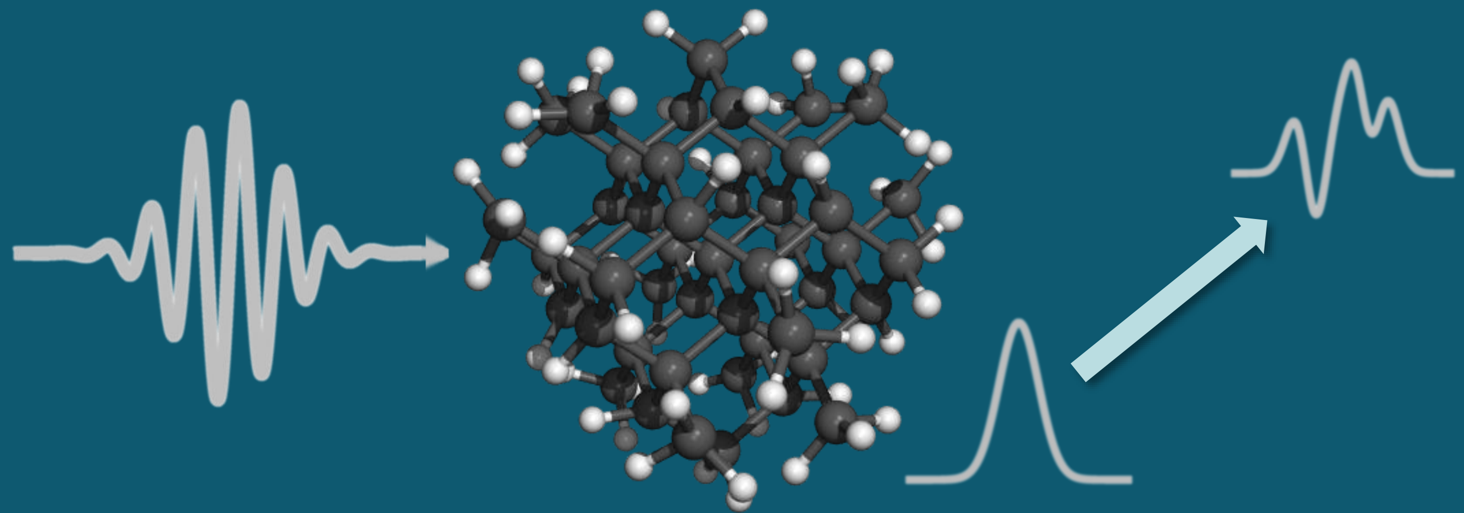
Efficient Implementation of RT-TDDFT on Siesta 4.0

F. D. Vila, M. Tzavala and J. J. Rehr



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Motivation

Why real-time?

Old approaches not well suited for new science:

- XFEL pulsed x-ray sources (FLASH, LCLS)
- Pump-probe experiments
- Interest in time-dependent response
- Direct access to time-domain

Real-time TDDFT advantages

- Can be more efficient than frequency space (large systems)
- Very versatile (pulses, transport, etc)
- More "physical/realistic"
- Easy access to NLO properties

Real-Time Time Dependent DFT

Direct numerical integration of the time-dependent Kohn-Sham equations in a time-dependent external electric field:

$$i\frac{\partial \Psi}{\partial t} = H(t)\,\Psi \qquad \Psi(t) = T\ \exp\left(-i\int_0^t H(t')dt'\right)\Psi(0)$$

$$H = -\frac{1}{2}\nabla^2 + V_{ext}(\mathbf{r},t) + V_H[\rho](\mathbf{r},t) + V_{xc}[\rho](\mathbf{r},t)$$

Optical properties are determined from the total dipole moment:

$$\mathbf{p}(t) = \int \rho(\mathbf{r},t)\,\mathbf{r}\,d^3\mathbf{r}$$

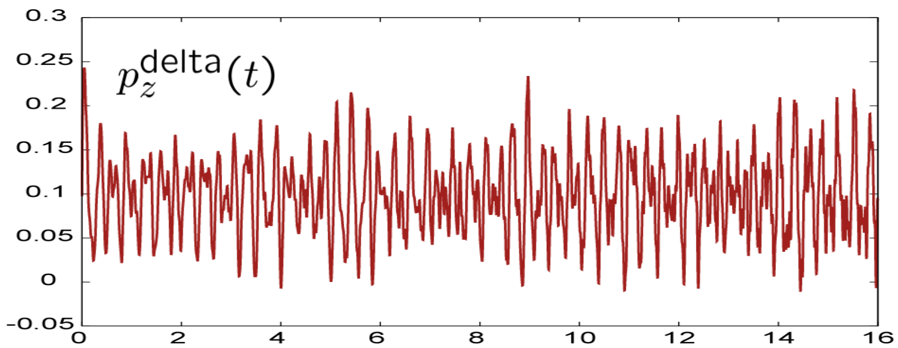
$$\underbrace{\chi_{ij}^{(1)}(\omega) = \delta p_i(\omega)/E_j(\omega) = \alpha_{ij}(\omega)}_{\text{Linear Response}} \qquad \underbrace{\sigma(\omega) \sim \omega\,\mathbf{Im}\,\langle \alpha(\omega)\rangle}_{\text{Absorption}}$$

