QUANTUM ESPRESSO FOUNDATION

QUANTUMESPRESSO

MAX CoE/ENCCS Workshop on efficient materials modelling on HPC
with Quantum ESPRESSO, Siesta, and Yambo

Hands-on session – Day 2

TDDFPT FOR HPC AND GPUs

Tommaso Gorni and Oscar Baseggio

t.gorni@cineca.it obaseggi@sissa.it

MAX

CINECA

ENCCS
EuroCC National Competence Centre Sweden

PAUL SCHERRER INSTITUT
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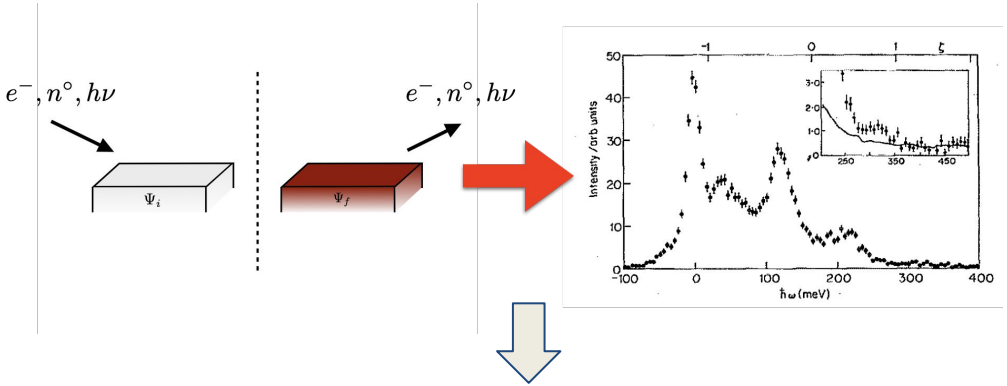
MARVEL
NATIONAL CENTRE OF COMPETENCE IN RESEARCH

Swiss National Science Foundation

Ab initio spectroscopy

Time-Dependent Density-Functional Perturbation Theory (TDDFpT)



- Optical absorption spectroscopy
- Electron energy loss spectroscopy (EELS)
- Inelastic X-ray scattering (IXS)
- Inelastic neutron scattering (INS)
- ...

Dynamical susceptibilities

$$\varphi_{\text{ext}}(t) \longrightarrow A(t) \approx A^\circ + A'(t)$$

$$A'(t) = \int \mathrm{d}t' \chi(t-t') \varphi_{\text{ext}}(t')$$

OUTLINE

1. How to compute the charge susceptibility with `turbo_eels.x`
2. How to compute the spin susceptibility with `turbo_magnon.x`

Time-Dependent Density-Functional Perturbation Theory

Kohn-Sham hamiltonian

Perturbing potential

$$(\hat{H}^{\circ} - \epsilon^{\circ}_{n,\mathbf{k}} - \hbar\omega)|\psi'_{n,\mathbf{k}+\mathbf{q}}(\omega)\rangle = -\hat{P}_{\mathcal{C}}\left[\hat{V}'_{\mathrm{Hxc},\mathbf{q}}(\omega) + \hat{V}'_{\mathrm{ext},\mathbf{q}}(\omega)\right]|\psi^{\circ}_{n,\mathbf{k}}\rangle$$

$$(\hat{H}^{\circ} - \epsilon^{\circ}_{n,-\mathbf{k}} + \hbar\omega)|\psi'^{*}_{n,-\mathbf{k}-\mathbf{q}}(-\omega)\rangle = -\hat{P}_{\mathcal{C}}\left[\hat{V}'_{\mathrm{Hxc},\mathbf{q}}(\omega) + \hat{V}'_{\mathrm{ext},\mathbf{q}}(\omega)\right]|\psi^{\circ *}_{n,-\mathbf{k}}\rangle$$

(2 X nbnd X nks) coupled equations

Resonant and anti-resonant response of the (n,k) state

Option 1: Sternheimer approach (PHonon-like)

