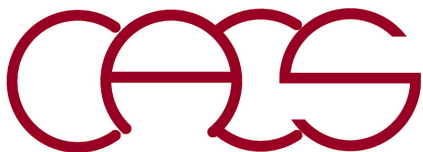# Multiresolution Methods

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Divide-&-conquer in continuum simulations
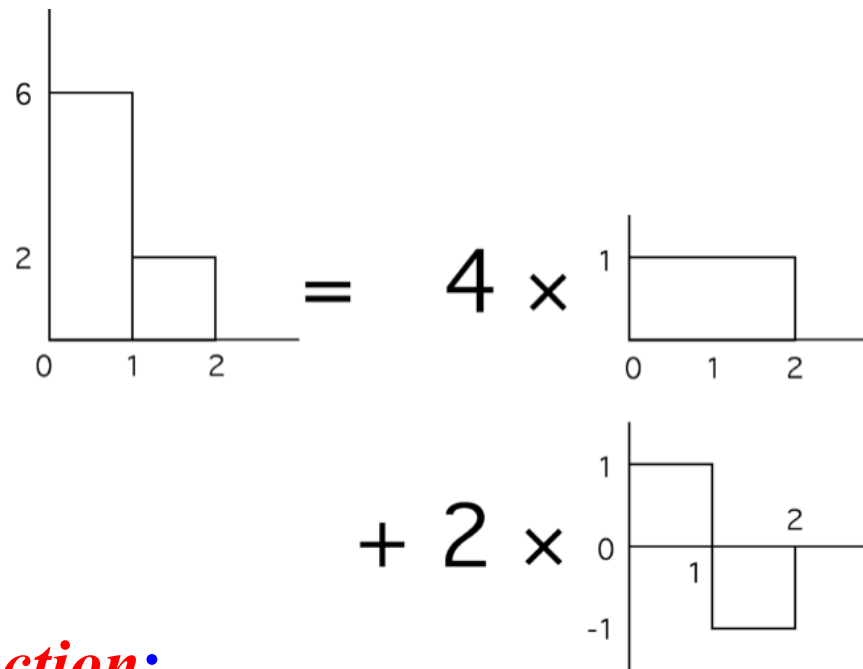
Discrete *vs*. Continuum Applications

Figure 2. Newton and Schrödinger's cat. Previously, classical physics and quantum chemistry belonged to rivalling worlds. The Nobel Laureates in Chemistry 2013 have opened a gate between those worlds and have brought about a flourishing collaboration.

https://www.nobelprize.org/prizes/chemistry/2013/summary/

- **Discrete simulation: *e.g.*, molecular dynamics (Newtonian mechanics)**
- **Continuum simulation: *e.g.*, quantum dynamics (Schrödinger equation); *cf.* image processing**
- **Multiresolution methods in the context of image processing**

Haar Wavelet Basis

- **One-dimensional "image":** `I[2]` = (6, 2)
- **Smooth component:** `s` = (6 + 2)/2 = 4
- **Detailed component:** `d` = (6 − 2)/2 = 2
- **Wavelet decomposition:** `I[]` = (6,2) = 4 × (1,1) + 2 × (1,–1)

- **Haar *scaling function*:**
  $\phi(x) = 1$ $(0 \leq x < 2)$; 0 (otherwise)
- **Haar *wavelet function*:**
  $\psi(x) = 1$ $(0 \leq x < 1)$; -1 $(1 \leq x < 2)$; 0 (otherwise)

Wavelet Decomposition

- **One-dimensional "image":**
  `I[16]=(1,2,5,9,1,9,2,2,2,3,5,7,4,2,1,1)`

- **Smooth component:**
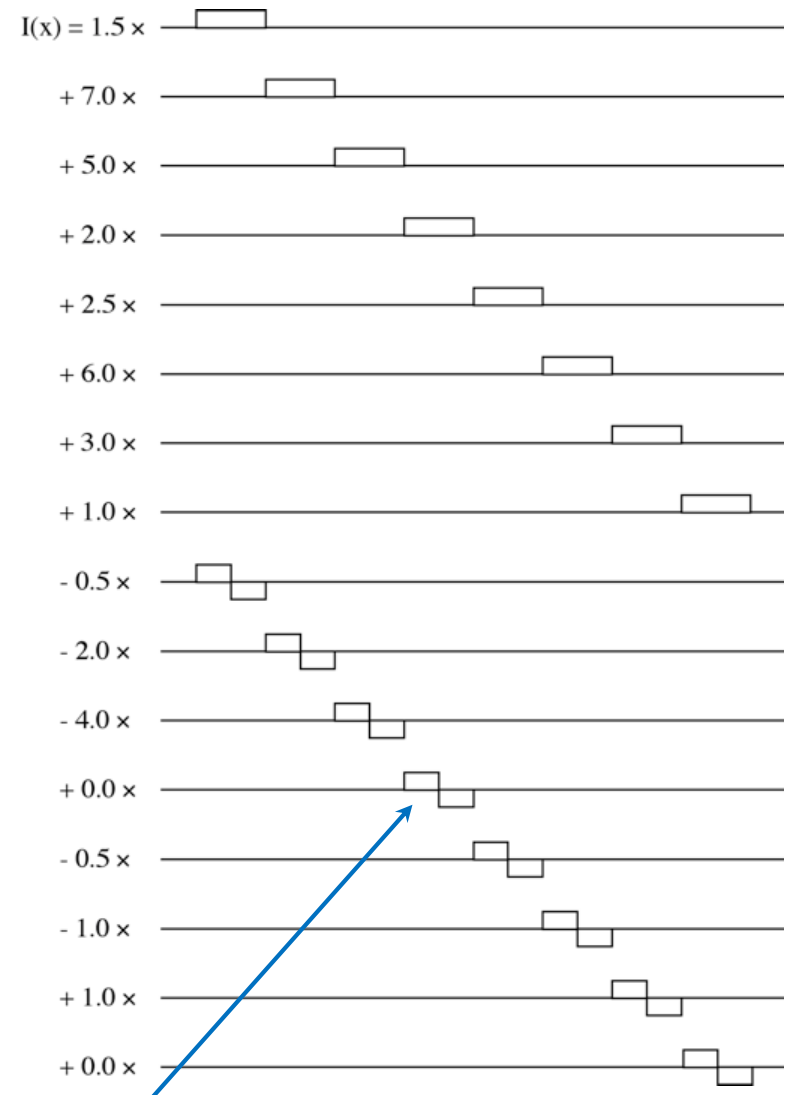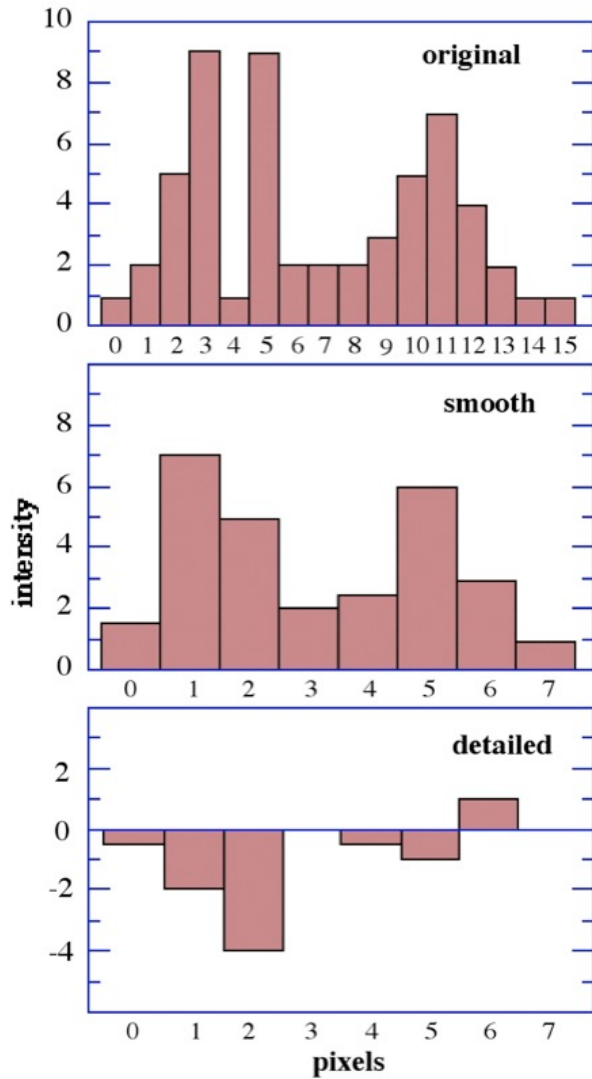 `s[i] = (I[2*i]+I[2*i+1])/2 (i=0,...,7)`

- **Detailed component:**
  `d[i] = (I[2*i]-I[2*i+1])/2 (i=0,...,7)`

- **Wavelet decomposition:**

```
I[16]   1   2   5   9   1   9   2   2   2   3   5   7   4   2   1   1
s[8]     1.5     7.0     5.0     2.0     2.5     6.0     3.0     1.0
d[8]       -0.5    -2.0    -4.0     0.0    -0.5    -1.0     1.0     0.0
```

Wavelet Decomposition

I[16]	1	2	5	9	1	9	2	2	2	3	5	7	4	2	1	1
s[8]	1.5		7.0		5.0		2.0		2.5		6.0		3.0		1.0	
d[8]		−0.5		−2.0		−4.0		0.0		−0.5		−1.0		1.0		0.0

Wavelets = spatially localized waves also localized in wavenumber

Multiresolution Analysis

- **Recursive wavelet decomposition:**

```
I[16] -> s[8], d[8]
        s[8] -> ss[4], sd[4]
                ss[4] -> sss[2], ssd[2]
                        sss[2] -> ssss[1], sssd[1]
```

- **Recursive vector-subspace decomposition:**

$$V^0 = V^1 + W^1; \text{s[8]} \in V^1; \text{d[8]} \in W^1$$
$$V^1 = V^2 + W^2; \text{ss[4]} \in V^2; \text{sd[4]} \in W^2$$
$$...$$

- **Multiresolution representation**

```
I[16] -> d[8], sd[4], ssd[2], sssd[1], ssss[1]
```
or
$$V^0 = V^1 + W^1$$
$$= V^2 + W^2 + W^1$$
$$= V^3 + W^3 + W^2 + W^1$$
$$= V^4 + W^4 + W^3 + W^2 + W^1$$

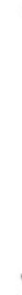**Very smooth**   **Progressively more details**