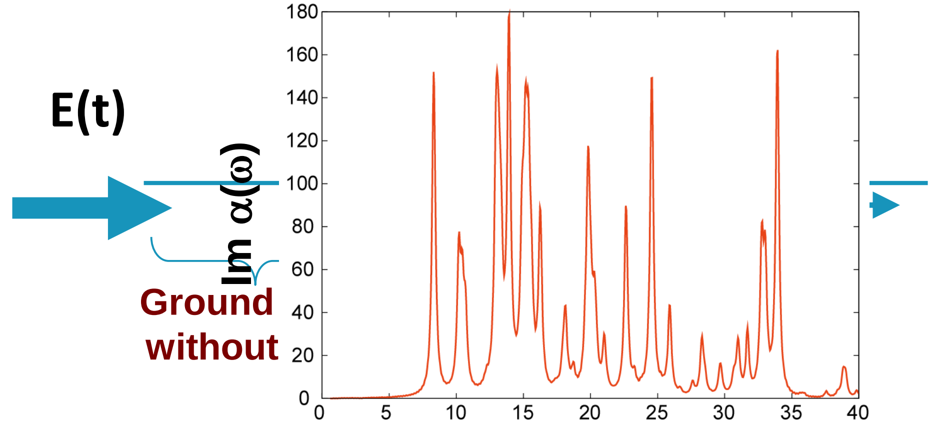
Linear Response | Absorption

Yabana and Bertsch, Phys. Rev. B 54, 4484 (1996)

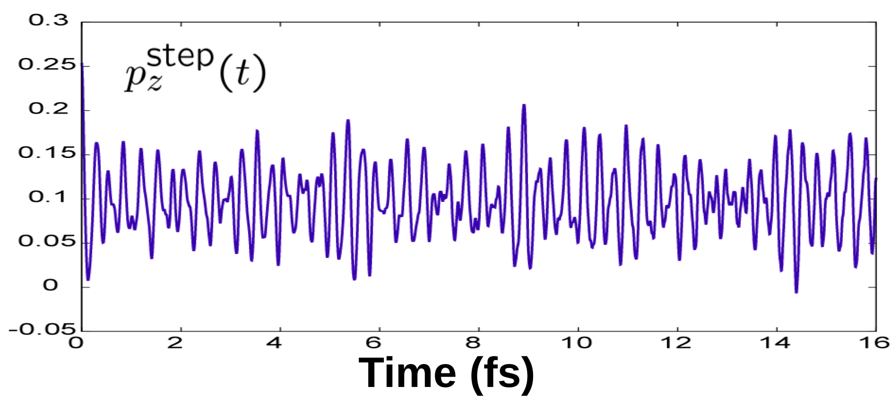
Optical Absorption in CO

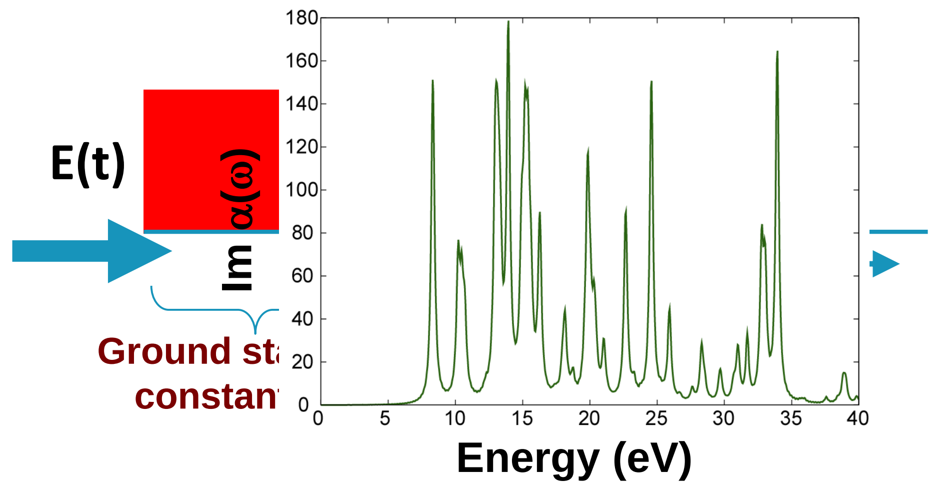
Delta Function (Unit Impulse at t=0)



