Magnetism: from DFT to Heisenberg model with TB2J and beyond

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June 02, 2025 @ ICN2
Advanced SIESTA workshop, Barcelona, Spain

Spin model

Heisenberg model

$$E = -\sum_{i \neq j} \left[J_{ij}^{iso} \vec{S}_i \cdot \vec{S}_j + \vec{S}_i \mathrm{J}_{ij}^{ani} \vec{S}_j + \vec{D}_{ij} \cdot \left(\vec{S}_i \times \vec{S}_j \right) \right] - \sum_i K_i (\vec{S}_i \cdot \vec{e}_i)^2 , \quad (1)$$

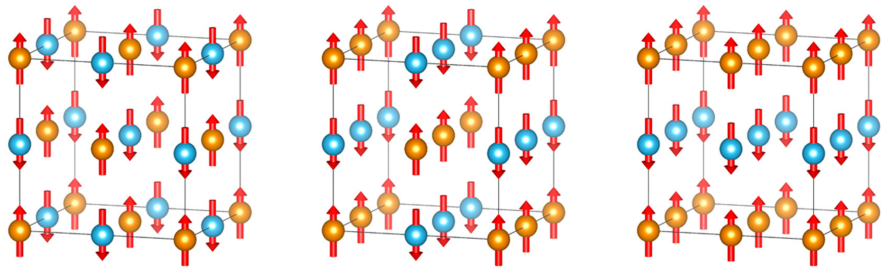
Methods for obtaining the parameters:

- Total energy methods.
- Generalized Bloch theorem (spin spiral method).
- Density functional perturbation theory (DFPT) method
- Cluster expansion method.
- Magnetic force theorem (LKAG) method.

Total energy method

The exchange parameters are obtained by fitting the DFT total energies of multiple magnetic configurations to the Heisenberg model.

- Based on total energy calculations
- Multiple magnetic configurations in supercells needed
- Exchange parameters obtained by fitting
- Good for short-range interactions
- Computationally expensive for many pairs

