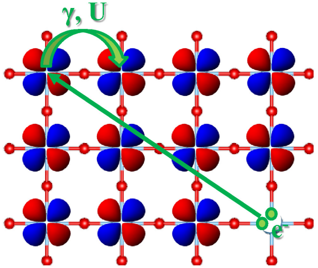

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Multipolar expansion!

We approximate the full charge density by a field of point charges and dipoles localized at the reference geometry

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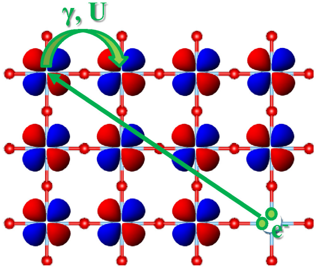

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We approximate the full charge density by a field of point charges and dipoles localized at the reference geometry

Model parameters are separated in long and short contributions.

The short-range part $\rightarrow$ quickly converging to zero
It also creates forces - long-range electron-lattice interaction

What can SP-DFT realistically do?

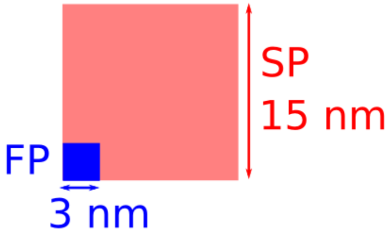
Good test $\rightarrow$ impurity concentration

Semiconductors $\rightarrow$ 10^{13}cm^{-3}-10^{18}cm^{-3}

What can SP-DFT realistically do?

Good test → impurity concentration

Semiconductors → 10^{13}cm^{-3}-10^{18}cm^{-3}



In FP size limit ∼700 atoms (3nm/cube side)

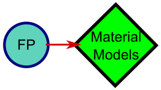
FP impurity density $\approx 3.7 \cdot 10^{19} cm^{-3}$

SP detailed model → 30000 atoms (12nm/cube side)
SP simple model → 60000 atoms (15nm/cube side)

SP impurity density $\approx 3.0 \cdot 10^{17} cm^{-3}$

The SP-DFT goal

Our goal is getting closer to the ideal of computer experiments...



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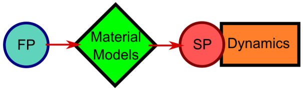

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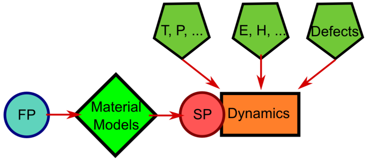

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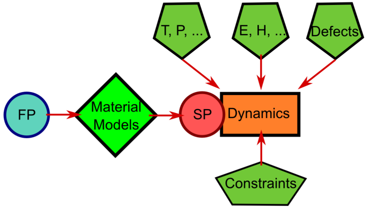

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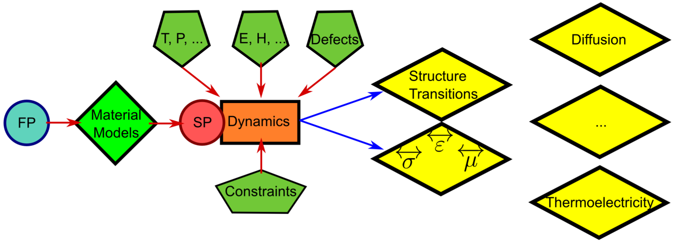

... with truly polyvalent models and the ability to understand and predict a wide variety of properties!

