Electronic excitation in semiconductor nanoparticles: A real-space quasiparticle perspective

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Overview of this talk

- **Objective:**
To describe electron-hole screening without using unoccupied states

- **Motivation:**
Calculation of unoccupied states are expensive. Judicious elimination of these states can lead to faster algorithm (e.g. WEST method by Galli et al.)

- **Strategy:**
Treating electron correlation in real-space representation by using explicitly correlated operators

- **Chemical applications:**
The developed method was used for calculations of optical gap and exciton binding energies

Charge-neutral excitation energy

- Matrix equation for excitation energies

$$\begin{bmatrix} \mathbf{A} & \mathbf{B} \\ \mathbf{B}^* & \mathbf{A}^* \end{bmatrix} \begin{bmatrix} \mathbf{x} \\ \mathbf{y} \end{bmatrix} = \omega \begin{bmatrix} \mathbf{1} & \mathbf{0} \\ \mathbf{0} & \mathbf{-1} \end{bmatrix} \begin{bmatrix} \mathbf{x} \\ \mathbf{y} \end{bmatrix}$$

- Electron-hole interaction kernel (Keh)

$$A_{ia,jb} = \delta_{ij}\delta_{ab}(\epsilon_a - \epsilon_i) + K^{\mathrm{eh}}_{ia,jb} \qquad B_{ia,jb} = K^{\mathrm{eh}}_{ia,jb}$$

- Can be obtained using linear-response (LR-TDDFT), MBPT (BSE), equation-of-motion methods (EOM-CC, EOM-GF), CIS, ADC,…

- Two important considerations:
  - [1] Choice of 1-particles basis functions
  - [2] Choice for treating e-e correlation

Effective single-particle Hamiltonian

- Non-interaction system

$$H_0 = \sum_i^N h_{\text{eff}}(i)$$

$$h_{\text{eff}} = \frac{-\hbar^2}{2m}\nabla^2 + v_{\text{ext}} + v_{\text{eff}}$$

Can be:

$$v_{\text{eff}} \in \{v_{\text{HF}}, v_{\text{KS}}, v_{\text{MBPT}}, v_{\text{ps}}, v_{\text{model}}, \ldots\}$$

$$H_0 |0\rangle = E_0^0 |0\rangle$$

$$H_0 |\Phi_i^a\rangle = E_n^0 |\Phi_i^a\rangle$$

$$\omega_{0n}^0 = E_n^0 - E_0^0$$

- Interacting system

$$W = V_{\text{ee}} - \sum_i^N v_{\text{eff}}(i)$$

$$W = \sum_{i<j}^N w(i,j)$$

$$H = H_0 + W$$

$$H |\Psi_0\rangle = E_0 |0\rangle$$

$$H |\Psi_n\rangle = E_n |\Psi_n\rangle$$

$$\omega_{0n} = E_n - E_0$$

Goal of today's talk: $\omega_{0n} = \omega_{0n}^0 + (?) + (?) + (?)$

Definitions: Correlation operator

- Intermediate normalization condition

$$\langle 0 | \Psi_0 \rangle = 1 \qquad \langle \Phi_i^a | \Psi_n \rangle = 1$$

- Electron-electron correlated is treated by operators that are local in real-space representation

$$\langle \mathbf{x} | G_{0,n} | \mathbf{x}' \rangle = G_{0,n}(\mathbf{x}) \delta(\mathbf{x} - \mathbf{x}')$$

$$| \Psi_0 \rangle = G_0 | 0 \rangle \qquad | \Psi_n \rangle = G_n | \Phi_i^a \rangle$$

- *G* is a two-body operator and is represented by a linear combination of Gaussian-type geminal functions

$$G_{0,n} = \sum_{i<j}^{N} g_{0,n}(i,j) \qquad g(1,2) = \sum_{k=1}^{N_g} b_k e^{-r_{12}^2 / d_k^2}$$

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See also: geminal correlator (Rassolov et al.), NEO-XCHF (Hammes-Schiffer et al.), geminal MCSCF (Varganov & Martinez), trans-correlated Hamiltonian, Jastrow functions in VMC…

Connection to Configuration Interaction (CI)

Configuration interaction (CI):

$$\Psi_{\mathrm{CI}} = \sum_{k}^{N_{\mathrm{CI}}} c_k \Phi_k$$

$\{c_k\}$: finite number of independly optimizable coefficients

Explicitly correlated wave function:

$$\Psi_G = G\Phi_0$$

$$G\Phi_0 = \sum_{k=0}^{\infty} \underbrace{|\Phi_k\rangle\langle\Phi_k|}_{1} G|\Phi_0\rangle = \sum_{k=0}^{\infty} \underbrace{\langle\Phi_k|G|\Phi_0\rangle}_{c_k^{\mathrm{G}}} |\Phi_k\rangle$$

(must be a functional of G)

The explicitly correlated wave function is an infinite-order CI expansion with constrained CI coefficients

Electron-hole interaction kernel

- The excitation energies for the interacting and non-interacting system are related by the W operator

$$E_n = \langle \Phi_i^a \,|\, H_0 + W \,|\, \Psi_n \rangle$$

$$E_0 = \langle 0 \,|\, H_0 + W \,|\, \Psi_0 \rangle$$

$$\omega_{0n} = \omega_{0n}^0 + \langle \Phi_i^a \,|\, W \,|\, \Psi_n \rangle - \langle 0 \,|\, W \,|\, \Psi_0 \rangle$$

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- Expressing in terms of non-interacting states using (G)

$$\omega_{0n} = \omega_{0n}^0 + \langle \Phi_i^a \mid WG_n \mid \Phi_i^a \rangle - \langle 0 \mid WG_0 \mid 0 \rangle$$

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- Expressing in term of vacuum expectation value

$$\omega_{0n} = \omega_{0n}^0 + \langle 0 \mid \{i^\dagger a\} WG_n \{a^\dagger i\} \mid 0 \rangle - \langle 0 \mid WG_0 \mid 0 \rangle$$

Can be simplified using diagrammatic techniques

Contribution from the linked terms

$$\langle 0 | \{i^\dagger a\} W G_n \{a^\dagger i\} | 0 \rangle$$

- Only fully contracted terms have non-zero contribution to this term (Wick's theorem)

- The set of all resulting Hugenholtz diagrams, can be factored into sets of linked and unlinked diagrams

- Subset #1: All linked diagrams (all vertices are connected)
- Subset #2: All unlinked diagrams

$$\langle 0 | \{i^\dagger a\} W G_n \{a^\dagger i\} | 0 \rangle = \langle 0 | \{i^\dagger a\} W G_n \{a^\dagger i\} | 0 \rangle_L + \langle 0 | W G_n | 0 \rangle$$

● ● ●

- Because (algebraically):

$$\langle 0 | \{i^\dagger a\} \{a^\dagger i\} | 0 \rangle = 1$$

$$\langle 0 | \{a^\dagger i\} | 0 \rangle = 0 \quad \text{(normal ordered)}$$

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Contribution from the linked terms

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- The set of all resulting Hugenholtz diagrams, can be factored into sets of linked and unlinked diagrams

- Subset #1: All linked diagrams (all vertices are connected)
- Subset #2: All unlinked diagrams

- Unlinked diagrams in excited state are exactly canceled by the ground state contributions

$$\langle 0 | \{i^\dagger a\} W G_n \{a^\dagger i\} | 0 \rangle - \langle 0 | W G_n | 0 \rangle = \langle 0 | \{i^\dagger a\} W G_n \{a^\dagger i\} | 0 \rangle_L$$

(Important point used in the next slide)

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Elimination of unlinked diagrams

- Adding zero to the expression…

$$\omega_{0n} = \omega_{0n}^{0} + \left[\langle 0|\{i^{\dagger}a\}WG_n\{a^{\dagger}i\}|0\rangle - \langle 0|WG_n|0\rangle\right] + \left[\langle 0|WG_n|0\rangle - \langle 0|WG_0|0\rangle\right]$$

- Only linked terms contribute in the following expression

$$\langle 0|\{i^{\dagger}a\}WG_n\{a^{\dagger}i\}|0\rangle - \langle 0|WG_n|0\rangle = \langle 0|\{i^{\dagger}a\}WG_n\{a^{\dagger}i\}|0\rangle_L$$

- Expression for the excitation energy

$$\omega_{0n} = \omega_{0n}^{0} + \langle 0|\{i^{\dagger}a\}WG_n\{a^{\dagger}i\}|0\rangle_L + \langle 0|W(G_n - G_0)|0\rangle$$

