RPA correlation energies from Lanczos chains and an optimal basis set: Theory and applications

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Nancy

Outline

- Introduction: Expressing the correlation energy in terms of response functions within the ACFDT
- Methodological development:
  - ✓ Elimination of the summations over empty states
  - ✓ The Lanczos algorithm to compute response functions
  - ✓ Efficient construction of the optimal basis set
- Applications to the binding curves of the benzene dimer
- Conclusions

Correlation energy within the adiabatic connection fluctuation and dissipation theorem

Within this formalism the density functional theory total energy is given by

$$E_{tot}[n] = T_{KS}[\{\phi_v\}] + E_H[n] + E_{e-I}[n] + E_{EXX}[\{\phi_v\}] + E_c[\{\phi_i\}]$$

Exact-exchange energy

Correlation energy

where the the correlation contribution is expressed as a function of the response function χ

$$E_c = -\frac{1}{2\pi}\int_0^1 d\lambda \int_0^\infty du \mathrm{Tr}\{v_c[\chi_\lambda(iu) - \chi_0(iu)]\}$$

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Interacting polarizability given by the Dyson-like equation

Independent-electron polarizability

$$\chi_\lambda = \chi_0 + \chi_0(\lambda v_c + f_{xc}^\lambda)\chi_\lambda$$

The Coulomb potential v_c is adiabatically switched on through the parameter λ

The random phase approximation (RPA)

$$E_c = -\frac{1}{2\pi}\int_0^1 d\lambda \int_0^\infty du \mathrm{Tr}\{v_c[\chi_\lambda(iu) - \chi_0(iu)]\}$$

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The random phase approximation (RPA)

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RPA

$$\chi_\lambda = \chi_0 + \chi_0(\lambda v_c + \cancel{X_c})\chi_\lambda$$

Accurate description of van der Waals dispersion forces

In the RPA the integration on the coupling constant λ can be evaluated analytically

$$E_c^{RPA} = \frac{1}{2\pi}\int_0^\infty du \mathrm{Tr}\{ln[1 - \chi_0(iu)v_c] + \chi_0(iu)v_c\}$$

$$= \frac{1}{2\pi}\int_0^\infty du \mathrm{Tr}\{ln[1 - \underbrace{v_c^{1/2}\chi_0(iu)v_c^{1/2}}_{\widetilde{\varepsilon}(iu)}] + \underbrace{v_c^{1/2}\chi_0(iu)v_c^{1/2}}_{\widetilde{\varepsilon}(iu)}\}$$

Logarithm of a large matrix!!!

Symmetric dielectric function

Challenges in the numerical calculation of RPA correlation energies

$$E_c^{RPA} = \frac{1}{2\pi}\int_0^{\infty} du \mathrm{Tr}\{ln[1 - v_c^{1/2}\chi_0(iu)v_c^{1/2}] + v_c^{1/2}\chi_0(iu)v_c^{1/2}\}$$

$$\chi_0(\mathbf{r},\mathbf{r}',iu) = 4Re\sum_{cv}\frac{\phi_v(\mathbf{r})\phi_c(\mathbf{r})\phi_c(\mathbf{r}')\phi_v(\mathbf{r}')}{iu + (\epsilon_v - \epsilon_c)}$$

v valence state
c conduction (virtual) state

- To evaluate χ_0 it is necessary to compute several conduction states ϕ_c. Convergence is slow with respect to this parameter
- It is necessary to compute an integral between 0 and $+\infty$
- It is necessary to store in memory and compute the logarithm of the large matrix χ_0
- The convergence is slow with respect to the basis set (number of plane-waves) used to represent χ_0

APPLICATIONS TO COMPLEX MATERIALS ARE VERY CHALLENGING

Challenges in the numerical calculation of RPA correlation energies

$$E_c^{RPA} = \frac{1}{2\pi}\int_0^{\infty} du \mathrm{Tr}\{ln[1 - v_c^{1/2}\chi_0(iu)v_c^{1/2}] + v_c^{1/2}\chi_0(iu)v_c^{1/2}\}$$

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v valence state
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- To evaluate χ_0 it is necessary to compute several conduction states ϕ_c. Convergence is slow with respect to this parameter → ELIMINATION OF THE EMPTY STATES (part1)
- It is necessary to compute an integral between 0 and +∞ → LANCZOS ALGORITHM (part 2)
- It is necessary to store in memory and compute the logarithm of the large matrix χ_0
- The convergence is slow with respect to the basis set (number of plane-waves) used to represent χ_0 → OPTIMAL BASIS SET (part 3)

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Elimination of the empty states in an (optimal) basis set representation

$$\chi_0(\mathbf{r},\mathbf{r}',iu) = 4Re\sum_{cv}\frac{\phi_v(\mathbf{r})\phi_c(\mathbf{r})\phi_c(\mathbf{r}')\phi_v(\mathbf{r}')}{iu+(\epsilon_v-\epsilon_c)}$$