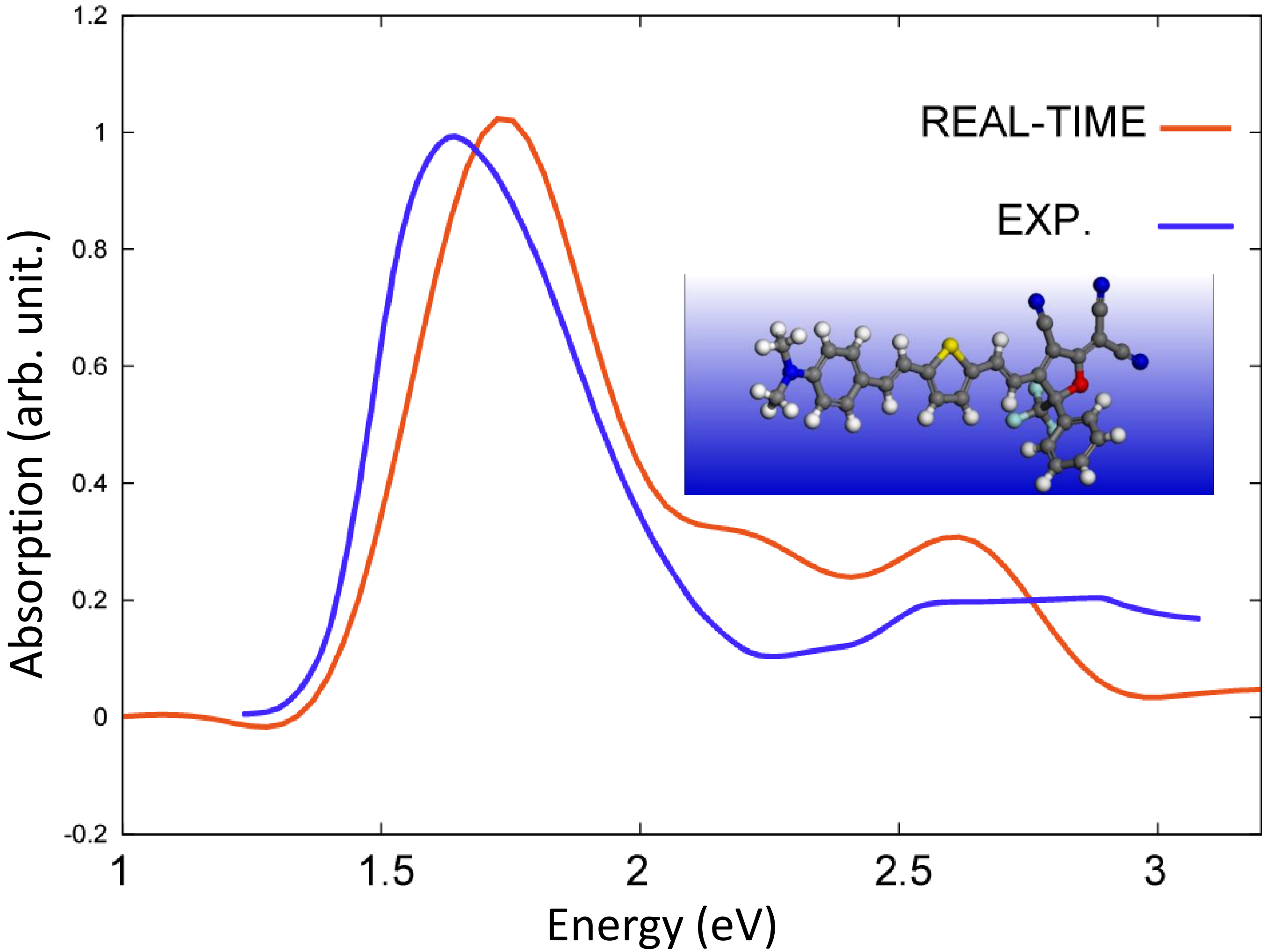


Step Function (Turn-off Constant E at t=0)





Linear Response: Chromophore YLD_156



Real-time (gas) absorption peak

ω_0 = 1.72 eV

(λ_0 = 721 nm)

Expt. ($CHCl_3$) absorption peak

ω_0 = 1.65 eV

(λ_0 = 753 nm)