Depends on particle-hole states

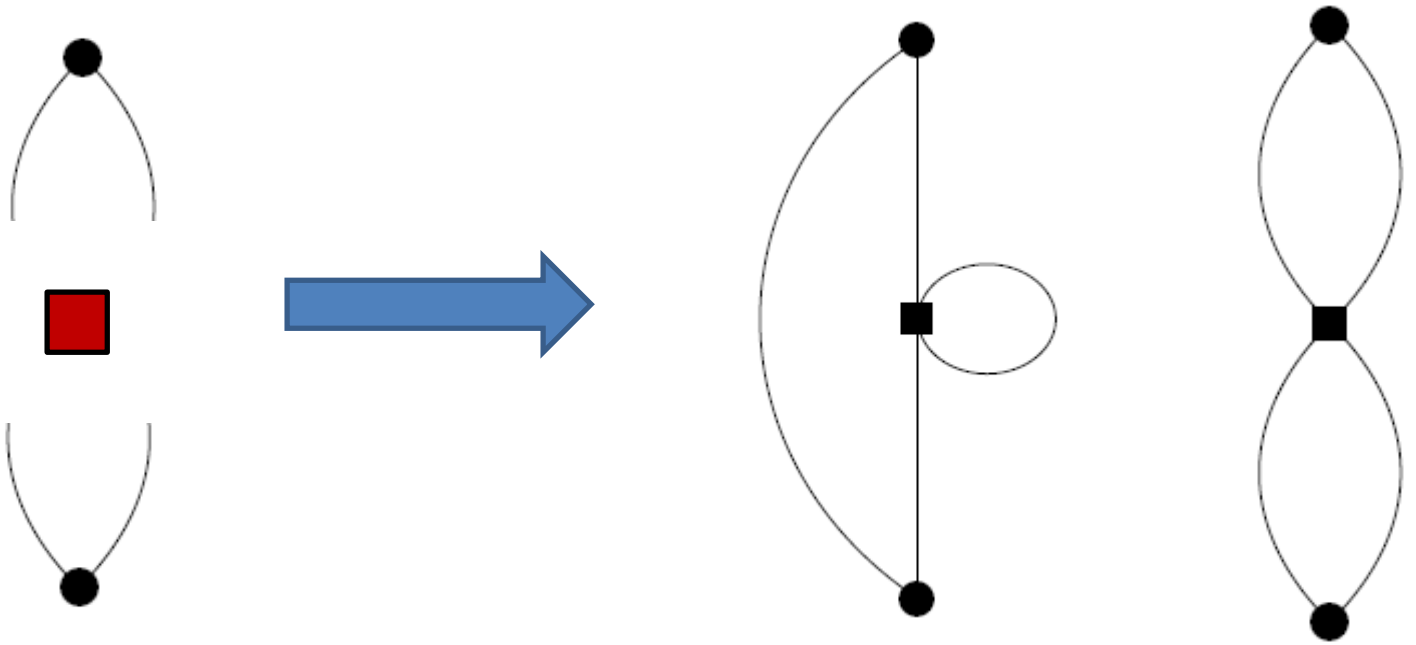
Depends only on occupied states

Generalized Hugenhotlz vertices

$$WG_n = \left[\sum_{i<j}^{N} w(i,j)\right]\left[\sum_{i<j}^{N} g_n(i,j)\right] = \sum_{i<j}^{N} \theta_n(i,j) + \sum_{i<j}^{N} \theta_n(i,j,k) + \sum_{i<j<k<l}^{N} \theta_n(i,j,k,l)$$

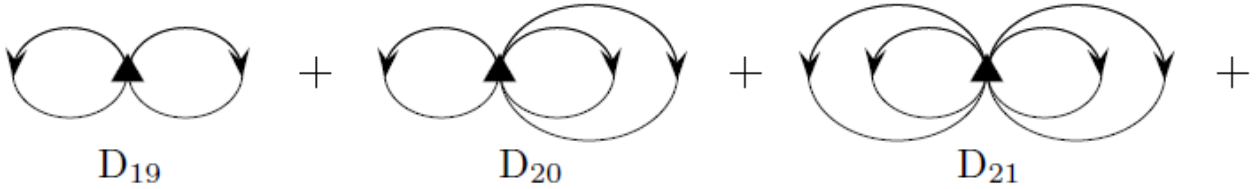
$$WG_n = \Omega_2 + \Omega_3 + \Omega_4$$

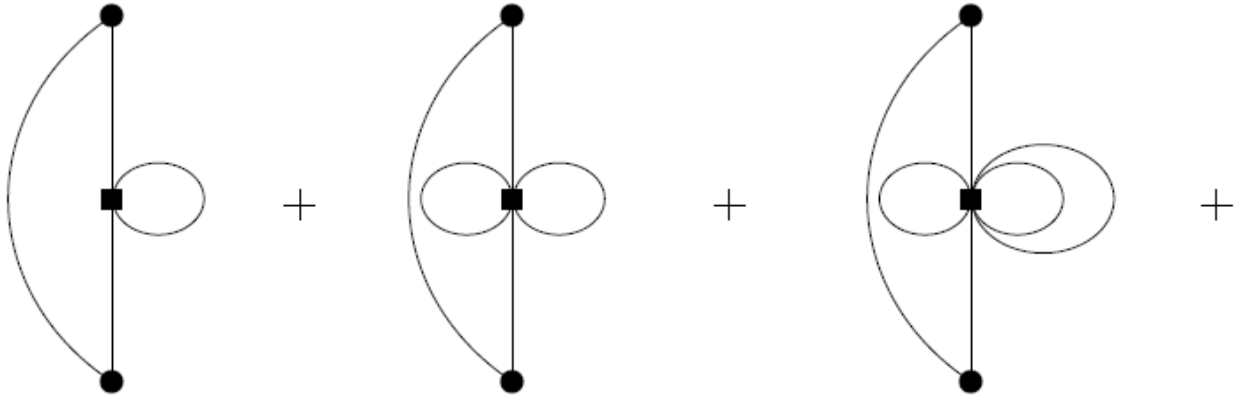
- Product of two two-body operators generates 2, 3, and 4-body operators



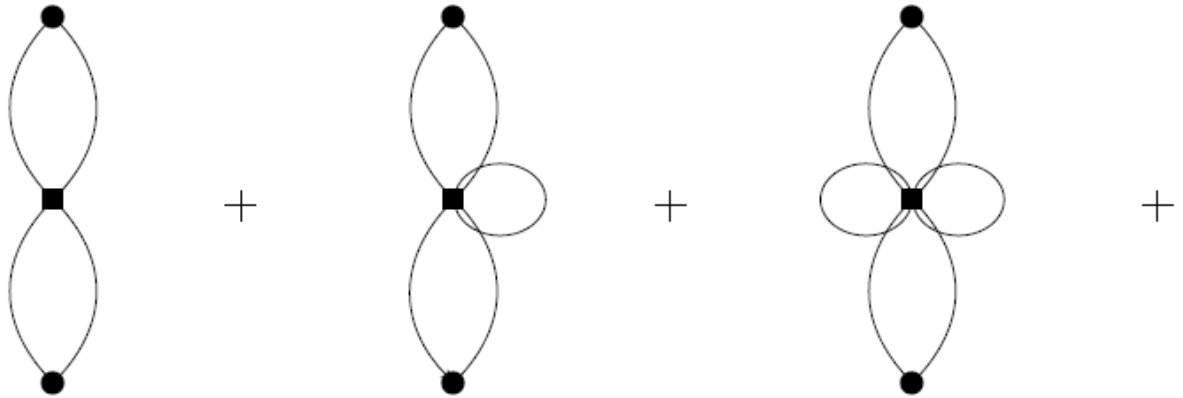
from 2-body vertex (Ω_2)

