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*Multiresolution  
Adaptive  
Numerical  
Scientific  
Simulation*

# *Multiresolution quantum chemistry*

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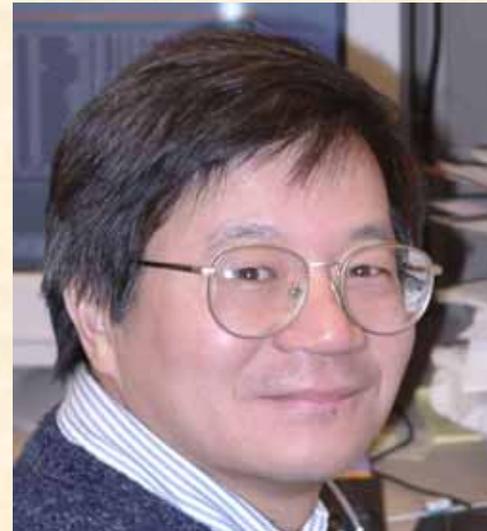
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# *The mathematicians ...*



**Gregory Beylkin**

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# *The funding*

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## SciDAC

Scientific Discovery through Advanced Computing

# *Outline*

- Multiresolution quantum chemistry
  - Objectives
  - Techniques
  - Capabilities
  - Futures
- Higher-level composition of scientific applications
  - Composition with functions and operators
  - Coding for the highly-threaded future with futures

# *Multiresolution chemistry objectives*

- Complete elimination of the basis error
  - One-electron models (e.g., HF, DFT)
  - Pair models (e.g., MP2, CCSD, ...)
- Correct scaling of cost with system size
- General approach
  - Readily accessible by students and researchers
  - Higher level of composition
  - No two-electron integrals – replaced by fast application of integral operators
- New computational approaches
- *Fast algorithms with guaranteed precision* <sup>6</sup>

# References

- The methods in this work are primarily based upon
  - Alpert, Beylkin, Grimes, Vozovoi (J. Comp. Phys., 2002)
  - B. Alpert (SIAM Journal on Mathematical Analysis 24, 246-262, 1993).
  - Beylkin, Coifman, Rokhlin (Communications on Pure and Applied Mathematics, 44, 141-183, 1991.)
  - Beylkin and Mohlenkamp, (Proc. Nat. Acad. 2002)
- The following are useful further reading
  - Daubechies, “*Ten lectures on wavelets*”
  - Walnut, “*An introduction to wavelets*”
  - Meyer, “*Wavelets, algorithms and applications*”
  - Burrus et al, “*Wavelets and Wavelet transforms*”

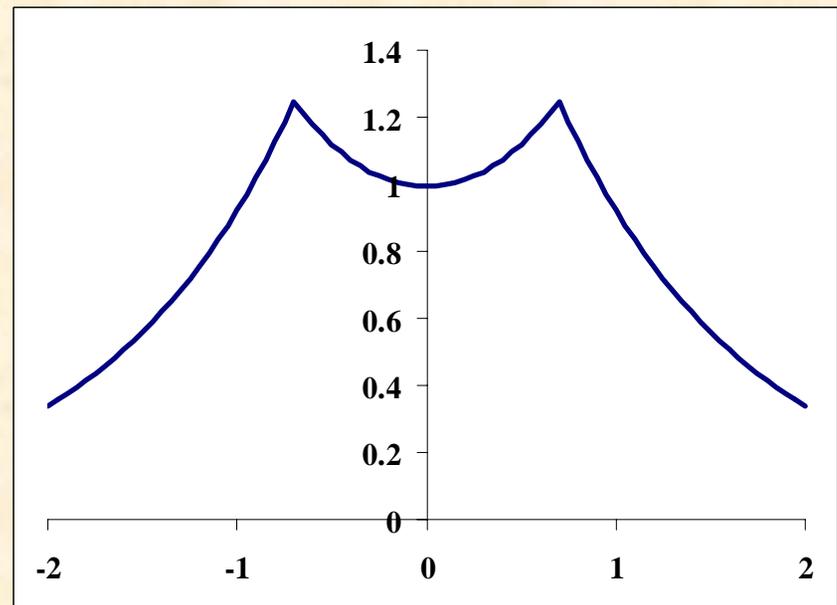
# Linear Combination of Atomic Orbitals (LCAO)

- Molecules are composed of (weakly) perturbed atoms
  - Use finite set of atomic wavefunctions as the basis
  - Hydrogen-like wavefunctions are exponentials
- E.g., hydrogen molecule ( $\text{H}_2$ )

$$1s(r) = e^{-|r|}$$

$$\phi(r) = e^{-|r-a|} + e^{-|r-b|}$$

- Smooth function of molecular geometry
- MOs: cusp at nucleus with exponential decay



# LCAO

- A fantastic success, but ...
- Basis functions have extended support
  - causes scaling problems – more about this later
- Basis set superposition error (BSSE)
  - incomplete basis on each center leads to over-binding as atoms are brought together
- Linear dependence problems
  - accurate calculations require balanced approach to a complete basis on every atom
  - molecular basis can have severe linear dependence

# *Essential techniques for fast computation*

- Multiresolution  $V_0 \subset V_1 \subset \dots \subset V_n$   
 $V_n = V_0 + (V_1 - V_0) + \dots + (V_n - V_{n-1})$
- Low-separation rank  
 $f(x_1, \dots, x_d) = \sum_{l=1}^M \sigma_l \prod_{i=1}^d f_i^{(l)}(x_i) + O(\varepsilon)$   
 $\|f_i^{(l)}\|_2 = 1 \quad \sigma_l > 0$
- Low-operator rank  
 $A = \sum_{\mu=1}^r \underline{u}_\mu \sigma_\mu \underline{v}_\mu^T + O(\varepsilon)$   
 $\sigma_\mu > 0 \quad \underline{v}_\mu^T \cdot \underline{v}_\lambda = \underline{u}_\mu^T \cdot \underline{u}_\lambda = \delta_{\mu\lambda}$

# *How to “think” multiresolution*

- Consider a ladder of function spaces

$$V_0 \subset V_1 \subset V_2 \subset \cdots \subset V_n$$

- E.g., increasing quality atomic basis sets, or finer resolution grids, ...

