DFPT with magnetic fields and noncollinear XC functionals

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Spin susceptibility

(a) General definition

We are considering **only the spin response** (no orbital magnetization)

$$\delta m_\alpha(r,t) = \int_V d^3 r_p \int_{-\infty}^{\infty} dt_p \; \chi_{\alpha\beta}(r,t,r_p,t_p)\delta B_\beta(r_p,t_p)$$

Magnetization density

Perturbation

Susceptibility tensor

(b) Why to compute it?

- For small q and ω it gives the excitation spectrum … magnons, spin (charge) density waves, etc.

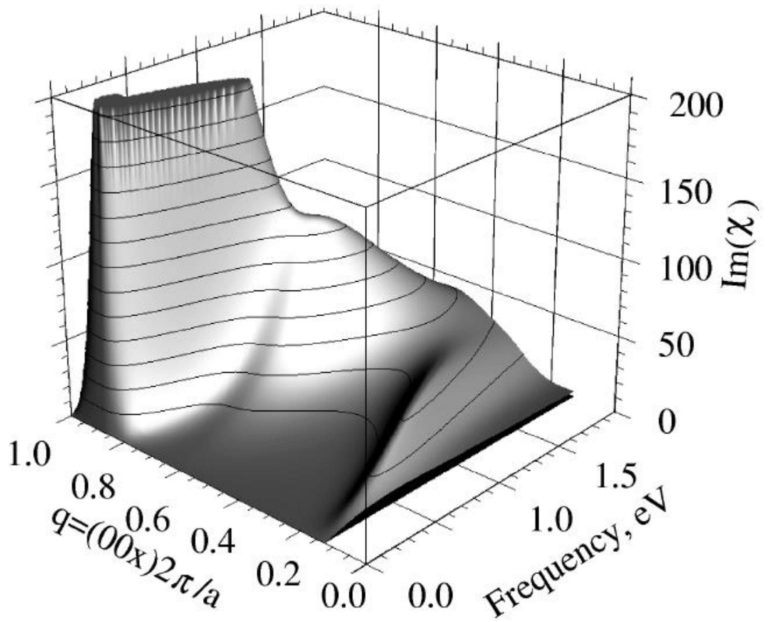
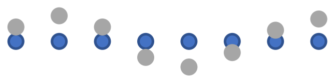


FIG. 3. Calculated Im[$\chi(\mathbf{q}, \omega)$] (Ry^{-1}) for Cr.

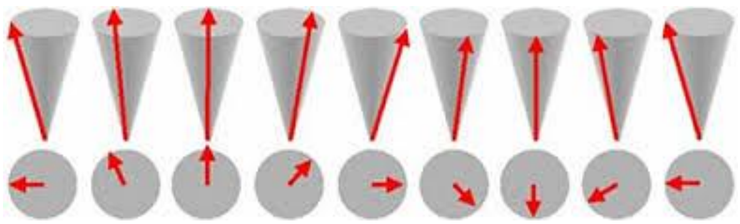
S. Savrasov Phys. Rev. Lett. **81** 2570 (1998)

(b) Why to compute it?

- Second principles (Multibinit!)
- *Fit effective Hamiltonian parameters (Heisenberg Hamiltonian etc.)*



Phonons

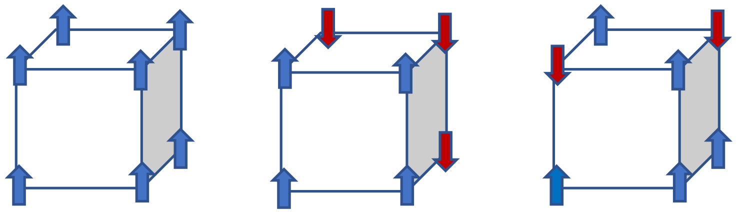


Spin waves

(b) Why to compute it?

- Second principles (Multibinit!)
- *Fit effective Hamiltonian parameters (Heisenberg Hamiltonian etc.)*

Frozen magnon



SSSDW DFT

$$\exp(i\,\mathbf{k}\cdot\mathbf{r}) \begin{pmatrix} \exp(-i\,\frac{1}{2}\,\mathbf{q}\cdot\mathbf{r})\; u^{(\uparrow)}_{\mathbf{k},\nu}(\mathbf{r}) \\ \exp(+i\,\frac{1}{2}\,\mathbf{q}\cdot\mathbf{r})\; u^{(\downarrow)}_{\mathbf{k},\nu}(\mathbf{r}) \end{pmatrix}$$

Phys. Rev. B 88, 134427 (2013)

DFPT

$$\hat{J}_{\alpha\beta}(q,\omega) = \hat{\chi}^{-1}_{\alpha\beta}(q,\omega)$$

Spin susceptibility

(c) How to compute from perturbation theory?