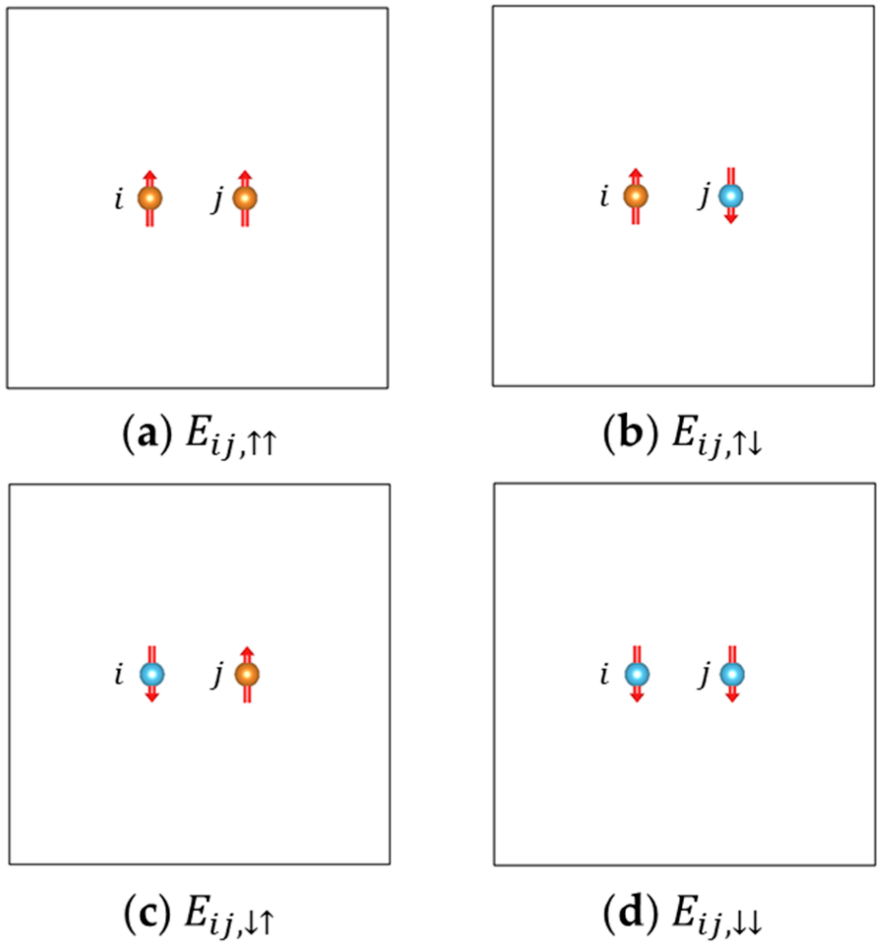


(a) G-type AFM (b) C-type AFM (c) A-type AFM

Four-State Method

The exchange parameters are calculated from energy differences between four carefully chosen magnetic states.

- Based on four magnetic states
- Each pair of J requires four calculations.
- Need large supercell for long-range J



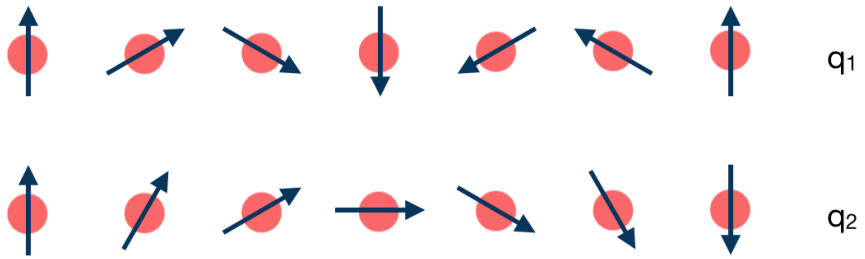
(a) $E_{ij,\uparrow\uparrow}$ (b) $E_{ij,\uparrow\downarrow}$

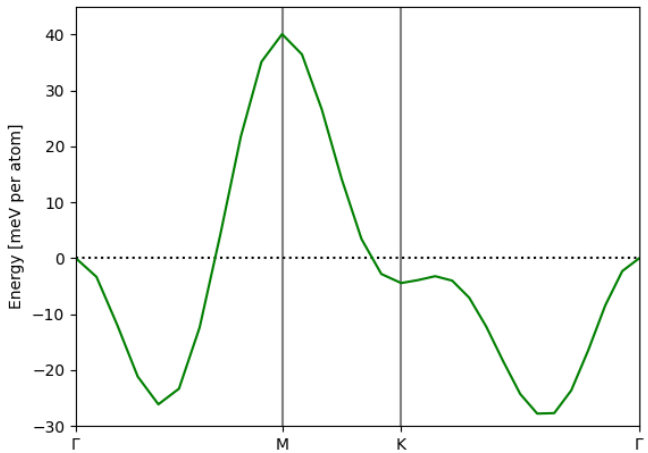
(c) $E_{ij,\downarrow\uparrow}$ (d) $E_{ij,\downarrow\downarrow}$

Spin Spiral Method

The exchange parameters are extracted from the energy dispersion of spin spiral states at different q-vectors using the generalized Bloch theorem.

- Based on generalized Bloch theorem
- Calculates energy vs q-vector
- Direct access to magnon dispersion
- Efficient for periodic structures
- Can handle long-range interactions

