Spin dynamics from second principles in Multibinit

Xu He, Nicole Helbig, Jordan Bieder, Philippe Ghosez, Matthieu Verstraete, and Eric Bousquet

University of Liège

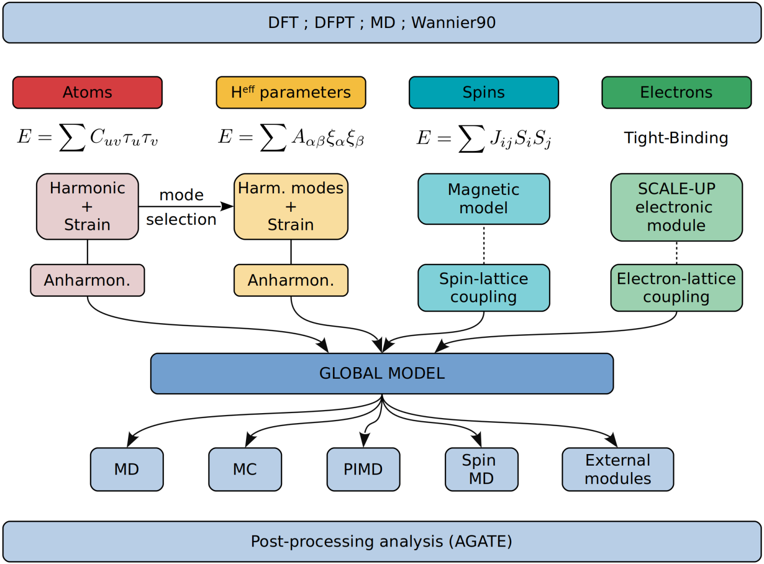
x.he@uliege.be

Abidev 2019

LIÈGE université

Outline

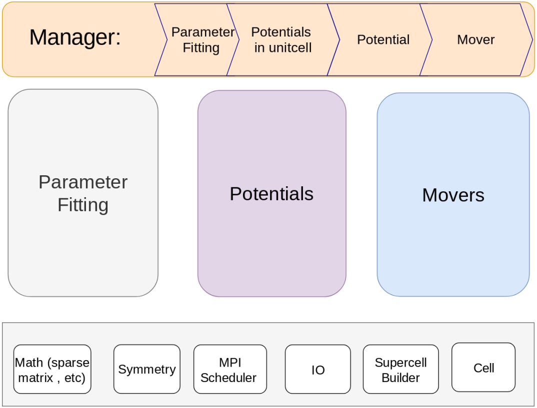
The MULTIBINIT project



Data structure in Multibinit

Physics problem:

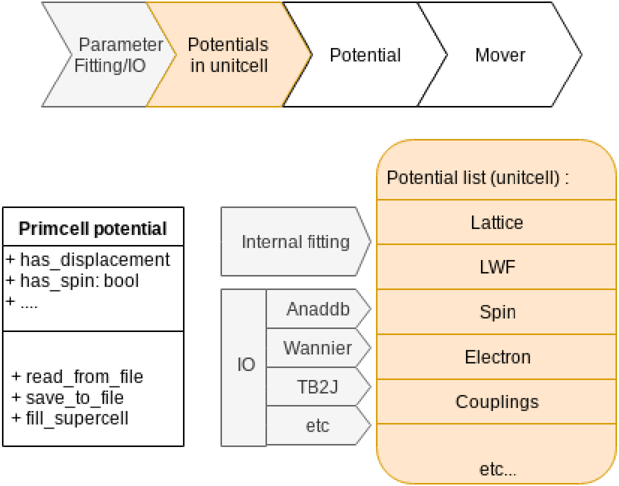
- Potential: $H = \sum H_c$
- Motion equation: $\frac{dx}{dt} = f(\frac{dH}{dx})$



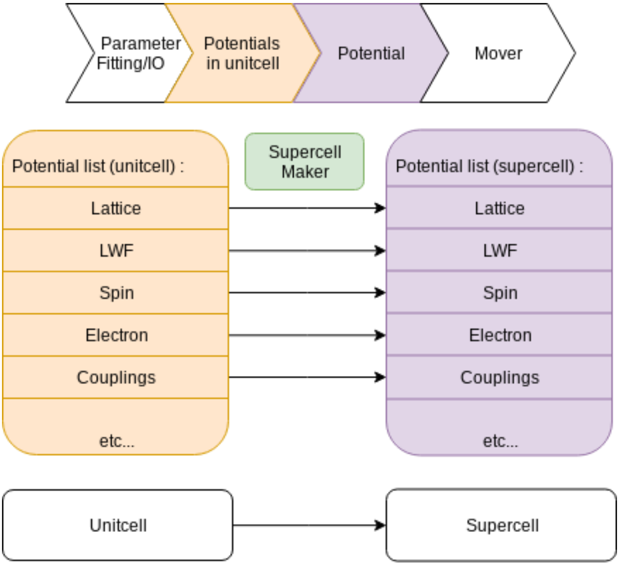
Design principles:

- Separate physics from implementation details.
- Each part can work as a black box.
- Keep consistency between potentials.

