Development of second-principles methods on spin models and electron-lattice models

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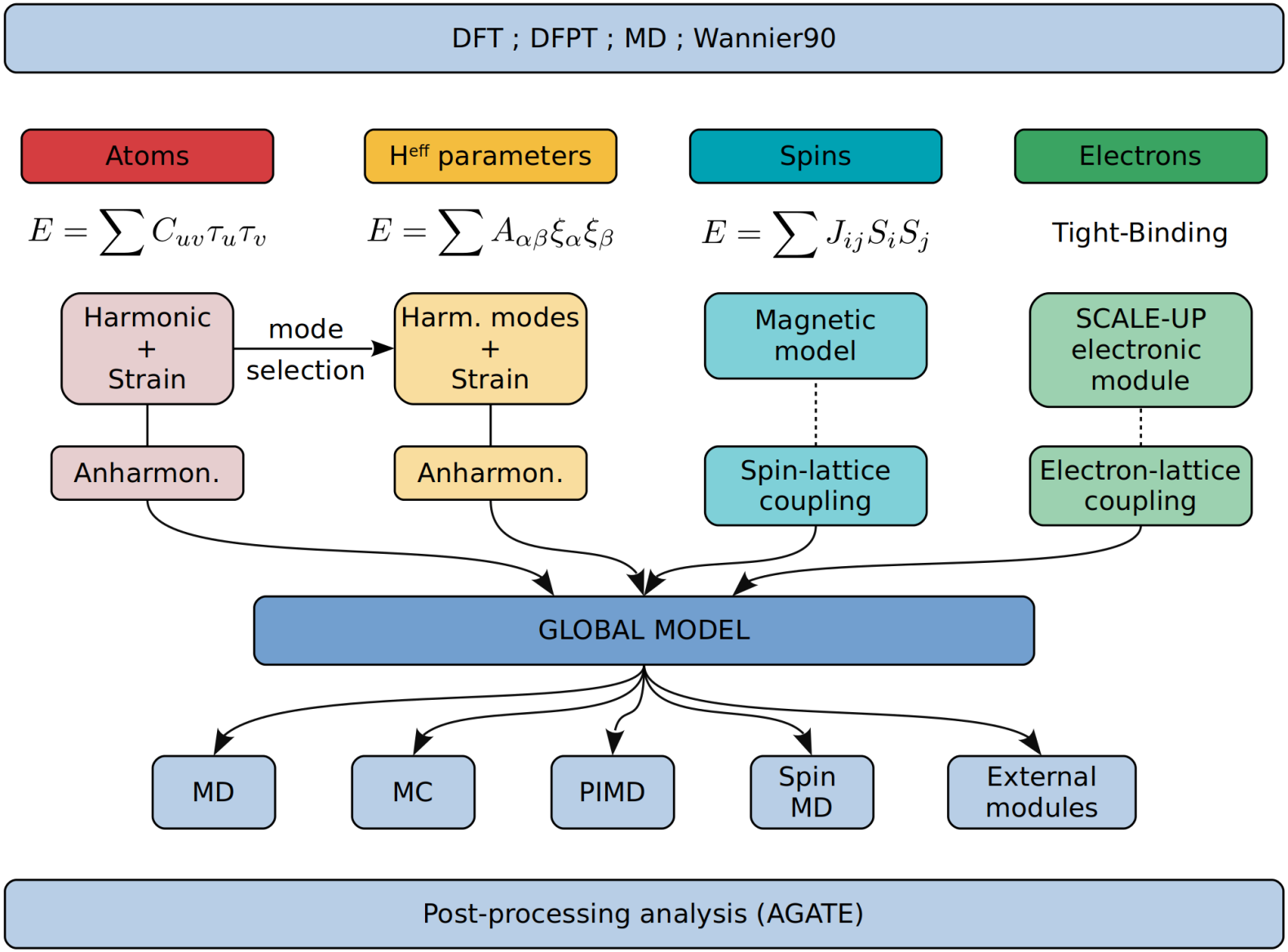
Overview


DFT ; DFPT ; MD ; Wannier90
Atoms
H^{eff} parameters
Spins
Electrons
$E = \sum C_{uv}\tau_u\tau_v$
$E = \sum A_{\alpha\beta}\xi_\alpha\xi_\beta$
$E = \sum J_{ij}S_iS_j$
Tight-Binding
Harmonic + Strain
mode selection
Harm. modes + Strain
Magnetic model
SCALE-UP electronic module
Anharmon.
Anharmon.
Spin-lattice coupling
Electron-lattice coupling
GLOBAL MODEL
MD
MC
PIMD
Spin MD
External modules
Post-processing analysis (AGATE)

Spin model

Energy

$$E = -\sum_{ij}(J_{ij}^{iso}\vec{S}_i \cdot \vec{S}_j + \sum_{uv} S_i^u J_{ij}^{ani,uv} S_j^v + \vec{D}_{ij}\vec{S}_i \times \vec{S}_j)$$