Contributing diagrams to the excitation energy

$\omega_k = \omega_k^0 +$  D_{19} D_{20} D_{21}

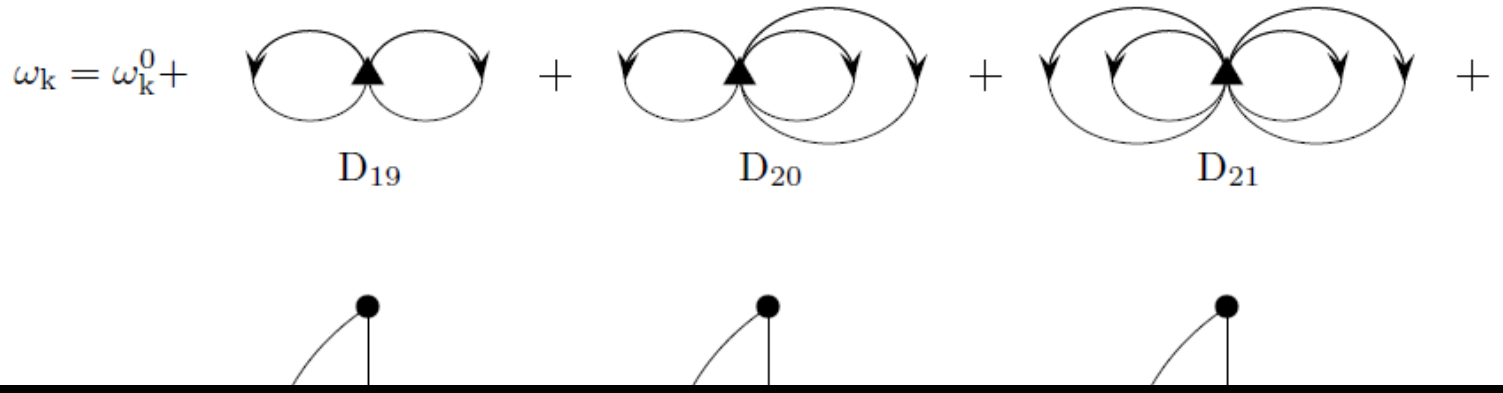


D_{22} D_{23} D_{24}

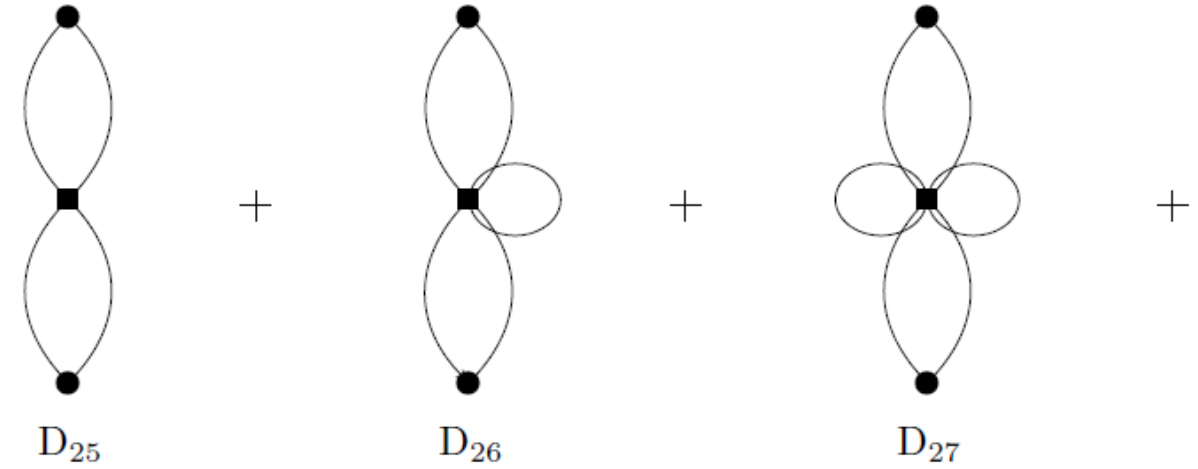


D_{25} D_{26} D_{27}

Contributing diagrams to the excitation energy



Key result:
Only connected diagrams contribute to the exciting energy and electron-hole interaction kernel



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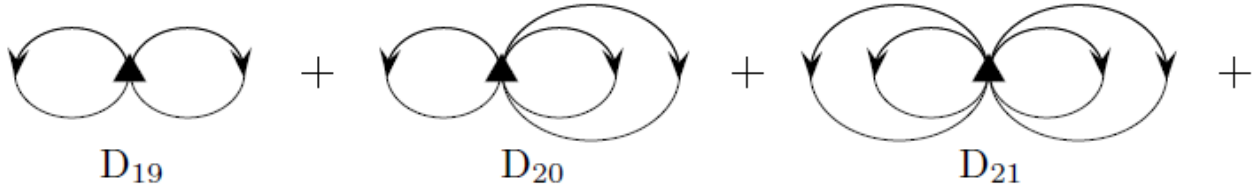
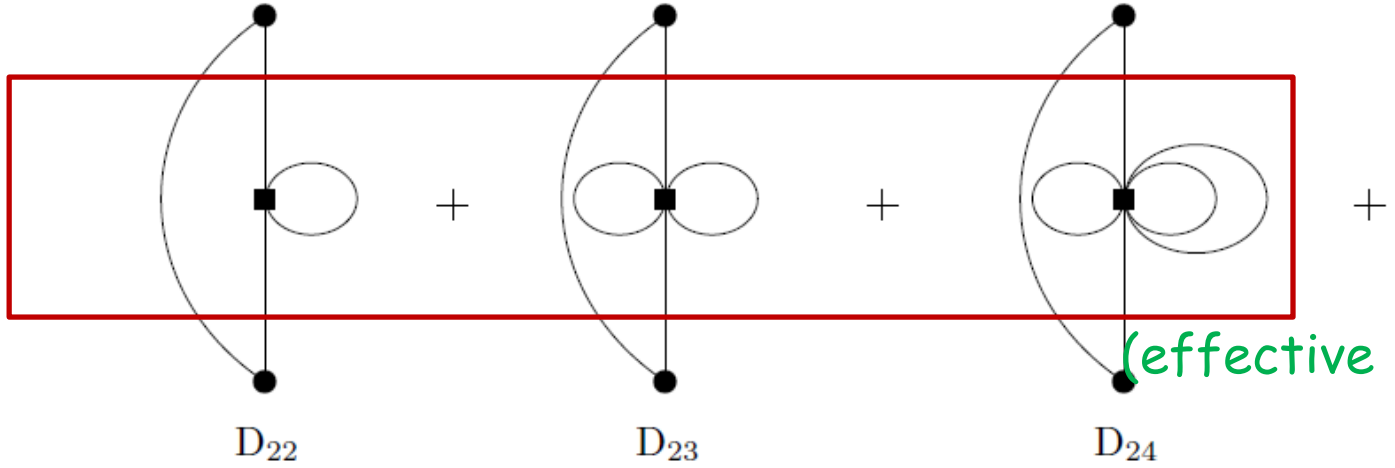
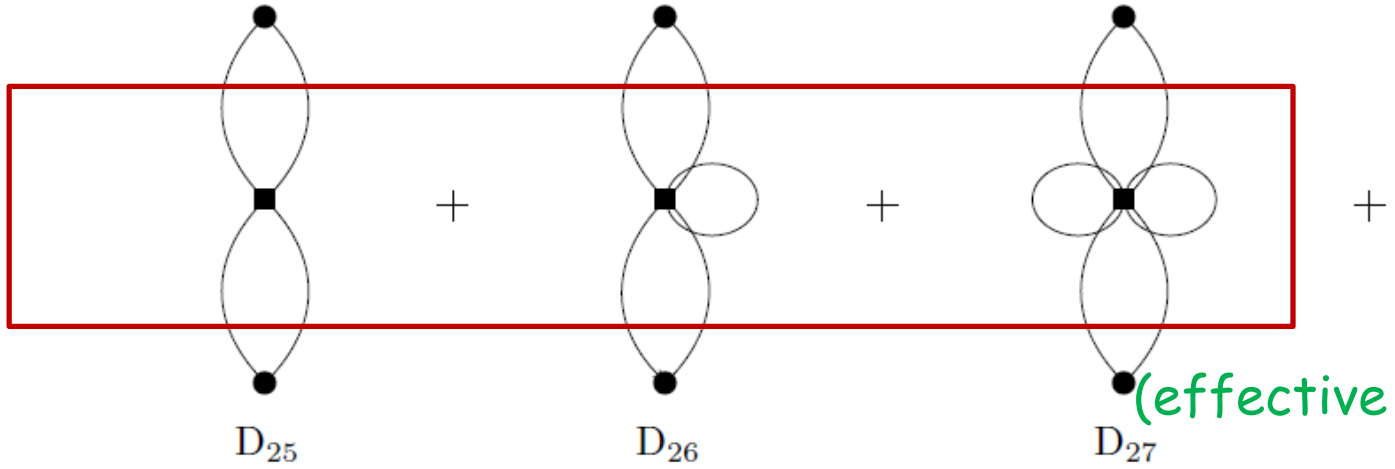
Contributing diagrams to the excitation energy

$\omega_k = \omega_k^0 +$ 

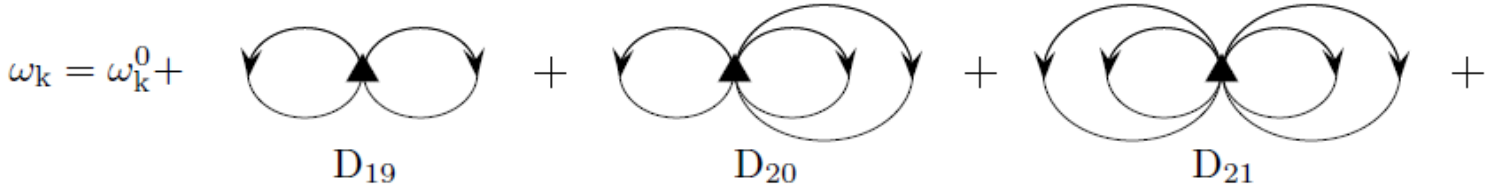
$U_e, U_h =$ 

(effective 1-body)

