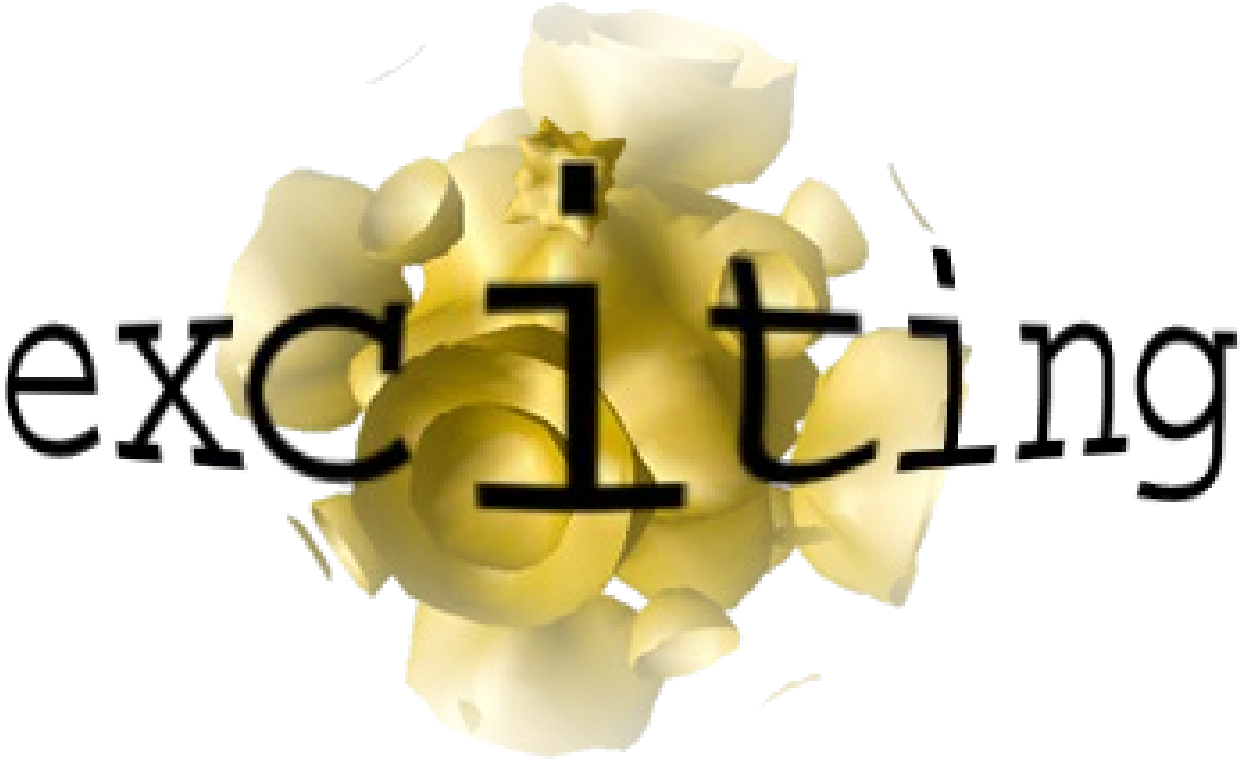
http://exciting-code.org



TDDFT in **exciting**

Santiago Rigamonti
Humboldt Universität zu Berlin
exciting Workshop, 2 August 2014, Berlin

TDDFT in a nutshell

Key quantity: dielectric function

$$\varepsilon_{\mathbf{G}\mathbf{G}'}(\mathbf{q},\omega)$$

TDDFT in a nutshell

Key quantity: dielectric function

$$\varepsilon_{\mathbf{G}\mathbf{G}'}(\mathbf{q},\omega)$$

In real space-time coordinates obtained from Fourier transform:

$$\varepsilon(\mathbf{r}t,\mathbf{r}'t') = \frac{1}{2\pi V}\sum_{\mathbf{q}}\sum_{\mathbf{G}\mathbf{G}'}\int_{\omega} d\omega e^{i(\mathbf{q}+\mathbf{G})\mathbf{r}}\varepsilon_{\mathbf{G}\mathbf{G}'}(\mathbf{q},\omega)e^{-i(\mathbf{q}+\mathbf{G}')\mathbf{r}'}e^{i\omega(t-t')}$$