Numerical Real-Time Evolution in Siesta 4

The ground state density ρ_0, overlap matrix S and Hamiltonian matrix $H(t)$ evaluated at each time-step using SIESTA:

$$i\frac{\partial \Psi}{\partial t} = H(t)\,\Psi \quad \rightarrow \quad i\underbrace{\frac{\partial c(t)}{\partial t}}_{} = S^{-1}H(t)c(t)$$

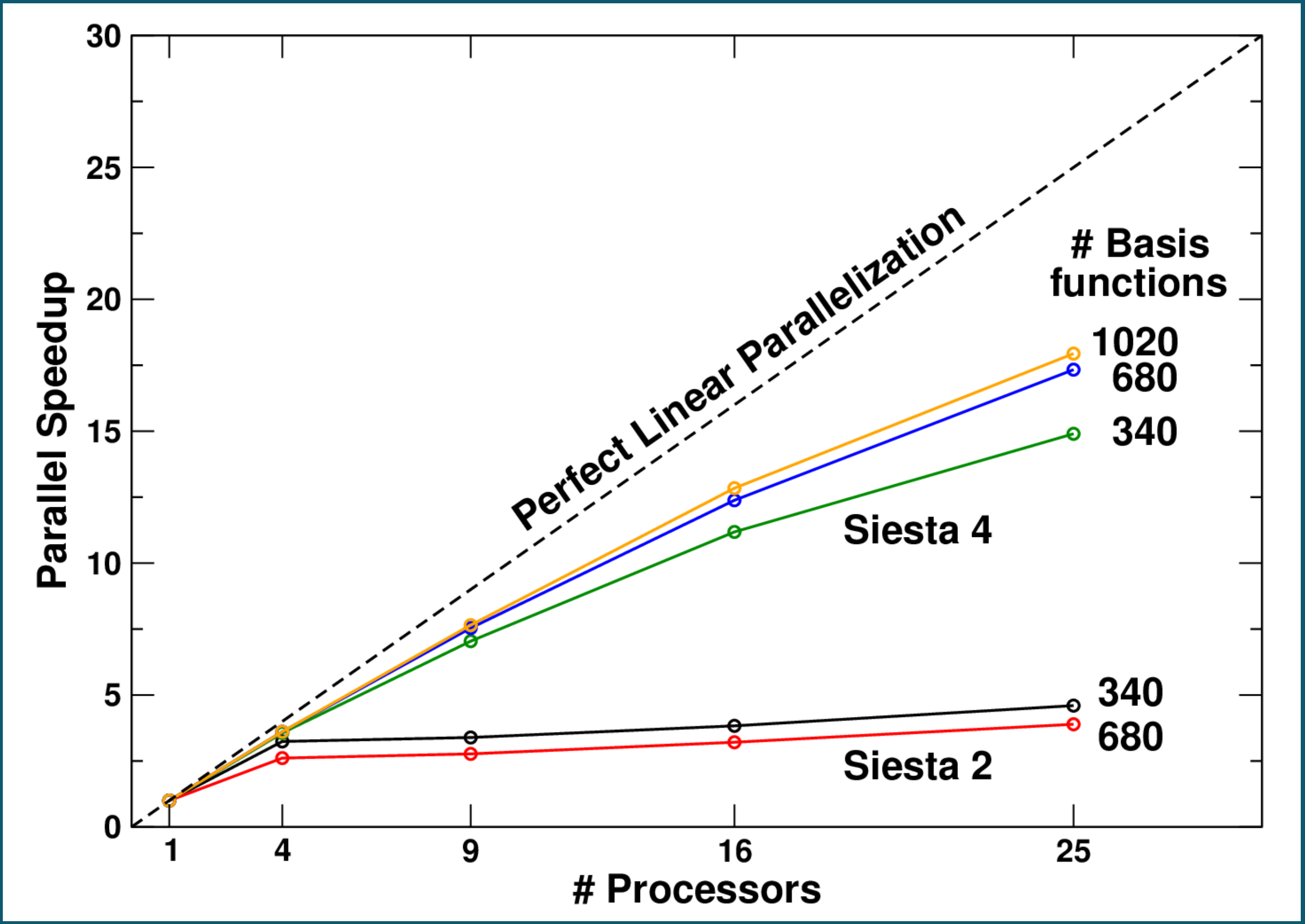
Orbital coefficients

Accurate and stable evolution using Crank-Nicolson propagator:

$$c(t+\Delta t) = \frac{1 - iS^{-1}H(\bar{t})\Delta t/2}{1 + iS^{-1}H(\bar{t})\Delta t/2}c(t) + \mathcal{O}(\Delta t^2)$$

Soler et al. J. Phys.: Condens. Matter 14, 2745 (2002)
Tsolakidis et al. Phys. Rev. B 66, 235416 (2002)