Wavelet Image Decomposition

- **Alternate row & column transformations**

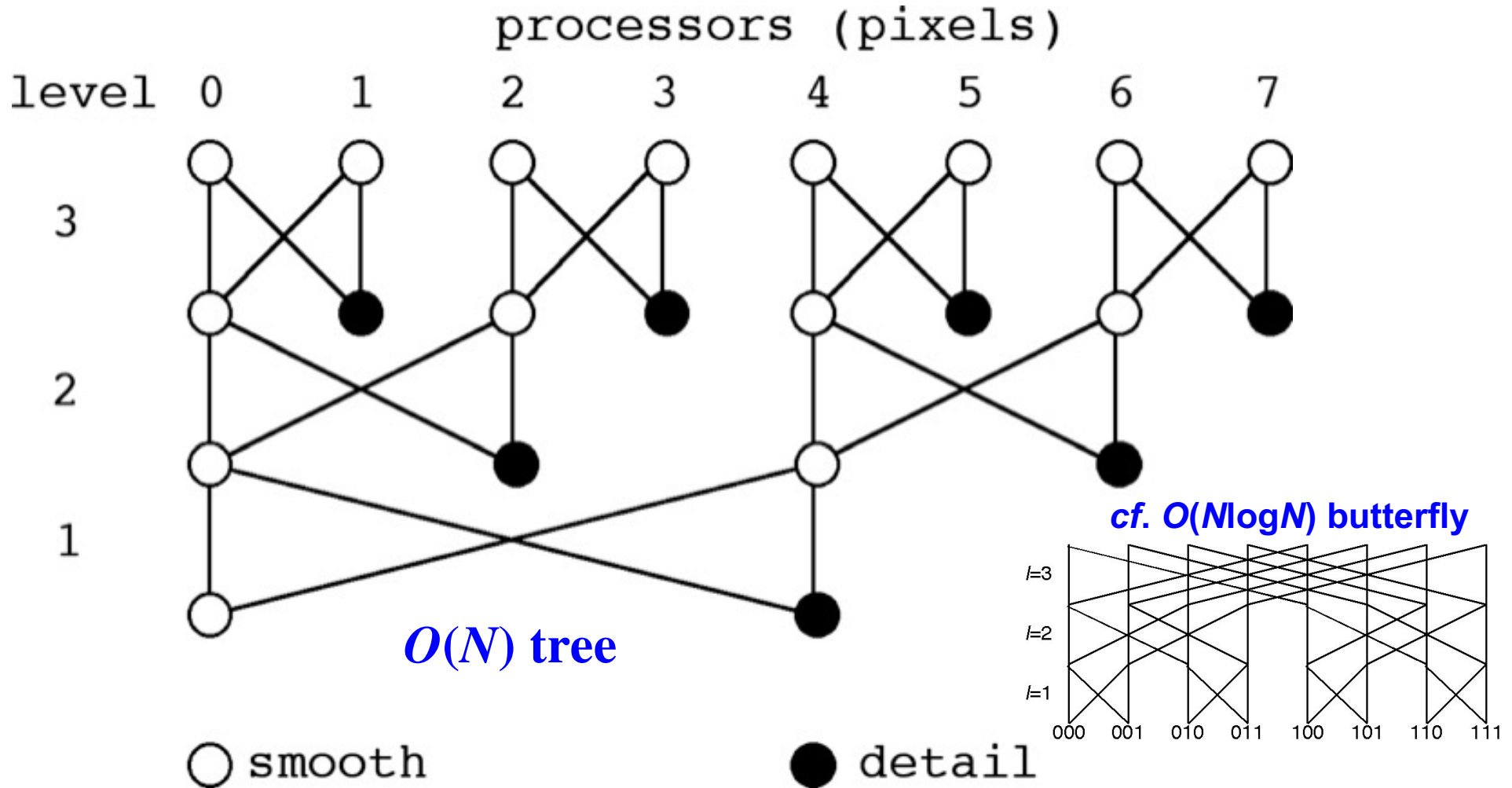transform rows →

transform columns ↓

- **Wavelet transform is invertible, like Fourier transform**

- **Wavelets are localized in both real & reciprocal (wavenumber) spaces, unlike plane waves in Fourier transform**

- **Wavelet transform is $O(N)$—scalable tree, while fast Fourier transform (FFT) is $O(N\log N)$—slightly less scalable hypercube**

- **Image compression? Discard sub-threshold wavelet-expansion coefficients**

Parallel Multiresolution Analysis

- **Local with spatial decomposition at fine scales**
- **Subtree masters own coarse smooth components**

- **Retain detailed components? Let subtree slaves do (*cf*. hypercube quicksort)**

Daubechies Wavelets

- **Moving window for smooth & detailed filters**

$$
\begin{bmatrix}
c_0 & c_1 & c_2 & c_3 & & & & & \\
c_3 & -c_2 & c_1 & -c_0 & & & & & \\
 & & c_0 & c_1 & c_2 & c_3 & & & \\
 & & c_3 & -c_2 & c_1 & -c_0 & & & \\
\vdots & \vdots & & & & & \ddots & & \\
 & & & & & & c_0 & c_1 & c_2 & c_3 \\
 & & & & & & c_3 & -c_2 & c_1 & -c_0 \\
c_2 & c_3 & & & & & & & c_0 & c_1 \\
c_1 & -c_0 & & & & & & & c_3 & -c_2
\end{bmatrix}
\begin{bmatrix}
y_1 \\ y_2 \\ y_3 \\ y_4 \\ y_5 \\ y_6 \\ y_7 \\ y_8 \\ y_9 \\ y_{10} \\ y_{11} \\ y_{12} \\ y_{13} \\ y_{14} \\ y_{15} \\ y_{16}
\end{bmatrix}
\longrightarrow
\begin{bmatrix}
s_1 \\ d_1 \\ s_2 \\ d_2 \\ s_3 \\ d_3 \\ s_4 \\ d_4 \\ s_5 \\ d_5 \\ s_6 \\ d_6 \\ s_7 \\ d_7 \\ s_8 \\ d_8
\end{bmatrix}
$$

```
C0 = (1+√3)/4√2 =  0.4829629131445341
C1 = (3+√3)/4√2 =  0.8365163037378079
C2 = (3-√3)/4√2 =  0.2241438680420134
C3 = (1-√3)/4√2 = -0.1294095225512604
```


DAUB4 $e_{10} + e_{58}$

Ingrid Daubechies

Iterative Solution of Linear Systems

$$\mathbf{Ax = b}$$

$$\mathbf{A} = \mathbf{L} + \mathbf{D} + \mathbf{U}$$

$$
\begin{bmatrix} & & & \\ X & & & \\ X & X & & \\ X & X & X & \end{bmatrix}
\begin{bmatrix} X & & & \\ & X & & \\ & & X & \\ & & & X \end{bmatrix}
\begin{bmatrix} & X & X & X \\ & & X & X \\ & & & X \\ & & & \end{bmatrix}
$$

- **Fixed-point equation**

$$\mathbf{x = D^{-1}[-(L+U)x + b]}$$

$$\mathbf{(D + L + U)x = b}$$
$$\mathbf{Dx = -(L + U)x + b}$$

- **Jacobi iteration**

$$\mathbf{x^{(n+1)} = D^{-1}[-(L+U)x^{(n)} + b]}$$

$$x_i^{(n+1)} = \frac{1}{a_{ii}}\left(-\sum_{\substack{j=1 \\ (j \neq i)}}^{N} a_{ij}x_j^{(n)} + b_i\right)$$

More Iterative Smoothing

$$x_i^{(n+1)} = \frac{1}{a_{ii}}\left(-\sum_{\substack{j=1 \\ (j\neq i)}}^{N} a_{ij}x_j^{(n)} + b_i\right) = x_i^{(n)} + \frac{1}{a_{ii}}\left(\overbrace{-\sum_{j=1}^{N} a_{ij}x_j^{(n)} + b_i}^{\text{residual}}\right)$$

$$\xrightarrow{\text{over|under relaxation}} x_i^{(n)} + \frac{\Delta}{a_{ii}}\left(-\sum_{j=1}^{N} a_{ij}x_j^{(n)} + b_i\right)$$
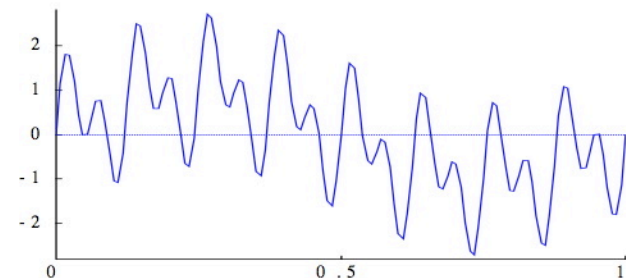
Over ($\Delta > 1$) or under ($\Delta < 1$) relaxation to accelerate convergence

- **More general fixed-point iteration**

$$\mathbf{x} \leftarrow \mathbf{x} + \mathbf{Z}\left(\overbrace{-\mathbf{Ax} + \mathbf{b}}^{\text{residual}}\right) = (\mathbf{I} - \mathbf{ZA})\mathbf{x} + \mathbf{Zb}$$

- **High-frequency residual (error) dies out quickly**

- Initial error:

- Error after 35 iteration sweeps:

Multigrid Method

- **Residual equation:**

  $A^{(l)}$: l-th level matrix
  v: Current guess
  e: error vector
  r: residual vector

$$A^{(l)}\overbrace{(v+e)}^{\text{exact}} = -4\pi e^2 n$$
$$-)\ A^{(l)}v = -4\pi e^2 n + r$$
$$\overline{A^{(l)}e = -r}$$

- **Smoothing:**

$$e \leftarrow \left[1 + Z^{(l)}A^{(l)}\right]e + Z^{(l)}r$$

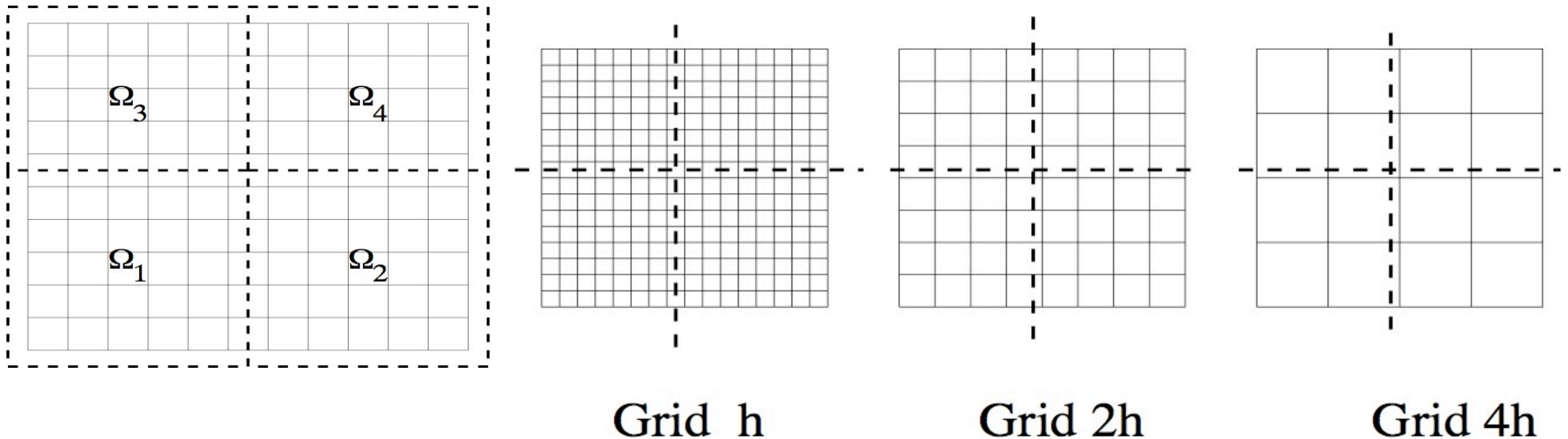- **Coarsening of residual & interpolation of error**

Achi Brandt

r: restrict (to coarser grid)
i: interpolate (to finer grid)

V-cycle

(S) smoothing   (S) $l = 3$

r   i

(S)   (S) $l = 2$

r i

E   exact solution $l = 1$

Full multigrid

$l = 4$

$l = 3$

$l = 2$

$l = 1$

Parallel Multigrid Method

- **Domain decomposition with boundary-layer caching**

Grid h   Grid 2h   Grid 4h

- **2D computational & communication costs (isogranular or weak scaling)**

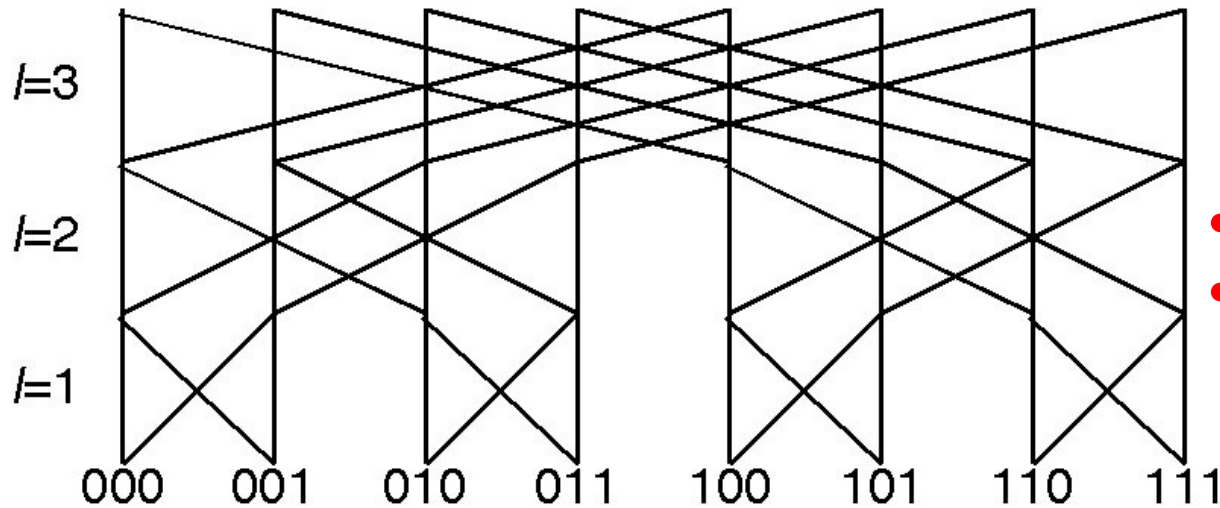$N \times N$ grids each on $P \times P$ processors: $T(N^2 P^2, P^2) = a \log NP + bN + cN^2$

