SCALE-UP: An implementation of Second-Principles DFT

Pablo García-Fernández, Jorge Íñiguez and Javier Junquera

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Collaborators

Cantabria University

Javier Junquera

Luxembourg Institute of Science and Technology

Jorge Iñiguez

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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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\[ \hat{H}\psi = E\psi \] or DFT
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Wealth of information that could be difficult to obtain experimentally
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Pablo García-Fernández

[Email]: garciapa@unican.es

SCALE-UP
The problem

Scale of interest

✔ Nanoscale ($\approx 10 - 100 \text{nm}$)

✗ DFT $\approx 1 \text{ nm}$
The problem

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Non-equilibrium states
 ✓ Resistivity
 ✓ Charge diffusion
 ✓ Reaction yield

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Disorder

✓ Domains
✓ Thermal
✓ Defects (polarons, impurities...)

Thermoelectrics, polarons...

Nanowires

Ferroelectric domains in BiFeO$_3$
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- Nanoscale ($\approx 10 - 100\, \text{nm}$)
- DFT $\approx 1\, \text{nm}$

Disorder

- Domains
- Thermal
- Defects (polarons, impurities...)

Non-equilibrium states

- Resistivity
- Charge diffusion
- Reaction yield

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 $\sim N^2$
Diagonalization $\sim N^3$
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Response to perturbations usually involves a few active electron/holes

Can we select the level of fidelity of our calculations?
Can we make it efficient?
Can we reliably parameterize it?

Second-principles Density Functional Methods

Pablo García-Fernández  garciapa@unican.es

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Basic concepts

We want to separate the active electrons that participate in physical properties from all others.
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Suppose an insulator doped with electrons or holes:
Basic concepts

We want to separate the active electrons that participate in physical properties from all others.

Suppose an insulator doped with electrons or holes:

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

The total density is separated in reference and deformation densities:

$$n_0 = \text{reference density}$$

$$\delta n = \text{deformation density}$$
Approximating the DFT energy

Our starting point is the DFT energy

\[
E_{\text{DFT}} = \sum_{jk} o_{jk} \langle \psi_{jk} | \hat{t} + v_{\text{ext}} | \psi_{jk} \rangle + \frac{1}{2} \int \int \frac{n(\vec{r}) n'(\vec{r}')}{|\vec{r} - \vec{r}'|} d^3 r d^3 r' + E_{\text{xc}}[n] + E_{\text{nn}}
\]

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

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

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

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

\[ E_{\text{xc}}[n] = E_{\text{xc}}[n_0] + \int \frac{\delta E_{\text{xc}}}{\delta n(\vec{r})} \left|_{n_0} \right. \delta n(\vec{r}) d^3r + \frac{1}{2} \int \int \frac{\delta^2 E_{\text{xc}}}{\delta n(\vec{r}) \delta n(\vec{r}')} \left|_{n_0} \right. \delta n(\vec{r}) \delta n(\vec{r}') d^3r d^3r' + \cdots \]
Approximating the DFT energy

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

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

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

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
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Based on atoms

\[ E_{\text{DFT}} \approx E^{(0)} + E^{(1)} + E^{(2)} + \ldots \]

Atoms \(\Rightarrow\) FP \(\Rightarrow\) Materials

Atoms + flexible detailed bonding = Materials

E_{\text{DFT}} \approx E^{(0)} + E^{(1)} + E^{(2)} + \ldots

Second-principles DFT approach

Material simulations allow for various approaches

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Based on atoms

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

Based on materials

\[ E_{\text{DFT}} \approx E^{(0)}_{\text{lattice}} + E^{(1)} + E^{(2)} + \ldots \]

Atoms \(\rightarrow\) FP \(\rightarrow\) Materials \(\rightarrow\) SP \(\rightarrow\) Large-scale

Accurate properties do not require bond-breaking!

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\[ E_{\text{DFT}} \approx E^{(0)} + E^{(1)} + E^{(2)} + \ldots \]

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Atoms \implies \text{FP} \implies \text{Materials} \implies \text{SP} \implies \text{Large-scale}

Accurate properties do not require bond-breaking!

Precise, small, material-adapted basis \implies \text{Wannier-like functions}

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

Reference

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

$$E^{(0)} = \sum_{j\bar{k}} o_{j\bar{k}}^0 \langle \psi_{j\bar{k}}^0 | \hat{t} + v_{\text{ext}} | \psi_{j\bar{k}}^0 \rangle + \frac{1}{2} \int \int \frac{n_0(\bar{r})n_0^\prime(\bar{r}^\prime)}{|\bar{r} - \bar{r}^\prime|} d^3rd^3r^\prime + E_{\text{xc}}[n_0] + E_{nn}$$
Energy terms: \( E = E^{(0)} + E^{(1)} + E^{(2)} \)

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

\[
E^{(0)} = \sum_{j\bar{k}} c_{jk}^0 \langle \psi_{j\bar{k}}^0 \rangle \hat{t} + v_{\text{ext}} \psi_{j\bar{k}}^0 \rangle + \frac{1}{2} \int \int \frac{n_0(\vec{r})n_0'(\vec{r}')}{|\vec{r} - \vec{r}'|} d^3r d^3r' + E_{\text{xc}}[n_0] + E_{nn}
\]

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.