Performance Issues in Siesta 2



Possible solution: Re-implement in Siesta 4

Objectives

- Complete rewrite of original TD code for SPEC collaboration
- Make modular to use on both Siesta 4 and NWChem
- Minimize "points of contact" with external code
- Reduce communications load
- Shift most TD parallel computing to ScaLAPACK
- Investigate scaling of both Siesta and TD components
- Find bottlenecks for different problem sizes

Amdhal's Law, Parallel Speedup and Efficiency

$$S = \frac{T_1}{T_N} = \frac{1}{f + (1 - f)/N}$$

$$E = \frac{S}{N}$$

S: Parallel Speedup
T_i: Time in i Processors
E: Parallel Efficiency
f: Fraction Serial Code
N: # of Processors

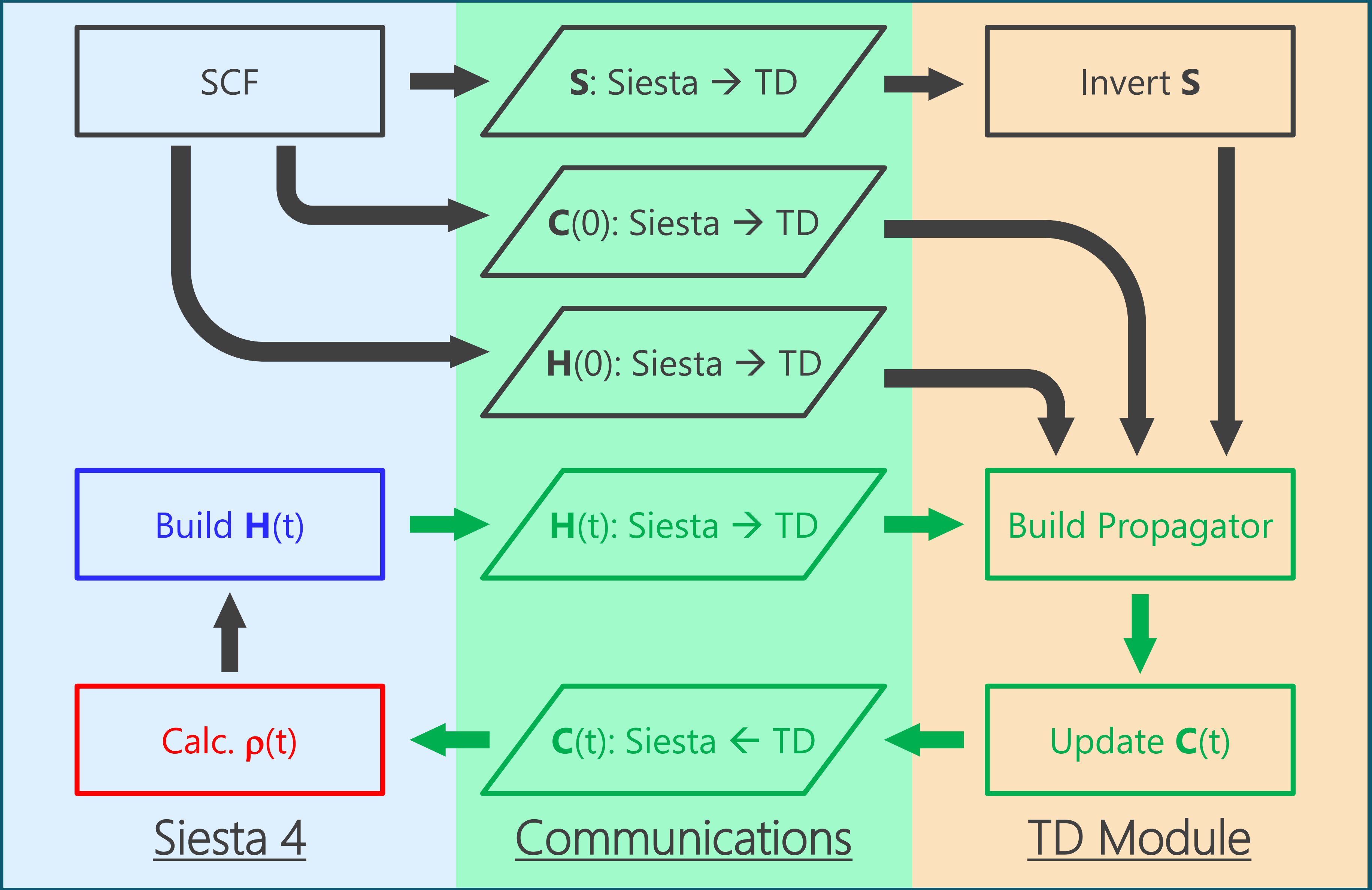
Uphill battle: Diminishing returns unless fully parallel

