

# Magnetism in Abinit

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- Origin of magnetism:
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  - Inside an atom
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1819: Hans Christian Oersted



First experimental evidence of the link between electric current and magnetic field.

# History of magnetism

1820: Jean-Baptiste Biot and Felix Savart



Mathematical expression of the magnetic field generated by an electric current flowing through a wire.

1907: Pierre-Ernest Weiss



First microscopic model to explain magnetism (molecular field).

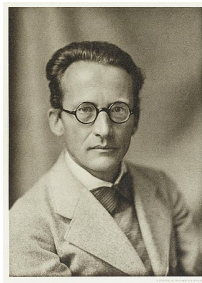
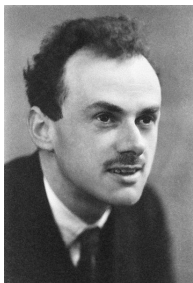
1930: Louis Néel



Extension of Weiss model to antiferromagnetism and ferrimagnetism.  
However, no explanation of the high temperature magnetic phase transitions.

# History of magnetism

Pauli, Dirac, Heisenberg, Schrödinger, *etc*



Magnetism cannot be explained without quantum mechanics (spin)!

all pictures up to this slide are from Wikipedia

# Origin of magnetism: Inside an atom

2 electrons with the same  $l$  but different  $m_l$  ( $\Phi_a$  and  $\Phi_b$  orbitals):

$$H = H_1 + H_2 + H_{12}$$

$$H_1 = -\frac{\hbar^2}{2m} \nabla_1^2 - \frac{Ze^2}{4\pi\epsilon_0 r_1}$$

$$H_2 = -\frac{\hbar^2}{2m} \nabla_2^2 - \frac{Ze^2}{4\pi\epsilon_0 r_2}$$

$$H_{12} = -\frac{e^2}{4\pi\epsilon_0 r_{12}}$$

$$\langle \phi_a(r_i) | H_i | \phi_a(r_i) \rangle = E_1$$

$$\langle \phi_b(r_i) | H_i | \phi_b(r_i) \rangle = E_2$$

$$\langle \phi_i | \phi_j \rangle = \delta_{ij}$$



# Origin of magnetism: Inside an atom

Each electron can have spin up and down states  $\longrightarrow$  4 spin-orbitals:

$$|\uparrow\uparrow\rangle \quad |\uparrow\downarrow\rangle \quad |\downarrow\uparrow\rangle \quad |\downarrow\downarrow\rangle$$

Such that we have the following Hamiltonian:

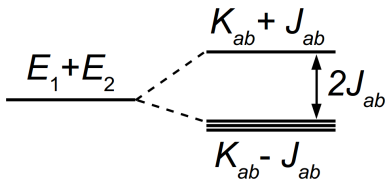
$$H = E_1 + E_2 + \begin{pmatrix} K_{ab} - J_{ab} & 0 & 0 & 0 \\ 0 & K_{ab} & -J_{ab} & 0 \\ 0 & -J_{ab} & K_{ab} & 0 \\ 0 & 0 & 0 & K_{ab} - J_{ab} \end{pmatrix}$$

where we have the Coulomb and Exchange integrals:

$$K_{ab} = \frac{e^2}{4\pi\epsilon_0} \int d^3r_1 \int d^3r_2 \frac{|\phi_a(r_1)|^2 |\phi_b(r_2)|^2}{|r_{12}|} \quad (= U)$$
$$J_{ab} = \frac{e^2}{4\pi\epsilon_0} \int d^3r_1 \int d^3r_2 \frac{\phi_a^*(r_1)\phi_b(r_1)\phi_b^*(r_2)\phi_a(r_2)}{|r_{12}|}$$

# Origin of magnetism: Inside an atom

The diagonalization gives a triplet (FM) and a singlet (AFM) states:



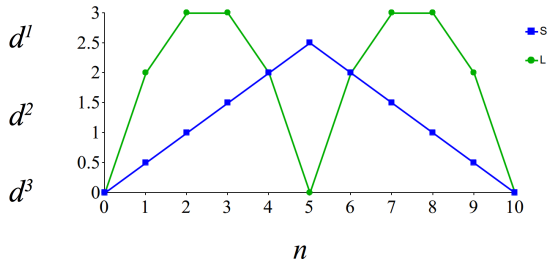
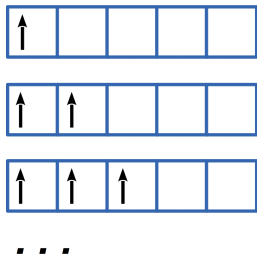
The FM triplet is the ground state.