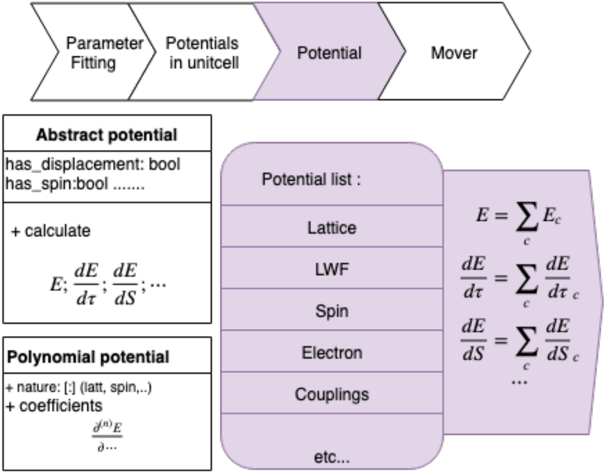
Unitcell potentials



Building supercell

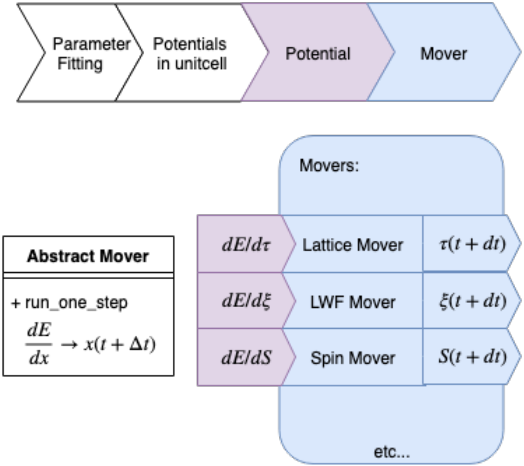


Potentials



Parameter Fitting
Potentials in unitcell
Potential
Mover
Abstract potential
has_displacement: bool
has_spin:bool ....
+ calculate
$E; \frac{dE}{d\tau}; \frac{dE}{dS}; \cdots$
Polynomial potential
+ nature: [:] (latt, spin,..)
+ coefficients
$\frac{\partial^{(n)} E}{\partial \cdots}$
Potential list :
Lattice
LWF
Spin
Electron
Couplings
etc...
$E = \sum_c E_c$
$\frac{dE}{d\tau} = \sum_c \frac{dE}{d\tau}_c$
$\frac{dE}{dS} = \sum_c \frac{dE}{dS}_c$
...

Movers



Parameter Fitting
Potentials in unitcell
Potential
Mover
Abstract Mover
+ run_one_step
$\frac{dE}{dx} \rightarrow x(t+\Delta t)$
Movers:
$dE/d\tau$
Lattice Mover
$\tau(t+dt)$
$dE/d\xi$
LWF Mover
$\xi(t+dt)$
dE/dS
Spin Mover
$S(t+dt)$
etc...

Spin part: hamiltonian

Hamiltonian

$$E = E_{exc} + E_{sia} + E_{DM} + E_{dd} + E_{ext}\,.$$

Effective magnetic field

The effective magnetic field (the spin torque) of $\vec{S}_i$:

$$\vec{H}_i = -\frac{1}{m_i}\frac{\partial E}{\partial \vec{S}_i}\,.$$

Motion equation: Stochastic Landau-Lifshitz-Gilbert Equation:

$$\frac{d\vec{S}_i}{dt} = -\gamma_L\left\{\vec{S}_i \times (\vec{H}_i + \vec{H}_i^{th}) + \lambda\vec{S}_i \times \left[\vec{S}_i \times (\vec{H}_i + \vec{H}_i^{th})\right]\right\},$$

Heisenberg model parameterization

Using Wannier function for local spin rotation perturbation:

$$E_{exc} = -\sum_{i \neq j} J_{ij} \vec{S}_i \cdot \vec{S}_j$$

$$J_{ij} = -\frac{1}{4\pi} \int_{-\infty}^{E_F} d\varepsilon \sum_{mm'm''m'''} \Im(\Delta_i^{mm'} G_{ij,\downarrow}^{m'm''} \Delta_j^{m''m'''} G_{ji,\uparrow}^{m'''m})$$

Where:

$G_{i'j',\sigma}^{mm'}(\varepsilon)$ is the green function in real space;

$\Delta_i^{mm'}$ is Hamiltonian differences in spin up and down.