- Direct TD-DFT $$i\frac{\partial}{\partial t}\varphi_k(\boldsymbol{r},t) = \hat{H}_{\mathrm{KS}}[n](\boldsymbol{r},t)\varphi_k(\boldsymbol{r},t)$$
- Sternheimer/Variational $$i\frac{\partial}{\partial t}\varphi_k^{(1)}(\boldsymbol{r},t) = \hat{H}_{\mathrm{KS}}^{(0)}[n^{(0)}](\boldsymbol{r})\varphi_k^{(1)}(\boldsymbol{r},t) + \left[\hat{H}_{\mathrm{KS}}^{(1)}[n](\boldsymbol{r},t) + v_{\mathrm{ext}}^{(1)}(\boldsymbol{r},t)\right]\varphi_k^{(0)}(\boldsymbol{r},t)$$
- Sum over states $$\varphi_{k,\omega}^{(1)} = \sum_{m\neq k} |\varphi_m^{(0)}\rangle \frac{\langle\varphi_m^{(0)}|\hat{H}_\omega^{(1)}|\varphi_k^{(0)}\rangle}{\varepsilon_m^{(0)} - \varepsilon_k^{(0)} + \omega}$$

(c) How to compute?

- Sternheimer/Variational

$$i\frac{\partial}{\partial t}\varphi_k^{(1)}(\boldsymbol{r},t) = \hat{H}_{\mathrm{KS}}^{(0)}[n^{(0)}](\boldsymbol{r})\varphi_k^{(1)}(\boldsymbol{r},t) + \left[\hat{H}_{\mathrm{KS}}^{(1)}[n](\boldsymbol{r},t) + v_{\mathrm{ext}}^{(1)}(\boldsymbol{r},t)\right]\varphi_k^{(0)}(\boldsymbol{r},t)$$

Conceptually simple + already implemented in Abinit
TD-DFPT deneralization
(Variation of the action "*S*" functional)

(d) Bibliography

Reviews

- “Aspects of Time-Dependent Perturbation Theory” *Rev. Mod. Phys.* P.W. Langhoff et al. (1972)
- “A unified formulation of the construction of variational principles” *Rev. Mod. Phys.* Gerjuoy et al. (1983)
- “Time-Dependent Density Functional Theory ” M.A.L. Marques et al. Lect. Notes Phys. 706 (Springer, 2006)

Implementations and theory

- K. L. Liu, S. H. Vosko, Can. J. Phys. 67, 1015 (1989)
- S. Y. Savrasov, Phys. Rev. Lett. 81, 2570 (1998)
- G. Vignale, Phys. Rev. A. 77, 062611 (2008)
- R. Requist, O. Pankratov, Phys. Rev. A 79, 032502 (2009)

(d) Bibliography

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- "Time-Dependent Density Functional Theory " M.A.L. Marques et al. Lect. Notes Phys. 706 (Springer, 2006)

Implementations and theory (spin response)

- K. L. Liu, S. H. Vosko, Can. J. Phys. 67, 1015 (1989) → Paramagnetic case, HEG, transverse spin susc.
- S. Y. Savrasov, Phys. Rev. Lett. 81, 2570 (1998) → "it is easy to show" style, implementation
- G. Vignale, Phys. Rev. A. 77, 062611 (2008) → An easier causality problem resolution
- R. Requist, O. Pankratov, Phys. Rev. A 79, 032502 (2009) → TD-DFPT

(d) Bibliography

- K. L. Liu, S. H. Vosko, Can. J. Phys. 67, 1015 (1989)
- S. Y. Savrasov, Phys. Rev. Lett. 81, 2570 (1998)

Paramagnetic case, HEG, transverse spin susc.