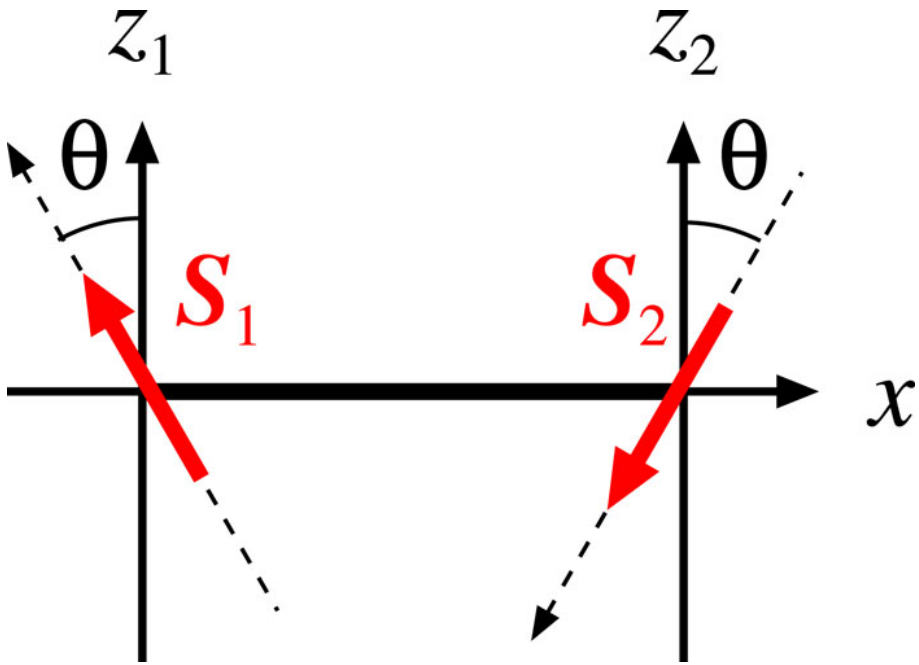




Magnetic Force Theorem Method

The exchange parameters are computed from the response of the system to small rotations of magnetic moments using Green's function perturbation theory.

- Based on perturbation theory
- Uses single-particle Green's function
- Requires only one DFT calculation
- Can compute long-range interactions
- Computationally efficient



What is TB2J

TB2J is a open source python package for calculating the magnetic interaction parameters in Heisenberg models from DFT. It use the magnetic force theorem and take the local rigid spin rotation as a perturbation in the Green's function method.

Features of TB2J

- It can Compute of isotropic/anisotropic exchange parameters.
- It requires only one or three DFT calcualtions in unitcell.
- It can compute exchange paramters up to a long distance.
- It can work with many DFT codes with Wannier interface.
- It can work with DFT codes with numerial atomic orbital basis (Siesta and OpenMX) directly.
- Orbital deomposition of exchange parameters.
- Magnon band structure.
- Output to several spin dynamics code.

LKAG method

Journal of Magnetism and Magnetic Materials 67 (1987) 65–74
North-Holland, Amsterdam

LOCAL SPIN DENSITY FUNCTIONAL APPROACH TO THE THEORY OF EXCHANGE INTERACTIONS IN FERROMAGNETIC METALS AND ALLOYS

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$$J_{ij} = \frac{1}{4\pi}\int^{\epsilon_F} d\epsilon \ \mathrm{Im}\ \mathrm{Tr}_L\left\{\Delta_i \hat{T}^{ij}_{\uparrow}\Delta_j \hat{T}^{ji}_{\downarrow}\right\}.$$

Magnetic local force theorem

In the local density approximation, the first variation to the total energy due to the spin density equals to the variation to the single particle energy (band energy). $\delta E_{TOT} = \delta E_{BAND}$

Electron Hamiltonian in localized basis set

The magnetic force theorem tells us if we can

- The band energy can be evaluated from single-particle Hamiltonian.
- The spin-rotation of the on-site potential can be used as an perturbation.
- The electron Hamiltonian in localized basis set provides both!

$$\mathrm{H}_{imjm'\sigma\sigma'}(\vec{R}) = \langle\psi_{im\sigma}(\vec{r})|\, H\, |\psi_{jm'\sigma'}(\vec{r}+\vec{R})\rangle\,, \tag{2}$$

$$\mathrm{S}_{imjm'\sigma\sigma'}(\vec{R}) = \langle\psi_{im\sigma}(\vec{r})|\psi_{jm'\sigma'}(\vec{r}+\vec{R})\rangle\,, \tag{3}$$