The Hamiltonian can be re-written in the Heisenberg form:

$$H = \text{constant} - 2J_{ab} \mathbf{S}_1 \cdot \mathbf{S}_2$$

# Origin of magnetism: Inside an atom

To have  $\langle S \rangle \neq 0$  one needs partially filled orbitals ex:  $d^n$  orbitals

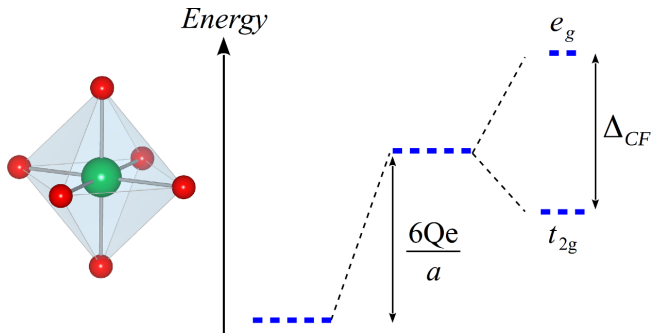


Hund's rules

Most of the crystals are magnetic because they contain Transition-Metal and Rare Earth atoms ( $d$  and  $f$  electrons partially filled)

# Origin of magnetism: Interaction with ligands

## Atom in a solid: Crystal Field Splitting

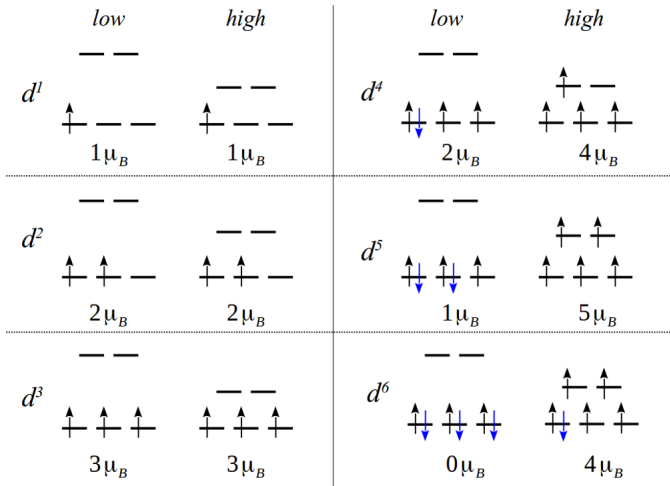


Atomic  $d$  (or  $f$ ) orbitals splitted due to the surrounding atoms.

Hund's rules still apply.

# Origin of magnetism: Interaction with ligands

High-spin and low-spin configurations:

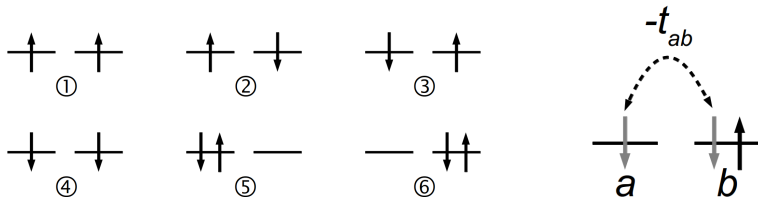


Depending on the size of  $\Delta_{CF}$  relative to  $U$ .

# Origin of magnetism: Between two atoms

The FM state is lower in energy inside an isolated atom (Hund's rules).

This is not necessarily the case between atoms, in  $H_2$  molecule:



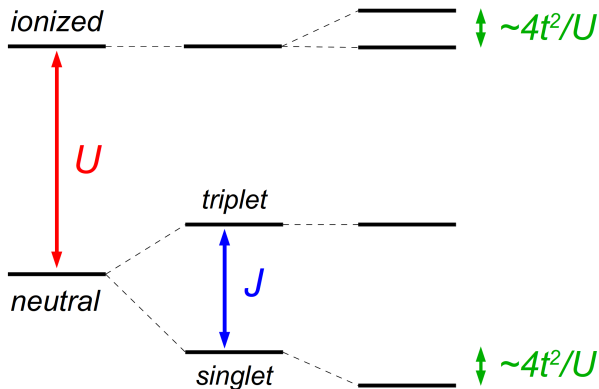
Similar Heisenberg Hamiltonian:  $H = \text{constant} - 2J_{ab}\mathbf{S}_1 \cdot \mathbf{S}_2$

but with the  $1s$  orbital overlap between the 2 atoms:  $J < 0$

The singlet AFM state is lower in energy!