TDDFT in a nutshell

Key quantity: dielectric function

$$\varepsilon_{\mathbf{G}\mathbf{G}'}(\mathbf{q},\omega)$$

Macroscopic dielectric function:

$$\varepsilon_M(\mathbf{q},\omega) = \frac{1}{\varepsilon^{-1}_{00}(\mathbf{q},\omega)}$$

TDDFT in a nutshell

Key quantity: dielectric function

$$\varepsilon_{\mathbf{G}\mathbf{G}'}(\mathbf{q},\omega)$$

Macroscopic dielectric function:

$$\varepsilon_M(\mathbf{q},\omega) = \frac{1}{\varepsilon^{-1}_{00}(\mathbf{q},\omega)}$$

and calculate several quantities:

$$L(\mathbf{q},\omega) = -Im\left[\frac{1}{\varepsilon_M(\mathbf{q},\omega)}\right] \longrightarrow \text{Loss function}$$

$$A(\mathbf{q},\omega) = Im\left[\varepsilon_M(\mathbf{q},\omega)\right] \longrightarrow \text{Optical absorption}$$

$$\sigma(\mathbf{q},\omega) = -i\frac{\omega}{4\pi}\left[\varepsilon_M(\mathbf{q},\omega) - 1\right] \longrightarrow \text{Conductivity}$$

...

TDDFT in a nutshell

Key quantity: dielectric function

$$\varepsilon_{\mathbf{G}\mathbf{G}'}(\mathbf{q},\omega)$$

Procedure of **`exciting`**

$$\delta n = \chi_0 \, \delta V_{KS} \qquad\qquad f_{xc} = \frac{\delta v_{xc}}{\delta n}$$

TDDFT in a nutshell

Key quantity: dielectric function

$$\varepsilon_{\mathbf{G}\mathbf{G}'}(\mathbf{q},\omega)$$

Procedure of **`exciting`**

$$\delta n = \chi_0 \, \delta V_{KS} \qquad f_{xc} = \frac{\delta v_{xc}}{\delta n}$$

$$\chi = \chi_0 + \chi_0 (v + f_{xc}) \chi$$

TDDFT in a nutshell

Key quantity: dielectric function

$$\varepsilon_{\mathbf{G}\mathbf{G}'}(\mathbf{q},\omega)$$

Procedure of **`exciting`**

$$\delta n = \chi_0 \, \delta V_{KS} \qquad f_{xc} = \frac{\delta v_{xc}}{\delta n}$$

$$\chi = \chi_0 + \chi_0 (v + f_{xc}) \chi$$

$$\varepsilon^{-1} = 1 + v\chi$$

TDDFT in a nutshell

Non-interacting KS response

$$\chi_{0,\mathbf{G}\mathbf{G}'}(\mathbf{q},\omega) = \frac{1}{N_{\mathbf{k}}\Omega}\sum_{nm\mathbf{k}}\frac{f_{n\mathbf{k}} - f_{m\mathbf{k}+\mathbf{q}}}{\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}+\mathbf{q}} + \omega + i\eta} M_{nm\mathbf{k}}(\mathbf{q},\mathbf{G}) M^*_{nm\mathbf{k}}(\mathbf{q},\mathbf{G}')$$

TDDFT in a nutshell

Non-interacting KS response

$$\chi_{0,\mathbf{G}\mathbf{G}'}(\mathbf{q},\omega) = \frac{1}{N_{\mathbf{k}}\Omega}\sum_{nm\mathbf{k}}\frac{f_{n\mathbf{k}} - f_{m\mathbf{k}+\mathbf{q}}}{\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}+\mathbf{q}} + \omega + i\eta} M_{nm\mathbf{k}}(\mathbf{q},\mathbf{G}) M^*_{nm\mathbf{k}}(\mathbf{q},\mathbf{G}')$$