Weak-scaling speedup & efficiency

$$S_{P^2} = \frac{N^2 P^2 T(N^2, 1)}{N^2 T(N^2, P^2)} = \frac{P^2 (cN^2)}{a \log NP + bN + cN^2} = \frac{P^2}{1 + \dfrac{b}{cN} + \dfrac{a}{cN} \log NP}$$

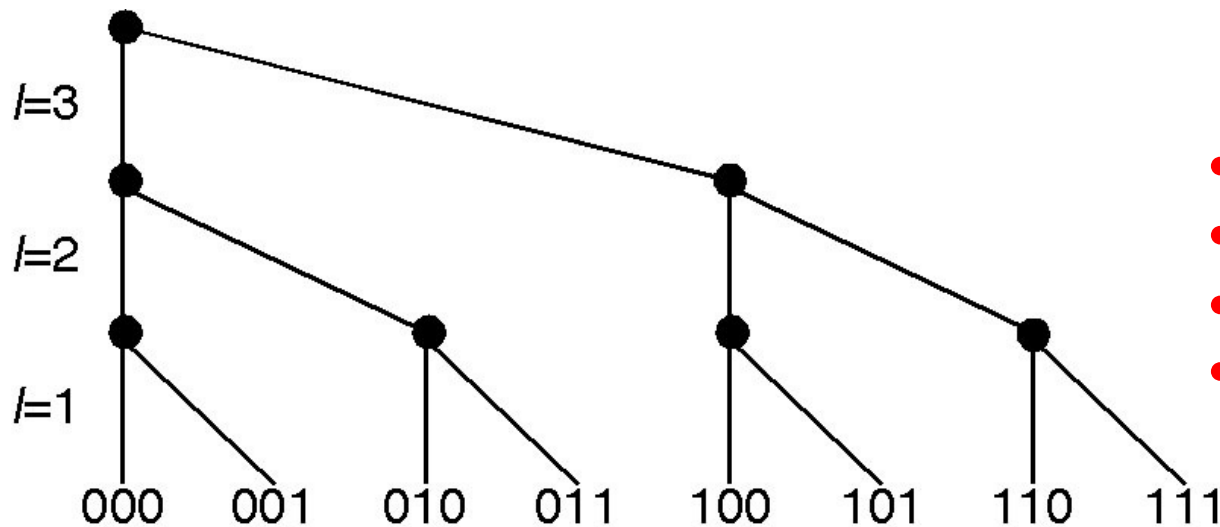$$E_{P^2} = \frac{S_{P^2}}{P^2} = \frac{1}{1 + \dfrac{b}{cN} + \dfrac{a}{cN} \log NP}$$

Global Communications

All-to-all (hypercube): $O(N\log N)$

- **Quicksort**
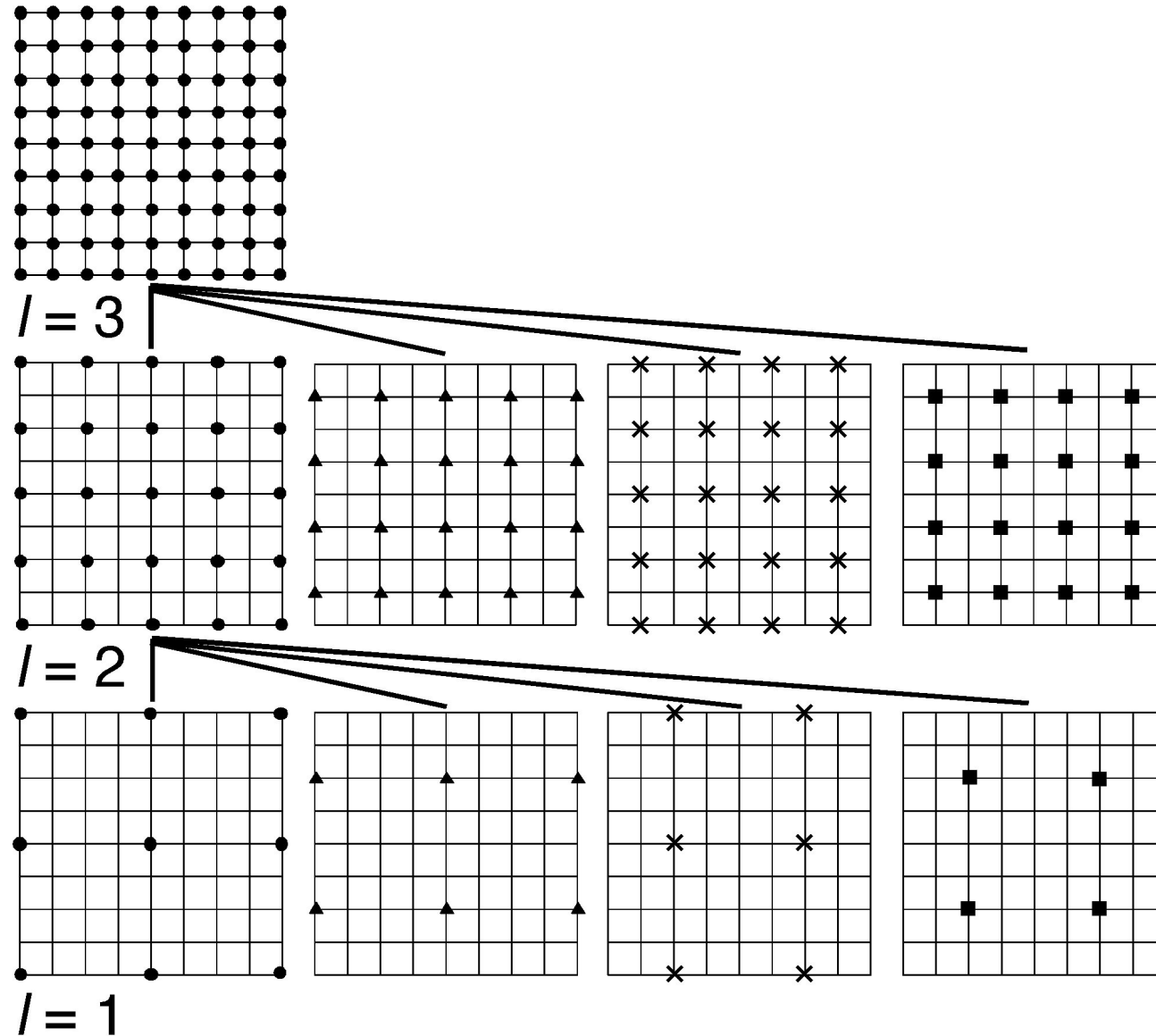- **Fast Fourier transform**
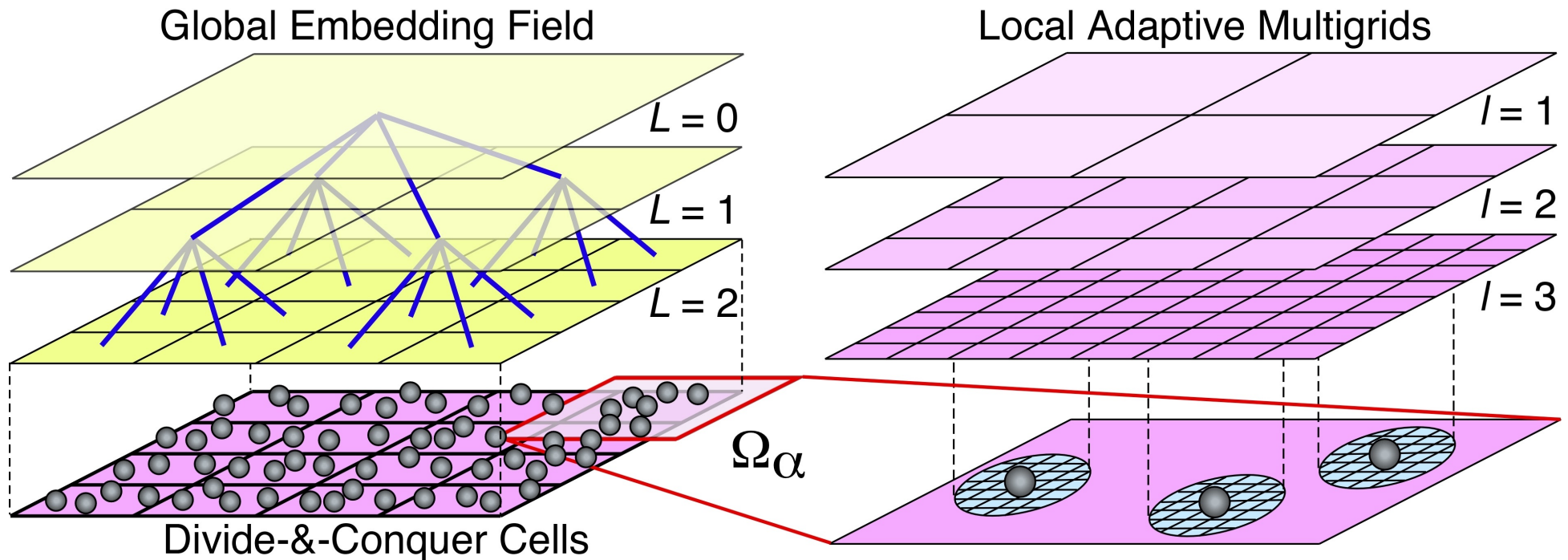
All-to-one (tournament): $O(N)$

- **Global reduction**
- **Fast multipole method**
- **Multigrid method**
- **Wavelets**

Solving the Idle Processor Problem

- **Parallel superconvergent multigrid:** Solve multiple coarse problems to accelerate the convergence [Frederickson & McBryan, '88]

l = 3

l = 2

l = 1

Divide-&-Conquer Algorithms

Global Embedding Field

$L = 0$

$L = 1$

$L = 2$

Divide-&-Conquer Cells

Ω_α

Local Adaptive Multigrids

$l = 1$

$l = 2$

$l = 3$

- *N-body problem*: $O(N^2) \rightarrow O(N)$
  - **> Space-time multiresolution molecular dynamics (MRMD): Fast multipole method & symplectic multiple time stepping**

- **Variable *N*-charge problem**: $O(N^3) \rightarrow O(N)$
  - **> Fast reactive force-field (F-ReaxFF) MD: Multilevel preconditioning**

- **Quantum *N*-body problem**: $O(C^N) \rightarrow O(N)$
  - **> DC density functional theory (DC-DFT): Adaptive multigrids**

A. Nakano *et al.*, *Int'l J. High Performance Comput. Appl.* **22**, 113 ('08)

Molecular Dynamics: *N*-Body Problem

- **Newton's equations of motion**

$$m_i \frac{d^2 \mathbf{r}_i}{dt^2} = -\frac{\partial E_{\mathrm{MD}}\left(\mathbf{r}^N\right)}{\partial \mathbf{r}_i} \quad (i = 1,\ldots,N)$$