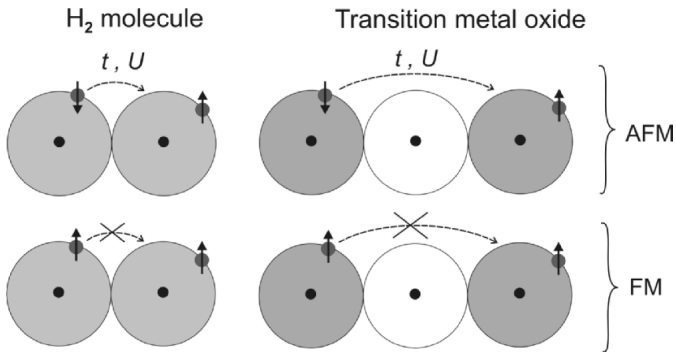
# Origin of magnetism: Between two atoms

The hopping process reinforce the AFM state:



# Origin of magnetism: Interaction through ligands

## Atoms interacting through the ligands: Superexchange



from Stöhr and Siegmann, *Magnetism*, Springer 2006

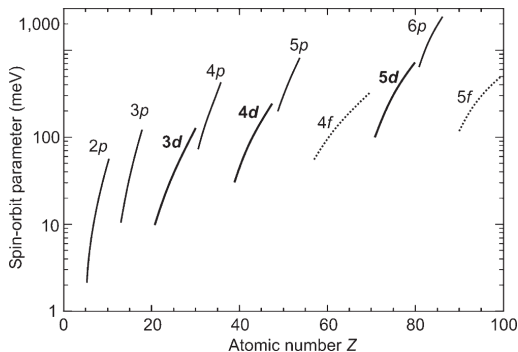
Heisenberg picture still holds (localised electrons,  $t \ll U$ ):  $H = JS_1 S_2$



# Spin-orbit coupling and spin orientations

Relativistic corrections gives an interaction between S and L:

$$\lambda \mathbf{L} \cdot \mathbf{S} = \frac{\mu_0 g_s Z e^2}{16\pi m^2 r^3} \mathbf{L} \cdot \mathbf{S} \quad \text{or} \quad \frac{g_s}{4m^2 c^2 r} \frac{dV}{dr} \mathbf{L} \cdot \mathbf{S}$$



In crystals, this term drives the easy/hard axis orientation of the spins  
It also drives the spin canting (Dzyaloshinskii-Moriya interaction)

# Heisenberg Model

For localised moments, the system can be reduced to an effective spin-spin interaction

2<sup>nd</sup> order Heisenberg model for localised magnetic moments:

$$H = -2 [J\mathbf{S} \cdot \mathbf{S}' + \mathbf{D} \cdot (\mathbf{S} \times \mathbf{S}') + \mathbf{S} \cdot \Phi \cdot \mathbf{S}']$$

- $J \mapsto$  Superexchange interaction (favors  $S \parallel S'$ )
- $\mathbf{D} \mapsto$  Dzyaloshinskii-Moriya interaction (favors  $S \perp S'$ )
- $\Phi \mapsto$  Single Ion Anisotropy (easy/hard spin orientation)

# Magnetism in DFT: Collinear case

DFT based on the charge density  $\rho(r)$

To enlarge DFT to (collinear) magnetism, decomposition of the density:

$$\rho = \rho(\uparrow) + \rho(\downarrow)$$

The Hohenberg and Kohn theorem generalizes with an energy functional:

$$E = E[\rho(\uparrow), \rho(\downarrow)]$$

With 2 Kohn-Sham equations to be solved, one for each spin-channel  $\sigma$ :

$$(T + V_{Ri}(r) + V_H(r) + V_{xc,\sigma}) \phi_{i\sigma}(r) = \epsilon_{i\sigma}(r)$$

with

$$V_{xc,\sigma} = \frac{\delta E_{xc}[\rho(\uparrow), \rho(\downarrow)]}{\delta \rho_{\sigma}(r)}$$

# Magnetism in DFT: Collinear case

Then minimizing the K-S equations we get the ground state with:

$$\rho = \rho(\uparrow) + \rho(\downarrow) \quad \text{and magnetization } m = \rho(\uparrow) - \rho(\downarrow)$$

Supposing the magnetic moments are localised around the atoms (this is often the case for *d* and *f* electrons), one can compute magnetization on each atom (*prtdensph* input flag in ABINIT)

Example of Fe bulk BCC:

```
Integrated electronic and magnetization densities in atomic spheres:
```

```
-----  
Note: Diff(up-dn) is a rough approximation of local magnetic moment
```

Atom	Radius	up_density	dn_density	Total(up+dn)	Diff(up-dn)
1	2.00000	8.152787	5.919175	14.071962	2.233612

```
-----  
Sum:                8.152787    5.919175    14.071962    2.233612
```

```
Total magnetization (from the atomic spheres):    2.233612
```

```
Total magnetization (exact up - dn):              2.228468
```

about  $2.23 \mu_B$ .