$$\max_{N \to \infty}(S) = \frac{1}{f}$$

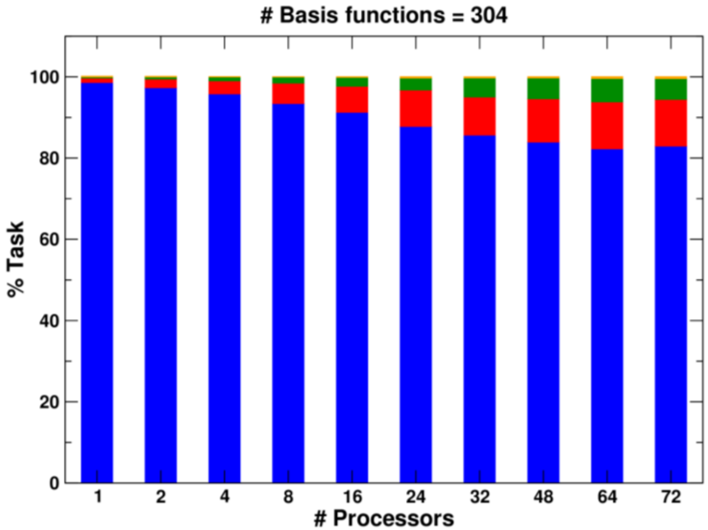
f (%)	$\max(S)$	$N\|_{E=0.6}$
1.00	100	68
0.10	1000	668
0.01	10000	6668

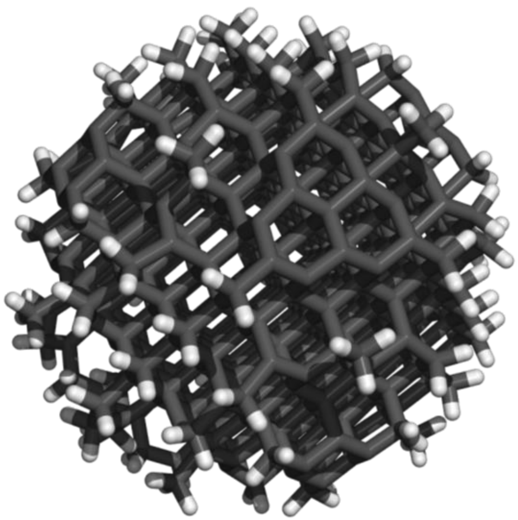
Our Current Aim ←

Data and Process Flow Diagram for New RT-TDDFT



Performance Results: RT-TDDFT Bottlenecks

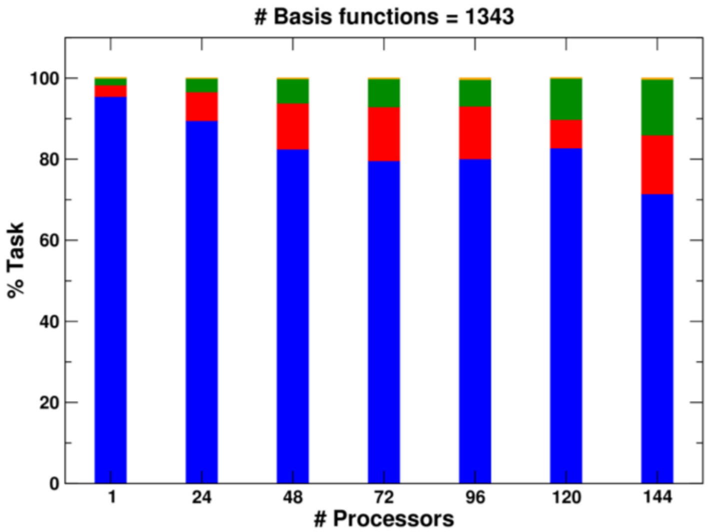


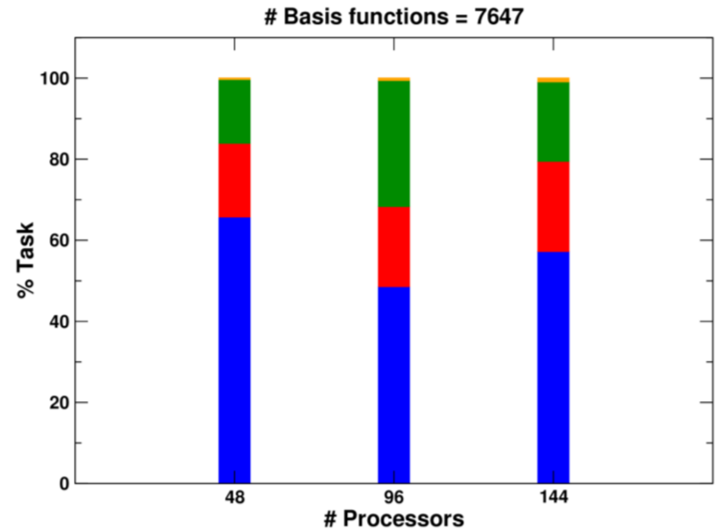


$C_{519}H_{180}$ with DZP Basis set (7647 Orbitals)