$K_{eh} =$ 

(effective 2-body)

Contributing diagrams to the excitation energy



- Contribution from different treatment of e-e correlation for ground and excited state wave function

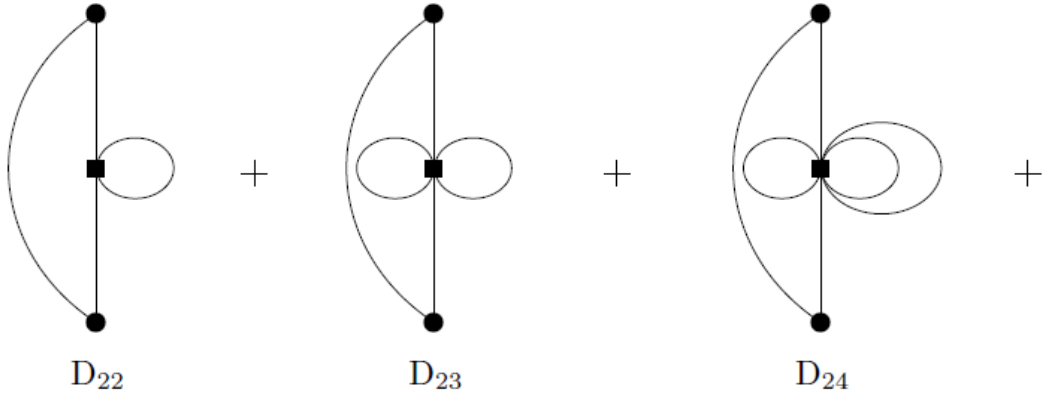
$$|\Psi_0\rangle = G_0 |0\rangle \qquad |\Psi_n\rangle = G_n |\Phi_i^a\rangle$$

$$\langle 0 | W(G_n - G_0) | 0 \rangle = \{D_{19} + D_{20} + D_{21}\}$$

- Impacts excitation energy
- Does not impact electron-hole interaction kernel
- Is zero if $G_n = G_0$

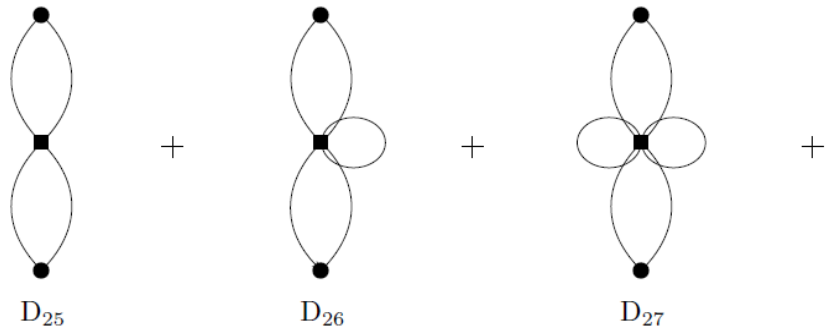
Contributing diagrams to the excitation energy

$U_e, U_h =$



- Effective 1-body (quasi) electron and hole operators
- Renormalizes quasiparticle energy levels due to e-e correlation
- Depends only on excited-state correlation operator G_n
- Impacts excitation energy
- Does not impact electron-hole interaction kernel

Contributing diagrams to the excitation energy

$K_{eh} =$ 

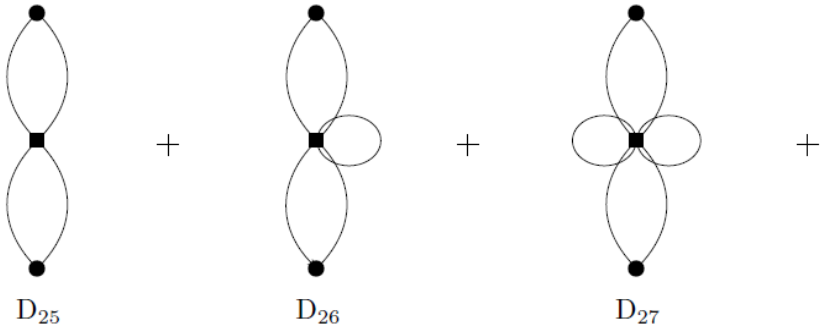
- It is an 2-particle operator that simultaneous operate on both (quasi) electron and hole states
- The loops represent renormalization of 3- and 4-body operators as effective 2-body operators
- All diagrams contribute to the electron-hole interaction kernel
- Depends only on excited-state correlation operator G_n

$$G_n = 0 \rightarrow K_{eh} = 0$$

(eh screening is a consequence of ee correlation)

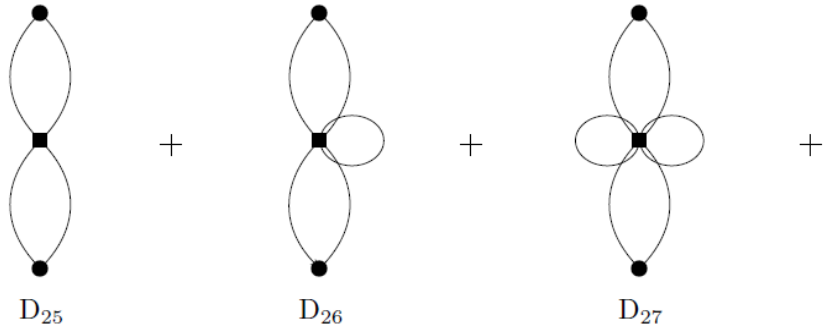
Interpretation of the closed-loops diagrams

$K_{eh} =$



- Closed-loops represent summation over occupied-state
- They represent effective 2-body operators generated from a 4-body operator by treating the additional coordinates at mean-field level

Interpretation of the closed-loops diagrams

$K_{eh} =$  + + +

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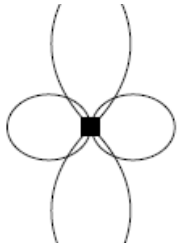
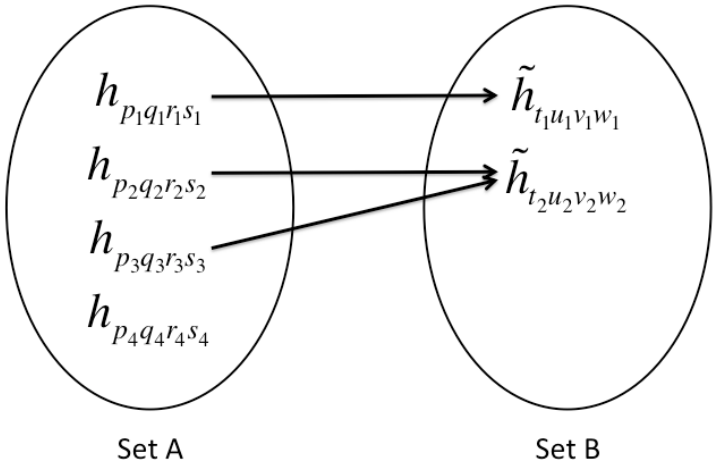


$$= \frac{1}{4!}\sum_{i,j\in \mathrm{occ}} \langle \chi_i(3)\chi_j(4) \,|\, \theta_n(1,2,3,4) \,|\, \chi_i(3)\chi_j(4)\rangle_A$$

Just-in-time (JIT) source code generation

$$I = \sum_{pqrs} h_{pqrs} \langle 0 | X_1 X_2 \ldots \{ p^\dagger q^\dagger sr \} \ldots Y_1 Y_2 | 0 \rangle$$



Many-to-one map

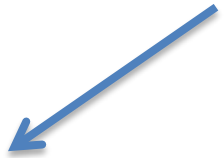
- Numerically zero mo integrals are eliminated
- Non-unique values are mapped to unique terms
- All non-zero & unique mo integerals are assigned a unique id
- The unique id of the mo integrals are used to consolidate terms in the reduction step

Key point: Incorporating molecular integrals in the reduction step

Computer assisted Wick's contraction

- The strings of second quantized operators were evaluated using generalized Wick's theorem

$$\langle 0 | \{X_1 X_2 \ldots\} \{\ldots Y_{N-1} Y_N\} | 0\rangle = \langle 0 | \overbrace{X_1 X_2 \ldots Y_{N-1} Y_N}^{\text{fully contracted}} | 0\rangle$$



- **Strategy#1**: All contractions are performed computationally
- **Strategy#2**: Contractions are performed diagrammatically and the implementation is done computationally

Just-in-time (JIT) source code generation

Disadvantages:

- Source code is generated every time the mo integrals are updated (new system and change of basis)