# Magnetism in DFT: Non-collinear case

Wave functions are described by spinors:

$$\phi_i(\mathbf{r}) = \begin{pmatrix} \phi_{i\uparrow} \\ \phi_{i\downarrow} \end{pmatrix}$$

Such as the density is a  $2 \times 2$  matrix:

$$\rho = \begin{pmatrix} \rho^{\uparrow\uparrow} & \rho^{\uparrow\downarrow} \\ \rho^{\downarrow\uparrow} & \rho^{\downarrow\downarrow} \end{pmatrix} = \frac{1}{2} \begin{pmatrix} n + m_z & m_x - im_y \\ m_x + im_y & n - m_z \end{pmatrix}$$

with  $n$  the electron density and  $m_i$  the magnetization density along the direction  $i$

$$n(\mathbf{r}) = \frac{1}{2} \text{Tr} \rho(\mathbf{r}) = \sum_{\alpha} \rho^{\alpha\alpha}(\mathbf{r}) \qquad \mathbf{m}(\mathbf{r}) = \sum_{\alpha\beta} \rho^{\alpha\beta}(\mathbf{r}) \cdot \boldsymbol{\sigma}_{\alpha\beta}$$

with the Pauli matrices  $\boldsymbol{\sigma}_{\alpha\beta} = (\sigma_x, \sigma_y, \sigma_z)$ :

$$\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \qquad \sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \qquad \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$$

# Magnetism in DFT: Non-collinear case

Kohn-Sham equations with spinors:

$$\sum_{\beta} H^{\alpha\beta} |\phi_i^{\beta}\rangle = \varepsilon_i |\phi_i^{\alpha}\rangle$$

where the Hamiltonian is a  $2 \times 2$  matrix:

$$H^{\alpha\beta} = T\delta_{\alpha\beta} + V(r)\delta_{\alpha\beta} + \int \frac{n(r')}{r-r'} dr' \delta_{\alpha\beta} + V_{xc}^{\alpha\beta}(r)$$

and:

$$V_{xc}^{\alpha\beta}(r) = \frac{\delta E_{xc}[\rho(r)]}{\delta \rho^{\alpha\beta}(r)}$$

$\rho^{\alpha\beta}$  is diagonal when  $m = m_z \mapsto$  collinear case.

# Magnetism in DFT: Non-collinear case

One needs SOC to couple directions (space rotations) to the spins:

- controlled by *so\_psp* flag for norm-conserving pseudopotential
- controlled by *pawspnorb* flag for PAW

(Mandatory to probe magnetic anisotropies or spin canting)

Magnetization on each atom and in each direction can also be estimated through *prtdensph=1* (default for >v8.8) flag, example of Fe bulk BCC:

```
Integrated electronic and magnetization densities in atomic spheres:
```

```
-----  
Note:      this is a rough approximation of local magnetic moments  
Atom  Radius      Total density      mag(x)      mag(y)      mag(z)  
  1    2.00000      14.078578      -0.000039   -0.000023   2.274108  
-----  
Total magnetization (spheres)      -0.000039   -0.000023   2.274108  
Total magnetization (exact)        -0.000039   -0.000023   2.270315
```

# Magnetism in practice with Abinit

*nsppol*, *nspinor* and *nspden* input flags:

	<i>nsppol</i>	<i>nspinor</i>	<i>nspden</i>
Non-magnetic	1	1	1
Collinear FM	2	1	2
Collinear AFM	1	1	2
Non-Collinear	1	2	4

Initialize *spinat* = ( $m_x$ ,  $m_y$ ,  $m_z$ ) for each atom  
(use *spinat* = (0, 0,  $m$ ) for collinear calculations)

SOC coupling controlled by *so\_psp* (norm-conserving) or *pawspnorb* for PAW  
(PAW SOC coupling amplitude can be artificially tuned with *spnorbscl* flag).

With SOC do not use (time-reversal) symmetry (*kptopt*=3 and *nsym*=1;  
*nsym*≠1 and *kptopt*=4 is under dev.)



DFT+ $U$  is in most cases necessary to get the magnetism right

Exemple for BiFeO<sub>3</sub>:

```
usepawu 1
lpawu   -1  2  -1  # Bi Fe 0
upawu   0.0 5.0 0.0 eV
jpawu   0.0 0.0 0.0 eV
```

$U = 5$  eV is applied on the Fe-d orbitals of Fe.

*NB1: DFT+ $U$  works only within PAW calculations in Abinit*

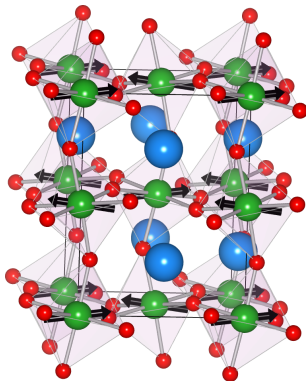
*NB2: Hybrid functionals are good too but cost more CPU time...*

*NB3: Be careful of multiple orbital occupation minima (see dmatpawu flag to control it, PRB 79, 235125 (2009))*

# Example of $\text{YCrO}_3$

$\text{YCrO}_3$  has a distorted perovskite structure ( $Pnma$  space group)