"it is easy to show" style, LMTO implementation

$$A_U[n,m_z] = G[n,m_z] + \frac{1}{2}\int_{t_0}^{t_1} d\tau \int d\mathbf{r}_1\, d\mathbf{r}_2\, n(\mathbf{r}_1\tau) \times u(\mathbf{r}_1 - \mathbf{r}_2) n(\mathbf{r}_2\tau) + \int_{t_0}^{t_1} d\tau \int d\mathbf{r}\, v_0(\mathbf{r}) n(\mathbf{r}\tau) - \int_{t_0}^{t_1} d\tau \int d\mathbf{r}\, B(\mathbf{r}\tau) m_z(\mathbf{r}\tau)$$

$$\int_{t_0}^{t_1} d\tau \langle \psi[n,m_z;\tau] | \hat{T} - i(\partial/\partial\tau) | \psi[n,m_z;\tau] \rangle + A_{XC}[n,m_z]$$

Action is stationary → 2^{nd} order action is stationary w.r.t 1^{st} order density

Time dependent variational principle

(d) Bibliography

- K. L. Liu, S. H. Vosko, Can. J. Phys. 67, 1015 (1989)
- S. Y. Savrasov, Phys. Rev. Lett. 81, 2570 (1998)

Action is stationary

2nd order action is stationary w.r.t
1st order density

~~action~~ quasienergy

Causality: which functional is stationary?!

e.g. G. Vignale, Phys. Rev. A. 77, 062611 (2008)

Time dependent response to AR perturbation

R. Requist, O. Pankratov, Phys. Rev. A 79, 032502 (2009)

Time dependent variational principle

(d) Bibliography

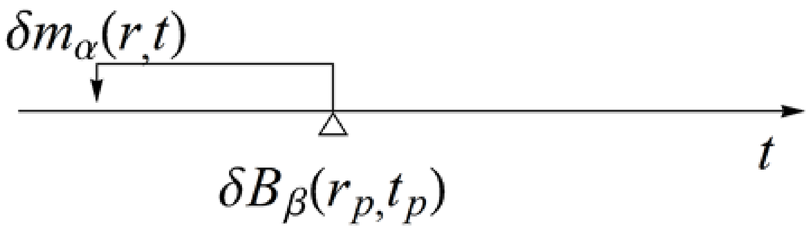
Time translational invariance

$$\chi_{\alpha,\beta}(r,t,r_p,t_p) = \Theta(t-t_p)\chi_{\alpha,\beta}(r,r_p,t-t_p)$$

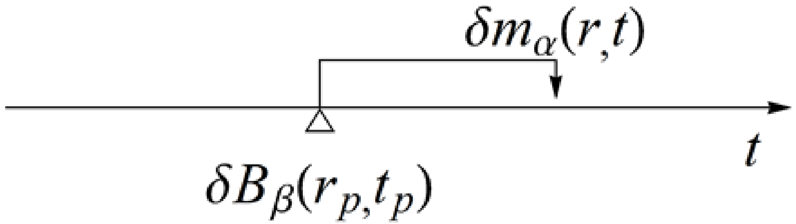
Heaviside

"causal loop"

"grandfather paradox"



Causal response



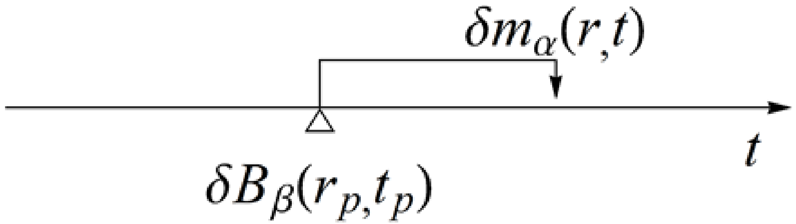
(d) Bibliography

Time translational invariance

$$\chi_{\alpha,\beta}(r,t,r_p,t_p) = \Theta(t-t_p)\chi_{\alpha,\beta}(r,r_p,t-t_p)$$

Heaviside

Causal response



(d) Bibliography

$$v[n;\mathbf{r},t] \equiv \frac{\delta A[n]}{\delta n(\mathbf{r},t)} \quad \Rightarrow \quad \frac{\delta v[n;\mathbf{r},t]}{\delta n(\mathbf{r}',t')} = \frac{\delta^2 A[n]}{\delta n(\mathbf{r},t)\,\delta n(\mathbf{r}',t')} \quad \Rightarrow \quad ?$$

(d) Bibliography

$$v[n;\mathbf{r},t] \equiv \frac{\delta A[n]}{\delta n(\mathbf{r},t)}$$

$$\frac{\delta v[n;\mathbf{r},t]}{\delta n(\mathbf{r}',t')} = \frac{\delta^2 A[n]}{\delta n(\mathbf{r},t)\,\delta n(\mathbf{r}',t')}$$

all OK for Adiabatically ramped harmonic perturbations

Physical meaning of the stationary functional changes

(d) Bibliography

R. Requist, O. Pankratov, Phys. Rev. A 79, 032502 (2009)

$$\Psi(t) = \xi_\tau(t)\exp\left[-i\int_{-\infty}^{t} dt' K_\tau(t')\right] \qquad K_\tau(t) = \frac{\langle \xi_\tau | \hat{H}_\tau(t) - i\partial_t | \xi_\tau \rangle}{\langle \xi_\tau | \xi_\tau \rangle}$$