Matrix elements of plane-waves / momentum

$$M_{nm\mathbf{k}}(\mathbf{q},\mathbf{G}) = \frac{\sqrt{4\pi}}{|\mathbf{q}+\mathbf{G}|}\left\langle n\mathbf{k}\left|e^{-i(\mathbf{q}+\mathbf{G})\mathbf{r}}\right|m\mathbf{k}+\mathbf{q}\right\rangle \xrightarrow{\mathbf{q},\mathbf{G}\to 0} \sqrt{4\pi}\frac{\mathbf{p}_{nm\mathbf{k}}}{\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}}}$$

Input parameters

```
<input>
   <title>Loss function</title>
   <structure ... />
   <groundstate ... />

   <xs
      xstype ="TDDFT"
      ngridk="8 8 8"
      nempty="30"
      gqmax="2.0"
      broad="0.004">

      <energywindow
         intv="0.0 2.0"
         points="500" />

      <tddft fxctype="RPA"/>

      <qpointset>
          <qpoint>
             0.0 0.0 0.0
          </qpoint>
      </qpointset>
   </xs>
</input>
```

Input parameters

```
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      <qpointset>
          <qpoint>
             0.0 0.0 0.0
          </qpoint>
      </qpointset>
   </xs>
</input>
```
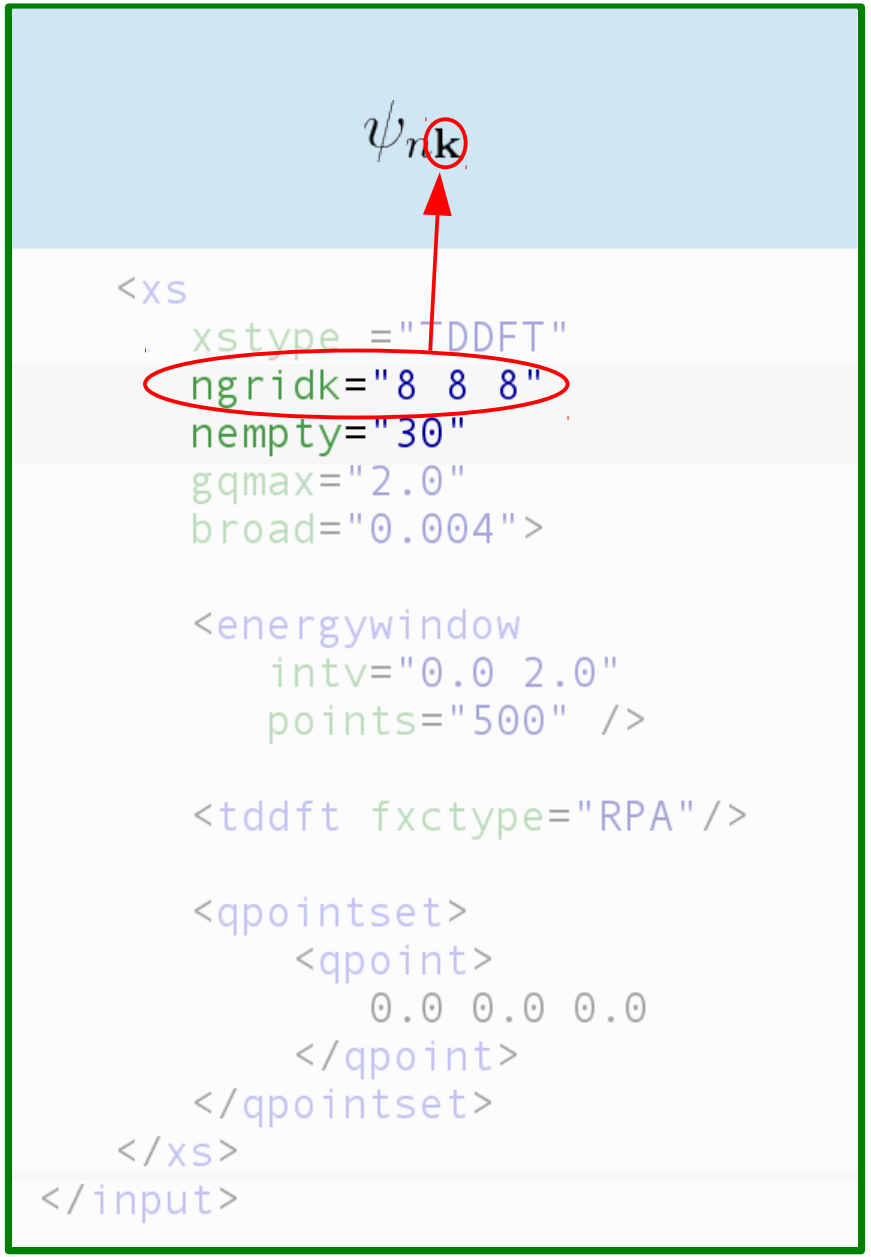

