Non-collinear magnetism in *Abinit* for Density Functional Perturbation Theory

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Frejus, France
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Density Functional Perturbation Theory

Why?

A general expression for the free energy

$$
\begin{aligned}
-g(\mathbf{E},\mathbf{H},\boldsymbol{\eta}) &= -g_0 + P_i^{(s)} E_i + M_i^{(s)} H_i + \frac{1}{2}\varepsilon_0 \varepsilon_{ik} E_i\, E_k + \frac{1}{2}\mu_0 \mu_{ik} H_i\, H_k + \alpha_{ik} E_i\, H_k + \\
&+ \frac{1}{2}\beta_{ijk} E_i\, H_j\, H_k + \frac{1}{2}\gamma_{ijk} H_i\, E_j\, E_k + \ldots
\end{aligned}
$$

The derivatives at the *first* order

$$
\begin{aligned}
F_k(\mathbf{E},\mathbf{H},\boldsymbol{\eta}) &= \frac{\partial g}{\partial \tau_k}\ ; \quad P_k^{(s)}(\mathbf{E},\mathbf{H},\boldsymbol{\eta}) = \frac{\partial g}{\partial E_k}\ ; \quad k,j = x,y,z \\
M_k^{(s)}(\mathbf{E},\mathbf{H},\boldsymbol{\eta}) &= \frac{\partial g}{\partial H_k}\ ; \quad \sigma_{k,j}(\mathbf{E},\mathbf{H},\boldsymbol{\eta}) = \frac{\partial g}{\partial \eta_{k,j}}
\end{aligned}
$$

Density Functional Perturbation Theory

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A general expression for the free energy

$$-g(\mathbf{E},\mathbf{H},\boldsymbol{\eta}) = -g_0 + P_i^{(s)}E_i + M_i^{(s)}H_i + \frac{1}{2}\varepsilon_0\varepsilon_{ik}E_i\,E_k + \frac{1}{2}\mu_0\mu_{ik}H_i\,H_k + \alpha_{ik}E_i\,H_k + \\ + \frac{1}{2}\beta_{ijk}E_i\,H_j\,H_k + \frac{1}{2}\gamma_{ijk}H_i\,E_j\,E_k + \ldots$$

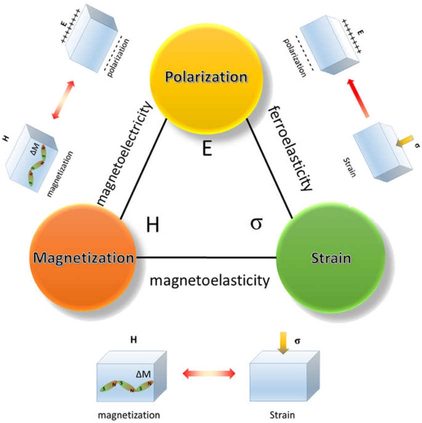
The derivatives at the *second* order

$\frac{\partial^2 g}{\partial \rightarrow \partial \downarrow}$	τ	$\mathbf{E}$	$\boldsymbol{\eta}$	$\mathbf{H}$
τ	IFC	Z^*	γ	Z_M^*
$\mathbf{E}$		ε^∞	e	α^∞
$\boldsymbol{\eta}$			c	Me
$\mathbf{H}$				χ_M

IFC : Interatomic Force Constant
Z^*: Born effective charge
γ: int. strain coupling
Z_M^*: Magnetic effective charge
ε^∞: dielectric constant
e: piezoelectric constant
α^∞: magneto-electric t.
Me: Magnetoelastic constant
χ_M: Magnetic susceptibility

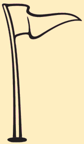
Accounting for Non-collinear Magnetic effects

Why?