The band energy is the integration of density state below Fermi energy.

$$E_{band} = \int^{E_f} \epsilon DOS(\epsilon) d\epsilon \tag{4}$$

Spin rotation as perturbation

Decomposing the Hamiltonian for atomic site i, $\mathrm{P}_i = \mathrm{H}_{ii}$, into charge and spin part:

$$\begin{aligned} \mathrm{P}_{imm'} &= p^0_{imm'}\mathrm{I} + \vec{p}_{imm'} \cdot \vec{\sigma}, \\ &= p^0_{imm'}\mathrm{I} + p_{imm'}\vec{e}_{imm'} \cdot \vec{\sigma}, \end{aligned} \tag{5}$$

where m and m' are the orbital indices. $\vec{\sigma}$ are the three Pauli matrices.
The change of H_i due to the rotation of spin is $p_i \delta\vec{e}_i \cdot \vec{\sigma}$ '

Perturbation with Green's function method

We can evaluate the band energy with the Green's function, which is:

$$\mathrm{G}(\vec{k},\epsilon) = \Big(\epsilon \mathrm{S}(\vec{k}) - \mathrm{H}(\vec{k})\Big)^{-1}, \tag{6}$$

where $\mathrm{H}(\vec{k}) = \sum_{\vec{R}} \mathrm{H}(\vec{R}) e^{i\vec{k}\cdot\vec{R}}$, and $\mathrm{S}(\vec{k}) = \sum_{\vec{R}} \mathrm{S}(\vec{R}) e^{i\vec{k}\cdot\vec{R}}$. Transforming $\mathrm{G}(\vec{k},\epsilon)$ to real space, we get:

$$\mathrm{G}(\vec{R},\epsilon) = \int_{BZ} \mathrm{G}(\vec{k},\epsilon) e^{-i\vec{k}\cdot\vec{R}}\, d\vec{k}. \tag{7}$$

Density of state: $n(\epsilon) = -\frac{1}{\pi} Im\, \mathrm{Tr}(\mathrm{G}(\epsilon))$

$$\Delta E = -\frac{1}{\pi}\, \mathrm{Im} \int_{-\infty}^{E_F} \mathrm{Tr}(\delta H G + \delta H G \delta H G + \cdots) \tag{8}$$

Spin-rotation perturbation Green's function method

Now we use the spin rotation as a perturbation. For the rotation of the spin at site i, the change in the energy up to the second order reads as

$$\delta E_i^{1spin} = -\frac{1}{\pi}\int_{-\infty}^{E_F} Im\,\mathrm{Tr}(\delta \mathrm{H}_i \mathrm{G} + \delta \mathrm{H}_i \mathrm{G} \delta \mathrm{H}_i \mathrm{G})\; d\epsilon. \tag{9}$$

Similarly, for the rotation of the spins at sites i and j, the change in energy is given by

$$\begin{aligned}\delta E_{ij}^{2spin} = -\frac{1}{\pi}\int_{-\infty}^{E_F} Im\,\mathrm{Tr}\,[&\delta \mathrm{H}_i \mathrm{G} + \delta \mathrm{H}_i \mathrm{G} \delta \mathrm{H}_i \mathrm{G} \\ &+\delta \mathrm{H}_j \mathrm{G} + \delta \mathrm{H}_j \mathrm{G} \delta \mathrm{H}_j \mathrm{G} \\ &+2\delta \mathrm{H}_i \mathrm{G} \delta \mathrm{H}_j \mathrm{G}\,]\; d\epsilon.\end{aligned} \tag{10}$$

The energy variation due to the two-spin interaction is then

$$\begin{aligned}\delta E_{ij} =& \delta E_{ij}^{2spin} - \delta E_i^{1spin} - \delta E_j^{1spin} = \\ &-\frac{2}{\pi}\int_{-\infty}^{E_F} Im\,\mathrm{Tr}(\delta \mathrm{H}_i \mathrm{G} \delta \mathrm{H}_j \mathrm{G})\; d\epsilon.\end{aligned} \tag{11}$$

