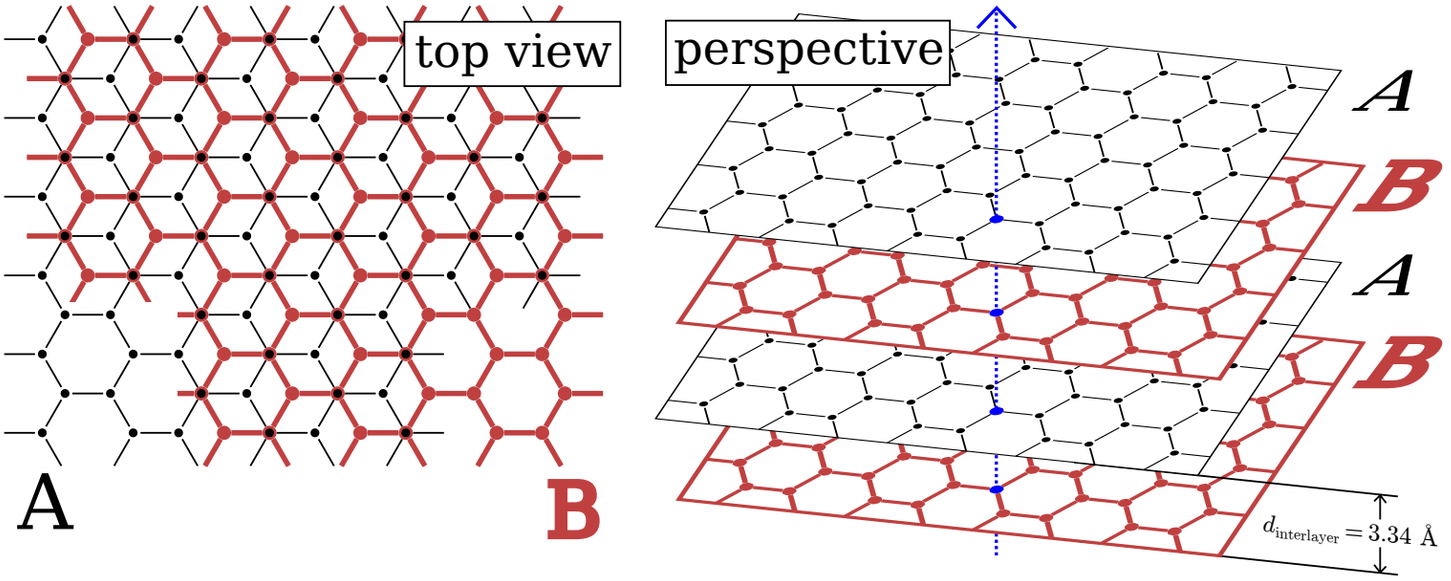
Diffusion Monte Carlo: Exponential Scaling of Computational Cost for Large Systems