Multiferroic Materials

FERROMAGNETIC	FERROELECTRIC
metal	*insulator*



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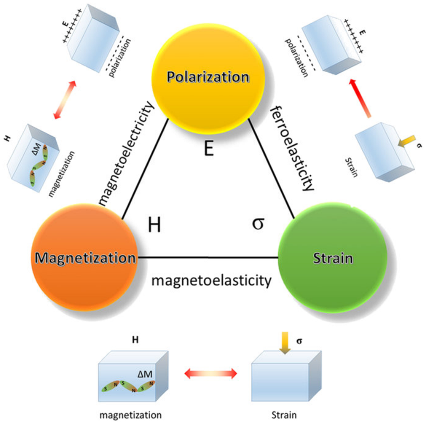
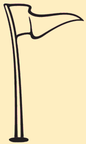


Multiferroic Materials

WEAK CANTING *insulator*

FERROELECTRIC *insulator*



Density matrix $\hat{\rho}$

Non-Collinear magnetism: an undefined global quantisation axis of the magnetisation

Ground-State (0) density matrix in real space

$$\hat{\rho}^{(0)} = |\psi^{(0)}\rangle \langle\psi^{(0)}| \quad \text{in the spin representation}$$

$$\hat{\rho}^{(0)}_{\alpha,\beta} = \langle\alpha|\psi^{(0)}\rangle \langle\psi^{(0)}|\beta\rangle \quad \text{with } |\alpha\rangle, |\beta\rangle = \uparrow, \downarrow$$

$$\rho^{(0)} = \begin{pmatrix} \psi^{(0)}_{\uparrow}\psi^{(0)*}_{\uparrow} & \psi^{(0)}_{\uparrow}\psi^{(0)*}_{\downarrow} \\ \psi^{(0)}_{\downarrow}\psi^{(0)*}_{\uparrow} & \psi^{(0)}_{\downarrow}\psi^{(0)*}_{\downarrow} \end{pmatrix} =$$

$$= \frac{1}{2}\left[\rho\, \delta_{\alpha\beta} + \boldsymbol{m}\cdot\boldsymbol{\sigma}_j\right] = \quad \text{with } j = x, y, z$$

$$= \frac{1}{2}\begin{pmatrix} \rho + m_z & m_x - i\, m_y \\ m_x + i\, m_y & \rho - m_z \end{pmatrix}.$$

Density matrix $\hat{\rho}$

Non-Collinear magnetism: an undefined global quantisation axis of the magnetisation

1^{th} order λ perturbed density matrix in real space

$$\hat{\rho}^{(1)} = \frac{\partial}{\partial\lambda}\left(\left|\psi^{(0)}\right\rangle\left\langle\psi^{(0)}\right|\right) =$$

$$= \left|\psi^{(1)}\right\rangle\left\langle\psi^{(0)}\right| + \left|\psi^{(0)}\right\rangle\left\langle\psi^{(1)}\right| \qquad \text{in the spin representation}$$

$$\hat{\rho}^{(1)}_{\alpha\beta} = \left\langle\alpha|\psi^{(1)}\right\rangle\left\langle\psi^{(0)}|\beta\right\rangle + \left\langle\alpha|\psi^{(0)}\right\rangle\left\langle\psi^{(1)}|\beta\right\rangle \quad \text{with } \left|\alpha\right\rangle, \left|\beta\right\rangle = \uparrow, \downarrow$$

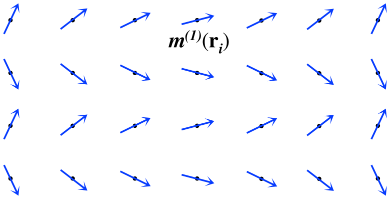
$$\rho^{(1)} = \begin{pmatrix} \psi_{\uparrow}^{(1)*}\psi_{\uparrow}^{(0)} + \psi_{\uparrow}^{(0)*}\psi_{\uparrow}^{(1)} & \psi_{\uparrow}^{(1)}\psi_{\downarrow}^{(0)*} + \psi_{\uparrow}^{(0)}\psi_{\downarrow}^{(1)*} \\ \psi_{\downarrow}^{(1)}\psi_{\uparrow}^{(0)*} + \psi_{\downarrow}^{(0)}\psi_{\uparrow}^{(1)*} & \psi_{\downarrow}^{(1)*}\psi_{\downarrow}^{(0)} + \psi_{\downarrow}^{(0)*}\psi_{\downarrow}^{(1)} \end{pmatrix} =$$