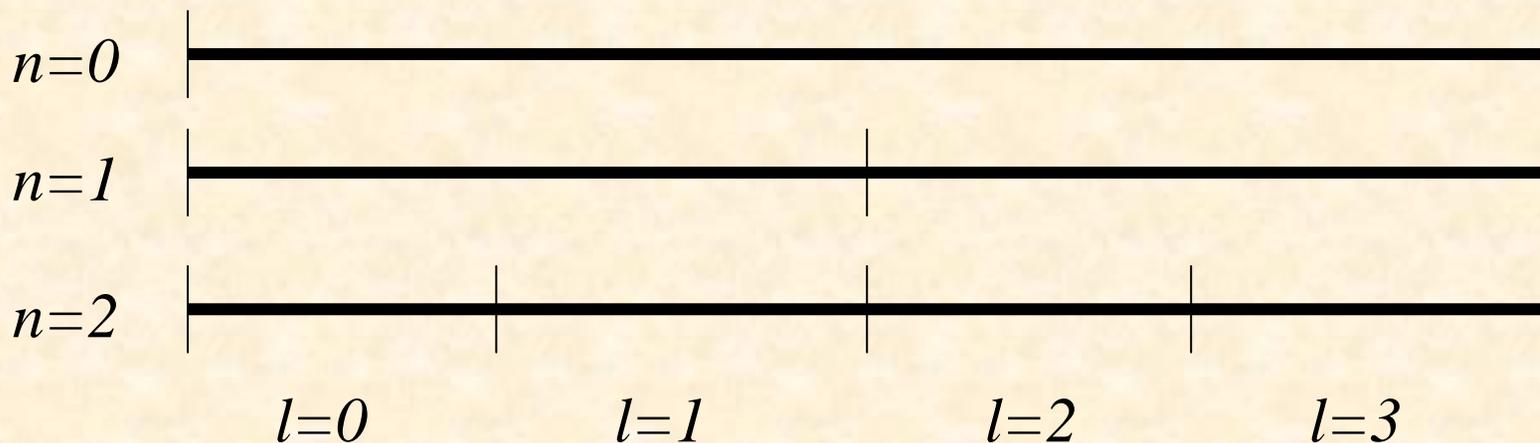
- Telescoping series

$$V_n = V_0 + (V_1 - V_0) + (V_2 - V_1) + \cdots + (V_n - V_{n-1})$$

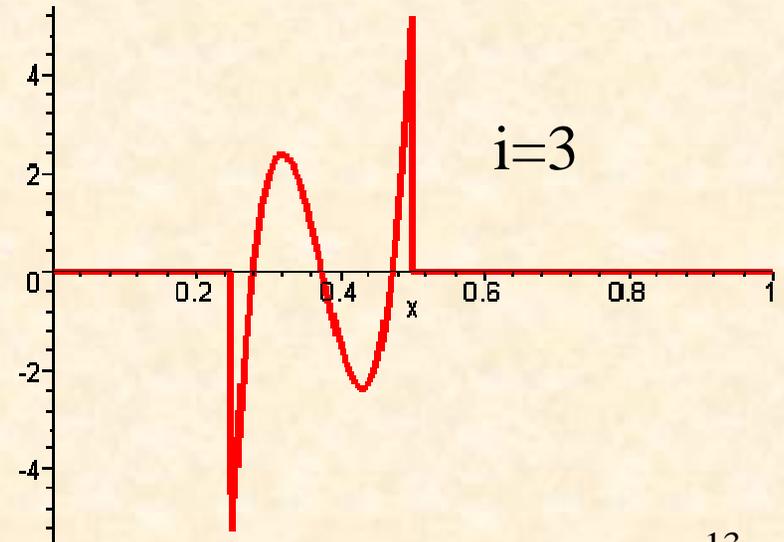
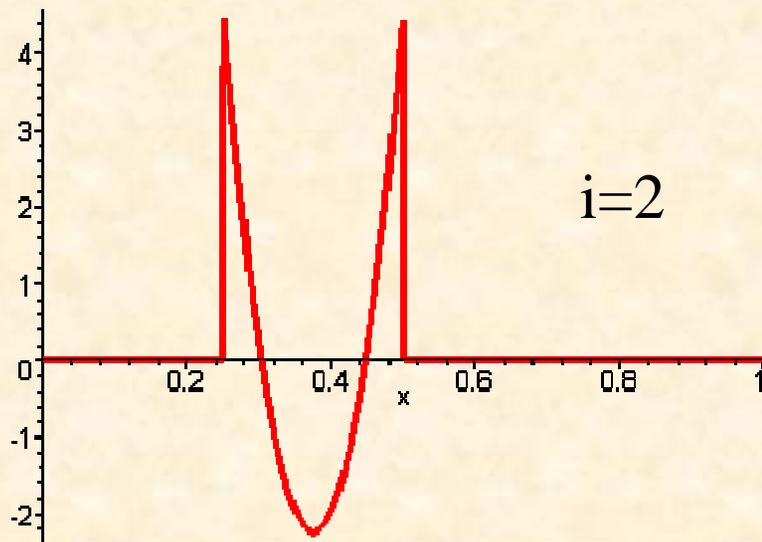
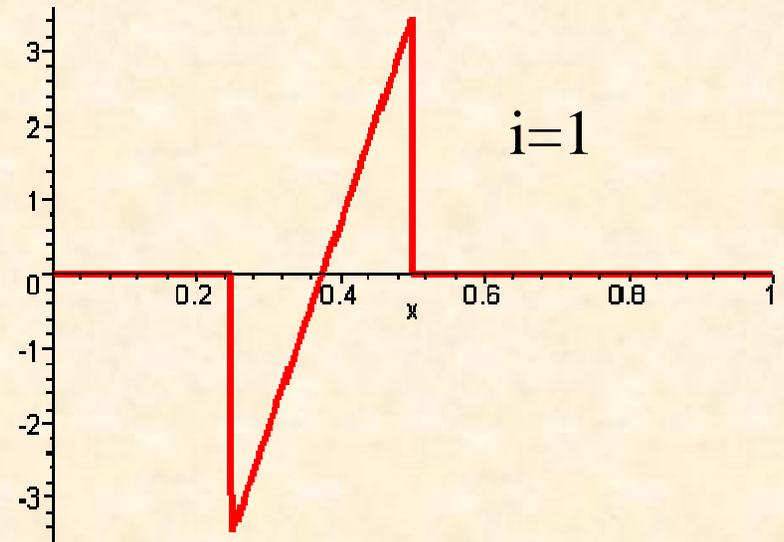
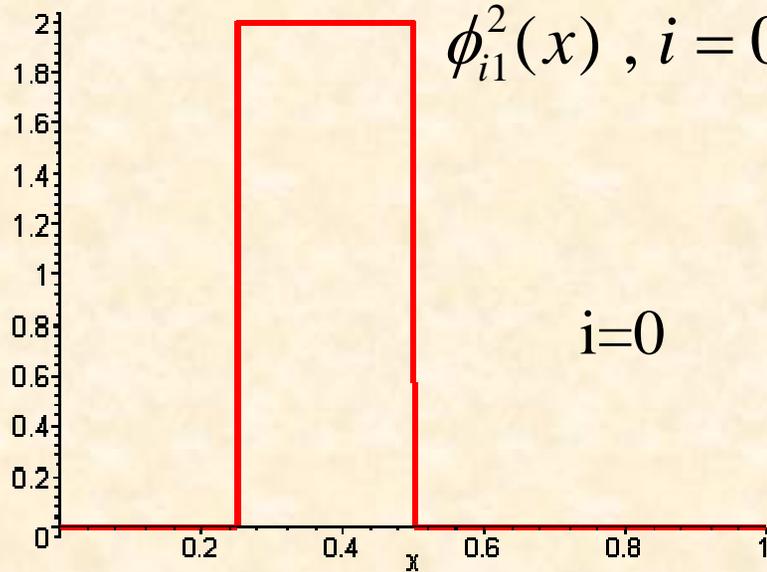
- Instead of using the most accurate representation, use the difference between successive approximations
- Representation on  $V_0$  small/dense; differences sparse
- Computationally efficient; possible insights

# Scaling Function Basis

- Divide domain into  $2^n$  pieces (level  $n$ )
  - Adaptive sub-division (local refinement)
  - $l^{\text{th}}$  sub-interval  $[l*2^{-n}, (l+1)*2^{-n}]$   $l=0, \dots, n-1$
- In each sub-interval define a polynomial basis
  - First  $k$  Legendre polynomials  $\phi_i(x) = \sqrt{2i+1}P_i(2x-1)$
  - Orthonormal, disjoint support  $\phi_{il}^n(x) = 2^{n/2} \phi_i(2^n x - l)$



# Scaling Function Basis - III



# *Multiwavelet Basis*

- An orthonormal basis to span  $W_{n-1} = V_n - V_{n-1}$

Haar (piecewise constant basis)  $\phi_l^{n-1} = 2^{-1/2} \left( \phi_{2l}^n + \phi_{2l+1}^n \right)$  sum

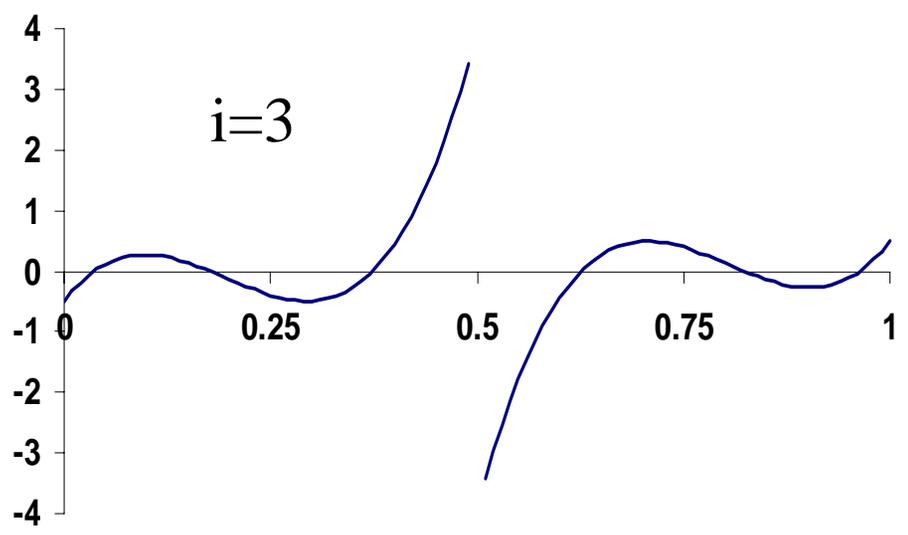
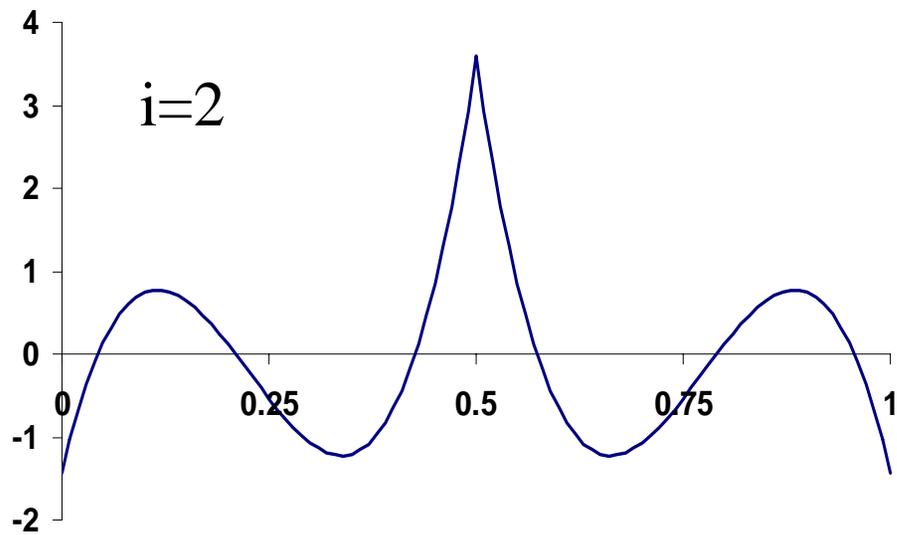
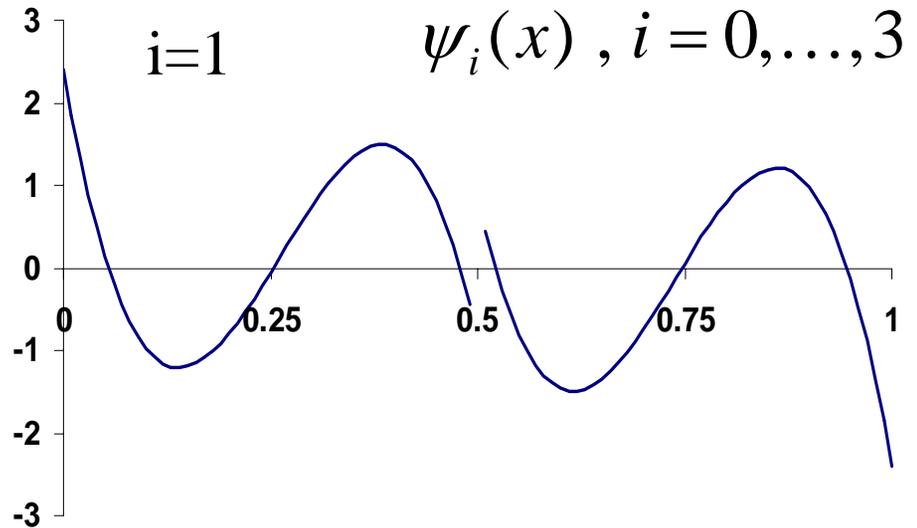
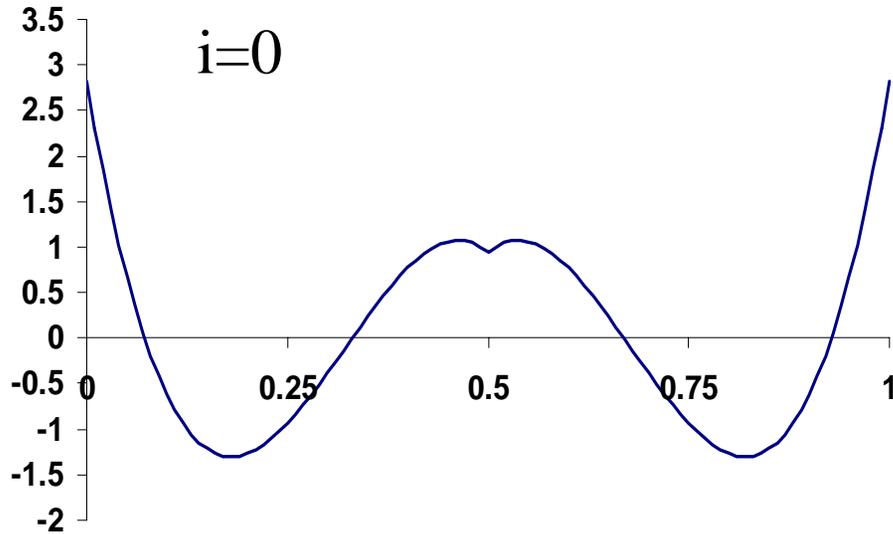
$\psi_l^{n-1} = 2^{-1/2} \left( \phi_{2l}^n - \phi_{2l+1}^n \right)$  difference