TB2J package:

`https://gitlab.abinit.org/xuhe/TB2J` Exchange parameter implemented. SIA, DMI term will be implemented soon.

- References:
  Lichtenstein *et al. JMMM 67, 65-74 (1987)*
  Katsnelson & Lichtenstein PRB 61, 8907 (2000)
  Korotin et. al. PRB 91, 224405 (2015)

Examples

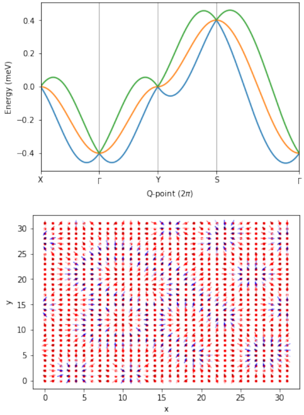


Figure: Magnon dispertion curve and spin configuration for a model system.

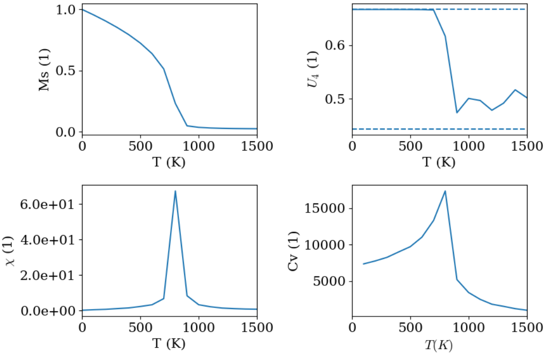


Figure: Thermodynamic property for $LaFeO_3$.

Spin-Lattice coupling: Hamiltonian

Construct lattice Hamiltonian for a reference spin configuration.
ΔE is the change in magnetic energy.

$$
\begin{aligned}
E =& E^{Ref}[\tau] && \textit{Lattice part}\\
&+\Delta E[\tau=0] && \textit{Spin part}\\
&+\sum_u \frac{\partial(\Delta E)}{\partial \tau_u}\tau_u && \textit{Coupling part 1}\\
&+\frac{1}{2}\sum_{u,v}\frac{\partial^2(\Delta E)}{\partial\tau_u\partial\tau_v}\tau_u\tau_v && \textit{Coupling part 2}
\end{aligned}
$$

In the Heisenberg model, $\Delta E = -\sum_{ij} J_{ij}(\vec{S}_i \cdot \vec{S}_j - \vec{S}_i^{Ref}\vec{S}_j^{Ref})$.
We define the spin-phonon coupling parameters:

$$O_{iju} = \frac{\partial J_{ij}}{\partial \tau_u}, \qquad T_{ijuv} = \frac{\partial J_{ij}}{\partial \tau_u \partial \tau_v}$$

Spin-lattice coupling: terms in spin and lattice dynamics

Forces from spin phonon coupling

The force difference from the reference state is:

$$\Delta F_u = \sum_{ij} O_{iju}(\vec{S}_i \cdot \vec{S}_j - \vec{S}_i^{Ref} \vec{S}_j^{Ref}) + \frac{1}{2} \sum_{ij,v} T_{ijuv}(\vec{S}_i \cdot \vec{S}_j - \vec{S}_i^{Ref} \vec{S}_j^{Ref}) \tau_v$$

Exchange parameter in distorted structure

The exchange difference from non-distorted structure:

$$\Delta J_{ij} = \sum_{u} O_{iju} \tau_u + \frac{1}{2} \sum_{uv} T_{ijuv} \vec{\tau}_u \vec{\tau}_v$$

Effective magnetic field:

$$\Delta H_i = 2 \frac{1}{M_i} \Delta J_{ij} \vec{S}_j$$

Spin-Lattice coupling parametrization

Parameters O and T fit by minimizing a target function in a supercell:

Method 1

- various spin configuration and displacements (labeled by *c*)
- calculate DFT forces for each structure, then target function is:

$$
\begin{aligned}
R &= \sum_c \Big| -\left(\Delta \vec{F}_u\right)_c + \sum_{i \neq j} \vec{O}_{iju} \left(\vec{S}_i \cdot \vec{S}_j - \vec{S}_i^{Ref} \cdot \vec{S}_j^{Ref}\right)_c \\
&+ \sum_{i \neq j, v} T_{ijuv} \left[\left(\vec{S}_i \cdot \vec{S}_j - \vec{S}_i^{Ref} \cdot \vec{S}_j^{Ref}\right) \tau_v\right]_c \Big|^2
\end{aligned}
$$