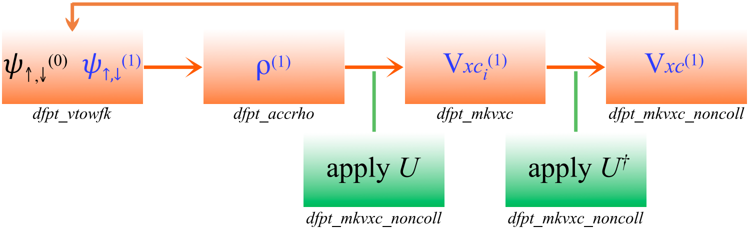
$$= \frac{1}{2}\left[\rho^{(1)}\,\delta_{\alpha\beta} + \boldsymbol{m}^{(1)}\cdot\boldsymbol{\sigma}_j\right] = \qquad \text{with } j = x, y, z$$

$$= \frac{1}{2}\begin{pmatrix} \rho^{(1)} + m_z^{(1)} & m_x^{(1)} - i\,m_y^{(1)} \\ m_x^{(1)} + i\,m_y^{(1)} & \rho^{(1)} - m_z^{(1)} \end{pmatrix}.$$

The Abinit DFPT: from the density matrix $\hat{\rho}^{(1)}$ to the *xc*-potential

A local quantisation axis of the magnetisation density





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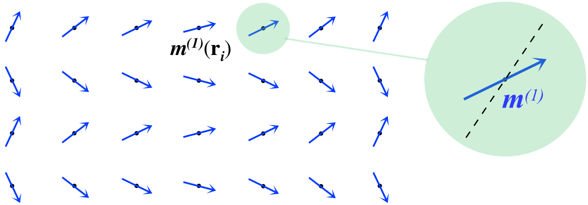

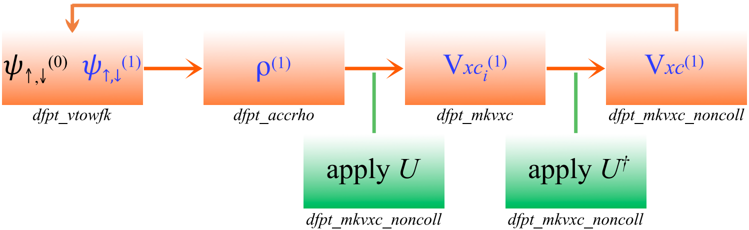

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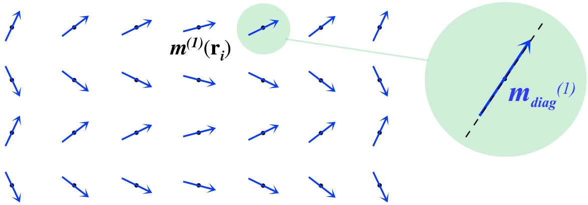

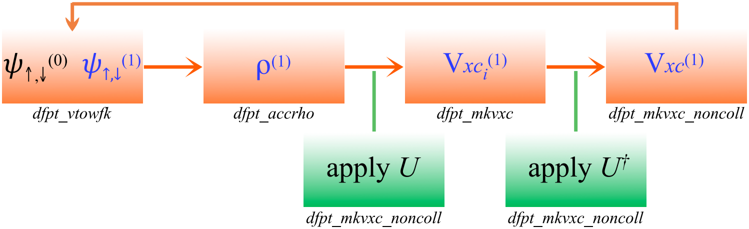

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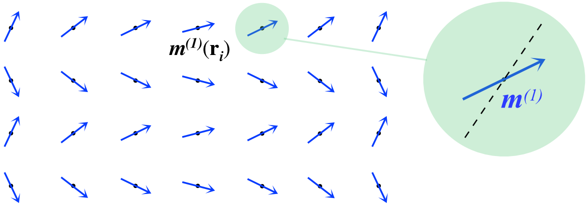

