Magnetic Excitations within TDDFpT

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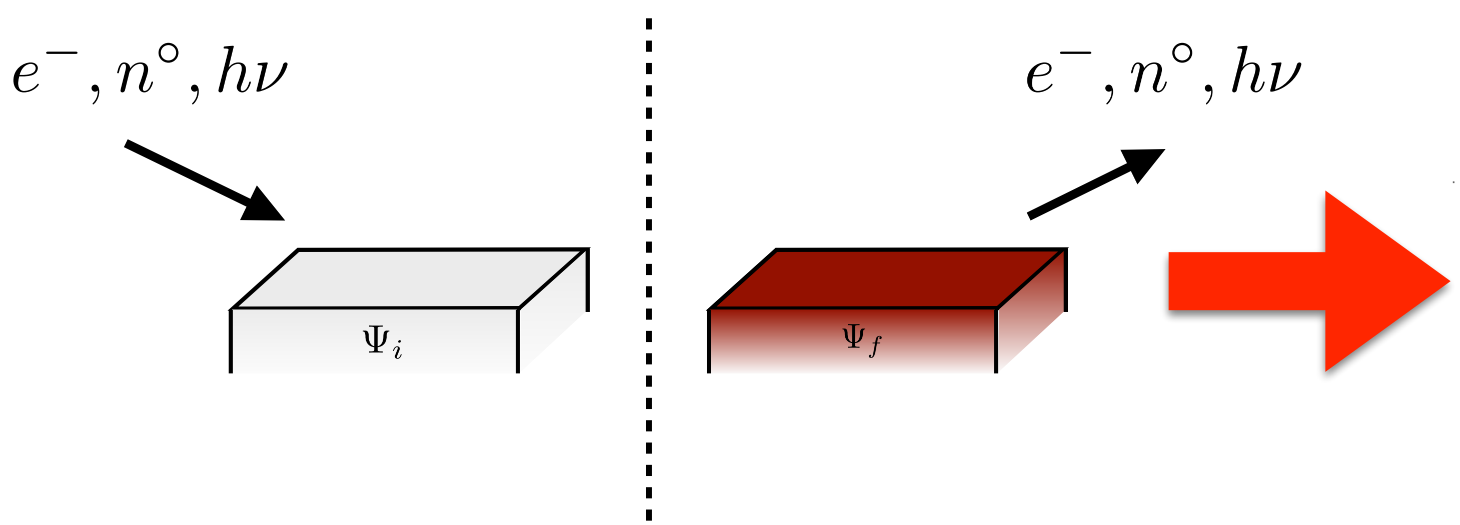
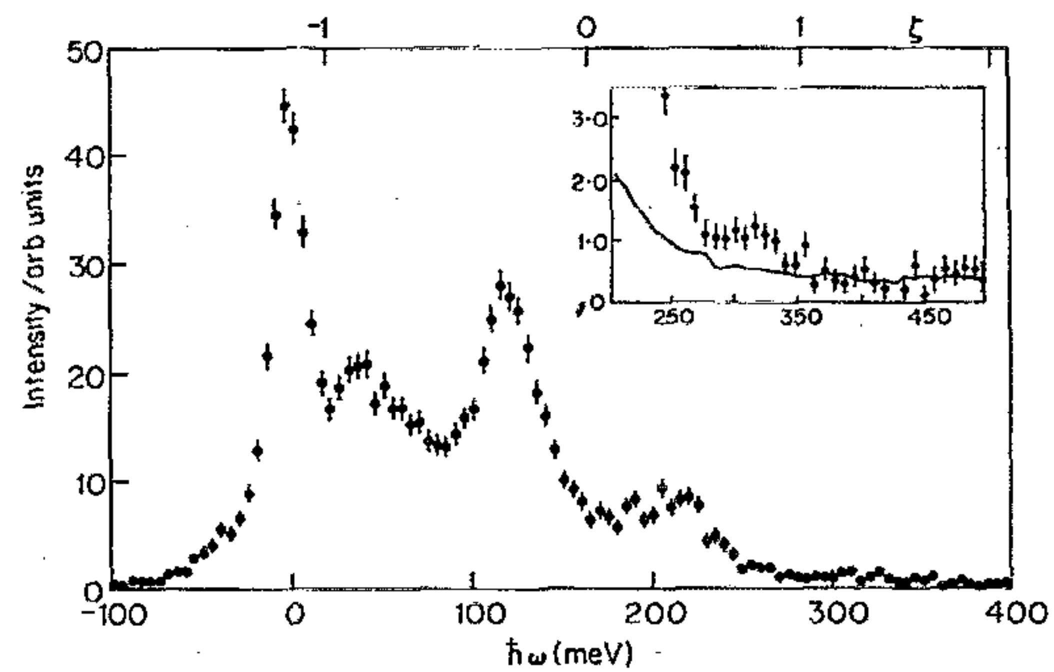
TDDFpT and spectroscopy