\( E_0(\eta, \{\vec{u}\}) \) 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)
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

\[
E^{(1)} = \sum_{jk} \left[ o_{jk} \langle \psi_{j\bar{k}} | \hat{h}_0 | \psi_{j\bar{k}} \rangle - o_{j\bar{k}}^{0} \langle \psi_{j\bar{k}}^{0} | \hat{h}_0 | \psi_{j\bar{k}}^{0} \rangle \right]
\]

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

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

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

\[
E^{(1)} = \sum_{j,k} \left( o_{j,k} \langle \psi_j | \hat{h}_0 | \psi_{j,k} \rangle - o_{j,k}^0 \langle \psi_j^0 | \hat{h}_0 | \psi_{j,k}^0 \rangle \right)
\]

\[
= \sum_{a,b} D_{ab} \gamma_{ab} \quad \text{(Wannier basis, } \chi_a \text{)}
\]

\[
\delta n(\vec{r}) = \sum_{a,b} D_{ab} \chi_a^*(\vec{r}) \chi_b(\vec{r})
\]

\( \gamma_{ab} \) takes the role of the hopping constant in TB schemes.

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\gamma_{ab} = \int d^3r \chi_a^*(\vec{r}) \hat{h}_0 \chi_b(\vec{r})
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Only depends on difference density!
Energy terms: $E = E^{(0)} + E^{(1)} + E^{(2)}$  

**Two electron**

- Full DFT energy for $n_0$
- Force field
- Multibinit

**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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\[
\begin{align*}
\left\{ \left[ D_{ab}^{\uparrow} + D_{ab}^{\downarrow} \right] \left[ D_{a'b'}^{\uparrow} + D_{a'b'}^{\downarrow} \right] U_{aba'b'} \\
+ \left[ D_{ab}^{\uparrow} - D_{ab}^{\downarrow} \right] \left[ D_{a'b'}^{\uparrow} - D_{a'b'}^{\downarrow} \right] I_{aba'b'} \right\}
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Energy terms: \( E = E^{(0)} + E^{(1)} + E^{(2)} \) Calculation

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**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, ...)
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
Electrostatics/Electron-lattice interactions

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Hartree/electron-nucleus

Local dipoles:

- Atomic displacement $\rightarrow Z^*$
- 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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- Hybridizations \( \to \langle \chi_a | \vec{r} | \chi_b \rangle \)

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

Model parameters \( \to \) long and short range contributions.

Pablo García-Fernández  
garciapa@unican.es
Geometry dependence - Forces

Geometry is involved by expanding $\gamma$ on the atomic positions:

$$\gamma_{ab}^{sr} = \gamma_{ab}^{0} + \sum_{\lambda\nu} \left[ \vec{f}_{ab,\lambda\nu} \cdot \delta\vec{r}_{\lambda\nu} + \delta\vec{r}_{\lambda\nu} \cdot \vec{g}_{ab,\lambda\nu} \cdot \delta\vec{r}_{\lambda\nu} + \ldots \right]$$

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

Magnetic Properties

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SPDFT captures doping and lattice screening!

Electronic/magnetic properties predicted at DFT level

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

2DEG at SrTiO$_3$/LaAlO$_3$ Interface

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

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SCALE-UP
Ferroelectric bubbles

SP can simulate ferroelectric bubble skyrmions in PbTiO$_3$/SrTiO$_3$

- ~ 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

- Fully integrated electron+lattice models
- Single-points, Dynamics (isokinetic, Langevin), Montecarlo
- SCF with convergence accelerators
- TDDFT: Real-time density propagation, $\tilde{E}(t)$ fields
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- SCALE-UP python module: Running and analysis
- Model building suite: MODELMAKER

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- Model building suite: MODELMAKER
- 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

- It can provide $E^{(0)}$, $E^{(1)}+E^{(2)}$ or $E^{(0)}+E^{(1)}+E^{(2)}$

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The connection between SCALE-UP and MULTIBININIT

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

- It can provide $E^{(0)}$, $E^{(1)}+E^{(2)}$ or $E^{(0)}+E^{(1)}+E^{(2)}$
- It has three main calls
  1. `scaleup_init`
  2. `calculate_energy`
  3. `calculate_forces`
The connection between SCALE-UP and MULTIBINIT

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- It can provide $E^{(0)}$, $E^{(1)}+E^{(2)}$ or $E^{(0)}+E^{(1)}+E^{(2)}$
- It has three main calls
  1. scaleup_init
  2. calculate_energy
  3. calculate_forces

Distribution SCALE-UP $\rightarrow$ Attendance to workshop
Rationale: Forming community, second-principles are not blackbox
Model construction

**Second Principles** depend on **First Principles** to create models

FP → Material Models → SP

FP-SP Interface

Very delicate process, we need:

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

Currently we have a Scale-Up - Siesta - Wannier90 Interface

Pablo García-Fernández
garciapa@unican.es
SCALE-UP
Model construction

Second Principles depend on First Principles to create models

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  1. Automated
  2. Few and clean input parameters
  3. Systematically improvable models

FP-SP Interface

Pablo García-Fernández
garciapa@unican.es
SCALE-UP
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  2. Few and clean input parameters
  3. Systematically improvable models
  4. Produces hopping, electron-lattice, electron-electron-parameters

Currently we have a SCALE-UP - SIESTA - WANNIER90 Interface
The first step to create a model is to create a FP training set.

Model Hamiltonian $\rightarrow$ capture FP physics

- **Bands:** Wannier Hamiltonian
- **Electron-lattice terms:** Change geometry on a supercell
- **Electron-electron terms:** Controlled perturbation of electrons

$$h_{ab}^s = \gamma_{ab}^{\text{RAG, sr}} + \delta\gamma_{ab}^{\text{el-lat, sr}} (\{\tilde{\mu}_\lambda\}) + \sum_{a'b'} (D_{a'b'}^U U_{aba'b'} \pm D_{a'b'}^l l_{aba'b'}) + \gamma_{ab}^{\text{lr}}$$
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\[
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\]
The Siesta-Wannier90 interface...

Javier Junquera

Pablo García-Fernández garciapa@unican.es SCALE-UP
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.
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!