$$\phi_{il}^{n-1}(x) = \sum_{j=0}^{k-1} \left( h_{ij}^{(0)} \phi_{j2l}^n(x) + h_{ij}^{(1)} \phi_{j2l+1}^n(x) \right)$$

$$\psi_{il}^{n-1}(x) = \sum_{j=0}^{k-1} \left( g_{ij}^{(0)} \phi_{j2l}^n(x) + g_{ij}^{(1)} \phi_{j2l+1}^n(x) \right)$$

# Multiwavelet Basis - III

$k=4$



# *Vanishing moments*

- Critically important property
  - Since  $W_n = V_{n+1} - V_n$  is orthogonal to  $V_n$  the first  $k$  moments of functions in  $W_n$  vanish, i.e.,

$$\int x^j \psi_i(x) dx = 0, \quad j = 0, \dots, k - 1$$

- Compact representation of smooth functions
  - Consider Taylor series ... the first  $k$  terms vanish and smooth implies higher order terms are small
- Compact representation of integral operators
  - E.g.,  $|r-s|^{-1}$  ... interaction decays as  $r^{-2k-1}$
- Derivatives vanish at origin in Fourier space
  - Diminishes effect of singularities at that point

# Three equivalent representations

- Scaling function basis (reconstructed)

$$f^n(x) = \sum_{l=0}^{2^n-1} \sum_{i=0}^{k-1} s_{il}^n \phi_{il}^n(x)$$

- Multi-wavelet basis (compressed)

$$f^n(x) = \sum_{i=0}^{k-1} s_{i0}^0 \phi_{i0}^0(x) + \sum_{n'=0}^{n-1} \sum_{l=0}^{2^{n'}-1} \sum_{i=0}^{k-1} d_{il}^{n'} \psi_{il}^{n'}(x)$$

- The function tabulated at the Gauss-Legendre quadrature points in each of the adaptively refined boxes

# *Please forget about wavelets*

- They are not central
- Wavelets are a convenient basis for spanning  $V_n - V_{n-1}$  and understanding its properties
- But you don't actually need to use them
  - MADNESS does still compute wavelet coefficients, but *Beylkin's new code does not*
- Please remember this ...
  - Discontinuous spectral element with multi-resolution and separated representations for fast computation with guaranteed precision.

# Truncation Error

- To satisfy the global error condition

$$\|f - f^n\|_2 \leq \varepsilon \|f\|_2$$

- Truncate according to

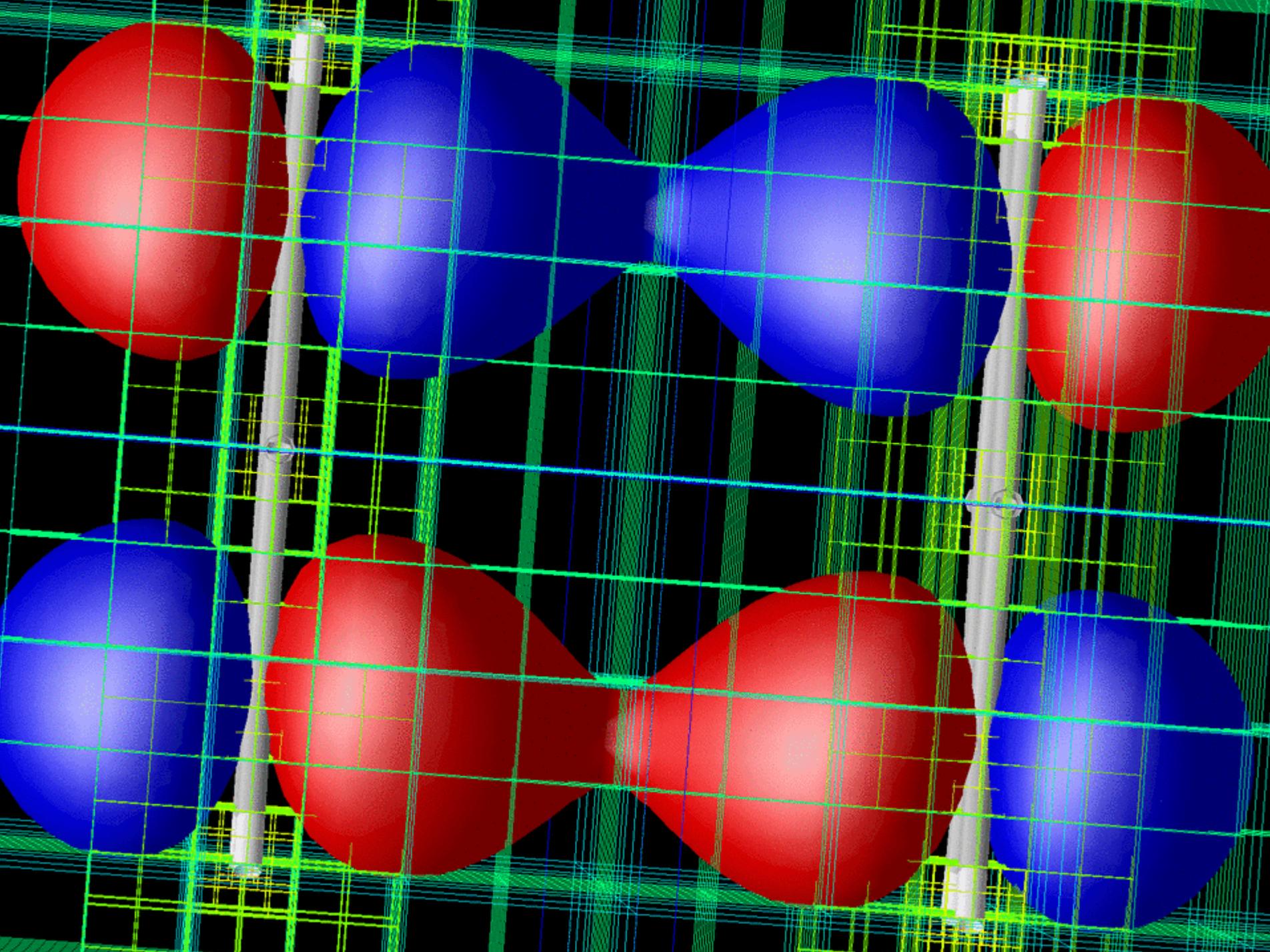
$$\|d_l^n\|_2 \leq 2^{-n/2} \varepsilon \|f\|_2$$