41 seconds per time step

8 hours full run





Build H(t)

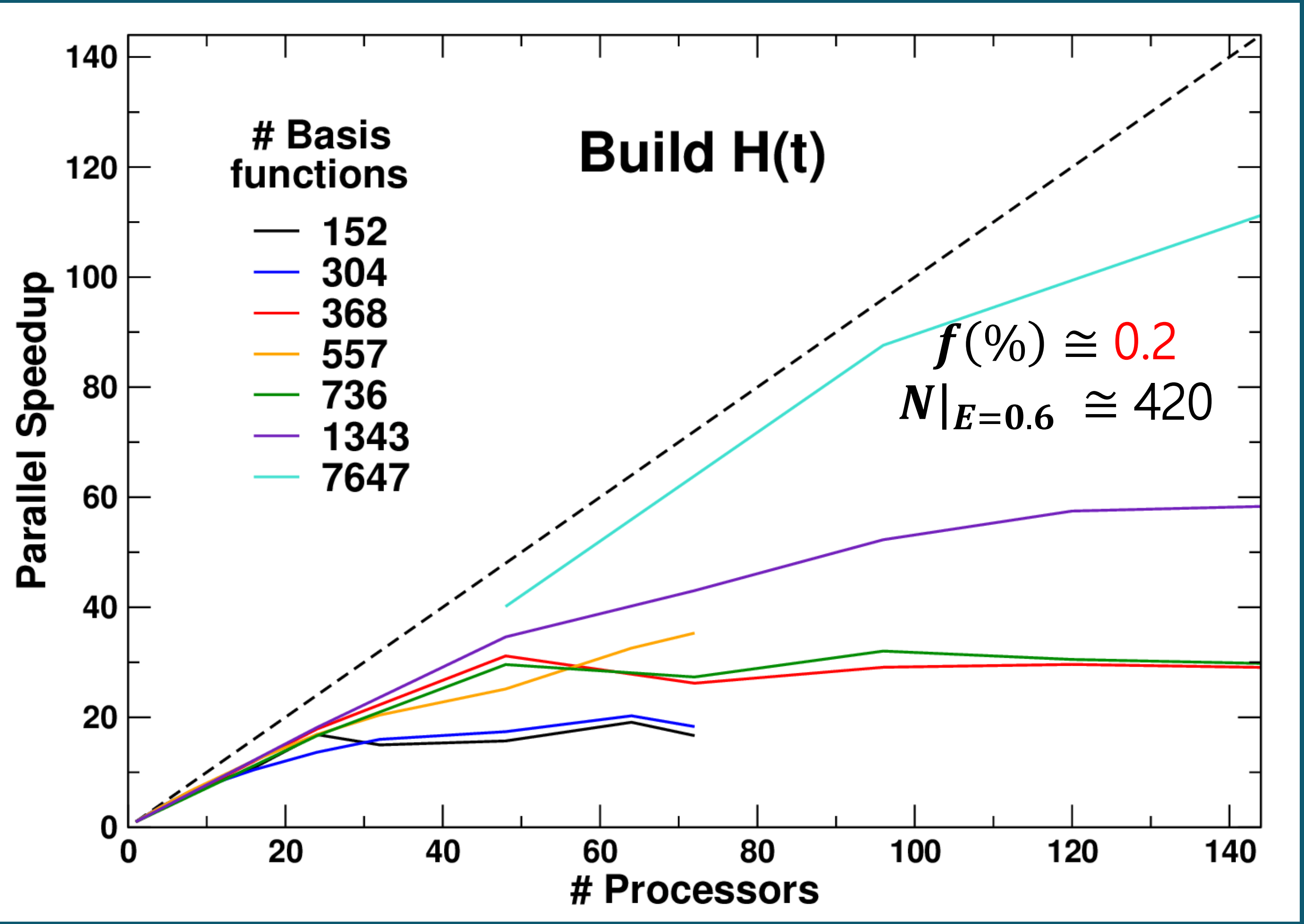
Calc. ρ(t)

Propag. C(t)