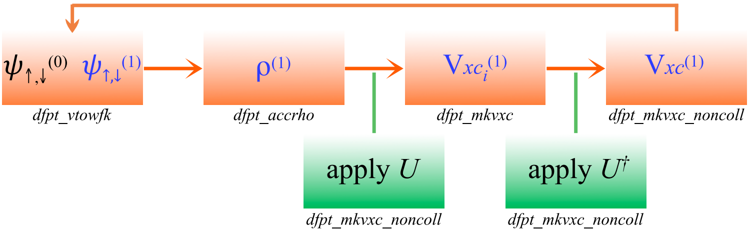

Searching for a local magnetisation quantisation axis

How Abinit locally treats a ground state density in real space

Ground State quantisation axis direction

$$\sum_{\alpha\beta} U^{\dagger(0)}_{i\alpha}\ \rho^{(0)}_{\alpha\beta}\ U^{(0)}_{\beta j} = \rho^{(0)}_i \delta_{ij}$$

$U^{(0)}$ is the spin-$1/2$ rotation matrix

$$n^{(0)}_{\uparrow,\downarrow} = \rho \pm m$$
$$m = \sqrt{m_x^2 + m_y^2 + m_z^2}$$

Searching for a local magnetisation quantisation axis

How Abinit locally treats a perturbed density in real space

1^{th} order quantisation axis direction

$$\sum_{\alpha\beta} U^{\dagger}_{i\alpha}\,\rho_{\alpha\beta}\,U_{\beta j} = \rho_i \delta_{ij}$$

$$\sum_{\alpha\beta} \left(U^{\dagger(0)}_{i\alpha} + \lambda U^{\dagger(1)}_{i\alpha}\right)\left(\rho^{(0)}_{\alpha\beta} + \lambda\rho^{(1)}_{\alpha\beta}\right)\left(U^{(0)}_{\beta j} + \lambda U^{(1)}_{\beta j}\right) = \left(\rho^{(0)}_i + \lambda\rho^{(1)}_i\right)\delta_{ij}$$

- unitarity of $U^{(0)}$
- unitarity of $\left(U^{(0)} + \lambda U^{(1)}\right)$
- analogous GS equation
- neglecting higher order terms

$$\sum_{\alpha} U^{\dagger(1)}_{i\alpha} U^{(0)}_{\alpha j}\left(\rho^{(0)}_j - \rho^{(0)}_i\right) + \sum_{\alpha\beta} U^{\dagger(0)}_{i\alpha}\rho^{(1)}_{\alpha\beta}U^{(0)}_{\beta j} = \rho^{(1)}_i\delta_{ij}$$

$$\begin{pmatrix} 0 & \triangle \\ \triangle^* & 0 \end{pmatrix} + \begin{pmatrix} \boxdot & -\triangle \\ -\triangle^* & \boxdot \end{pmatrix} = \begin{pmatrix} \boxdot & 0 \\ 0 & \boxdot \end{pmatrix}$$

Searching for a local magnetisation quantisation axis

1^{th} order quantisation axis direction

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Estimation of the 1^{th} order xchange-correlation potential

A(n) (in)complete tranformation

Recovering the original direction on the local xc-potential

$$\begin{pmatrix} 0 & \triangle \\ \triangle^* & 0 \end{pmatrix} + \begin{pmatrix} \boxdot & -\triangle \\ -\triangle^* & \boxdot \end{pmatrix} = \begin{pmatrix} \boxdot & 0 \\ 0 & \boxdot \end{pmatrix}$$

$$V_{xc}^{\alpha\beta}\left(\rho_{\alpha\beta}^{(0)} + \lambda\rho_{\alpha\beta}^{(1)}\right) = \sum_i \left(U_{\alpha i}^{(0)} + \lambda U_{\alpha i}^{(1)}\right)\left(V_i^{(0)} + \lambda V_i^{(1)}\right)\left(U_{i\beta}^{\dagger(0)} + \lambda U_{i\beta}^{\dagger(1)}\right)$$

$$V_{xc}^{\alpha\beta(1)} = \sum_i \left[U_{\alpha i}^{(0)} V_i^{(0)} U_{i\beta}^{(1)\dagger} + U_{\alpha i}^{(1)} V_i^{(0)} U_{i\beta}^{(0)\dagger} + U_{\alpha i}^{(0)} V_i^{(1)} U_{i\beta}^{(0)\dagger}\right]$$