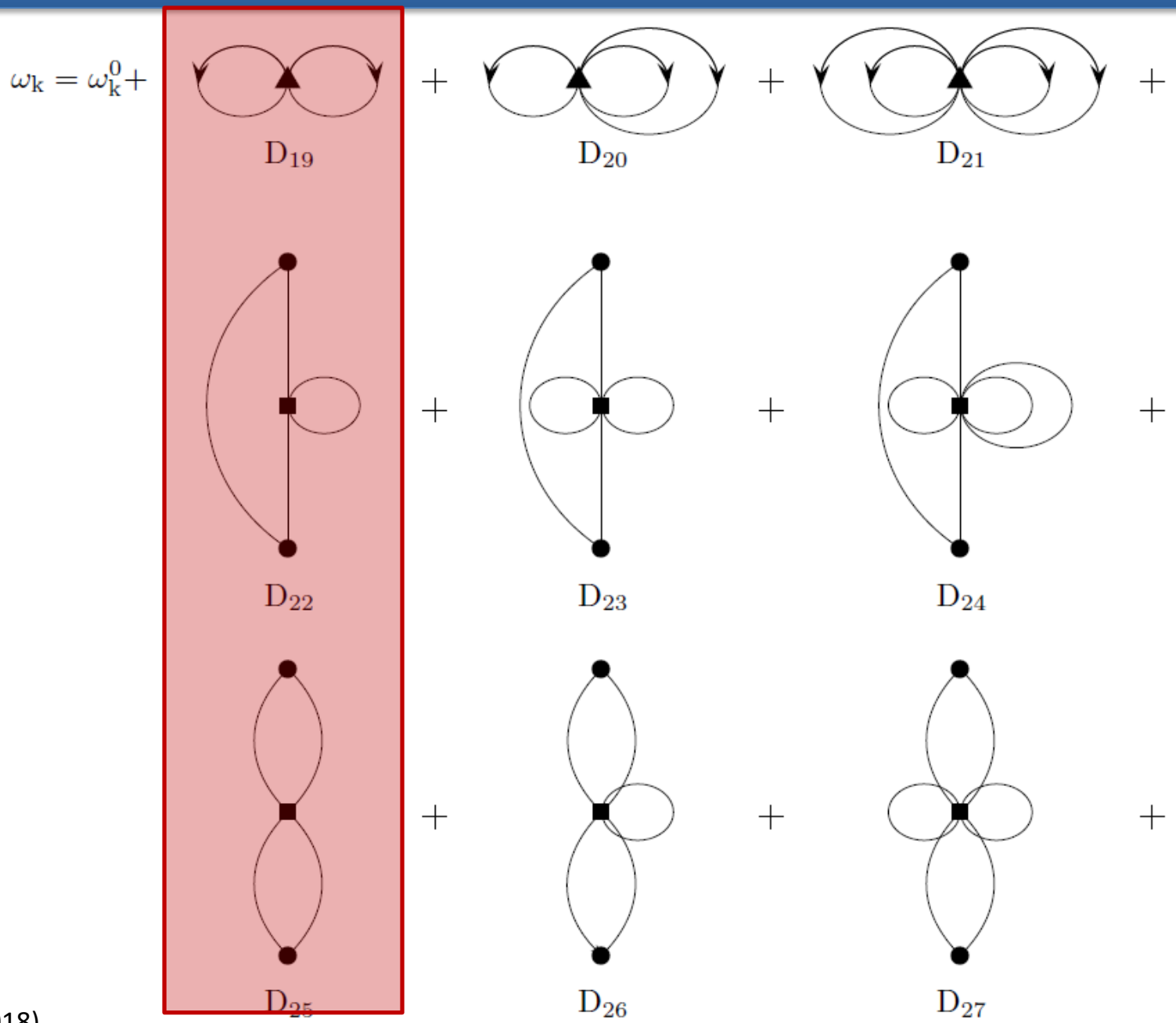
- Can be impractical for large codes that have long compilation time

Advantages:

- The generated source code is optimized for the specific system

- Can reduce the overall memory footprint

Chemical application using first-order diagrams



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Application to chemical systems

$$K_{\mathrm{eh}}^{(I)} = w(1,2)g(1,2)(1-P_{12})$$

$$\text{approximations}: \begin{cases} G_n = G_0 \\ K_{\mathrm{eh}}^{(II,III)} = U_{\mathrm{e,h}}^{(II,III)} = 0 \end{cases}$$

- Excitation energies in small molecules and clusters

$\omega_{0n}[\text{this work}] - \omega_{0n}[\text{EOM-CCSD}]$

System	Energy difference in eV
Ne	0.06
H_2O	0.03

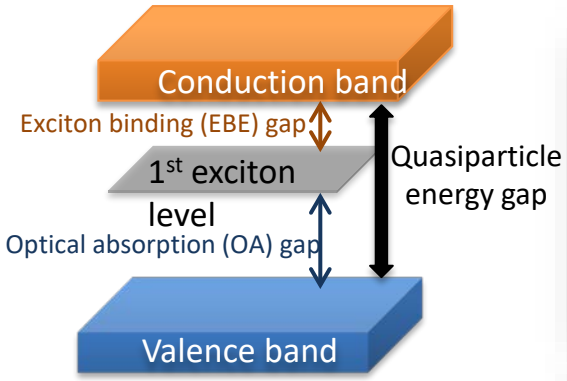
$\omega_{0n}[\text{this work}] - \omega_{0n}[\text{GW/BSE}]$

System	Energy difference in eV	
Cd_6Se_6	0.04	(Ref. 1 & 3)
$Cd_{20}Se_{19}$	-0.04	(Ref. 2 & 3)

Results from this work show reasonable agreement with many-body methods that use unoccupied states

1 Noguchi, Sugino, Nagaoka, Ishii, Ohno, JCP, 137, 024306 (2012)
2 Wang, Zunger, PRB, 53, 9579 (1996)
3 Bayne, Chakraborty, to be submitted (this work)

Exciton binding energies in CdSe clusters



Determination of the Exciton Binding Energy in CdSe Quantum Dots

Robert W. Meulenberg,[†,§,*] Jonathan R.I. Lee,[†,*] Abraham Wolcott,[‡] Jin Z. Zhang,[‡] Louis J. Terminello,[†] and Tony van Buuren[†]

VOL. 3 ▪ NO. 2 ▪ 325–330 ▪ 2009 ACSNANO

Size-Dependent Valence and Conduction Band-Edge Energies of Semiconductor Nanocrystals

Jacek Jasieniak,[†,*] Marco Califano,[‡] and Scott E. Watkins[†]