- **Reliable interatomic potential**

$$E_{\mathrm{MD}} = \sum_{i<j} u_{ij}\left(r_{ij}\right) + \sum_{i,j<k} v_{jik}\left(\mathbf{r}_{ij},\mathbf{r}_{ik}\right)$$
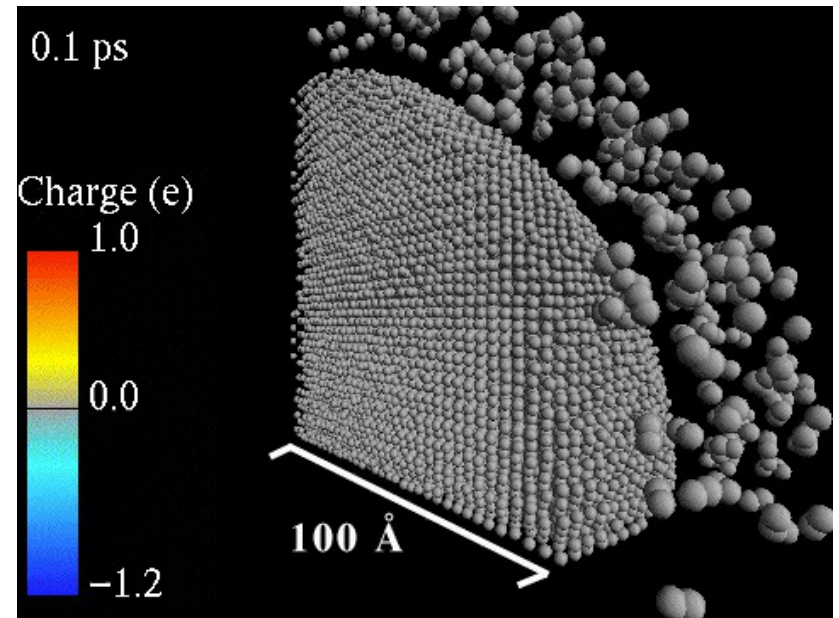

- *N*-body problem
  Long-range electrostatic interaction — *O(N²)*

$$\text{Evaluate } V_{\mathrm{es}}(\mathbf{x}) = \sum_{j=1}^{N} \frac{q_j}{|\mathbf{x}-\mathbf{x}_j|} \text{ at } \mathbf{x} = \mathbf{x}_i \ (i = 1,\ldots,N)$$

- *O(N)* **space-time multiresolution MD (MRMD) algorithm**
  1. **Fast multipole method (FMM)** [Greengard & Rokhlin, '87]
  2. **Symplectic multiple time stepping (MTS)** [Tuckerman *et al*., '92]

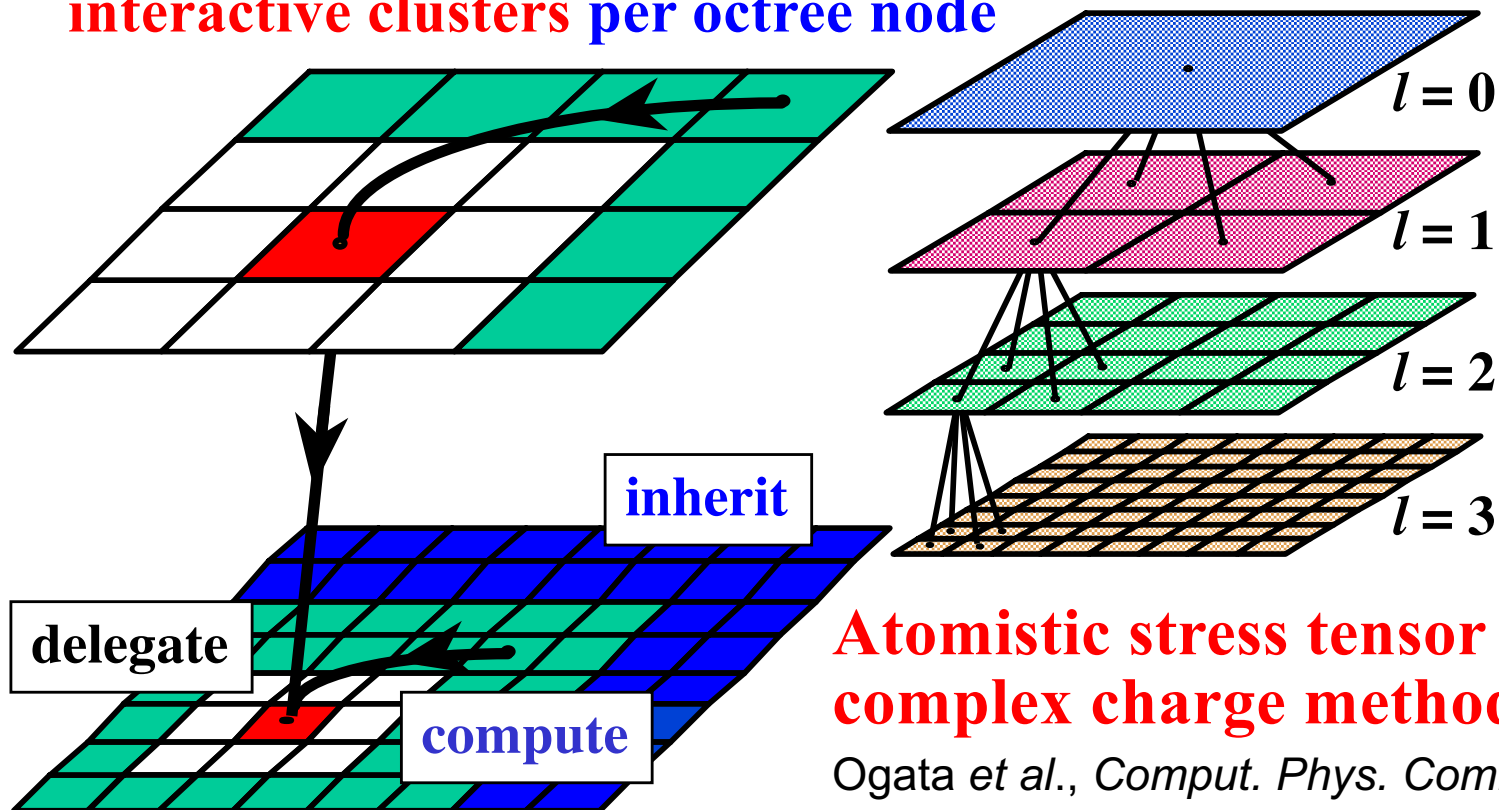A. Nakano, *Comput. Phys. Commun.* **83**, 197 ('94)

Spatial Locality: Fast Multipole Method

1. Clustering: Encapsulate far-field information using multipoles

$$V(\mathbf{x}) = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} \left\{ \sum_{i=1}^{N} q_i r_i^l Y_l^{*m}(\theta_i, \phi_i) \right\} \frac{Y_l^m(\theta, \phi)}{r^{l+1}}$$

2. Hierarchical abstraction: Octree data structure

3. $O(N)$ algorithm: Constant number of interactive clusters per octree node

(r_i, θ_i, ϕ_i)

(r, θ, ϕ)

$l = 0$

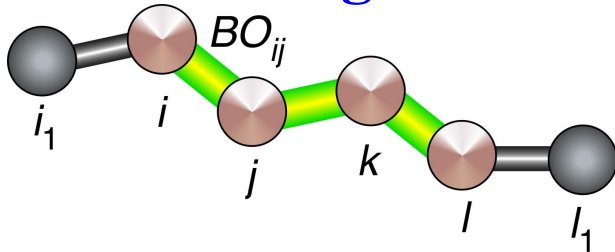$l = 1$

$l = 2$

$l = 3$

inherit

delegate

compute

Atomistic stress tensor by a novel complex charge method (CCM)

Ogata *et al.*, *Comput. Phys. Commun.* **153**, 445 ('03)

Reactive Force-Field (ReaxFF) MD: Variable N-Charge Problem

- **Reactive bond order potential energy:** $E_{\text{bond}}(\{r_{ij}\},\{r_{ijk}\},\{r_{ijkl}\},\{BO_{ij}\})$
  → **Bond breakage & formation**

- **Charge-equilibration (QEq)**
  → **Charge transfer**
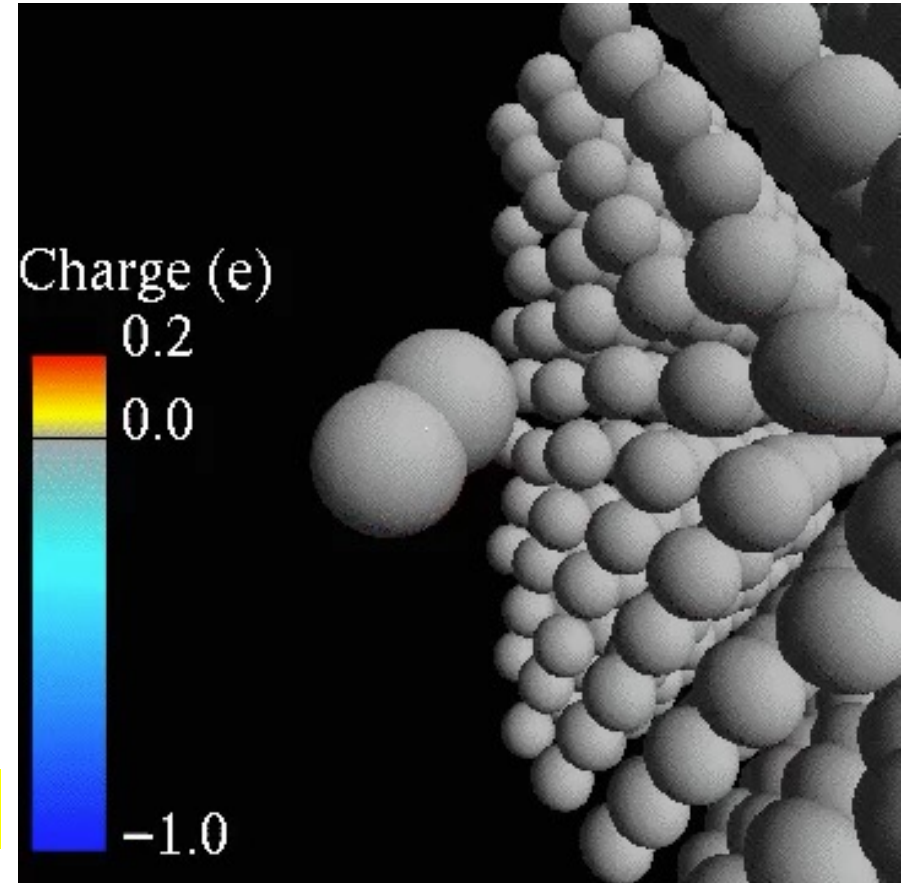 Determine atomic charges
  $\{q_i \mid i = 1, ..., N\}$ every MD step
  to minimize $E_{\text{ES}}(\mathbf{r}^N, q^N)$ with
  charge-neutrality constraint:
  $\sum_i q_i = 0$
  — Dense linear system: $M\,q = -\chi$
  $O(N^3)$!

$$E_{\text{ES}}(\mathbf{r}^N, q^N) = \sum_i \left(\chi_i q_i + \frac{1}{2} J_i q_i^2 \right) + \sum_{i<j} \int d\mathbf{x} \int d\mathbf{x}' \frac{\rho_i(q_i; \mathbf{x} - \mathbf{r}_i)\rho_j(q_j; \mathbf{x}' - \mathbf{r}_j)}{|\mathbf{x} - \mathbf{x}'|}$$

Fast Reactive Force-Field Algorithm

- $O(N)$ **fast reactive force-field (F-ReaxFF) algorithm**
  1) **Fast multipole method**
  2) **Temporal locality,** $q_i^{(init)}(t+\Delta t) = q_i(t)$

$$M\,q = -\chi$$

$$V_{es}(\mathbf{x}_i) = \sum_{j=1}^{N} \frac{q_j}{|\mathbf{x}_i - \mathbf{x}_j|}$$