Effective magnetic field

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

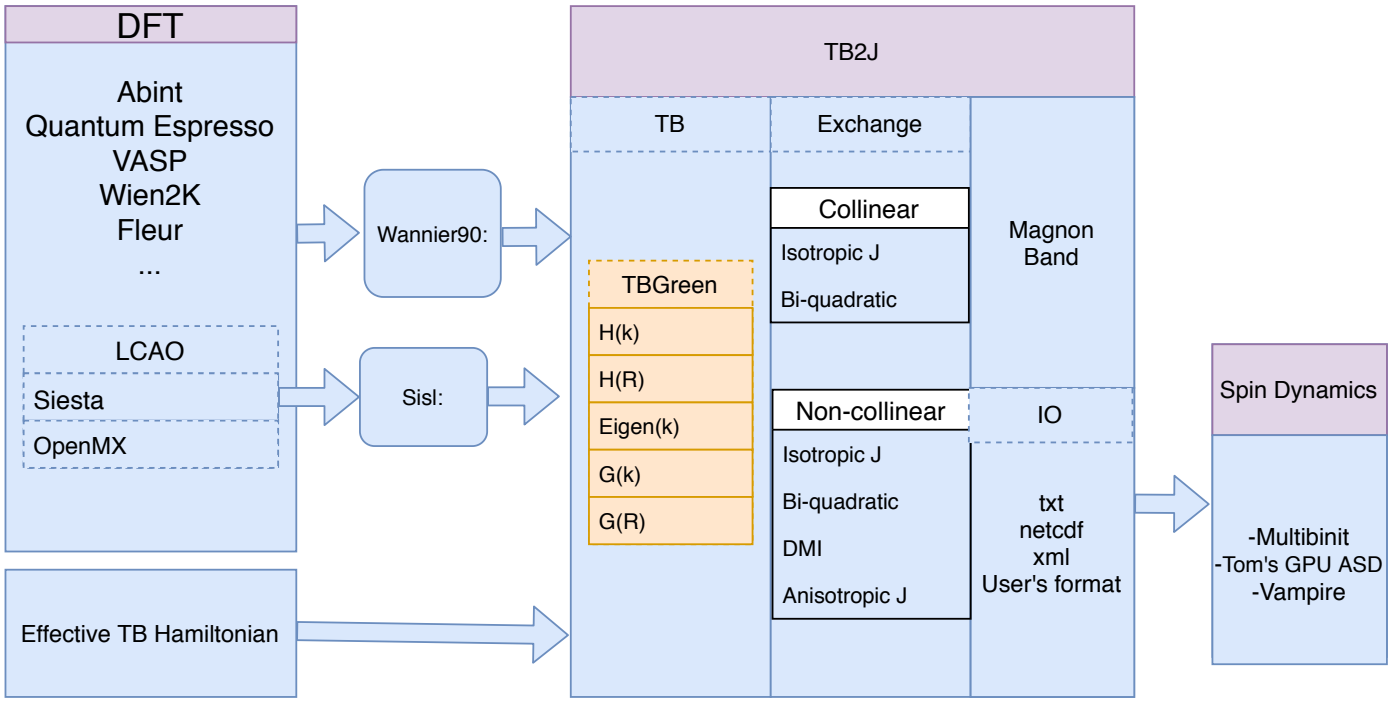
$$\vec{H}_i = -\frac{1}{m_i}\frac{\partial E}{\partial \vec{S}_i} .$$

Stochastic Landau-Lifshitz-Gilbert Equation:

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

TB2J: a python package for computing magnetic interaction parameters

DFT $\rightarrow$ TB $\rightarrow$ Green's function + perturbation $\rightarrow$ J



Use localized basis set for spin rotation perturbation.

$$\begin{aligned} \mathrm{H}_{imjm'\sigma\sigma'}(\vec{R}) = &\quad \langle \psi_{im\sigma}(\vec{r})|\, H\, |\psi_{jm'\sigma'}(\vec{r}+\vec{R})\rangle \\ \mathrm{S}_{imjm'\sigma\sigma'}(\vec{R}) = &\quad \langle \psi_{im\sigma}(\vec{r})|\psi_{jm'\sigma'}(\vec{r}+\vec{R})\rangle \end{aligned}$$

Decompose H_i into scalar (charge) part and vector (spin) part.

$$\begin{aligned} \mathrm{H}_{imm'} &= p^0_{imm'}\mathrm{I} + \vec{p}_{imm'}\cdot\vec{\sigma} \\ &= p^0_{imm'}\mathrm{I} + p_{imm'}\vec{e}_{imm'}\cdot\vec{\sigma} \end{aligned}$$

i, j: site indices. m: orbital index, σ: spin index.

Rigid rotation of exchange field:

$$\delta\mathrm{H}_{imm'} = p_{imm'}\delta\vec{e}_{imm'}$$

Green's function in k-space:

$$\mathrm{G}(\vec{k},\epsilon) = \Big(\epsilon \mathrm{S}(\vec{k}) - \mathrm{H}(\vec{k})\Big)^{-1}$$

In real space:

$$\mathrm{G}(\vec{R},\epsilon) = \int_{BZ} \mathrm{G}(\vec{k},\epsilon) e^{-i\vec{k}\cdot\vec{R}}\, d\vec{k}$$

Decompose G_{ij} into scalar and vector parts:

$$\mathrm{G}_{im,jm'} = G^0_{im,jm'}\mathrm{I} + \vec{G}_{im,jm'} \cdot \vec{\sigma}$$

Perturbing two spin rotations, the energy difference due to the interaction between i and j :

$$\delta E_{ij} = -\frac{2}{\pi} \int_{-\infty}^{E_F} Im\, \mathrm{Tr}(\delta \mathrm{H}_i \mathrm{G}_{ij} \delta \mathrm{H}_j \mathrm{G}_{ji})\, d\epsilon$$