VOL. 5 ▪ NO. 7 ▪ 5888–5902 ▪ 2011 ACSNANO

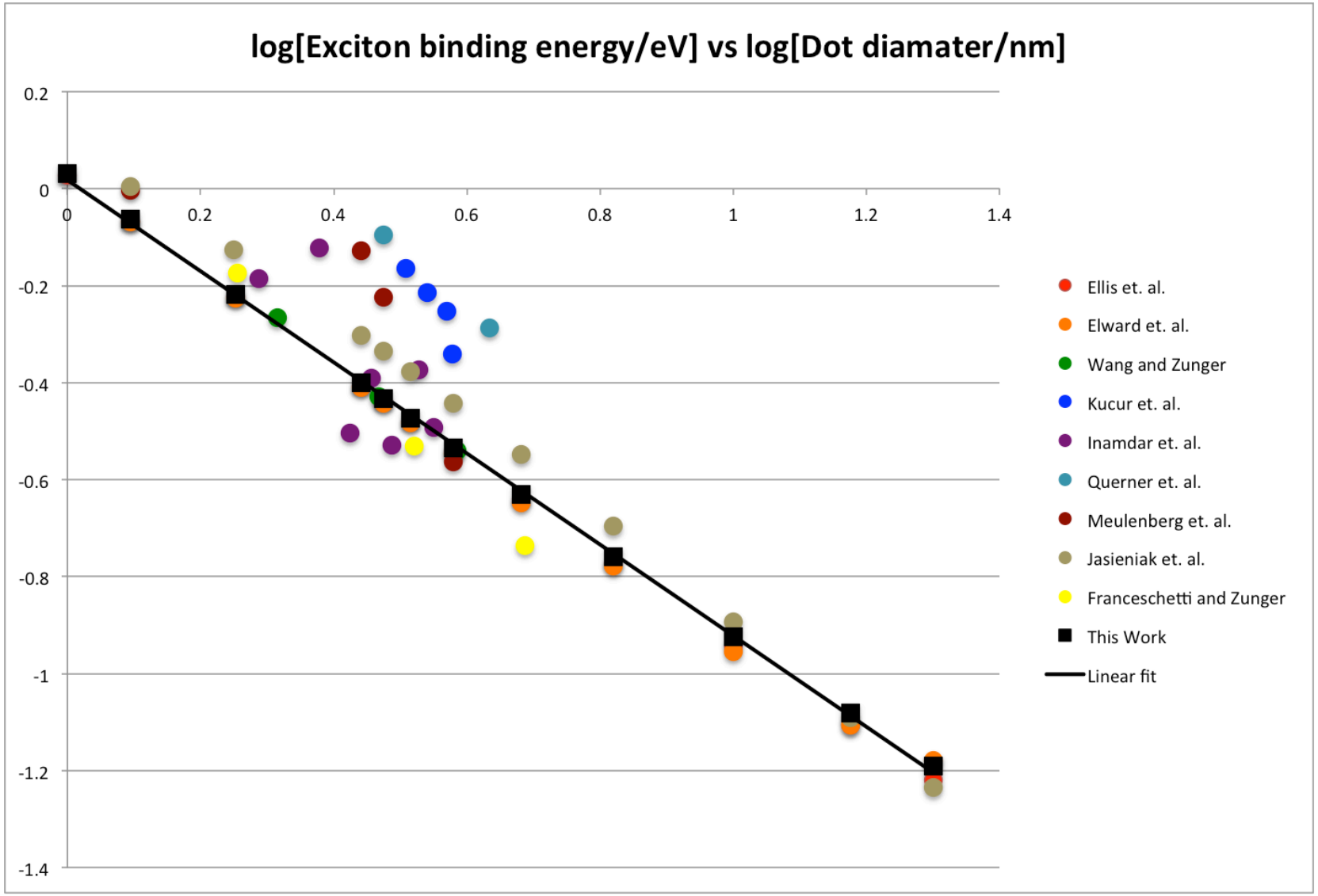
VOLUME 78, NUMBER 5 PHYSICAL REVIEW LETTERS 3 FEBRUARY 1997

Direct Pseudopotential Calculation of Exciton Coulomb and Exchange Energies in Semiconductor Quantum Dots

Alberto Franceschetti and Alex Zunger
National Renewable Energy Laboratory, Golden, Colorado 80401
(Received 23 August 1996)

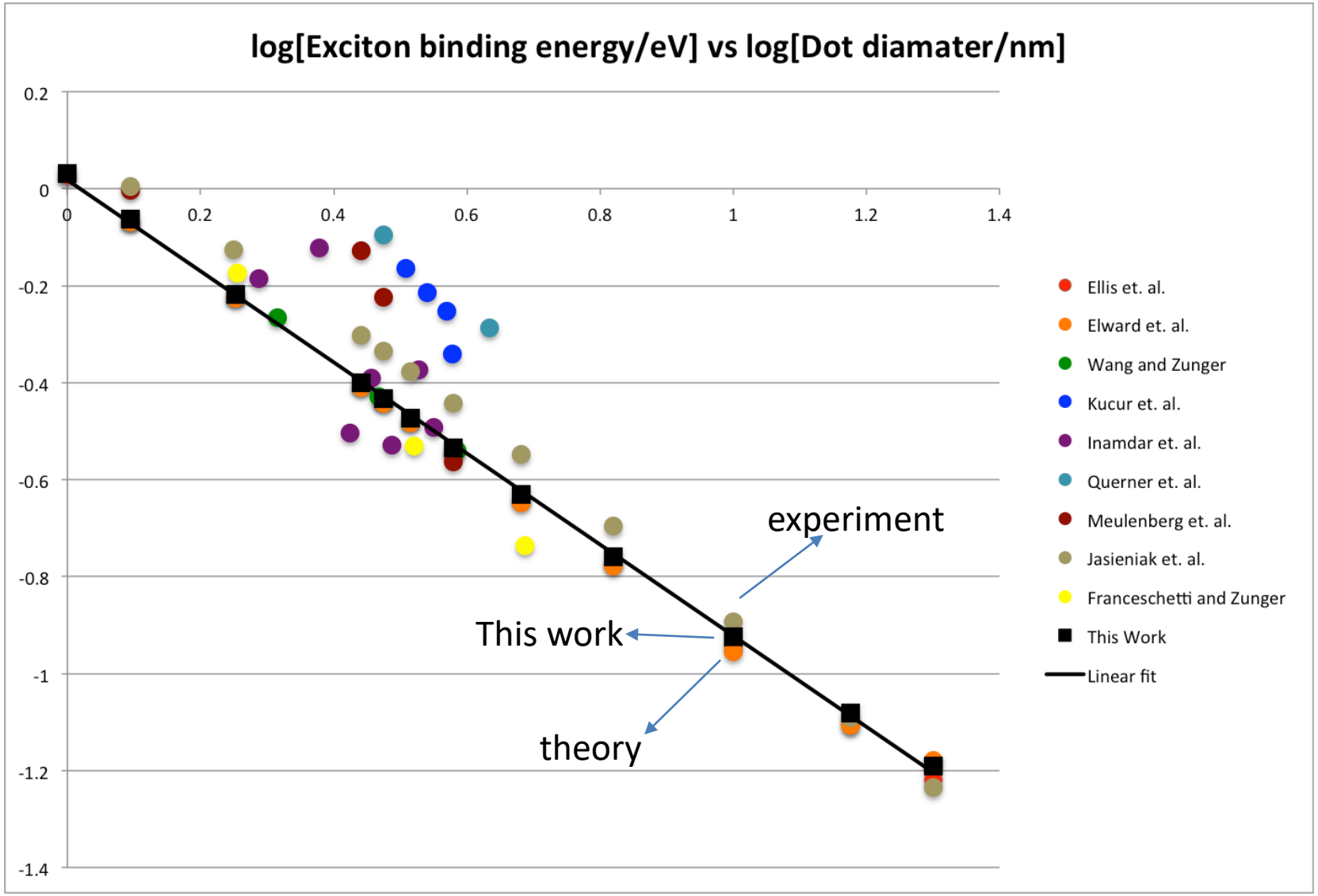
Exciton binding energies in CdSe quantum dots

$$E_{\text{binding}} = \omega_{0n}^{0} - \omega_{0n}$$



Exciton binding energies in CdSe quantum dots

$$E_{\text{binding}} = \omega_{0n}^{0} - \omega_{0n}$$



Summary

- It was shown that the electron-hole interaction kernel (eh-kernel) can be expressed without using unoccupied states.
- The derivation was performed using a two-body correlation operator which is local in real-space representation.
- Using diagrammatic techniques, it was shown that the eh-kernel can be expressed only in terms of linked-diagrams.
- The derived expression provides a route to make additional approximations to the eh-kernel
- The 1st order approximation of eh-kernel was used for calculating electron-hole binding energies and excitation energies in atoms, molecules, clusters, and quantum dots.

Title here

Deformation potential: Which basis?

$$\mathbf{h}^{\eta} = \mathbf{h}^{0} + \mathbf{v}^{\eta}_{\mathrm{def}}$$

Space-filling basis functions

- Examples: plane-waves, real-space grid, distributed Guassian functions, Harmonic osc. basis, particle-in-box basis
- Both deformed and reference Hamiltonian use identical basis functions

Atom-centered basis functions

- Deformed and reference Hamiltonian use different basis functions
- We transform into the eigenbasis of the reference Hamiltonian

Transformation to ref. eigenbasis-I

- Step #1: Get quantities from the converged SCF calculation on reference structure

$$\mathbf{F}^0\mathbf{C}^0 = \boldsymbol{\lambda}^0\mathbf{S}^0\mathbf{C}^0$$

- Step #2: Perform symmetric or orthogonal transformation such that the S matrix is diagonal in that basis

$$\mathbf{X}^{0\dagger}\mathbf{S}^0\mathbf{X} = \mathbf{I}$$

$$\mathbf{X}^{0\dagger}\mathbf{F}^0\mathbf{X} = \tilde{\mathbf{F}}^0$$

(single tilde transformation)

- Step #3: Find the U matrix that diagonalizes the transformed Fock matrix

$$\tilde{\tilde{\mathbf{F}}}^0 \equiv [\mathbf{U}^0]^\dagger\tilde{\mathbf{F}}^0\mathbf{U}^0 = \boldsymbol{\lambda}^0$$

(double tilde transformation)

- The U^0 matrix is the matrix needed to transform operators in the eigenbasis of the reference structure

Transformation to ref. eigenbasis-II

- Step #4: Get quantities from the converged SCF calculation on the deformed structure

$$\mathbf{F}^{\eta}\mathbf{C}^{\eta} = \boldsymbol{\lambda}^{\eta}\mathbf{S}^{\eta}\mathbf{C}^{\eta}$$

- Step #5: Perform orthogonal transformation

$$\mathbf{X}^{0\dagger}\mathbf{S}^{\eta}\mathbf{X} = \mathbf{I}$$

$$\mathbf{X}^{0\dagger}\mathbf{F}^{\eta}\mathbf{X} = \tilde{\mathbf{F}}^{\eta}$$

- Step #6: Transform the Fock in the eigenbasis of the reference Hamiltonian

$$\tilde{\tilde{\mathbf{F}}}^{\eta} = [\mathbf{U}^{0}]^{\dagger}\tilde{\mathbf{F}}^{\eta}\mathbf{U}^{0}$$