In the optimal (as small as possible) basis set $\{U_i\}$ becomes
(for the moment we assume that this basis exists)

$$\begin{aligned}\chi_{ij} &= \int U_i(\mathbf{r})\chi_0(\mathbf{r},\mathbf{r}',iu)U_j(\mathbf{r}')d\mathbf{r}d\mathbf{r}' \\ &= 4Re\sum_{cv}\frac{\langle\phi_v|U_i|\phi_c\rangle\langle\phi_c|U_j|\phi_v\rangle}{iu+(\epsilon_v-\epsilon_c)}\end{aligned}$$

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Projector on the virtual state subspace $\hat{Q}$

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$$= 4Re\sum_{v}\langle\phi_v U_i\hat{Q}[iu+(\epsilon_v-\hat{H})]^{-1}\hat{Q}U_j\phi_v\rangle$$

By using the projector Q it is NOT NECESSARY to explicitly refer to virtual states

$$\hat{Q} = 1-\sum_v|\phi_v\rangle\langle\phi_v|.$$

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$$\chi_0(\mathbf{r},\mathbf{r}',iu) = 4Re\sum_{cv}\frac{\phi_v(\mathbf{r})\phi_c(\mathbf{r})\phi_c(\mathbf{r}')\phi_v(\mathbf{r}')}{iu+(\epsilon_v-\epsilon_c)}$$

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The price to pay for the elimination of the empty states is that now we need to solve a different non-Hermitian linear system for *each* U_j and *iu*

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Lanczos algorithm for response functions

$$\chi_{ij} = 4Re \sum_v \langle \phi_v U_i | \hat{Q}(iu + \epsilon_v - H)^{-1} \hat{Q} | U_j \phi_v \rangle \approx \langle \zeta_{ij}^l | (iu + T_j^l)^{-1} | e^l \rangle$$

LANCZOS

- ❑ The algorithm iteratively reduces the very large matrix *H* to the small tridiagonal matrix *T*
- ❑ The tridiagonal matrix *T* does not depend on *iu*: Once *T* is evaluated we can easily compute χ_{ij} for several values of *iu*
- ❑ The full diagonalization of *H* is avoided: Only the necessary information is extracted
- ❑ No need to deal with non-Hermitian algorithms

-D. Rocca, R. Gebauer, Y. Saad and S. Baroni, *JCP* (2008)
-P. Umari, G. Stenuit, and S. Baroni, PRB (2010)
-H. V. Nguyen, T. A. Pham, D. Rocca, and G. Galli, PRB-RC (2012)

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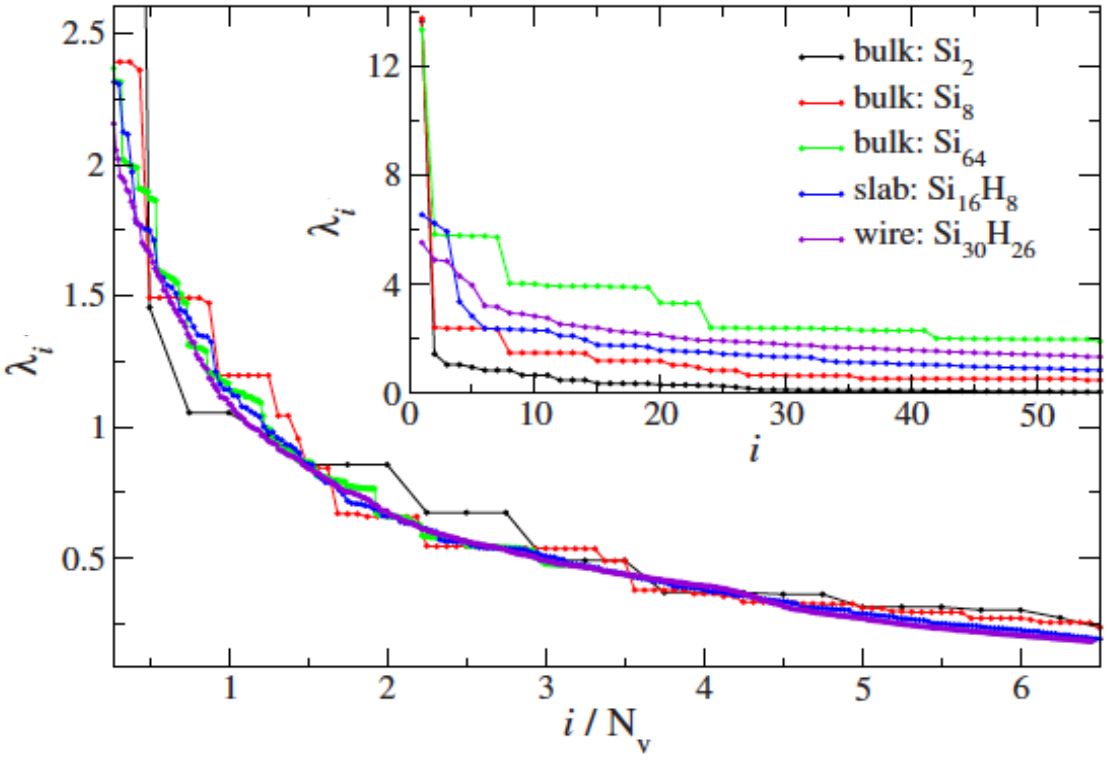
Eigenvalues and eigenvectors of χ_0 as the optimal basis set: Iterative diagonalization

$$\chi_{ij} = \int U_i(\mathbf{r})\chi_0(\mathbf{r},\mathbf{r}',iu)U_j(\mathbf{r}')d\mathbf{r}d\mathbf{r}' = \lambda_i\delta_{ij}$$