Method 2

- displace atoms, obtain Wannier functions
- calculate new exchange, the target function is:

$$
R = \sum_c \left\{ -(J_{ij})_c + J_{ij}[\tau = 0] + \sum_u \vec{O}_{iju} (\vec{\tau}_u)_c + \frac{1}{2} \sum_{uv} (\vec{\tau}_u)_c \mathbf{T}_{ijuv} (\vec{\tau}_v)_c \right\}^2
$$

Method 3

- Using perturbation theory to calculate O and T from electron-phonon coupling :

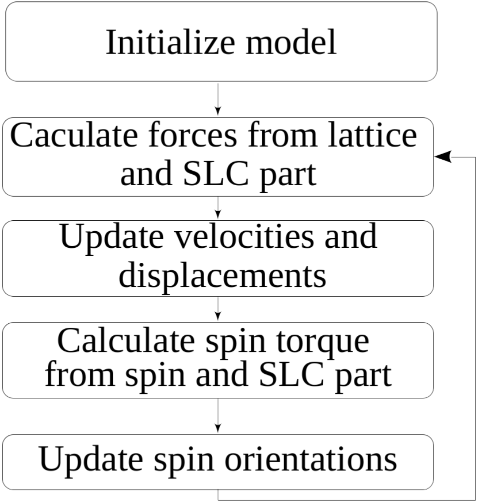
$$\mathscr{O}_{iju} = \Delta_i \sum_{rs} [G^0_{ir\uparrow} \frac{dH_{rs\uparrow}}{d\tau} G^0_{sj\uparrow}] \Delta_j G^0_{ji\downarrow} + \Delta_i G^0_{ij\uparrow} \Delta_j \sum_{rs} [G^0_{jr\downarrow} \frac{dH_{rs\downarrow}}{d\tau} G^0_{si\downarrow}]$$
$$+ \frac{d\Delta_i}{d\tau_u} G^0_{ij\uparrow} \Delta_j G^0_{ji\downarrow} + \Delta_i G^0_{ij\uparrow} \frac{d\Delta_j}{d\tau_u} G^0_{ji\downarrow}$$

$$(partial)\mathscr{T}_{ijuv} = \Delta_i \sum_{rs} [G^0_{ir\uparrow} \frac{dH_{rs\uparrow}}{d\tau_u} G^0_{sj\uparrow}] \Delta_j \sum_{pq} [G^0_{jp\downarrow} \frac{dH_{pq\downarrow}}{d\tau_v} G^0_{qi\downarrow}] + \overleftrightarrow{uv}$$
$$+ \frac{1}{2} \Delta_i \sum_{rs} [G^0_{ir\uparrow} \frac{d^2 H_{rs\uparrow}}{d\tau_u d\tau_v} G^0_{sj\uparrow}] \Delta_j G^0_{ji\downarrow}$$
$$+ \frac{1}{2} \Delta_i G^0_{ij\uparrow} \Delta_j \sum_{rs} [G^0_{jr\downarrow} \frac{d^2 H_{rs\downarrow}}{d\tau_u d\tau_v} G^0_{si\downarrow}]$$

$$O_{iju} = -\frac{1}{4\pi} \Im \int_{-\infty}^{E_F} d\varepsilon \, Tr \mathscr{O}_{iju} \qquad T_{ijuv} = -\frac{1}{4\pi} \Im \int_{-\infty}^{E_F} d\varepsilon \, Tr \mathscr{T}_{ijuv}$$

Coupled dynamics

The procedure of spin and lattice coupled dynamics:



Postprocessing
Observables affected by SLC:

- changes of average positions/magnetic moments
- shift/linewidth in phonon/magnon dispertion curve

Conclusions

- Multibinit Generic data structure:
  - Implemented
  - Used by spin dynamics.
- Spin dynamics features:
  - Hamiltonian with exchange, DMI, SIA, external field.
  - LLG equation; Monte-Carlo.
  - Parameters: TB2J (Python code interfaced with Wannier90, available on gitlab).
    - Exchange: implemented.
    - DMI and SIA will follow soon.
  - Observables: spin temperature, macroscopic magnetic moment, specific heat, magnetic susceptibility, correlation functions, magnon band structure.
  - Documentation ready.
- Spin-lattice coupling
  - Algorithm being tested in a python prototype code.
  - Parameters fitting: Multiple methods yet to be tested.
  - Implementation in Multibinit started.