- Step #7: Calculate the deformation potential

$$\mathbf{V}_{\mathrm{def}}^{\eta} = \tilde{\tilde{\mathbf{F}}}^{\eta} - \tilde{\tilde{\mathbf{F}}}^{0}$$

Info#: Contributing diagrams to the excitation energy

$$(G_n - G_0) = \sum_{i<j}^{N} g_n(i,j) - g_0(i,j) = \sum_{i<j}^{N} \tilde{g}(i,j)$$

$$W(G_n - G_0) = \left[\sum_{i<j}^{N} w(i,j)\right]\left[\sum_{i<j}^{N} \tilde{g}(i,j)\right] = \sum_{i<j}^{N} \theta_n(i,j) + \sum_{i<j}^{N} \theta_n(i,j,k) + \sum_{i<j<k<l}^{N} \theta_n(i,j,k,l)$$

$$\begin{aligned}\langle 0 | W(G_n - G_0) | 0 \rangle = & \frac{1}{2}\sum_{i_1 i_2}^{N} \langle i_1 i_2 | \theta_2 (1 - P_{12}) | i_1 i_2 \rangle \\ & + \frac{1}{3!}\sum_{i_1 i_2 i_3}^{N} \langle i_1 i_2 i_3 | \theta_3 \sum_{k=1}^{3!} (-1)^{p_k} \hat{P}_k | i_1 i_2 i_3 \rangle \\ & + \frac{1}{4!}\sum_{i_1 i_2 i_3 i_4}^{N} \langle i_1 i_2 i_3 i_4 | \theta_4 \sum_{k=1}^{4!} (-1)^{p_k} \hat{P}_k | i_1 i_2 i_3 i_4 \rangle\end{aligned}$$

Info#: Determination of geminal parameters

- For quantum dots, G was obtained from parabolic QD

$$H_{\text{model}} = T_e + T_h + V_{eh} + V_{\text{harm}}$$

$$\min_G \frac{\langle \chi_e \chi_h | G^\dagger H_{\text{model}} G | \chi_e \chi_h \rangle}{\langle \chi_e \chi_h | G^\dagger G | \chi_e \chi_h \rangle} \Rightarrow G_{\text{model}}$$

$$G_n \simeq G_{\text{model}} \qquad G_0 = G_n$$

- For small molecules and clusters, G was obtained varaitionally

$$\min_G \frac{\langle 0 | G^\dagger H G | 0 \rangle}{\langle 0 | G^\dagger G | 0 \rangle} \Rightarrow G_0$$

$$G_n = G_0$$

If we are ready to admit unoccupied states

Infinite-order diagrammatic summation approach to explicitly correlated congruent transformed Hamiltonian

Phys. Rev. A **89**, 032515 (2014)

Mike Bayne,[1] John Drogo,[2] and Arindam Chakraborty[1,*]

Challenge: Avoiding 3, 4, 5, 6-particle integrals

Step 1: Project the correlation function in a finite basis

$$E = \langle \Phi_0 \,|\, G^\dagger H G \,|\, \Phi_0 \rangle = \sum_{kk'}^{M} G_{0k} H_{kk'} G_{k'0}$$

Step 2: Write the energy expression term of diagrams

$$E = \sum_{kk}^{M} \; D_1 + D_2 + \ldots + D_{36}$$

Step 3: Obtain a renormalized 2-body operator by performing infinite-order summation over a subset of diagrams

$$E = \underbrace{\lim_{M\to\infty} \sum_{kk}^{M} \; D_1 + \ldots + D_{10}}_{g(r_{12}) r_{12}^{-1} g(r_{12})} + \sum_{kk}^{M} \; D_{11} + \ldots + D_{36}$$

Partial infinite-order diagrammatic summation

$$E_{\mathrm{XCHF}} = \frac{\lim_{M\to\infty}\sum_{k}^{M} D_k}{\lim_{M\to\infty}\sum_{k}^{M} D_k}$$

All diagrams are added to infinite order

$$E_{\mathrm{PCTH\text{-}PIOS}} = \frac{\left[\lim_{M\to\infty}\sum_{k}^{M} D_k\right] + \sum_{k}^{M} D_k}{\left[\lim_{M\to\infty}\sum_{k}^{M} D_k\right] + \sum_{k}^{M} D_k}$$

