Update on linear-response TDDFPT codes

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Outline

1. Absorption spectroscopy for finite systems: turboTDDFT code

2. Electron Energy Loss Spectroscopy for periodic solids: turboEELS code

3. Inelastic Neutron Scattering in periodic solids

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1. Absorption spectroscopy for finite systems: turboTDDFT code

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Developers' of the turboTDDFT code*

- Stefano Baroni (2004 - present), coordinator.
- Ralph Gebauer (2004 - present)
- Brent Walker (2004 - 2006)
- Dario Rocca (2006 - 2012)
- Baris Malcioglu (2008 - 2010)
- Simon Binnie (2010 - 2012)
- Xiaochuan Ge (2010 - 2014)
- Iurii Timrov (2010 - present)
- Tommaso Gorni (2014 - present)

*The timing is approximate.

Structure of the turboTDDFT code

- **turboTDDFT** is the open-source code for a calculation of the absorption spectra of finite systems containing up to several hundreds of atoms.
- **turboTDDFT** is based on the Time-Dependent Density Functional Perturbation Theory (TDDFPT).
- There are two flavours of the **turboTDDFT** code:
  - `turbo_lanczos.x`
  - `turbo_davidson.x`

  Both codes give the same absorption spectrum but have their Pros and Cons (next slide).

  Both codes are residing in the same directory `TDDFPT/src` and are using many same routines in `TDDFPT`, `PH`, and `PW`.

Pros & Cons

Liouville-Lanczos approach

(**turbo_lanczos.x**)

Solution method: Computes an absorption coefficient directly without computing eigenvalues of the Liouvillian, using the Lanczos recursion algorithm.

☺ Spectrum is computed *at once* in the large frequency range (only one Lanczos recursion is needed).

☺ No empty states (use of DFPT techniques)

☺ No large-matrix inversions (Lanczos recursion instead)

☹ No information about individual electronic transitions

Casida-Davidson approach

(**turbo_davidson.x**)

Solution method: Computes eigenvalues of the Liouvillian by diagonalizing it, and then computes the absorption coefficient from them.

☹ Many eigenvalues are required to compute spectrum in the large energy range.

☺ No empty states (use of DFPT techniques)

☹ Large-matrix diagonalization (Davidson-like solver) $\Rightarrow$ high RAM

☺ Information about individual electronic transitions

Restrictions of the turboTDDFT code

- Linear-response regime.
- Use of only the Γ point (`gamma_only`) and supercells.
  $k = 0$ or many k points algorithm seems working well, but nobody have ever performed an extensive testing. It may be used at your own risk!
- Supports NC and US pseudopotentials. No PAW.
- Hybrid functional are supported, but only with NC PP's (*S. Binnie*).
- Adiabatic XC kernels only.
- Only unpolarized systems are allowed (`nspin=1`). Collinear (`nspin=2`) and noncollinear (`noncolin=.true.`) cases are not supported.
- No relativistic effects: spin-orbit coupling is not implemented.
- Parallelization: PWs, bands, task groups (only NC PP's, experimental). No OpenMP.

New features of the turboTDDFT code (I)

What new was done during 2014?

- Implemented a subroutine (`compute_d0psi_rs`) which calculates $\hat{P}_c\,\mathbf{r}\,\varphi_v(\mathbf{r})$ in the real space. This can be used only for finite systems! This is need when hybrid functionals are used, because the usual trick with the commutator $[\hat{H}, \hat{\mathbf{r}}]$ misses a contribution from the exact exchange potential. (*X. Ge*)
- Tried to implement $[\hat{V}_{\mathrm{EXX}}, \hat{\mathbf{r}}]$ for hybrid functionals using 1) spherically truncated Coulomb potential, and 2) Martyna-Tuckerman partitioning of the Coulomb potential, but have not succeeded so far to agree with the real-space method mentioned above. (*I. Timrov*)
- Martyna-Tuckerman periodic-boundary correction for charged molecules is now supported. (*I. Timrov*)

  Recall: The supercell size must be *at least two times* larger than the molecule size!

New features of the turboTDDFT code (II)

What new was done during 2014?

- turboTDDFT has been interfaced with the `Environ` module. (*I. Timrov*)

  I. Timrov, O. Andreussi, A. Biancardi, N. Marzari, S. Baroni, *J. Chem. Phys.* (2015), in press. $\longrightarrow$ poster

  What does it give? $\Rightarrow$ A possibility to compute optical absorption spectra of molecules in various solvents (e.g. water) using the revised self-consistent continuum solvation model of O. Andreussi *et al.* (JCP **136**, 064102 (2012)).

  However, the `Environ` module is not publically distributed yet (coordinated by O. Andreussi).

Ongoing work:

- Speeding up TDDFPT calculations with optimally reduced PW basis sets. (*T. Gorni, I. Timrov, S. Baroni*) $\longrightarrow$ poster

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turboEELS code (I)

- **Developers:** Iurii Timrov, Nathalie Vast, Ralph Gebauer, Stefano Baroni
- turboEELS was created during I. Timrov's PhD thesis at the École Polytechnique (Paris) during 2009-2013, in strong collaboration with SISSA and ICTP.

  I. Timrov's PhD thesis:
  https://pastel.archives-ouvertes.fr/pastel-00823758

  See also: *I. Timrov, N. Vast, R. Gebauer, and S. Baroni, PRB* **88**, *064301 (2013).*
- turboEELS is not yet public. Is planned to be public during next months (will be re-implemented in the SVN version of Quantum ESPRESSO on http://qe-forge.org/gf/project/q-e/).

  I. Timrov, N. Vast, R. Gebauer, and S. Baroni, Comput. Phys. Commun., in preparation.

turboEELS code (II)

- **turboEELS** is the code for a calculation of the EELS (IXSS) spectra at $\mathbf{q} \neq \mathbf{0}$ of periodic solids, containing up to several hundreds of atoms in the unit cell.
- **turboEELS** $\Rightarrow$ calculation of plasmons and single-particle excitations.
- **turboEELS** is based on the Liouville-Lanczos approach to TDDFPT.
- **turboEELS** resides in the directory `TDDFPT/src`, and is strongly linked to turboTDDFT, `PH`, and `PW`.

Restrictions of the turboEELS code

- Linear-response regime.
- Uses general **k** points algorithm and allows finite transferred momenta ($\mathbf{q} \neq \mathbf{0}$).
- Supports NC and US pseudopotentials. No PAW.
- No hybrid functionals.
- Adiabatic XC kernels only.
- Unpolarized (`nspin=1`) and noncollinear (`noncolin=.true.`) cases are allowed. Collinear case (`nspin=2`) is not supported.
- Relatifictic effects: spin-orbit coupling is supported (only with NC PP's). Application to Bi: *I. Timrov, N. Vast, R. Gebauer, and S. Baroni, in preparation.*
- Symmetry is exploited (small group of **q**).
- Parallelization: PWs, **k** points. No OpenMP.

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Magnons

- **Developers:** Tommaso Gorni, Iurii Timrov, Andrea Dal Corso, Ralph Gebauer, Stefano Baroni
- The project has been started recently. Aim: calculations of magnons (spin waves) at finite **q**, which can be observed in INS experiments.
- Use of the Liouville-Lanczos approach to TDDFPT. It must be extented to magnetic systems (no time-reversal symmetry).
- Noncollinear case (`noncolin=.true.`)
- $+\mathbf{q}$ and $-\mathbf{q}$ perturbations must be treated simultaneously (not present in `PH`), or, equivalently, time-dependent Kohn-Sham equation and the complex-conjugate one must be solved simultaneously.

  Work in progress...