- Invert the linear system **for each frequency** to build the response density matrix

$$\hat{\rho}'_{\mathbf{q}}(\omega) = \sum_{n,\mathbf{k}}^{\mathrm{occ.}}\left[|\psi'_{n,\mathbf{k}+\mathbf{q}}(\omega)\rangle\langle\psi^{\circ}_{n,\mathbf{k}}| + |\psi'^{*}_{n,-\mathbf{k}-\mathbf{q}}(-\omega)\rangle\langle\psi^{\circ *}_{n,-\mathbf{k}}|\right]$$

Cost of static linear response X number of frequencies

- Compute the dynamical susceptibility

$$n'(\mathbf{q},\omega) = \boxed{\chi(\mathbf{q},\mathbf{q},\omega)}V'_{\mathrm{ext},\mathbf{q}}(\omega) = \mathrm{Tr}\left[\hat{n}^{\dagger}_{\mathbf{q}}\hat{\rho}'_{\mathbf{q}}(\omega)\right]$$

$\propto$ EELS cross section

Time-Dependent Density-Functional Perturbation Theory

Kohn-Sham hamiltonian

Perturbing potential

$$(\hat{H}^{\circ} - \epsilon^{\circ}_{n,\mathbf{k}} - \hbar\omega)|\psi'_{n,\mathbf{k}+\mathbf{q}}(\omega)\rangle = -\hat{P}_{\mathcal{C}}\left[\hat{V}'_{\mathrm{Hxc},\mathbf{q}}(\omega) + \hat{V}'_{\mathrm{ext},\mathbf{q}}(\omega)\right]|\psi^{\circ}_{n,\mathbf{k}}\rangle$$

$$(\hat{H}^{\circ} - \epsilon^{\circ}_{n,-\mathbf{k}} + \hbar\omega)|\psi'^{*}_{n,-\mathbf{k}-\mathbf{q}}(-\omega)\rangle = -\hat{P}_{\mathcal{C}}\left[\hat{V}'_{\mathrm{Hxc},\mathbf{q}}(\omega) + \hat{V}'_{\mathrm{ext},\mathbf{q}}(\omega)\right]|\psi^{\circ *}_{n,-\mathbf{k}}\rangle$$

(2 X nbnd X nks) coupled equations

Resonant and anti-resonant response of the (n,k) state

Option 2: Lanczos approach

- Recast as a unique linear problem $\left(\hbar\omega - \mathcal{L}_{\mathbf{q}}(\omega)\right) \cdot \hat{\rho}'_{\mathbf{q}}(\omega) = \left[\hat{V}'_{\mathrm{ext},\mathbf{q}}(\omega)\,,\hat{\rho}^{\circ}\right]$

- The response density matrix can be represented as an array of response orbitals (‘batch’).

$$\hat{\rho}'_{\mathbf{q}}(\omega) \rightarrow \begin{pmatrix} \psi'_{n,\mathbf{k}+\mathbf{q}}(\omega) \\ \psi'^{*}_{n,-\mathbf{k}-\mathbf{q}}(-\omega) \end{pmatrix}_{n,\mathbf{k}}$$

batch size = (2 x npw x nbnd x nks) complex numbers

- The action of the Liouvillian on the batch costs roughly twice a static linear response step.

making use of time-reversal symmetry (standard batch rotation)

Time-Dependent Density-Functional Perturbation Theory

Option 2: Lanczos approach

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

For adiabatic xc kernels

- Tridiagonalize the Liouvillian via Lanczos recursion (computationally intensive part).

$$\begin{aligned}\beta_{i+i}\mathbf{V}_{i+1} &= \mathcal{L}_{\mathbf{q}}\mathbf{V}_i - \gamma_i\mathbf{V}_{i-1}\\ \gamma_{i+i}\mathbf{U}_{i+1} &= \mathcal{L}^{\dagger}_{\mathbf{q}}\mathbf{U}_i - \beta_i\mathbf{U}_{i-1}\end{aligned}$$

$$\mathcal{L}_{\mathbf{q}} \approx T^N_{\mathbf{q}} = \begin{pmatrix} 0 & \gamma_2 & 0 & \cdots & 0 \\ \beta_2 & 0 & \gamma_3 & 0 & 0 \\ 0 & \beta_3 & 0 & \ddots & 0 \\ \vdots & 0 & \ddots & \ddots & \gamma_N \\ 0 & 0 & 0 & \beta_N & 0 \end{pmatrix}$$

- Invert at **any desired frequency** at a negligible computation cost.

