Issues with simulations of non-collinear magnets in DFT

Eric Bousquet and Nicola Spaldin

Materials Theory, ETH Zürich

eric.bousquet@mat.ethz.ch

5th Abinit Developer Workshop, Han-Sur-Lesse, Belgium, 14/04/2011

UCSB

ETH

Eidgenössische Technische Hochschule Zürich
Swiss Federal Institute of Technology Zurich

DFT and non-collinear magnetism

General density within non collinear magnetism:

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

$n =$ electronic density ; $m =$ magnetization density

Need to take into account the Spin-Orbit coupling.

Potential:

$$V = \begin{pmatrix} V^{\uparrow\uparrow} & V^{\uparrow\downarrow} \\ V^{\downarrow\uparrow} & V^{\downarrow\downarrow} \end{pmatrix}$$

Non-collinear magnetism and projects

- Non-collinear magnets:
  - Spin Canting
  - Spin Spiral
  - ...
- Magnetocrystalline Anisotropy:
  - Magnetostriction
  - Magnetoelastic coef
  - ...
- Response to magnetic/electric field:
  - Magnetic/Electric susceptibility
  - Magnetoelectric response
  - Ferrotoroidal response
- Get these responses with DFPT

Localised orbitals: LSDA+U

In the collinear scheme
LSDA+U Liechtenstein:

$$V^{\sigma}_{LSDA+U} = V^{\sigma}_{LSDA} + V^{\sigma}_{Coulomb} - V^{\sigma}_{double-count}$$

$$V^{\sigma}_{Coulomb} = V_U n^{-\sigma} + (V_U - V_J) n^{\sigma}$$

$$V^{\sigma}_{double-count} = -U\left(N - \frac{1}{2}\right) + \frac{1}{2} J (N^{\sigma} - 1) \quad .$$

(FLL double counting)

LSDA+U and non-collinear magnetism

LSDA+U Liechtenstein:

Diagonal terms:

$$\sigma = \uparrow\uparrow, \downarrow\downarrow \qquad V^{\sigma}_{Coulomb} = V_U n^{-\sigma} + (V_U - V_J) n^{\sigma}$$

$$\sigma = \uparrow\uparrow, \downarrow\downarrow \qquad V^{\sigma}_{double-count} = -U\left(N - \frac{1}{2}\right) + \frac{1}{2} J (N-1) \pm \frac{1}{2} J m_z$$

LSDA+U and non-collinear magnetism

LSDA+U Liechtenstein:

Diagonal terms:

$$\sigma = \uparrow\uparrow, \downarrow\downarrow \qquad V^{\sigma}_{Coulomb} = V_U n^{-\sigma} + (V_U - V_J) n^{\sigma}$$

$$\sigma = \uparrow\uparrow, \downarrow\downarrow \qquad V^{\sigma}_{double-count} = -U\left(N - \frac{1}{2}\right) + \frac{1}{2}J(N-1) \pm \frac{1}{2}Jm_z$$

Off-diagonal terms:

$$\sigma = \uparrow\downarrow, \downarrow\uparrow \qquad V^{\sigma}_{Coulomb} = -V_J n^{\sigma}$$

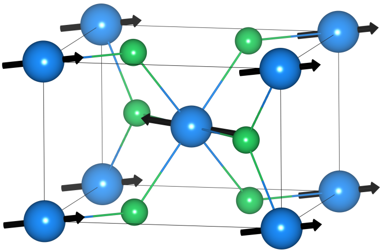
$$\sigma = \uparrow\downarrow, \downarrow\uparrow \qquad V^{\sigma}_{double-count} = \frac{1}{2}J\left(m_x \pm i m_y\right)$$

LSDA+U Dudarev = Liechtenstein with $J = 0$

$\longrightarrow$ No correction for $\sigma = \uparrow\downarrow, \downarrow\uparrow$!

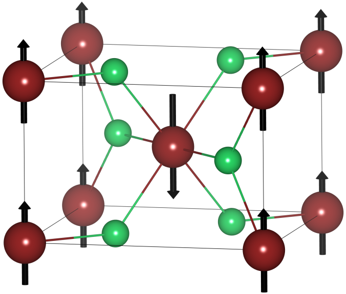
Magnetocrystalline anisotropy: Difluorites

NiF_2



Spins in-plane,
canting out-of-plane

MnF_2, CoF_2, FeF_2

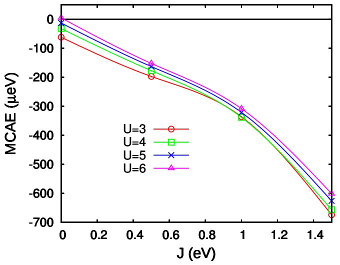


Spins out-of-plane,
no canting

MCAE = E(in-plane) - E(out-of-plane)