TDDFpT → first-principle description of numerous **spectroscopies**





- Optical absorption
- Electron energy loss spectroscopy (EELS)
- Inelastic X-ray scattering (IXS)
- Inelastic neutron scattering (INS)
- Spin-polarized electron energy loss spectroscopy (SPEELS)

→ **Magnetic excitations**

Magnetic excitations

- Contribute to specific heat ($\sim T^{3/2}$)
- May provide a coupling mechanism in high-T_C superconductivity
- Magnonics ==> circuits of magnetic materials, spin waves as information carriers
- Influence speed of information read&write in spintronic devices
- First-principle description of magnetic excitations in complex materials is still an open challenge

TDDFpT codes for modelling non-magnetic excitations

TDDFpT

turboTDDFT | **turboEELS**

Optical absorption

- Finite and extended (untested) systems
- Unpolarized case only
- NC and US PP
- Hybrids with NC PP only
- No empty states needed
- Two alternative approaches: Lanczos and Davidson

EELS and IXS

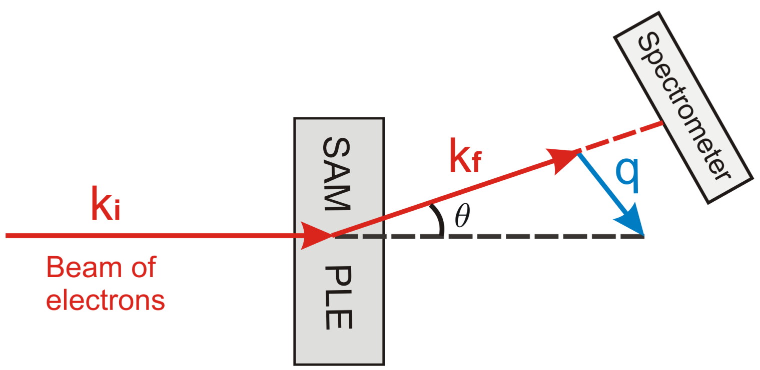
- Extended systems
- Unpolarized and non-collinear case
- NC and US PP
- Hybrids with NC PP only
- No empty states needed
- Lanczos approach

EELS for non-magnetic systems

Interaction part | External perturbation

$$(\hat{h}^{\circ}_{\mathbf{k+q}} - \varepsilon_{n\mathbf{k}} - \omega)\tilde{u}'_{n\mathbf{k+q}}(\mathbf{r},\omega) + \hat{P}_{\mathcal{C}}^{\mathbf{k+q}}\,\tilde{v}'_{\mathrm{Hxc},\mathbf{q}}(\mathbf{r},\omega)u^{\circ}_{n\mathbf{k}}(\mathbf{r}) = -\hat{P}_{\mathcal{C}}^{\mathbf{k+q}}\,\tilde{v}'_{\mathrm{ext},\mathbf{q}}(\mathbf{r},\omega)u^{\circ}_{n\mathbf{k}}(\mathbf{r})$$