Secular phase

Assumptions

$$\delta B_\beta(r,t) = (e^{i\omega t + iqr} + c.c.) \sum_G \lambda_G e^{iGr}$$

Incommensurate

Periodic

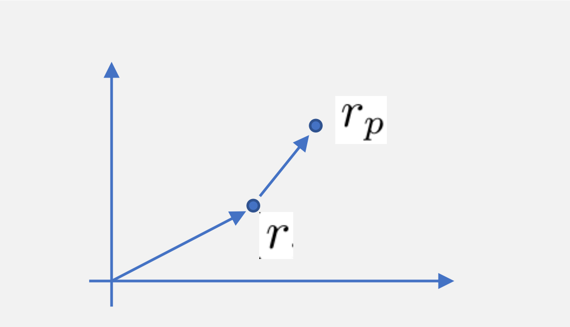
General considerations

$$\delta m_\alpha(r,t) = \int_V d^3 r_p \int_{-\infty}^{\infty} dt_p \Theta(t-t_p)\chi_{\alpha,\beta}(r,r_p,t-t_p)(e^{i\omega t_p + iqr_p} + c.c.)\sum_G \lambda_G e^{iGr_p} =$$

$$= \sqrt{2\pi} e^{i\omega t}\int_V d^3 r_p X_{\alpha\beta}(r,r_p,\omega)e^{iqr_p}\sum_G \lambda_G e^{iGr} + \{\omega \to -\omega, q \to -q\} =$$

$$= (2\pi)^2\left(e^{i\omega t + iqr}\sum_G \lambda_G X_{\alpha\beta}(r,G+q,\omega)e^{iGr} + e^{-i\omega t - iqr}\sum_G \lambda_G X_{\alpha\beta}(r,G-q,\omega)e^{iGr}\right)$$

$$X_{\alpha\beta}(r,r_p,\omega) = X_{\alpha\beta}(r,r-r_p,\omega)$$



General considerations

$$\delta B_\beta(r,t) = (e^{i\omega t + iqr} + c.c.)\sum_G \lambda_G e^{iGr}$$

$\lambda_G = \lambda^*_{-G}$ is not zero for one single G value

$$\delta m_\alpha(r,t) = (2\pi)^2 \left(\lambda_G e^{i\omega t} e^{i(q+G)r} X_{\alpha\beta}(r, G+q, \omega) + \lambda^*_G e^{-i\omega t} e^{-i(q+G)r} X_{\alpha\beta}(r, -G-q, \omega)\right)$$

Spin susceptibility

General considerations

$$\delta B_\beta(r,t) = (e^{i\omega t + iqr} + c.c.)\sum_G \lambda_G e^{iGr}$$

$\lambda_G = \lambda^*_{-G}$ is not zero for one single G value

$$\delta m_\alpha(r,t) = (2\pi)^2 \left(\lambda_G e^{i\omega t} e^{i(q+G)r} X_{\alpha\beta}(r, G+q,\omega) + \lambda^*_G e^{-i\omega t} e^{-i(q+G)r} X_{\alpha\beta}(r,-G-q,\omega)\right)$$

Periodic part

Monochromatic

Incommensurate part

$$\delta m_\alpha(r,t) = e^{i\omega t + iqr} \times (periodic\ function)$$

General considerations

$$\delta m_\alpha(r,t) = e^{i\omega t + iqr} \times (periodic\ function)$$

$$\int d^3r \int dt \left(\delta m_\alpha(r,t) e^{-i\omega t - iqr}\right) e^{-iG'r} = X_{\alpha,\beta}(G' - G, G + q, \omega)$$

General considerations

$$\int d^3r \int dt \left(\delta m_\alpha(r,t) e^{-i\omega t - iqr}\right) e^{-iG'r} = X_{\alpha,\beta}(G' - G, G + q, \omega)$$

$$\int d^3r \int dt \left(\delta m_\alpha(r,t) e^{-i\omega t - iqr}\right) = X_{\alpha,\beta}(G = 0, q, \omega)$$