- **Multilevel preconditioned conjugate gradient (MPCG) method**
  1) **Split Coulomb matrix:** $M = M_{near} + M_{far}$
  2) **Sparse near-field preconditioner:** $M_{near}^{-1} M\,q = -M_{near}^{-1}\chi$
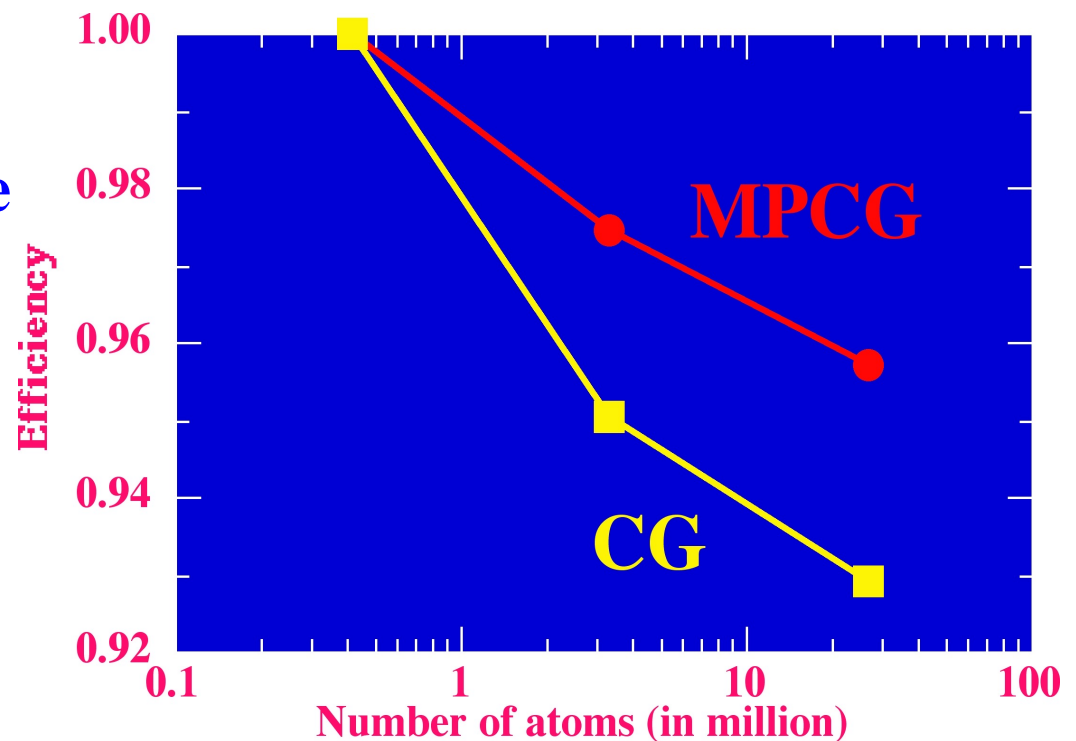
Results:

- **20% speed up of convergence**

- **Enhanced data locality: Improved parallel efficiency 0.93 → 0.96 for 26.5M-atom Al_2O_3 on 64 Power nodes**

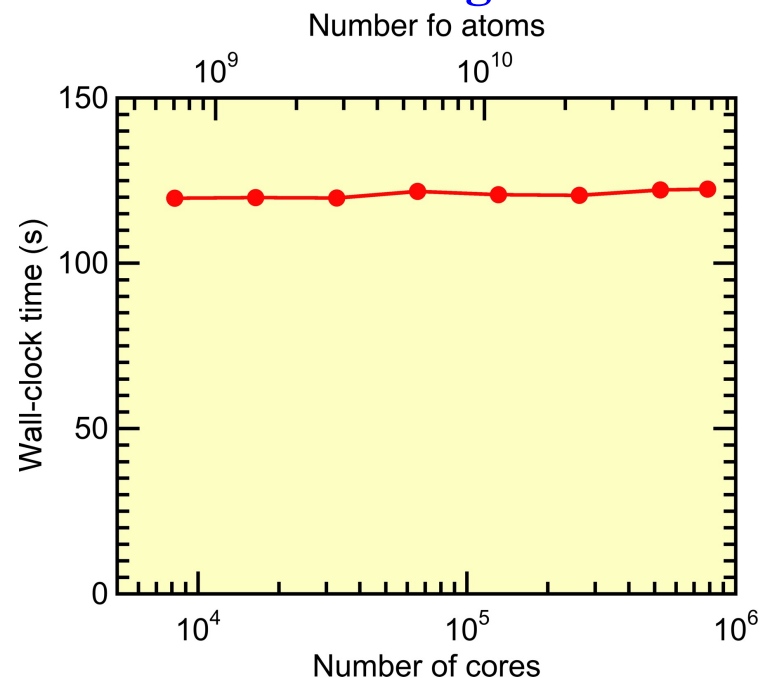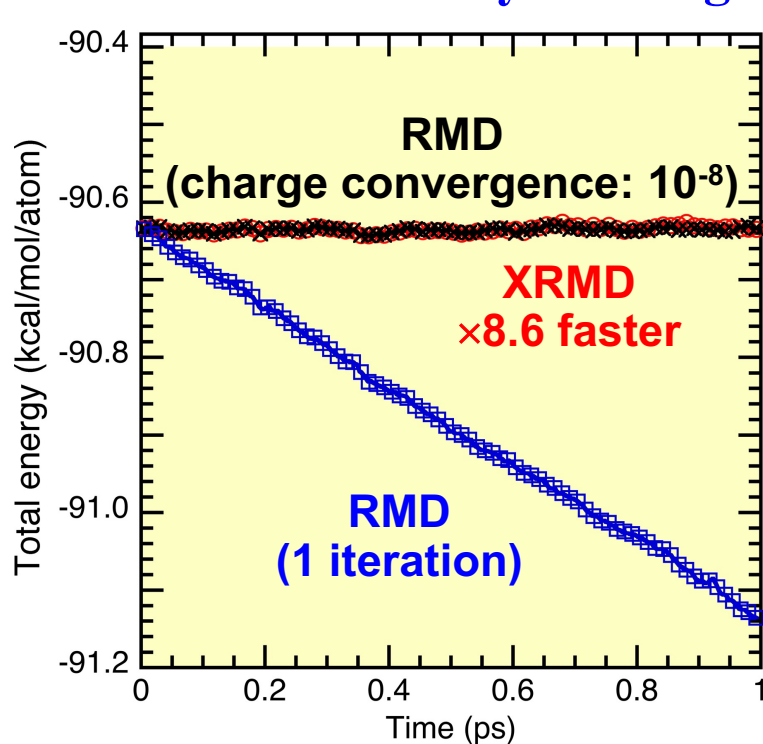A. Nakano, *Comput. Phys. Commun.* **104**, 59 ('97)

Extended-Lagrangian RMD (XRMD)

- **Eliminated speed-limiting iteration for charge-equilibration in reactive molecular dynamics (RMD) by adapting an extended-Lagrangian scheme proposed for QMD** [P. Souvatzis & A. Niklasson, *J. Chem. Phys.* **140**, 044117 ('14)]

$$L_{\mathrm{XRMD}} = L_{\mathrm{RMD}} + \frac{\mu}{2}\Sigma_i \dot{\theta}_i^2 - \frac{\mu\omega^2}{2}\Sigma_i(\theta_i - q_i)^2$$

Auxiliary charge: dynamic variable

Physical charge

- **Extended-Lagrangian RMD (XRMD) achieves the same energy conservation as fully converged RMD but an order-of-magnitude faster**

Nomura *et al.*, *Comput. Phys. Commun.* **192**, 91 ('15)

- **Parallel efficiency 0.977 on 786,432 Blue Gene/Q cores for 67.6 billion atoms**

Quantum Molecular Dynamics (QMD)

$$M_I \frac{d^2}{dt^2} \mathbf{R}_I = -\frac{\partial}{\partial \mathbf{R}_I} E[\{\mathbf{R}_I\}, \psi(\mathbf{r}_1 \ldots, \mathbf{r}_N)] \ (I = 1, \ldots, N_{\mathrm{atom}})$$

First molecular dynamics using an empirical interatomic interaction

A. Rahman, *Phys. Rev.* **136**, A405 ('64)

$$\psi(\mathbf{r}_1 \ldots, \mathbf{r}_N) \leftarrow \mathrm{argmin} \ E[\{\mathbf{R}_I\}, \psi(\mathbf{r}_1 \ldots, \mathbf{r}_N)]$$

Density functional theory (DFT)

Hohenberg & Kohn, *Phys. Rev.* **136**, B864 ('64)

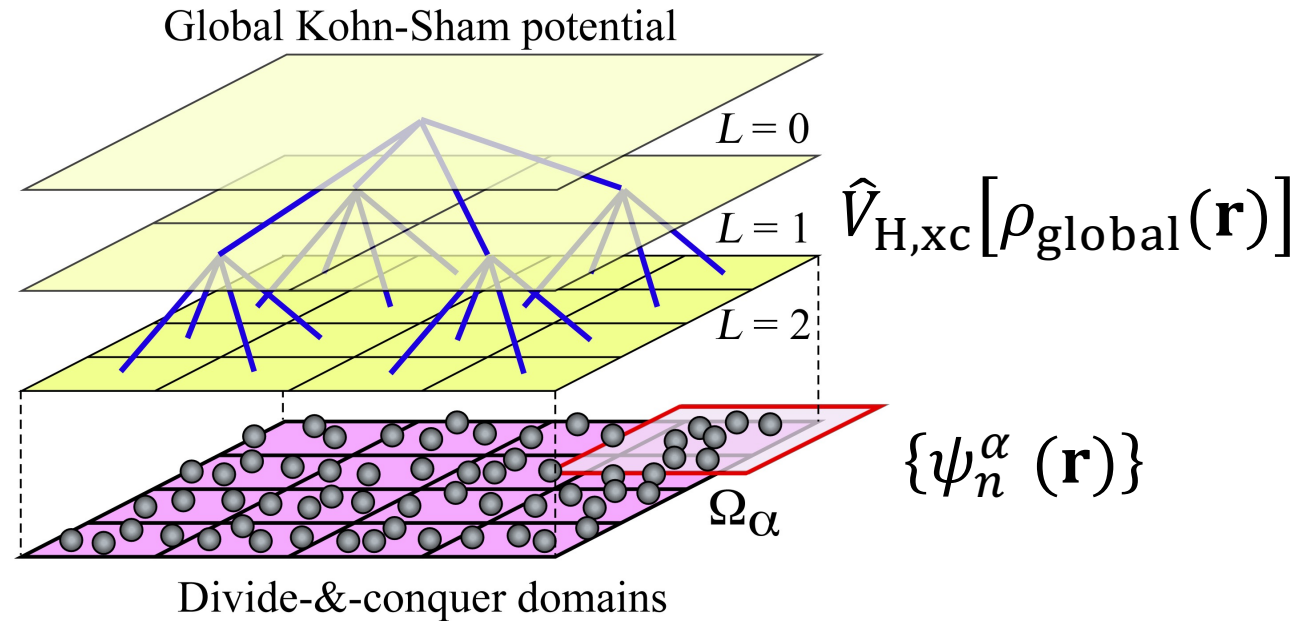W. Kohn, *Nobel chemistry prize*, '98

$$O(C^N) \qquad \rightarrow \qquad O(N^3)$$

1 N-electron problem   **N 1-electron problems**

intractable   **tractable**

$$\psi(\mathbf{r}_1 \ldots, \mathbf{r}_N) \qquad \{\psi_i(\mathbf{r}) | i = 1, \ldots, N\}$$

$O(N)$ DFT algorithms

- **Divide-&-conquer DFT** [W. Yang, *Phys. Rev. Lett.* **66**, 1438 ('91); F. Shimojo *et al.*, *Comput. Phys. Commun.* **167**, 151 ('05); *Phys Rev. B* **77**, 085103 ('08); *Appl. Phys. Lett.* **95**, 043114 ('09); *J. Chem. Phys.* **140**, 18A529 ('14)]

- **Quantum nearsightedness principle** [W. Kohn, *Phys. Rev. Lett.* **76**, 3168 ('96)]

- **A recent review** [Bowler & Miyazaki, *Rep. Prog. Phys.* **75**, 036503 ('12)]

Divide-&-Conquer Density Functional Theory

Global Kohn-Sham potential

$$\hat{V}_{\mathrm{H,xc}}\big[\rho_{\mathrm{global}}(\mathbf{r})\big]$$

$$\{\psi_n^\alpha(\mathbf{r})\}$$

$$\Omega_\alpha$$

Divide-&-conquer domains

- **Overlapping spatial domains:** $\Omega = \cup_\alpha \Omega_\alpha$
- **Domain Kohn-Sham equations**