$$(\hat{h}^{\circ}_{\mathbf{k+q}} - \varepsilon_{n\mathbf{k}} + \omega)\tilde{u}'^{*}_{n\mathbf{-k-q}}(\mathbf{r},-\omega) + \hat{P}_{\mathcal{C}}^{\mathbf{k+q}}\,\tilde{v}'_{\mathrm{Hxc},\mathbf{q}}(\mathbf{r},\omega)u^{\circ}_{n\mathbf{k}}(\mathbf{r}) = -\hat{P}_{\mathcal{C}}^{\mathbf{k+q}}\,\tilde{v}'_{\mathrm{ext},\mathbf{q}}(\mathbf{r},\omega)u^{\circ}_{n\mathbf{k}}(\mathbf{r})$$



$$(\omega - \hat{\mathcal{L}}_{\mathbf{q}}) \cdot \hat{\rho}'_{\mathbf{q}} = [\hat{\tilde{v}}'_{\mathrm{ext},\mathbf{q}}(\omega), \hat{\rho}^{\circ}]$$

$$\hat{\mathcal{L}}_{\mathbf{q}} \cdot \hat{\rho}'_{\mathbf{q}} \equiv [\hat{h}^{\circ}, \hat{\rho}'_{\mathbf{q}}] + [\hat{\tilde{v}}'_{\mathrm{Hxc},\mathbf{q}}, \hat{\rho}^{\circ}]$$

Use of time-reversal symmetry

- Lanczos algorithm for real matrices
- Factor-2 gain (batch rotation)

Charge-density susceptibility
(density-density response function)

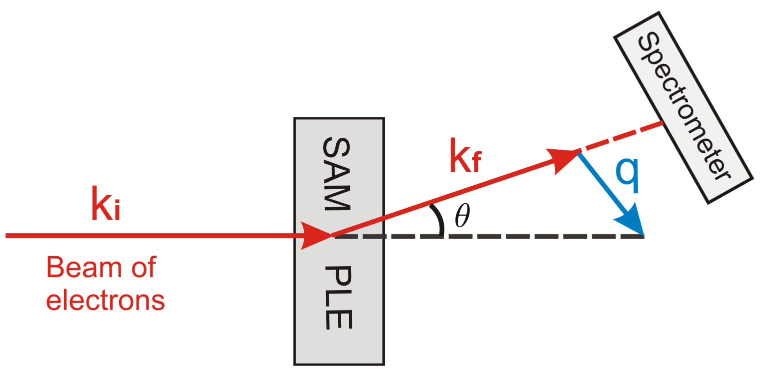
$$\chi(\mathbf{q},\omega) = \mathrm{Tr}\left[\hat{n}_{\mathbf{q}}(\omega - \mathcal{L})^{-1} \cdot [\hat{n}_{\mathbf{q}}, \hat{\rho}^{\circ}]\right]$$