ONLY FEW EIGENPAIRS ARE NECESSARY



H. F Wilson, F. Gygi, and G. Galli, *PRB* (2008)
D. Lu, Y. Li, D. Rocca, G. Galli, PRL (2009)

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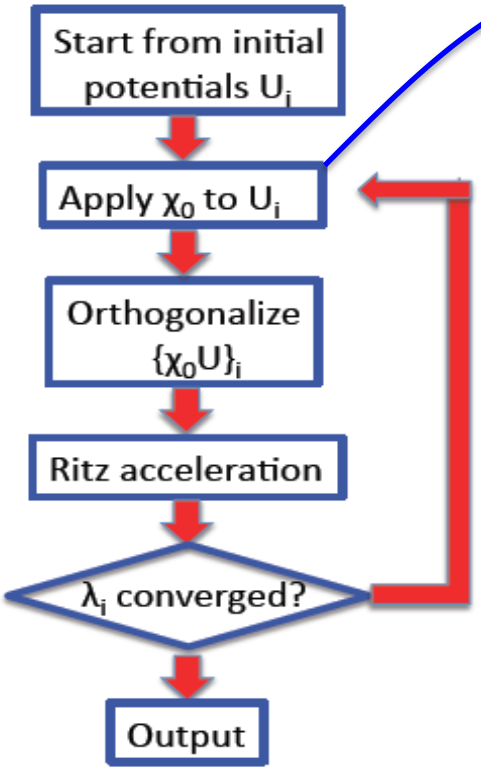
Iterative diagonalization of χ_0



It is not necessary to know explicitly the matrix in order to apply it to a vector

$$\int \chi_0(\mathbf{r},\mathbf{r}',iu)U_j(\mathbf{r}')d\mathbf{r}'$$

$$= 4Re\sum_{cv}\frac{\phi_v(\mathbf{r})\phi_c(\mathbf{r})\langle\phi_c|U_j|\phi_v\rangle}{iu+(\epsilon_v-\epsilon_c)}$$

$$= 4Re\sum_{v}\phi_v(\mathbf{r})\hat{Q}(\mathbf{r})[iu+(\epsilon_v-\hat{H})]^{-1}\hat{Q}|U_j\phi_v\rangle$$

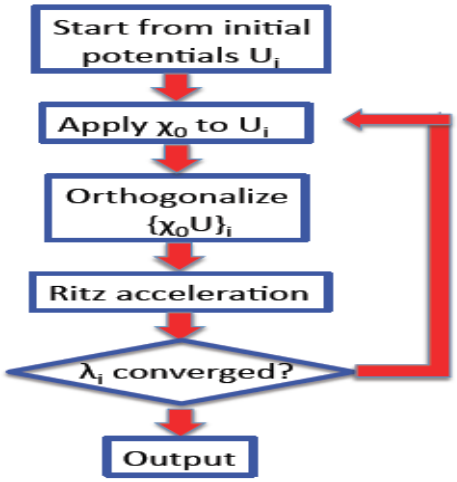
Drawbacks:

- The response function has to be diagonalized for each value of the imaginary energy *iu*
- Every time the response function matrix is applied to a vector a non-Hermitian linear system has to be solved

D. Lu, Y. Li, D. Rocca, G. Galli, PRL (2009)

Use of a frequency independent optimal basis set and the Lanczos algorithm

First step: Diagonalization of χ_0 in the static case (iu=0)



Second step: The static eigenvectors of χ_0 are used as a basis set and the dynamical effects are introduced by the Lanczos algorithmm

$$\chi_{ij} = 4Re\sum_v \langle \phi_v U_i | \hat{Q}(iu + \epsilon_v - H)^{-1}\hat{Q}|U_j\phi_v\rangle$$
$$\approx \langle \zeta_{ij}^l | (iu + T_j^l)^{-1} | e^l \rangle$$

- The static eigenpotentials of χ_0 are a good basis set also at finite frequency. This has been demonstrated for GW calculations
- However, a considerable effort is still necessary to build the basis set

H.-V. Nguyen, T. A. Pham, D. Rocca et G. Galli, PRB-Rap. Comm. (2012)
T. A. Pham, H.-V. Nguyen, D. Rocca et G. Galli, PRB (2013)