$\text{YCrO}_3$  is a non-collinear AFM (wFM and wAFM)



# Collinear case of $\text{YCrO}_3$ (input)

```
natom 20
ntypat 3
znucl 39 24 8
typat 4*1 4*2 12*3
```

```
acell 5.518 7.54 5.247 Angstrom
```

```
xred
```

```
0.07064 0.75000 0.48013 # Y
0.42935 0.25000 0.98013 # Y
0.57064 0.75000 0.01986 # Y
0.92935 0.25000 0.51986 # Y
0.00000 0.00000 0.00000 # Cr
0.50000 0.00000 0.50000 # Cr
0.00000 0.50000 0.00000 # Cr
0.50000 0.50000 0.50000 # Cr
0.46054 0.75000 0.61179 # 0
0.03945 0.25000 0.11179 # 0
0.96054 0.75000 0.88820 # 0
0.53945 0.25000 0.38820 # 0
0.30409 0.55780 0.19616 # 0
0.19590 0.44219 0.69616 # 0
0.80409 0.94219 0.30383 # 0
0.69590 0.05780 0.80383 # 0
0.69590 0.44219 0.80383 # 0
0.80409 0.55780 0.30383 # 0
0.19590 0.05780 0.69616 # 0
0.30409 0.94219 0.19616 # 0
```

```
nspden 2 # (Collinear)
nspol 1 # (AFM)
nspinor 1 # no spinorial WF
spinat
12*0 # Y
0.0 0.0 5.0 # Cr1 (d3)
0.0 0.0 -5.0 # Cr2 (d3)
0.0 0.0 -5.0 # Cr3 (d3)
0.0 0.0 5.0 # Cr4 (d3)
36*0 # 0
prtdensph 1
```

# Collinear case YCrO<sub>3</sub> (output)

Integrated electronic and magnetization densities in atomic spheres:

---

Note: Diff(up-dn) is a rough approximation of local magnetic moment

Atom	Radius	up_density	dn_density	Total(up+dn)	Diff(up-dn)
1	2.21000	4.011174	4.011174	8.022348	-0.000000
2	2.21000	4.011174	4.011174	8.022348	-0.000000
3	2.21000	4.011174	4.011174	8.022348	-0.000000
4	2.21000	4.011174	4.011174	8.022348	-0.000000
5	2.10818	7.280490	4.827179	12.107670	2.453311
6	2.10818	4.827179	7.280490	12.107670	-2.453311
7	2.10818	4.827179	7.280490	12.107670	-2.453311
8	2.10818	7.280490	4.827179	12.107670	2.453311
9	1.41465	2.281923	2.281923	4.563846	0.000000
10	1.41465	2.281923	2.281923	4.563846	0.000000
11	1.41465	2.281923	2.281923	4.563846	0.000000
12	1.41465	2.281923	2.281923	4.563846	0.000000
13	1.41465	2.273977	2.274262	4.548239	-0.000285
14	1.41465	2.274262	2.273977	4.548239	0.000285
15	1.41465	2.273977	2.274262	4.548239	-0.000285
16	1.41465	2.274262	2.273977	4.548239	0.000285
17	1.41465	2.273977	2.274262	4.548239	-0.000285
18	1.41465	2.274262	2.273977	4.548239	0.000285
19	1.41465	2.273977	2.274262	4.548239	-0.000285
20	1.41465	2.274262	2.273977	4.548239	0.000285

---

Sum:		67.580681	67.580681	135.161363	-0.000000
Total magnetization (from the atomic spheres):					-0.000000
Total magnetization (exact up - dn):					-0.000000

# Non-collinear case of YCrO<sub>3</sub> (input with spins along z)

```
natom 20
ntypat 3
znucl 39 24 8
typat 4*1 4*2 12*3

acell 5.518 7.54 5.247 Angstrom
xred
  0.07064 0.75000 0.48013 # Y
  0.42935 0.25000 0.98013 # Y
  0.57064 0.75000 0.01986 # Y
  0.92935 0.25000 0.51986 # Y
  0.00000 0.00000 0.00000 # Cr
  0.50000 0.00000 0.50000 # Cr
  0.00000 0.50000 0.00000 # Cr
  0.50000 0.50000 0.50000 # Cr
  0.46054 0.75000 0.61179 # 0
  0.03945 0.25000 0.11179 # 0
  0.96054 0.75000 0.88820 # 0
  0.53945 0.25000 0.38820 # 0
  0.30409 0.55780 0.19616 # 0
  0.19590 0.44219 0.69616 # 0
  0.80409 0.94219 0.30383 # 0
  0.69590 0.05780 0.80383 # 0
  0.69590 0.44219 0.80383 # 0
  0.80409 0.55780 0.30383 # 0
  0.19590 0.05780 0.69616 # 0
  0.30409 0.94219 0.19616 # 0

nspden 4 # (Non-collinear)
nspol 1
nspinor 2
so_psp 3*1 # (SOC for norm-conserving)
spinat
12*0 # Y
0.0 0.0 5.0 # Cr1 (d3)
0.0 0.0 -5.0 # Cr2 (d3)
0.0 0.0 -5.0 # Cr3 (d3)
0.0 0.0 5.0 # Cr4 (d3)
36*0 # 0
prtdensph 1
```