Euler Lagrange equations

$$\begin{pmatrix} \hat{H}_{\mathrm{KS}}^{(0)} - \varepsilon_k^{(0)} + \omega & 0 \\ 0 & \hat{H}_{\mathrm{KS}}^{(0)} - \varepsilon_k^{(0)} - \omega \end{pmatrix} \begin{pmatrix} \varphi_{k,+\omega}^{(1)} \\ \varphi_{k,-\omega}^{(1)} \end{pmatrix} = - \begin{pmatrix} \left(v_{\mathrm{Hxc},+\omega}^{(1)} + v_{\mathrm{ext},+\omega}^{(1)} - \varepsilon_{k,+\omega}^{(1)} \right) \varphi_k^{(0)} \\ \left(v_{\mathrm{Hxc},-\omega}^{(1)} + v_{\mathrm{ext},-\omega}^{(1)} - \varepsilon_{k,-\omega}^{(1)} \right) \varphi_k^{(0)} \end{pmatrix}$$

Ansatz

$$\varphi(\boldsymbol{r}, t) = \mathrm{e}^{-\mathrm{i}\varepsilon^{(0)} t - \mathrm{i}\lambda \Delta\varepsilon^{(1)}(t)} \times \left\{ \varphi^{(0)}(\boldsymbol{r}) + \lambda \left[\varphi_{+\omega}^{(1)}(\boldsymbol{r}) \mathrm{e}^{\mathrm{i}\omega t} + \varphi_{-\omega}^{(1)}(\boldsymbol{r}) \mathrm{e}^{-\mathrm{i}\omega t} \right] \right\}$$

$$\Delta\varepsilon^{(1)}[n](t) = \int_{-\infty}^{t} \mathrm{d}t \langle \varphi^{(0)} | \hat{H}_{\mathrm{KS}}^{(1)}[n](t) + v_{\mathrm{ext}}^{(1)}(t) | \varphi^{(0)} \rangle .$$

Euler Lagrange equations

$$\begin{pmatrix} \hat{H}_{\mathrm{KS}}^{(0)} - \varepsilon_k^{(0)} + \omega & 0 \\ 0 & \hat{H}_{\mathrm{KS}}^{(0)} - \varepsilon_k^{(0)} - \omega \end{pmatrix} \begin{pmatrix} \varphi_{k,+\omega}^{(1)} \\ \varphi_{k,-\omega}^{(1)} \end{pmatrix} = - \begin{pmatrix} \left(v_{\mathrm{Hxc},+\omega}^{(1)} + v_{\mathrm{ext},+\omega}^{(1)} - \varepsilon_{k,+\omega}^{(1)} \right) \varphi_k^{(0)} \\ \left(v_{\mathrm{Hxc},-\omega}^{(1)} + v_{\mathrm{ext},-\omega}^{(1)} - \varepsilon_{k,-\omega}^{(1)} \right) \varphi_k^{(0)} \end{pmatrix}$$

$\omega + i\eta$ $\qquad$ $\omega - i\eta$

Ansatz

$$\varphi(\boldsymbol{r}, t) = \mathrm{e}^{-\mathrm{i}\varepsilon^{(0)} t - \mathrm{i}\lambda \Delta\varepsilon^{(1)}(t)} \times \left\{ \varphi^{(0)}(\boldsymbol{r}) + \lambda \left[\varphi_{+\omega}^{(1)}(\boldsymbol{r}) \mathrm{e}^{\mathrm{i}\omega t} + \varphi_{-\omega}^{(1)}(\boldsymbol{r}) \mathrm{e}^{-\mathrm{i}\omega t} \right] \right\}$$

$$\Delta\varepsilon^{(1)}[n](t) = \int_{-\infty}^{t} \mathrm{d}t \, \langle \varphi^{(0)} | \hat{H}_{\mathrm{KS}}^{(1)}[n](t) + v_{\mathrm{ext}}^{(1)}(t) | \varphi^{(0)} \rangle.$$

Euler Lagrange equations

$$\begin{pmatrix} \hat{H}_{\mathrm{KS}}^{(0)} - \varepsilon_k^{(0)} + \omega & 0 \\ 0 & \hat{H}_{\mathrm{KS}}^{(0)} - \varepsilon_k^{(0)} - \omega \end{pmatrix} \begin{pmatrix} \varphi_{k,+\omega}^{(1)} \\ \varphi_{k,-\omega}^{(1)} \end{pmatrix} = - \begin{pmatrix} \left(v_{\mathrm{Hxc},+\omega}^{(1)} + v_{\mathrm{ext},+\omega}^{(1)} - \varepsilon_{k,+\omega}^{(1)} \right) \varphi_k^{(0)} \\ \left(v_{\mathrm{Hxc},-\omega}^{(1)} + v_{\mathrm{ext},-\omega}^{(1)} - \varepsilon_{k,-\omega}^{(1)} \right) \varphi_k^{(0)} \end{pmatrix}$$