- Above is rather conservative – often use

$$\|d_l^n\|_2 \leq \varepsilon$$

- For accurate function & derivative

$$\|d_l^n\|_2 \leq 2^{-n} \varepsilon$$



# *Integral Formulation*

- Solving the integral equation
  - Eliminates the derivative operator and related “issues”
  - Converges as fixed point iteration *with no preconditioner*

$$\left(-\frac{1}{2}\nabla^2 + V\right)\Psi = E\Psi$$

$$\begin{aligned}\Psi &= -2\left(-\nabla^2 - 2E\right)^{-1} V\Psi \\ &= -2G^*(V\Psi)\end{aligned}$$

$$(G^* f)(r) = \int ds \frac{e^{-k|r-s|}}{4\pi|r-s|} f(s) \quad \text{in 3D ; } k^2 = -2E$$

Such Green's Functions (bound state Helmholtz, Poisson) can be rapidly and accurately applied with a single, sparse matrix vector product.<sup>21</sup>

# Separated form for integral operators

$$T * f = \int ds K(r - s) f(s)$$

- Approach in current prototype code
  - Represent the kernel over a finite range as a sum of Gaussians

$$r_{ii',jj',kk'}^{nl} = \sum_i \omega_i X_{ii'}^{nl_x} Y_{ii'}^{nl_y} Z_{ii'}^{nl_z} + O(\varepsilon) \quad \text{in 3D}$$

$$K(r) = \sum_i \omega_i e^{-t_i r^2} + O(\varepsilon)$$

- Only need compute 1D transition matrices (X,Y,Z)
- SVD the 1-D operators (low rank away from singularity)
- Apply most efficient choice of low/full rank 1-D operator
- Even better algorithms not yet implemented

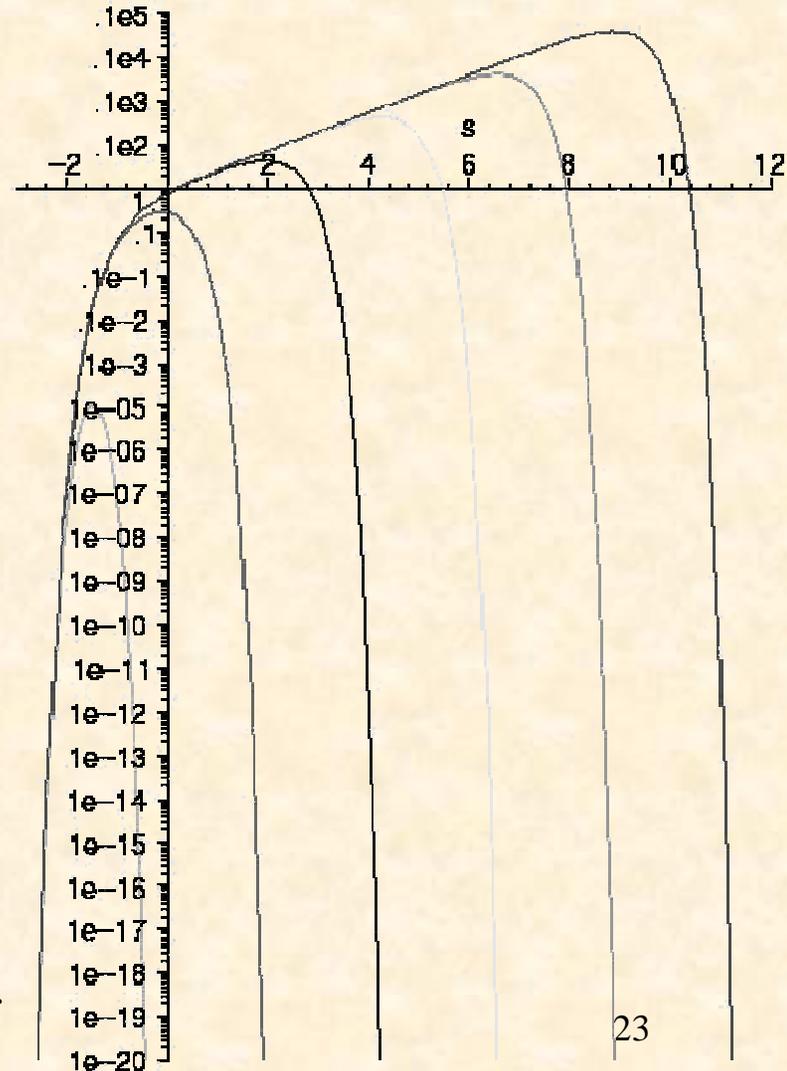
# Accurate Quadratures

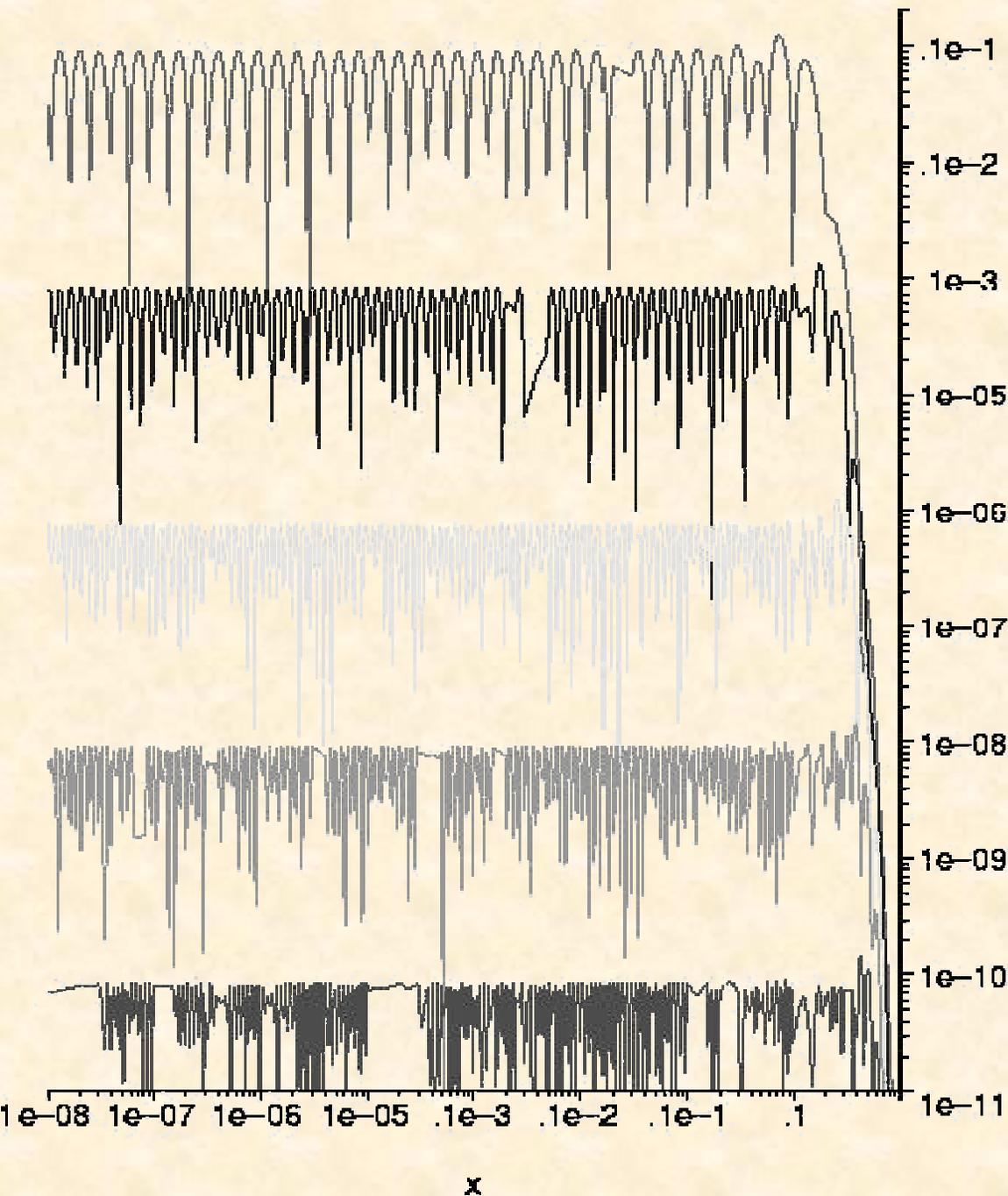
$$\frac{e^{-\mu r}}{r} = \frac{2}{\sqrt{\pi}} \int_0^{\infty} e^{-x^2 t^2 - \mu^2 / 4t^2} dt$$

$$= \frac{2}{\sqrt{\pi}} \int_{-\infty}^{\infty} e^{-x^2 e^{2s} - \mu^2 e^{-2s} / 4 + s} ds$$

- Trapezoidal quadrature
  - Geometric precision for periodic functions with sufficient smoothness.

The kernel for  $x=1e-4, 1e-3, 1e-2, 1e-, 1e0$ .  
 The curve for  $x=1e-4$  is the rightmost





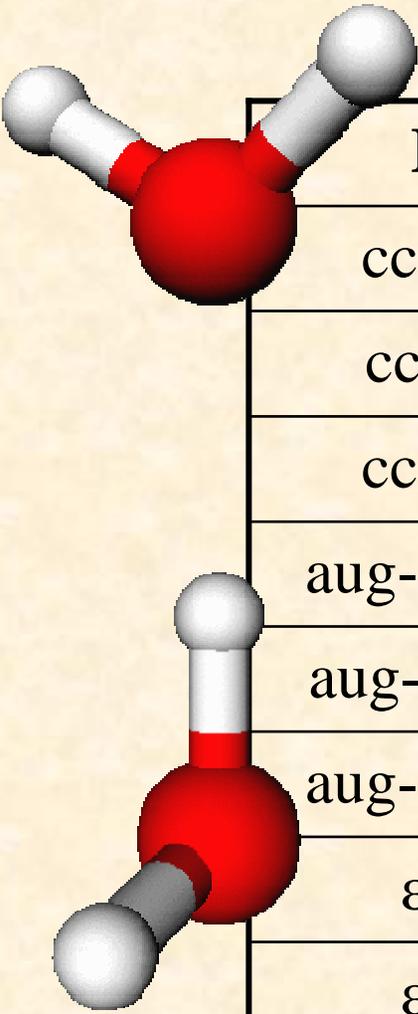
Automatically generated representations of  $\exp(-30r)/r$  accurate to  $1e-10$ ,  $1e-8$ ,  $1e-6$ ,  $1e-4$ ,  $1e-2$  (relative error) for  $r$  in  $[1e-8, 1]$  (92, 74, 57, 39 and 21 terms, respectively).