xstype: “TDDFT” or “BSE”

Input parameters



One-step ground-sate calculation triggered with saved density and potential

`ngridk` same meaning as `groundstate/@ngridk` attribute

Input parameters

$\psi_{n\mathbf{k}}$

```
<xs
   xstype ="TDDFT"
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   broad="0.004">

   <energywindow
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   <qpointset>
      <qpoint>
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      </qpoint>
   </qpointset>
</xs>
</input>
```

One-step ground-sate calculation triggered with saved density and potential

`ngridk` same meaning as `groundstate/@ngridk` attribute

`nempty` number of empty bands

$$n_{\max} = N_{\text{occ}} + N_{\text{empty}}$$

Input parameters

$$\langle n\mathbf{k} \left| e^{-i(\mathbf{q}+\mathbf{G})\mathbf{r}} \right| m\mathbf{k}+\mathbf{q} \rangle$$

$$\langle n\mathbf{k} \left| \hat{\mathbf{p}} \right| m\mathbf{k} \rangle$$

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</input>
```

Matrix elements of plane-waves and momentum calculated

Input parameters

$$\langle n\mathbf{k} \left| e^{-i(\mathbf{q}+\mathbf{G})\mathbf{r}} \right| m\mathbf{k}+\mathbf{q} \rangle$$

$$\langle n\mathbf{k} \left| \hat{\mathbf{p}} \right| m\mathbf{k} \rangle$$

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Matrix elements of plane-waves and momentum calculated

$$|\mathbf{G}| < G_{max}$$

Input parameters

$$\langle n\mathbf{k} \left| e^{-i(\mathbf{q}+\mathbf{G})\mathbf{r}} \right| m\mathbf{k}+\mathbf{q} \rangle$$

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Matrix elements of plane-waves and momentum calculated

$$|\mathbf{G}| < G_{max}$$

Input parameters

$$\chi_{0,\mathbf{GG'}}(\mathbf{q},\omega) = \frac{1}{N_\mathbf{k}\Omega}\sum_{nm\mathbf{k}}\frac{f_{n\mathbf{k}} - f_{m\mathbf{k+q}}}{\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k+q}} + \omega + i\eta} M_{nm\mathbf{k}}(\mathbf{q},\mathbf{G}) M^*_{nm\mathbf{k}}(\mathbf{q},\mathbf{G'})$$

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   <xs
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      nempty="30"
      gqmax="2.0"
      broad="0.004">

      <energywindow
         intv="0.0 2.0"
         points="500" />

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      <qpointset>
          <qpoint>
             0.0 0.0 0.0
          </qpoint>
      </qpointset>
   </xs>
</input>
```

Adiabatic turning-on of the perturbation

Finite lifetime broadening: scattering processes and finite experimental resolution

Input parameters

$$\chi_{0,\mathbf{GG'}}(\mathbf{q},\omega) = \frac{1}{N_\mathbf{k}\Omega}\sum_{nm\mathbf{k}}\frac{f_{n\mathbf{k}} - f_{m\mathbf{k+q}}}{\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k+q}} + \omega + i\eta} M_{nm\mathbf{k}}(\mathbf{q},\mathbf{G}) M^*_{nm\mathbf{k}}(\mathbf{q},\mathbf{G'})$$

```
<xs
   xstype="TDDFT"
   ngridk="8 8 8"
   nempty="30"
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   <energywindow
      intv="0.0 2.0"
      points="500" />

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   <qpointset>
      <qpoint>
         0.0 0.0 0.0
      </qpoint>
   </qpointset>
</xs>
</input>
```