Construction of the optimal basis set: A few observations

Let us consider the static response function (*iu=0*)

$$\chi_{ij} = \int U_i(\mathbf{r})\chi_0(\mathbf{r},\mathbf{r}')U_j(\mathbf{r}')d\mathbf{r}d\mathbf{r}'$$
$$= 4Re\sum_{cv}\frac{\langle\phi_v|U_i|\phi_c\rangle\langle\phi_c|U_j|\phi_v\rangle}{\epsilon_v-\epsilon_c}$$

The basis set elements needs to accurately represent the products

$$\phi_v(\mathbf{r})\phi_c(\mathbf{r})$$

The products of valence (v) and conduction (c) states have a strong linear dependence

Keeping into account the weight

$$\frac{1}{\epsilon_v-\epsilon_c}$$

D. Rocca, JCP (2014), to appear in the special issue Advances in DFT Methodology

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$$= 4Re\sum_{cv}\frac{\langle\phi_v|U_i|\phi_c\rangle\langle\phi_c|U_j|\phi_v\rangle}{\epsilon_v-\epsilon_c}$$

Important Contribution!!!

$$\phi_{HOMO}(\mathbf{r})\phi_{LUMO}(\mathbf{r})$$

$$\frac{1}{\epsilon_{HOMO}-\epsilon_{LUMO}}$$

Small contribution (because of the weight)

$$\phi_{HOMO}(\mathbf{r})\phi_{LUMO+50000}(\mathbf{r})$$

Just an example

$$\frac{1}{\epsilon_{HOMO}-\epsilon_{LUMO+50000}} \approx 0$$

D. Rocca, JCP (2014), to appear in the special issue Advances in DFT Methodology

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Just an example

$$\frac{1}{\epsilon_{HOMO}-\epsilon_{LUMO+50000}} \approx 0$$

Only kinetic energy

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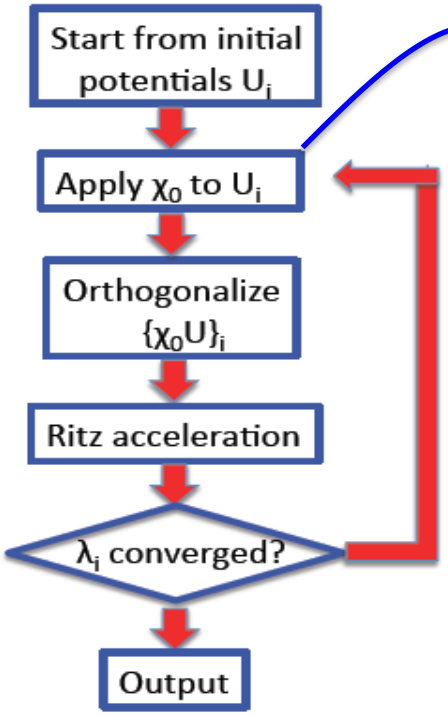
Construction of the optimal basis set by the iterative diagonalization of a χ_0 containing only the kinetic energy term



$$\chi_0 \cdot U_j = \int \chi_0(\mathbf{r},\mathbf{r}',iu)U_j(\mathbf{r}')d\mathbf{r}d\mathbf{r}'$$

$$= 4Re\sum_{cv}\frac{\phi_v(\mathbf{r})\phi_c(\mathbf{r})\langle\phi_c|U_j|\phi_v\rangle}{\epsilon_v-\epsilon_c}$$

$$= 4Re\sum_{v}\phi_v(\mathbf{r})\hat{Q}(\mathbf{r})(\epsilon_v-\hat{H})^{-1}\hat{Q}(\mathbf{r})|U_j\phi_v\rangle$$

$$\approx 4Re\sum_{v}\phi_v(\mathbf{r})\hat{Q}(\mathbf{r})(\epsilon_v+\boxed{\nabla^2/2})^{-1}\hat{Q}(\mathbf{r})|U_j\phi_v\rangle$$

KINETIC ENERGY ONLY

- The kinetic energy is diagonal in reciprocal space (proportional to G^2): It is not necessary to solve a linear system to apply χ_0 and the implementation becomes simple and efficient
- This procedure is used only to build the optimal basis set

D. Rocca, JCP (2014), to appear in the special issue Advances in DFT Methodology

Details of the Implementation

- Implemented in the QUANTUM ESPRESSO package that uses plane-waves and pseudopotentials
- The RPA calculations are performed in a non-self-consistent way starting from LDA or GGA orbitals and energies