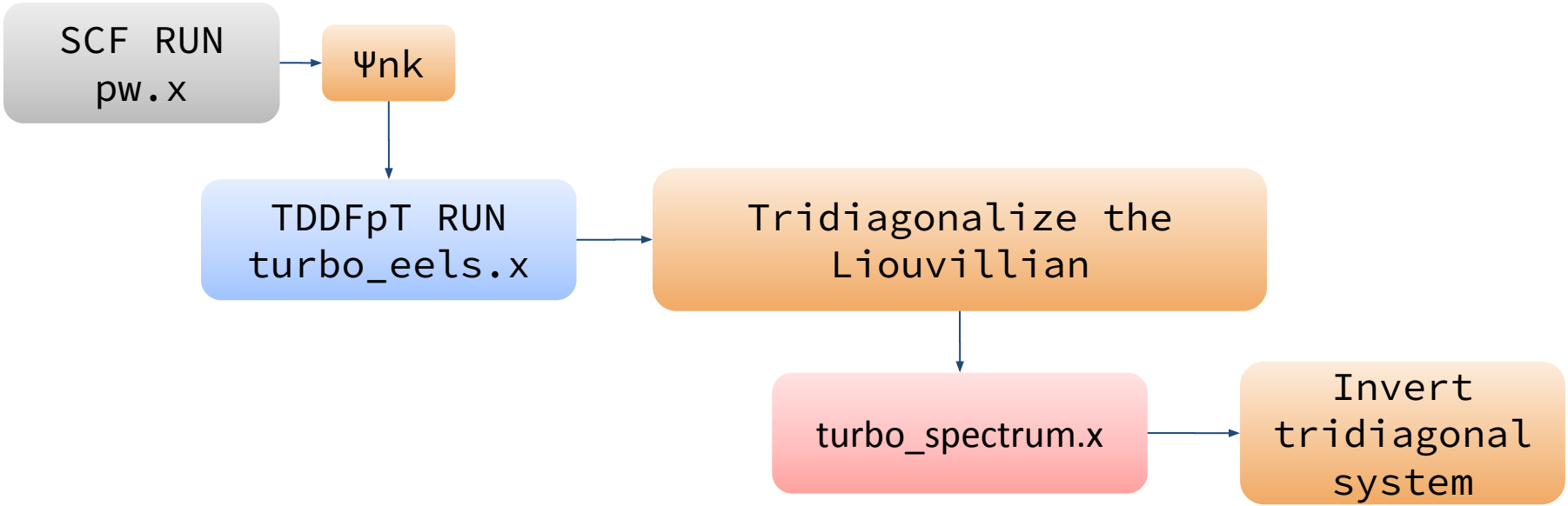
$$\chi(\mathbf{q},\mathbf{q},\omega) \approx \left\langle u\,,(\hbar\omega + i\eta - T^N_{\mathbf{q}})^{-1} v\right\rangle$$

Imaginary shift to regularize poles (broadening term)

Batches (npw x nbnd x nks) complex numbers

EXERCISE: TURBO_EELS.X

The turbo_eels workflow for modes at a single q point



EXERCISE: EELS IN BULK SILICON

Go to the directory with the input files:

```
cd  Day-2/exercise_eels
```

In this directory you will find:

- `README.md` – File describing how to do the exercise
- `pw.Si.scf.in` – Input file for the SCF ground-state calculation
- `turbo_eels.Si.tddfpt.in` – Input file for the EELS calculation
- `turbo_spectrum.Si.pp.in` – Input file for post processing calculation
- `reference` – Directory with the reference results

PWSCF SIMULATION, STEP 1

1. `cd Day-2/exercise-eels`

   Perform a self-consistent field ground-state calculation for silicon using the *pw.x* *program.*