Low-energy *scattering* states also possible (but stronger dependence on range)

Periodic systems (cubic subgroups) straightforward.

# Water dimer LDA

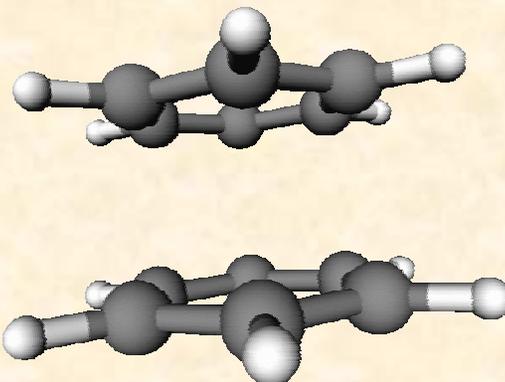
*aug-cc-pVTZ geometry, kcal/mol.*



<b>Basis</b>	<b>Uncorrected</b>	<b>BSSE</b>	<b>Corrected</b>
cc-pVDZ	-11.733	-3.958	-7.775
cc-pVTZ	-9.464	-1.654	-7.810
cc-pVQZ	-8.708	-0.821	-7.888
aug-cc-pVDZ	-8.187	-0.382	-7.805
aug-cc-pVTZ	-7.992	-0.086	-7.906
aug-cc-pVQZ	-7.995	-0.054	-7.941
$\epsilon=10^{-3}$	-6.483		
$\epsilon=10^{-5}$	-7.932		
$\epsilon=10^{-7}$	-7.943		

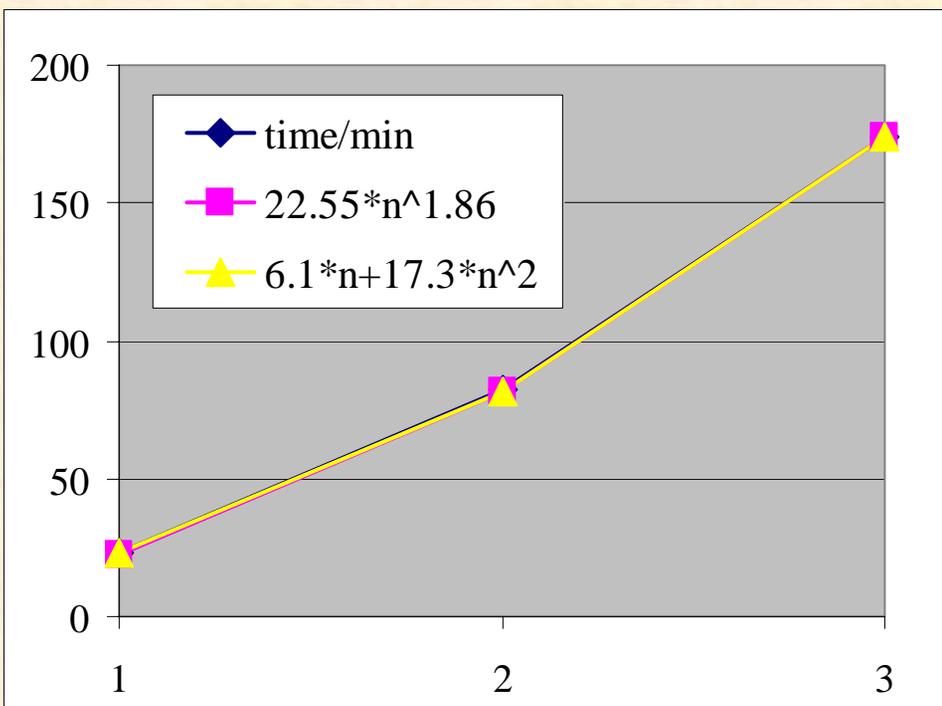
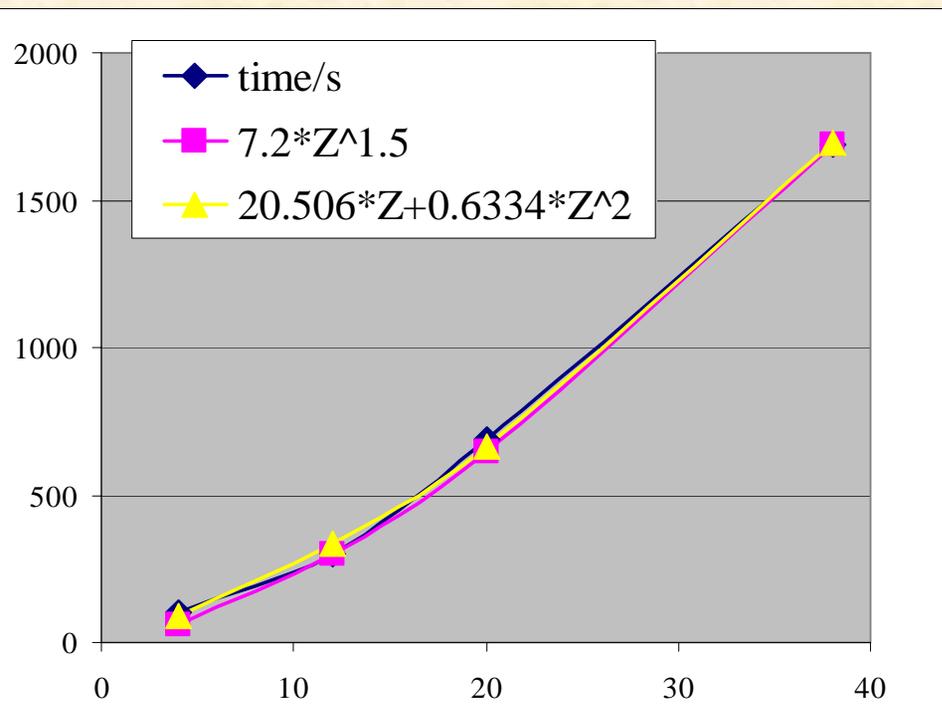
# Benzene dimer LDA

*aug-cc-pVDZ geometry, kcal/mol.*



Basis	Uncorrected	BSSE	Corrected
cc-pVDZ	-1.506	-1.035	-0.471
cc-pVTZ	-1.271	-0.387	-0.884
cc-pVQZ	-1.074	-0.193	-0.881
aug-cc-pVDZ	-1.722	-0.698	-1.024
aug-cc-pVTZ	-1.159	-0.193	-0.966
$\epsilon=10^{-5}$	-0.872		
$\epsilon=10^{-7}$	-0.956		
$\epsilon=10^{-9}$	-0.956		

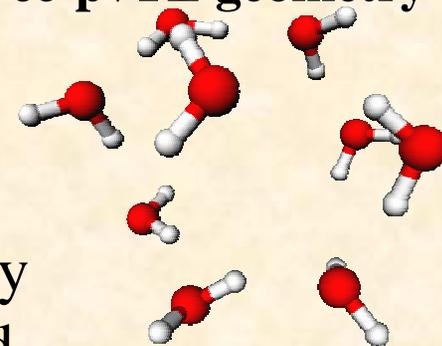
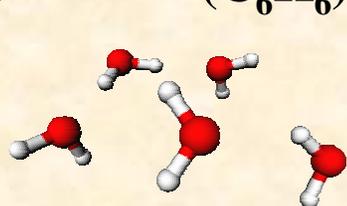
# LDA scaling with Z and system size (energy $\epsilon=10^{-5}$ )



Rare earth atoms  $Z=4,12,20,38$

$(C_6H_6)_n$  MP2 aug-cc-pvTZ geometry

$(H_2O)_n$   $n=5,9 \dots t = O(n^{1.1})$



Stacked benzene – MOs are delocalized by symmetry

Water cluster – MOs are asymptotically localized

(long tail is smooth so is inexpensively treated)<sup>27</sup>

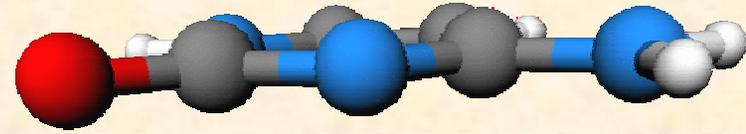
# *High-precision Hartree-Fock geometry for water*

- Pahl and Handy Mol. Phys. 100 (2002) 3199
  - Plane waves + polynomials for the core
  - Finite box ( $L=18$ ) requires extrapolation
  - Estimated error  $3\mu\text{H}$ ,  $1e-5$  Angstrom
- $k=11$ ,  $\text{conv.tol}=1e-8$ ,  $\epsilon=1e-9$ ,  $L=40$ 
  - Max. gradient =  $3e-8$ , RMS step= $5e-8$
  - Difference to Pahl  $10\mu\text{H}$ ,  $4e-6$  Angstrom, 0.0012