Exchange parameters from Green's function method

For the spin-rotation perturbation, $\delta \mathrm{H}_i = p_i \delta \vec{e}_i \cdot \sigma$ With some algebra, we get

$$\begin{aligned} \delta E_{ij} = & -2\,\mathrm{Im}[A_{ij}^{00} - \sum_{u=x,y,z} A_{ij}^{uu}]\delta\vec{e}_i \cdot \delta\vec{e}_j \\ & - 2 \sum_{u,v\in x,y,z} \delta e_i^u\, \mathrm{Im}[A_{ij}^{uv} + A_{ij}^{vu}]\delta e_j^v \\ & - 2\vec{d}_{ij} \cdot (\delta\vec{e}_i \times \delta\vec{e}_j) \end{aligned} \tag{12}$$

in which the 4×4 matrix A_{ij} is defined as

$$A_{ij}^{uv} = -\frac{1}{\pi} \int_{-\infty}^{E_F} \mathrm{Tr}\{\mathrm{p}_i \mathrm{G}_{ij}^u \mathrm{p}_j \mathrm{G}_{ji}^v\}\, d\epsilon, \tag{13}$$

where $u, v \in \{0, x, y, z\}$ and $d_{ij}^u = \mathrm{Re}(A_{ij}^{0u} - A_{ij}^{u0})$.
The δE_{ij} in equation 12 look exactly like the exchange and DMI terms!

$$\delta E_{ij}^{Heisenberg} = -2J_{ij}^{iso}\delta\vec{e}_i \cdot \delta\vec{e}_j - 2\delta\vec{e}_i \mathrm{J}_{ij}^{ani} \delta\vec{e}_j - 2\vec{D}_{ij} \cdot (\delta\vec{e}_i \times \delta\vec{e}_j) \tag{14}$$

Computing J^{iso}, $\vec{D}$, and $\mathrm{J}^{\mathrm{ani}}$

We can map the equation 12 to the Heisenberg model form, and the J^{iso}, $\vec{D}$, and $\mathrm{J}^{\mathrm{ani}}$ can be computed with:

$$J_{ij}^{iso} = \mathrm{Im}(A_{ij}^{00} - A_{ij}^{xx} - A_{ij}^{yy} - A_{ij}^{zz}), \tag{15}$$