Other

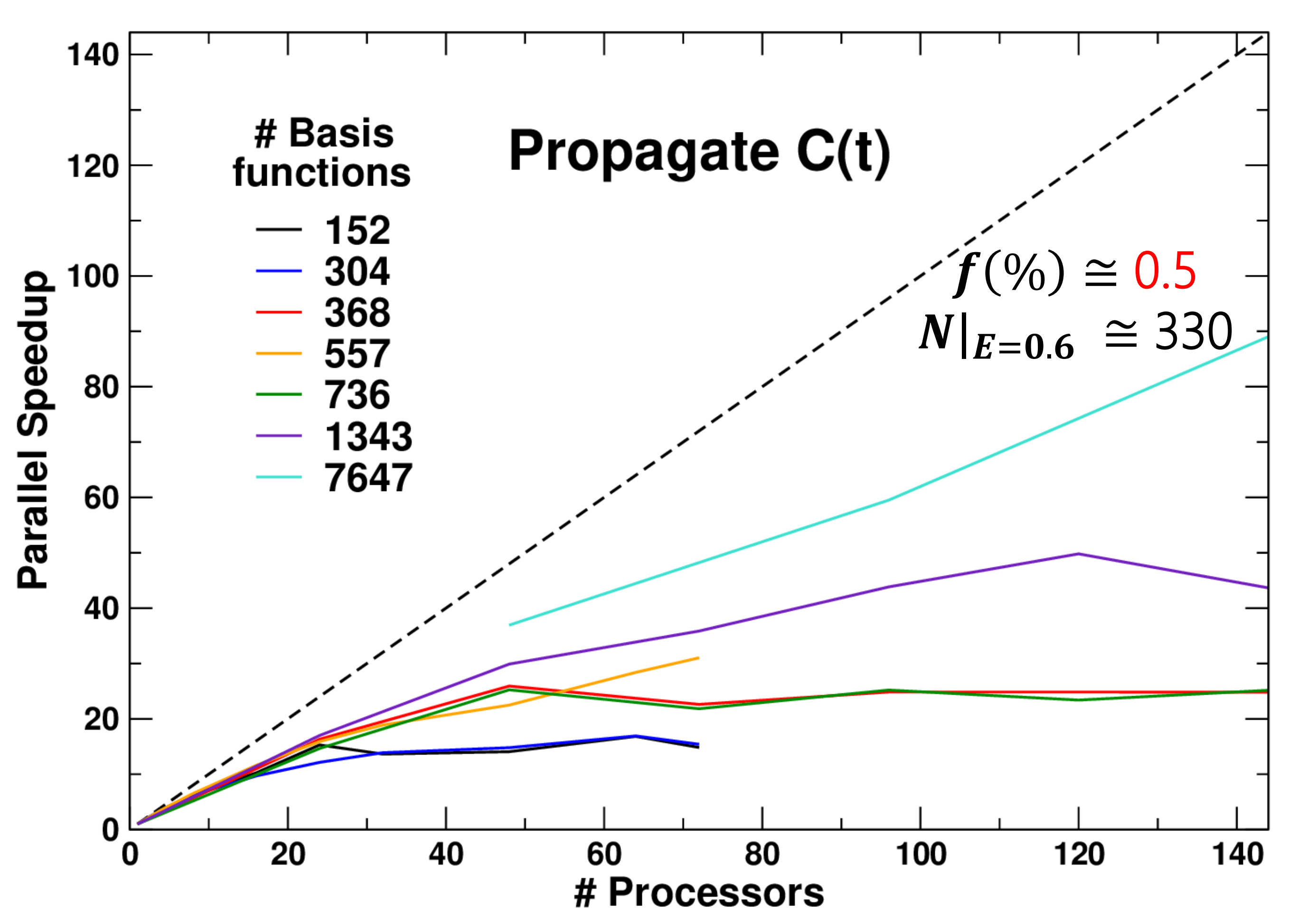
Main bottleneck: Construction of Hamiltonian

Performance Results: Scaling of Siesta 4 Bottleneck



Almost target scaling for internal Siesta bottleneck

Performance Results: Scaling of TD Module Bottleneck



Need scaling improvements for TD bottleneck

Next Steps

What next for better performance?

Streamline communications:
- Remove conversion between Siesta and TD data formats

Try other parallel matrix libraries:
- DPLASMA
- Elemental
- Specifically optimized versions of routines needed (MM, Minv)

Try a better scaling DFT engine
- NWChem (as part of SPEC initiative)

Summary

Developed a new implementation of RT-TDDFT:

- Faster performance:
  - Speedup 10-100 times better than in Siesta 2
  - Smaller communication load

- Better code:
  - Modular implementation
  - Usable with other DFT engines
  - Easier to maintain and expand
  - Makes use of processor optimized ScaLAPACK routines

- Room for improvement from both:
  - Siesta 4
  - TD module

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