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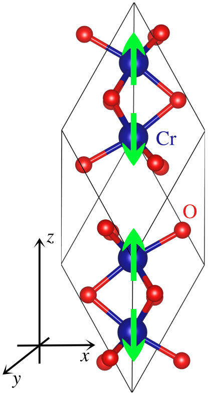
As a first approximation we used just the last term

Application on Cr_2O_3

A collinear antiferromagnet as a test case for non-collinear DFPT



		FROZEN		TOTAL	
		FD	DFPT	FD	DFPT
x, y, z	$\mathbf{m} \parallel z$	23201.036772	23204.0723534	7.71138685414	7.711369006
	$\mathbf{m} \parallel x$	23201.036746	23204.0723507	7.71138847221	7.779709388

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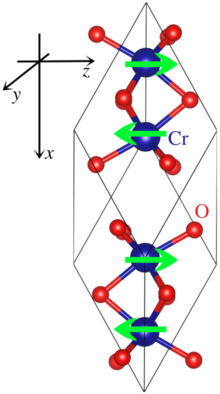

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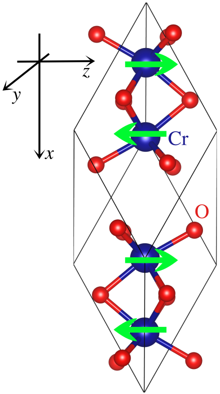

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Correctly working for diagonal density matrices!

$$\begin{pmatrix} 0 & \triangle \\ \triangle^* & 0 \end{pmatrix} + \begin{pmatrix} \boxdot & -\triangle \\ -\triangle^* & \boxdot \end{pmatrix} = \begin{pmatrix} \boxdot & 0 \\ 0 & \boxdot \end{pmatrix}$$

The $U^{(0)}$ matrix gives an incomplete estimation of the off-diagonal elements

Estimation of the 1^{th} order xchange-correlation potential

A complete tranformation

We need $U^{(1)}$ to correctly obtain the $V_{xc}^{(1)}$ off-diagonal terms

Local density matrix diagonalisation:

$$\sum_{\alpha} U_{i\alpha}^{\dagger(1)} U_{\alpha j}^{(0)} \left(\rho_j^{(0)} - \rho_i^{(0)}\right) + \sum_{\alpha\beta} U_{i\alpha}^{\dagger(0)} \rho_{\alpha\beta}^{(1)} U_{\beta j}^{(0)} = \rho_i^{(1)} \delta_{ij}$$

Local non-collinear *xc*-potential:

$$V_{xc}^{\alpha\beta(1)} = \sum_i \left[U_{\alpha i}^{(0)} V_i^{(0)} U_{i\beta}^{(1)\dagger} + U_{\alpha i}^{(1)} V_i^{(0)} U_{i\beta}^{(0)\dagger} + U_{\alpha i}^{(0)} V_i^{(1)} U_{i\beta}^{(0)\dagger} \right]$$

Conclusions

1. GS and perturbed density matrix formalisms.
2. The way Abinit handles the non-collinear density-to-*xc*-potential calculation.
3. The locally collinear approximation works (untill now) for systems with collinear magnetic moments (along z).
4. We are working to get the full non-collinear *xc*-potential estimating explicitly the effect of the $U^{(1)}$.

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Perspectives

1. Extend the formalism for $\boldsymbol{q} \neq 0$.
2. SOC?
3. Extend the formalism for PAW.
4. Full non-collinear xc functional?
5. Perturbation with magnetic field.

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Next talk: application of a Zeeman magnetic field in DFPT

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