Plug in the spin rotation perturbation is $\delta \mathrm{H}_i = p_i \delta \vec{e}_i$, we get:

$$\begin{aligned}\delta E_{ij} = &-2\,\mathrm{Im}[A_{ij}^{00} - \sum_{u=x,y,z} A_{ij}^{uu}]\delta\vec{e}_i \cdot \delta\vec{e}_j \\ &- 2 \sum_{u,v\in x,y,z} \delta e_i^u\, \mathrm{Im}(A_{ij}^{uv} + A_{ij}^{vu})\delta e_j^v \\ &- 2\,\mathrm{Re}(A_{ij}^{0u} - A_{ij}^{u0}) \cdot (\delta\vec{e}_i \times \delta\vec{e}_j)\end{aligned}$$

where

$$A_{ij}^{uv} = -\frac{1}{\pi}\int_{-\infty}^{E_F} \mathrm{Tr}\{\mathrm{p}_i \mathrm{G}_{ij}^u \mathrm{p}_j \mathrm{G}_{ji}^v\}\, d\epsilon$$

and $u, v \in (0, x, y, z)$
Comparing with the Heisenberg model, we get:

$$\begin{aligned}J_{ij}^{iso} &= \mathrm{Im}(A_{ij}^{00} - A_{ij}^{xx} - A_{ij}^{yy} - A_{ij}^{zz}) \\ J_{ij}^{ani,uv} &= \mathrm{Im}(A_{ij}^{uv} + A_{ij}^{vu}) \\ D_{ij}^u &= \mathrm{Re}(A_{ij}^{0u} - A_{ij}^{u0}),\end{aligned}$$

TB2J: usage and outputs

Prepare Wannier Hamiltonian files and run:

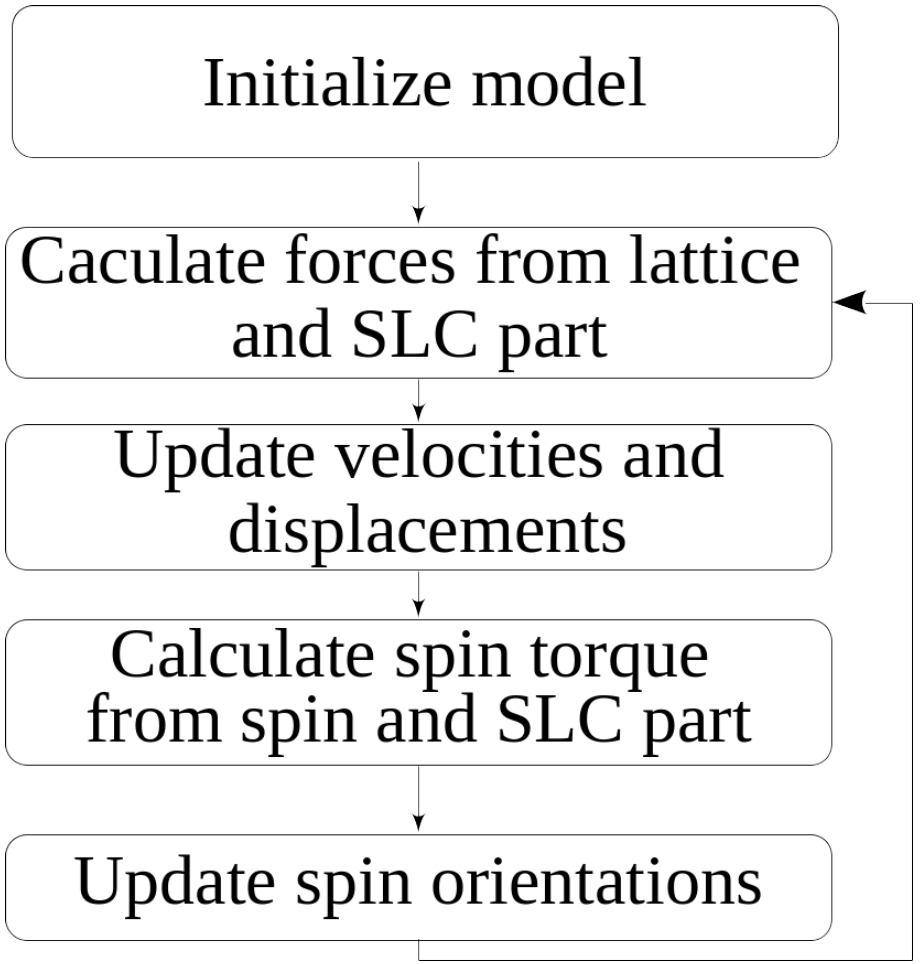
```
wann2J.py --spinor --prefix_spinor abinito --elements Fe
          --posfile abinit.in --efermi 6.15 --kmesh 5 5 5
```

```
================================================================================================
Information:
Exchange parameters generated by TB2J 0.2.5.
================================================================================================
Cell (Angstrom):
0.030   3.950   3.950
3.950   0.030   3.950
3.950   3.950   0.030

================================================================================================
Atoms:
(Note: charge and magmoms only count the wannier functions.)
Atom_number       x          y          z      w_charge    M(x)      M(y)      M(z)
Bi1             0.2413     0.2413     0.2413     2.1538   -0.0015    0.0000   -0.0044
Bi2             4.2060     4.2060     4.2060     2.1538    0.0000    0.0000    0.0044
Fe1             2.0165     2.0165     2.0165     6.3564   -0.0260    0.0000    3.7927
Fe2             5.9812     5.9812     5.9812     6.3564   -0.0176    0.0000   -3.7927
O1              5.5238     2.1558     3.9388     4.8306   -0.0013    0.0000   -0.0705
.....
Total                                            46.0038   -0.0493    0.0000   -0.0000

================================================================================================
Exchange:
i           j           R       J_iso(meV)       vector         distance(A)
------------------------------------------------------------------------------------------------
Fe2         Fe1   (0,   1,   1)  -28.9371 ( 3.934,  0.015,  0.015)  3.934
J_iso: -28.9371
[Testing!] Jprime: -59.620,  B: -15.342
[Testing!] DMI: (-0.5191  1.3581  0.1090)
[Testing!] J_ani:
[[ 0.    -0.002  0.047]
[-0.002  0.    -0.154]
[ 0.047 -0.154  0.   ]]
```