EELS for non-magnetic systems

Interaction part

External perturbation

$$
\begin{aligned}
(\hat{h}^{\circ}_{\mathbf{k}+\mathbf{q}} - \varepsilon_{n\mathbf{k}} - \omega)\tilde{u}'_{n\mathbf{k}+\mathbf{q}}(\mathbf{r},\omega) \boxed{+ \hat{P}_{c}^{\mathbf{k}+\mathbf{q}}\, \tilde{v}'_{\mathrm{Hxc},\mathbf{q}}(\mathbf{r},\omega) u^{\circ}_{n\mathbf{k}}(\mathbf{r})} &= \boxed{-\hat{P}_{c}^{\mathbf{k}+\mathbf{q}}\, \tilde{v}'_{\mathrm{ext},\mathbf{q}}(\mathbf{r},\omega) u^{\circ}_{n\mathbf{k}}(\mathbf{r})} \\
(\hat{h}^{\circ}_{\mathbf{k}+\mathbf{q}} - \varepsilon_{n\mathbf{k}} + \omega)\tilde{u}'^{*}_{n-\mathbf{k}-\mathbf{q}}(\mathbf{r},-\omega) \boxed{+ \hat{P}_{c}^{\mathbf{k}+\mathbf{q}}\, \tilde{v}'_{\mathrm{Hxc},\mathbf{q}}(\mathbf{r},\omega) u^{\circ}_{n\mathbf{k}}(\mathbf{r})} &= \boxed{-\hat{P}_{c}^{\mathbf{k}+\mathbf{q}}\, \tilde{v}'_{\mathrm{ext},\mathbf{q}}(\mathbf{r},\omega) u^{\circ}_{n\mathbf{k}}(\mathbf{r})}
\end{aligned}
$$



~~Use of time-reversal symmetry~~

- ~~Lanczos algorithm for real matrices~~
- ~~Factor 2 gain (batch rotation)~~

$$
\boxed{
\begin{aligned}
(\omega - \hat{\mathcal{L}}_{\mathbf{q}}) \cdot \hat{\rho}'_{\mathbf{q}} &= [\hat{\tilde{v}}'_{\mathrm{ext},\mathbf{q}}(\omega), \hat{\rho}^{\circ}] \\
\hat{\mathcal{L}}_{\mathbf{q}} \cdot \hat{\rho}'_{\mathbf{q}} &\equiv [\hat{h}^{\circ}, \hat{\rho}'_{\mathbf{q}}] + [\hat{\tilde{v}}'_{\mathrm{Hxc},\mathbf{q}}, \hat{\rho}^{\circ}]
\end{aligned}
}
$$

No time-reversal symmetry when modelling magnetic excitations

Charge-density susceptibility
(density-density response function)

$$
\boxed{\chi(\mathbf{q},\omega) = \mathrm{Tr}\left[\hat{n}_{\mathbf{q}}(\omega - \mathcal{L})^{-1} \cdot [\hat{n}_{\mathbf{q}}, \hat{\rho}^{\circ}]\right]}
$$

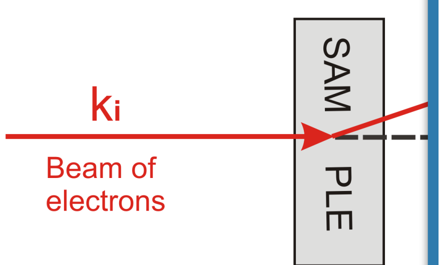
EELS for non-magnetic systems

Interaction part

External perturbation

$(\hat{h}^{\circ}_{\mathbf{k+q}} - \varepsilon$ … $_{n\mathbf{k}}(\mathbf{r})$

$(\hat{h}^{\circ}_{\mathbf{k+q}} - \varepsilon_{n\mathbf{k}}$ … $_{n\mathbf{k}}(\mathbf{r})$



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excitations

Generalized to treat magnetic systems

- Compute response to an external magnetic field
- Non-collinear magnetism (spinors, $n(\mathbf{r}), \mathbf{m}(\mathbf{r}), v_{\mathrm{Hxc}}(\mathbf{r}), \mathbf{b}_{\mathrm{xc}}(\mathbf{r})$)
- Lanczos algorithm generalized to complex algebra
- No time-reversal symmetry ==> need of KS wave functions at $\mathbf{k}, \mathbf{k+q}, \mathbf{k-q}$

Magnetization-density susceptibility
(magnetization-magnetization response function)

$$\boxed{\chi_{\alpha\beta}(\mathbf{q},\omega) = \mathrm{Tr}\left[\hat{m}^{\alpha}_{\mathbf{q}}(\omega-\mathcal{L})^{-1}\cdot[\hat{m}^{\beta}_{\mathbf{q}},\hat{\rho}^{\circ}]\right]} \longrightarrow \boxed{\text{Magnons}}$$