# Non-collinear case $\text{YCrO}_3$ (output from spinat along z)

Atom	Radius	Total density	mag(x)	mag(y)	mag(z)
1	2.00000	7.451331	-0.000000	0.000277	-0.000000
2	2.00000	7.451331	0.000000	0.000277	-0.000000
3	2.00000	7.451331	-0.000000	0.000277	-0.000000
4	2.00000	7.451331	-0.000000	0.000277	-0.000000
5	2.00000	11.787209	-0.072667	0.067117	2.434474
6	2.00000	11.787209	-0.074021	0.066454	-2.434451
7	2.00000	11.787209	0.072650	0.066488	-2.434491
8	2.00000	11.787209	0.074038	0.067083	2.434433
9	2.00000	6.291359	0.000000	0.001070	-0.000000
10	2.00000	6.291361	-0.000000	0.001070	-0.000000
11	2.00000	6.291357	-0.000000	0.001070	-0.000000
12	2.00000	6.291359	0.000000	0.001070	0.000000
13	2.00000	6.247049	0.001146	0.001165	-0.000078
14	2.00000	6.247047	0.001146	0.001165	0.000077
15	2.00000	6.247048	-0.001146	0.001165	-0.000077
16	2.00000	6.247049	-0.001146	0.001165	0.000077
17	2.00000	6.247047	0.001146	0.001165	-0.000078
18	2.00000	6.247049	0.001146	0.001165	0.000077
19	2.00000	6.247048	-0.001146	0.001165	-0.000077
20	2.00000	6.247047	-0.001145	0.001165	0.000077
-----					
Total magnetization (spheres)			-0.000001	0.281846	-0.000037
Total magnetization (exact)			-0.000001	0.309096	-0.000041

$\text{YCrO}_3$  spins along the z-axis (AFM G-type); wFM canting along the y axis; wAFM canting along x. [Spin-Orbit coupling necessary to describe the canting and easy-axis](#)

# Non-collinear case $\text{YCrO}_3$ (from spinat along $x/y$ )

starting with spinat along  $x$  direction:

Atom	Radius	Total density	mag(x)	mag(y)	mag(z)
5	2.00000	11.787209	2.434622	0.061122	0.072067
6	2.00000	11.787209	-2.434625	-0.061157	0.071949
7	2.00000	11.787209	-2.434653	0.059917	-0.072040
8	2.00000	11.787209	2.434656	-0.059960	-0.071924
-----					
Total magnetization (spheres)			-0.000000	-0.000084	0.000057
Total magnetization (exact)			-0.000000	-0.000092	0.000062

starting with spinat along  $y$  direction:

Atom	Radius	Total density	mag(x)	mag(y)	mag(z)
5	2.00000	11.787205	-0.065491	2.434621	-0.068435
6	2.00000	11.787205	0.065466	-2.434589	-0.069587
7	2.00000	11.787205	-0.065497	-2.434588	-0.069588
8	2.00000	11.787205	0.065542	2.434620	-0.068437
-----					
Total magnetization (spheres)			0.000022	0.000067	-0.291274
Total magnetization (exact)			0.000024	0.000073	-0.319436

Magnetocrystalline anisotropy (MCA) energy can be calculated:

spin direction	Energy ( $\mu\text{eV}$ )
001	0
010	99
100	199

# Convergence tips

In case of convergence troubles for magnetic systems:

- *diemixmag* flag can be reduced more than *diemix* (could be down to 0.1 for non-collinear cases).
- Increase *nline*
- Use of *nnscl* < 0 (very useful for non-collinear cases with “noisy” magnetization)
- Use *nnscl* > 1 (in case *nnscl* < 0 appears not sufficient, only for very troublesome cases)
- Using metallic occupation (*occopt*=3,7) and a smearing (*tsmear*) could help to converge some systems even if not metallic.
- It is often necessary to start with an initial moment amplitude (*spinat*) larger than the physical one (also useful to target high/low spin configurations).
- GGA is often more “noisy” than LDA...