$\omega + i\eta$ $\omega - i\eta$

- Solution for two different frequencies for each point of (ω, q) grid
- Phase factorization works in the same way as in the static case
- First order external potential is a constant matrix:

$$v_{ext}^{(1)} = \frac{1}{2}\vec{\sigma}$$

Implementation strategy

$$(\omega = 0, q = 0)$$

↓

$$(\omega = 0, q \neq 0)$$

↓

$$(\omega \neq 0, q \neq 0)$$

↓

Physics (+ tests of xc flavors)

Implementation strategy

$$(\omega = 0, q = 0)$$

↓

$$(\omega = 0, q \neq 0)$$

↓

$$(\omega \neq 0, q \neq 0)$$

↓

Physics (+ tests of xc flavors)

Implementation strategy

$$(\omega = 0, q = 0)$$

$$(\omega = 0, q \neq 0)$$

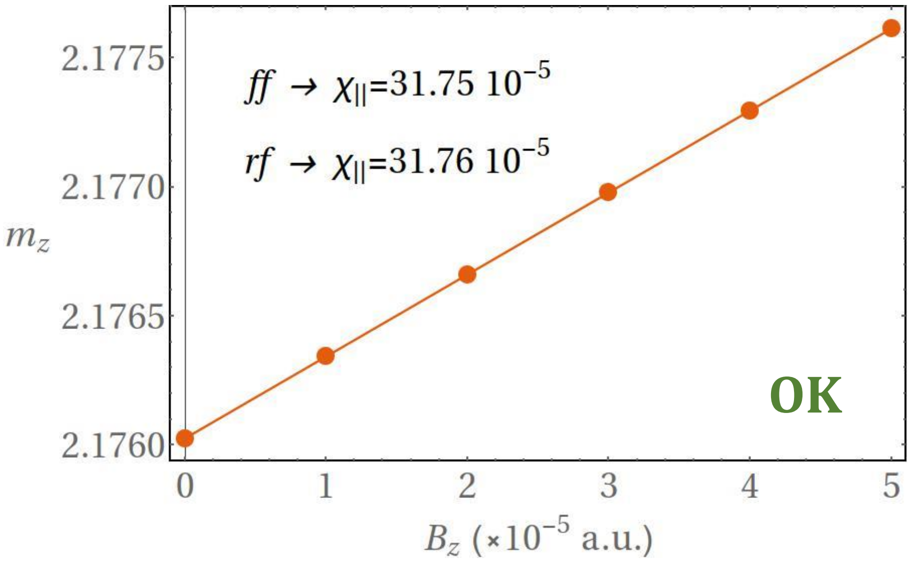
$$(\omega \neq 0, q \neq 0)$$

Physics (+ tests of xc flavors)

- rfmagn = 1

  ipert = natom + 5 rfuser: natom + 6
  natom + 7

- Fe bcc



$(\omega = 0, q = 0)$

↓

$(\omega = 0, q \neq 0)$

↓

$(\omega \neq 0, q \neq 0)$

↓

Physics (+ tests of xc flavors)