Coupled spin-lattice dynamics

Spin-lattice coupling Hamiltonian:

$$
\begin{aligned}
H_{\mathrm{slc}} &= -\sum_{i,u} L_{iu}\vec{S}_i\tau_u \\
&- \frac{1}{2}\sum_{i,u,v} N_{iuv}\vec{S}_i\tau_u\tau_v \\
&- \sum_{i\neq j,u} O_{iju}\vec{S}_i\vec{S}_j\tau_u \\
&- \frac{1}{2}\sum_{i\neq j,u,v} T_{ijuv}\vec{S}_i\vec{S}_j\tau_u\tau_v .
\end{aligned}
$$



More complicated coupled-spin-lattice mover might be needed.

Spin-lattice coupling parameters downfolded from electron-phonon coupling parameters

$$\delta A_{ij}^{uv} = -\frac{1}{\pi}\int_{-\infty}^{E_F} \mathrm{Tr}\,[\ \delta \mathrm{p}_i \mathrm{G}_{ij}^{u} \mathrm{p}_j \mathrm{G}_{ji}^{v} + \mathrm{p}_i \delta \mathrm{G}_{ij}^{u} \mathrm{p}_j \mathrm{G}_{ji}^{v} + \mathrm{p}_i \mathrm{G}_{ij}^{u} \delta \mathrm{p}_j \mathrm{G}_{ji}^{v} + \mathrm{p}_i \mathrm{G}_{ij}^{u} \mathrm{p}_j \delta \mathrm{G}_{ji}^{v}\ ]$$

Electron-phonon coupling parameters as perturbation.

$$\delta \mathrm{H} = \frac{dH}{d\tau}\delta\tau + \frac{1}{2}\frac{d^2H}{d\tau^2}\delta\tau^2 + \cdots$$

$\delta \mathrm{p}$ is from the onsite part of δH.

$$\delta \mathrm{G} = \mathrm{G}\delta\mathrm{H}\mathrm{G} + \mathrm{G}\delta\mathrm{H}\mathrm{G}\delta\mathrm{H}\mathrm{G} + \cdots$$

The variation to the exchange parameters can thus be written as:

$$\delta J_{ij}^{iso} = \mathrm{Im}(\delta A_{ij}^{00} - \sum_{u=x,y,z} \delta A_{ij}^{uu}),$$

$$\delta J_{ij}^{ani,uv} = \mathrm{Im}(\delta A_{ij}^{uv} + \delta A_{ij}^{vu}),$$

$$\delta D_{ij}^{u} = \mathrm{Re}(\delta A_{ij}^{0u} - \delta A_{ij}^{u0}),$$

TB2J Links

TB2J Documentation:

`https://tb2j.readthedocs.io/`

TB2J GIT repo:

`https://github.com/mailhexu/TB2J`

TB2J Forum:

`https://groups.google.com/g/tb2j`

TB2J Examples:

`https://github.com/mailhexu/TB2J_examples`

TB2J Paper:

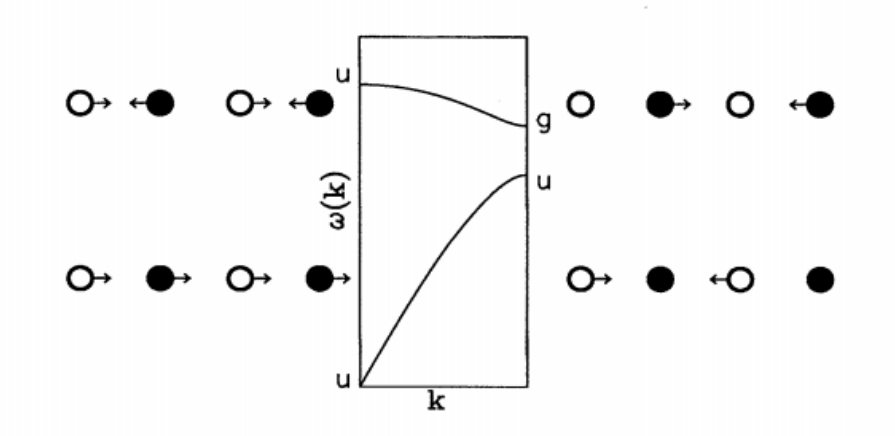
Computer Physics Communications, 107938 (2021).