Lanczos algorithm with complex algebra

1) Initialize

$$q_1 = p_1 = v \quad \begin{pmatrix} \left\{ \hat{P}_{\mathcal{C}}^{\mathbf{k}+\mathbf{q}} \tilde{v}_{\mathrm{ext},\mathbf{q}}(\omega) u^{\circ}_{n\mathbf{k}}(\mathbf{r}) \right\} \\ \left\{ \hat{\mathrm{T}} \hat{P}_{\mathcal{C}}^{\mathbf{k}-\mathbf{q}} \tilde{v}_{\mathrm{ext},-\mathbf{q}}(-\omega) u^{\circ}_{n\mathbf{k}}(\mathbf{r}) \right\} \end{pmatrix}$$

$$u \quad \begin{pmatrix} \left\{ \hat{O}_{\mathbf{q}}(\omega) u^{\circ}_{n\mathbf{k}}(\mathbf{r}) \right\} \\ \left\{ \hat{\mathrm{T}} \hat{O}_{-\mathbf{q}}(-\omega) u^{\circ}_{n\mathbf{k}}(\mathbf{r}) \right\} \end{pmatrix}$$

$$\chi = \begin{array}{c c} & \begin{array}{cccc} \phantom{\chi_{\circ\circ}} & \phantom{\chi_{\circ x}} & v & \phantom{\chi_{\circ z}} \end{array} \\ & \begin{pmatrix} \chi_{\circ\circ} & \chi_{\circ x} & \chi_{\circ y} & \chi_{\circ z} \\ \chi_{x\circ} & \chi_{xx} & \chi_{xy} & \chi_{xz} \\ \chi_{y\circ} & \chi_{yx} & \chi_{yy} & \chi_{yz} \\ \chi_{z\circ} & \chi_{zx} & \chi_{zy} & \chi_{zz} \end{pmatrix} \begin{array}{c} \\ \\ u \\ \\ \end{array} \end{array}$$

2) Iterate

$$\beta_{i+1} q_{i+1} = \hat{\mathcal{L}}\, q_i - \alpha_i q_i - \gamma_i q_{i-1}$$

$$\gamma^*_{i+1} p_{i+1} = \hat{\mathcal{L}}^{\dagger} p_i - \alpha^*_i p_i - \beta^*_i p_{i-1}$$

$$\left[\text{store} \quad \alpha_i, \beta_i, \gamma_i, \zeta_i = \langle q_i, u \rangle \right]$$

$$T^N = \begin{pmatrix} \alpha_1 & \gamma_2 & 0 & \cdots & 0 \\ \beta_2 & \alpha_2 & \gamma_3 & 0 & \vdots \\ 0 & \beta_3 & \alpha_3 & \ddots & 0 \\ \vdots & 0 & \ddots & \ddots & \gamma_N \\ 0 & \cdots & 0 & \beta_N & \alpha_N \end{pmatrix}$$

3) Post-process

$$\langle u, (\omega - \hat{\mathcal{L}})^{-1} v \rangle \approx \sum_{i=1}^{N} \zeta^*_i \left[(\omega - T^N)^{-1} \right]_{i1}$$

Lanczos algorithm with complex algebra

1) Initialize

$$q_1 = p_1 = v \quad \rightarrow \quad \begin{pmatrix} \left\{ \hat{P}_{\mathcal{C}}^{\mathbf{k}+\mathbf{q}} \tilde{v}_{\mathrm{ext},\mathbf{q}}(\omega) u^{\circ}_{n\mathbf{k}}(\mathbf{r}) \right\} \\ \left\{ \hat{\mathrm{T}} \hat{P}_{\mathcal{C}}^{\mathbf{k}-\mathbf{q}} \tilde{v}_{\mathrm{ext},-\mathbf{q}}(-\omega) u^{\circ}_{n\mathbf{k}}(\mathbf{r}) \right\} \end{pmatrix}$$