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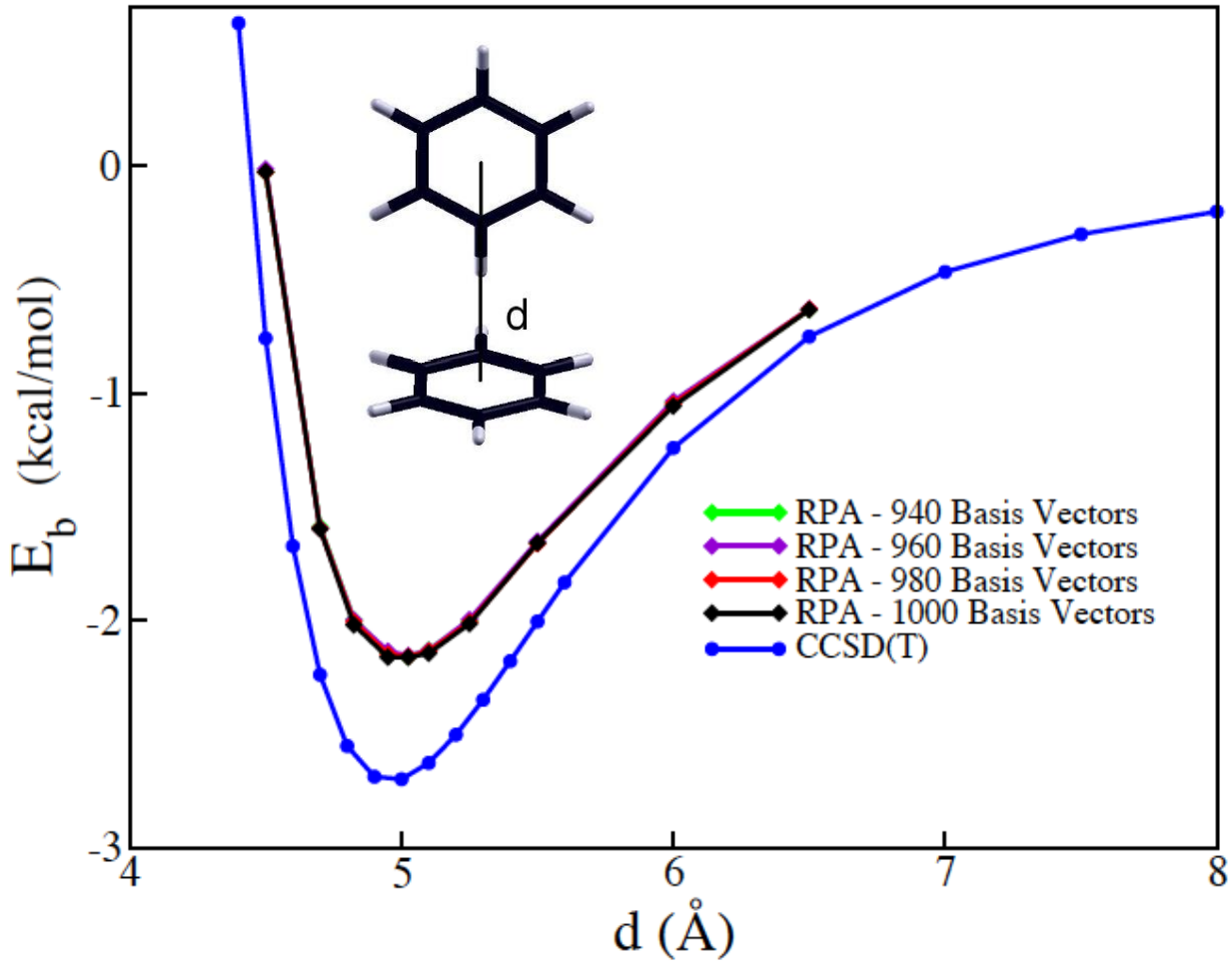
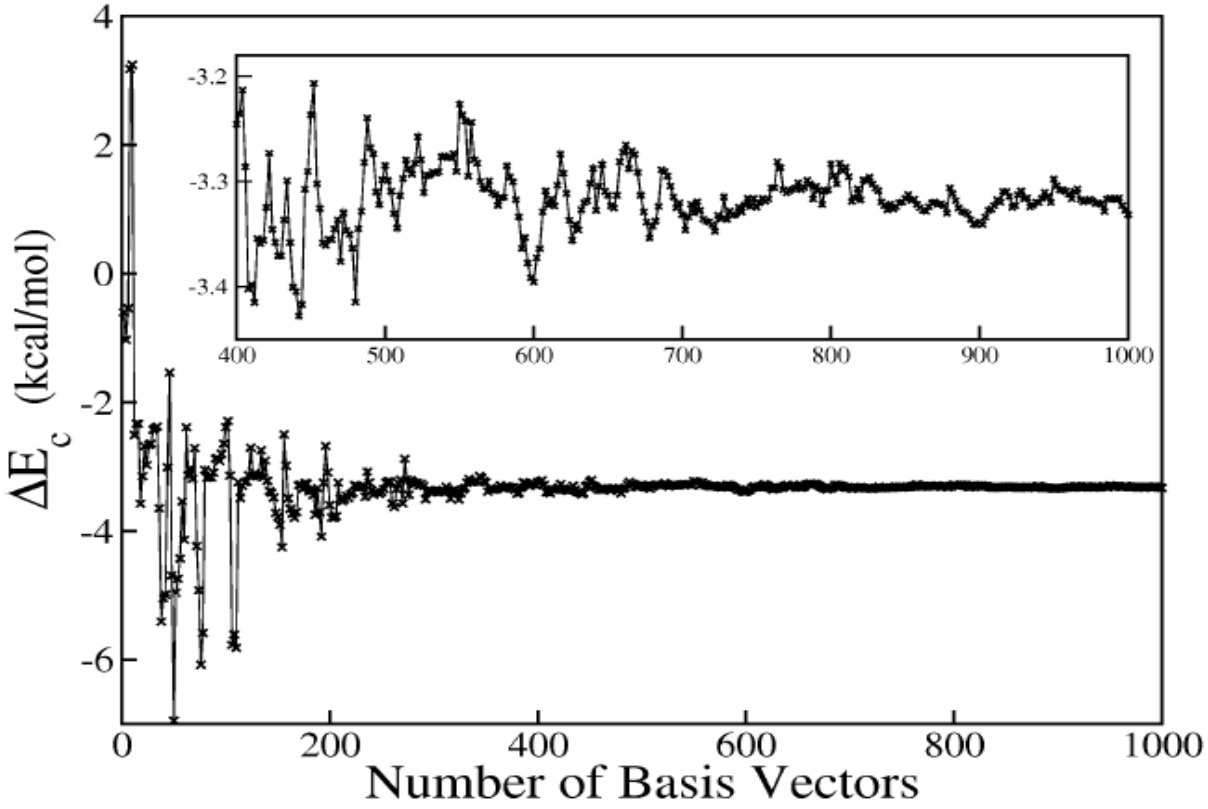
RPA binding curve of the t-shaped configuration of the benzene dimer