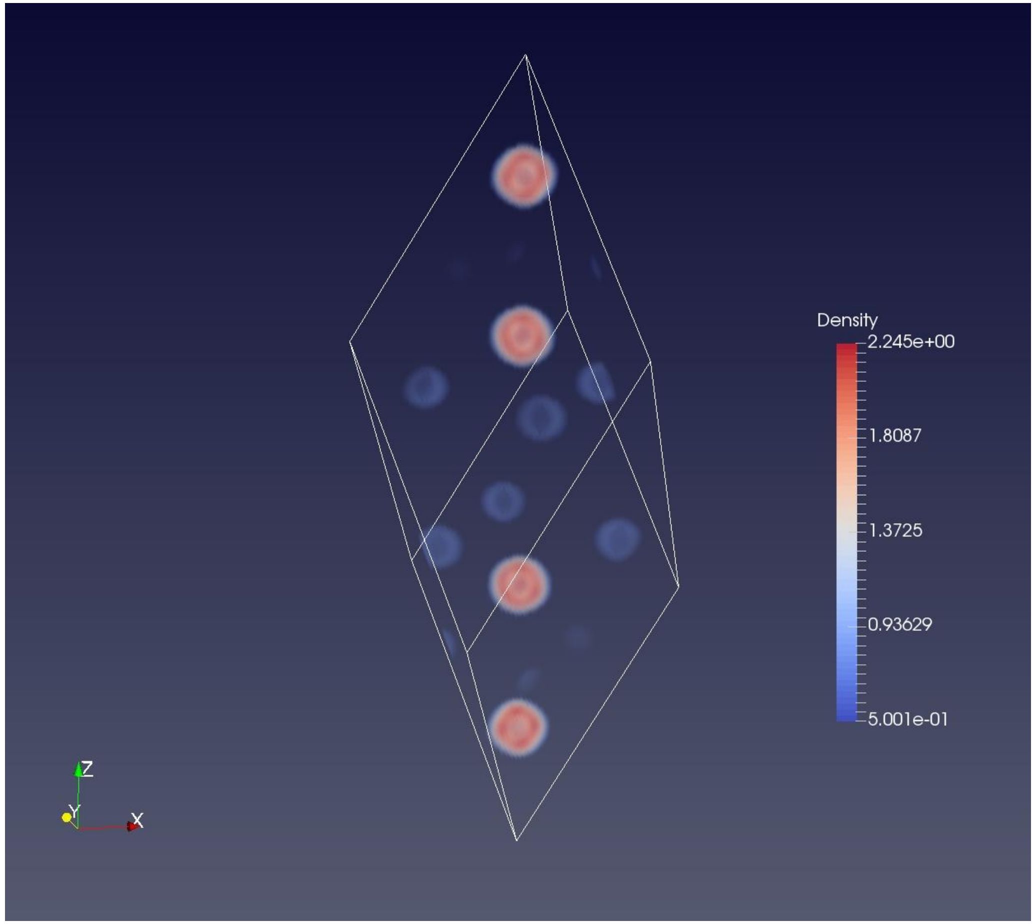
- rfmagn = 1

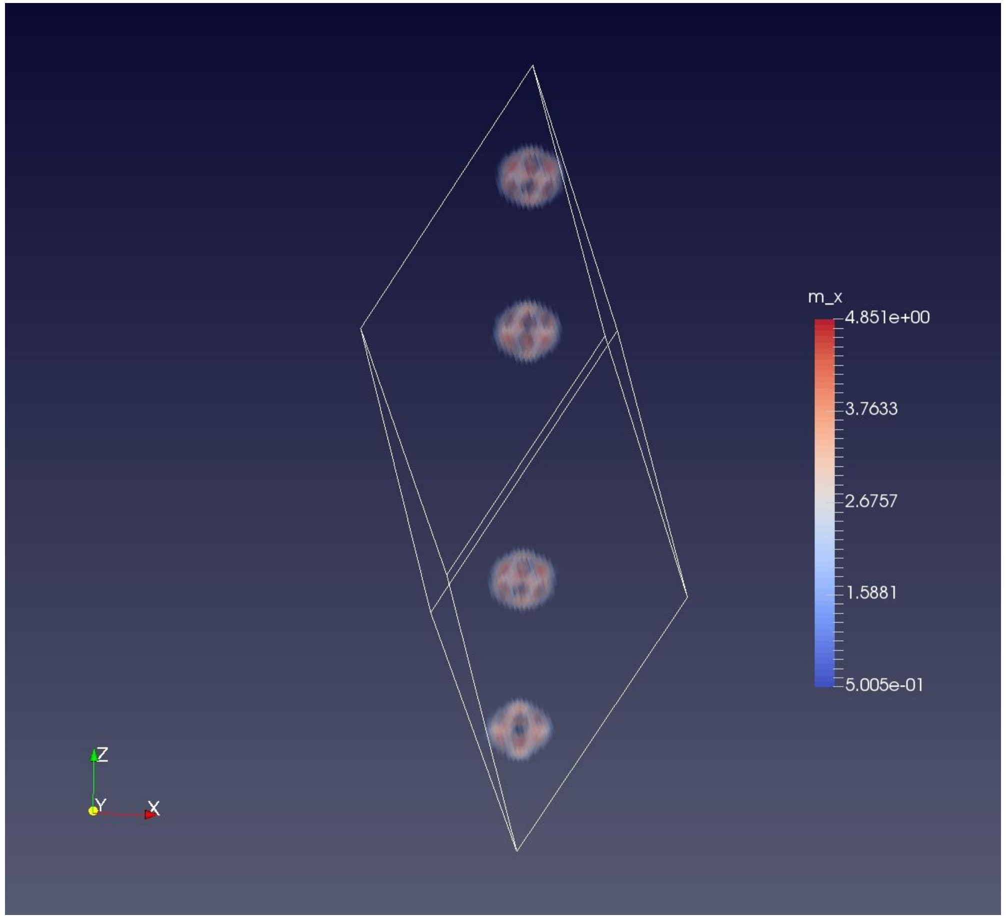
  ipert = natom + 5 rfuser: natom + 6
  natom + 7