# Constrained magnetic moment calculations

Constrain the direction of the magnetic moments (*magconon* flag =1) :

$$E = E_{KS} + \sum_i \lambda \left[ \mathbf{m}_i - \mathbf{m}_i^0 (\mathbf{m}_i^0 \cdot \mathbf{m}_i) \right]^2$$

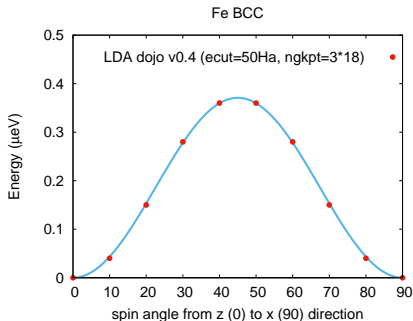
Constrain the direction and the amplitude of the magnetic moments (*magconon* flag =2) :

$$E = E_{KS} + \sum_i \lambda \left[ \mathbf{m}_i - \mathbf{m}_i^0 \right]^2$$

with  $\lambda$  the strength of the Lagrange multiplier (*magcon\_lambda* flag) and  $\mathbf{m}_i^0$  the desired magnetic moment on each atom  $i$  (given by *spinat*).

# Constrained magnetic moment calculations

Useful to explore the spin energy landscape (direction constraint):



```
diemixmag 0.6
diemix 0.7
nline 8
magconon 1
magcon_lambda 0.001
ndtset 10
spinat1 0.0000000 0.0 3.0000000
spinat2 0.5209445 0.0 2.9544233
spinat3 1.0260604 0.0 2.8190779
spinat4 1.5000000 0.0 2.5980762
spinat5 1.9283628 0.0 2.2981333
spinat6 2.2981333 0.0 1.9283628
spinat7 2.5980762 0.0 1.5000000
spinat8 2.8190779 0.0 1.0260604
spinat9 2.9544233 0.0 0.5209445
spinat10 3.0000000 0.0 0.0000000
```

And also to compute the magnetocrystalline anisotropy energy.

(For multiple local minima of complex better to use meta-heuristic approach, see J. Chem. Theory Comput. 14, 4455 (2018))

Can be useful to control high/low spin configurations (amplitude constraint).

# Applied magnetic field

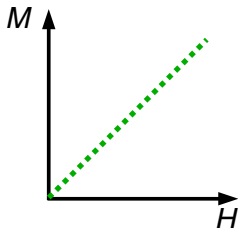
Applying a (Zeeman) magnetic field on the spins gives (*zeemanfield* flag):

$$V_H = -\mu_B\mu_0 \begin{pmatrix} H_z & H_x + iH_y \\ H_x - iH_y & -H_z \end{pmatrix}$$

Allows to access to:

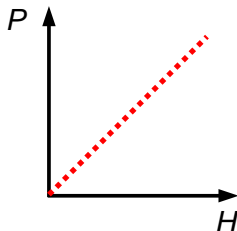
Linear magnetic susceptibility tensor

$$\mathbf{M} = \chi\mathbf{H}$$



Linear magnetoelectric tensor

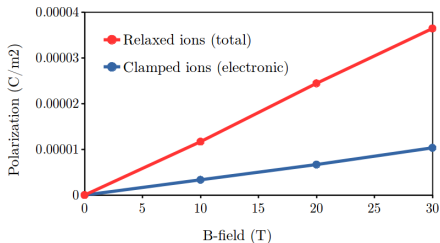
$$\mathbf{P} = \alpha\mathbf{H}$$



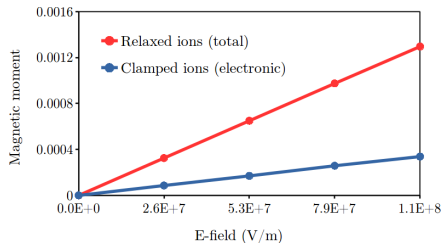
As well as non-linear responses!

# Example: Magnetolectric response of $\text{Cr}_2\text{O}_3$

Applied B-field induces Polarization vs applied E-field induces Magnetization:



$$\alpha_{elec}^{(B)} = 0.36 \text{ ps/m}; \alpha_{tot}^{(B)} = 1.48 \text{ ps/m}$$



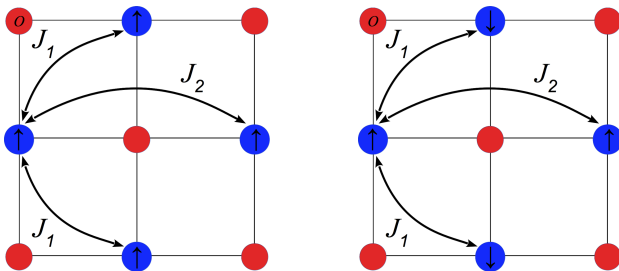
$$\alpha_{elec}^{(E)} = 0.39 \text{ ps/m}; \alpha_{tot}^{(E)} = 1.48 \text{ ps/m}$$

Perfect agreement between E-field and B-field responses (as it should be!).