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- Parallelization: Hybrid scheme, MPI+OPENMP
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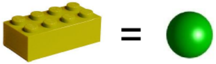
- Future: Spin-orbit, defects

Running Scale-Up

Running is analogous to ab initio code with a key difference

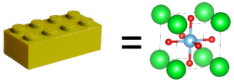
First-principles



Based on atoms

Pseudopotential/basis files

Second-principles



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Material xml file

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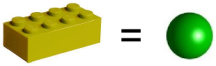

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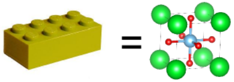

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There are two main ways of running Scale-Up :

1. Calling the code from the terminal, requires input file

```
$ more input.fdf
System_name a_linear_chain
Parameter_file linear_chain.xml
%block Supercell
1 1 2
%endblock Supercell
```

```
$ scaleup.x < input.fdf > output
```

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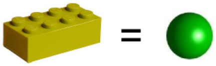

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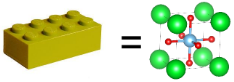

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```
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```

2. Using the python scaleup module in myscript.py

```
$ python myscript.py
```

Running SCALE-UP from the terminal

SCALE-UP uses one environment variable:

SCALEUPHOME $\rightarrow$ points to SCALE-UP root directory

```
$ ls $SCALEUPHOME
bin dev docs interface scripts src test
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   ```
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   ```
4. src contains the source code
5. test contains examples and tutorials
6. interface contains the python scaleup.py module
7. dev contains some tools used during development

The input file: fdf format

SCALE-UP input is given in text file using SIESTA's fdf format...

- Keywords: A keyword string and a value (integer, real, word)

  Geometrymode single-point
  MaximumSCFiter 100

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  ```
  %block band_path
  2
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  ```

- Physical: A keyword string with value (real) and the unit.

  ```
  Temperature 100.0 kelvin
  ```

Extra auxiliary files may be required to input initial geometries, etc.

Basic input and running

SCALE-UP can be run as a serial program:

```
$ $SCALEUPHOME/bin/scaleup_serial.x < input.fdf > output
```

or as a full parallel MPI Fortran program:

```
$ mpirun -n 4 $SCALEUPHOME/bin/scaleup.x < input.fdf > output
```

Your current compilation is serial only!

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Let's have a look at the main keywords!

Minimum input

A minimum Scale-Up input contains:

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1. No_lattice Do not read parameters for $E^{(0)}$ from file
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3. mode
    - single_point (default)
    - monte_carlo
    - dynamics
    - optimization

Basic keywords

Some useful keywords are:

- geometry_restart filename read initial geometry
- read_orbocc filename read initial orbital occupation
- print_bands int print bands file
- print_forces int output the forces to file
- temperate real kelvin The temperature
- %block static_electric_field The electric field

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SCF keywords:

- Magnetic Carry out spin-polarized calculations
- MaximumSCFiter int Maximum SCF iterations before aborting
- SCFthreshold real convergence criterion for SCF (def. 10^{-3})
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Check all the keywords in the manual!

```
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```

The visualization tools

The visualization tools are in $SCALEUPHOME/scripts:

scaleup_utils.py

```
$ python $SCALEUPHOME/scripts/scaleup_utils.py -help
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```

1. Geometry takes .REF and .restart files and outputs xcrysden
   - Allows representing absolute geometry
   - Allows representing reference geometry and distortion
2. DOS takes a .ener file and plots the DOS
3. bands takes a .bands file and plots the bands
4. current takes current/polarization file and plots conductivity
5. other we are expanding it to take:
   - .elec files to plot magnetization, electrostatic potentials, atomic dipoles in xcrysden format
   - .orbocc files to plot electron and hole densities in space

The python interface

SCALE-UP contains a python interface to the Fortran code

Check: $PYTHONPATH="$SCALEUPHOME/interface"

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To use it simple load the py_scaleup module:

```
$ more my_python_scup_script.py
"""
A simple python script using the py_scaleup module
"""
import py_scaleup as sclup
# Initialize an scale-up simulation of SrTiO3 in a 2x2x1 supercell
# with just lattice degrees of freedom
 scaleupsim=sclup.scaleup('srtio3_full_lat.xml',supercell=[2,2,1],
lattice=True,electrons=False)
# Change an atom position and calculate energy
scaleupsim.displacements[9]=0.1
energy=scaleupsim.get_energy()
```

At present functionality is limited but will quickly be expanded

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Check definition of the scaleup class!

```
$ vi $SCALEUPHOME/interface/py_scaleup.py
```