- Open and check `pw.Si.scf.in`
- Open and check `submit_pw.slurm`
- Submit the job file

```
&control
   calculation  = 'scf'
   restart_mode = 'from_scratch'
   prefix       = 'Si'
   pseudo_dir   = '../../pseudo'
   outdir       = './tempdir'
   verbosity    = 'high'
/
&system
   ibrav     = 2
   celldm(1) = 10.26
   nat       = 2
   ntyp      = 1
   ecutwfc   = 20.0
/
&electrons
   conv_thr =  1.0d-10
/
ATOMIC_SPECIES
 Si  28.08  Si.upf
ATOMIC_POSITIONS alat
 Si 0.00 0.00 0.00
 Si 0.25 0.25 0.25
K_POINTS automatic
 12 12 12 0 0 0
```

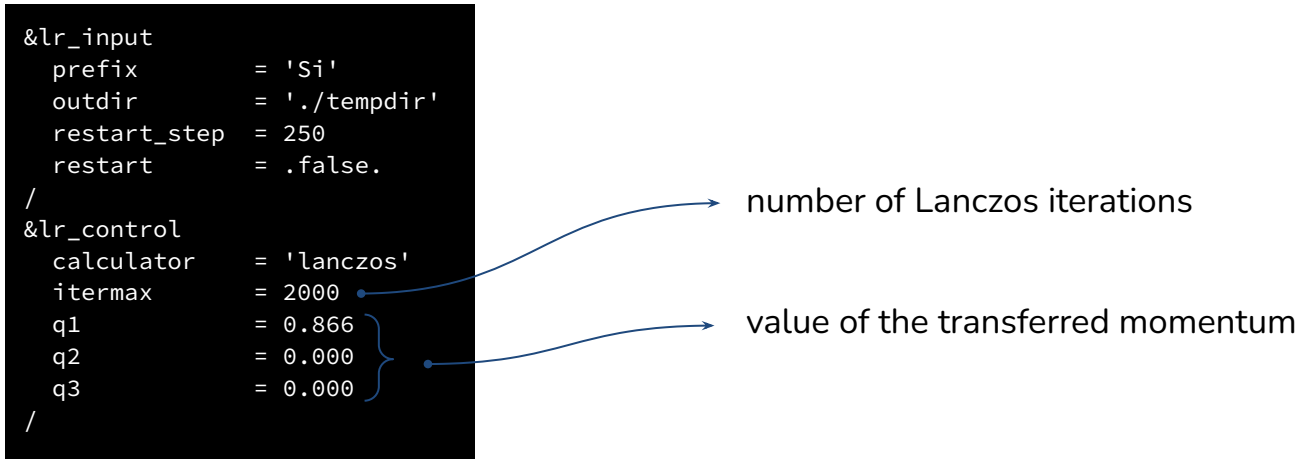
EELS CALCULATION, STEP 2

2. `cd Day-2/exercise-eels`

Perform a Lanczos coefficients calculation using the `turbo_eels.x` program.

- Open and check `turbo_eels.Si.tddfpt.in`

```
&lr_input
  prefix        = 'Si'
  outdir        = './tempdir'
  restart_step  = 250
  restart       = .false.
/
&lr_control
  calculator    = 'lanczos'
  itermax       = 2000
  q1            = 0.866
  q2            = 0.000
  q3            = 0.000
/
```



- Submit the job file `submit_eels.slurm`

SPECTRUM CALCULATION, STEP 3

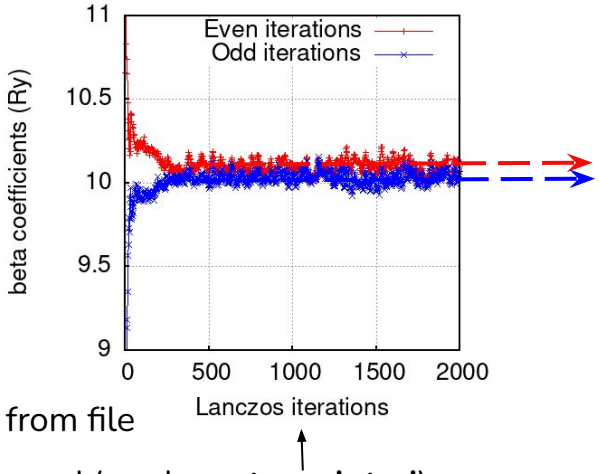
3. `cd Day-2/exercise-eels`

Perform spectrum using the `turbo_spectrum.x` program.