Good agreement with experiments:  $exp \simeq 1.6 \text{ ps.m}^{-1}$

# Calculation of superexchange constants

Superexchange constants  $J$  can be estimated. Ex: Rock-Salt oxides



One needs to compute the energy for ferro and antiferro in order to extract  $J_1$  and  $J_2$  from:

$$E = E_0 + S \sum_i J_i S_i$$

Ex. for NiO:  $J_1 \simeq 17$  meV and  $J_2 \simeq -0.4$  meV (exp:  $J_1 \simeq 19$  meV)

PRB 84, p.224429 (2011), PRB 86, 094413 (2012)

# Calculation of superexchanges

Superexchange constants  $J$  can also be estimated through Wannier→Green's functions (PRB 91, 224405 (2015)):

$$J_{ij}^{kk'} = -\frac{1}{2\pi} \int_{-\infty}^{E_F} \sum_{kk'} \text{Im}(\tilde{\Delta}_i^{kk} \tilde{G}_{ij,\downarrow}^{kk'} \tilde{\Delta}_j^{k'k'} \tilde{G}_{ji,\uparrow}^{k'k})$$

This has been implemented in the TB2J script (He Xu), see doc and tuto at [gitlab.abinit.org/xuhe/TB2J](http://gitlab.abinit.org/xuhe/TB2J) (still under dev. but can be used in production to compute J).

Output example for SrMnO<sub>3</sub>:

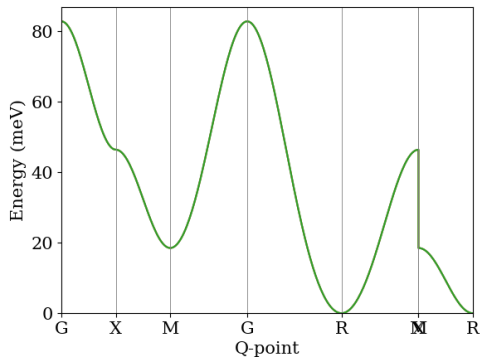
Exchange:

```
i=0(Mn1), j=0(Mn1), R=(-1, 0, 0), J=-6.9178 meV, vec=(-3.810, 0.000, 0.000), distance=3.810
i=0(Mn1), j=0(Mn1), R=( 0,-1, 0), J=-6.9178 meV, vec=( 0.000,-3.810, 0.000), distance=3.810
i=0(Mn1), j=0(Mn1), R=( 0, 1, 0), J=-6.9178 meV, vec=( 0.000, 3.810, 0.000), distance=3.810
i=0(Mn1), j=0(Mn1), R=( 1, 0, 0), J=-6.9178 meV, vec=( 3.810, 0.000, 0.000), distance=3.810
i=0(Mn1), j=0(Mn1), R=( 0, 0, 1), J=-6.8353 meV, vec=( 0.000, 0.000, 3.810), distance=3.810
i=0(Mn1), j=0(Mn1), R=( 0, 0,-1), J=-6.8353 meV, vec=( 0.000, 0.000,-3.810), distance=3.810
i=0(Mn1), j=0(Mn1), R=(-1, 0, 1), J=-0.5538 meV, vec=(-3.810, 0.000, 3.810), distance=5.388
i=0(Mn1), j=0(Mn1), R=( 1, 0,-1), J=-0.5538 meV, vec=( 3.810, 0.000,-3.810), distance=5.388
i=0(Mn1), j=0(Mn1), R=( 1, 0, 1), J=-0.5538 meV, vec=( 3.810, 0.000, 3.810), distance=5.388
i=0(Mn1), j=0(Mn1), R=( 0, 1, 1), J=-0.5538 meV, vec=( 0.000, 3.810, 3.810), distance=5.388
i=0(Mn1), j=0(Mn1), R=( 0,-1,-1), J=-0.5538 meV, vec=( 0.000,-3.810,-3.810), distance=5.388
i=0(Mn1), j=0(Mn1), R=( 0,-1, 1), J=-0.5538 meV, vec=( 0.000,-3.810, 3.810), distance=5.388
```

# Calculation of superexchanges

TB2J can output the (static) spin dispersion or magnon (under debug stage).

Example for  $\text{SrMnO}_3$  in its FM phase:



# Conclusions

## DFT + spins:

- Collinear magnetism: easy to handle
- Non-Collinear magnetism: often less easy
- DFT+ $U$  or Hybrid functionals are often necessary for magnetic systems
- Allows to compute:
  - (Super) Exchange interaction between spins ( $J$ ).
  - Spin canting / Dzyaloshinsky-Moriya interaction ( $D$ )
  - Magnetic anisotropy
  - Response under Zeeman field (magnetic and magnetoelectric susceptibilities)

## Other features:

- DFPT with magnetic field (see rfmagn input flag, only NC LDA)
- Spin dynamics with MULTIBINIT (testing level)
- TB2J for  $J$  and DMI (under dev.).
- ...