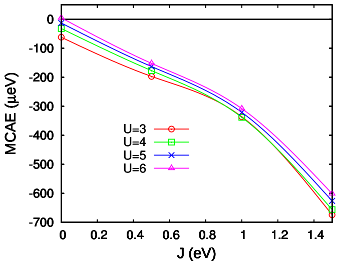
Magnetocrystalline anisotropy: Difluorites

NiF_2

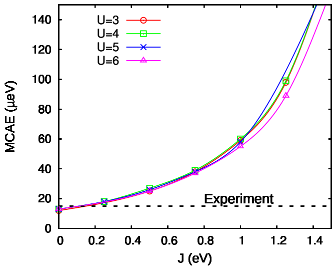


Magnetocrystalline anisotropy: Difluorites

NiF_2

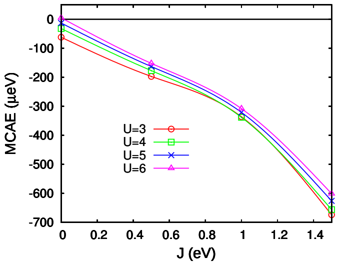


MnF_2

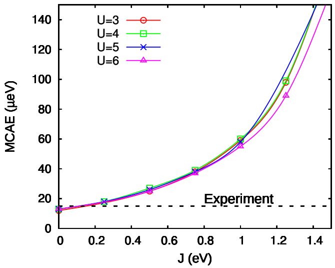


Magnetocrystalline anisotropy: Difluorites

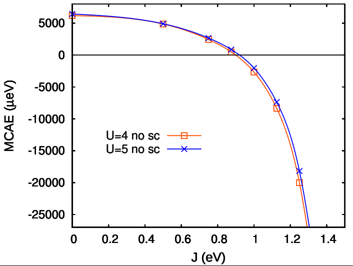
NiF_2



MnF_2

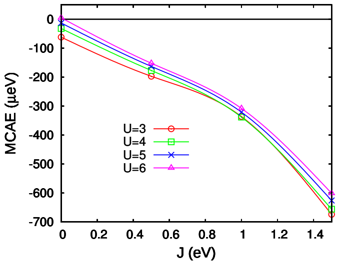


CoF_2

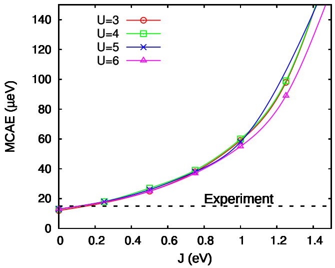


Magnetocrystalline anisotropy: Difluorites

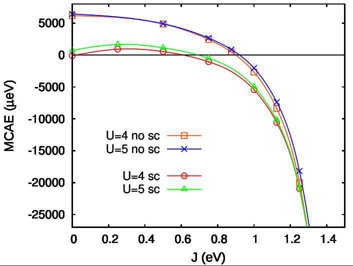
NiF_2



MnF_2

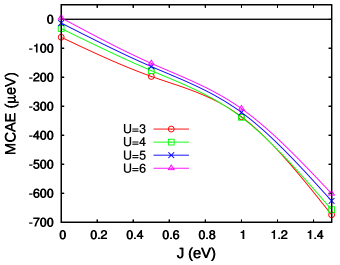


CoF_2

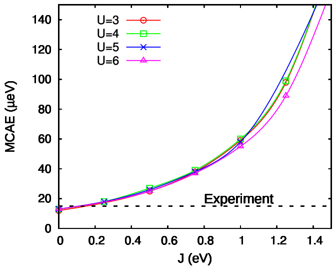


Magnetocrystalline anisotropy: Difluorites

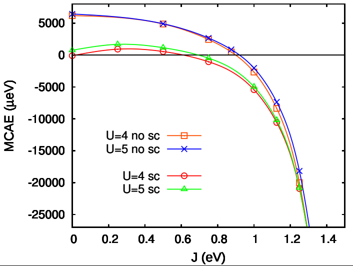
NiF_2



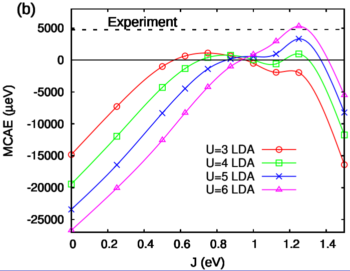
MnF_2



CoF_2

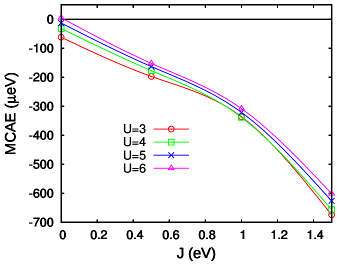


FeF_2

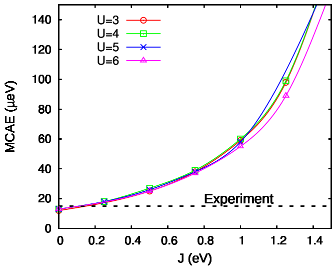


Magnetocrystalline anisotropy: Difluorites

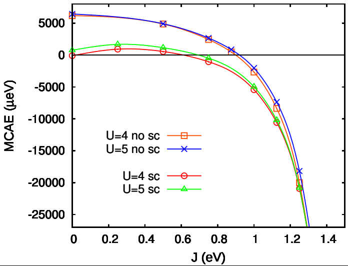
NiF_2



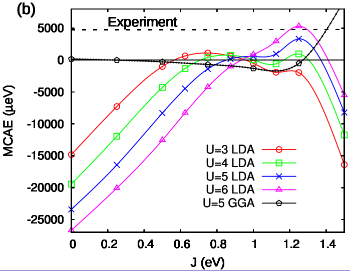
MnF_2



CoF_2

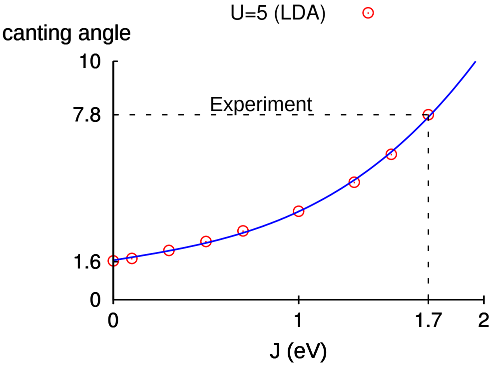


FeF_2



Strong J dependence of the canting angle

- $LiNiPO_4$: Small effect of U but canting angle $\propto J^3$:



Similar J-dependence for other systems with spin canting ($BaNiF_4$, $BiFeO_3$, ...)

Bousquet and Spaldin, PRB 82, 220402(R) (2010)

Origin of the J dependence:

Collinear spins along z:

$$\rho = \begin{pmatrix} n+m_z & 0 \\ 0 & n-m_z \end{pmatrix} \qquad V_{LSDA+U} \equiv \begin{pmatrix} -Un+Jm_z & \times \\ \times & -Un-Jm_z \end{pmatrix}$$

Origin of the J dependence:

Collinear spins along z:

$$\rho = \begin{pmatrix} n+m_z & 0 \\ 0 & n-m_z \end{pmatrix} \qquad V_{LSDA+U} \equiv \begin{pmatrix} -Un+Jm_z & \times \\ \times & -Un-Jm_z \end{pmatrix}$$

Spin component along x (Canting or MCA):

$$\rho = \begin{pmatrix} n+m_z & m_x \\ m_x & n-m_z \end{pmatrix} \qquad V_{LSDA+U} \equiv \begin{pmatrix} -Un+Jm_z & Jm_x \\ Jm_x & -Un-Jm_z \end{pmatrix}$$

J acts directly on m_x

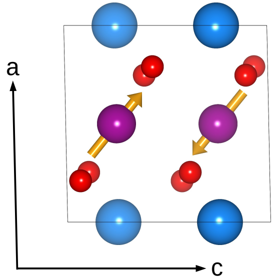
Beyond LSDA+U

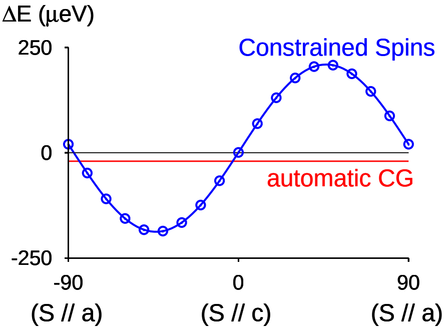
- Problem of predictability of the LSDA+U with non-colinear magnetism
- Particularly large J dependence
- Extremly important for MCAE, magnetoelectric response, magnetostriction, piezomagnetism, ...
- Fine tuning of U and J: Imposible without experimental measurements!
- Solutions:
  - Self consistent U and J ?
  - Hybrids ?
- Challenging case for testing the correctness of new exchange correlation functionals

Technical problem ...

- GS of non-collinear spins: Global minimum with (a lot of) local minima
- Energy differences: 1–100 μeV!

$MnWO_4$





- Clasical Conjugate Gradient: not trustable!

In ABINIT?

$\surd$: Working $\surd$: Problem $\triangleright$: in process $\times$: Not done

- PAW/NCPSP + non-coll + soc: $\surd$ (noise on spin orientation $\triangleright$ XG, MT, EB)
- LDA$+U$: $\surd$ (need the correct double-counting $\triangleright$ BA, EB)
- Constrained magnetic moment: $\triangleright$ (I. Lukacevic, MV, EB)
- Finite magnetic field: $\triangleright$ (K. Delaney, EB)
- Finite electric field: $\surd$; but:
- Berry phase + non-coll + soc: $\surd$
- Alternative Algo for SCF: $\times$?
- DFPT + non-coll + soc: $\times$