- Open and check `turbo_spectrum.Si.pp.in`

```
&lr_input
  prefix        = 'Si'
  outdir        = './tmp'
  eels          = .true.
  units         = 1
  itermax0      = 2000
  itermax       = 10000
  extrapolation = 'osc'
  epsil         = 0.03675
  start         = 0.d0
  increment     = 0.05d0
  end           = 50.d0
/
```

- the unit system used (1=eV)
- number of Lanczos coefficients read from file
- total number of Lanczos coefficients used (read + **extrapolated**)
- extrapolation scheme
- broadening term (Ry)
- spectrum energy grid (eV)



- Submit the job file `submit_spectrum.slurm`

IMPLEMENTED FEATURES

- ✓ Metals and insulators
- ! Non-magnetic systems only
- ✓ Spin-orbit coupling (fully relativistic approach)
- ✓ LDA, GGA functionals (+U, work in progress)
- ✓ Norm-conserving and ultrasoft pseudopotentials
- ✓ Use of symmetries to reduce k-points (nks)
- ✓ R&G, k-point parallelization
- ✓ GPU offloading (new!)

Time-Dependent Density-Functional Perturbation Theory

Kohn-Sham hamiltonian

Spinor wavefunctions

Perturbing potential (magnetic field)

$$(\hat{H}^{\circ} - \epsilon^{\circ}_{n,\mathbf{k}} - \hbar\omega)|\psi'_{n,\mathbf{k}+\mathbf{q}}(\omega)\rangle = -\hat{P}_c\left[\hat{V}'_{\mathrm{Hxc},\mathbf{q}}(\omega) + \hat{V}'_{\mathrm{ext},\mathbf{q}}(\omega)\right]|\psi^{\circ}_{n,\mathbf{k}}\rangle$$

$$(\hat{H}^{\circ} - \epsilon^{\circ}_{n,-\mathbf{k}} + \hbar\omega)\hat{T}|\psi'_{n,-\mathbf{k}-\mathbf{q}}(-\omega)\rangle = -\hat{P}^{+}_{c}\left[\hat{V}'^{+}_{\mathrm{Hxc},\mathbf{q}}(\omega) + \hat{V}'^{+}_{\mathrm{ext},\mathbf{q}}(\omega)\right]\hat{T}|\psi^{\circ}_{n,-\mathbf{k}}\rangle$$

Time-reversal operator

$\hat{O}^{+}_{\mathbf{q}}(\omega) = \hat{T}\hat{O}_{-\mathbf{q}}(-\omega)\hat{T}^{-1}$

Resonant and anti-resonant response of the (n,k) state

- Recast as a unique linear problem $\left(\hbar\omega - \mathcal{L}_{\mathbf{q}}(\omega)\right) \cdot \hat{\rho}'_{\mathbf{q}}(\omega) = \left[\hat{V}'_{\mathrm{ext},\mathbf{q}}(\omega)\,,\hat{\rho}^{\circ}\right]$

- The response density matrix can be represented as an array of response orbitals (‘batch’). $\hat{\rho}'_{\mathbf{q}}(\omega) \rightarrow \begin{pmatrix} \psi'_{n,\mathbf{k}+\mathbf{q}}(\omega) \\ \hat{T}\psi'_{n,-\mathbf{k}-\mathbf{q}}(-\omega) \end{pmatrix}_{n,\mathbf{k}}$

batch size
=
(**4** x npw x nbnd x nks)
complex numbers

- The action of the Liouvillian on the batch costs roughly twice a static linear response step.