$$u \quad \rightarrow \quad \begin{pmatrix} \left\{ \hat{O}_{\mathbf{q}}(\omega) u^{\circ}_{n\mathbf{k}}(\mathbf{r}) \right\} \\ \left\{ \hat{\mathrm{T}} \hat{O}_{-\mathbf{q}}(-\omega) u^{\circ}_{n\mathbf{k}}(\mathbf{r}) \right\} \end{pmatrix}$$

$$\chi = \begin{array}{c c} & \begin{array}{cccc} \phantom{\chi_{\circ\circ}} & \phantom{\chi_{\circ x}} & v & \phantom{\chi_{\circ z}} \end{array} \\ & \begin{pmatrix} \chi_{\circ\circ} & \chi_{\circ x} & \chi_{\circ y} & \chi_{\circ z} \\ \chi_{x\circ} & \chi_{xx} & \chi_{xy} & \chi_{xz} \\ \chi_{y\circ} & \chi_{yx} & \chi_{yy} & \chi_{yz} \end{pmatrix} & u \end{array}$$

- $\hat{h}^{\circ}$ applied $2\times$`nbnd_occ`$\times$`nksq` times
- 2 computations of response potentials

2) Iterate

$$\beta_{i+1} q_{i+1} = \hat{\mathcal{L}}\, q_i - \alpha_i q_i - \gamma_i q_{i-1}$$

$$\gamma^*_{i+1} p_{i+1} = \hat{\mathcal{L}}^{\dagger} p_i - \alpha^*_i p_i - \beta^*_i p_{i-1}$$

$$\left[\text{store} \quad \alpha_i, \beta_i, \gamma_i, \zeta_i = \langle q_i, u \rangle \right]$$

$$T^N = \begin{pmatrix} \alpha_1 & \gamma_2 & 0 & \cdots & 0 \\ \beta_2 & \alpha_2 & \gamma_3 & 0 & \vdots \\ 0 & \beta_3 & \alpha_3 & \ddots & 0 \\ \vdots & 0 & \ddots & \ddots & \gamma_N \\ 0 & \cdots & 0 & \beta_N & \alpha_N \end{pmatrix}$$

3) Post-process

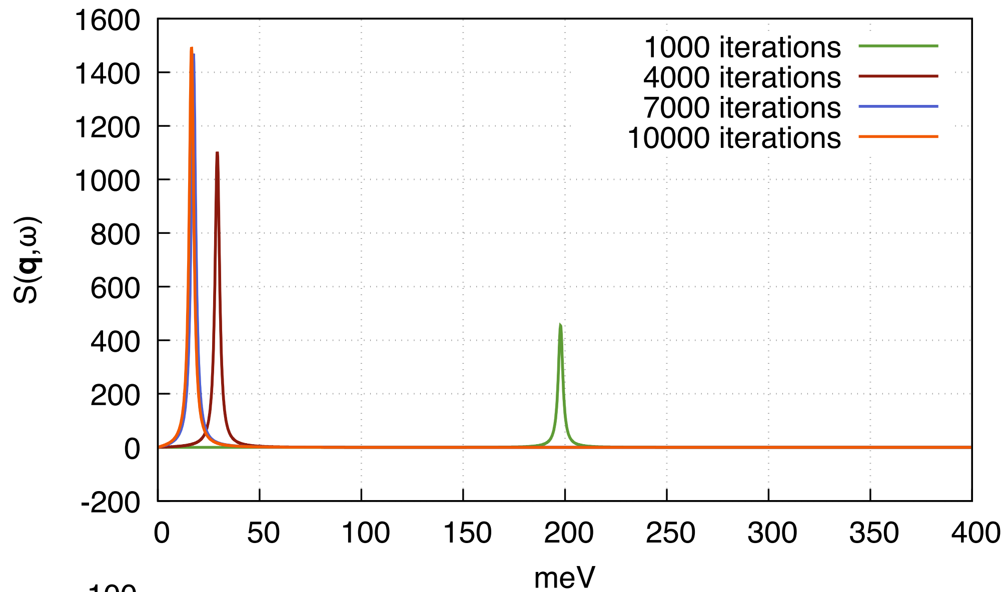
$$\langle u, (\omega - \hat{\mathcal{L}})^{-1} v \rangle \approx \sum_{i=1}^{N} \zeta_i^* \left[(\omega - T^N)^{-1} \right]_{i1}$$