Input parameters

$$\chi_{0,\mathbf{GG'}}(\mathbf{q},\omega) = \frac{1}{N_{\mathbf{k}}\Omega}\sum_{nm\mathbf{k}}\frac{f_{n\mathbf{k}} - f_{m\mathbf{k+q}}}{\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k+q}} + \omega + i\eta}M_{nm\mathbf{k}}(\mathbf{q},\mathbf{G})M^{*}_{nm\mathbf{k}}(\mathbf{q},\mathbf{G'})$$

```
<xs
   xstype ="TDDFT"
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   nempty="30"
   gqmax="2.0"
   broad="0.004">

   <energywindow
      intv="0.0 2.0"
      points="500" />

   <tddft fxctype="RPA"/>

   <qpointset>
      <qpoint>
         0.0 0.0 0.0
      </qpoint>
   </qpointset>
</xs>
</input>
```

Input parameters

$$f_{xc} = \frac{\delta v_{xc}}{\delta n}$$

```
<input>
  <xs
    xstype ="TDDFT"
    ngridk="8 8 8"
    nempty="30"
    gqmax="2.0"
    broad="0.004">

    <energywindow
      intv="0.0 2.0"
      points="500" />

    <tddft fxctype="RPA"/>

    <qpointset>
      <qpoint>
        0.0 0.0 0.0
      </qpoint>
    </qpointset>
  </xs>
</input>
```

$$\chi = \chi_0 + \chi_0(v + f_{xc})\chi$$

Input parameters

$$f_{xc} = \frac{\delta v_{xc}}{\delta n}$$

```
<xs
   xstype ="TDDFT"
   ngridk="8 8 8"
   nempty="30"
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   <energywindow
      intv="0.0 2.0"
      points="500" />

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   <qpointset>
      <qpoint>
         0.0 0.0 0.0
      </qpoint>
   </qpointset>
</xs>
</input>
```

fxctype

- “RPA”
- “LRCstatic”
- “LRCdyn”
- ”ALDA”
- “MB1”

Input parameters

$$f_{xc} = \frac{\delta v_{xc}}{\delta n}$$

```
<input>
  <xs
    xstype ="TDDFT"
    ngridk="8 8 8"
    nempty="30"
    gqmax="2.0"
    broad="0.004">

    <energywindow
      intv="0.0 2.0"
      points="500" />

    <tddft fxctype="RPA"/>

    <qpointset>
      <qpoint>
        0.0 0.0 0.0
      </qpoint>
    </qpointset>
  </xs>
</input>
```

fxctype

- "RPA"
- "LRCstatic"
- "LRCdyn"
- "ALDA"
- "MB1"

$$f_{xc}^{\mathrm{RPA}} \equiv 0$$

$$f_{xc}^{\mathrm{LRCstatic}} = -\frac{\alpha}{4\pi} v$$

$$f_{xc}^{\mathrm{LRCdyn}} = -\frac{1}{4\pi}(\alpha + \beta\omega^2) v$$

$$f_{xc}^{\mathrm{ALDA}} = \left.\frac{\delta v_{xc}^{\mathrm{LDA}}}{\delta n}\right|_{n=n_{gs}(\mathbf{r})}$$

Input parameters

$$f_{xc} = \frac{\delta v_{xc}}{\delta n}$$

```
<input>
  <xs
    xstype ="TDDFT"
    ngridk="8 8 8"
    nempty="30"
    gqmax="2.0"
    broad="0.004">

    <energywindow
      intv="0.0 2.0"
      points="500" />

    <tddft fxctype="RPA"/>

    <qpointset>
      <qpoint>
        0.0 0.0 0.0
      </qpoint>
    </qpointset>
  </xs>
</input>
```

tddft/@intraband:

- "true"
- "false"

$$\chi_{0,\mathbf{GG'}}(\mathbf{q},\omega) = \frac{1}{N_{\mathbf{k}}\Omega}\sum_{nm\mathbf{k}} \ldots$$

tddft/@drude:

- "wp wtau"

$$\chi_0^{\text{Drude}}(\omega) = \frac{1}{\omega}\frac{\omega_p^2}{\omega + i\omega_\tau}$$

Output files

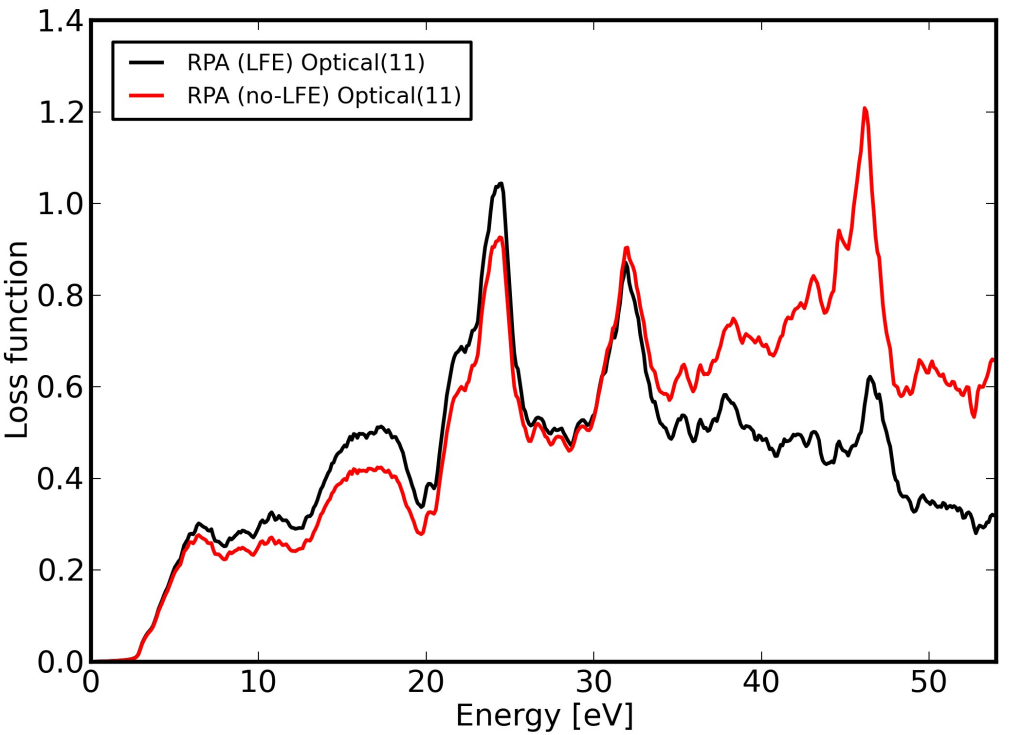
`PROP_[NLF_]FXCXXX_OCYY_QMTZZZ.OUT[.xml]`

`PROP`	calculated property, e.g. EPSILON, LOSS, SIGMA, …
`XXX`	exchange correlation kernel, e.g. RPA, ALDA, …
`YY`	tensor component, e.g. 11, 12, 13, 21, 22,...
`ZZZ`	**q** points from the `qpointset` block, e.g. 001, 002, …
`NLF`	local-field effects neglected
`.xml`	xml output

Example: EPSILON_FXCRPA_OC11_QMT001.OUT

Example

```
<input>

 <title>Loss function of Ag</title>
 <structure> ...</structure>
 <groundstate> ... </groundstate>

 <xs
    xstype ="TDDFT"
    ngridk="16 16 16"
    vkloff="0.097 0.273 0.493"
    nempty="30"
    gqmax="2.0"
    broad="0.004"
    tevout="true">

  <energywindow
     intv="0.0 2.0"
     points="500" />

  <tddft fxctype="RPA"/>

  <qpointset>
    <qpoint> 0.0 0.0 0.0 </qpoint>
  </qpointset>

 </xs>
</input>
```



Parallelization

Wavefunctions

k-points: MPI
diagonalization: OpenMP

Matrix elements

k-points: MPI

Kohn-Sham response

frequencies: MPI

xc kernel, Dyson equation

frequencies (MPI)

Thanks!