<b><u>Basis</u></b>	<b><u>OH</u></b>	<b><u>HOH</u></b>	<b><u>Energy</u></b>
$k=11$	0.939594	106.3375	-76.06818006
Pahl	0.939598	106.3387	-76.068170
cc-pVQZ	0.93980	106.329	-76.066676

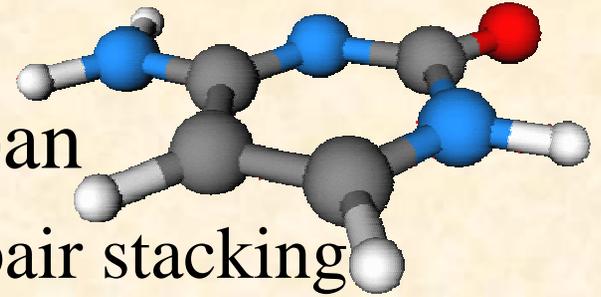
# *Current Capabilities*

- Open/closed shell Hartree-Fock and DFT
  - Wide range of GGAs, hybrid ( $O(N)$  HF exchange), and asymptotically corrected functionals
  - Energies and analytic derivatives
  - Full TDDFT and RPA for excitation energies \*\*\*\*\*
  - Abelian point groups
  - Parallel execution on shared memory computers
  - Interfaces to NWChem and GAMESS-US
- Working prototypes for computing in 6D
  - Direct solution of the pair equation for polyatomic systems (initial target is basis-set limit MP2)



## *Prototype code in use by ...*

- Hideo Sekino at Toyohashi, Japan
  - Benchmark calculations of base pair stacking
  - Electron transport in molecules
- Schaefer and Allen U. Georgia, Athens, USA
  - Benchmark HF energies and structures
- Tennant, U. Sheffield, England
  - New solvation models



# *High-level composition using functions and operators*

- Conventional quant. chem. uses explicitly indexed sparse arrays of matrix elements
  - Complex, tedious and error prone

- Python classes for Function and Operator

- in 1,2,3,6 and general dimensions

$$H\varphi = -\frac{1}{2}\nabla^2\varphi + V\varphi$$

- wide range of operations

$$J(r) = G^* \rho$$

```
Hpsi = -0.5*DelSq*psi+ V*psi
```

```
J = Coulomb.apply(rho)
```

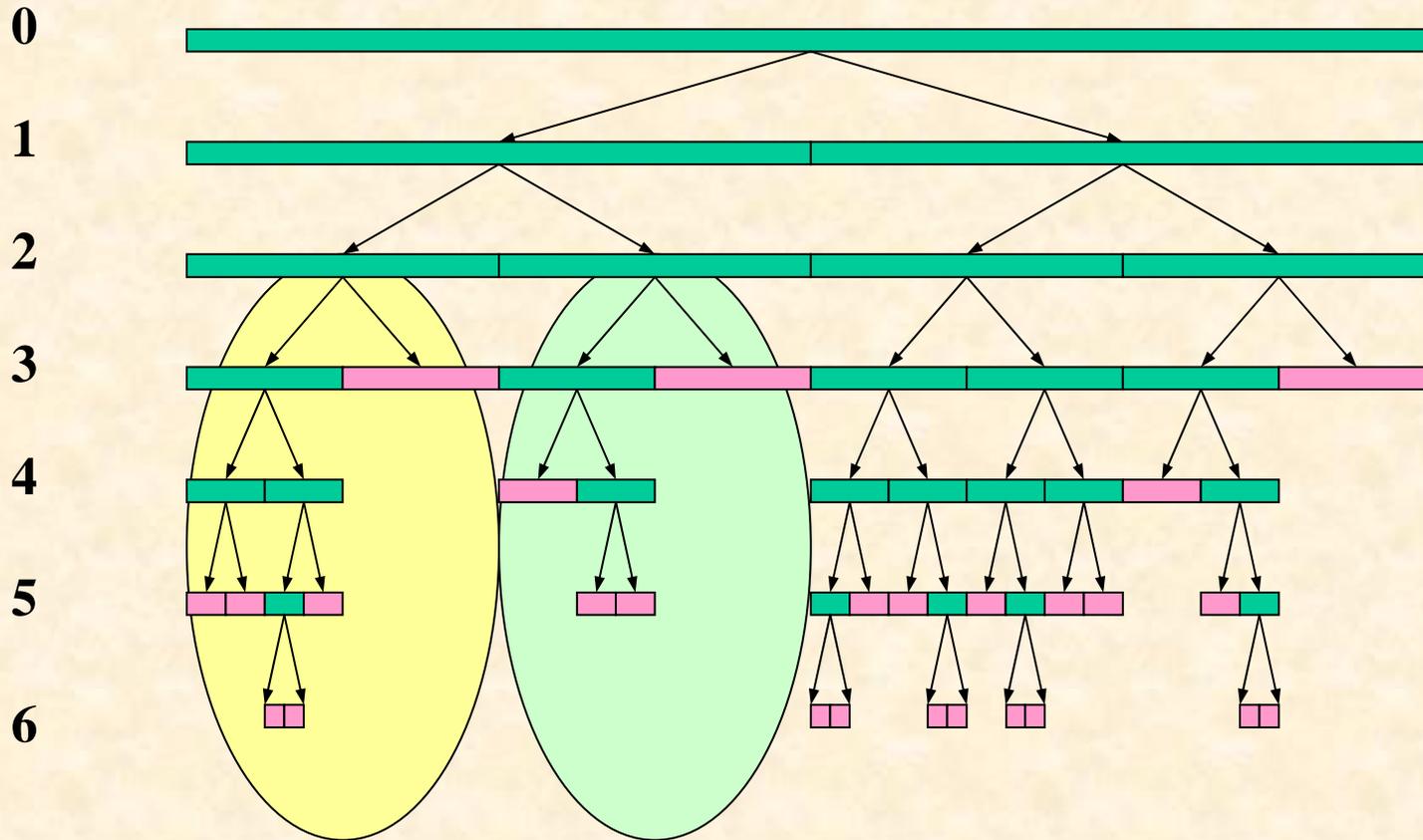
$$= \int \frac{\rho(s)}{|r-s|} ds$$

- All with guaranteed speed and precision

# *New solver being developed*

- Working with localized orbitals
  - $O(1)$  application of operators to one orbital
  - $O(N)$  computation of Coulomb potential (already)
  - $O(N)$  computation of Fock-like matrices
    - As a result of localized orbitals
  - More robust convergence
- Near total rewrite in C++
  - Two-levels of parallelism targeting massively parallel computer using multi-processor nodes
  - In anticipation of highly-threaded processors

# 1-D Example Sub-Tree Parallelism



Both sub-trees can be done in parallel.

In 3-D nodes split into 8 children ... in 6-D there are 64 children

# *Next Generation Languages/Mechanisms*

- DARPA HPCS – P is productivity not performance
- X10 (IBM)
  - Derivative of Java
  - Futures/Async(Dynamic Scheduling)
  - Var@Place (notion of locality)
- Chapel (Cray) (funded under DARPA HPCS)
  - Roots in ZPL, MTA stuff, and others
  - Futures
  - Domains (locality and scheduling)
- Fortress (Sun)

# *Futures*

- Parallel language construct that simplifies parallel execution with a dependency upon result
  - Store an unevaluated expression in a variable of type Future
  - May be executed by another thread as resources permit
  - Reference to the result forces it to be evaluated if it has not yet been done so.
- Implementation as templated C++ class with thread pool (similar interface to Java)

# *Recursive composition with Futures*

## Sequential

```
void Function::_reconstruct(OctTreeT *t) {  
    Tensor<double> ss = unfilter(t->data());  
  
    FORIJK(OctTreeT *child = t->child(i,j,k);  
        if (child) {  
            child->data()...;  
            _reconstruct(child);  
        }  
        else {  
            t->insert_child(...);  
        }  
    });  
};
```

## Multithreaded

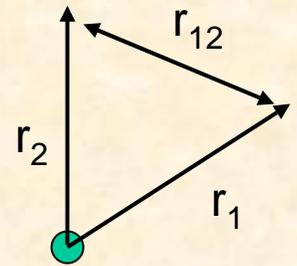
```
void Function::_reconstruct(OctTreeT *t) {  
    Tensor<double> ss = unfilter(t->data());  
    Future<void,Function,OctTreeT*> fut[2][2][2];  
    FORIJK(OctTreeT *child = t->child(i,j,k);  
        if (child) {  
            child->data()...;  
            fut[i][j][k].start(&Function::_reconstruct, this, child);  
        }  
        else {  
            t->insert_child(...);  
        }  
    });  
    FORIJK( fut[i][j][k].force(); );  
};
```

# Electron correlation

- All defects in the mean-field model are ascribed to electron correlation
- Consideration of singularities in the Hamiltonian imply that for a two-electron singlet atom (e.g., He)

$$\Psi(r_1, r_2, r_{12}) = 1 + \frac{1}{2} r_{12} + O(r_{12}^2) \quad \text{as } r_{12} \rightarrow 0$$

- Include the inter-electron distance in the wavefunction



- E.g., Hylleraas 1938 wavefunction for He

$$\Psi(r_1, r_2, r_{12}) = e^{-\zeta(r_1+r_2)} (1 + ar_{12} + \dots)$$

- Potentially very accurate, but not systematically improvable, and (until recently) not computationally feasible for many-electron systems