- ❑ 60 Ry cut-off
- ❑ 165000 plane-waves for the wavefunctions
- ❑ 1.3 millions of PWs for the charge/potential
- ❑ Curves are shown for different sizes of the optimal basis set
- ❑ RPA gives the correct equilibrium distance and 80% of the coupled clusters binding energy

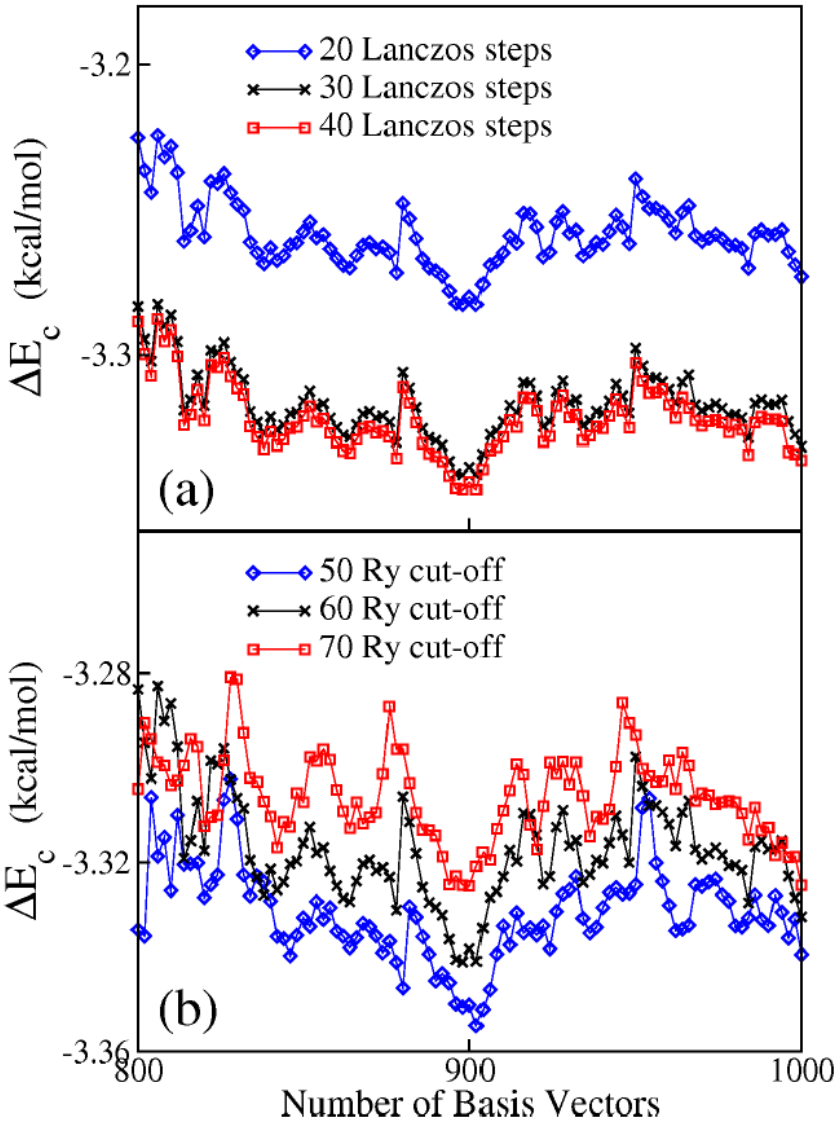
ONLY 1000 BASIS VECTORS ARE SUFFICIENT TO CONVERGE THE BINDING CURVE WITHIN 0.05 kcal/mol

Convergence of the correlation energy difference as a function of the basis set size (d=3.9Å)