Time-Dependent Density-Functional Perturbation Theory

Lanczos approach

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

For adiabatic xc kernels

- Tridiagonalize the Liouvillian via Lanczos recursion (computational intensive part).

$$\beta_{i+i}\mathbf{V}_{i+1} = \mathcal{L}_{\mathbf{q}}\mathbf{V}_i - \alpha_i\mathbf{V}_i - \gamma_i\mathbf{V}_{i-1}$$
$$\gamma^*_{i+i}\mathbf{U}_{i+1} = \mathcal{L}^{\dagger}_{\mathbf{q}}\mathbf{U}_i - \alpha_i\mathbf{U}_i - \beta_i\mathbf{U}_{i-1}$$

$$\mathcal{L}_{\mathbf{q}} \approx T^N_{\mathbf{q}} = \begin{pmatrix} \alpha_1 & \gamma_2 & 0 & \cdots & 0 \\ \beta_2 & \alpha_2 & \gamma_3 & 0 & 0 \\ 0 & \beta_3 & \alpha_3 & \ddots & 0 \\ \vdots & 0 & \ddots & \ddots & \gamma_N \\ 0 & 0 & 0 & \beta_N & \alpha_N \end{pmatrix}$$

- Invert at **any desired frequency** at a negligible computation cost.

INS cross section
$\propto$
anti-hermitian part of
$\chi_{\lambda\mu}(\mathbf{q},\mathbf{q},\omega)$

Magnetization component

$$\chi_{\lambda\mu}(\mathbf{q},\mathbf{q},\omega) \approx \left\langle u^{\lambda}\,,(\hbar\omega + i\eta - T^N_{\mathbf{q}})^{-1} v^{\mu}\right\rangle$$