Lattice Wannier functions

$$E = \sum_{\alpha\beta} A_{\alpha\beta}\xi_\alpha\xi_\beta + \sum_{\alpha\beta,mn} B_{\alpha\beta,mn}\xi_\alpha^m\xi_\beta^n$$

Lattice distortion representation:

- phonon modes
- atomic displacements
- Lattice Wannier function



Phys Rev B, 52(18), 13236, 1995

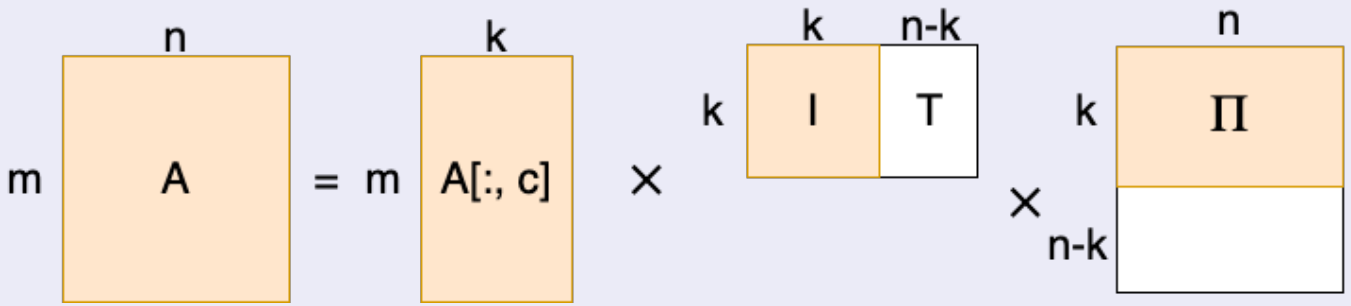
Why LWF

- Allows for disorder (compared with single phonon modes).
- Fewer degrees of freedom than full lattice model
- Easier to fit higher order terms.
- Clear physical meaning.

SCDM-k method for building LWF

Select column density matrix

- Columns of density matrix are localized in local basis set.
- The rank of the density matrix is small.
- Rank revealing QR decomposition



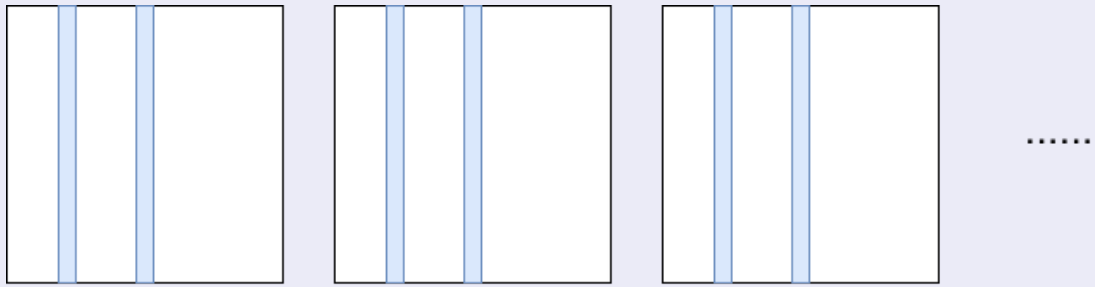
Disentanglement

$$\rho = \Psi \mathrm{f}(E) \Psi^*$$

f: Fermi function / Gaussian / etc

Into k-space

Select one *k* point as anchor point, and choose the same columns for all k-points.



Advantages

- No gauge problem
- Easy to use
- Parallel over k
- Easy to implement
- Can be used as initial guess for MLWF

Refs: Damle and Lin, et al . Chem. Theory Comp. 2015, 11, 4, 1463–1469 ; Multiscale Modeling and Simulation, 2018; J.Comp. Phys. 2016.12.053;

Banddownfolder: a python package for building lattice/electron Wannier functions models.