$$J_{ij}^{ani,uv} = \mathrm{Im}(A_{ij}^{uv} + A_{ij}^{vu}), \tag{16}$$

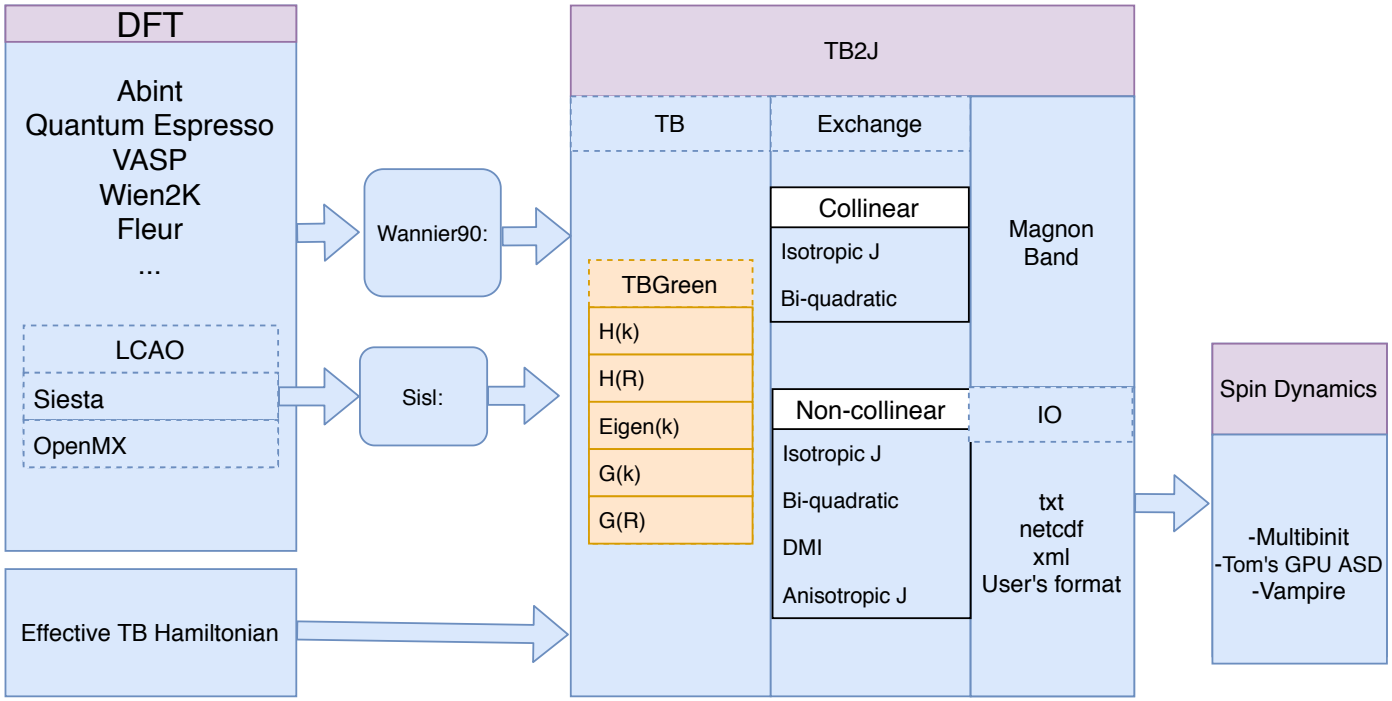
$$D_{ij}^{u} = \mathrm{Re}(A_{ij}^{0u} - A_{ij}^{u0}), \tag{17}$$

In the collinear limit without SOC, the anisotropic exchange and DMI vanishes. If we define $\Delta_i = p_i^{\uparrow} - p_i^{\downarrow}$, the isotropic exchange becomes the same as in the LKAG formula:

$$J_{ij}^{iso} = -\frac{1}{4\pi} \int_{-\infty}^{E_F} \mathrm{Im}\,\mathrm{Tr}\left\{ \Delta_i \mathrm{G}_{ij}^{\uparrow} \Delta_j \mathrm{G}_{ji}^{\downarrow} \right\} d\epsilon. \tag{18}$$

TB2J: the implementation

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



Hands-on section: Using TB2J for computing exchange parameters

Examples

- bcc Fe: basic usage of TB2J-SIESTA.
- CrI3 (1): collinear spin and isotropic exchange.
- CrI3 (2): spin-orbit coupling and anisotropic exchange and DMI
- CrI3 (3): New implementation of TB2J-SIESTA (Magnetocrystalline anisotropy and exchange parameters).

TB2J: NOT a blackbox

DFT calculation

- Imperfect density functional
- The values of U in DFT+U.

Heisenberg model

- Does the Heisenberg model include the key ingredients?
- Higher order term: biquadratic, four-spin, etc.
- Spin-lattice interaction.
- Non-local spin.

Magnetic force theorem

- Rigid spin approximation.
- The basis in tight binding model may not be the "unit" of spin.
- Ligand magnetization.
- Only exact when close to ground state.

What are Magnons?

Definition

Magnons are quantized spin waves - collective excitations of the magnetic moments in a material. They are the quantum equivalent of classical spin waves.