- ΔE_c is the difference between the correlation energy of the dimer and the correlation energy of the monomer
- The quantity that actually converges rapidly as a function of the basis set size is the ENERGY DIFFERENCE

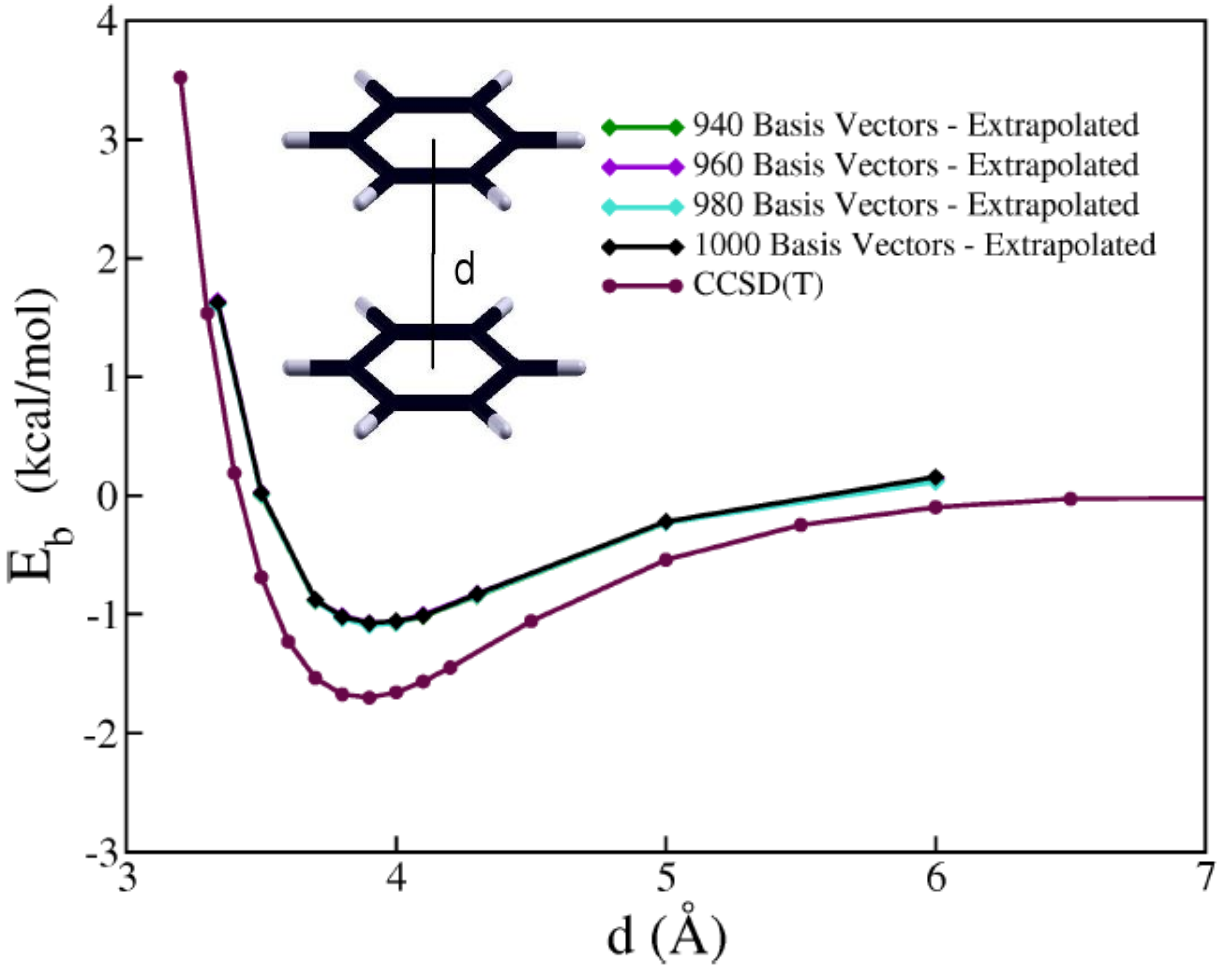
Convergence with respect to the number of Lanczos steps and the kinetic energy cutoff (d=3.9Å)



- By increasing the Lanczos steps from 30 to 40 the energy shifts by only 0.005 kcal/mol
- This means that the 165000x165000 Hamiltonian *H* can be approximated by a 30x30 tridiagonal matrix *T*.