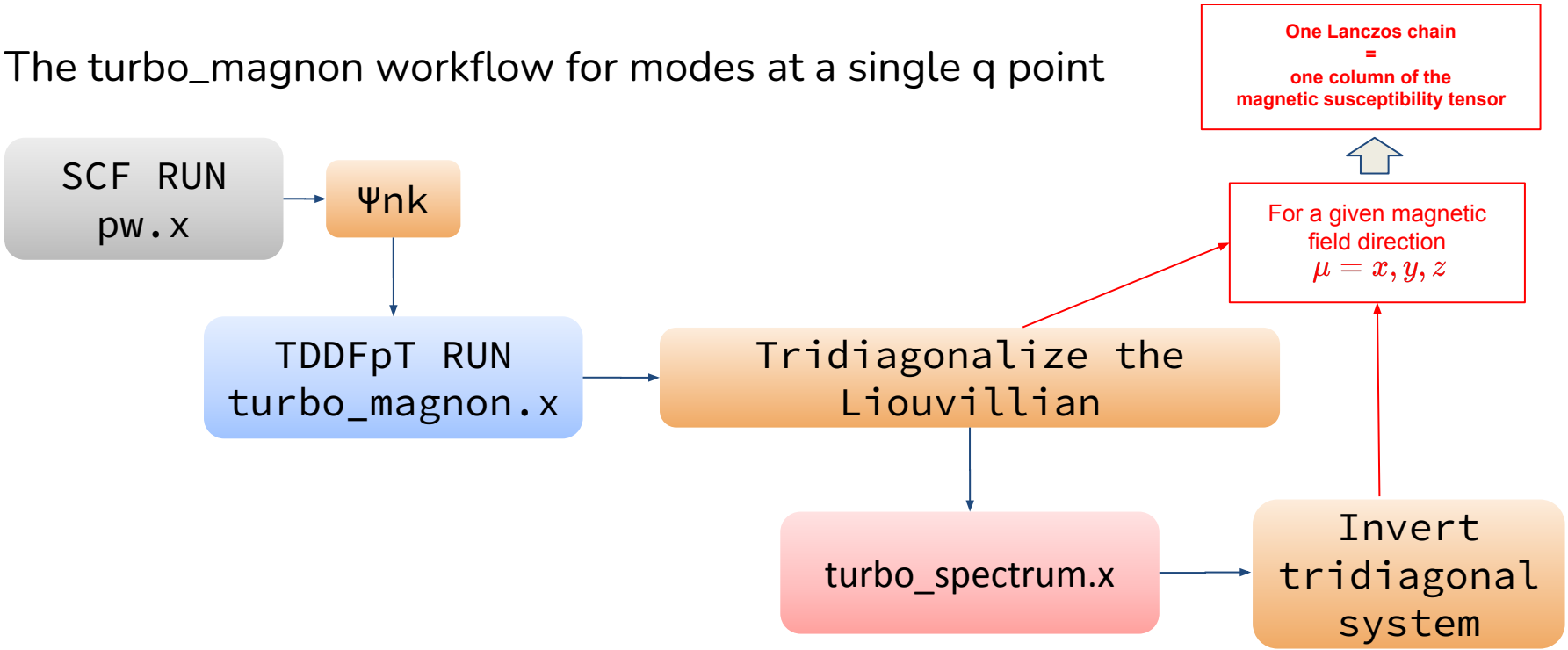
Magnetic field component

Imaginary shift to regularize poles
(broadening term)

Batches
(4 x npw x nbnd x nks)
complex numbers

EXERCISE: TURBO_MAGNON.X

The turbo_magnon workflow for modes at a single q point



EXERCISE: MAGNONS IN BULK IRON

Go to the directory with the input files:

```
cd  Day-2/exercise_magnon
```

In this directory you will find:

- `README.md` – File describing how to do the exercise
- `pw.Fe.scf.in` – Input file for the SCF ground-state calculation
- `turbo_magnon.Fe.tddfpt.in` – Input file for the magnon calculation
- `turbo_spectrum.Fe.pp.in` – Input file for post processing calculation
- `reference` – Directory with the reference results

PWSCF SIMULATION, STEP 1

1. `cd Day-2/exercise-magnon`

   Perform a self-consistent field ground-state calculation for iron using the *pw.x* *program.*

- Open and check `pw.Fe.scf.in`
- Open and check `submit_pw.slurm`
- Submit the job file

```
 &control
    calculation  = 'scf'
    restart_mode = 'from_scratch'
    prefix       = 'Fe'
    outdir       = './tempdir'
    pseudo_dir   = '../../pseudo'
    verbosity    = 'high'
 /
 &system
    nosym           = .true.
    noinv           = .true.
    noncolin        = .true.
    lspinorb        = .false.
    ibrav           = 3
    celldm(1)       = 5.406
    nat             = 1
    ntyp            = 1
    ecutwfc         = 40
    occupations     = 'smearing'
    smearing        = 'gaussian'
    degauss         = 0.01
    starting_magnetization(1) = 0.15
 /
 &electrons
    mixing_beta     = 0.3
    conv_thr        = 1.d-12
 /
ATOMIC_SPECIES
Fe  55.85   Fe.pz-n-nc.UPF
ATOMIC_POSITIONS alat
Fe  0.00000000 0.00000000 0.00000000
K_POINTS automatic
 4 4 4 0 0 0
```

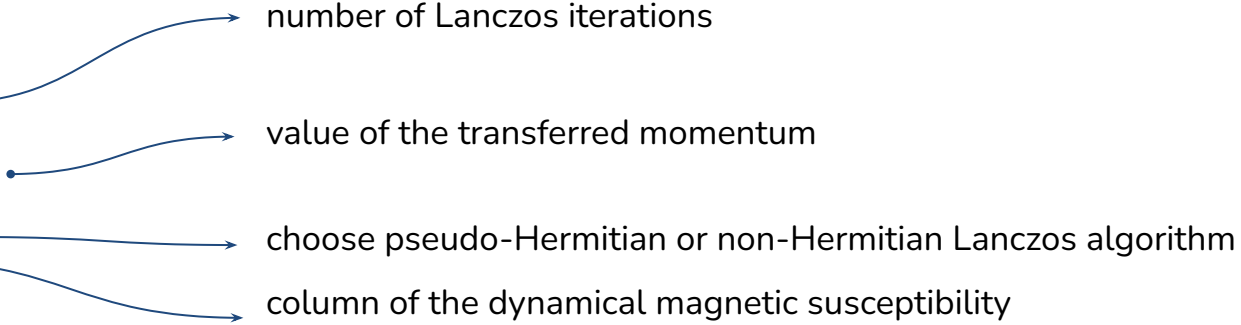
MAGNON CALCULATION, STEP 2

2. `cd Day-2/exercise-magnon`

Perform a Lanczos coefficients calculation using the `turbo_magnon.x` program.