Demo

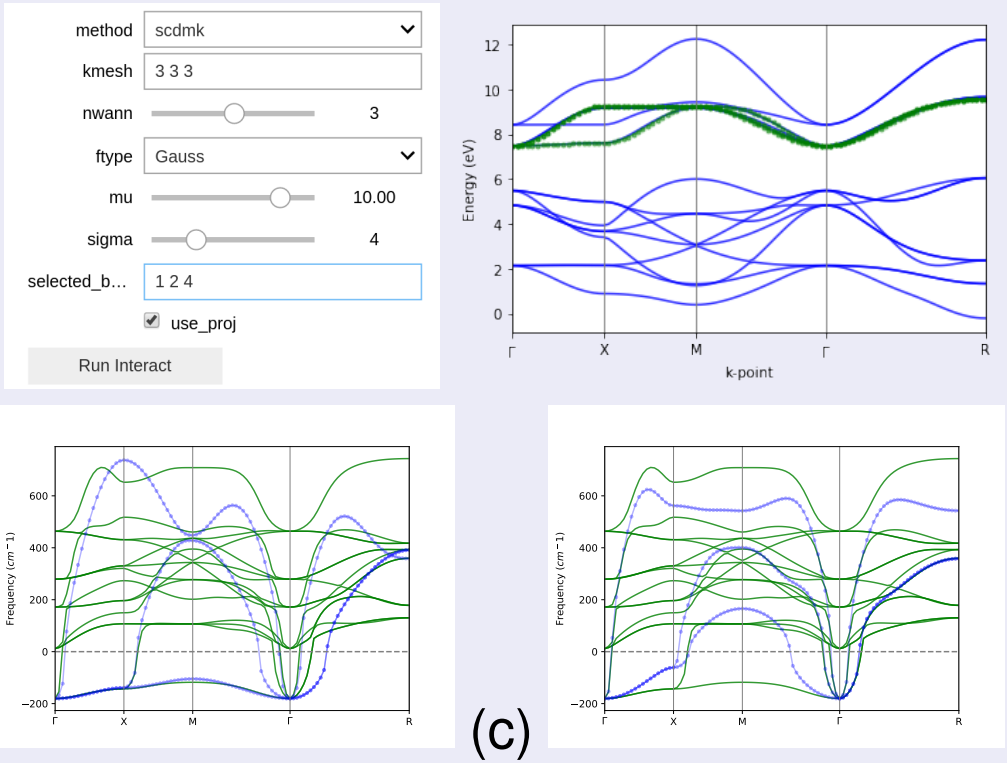


Figure: (a) Electron Wannier function for SrTiO3. (b,c) Lattice Wannier for $BaTiO_3$ using (b) SCDM-k method and (c) projected Wannier function.

Status

- SCDM-k/projected wannier function + disentanglement.
- Python and Fortran version algorithm implemented.
- LO-TO splitting not yet implemented but should be easy.
- Integration in ABINIT on the way.

Electron-lattice coupled models

Two types of electron-lattice models

- Active electron part
- Passive electron part

Electrons as active part

$$\begin{aligned} E(\tau,\rho) &= E^{ref}_{latt}(\tau) - E^{ref}_{elec}(\tau,\rho^{ref}) + E_{elec}(\tau,\rho) \\ H_{elec}(\tau) &= H^{ref}_{elec}(\tau) + H_{ee} \end{aligned}$$

E^{ref}_{latt} and H^{ref}_{elec} are fit to the DFT data with a reference electronic structure, e.g. weakly correlated, non-magnetic DFT.
H_{elec} : H^{ref}_{elec} + correction (e.g. Hubbard U) .

Electrons as passive part

$$E = E_{latt}$$

E_{latt} are fit to DFT calculation with target states.
Electron hamiltonian takes τ from lattice dynamics and does not feed back force.

Electron models: tight binding + electron-lattice-coupling + U

$$\begin{aligned} H^{ref}_{elec}(\tau) &= \sum_{ij}(t^{ref}_{ij}(\tau = 0) + \sum_{u} g_{ij,u}\tau_u)c^{\dagger}_i c_j \\ H_{elec}(\tau) &= H^{ref}(\tau) + \sum_{i} U n^{\uparrow}_i n^{\downarrow}_i \end{aligned}$$

Electron-lattice coupled models: Examples of VO_2

VO_2 : metal-insulator transition accompanied by R-M1 phase transition at 340K.

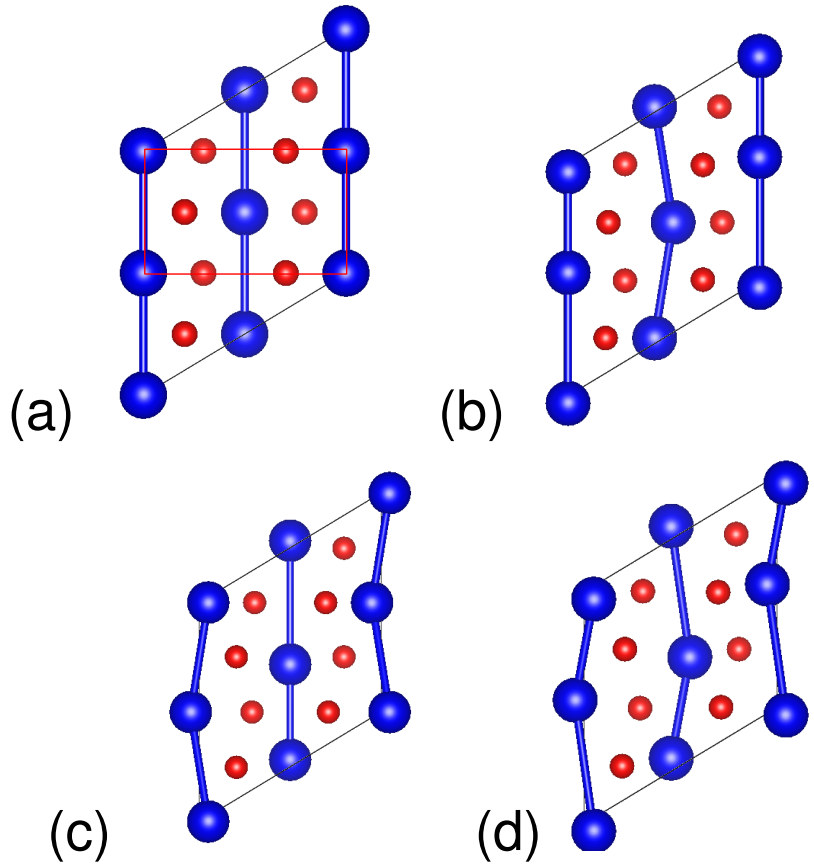


Figure: (a) The R phase. (b) and (c) two degenerate phonon mode distortions at $\mathbf{q} = R$. (d) M1 phase.