Some diagrams are added to infinite order

$$E_{\mathrm{PCTH}} = \frac{\sum_{k}^{M} D_k}{\sum_{k}^{M} D_k}$$

All diagrams are added to finite order

Info#: Summation over intermediate particle-hole states

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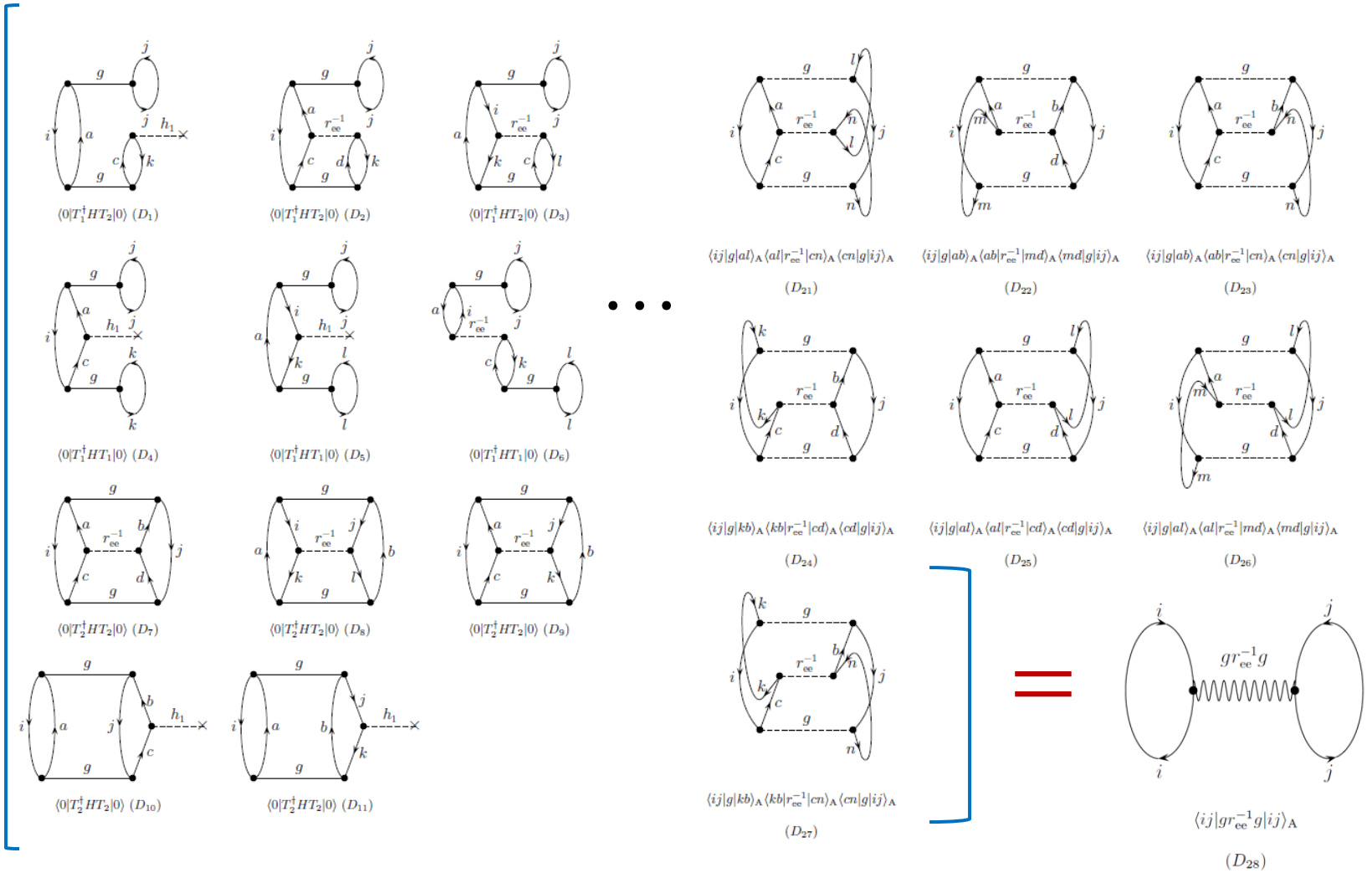
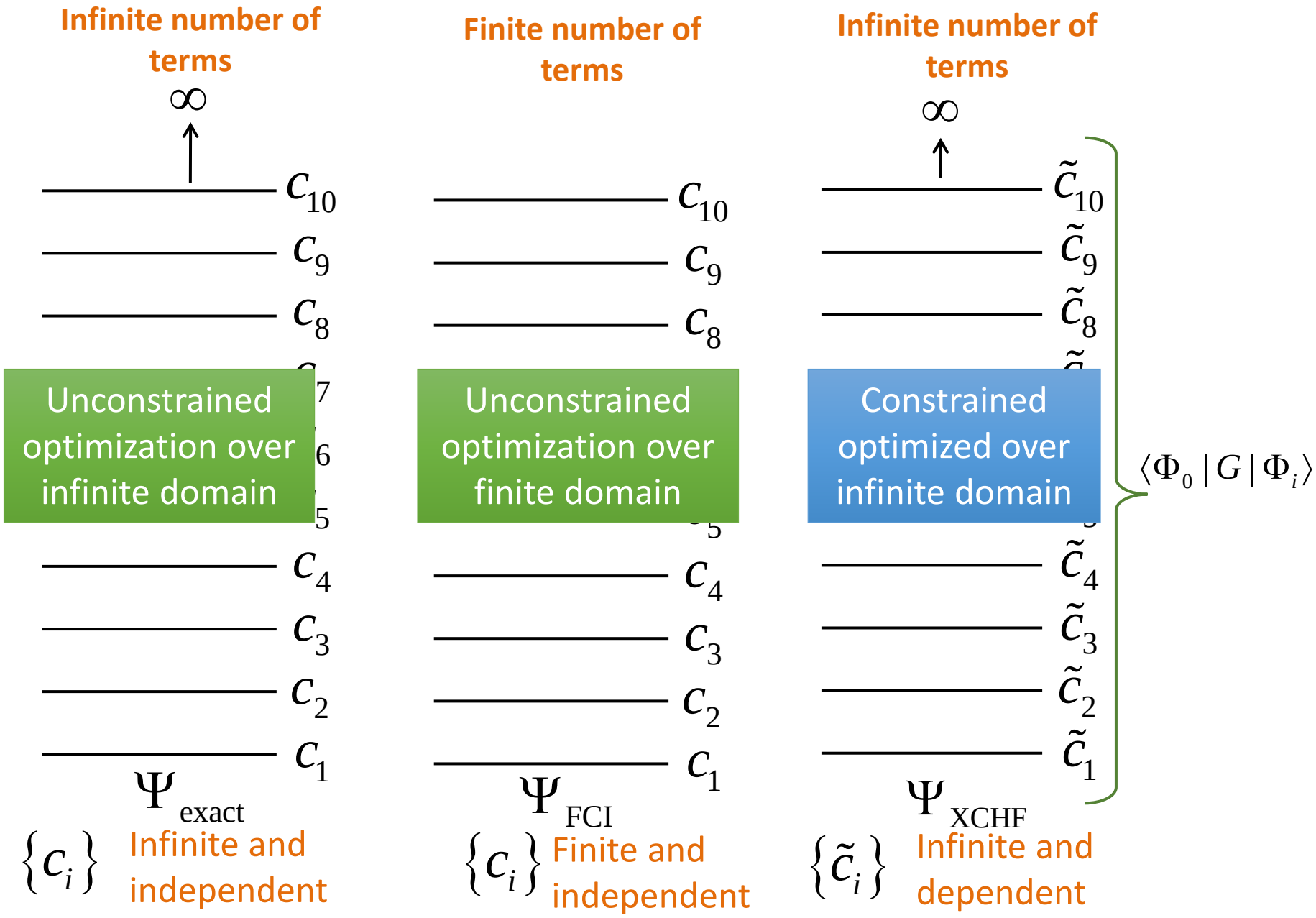
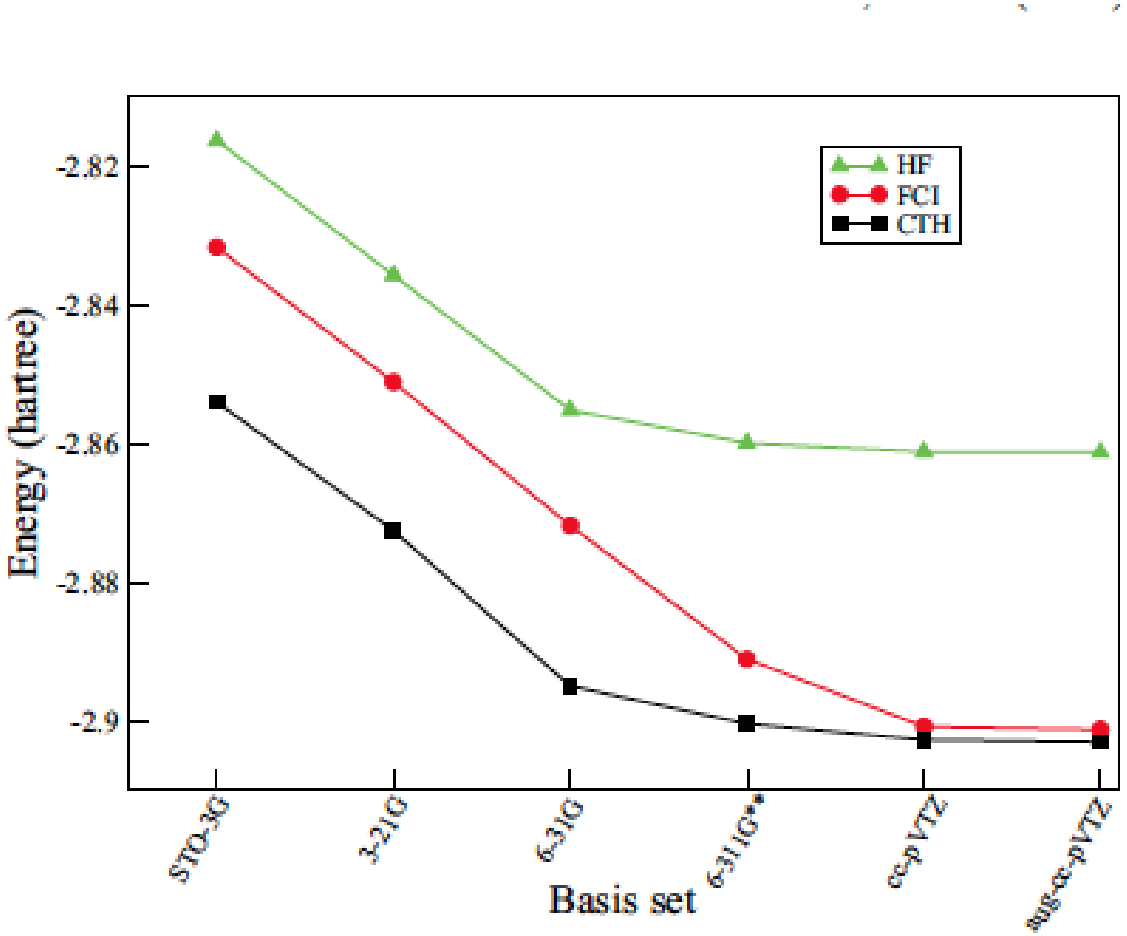
$$\sum_{a,b,c,d}^{\infty}$$



Comparison of the three methods



Ground state energy of helium atom



3 Key Points

- FCI - multi-determinant minimization.

XCHF - single determinant minimization.

- XCHF gives lower energy than FCI for all the basis sets shown.
- XCHF converges much faster with respect to size 1-particle basis

1. Elward, Hoja, Chakraborty Phys. Rev. A. 86, 062504 (2012)

See also: Geminal augmented MCSCF for H2, QMC calculations on H2O
Varganov and Martinez, JCP, **132**, 054103 (2010) ; Xu and Jordan, JPCA, **114**, 1365 (2010)

Electron-hole Hamiltonian

$$H = \underbrace{T_e + V_{ee} + V_e^{ext}}_{\text{electronic subsystem}} + \underbrace{T_h + V_{hh} + V_h^{ext}}_{\text{hole subsystem}} + \underbrace{V_{eh}}_{\text{interaction term}}$$

$$H = H_0 + V_{eh}$$

Coulomb attraction term is responsible for electron-hole coupling

This talk:
Explicitly correlated Hartree-Fock

- **Configuration interaction** (CI): Zunger, Efros, Sundholm, Wang, Bester, Rabani, Franceschetti, Califano, Bittner, Hawrylak,…
- **Many-body perturbation theory** (MBPT): Baer, Neuhauser, Galli,...
- **Quantum Monte Carlo method** (QMC): Hybertsen, Shumway,…
- **GW+BSE**: Louie,Chelikowsky, Galli, Rohlfing , Rubio, …
- **All-electron TDDFT/DFT**: Prezhdo, Tretiak, Kilina, Akimov, Ullrich, Li,….