Global-local self-consistent field (SCF) iteration

$$\left(-\frac{1}{2}\nabla^2 + \hat{V}_{\mathrm{ion}} + \hat{V}_{\mathrm{H,xc}}\big[\rho_{\mathrm{global}}(\mathbf{r})\big]\right)\psi_n^\alpha(\mathbf{r}) = \epsilon_n^\alpha \psi_n^\alpha(\mathbf{r})$$
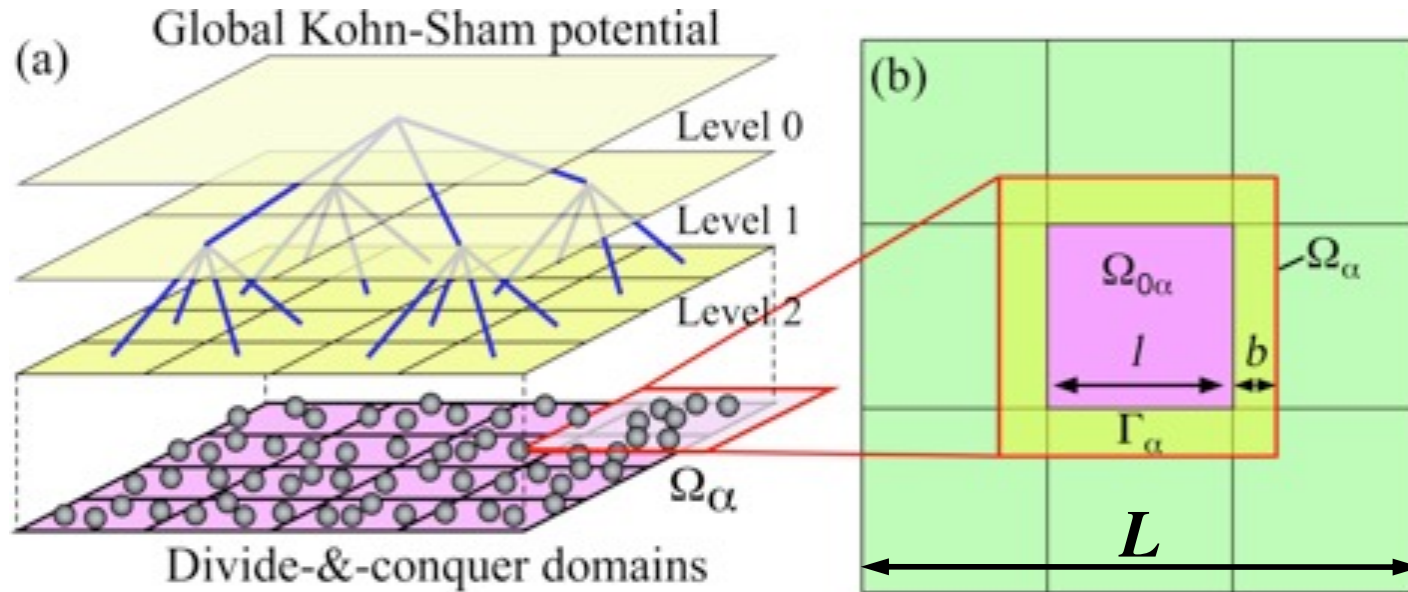
- **Global & domain electron densities**

$$\rho_{\mathrm{global}}(\mathbf{r}) = \Sigma_\alpha\, p_\alpha(\mathbf{r})\rho_\alpha(\mathbf{r}) \quad\longleftarrow\quad \rho_\alpha(\mathbf{r}) = \Sigma_n \lfloor\psi_n^\alpha\rfloor^2\, \Theta(\mu - \epsilon_n^\alpha)$$

Domain support function

$$\Sigma_\alpha\, p_\alpha(\mathbf{r}) = 1$$

Global chemical potential

$$N = \int d\mathbf{r}\, \rho_{\mathrm{global}}(\mathbf{r})$$

Optimization of Divide-&-Conquer DFT

- **Computational parameters of DC-DFT = domain size (l) + buffer thickness (b)**

(a) Global Kohn-Sham potential
Level 0
Level 1
Level 2
Ω_α
Divide-&-conquer domains

(b) $\Omega_{0\alpha}$ Ω_α l b Γ_α L

- **Complexity analysis to optimize the domain size l**

$$l_* = \mathrm{argmin}\left(T_{\mathrm{comp}}(l)\right) = \mathrm{argmin}\left(\left(\frac{L}{l}\right)^3 (l + 2b)^{3\nu}\right) = \frac{2b}{\nu - 1}$$

Per-domain computational complexity of DFT = $O(n^\nu)$: $\nu = 2$ or 3 ($n <$ or $> 10^3$)

- **Error analysis: Buffer thickness b is dictated by the accuracy requirement**

$$b = \lambda \ln\left(\max\{|\Delta\rho_\alpha(\mathbf{r})||\mathbf{r} \in \partial\Omega_\alpha\}\right)/\varepsilon\langle\rho_\alpha(\mathbf{r})\rangle$$

Decay length $\rho_\alpha(\mathbf{r}) - \rho_{\mathrm{global}}(\mathbf{r})$ Error tolerance
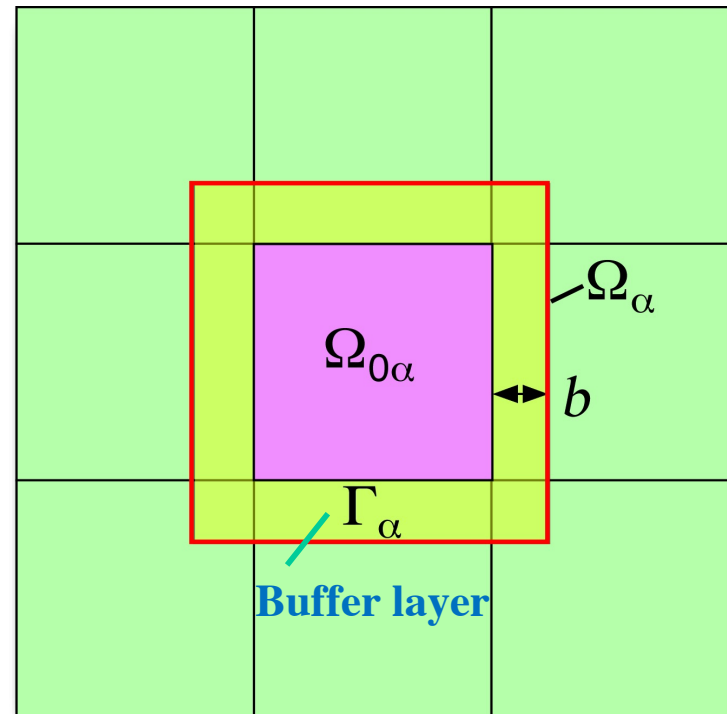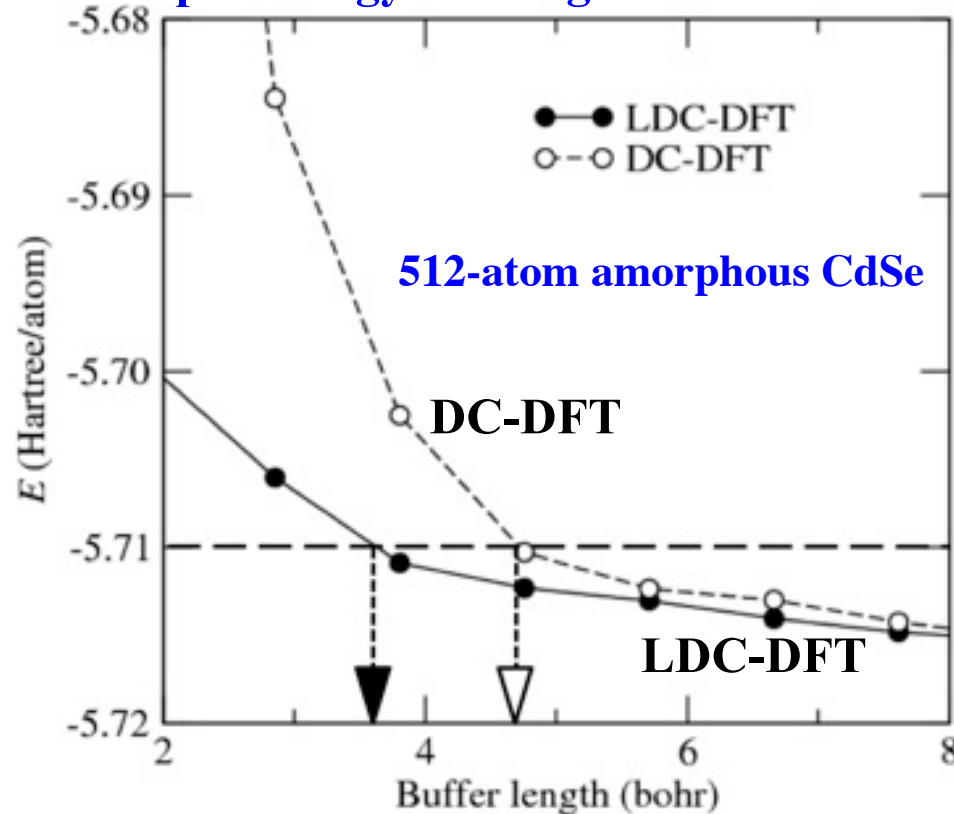
cf. quantum nearsightedness [Kohn, *Phys. Rev. Lett.* **76**, 3168 ('96)]

Lean Divide-&-Conquer (LDC) DFT

- **Density-adaptive boundary potential to reduce the $O(N)$ prefactor**

$$v_\alpha^{\mathrm{bc}}(\mathbf{r}) = \int d\mathbf{r}' \frac{\partial v(\mathbf{r})}{\partial \rho(\mathbf{r}')} \left(\rho_\alpha(\mathbf{r}) - \rho_{\mathrm{global}}(\mathbf{r}) \right) \cong \frac{\rho_\alpha(\mathbf{r}) - \rho_{\mathrm{global}}(\mathbf{r})}{\xi}$$

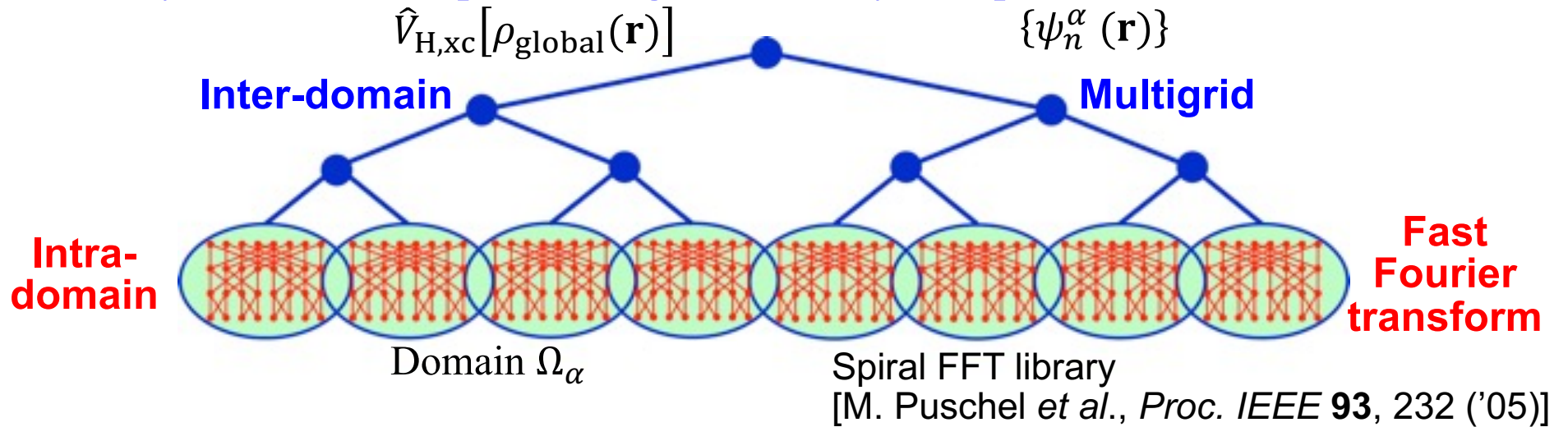- **More rapid energy convergence of LDC-DFT compared with nonadaptive DC-DFT**