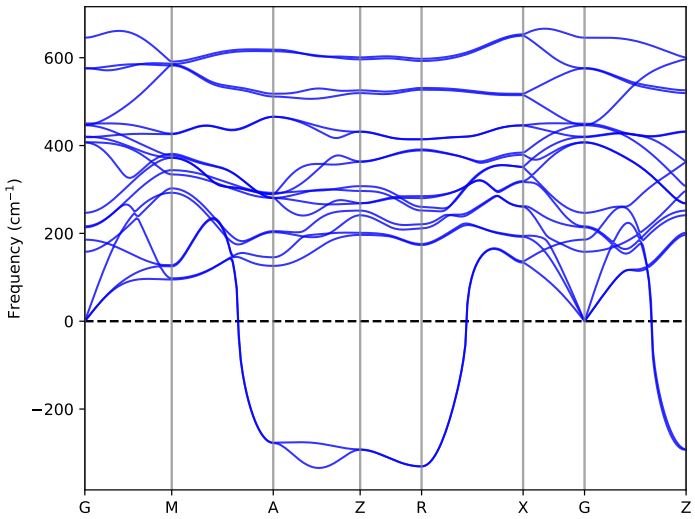


Figure: Phonon band structure of R phase VO_2.

Modelling MIT in VO_2

Fitting of LWF

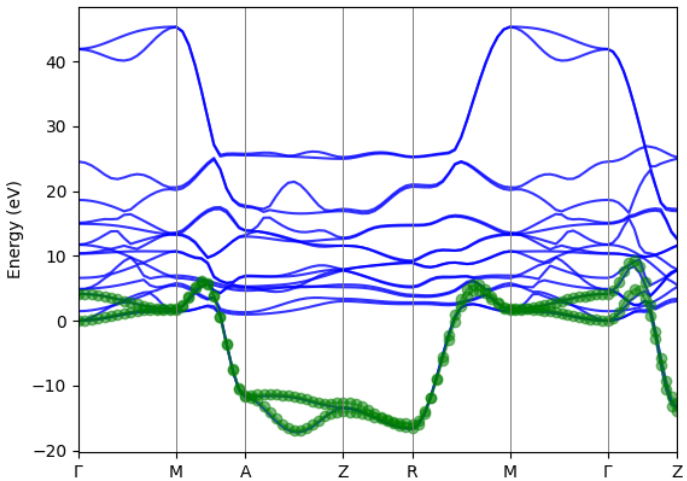


Figure: Eigenvalue of IFC, and the LWF Hamiltonian

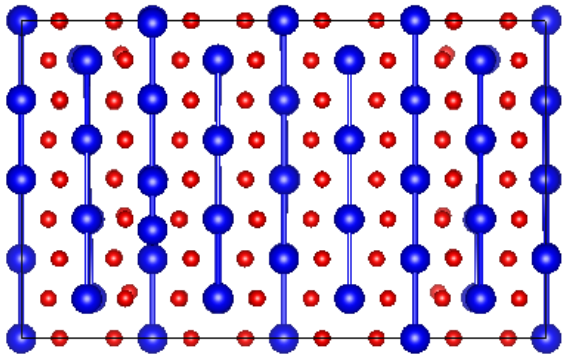


Figure: Supercell with one LWF populated.

Structural and electronic phase transitions in a global view:
The growth of the R/M and metal/insulator domains.