Inexpensive, done in serial

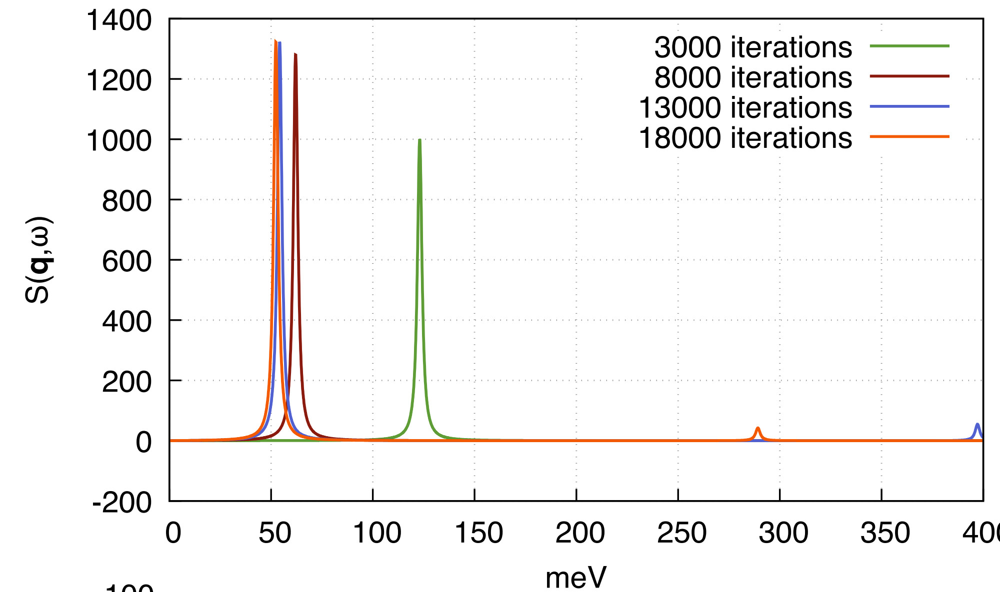
Testing: magnetic susceptibility of bcc iron