- **Factor 2.03 (for $\nu = 2$) ~ 2.89 (for $\nu = 3$) reduction of the computational cost with an error tolerance of 5×10^{-3} a.u. (per-domain complexity: n^ν)**
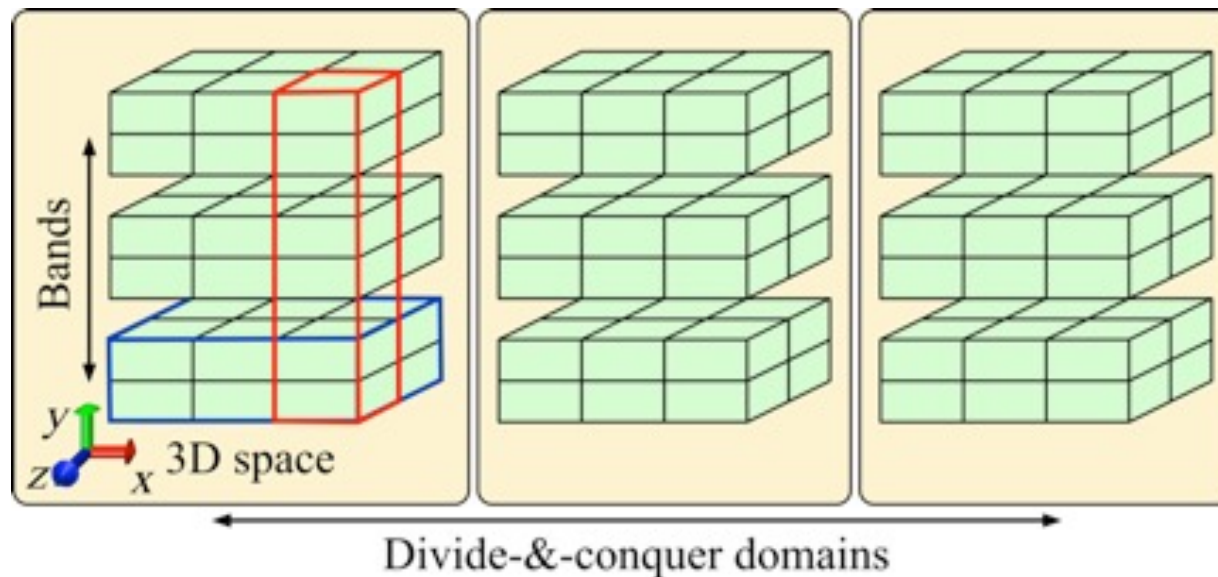
F. Shimojo *et al.*, *J. Chem. Phys.* **140**, 18A529 ('14);
Phys. Rev. B **77**, 085103 ('08); *Comput. Phys. Commun.* **167**, 151 ('05)

Hierarchical Computing

- **Globally scalable (real-space multigrid) + locally fast (plane wave) electronic solver**

$$\hat{V}_{\mathrm{H,xc}}[\rho_{\mathrm{global}}(\mathbf{r})] \qquad\qquad \{\psi_n^{\alpha}(\mathbf{r})\}$$

Inter-domain

Multigrid

Intra-domain

Fast Fourier transform

Domain Ω_{α}

Spiral FFT library
[M. Puschel *et al.*, *Proc. IEEE* **93**, 232 ('05)]

- **Hierarchical band (*i.e.*, Kohn-Sham orbital) + space + domain (BSD) decomposition**

Bands

3D space

Divide-&-conquer domains

Floating Point Performance

- **Transform from band-by-band to all-band computations to utilize a matrix-matrix subroutine (DGEMM) in the level 3 basic linear algebra subprograms (BLAS3) library**

- **Algebraic transformation of computations**

Example: Nonlocal pseudopotential operation
D. Vanderbilt, *Phys. Rev. B* **41**, 7892 ('90)

$$\hat{v}_{\text{nl}}|\psi_n^\alpha\rangle = \sum_I^{N_{\text{atom}}} \sum_{ij}^{L_{\text{max}}} |\beta_{i,I}\rangle D_{ij,I} \langle\beta_{j,I}|\psi_n^\alpha\rangle \quad (n = 1, \dots, N_{\text{band}})$$

$$\Psi = \left[|\psi_1^\alpha\rangle, \dots, |\psi_{N_{\text{band}}}^\alpha\rangle\right] \quad \widetilde{\mathbf{B}}(i) = \left[|\beta_{i,1}\rangle, \dots, |\beta_{i,N_{\text{atom}}}\rangle\right] \quad \left[\widetilde{\mathbf{D}}(i,j)\right]_{I,J} = D_{ij,I}\delta_{IJ}$$
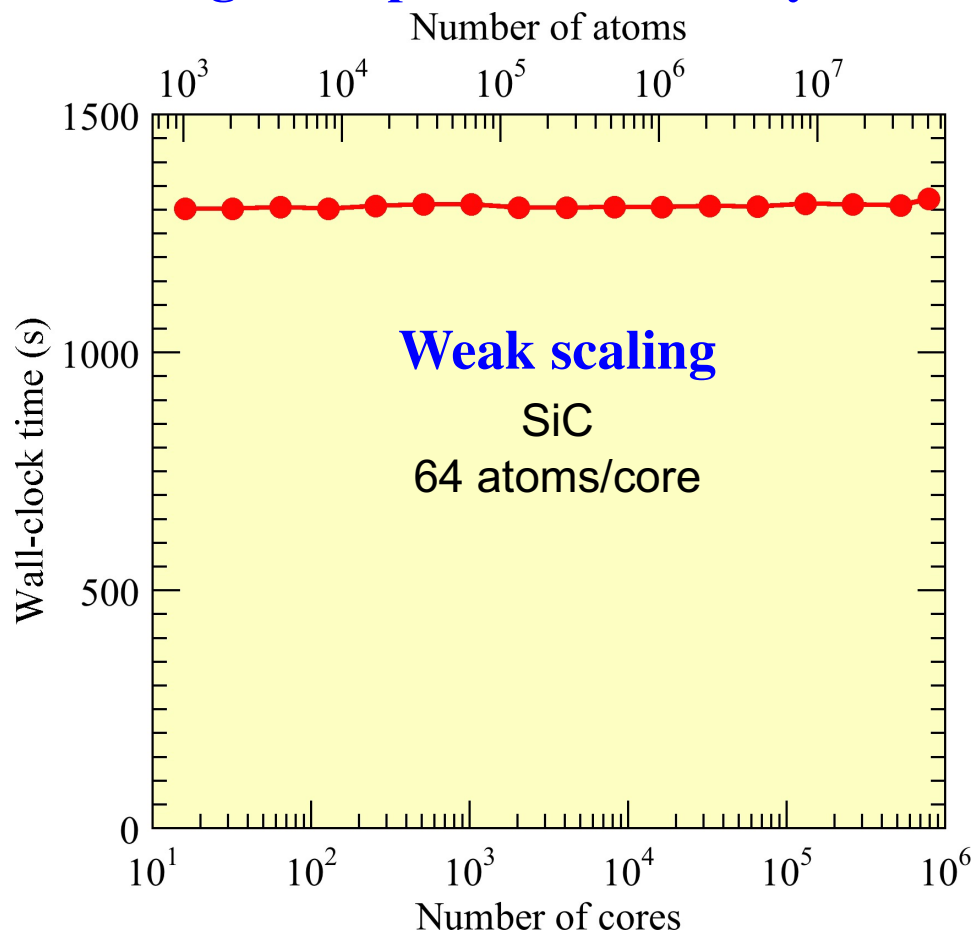
$$\hat{v}_{\text{nl}}\Psi = \sum_{i,j}^{L} \widetilde{\mathbf{B}}(i)\widetilde{\mathbf{D}}(i,j)\widetilde{\mathbf{B}}(j)^T$$

- **50.5% of the theoretical peak FLOP/s performance on 786,432 Blue Gene/Q cores (entire Mira at the Argonne Leadership Computing Facility)**

- **55% of the theoretical peak FLOP/s on Intel Xeon E5-2665**

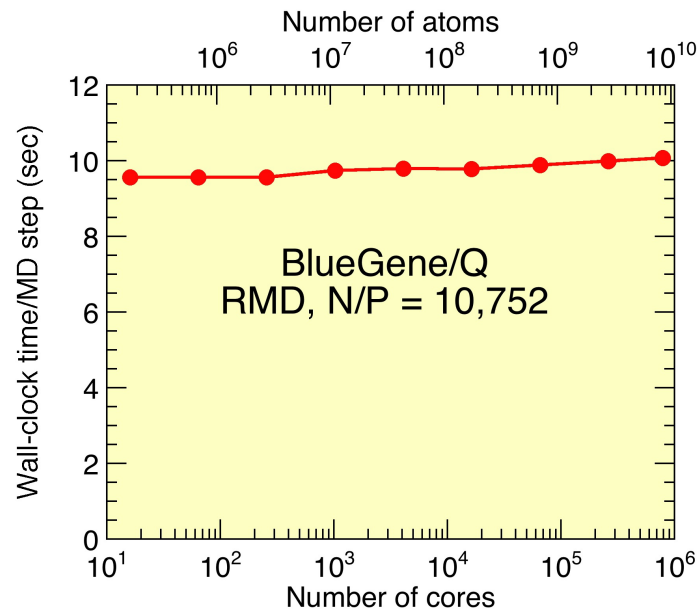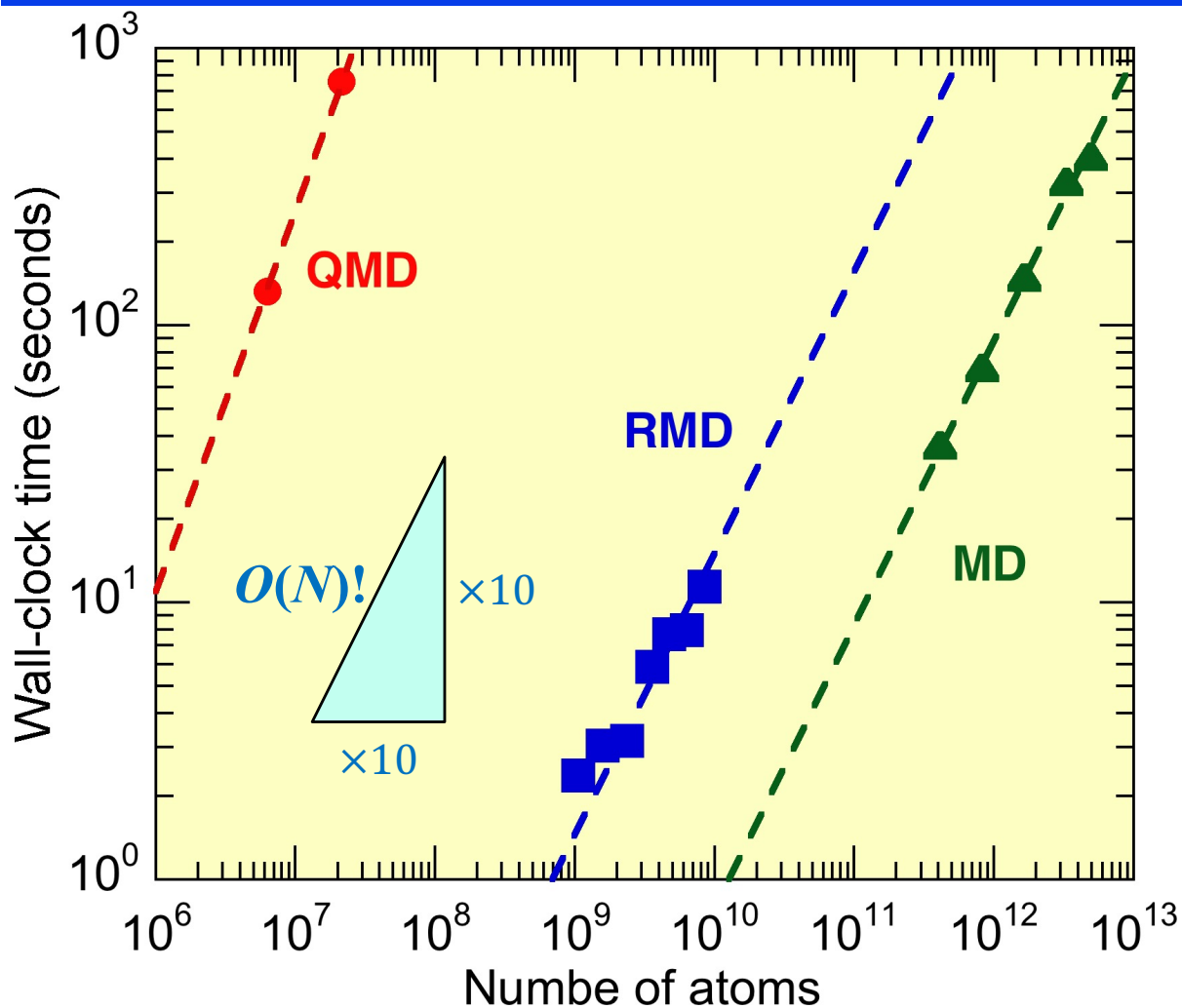K. Nomura *et al.*, *IEEE/ACM Supercomputing, SC14* ('14)