- Cr_2O_3 $\chi_{||} = 0$ → **OK**

Implementation strategy

- Cr_2O_3 transverse susceptibility





Implementation strategy

$$(\omega = 0, q = 0)$$

$$(\omega = 0, q \neq 0)$$

$$(\omega \neq 0, q \neq 0)$$

Physics (+ tests of xc flavors)

- rfmagn = 1

  ipert = natom + 5   rfuser: natom + 6
  natom + 7

- NC + only longitudinal response is OK

- Many things can be probed even in this simplest case

  Dynamic magnetic charges (spin part)

  Phonon induced first order magnetization

$(\omega = 0, q = 0)$

↓

$(\omega = 0, q \neq 0)$

↓

$(\omega \neq 0, q \neq 0)$

↓

Physics (+ tests of xc flavors)

Many people believe that if it ain't broke, don't fix it...

... if it ain't broke, it doesn't have enough features yet

$(\omega = 0, q = 0)$

↓

$(\omega = 0, q \neq 0)$

↓

$(\omega \neq 0, q \neq 0)$

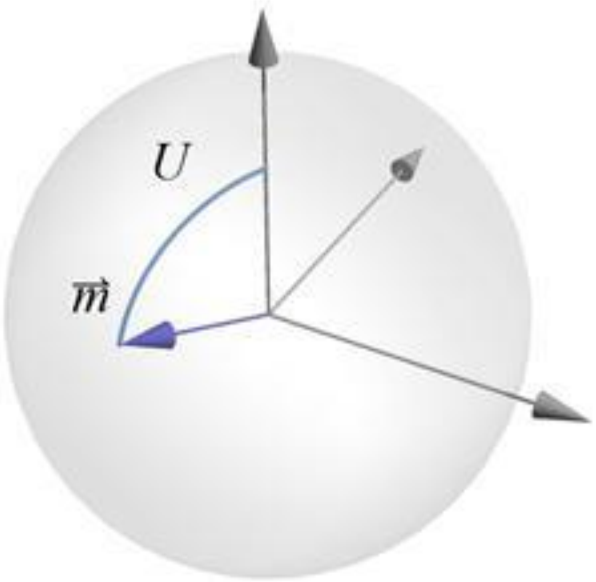
↓

Physics (+ tests of xc flavors)

PAW?

NC LDA+U?

LSDA/GGA



Kettle principle:

use collinear xc functionals

$$U_{\alpha\beta} \sim 1/|m|$$

$$v_{xc} = v_x c[|m| = 0] + |m| \frac{dv_{xc}}{dm}$$

$$\{n, \vec{m}\} \rightarrow \{n_{\pm}, \gamma_{\pm}, \gamma_{\text{mix}}, \tau_{\pm}, \nabla^2 n_{\pm}\}$$

I. W. Bulik et al. Phys. Rev. B 87, 035117 (2013)

Other approaches

~~Kettle principle~~

PRL **98,** 196405 (2007) PHYSICAL REVIEW LETTERS week ending 11 MAY 2007

First-Principles Approach to Noncollinear Magnetism: Towards Spin Dynamics

S. Sharma,[1,2,5,*] J. K. Dewhurst,[2,3] C. Ambrosch-Draxl,[2,4] S. Kurth,[5] N. Helbig,[1,5] S. Pittalis,[5] S. Shallcross,[6] L. Nordström,[7] and E. K. U. Gross[5]

PRL **111,** 156401 (2013) PHYSICAL REVIEW LETTERS week ending 11 OCTOBER 2013

Transverse Spin-Gradient Functional for Noncollinear Spin-Density-Functional Theory

F. G. Eich[1,2,*] and E. K. U. Gross[1]

Thank you for your attention

Other developments: Hybrid MC
(better sampling, faster than MMC, equilibrium properties?)