- The kinetic energy cut-off used (60 Ry) for the wavefunctions might be responsible of an error of ≈0.01 kcal/mol

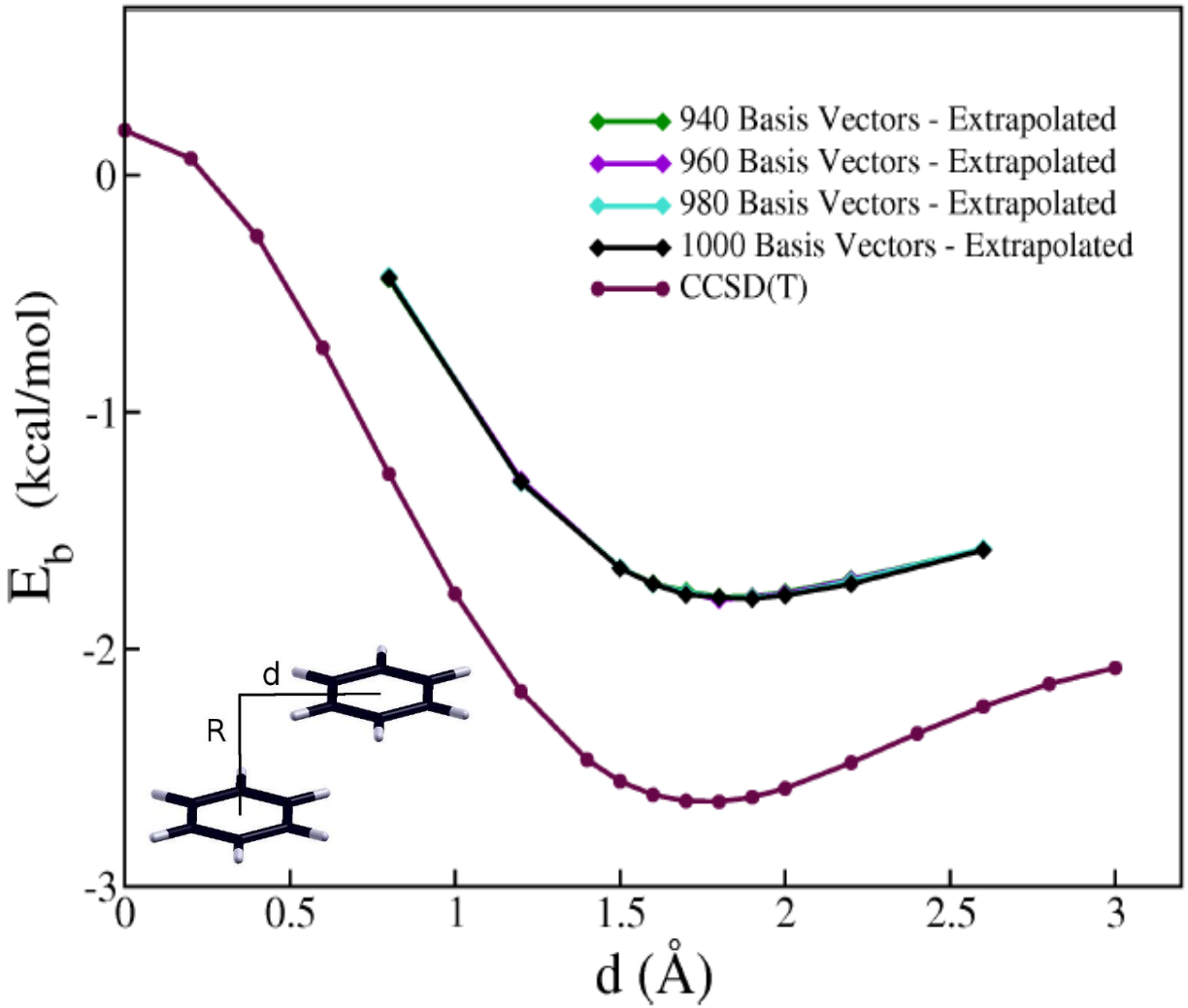
RPA binding curve of the sandwich configuration of the benzene dimer



- ❑ 60 Ry cut-off
- ❑ 123000 plane-waves for the wavefunctions
- ❑ Almost 1 million of PWs for the charge/potential
- ❑ Curves are shown for different sizes of the optimal basis set
- ❑ RPA gives the correct equilibrium distance and 64% of the coupled clusters binding energy

ONLY 1000 BASIS VECTORS ARE SUFFICIENT TO CONVERGE THE BINDING CURVE WITHIN 0.05 kcal/mol

RPA binding curve of the parallel-displaced configuration of the benzene dimer (R=3.4Å)



- ❑ 60 Ry cut-off
- ❑ 143000 plane-waves for the wavefunctions
- ❑ 1.1 millions of PWs for the charge/potential
- ❑ Curves are shown for different sizes of the optimal basis set
- ❑ RPA gives the correct equilibrium distance and 68% of the coupled clusters binding energy

ONLY 1000 BASIS VECTORS ARE SUFFICIENT TO CONVERGE THE BINDING CURVE WITHIN 0.05 kcal/mol

Conclusions

- A new efficient method to compute the RPA correlation energies has been introduced that improves over the shortcomings of previous implementations and avoids extrapolation techniques
- A few examples of the application of this method to non-trivial systems have been shown
- Future work will concern the extension of this method to treat periodic systems and the implementation of methods beyond the RPA

D. Rocca, JCP (2014), to appear in the special issue Advances in DFT Methodology

Eigenvalues and eigenvectors of χ_0 as the optimal basis set: Iterative diagonalization