Parallel Performance

- **Weak-scaling parallel efficiency is 0.984 on 786,432 Blue Gene/Q cores for a 50,331,648-atom SiC system**

- **Strong-scale parallel efficiency is 0.803 on 786,432 Blue Gene/Q cores**

- **62-fold reduction of time-to-solution** [441 s/SCF-step for 50.3M atoms] **from the previous state-of-the-art** [55 s/SCF-step for 102K atoms, Osei-Kuffuor *et al.*, *PRL* '14]

K. Nomura *et al.*, *IEEE/ACM Supercomputing, SC14* ('14)

Scalable Simulation Algorithm Suite

QMD (quantum molecular dynamics): DC-DFT

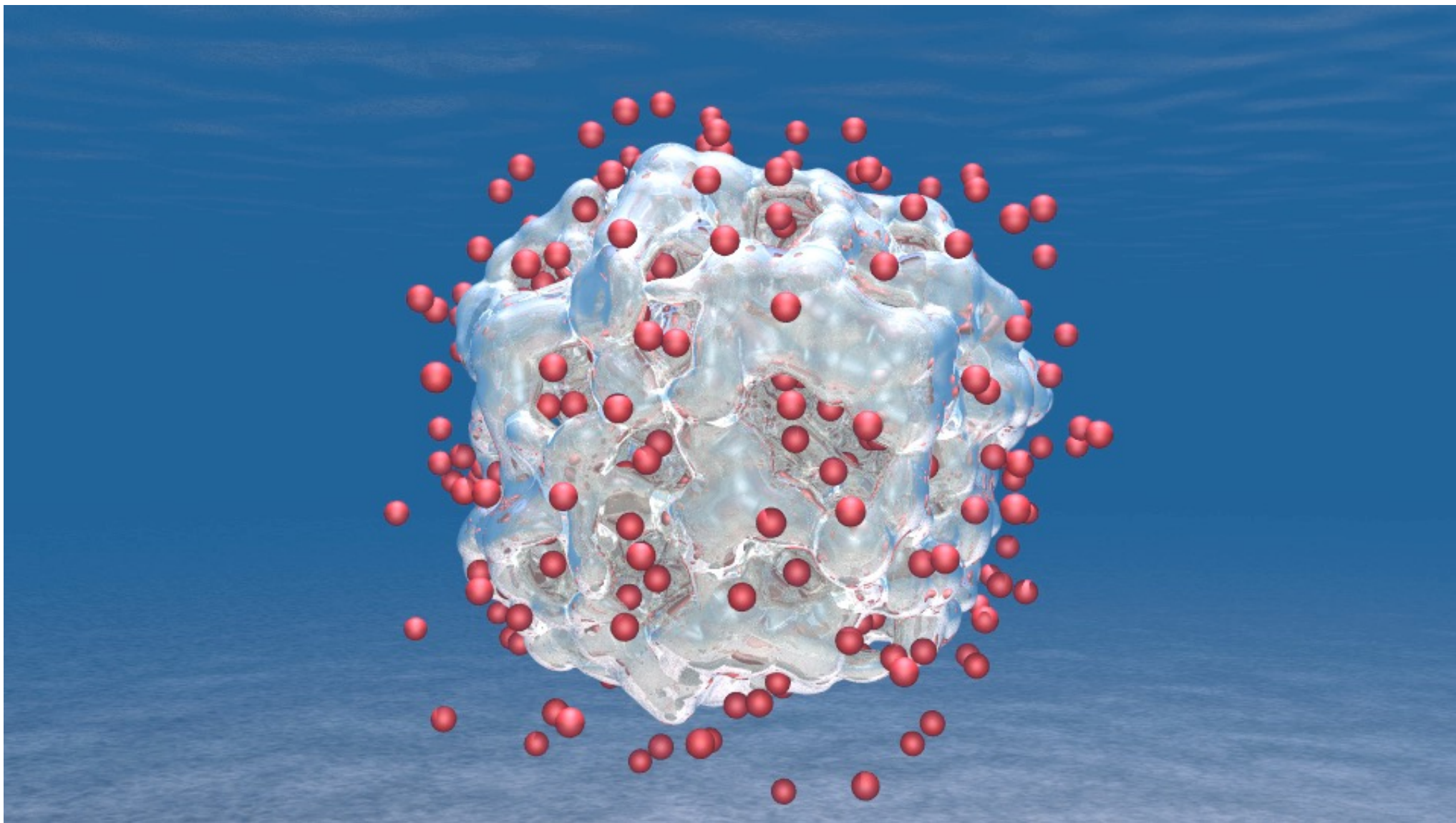**RMD (reactive molecular dynamics): F-ReaxFF**

MD (molecular dynamics): MRMD

- **4.9 trillion-atom space-time multiresolution MD (MRMD) of SiO$_2$**
- **8.5 billion-atom fast reactive force-field (F-ReaxFF) RMD of RDX**
- **1.9 trillion grid points (21.2 million-atom) DC-DFT QMD of SiC**
  parallel efficiency 0.98 on 786,432 BlueGene/Q cores

H$_2$ Production from Water Using LiAl Particles

16,661-atom QMD simulation of Li$_{441}$Al$_{441}$ in water on 786,432 IBM Blue Gene/Q cores

21,140 time steps (129,208 self-consistent-field iterations)

Divide-Conquer-Recombine Algorithms

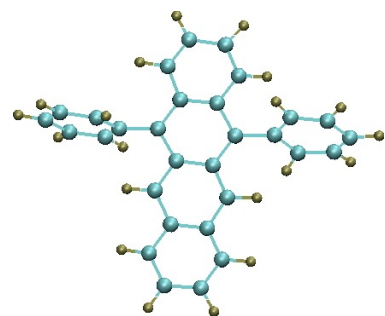M. Kunaseth *et al.*, *ACM/IEEE SC13* ('13)

Globally informed local DC-DFT solutions are used in the recombine phase as compact bases to synthesize global properties in broad applications:

- **High-order inter-molecular-fragment correlation** [S. Tanaka *et al.*, '13]

- **Global frontier orbitals (HOMO & LUMO)** [S. Tsuneyuki *et al.*, '09, '13]

- **Global charge migration** [H. Kitoh-Nishioka *et al.*, '12; C. Gollub *et al.*, '12]

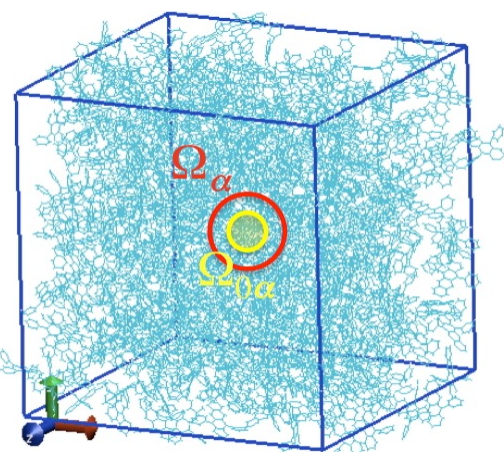- **Global exciton dynamics** [W. Mou *et al.*, '13]

F. Shimojo *et al.*, *J. Chem. Phys.* **140**, 18A529 ('14); K. Nomura *et al.*, *IEEE/ACM SC14* ('14)
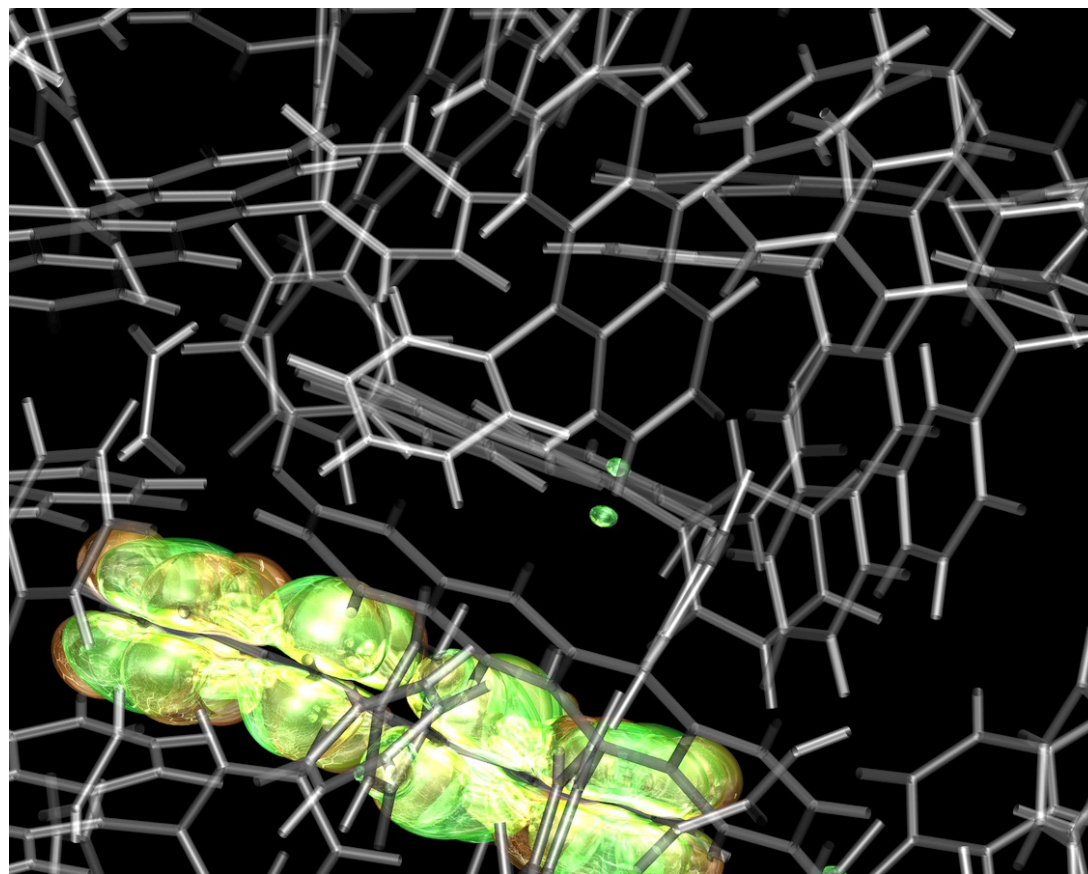
Singlet Fission in Amorphous DPT

- **Photo-current doubling by splitting a singlet exciton into 2 triplet excitons**
- **Singlet fission in mass-produced disordered organic solid → efficient low-cost solar cells**
- **Exp'l breakthrough: SF found in amorphous diphenyl tetracene (DPT)**

DPT molecule

Amorphous DPT

Quasi-electron

Quasi-hole

- **Divide-conquer-recombine nonadiabatic QMD (phonon-assisted exciton dynamics) + time-dependent perturbation theory (singlet-fission rate) + kinetic Monte Carlo calculations of exciton population dynamics in 6,400-atom amorphous DPT**