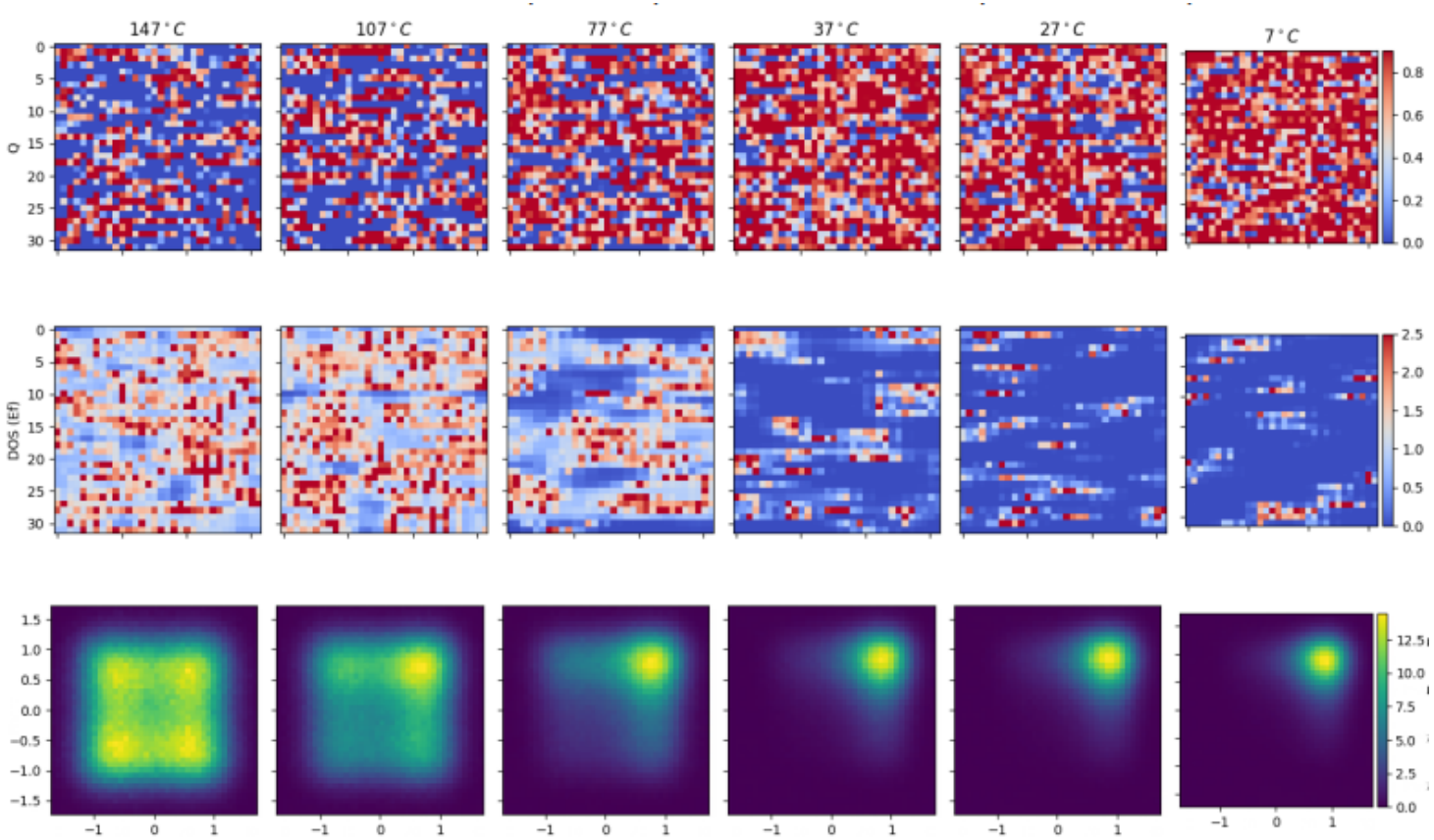


Figure: Top panel: The amplitudes of the LWF in each cell. Middle panel: local density of states at Fermi energy in each cell. Bottom panel: the histograms of the (ξ_1, ξ_2).

Structural and electronic phase transitions in a global view:

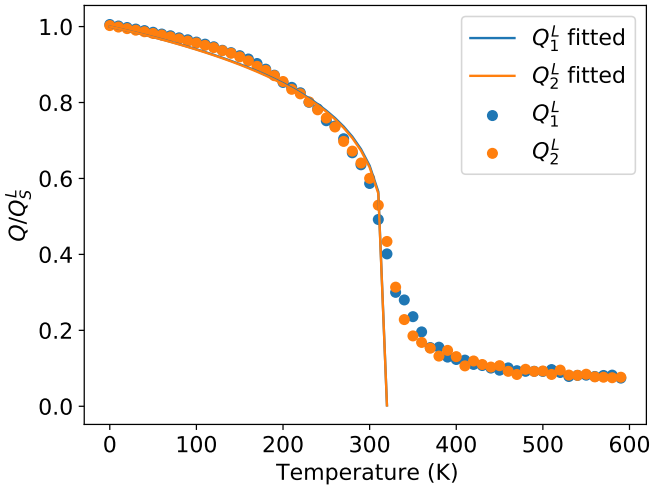


Figure: Fourier component of the LWF amplitudes at $q = R$.

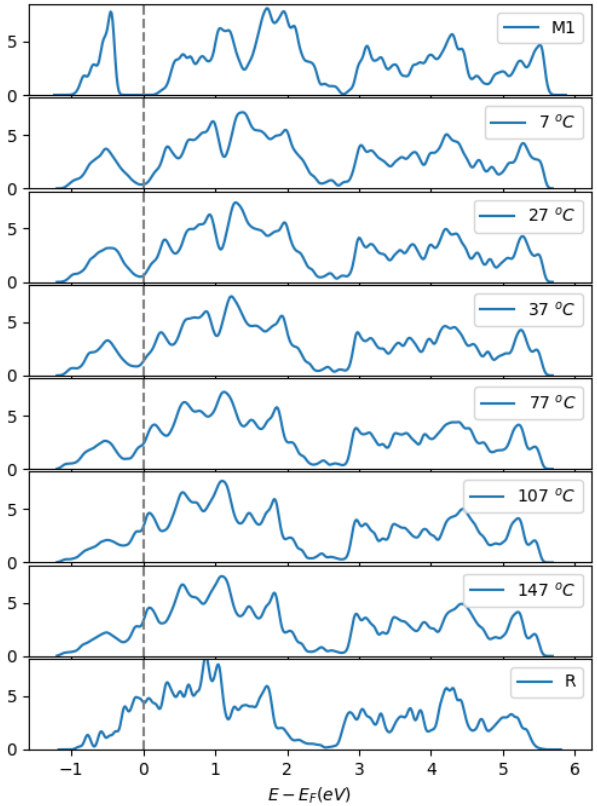


Figure: Total DOS as function of temperature.

Summary

Spin

- New features in TB2J: DMI, Anisotropic exchange.
- Method of getting spin-lattice coupling parameters by downfolding electron-phonon coupling.
- Spin-lattice coupled dynamics implemented in MULTIBINIT.

LWF

- Banddownfolder: package for easy building WF models.
- LWF dynamics implemented in MULTIBINIT.

Electron-lattice

- Tools for building electron-lattice coupled models.
- Electron band structure in thermalized lattice.
- Methods implemented in a python package.