Linear Spin Wave Theory I: Foundations

Basic Assumptions

- Small deviations from ground state configuration
- spin wave have a single wave vector
- Spin operators can be mapped to bosonic operators (Holstein-Primakoff transformation)
- Neglect higher-order terms in the expansion
- Valid at low temperatures where excitations are weak

Linear Spin Wave Theory II: Calculation

Holstein-Primakoff Transformation

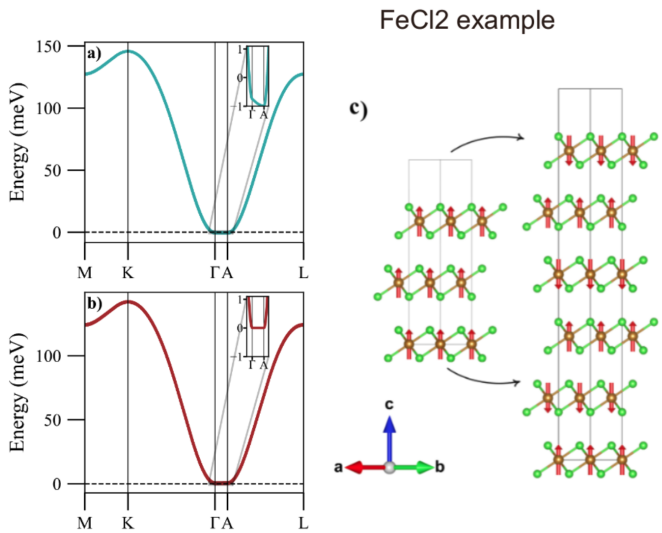
- Maps spin operators to bosonic creation/annihilation operators:
- Preserves spin commutation relations
- Enables quantum mechanical treatment of spin waves

Procedure for Computing Magnon Bands

1. Start with Heisenberg Hamiltonian: $\mathcal{H} = -\sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j$
2. Apply Holstein-Primakoff transformation
3. Fourier transform to k-space: $a_k = \frac{1}{\sqrt{N}} \sum_i e^{-i\vec{k}\cdot\vec{R}_i} a_i$
4. Diagonalize the resulting quadratic Hamiltonian: $\mathcal{H} = \sum_k \hbar\omega_k a_k^\dagger a_k$

Application: Finding Magnetic Ground State

- Determination of magnetic ground state is crucial for:
  - Understanding magnetic properties
  - Validating theoretical models
  - Predicting material behavior
- Procedure:
  - DFT calculation for magnetic structure
  - Computing exchange parameters and magnon band structure
  - construct the new trail magnetic structure from the negative magnon eigenstate.
  - Iterate until convergence



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Systematic determination of a material's magnetic ground state from first principles

Andres Tellez-Mora[1✉], Xu He[2], Eric Bousquet[2], Ludger Wirtz[3] and Aldo H. Romero[1,3]

We present a self-consistent method based on first-principles calculations to determine the magnetic ground state of materials, regardless of their dimensionality. Our methodology is founded on satisfying the stability conditions derived from the linear spin wave theory (LSWT) by optimizing the magnetic structure iteratively. We demonstrate the effectiveness of our method by successfully predicting the experimental magnetic structures of NiO, $FePS_3$, FeP, MnF_2, $FeCl_2$, and CuO. In each case, we compared our results with available experimental data and existing theoretical calculations reported in the literature. Finally, we discuss the validity of the method and the possible extensions.

npj Computational Materials (2024)10:20; https://doi.org/10.1038/s41524-024-01202-z

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Work mostly done by Andres Tellez Mora:

- Systematic approach to ground state determination
- Integration of multiple computational methods
- Validation through experimental data

The Role of Ligand

Challenge in Metal-Ligand Systems

- Strong hybridization between metal and ligand states
- Ligand magnetic moments cannot be neglected
- Simple spin rotation perturbation becomes incomplete
- Traditional Hamiltonian needs modification

Solution

- Explicit treatment of ligand spins required
- Consider metal-ligand hybridization effects
- Modified perturbation scheme needed
- More complete description of magnetic interactions

Ligand Spin Effects in Heisenberg Models: A Löwdin Partitioning Method

Andres Tellez-Mora,[1,*] Xu He,[2] Eric Bousquet,[2] and Aldo H. Romero[1]

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(Dated: October 2024)

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).