- Open and check `turbo_magnon.Fe.tddfpt.in`

```
 &lr_input
   prefix       = 'Fe'
   outdir       = './tempdir'
   restart_step = 200
   restart      = .false.
 /
&lr_control
    itermax           = 5000
    q1                = 0.1d0
    q2                = 0.1d0
    q3                = 0.0d0
    pseudo_hermitian = .true.
    ipol              = 2
/
```



- Submit the job file `submit_magnon.slurm`

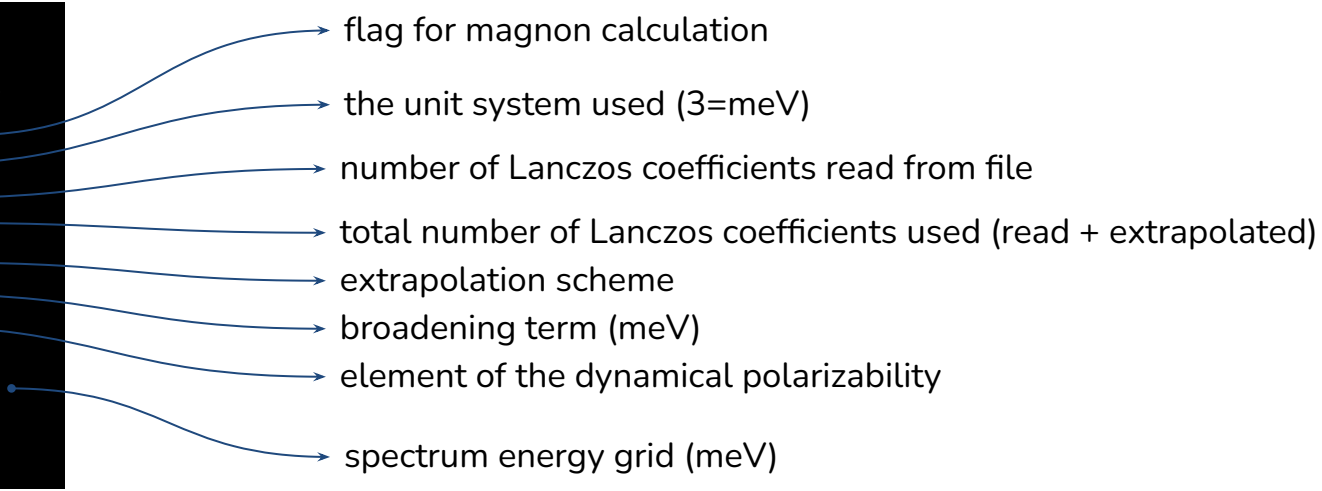
SPECTRUM CALCULATION, STEP 3

3. `cd Day-2/exercise-magnon`

Perform spectrum using the `turbo_spectrum.x` program.

- Open and check `turbo_spectrum.Fe.pp.in`

```
&lr_input
  prefix        = 'Fe'
  outdir        = './tempdir'
  magnons       = .true.
  units         = 3
  itermax0      = 5000
  itermax       = 15000
  extrapolation = 'osc'
  epsil         = 1.0
  ipol          = 2
  start         = 0.d0
  increment     = 1.d0
  end           = 28.d0
/
```



- Submit the job file `submit_spectrum.slurm`
- *grep chi_2_2 Femag.plot_chi.dat &> Chi_2_2.dat & and use the script for gnuplot*

IMPLEMENTED FEATURES

- ✓ Metals and insulators
- ✓ Spin-orbit coupling (fully relativistic approach)
- ✓ LDA functionals (+U, work in progress)
- ✓ Norm-conserving pseudopotentials only
- ! No symmetry is used (set noinv = .true. and nosym = .true. in the pw input)
- ✓ R&G, k-point parallelization
- ✓ GPU offloading (new, work in progress)