# Conventional approach

- The two-electron wave function is expanded as a product of one-particle functions (orbitals)

$$\Psi(r_1, r_2) = \sum_{ij} c_{ij} \phi_i(r_1) \phi_j(r_2)$$

- Can prove for atoms, that if saturate the atomic basis up to some angular momentum  $L$ , then

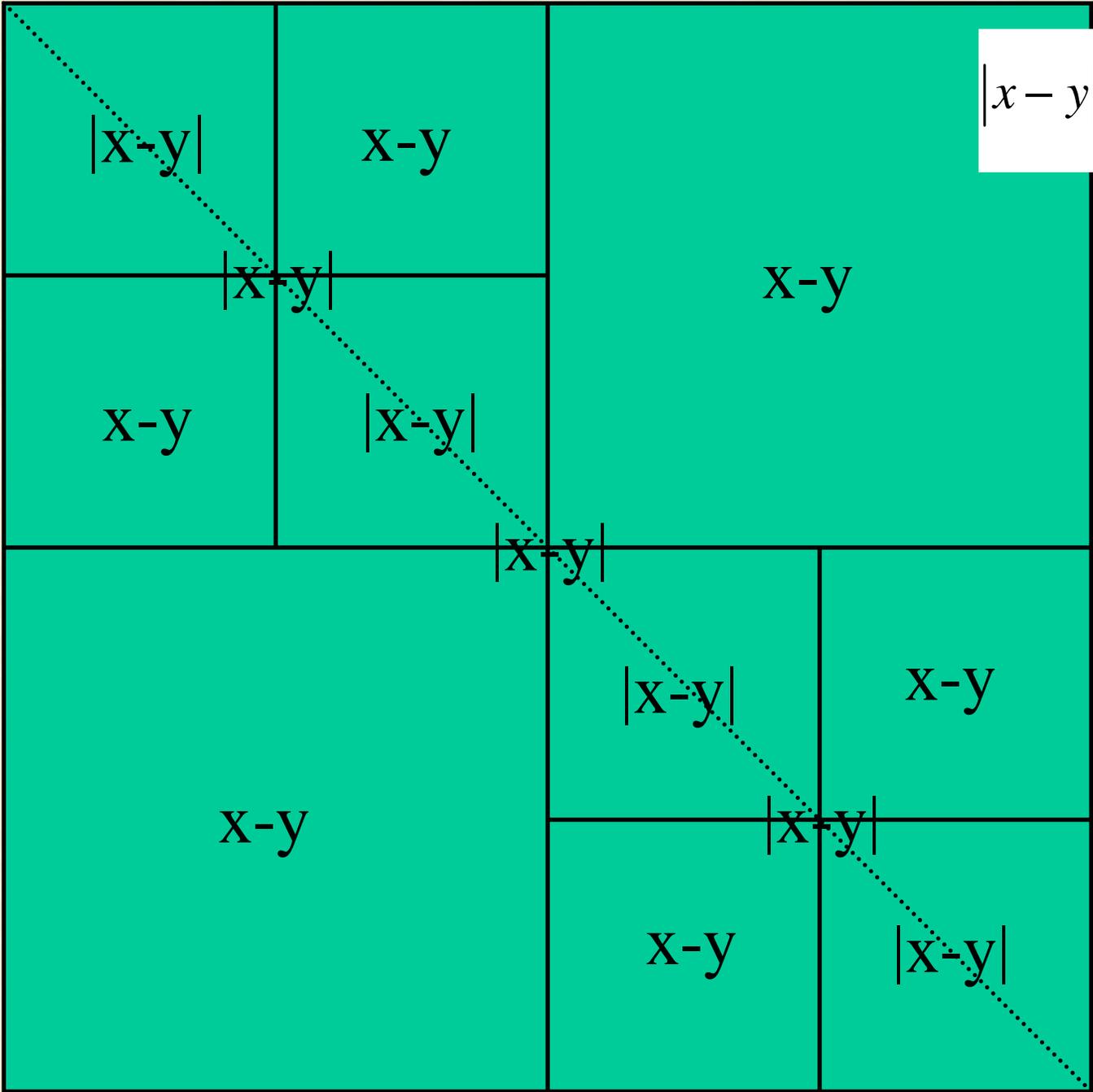
$$\Delta E_{corr} \propto (L + 1)^{-3}$$

L	d	f	g	h
$(L+1)^{-3}$	0.04	0.016	0.008	0.0046

- Correlation consistent basis sets (Dunning) are currently the best choice – cost is  $O(\varepsilon^{-4})$
- Explicitly correlated wave functions yields  $O(\varepsilon^{-2.4})$
- Fully numerical promises  $O(\log \varepsilon^{-1})$

$x \rightarrow$

$y \downarrow$



$$|x - y| = \sum_{\mu=1}^r f_{\mu}(x) g_{\mu}(y)$$

$r =$  separation rank

In 3D, ideally must be one box removed from the diagonal

Diagonal box has full rank

Boxes touching diagonal (face, edge, or corner) have increasingly low rank

Away from diagonal  $r = O(-\log \epsilon)$

# *Multiresolution solver of two-electron Schrödinger equation*

- Wavefunction in 6-D multiresolution representation
- Solve integral equation
  - The 6D GF nominally has 12 indices! Separated representation of operator accurate and efficient
- Partly or fully use SVD to represent 6-D tensor coefficient sets
  - Blocks separated from the diagonal have low rank (1 or 2, the full rank being  $k^3$ )
  - Directly analogous to linear CI expansion but not global
- Can compute directly in this form, but other refinements make it much more practical

# Analytic removal of cusp(s)

- Two-electron Hamiltonian
 
$$H = -\frac{1}{2}\Delta_1 - \frac{1}{2}\Delta_2 + V(r_1, r_2) + \frac{1}{|r_1 - r_2|}$$
- Two-electron wavefunction
 
$$\Psi(r_1, r_2) = e^{u(|r_1 - r_2|)} \phi(r_1, r_2)$$
- Transformed problem
 
$$e^{-u} H e^u \phi = -\frac{1}{2}\Delta_1 \phi - \frac{1}{2}\Delta_2 \phi - u' \frac{r_1 - r_2}{|r_1 - r_2|} \cdot (\nabla_1 \phi - \nabla_2 \phi) + \left( V - ((u')^2 + u'') + \frac{1}{|r_1 - r_2|} (1 - 2u') \right) \phi$$
- Choose  $u$  to eliminate singularity at  $r_{12}=0$
- Quantum Monte Carlo calculations
  - Best to eliminate all  $r_1=0, r_2=0, r_{12}=0, r_1=r_2=r_{12}=0$

# Smoothed potential and wave function

- Similarity-transformed Hamiltonian with correlation factor (cf. transcorrelated Hamiltonian)
  - The effective wavefunction  $\Phi$  as well as the transformed Hamiltonian is smoothed at  $r_{12}=0$