Magnon peak

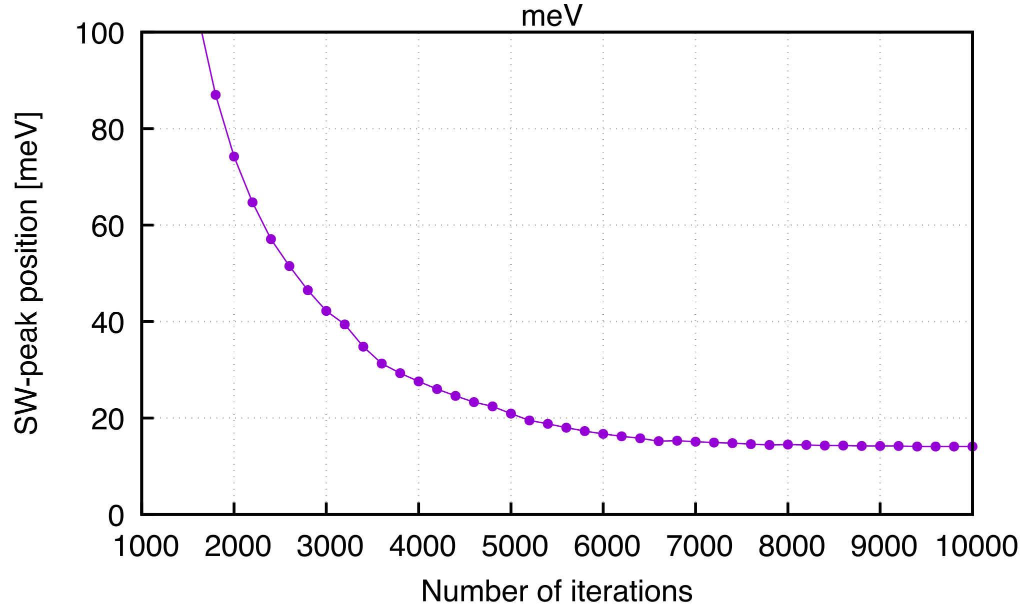
$\mathbf{q} = (0.09, 0, 0)2\pi/a$

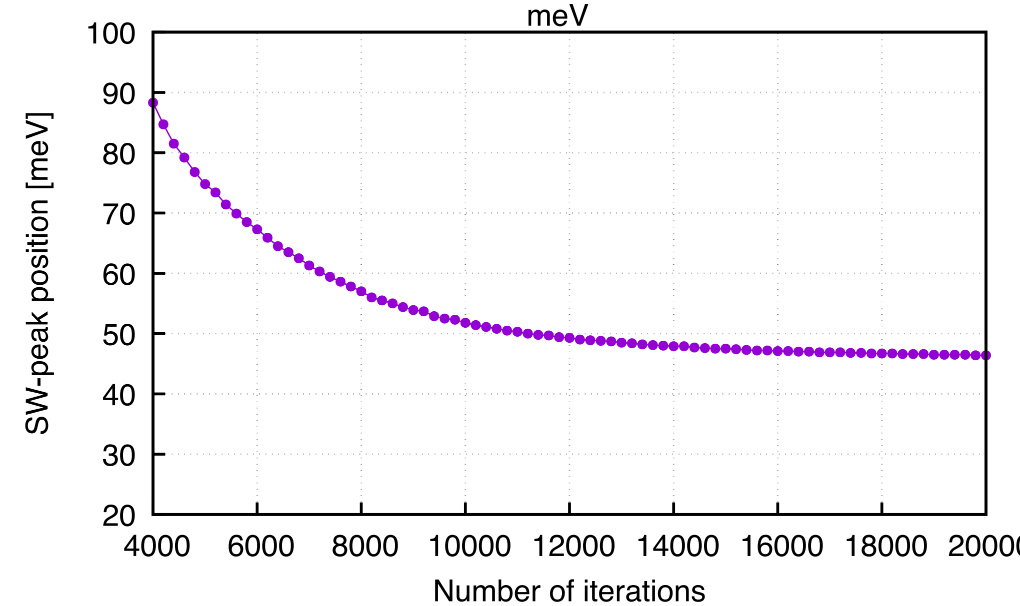


$\mathbf{q} = (0.18, 0, 0)2\pi/a$



Magnon peak position wrt iterations





Slow convergence wrt number of iterations ==> work in progress to speed it up

Implementation

- Implemented in the development version of QE(v. 5.3)
- Source code is ~ 10 FORTRAN files in TDDFPT/src
- Most of the routines are variations of the EELS code —- can be put together in the near future
- Only modification involving shared routines ==> LR_modules/incdrhoscf.f90
- Writes to disk also time-reversed of the unperturbed KS orbitals, now done in separate files *.Twfc
- Parallelized over G-vectors and k-points

Features

- Magnetic excitations within the Liouville-Lanczos approach
- Non-collinear only code (with spin-orbit)
- No empty states needed
- Extended systems
- All ingredients for finite systems already there, needs just some copy&paste + testing
- NC PP only
- Hybrids not yet supported

Thanks for your attention!