$$\bar{H} = e^{-u(r_{12})} \hat{H} e^{u(r_{12})}$$

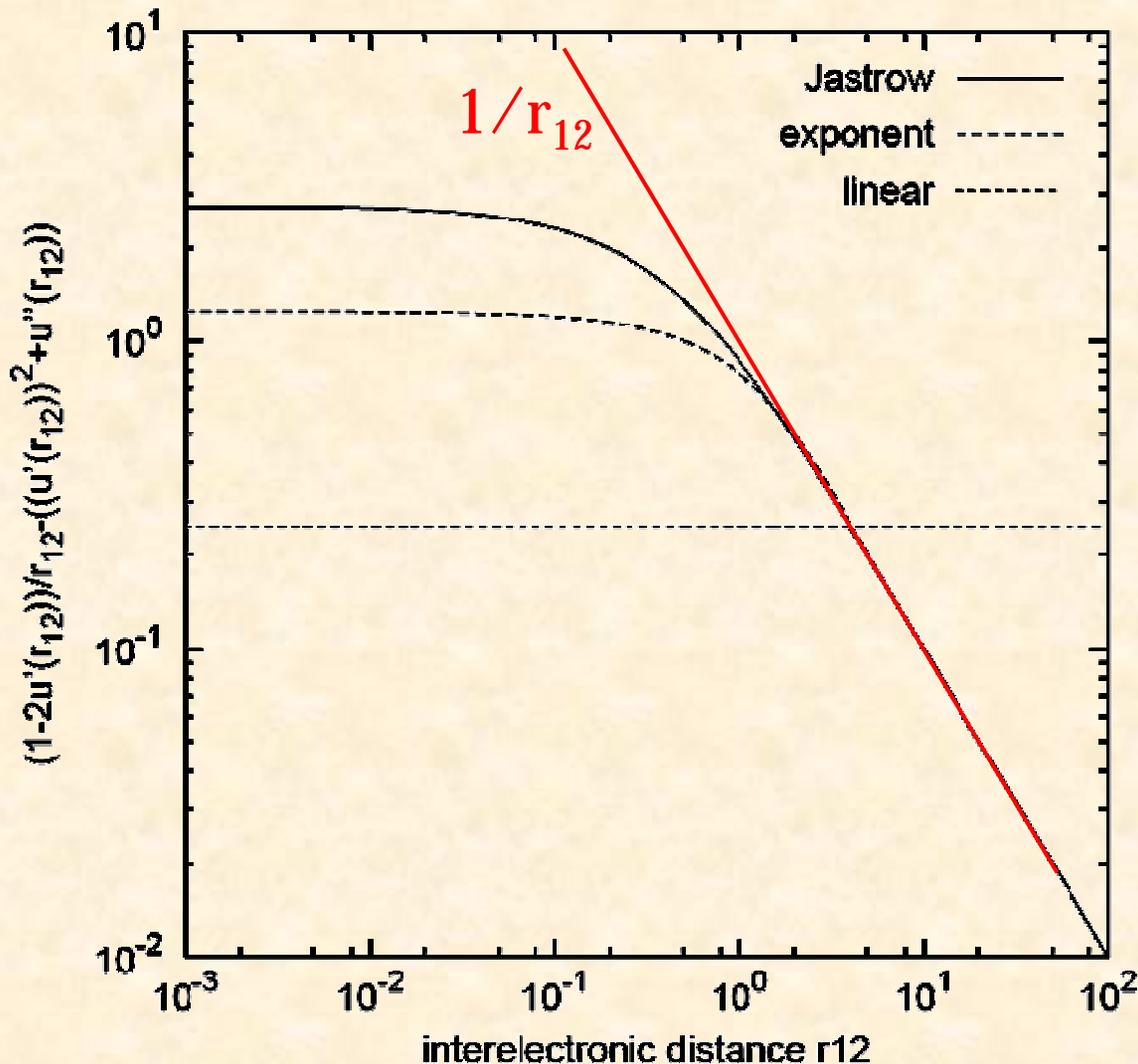
Correlation factor:  $u(r_{12})$

$$\Psi(r_1, r_2) = e^{u(r_{12})} \Phi(r_1, r_2)$$

- Electron-electron repulsion is smoothed

$$\frac{1}{r_{12}} \rightarrow \frac{1}{r_{12}} (1 - 2u'(r_{12})) - u'(r_{12})^2 - u''(r_{12}) - u'(r_{12}) \frac{\mathbf{r}_1 - \mathbf{r}_2}{r_{12}} \cdot (\nabla_1 - \nabla_2)$$

# Smoothed interelectron potential



- Correlation factor

- Jastrow

$$u(r_{12}) = \frac{ar_{12}}{1 + br_{12}}$$

- Exponential

$$u(r_{12}) = a(1 - e^{-br_{12}})$$

- Linear

$$u(r_{12}) = \frac{1}{2}r_{12}$$

# *Solve for the correlation correction to the HF wavefunction*

- The smoothed wavefunction is separated into Hartree-Fock wavefunction and its perturbation.
  - The perturbative wavefunction is numerically smaller than Hartree-Fock wavefunction.

$$\Phi(r_1, r_2) = \Phi^{HF}(r_1, r_2) + \underline{\delta\Phi(r_1, r_2)}$$

$$\left\{ \begin{array}{l} \|\Phi^{HF}(r_1, r_2)\|_2 = 1.0 \\ \|\underline{\delta\Phi(r_1, r_2)}\|_2 \approx 0.1 \quad \text{For He atom} \end{array} \right.$$

# *Preliminary results for He atom*

	Variational E	$\Delta E$	residual
<b>HF</b>	<b>-2.861 61</b>		
<b>Iter. 0</b>	<b>-2.871 08</b>		<b>0.414 73</b>
<b>1</b>	<b>-2.894 92</b>	<b>-0.023 84</b>	<b>0.017 28</b>
<b>2</b>	<b>-2.900 43</b>	<b>-0.005 51</b>	<b>0.007 94</b>
<b>3</b>	<b>-2.902 18</b>	<b>-0.001 75</b>	<b>0.003 84</b>
<b>4</b>	<b>-2.902 88</b>	<b>-0.000 70</b>	<b>0.002 02</b>
<b>5</b>	<b>-2.903 20</b>	<b>-0.000 32</b>	<b>0.001 25</b>
<b>6</b>	<b>-2.903 39</b>	<b>-0.000 20</b>	<b>0.000 91</b>
<b>...</b>	<b>...</b>	<b>...</b>	<b>...</b>
<b>12</b>	<b>-2.903 73</b>	<b>-0.000 04</b>	<b>0.000 36</b>
<b>13</b>	<b>-2.903 73</b>	<b>+0.000 004</b>	<b>0.000 32</b>
<b>14</b>	<b>-2.903 77</b>	<b>-0.000 04</b>	<b>0.000 28</b>

## **Computational details:**

- 5-th order multiwavelets
- Wavelet threshold:  $2 \times 10^{-5}$
- SVD threshold:  $2 \times 10^{-6}$
- Exponential correlation factor

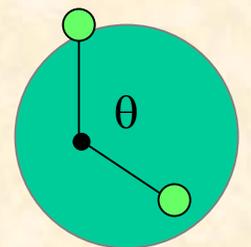
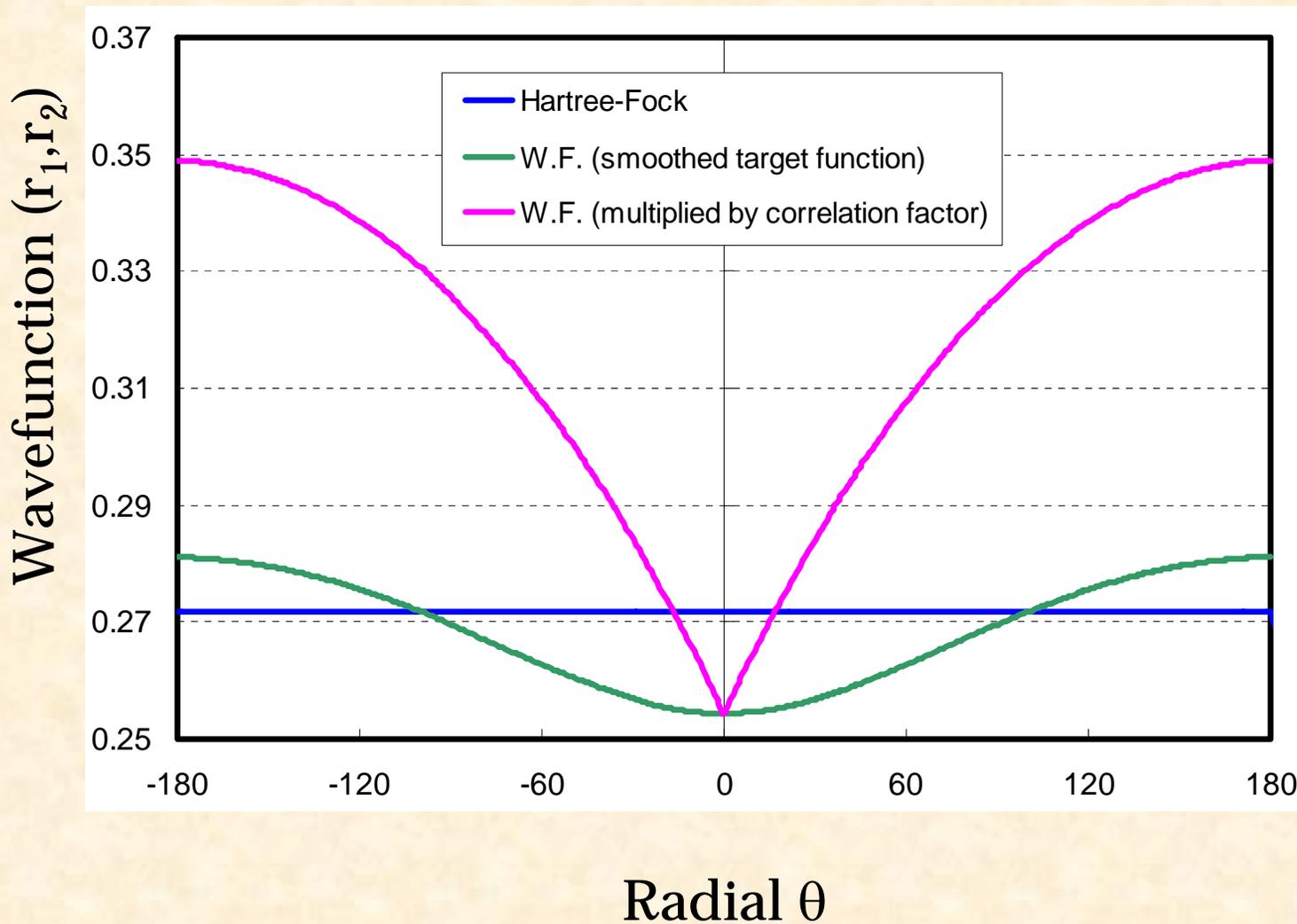
## **Perturbative wavefunction:**

- Maximum refinement:  $n=4$
- Memory: 132M in full SVD form

<b>exact</b>	<b>-2.903 74 (E(HF)=-2.861 68)</b>
<b>Hylleraas (6 terms)</b>	<b>-2.903 24</b>
<b>Löwdin and Redei</b>	<b>-2.895 4</b>
<b>cc-pV6Z</b>	<b>-2.903 48 (FCI) (E(HF)= -2.861 67)</b>

-Energy is variational  
(small non-variational is  
just truncation err)

# *Coulomb hole (He)*



$$r_1 = r_2 = 0.5a_0$$

# Summary

- Multiresolution analysis provides a general framework for computational chemistry
  - Accurate and efficient with high-level composition
  - Multiwavelets provide high-order convergence and readily accommodate singularities/boundary conditions
  - General framework readily accessible to researchers
  - Real impact will be application to many-body models
- Separated form for operators and functions
  - Critical for efficient computation in higher dimension
- Precision is guaranteed
  - Excited states, non-linear response, ...