→ arXiv:0906.0501 ←

Norbert Nemec

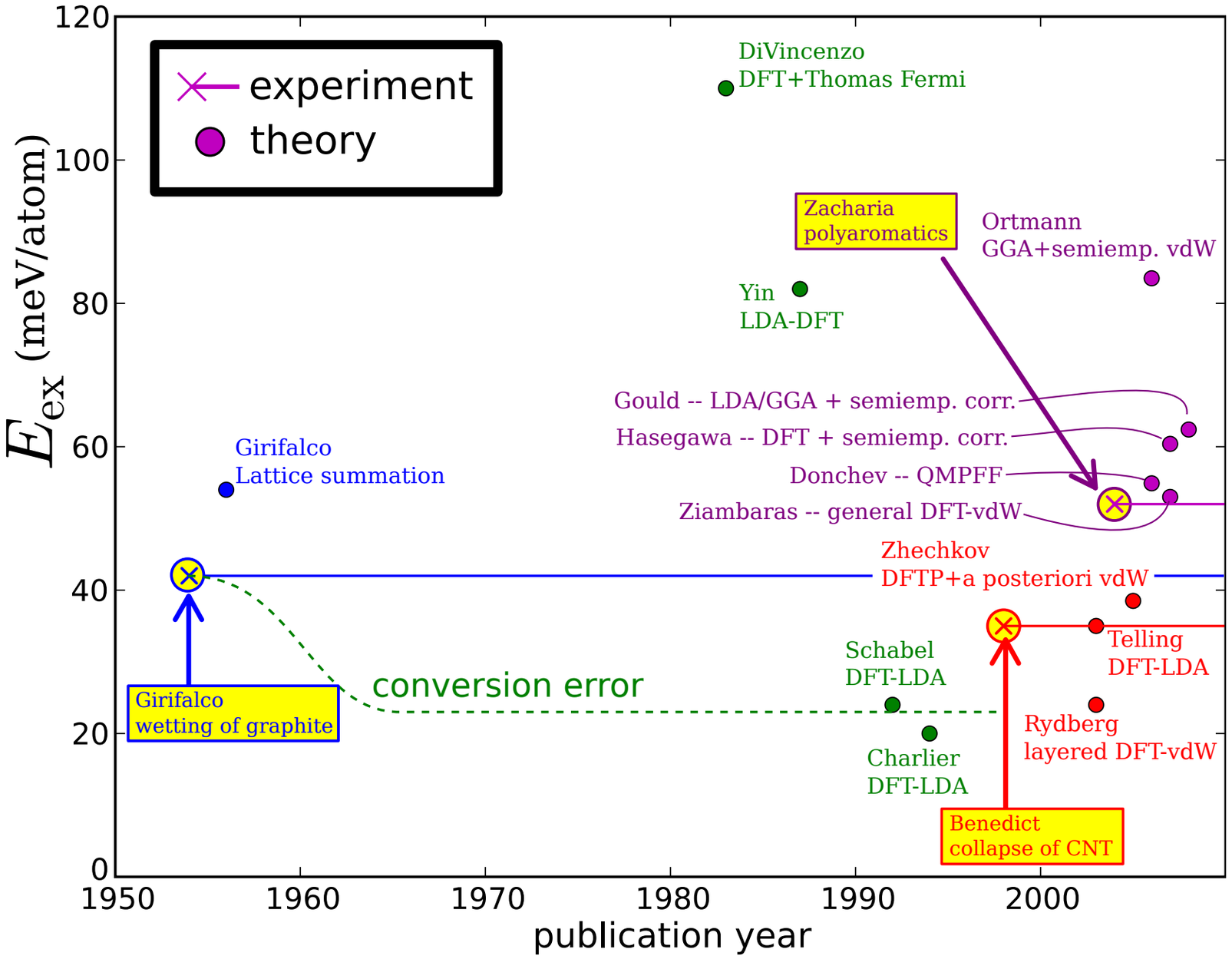
QMC in the Apuan Alps
Vallico Sotto, 30th July 2009

Graphite



intralayer bond length:	$d_{CC} = 1.4196\,\text{Å}$
atomization energy	$E_{\text{at}} = 7.374\,\text{eV/atom}$
intralayer isotrop. elastic constant:	$C_{11} + C_{22} = 1240\,\text{GPa}$
interlayer distance:	$d_{\text{interlayer}} = 3.34\,\text{Å}$
exfoliation energy:	$E_{\text{ex}} = 35 \ldots 52\,\text{meV/atom}$
interlayer elastic constant:	$C_{33} = 36.5\,\text{GPa}$

Exfoliation energy of graphite



Dispersion forces for thin layers

TABLE I. Asymptotic vdW energy of parallel structures. K and D_0 are constants.

System	Present	Standard
1D metals[a]	$-\mathbf{D^{-2}(\ln(KD))^{-3/2}}$	$-D^{-5}$
1D insulators [9]	$-D^{-5}$	$-D^{-5}$
2D metals [10,11]	$-\mathbf{D^{-5/2}}$	$-D^{-4}$
π-conjugated layers[a]	$-\mathbf{D^{-3}}$	$-D^{-4}$
1 metallic, 1 π layer[a]	$-\mathbf{D^{-3}\ln(D/D_0)}$	$-D^{-4}$
2D insulators [6]	$-D^{-4}$	$-D^{-4}$
Thick metals or ins. [11]	$-D^{-2}$	$-D^{-2}$

[a]* Denotes new derivations given here.

[from Dobson, White and Rubio, Phys. Rev. Lett. **96**, 073201 (2006)]

Diffusion quantum Monte Carlo (DMC)

N-particle Schrödinger equation:

$$i\partial_t \Psi(\boldsymbol{r}) = -\frac{1}{2}\nabla^2\Psi(\boldsymbol{r}) + V(\boldsymbol{r})\Psi(\boldsymbol{r})$$

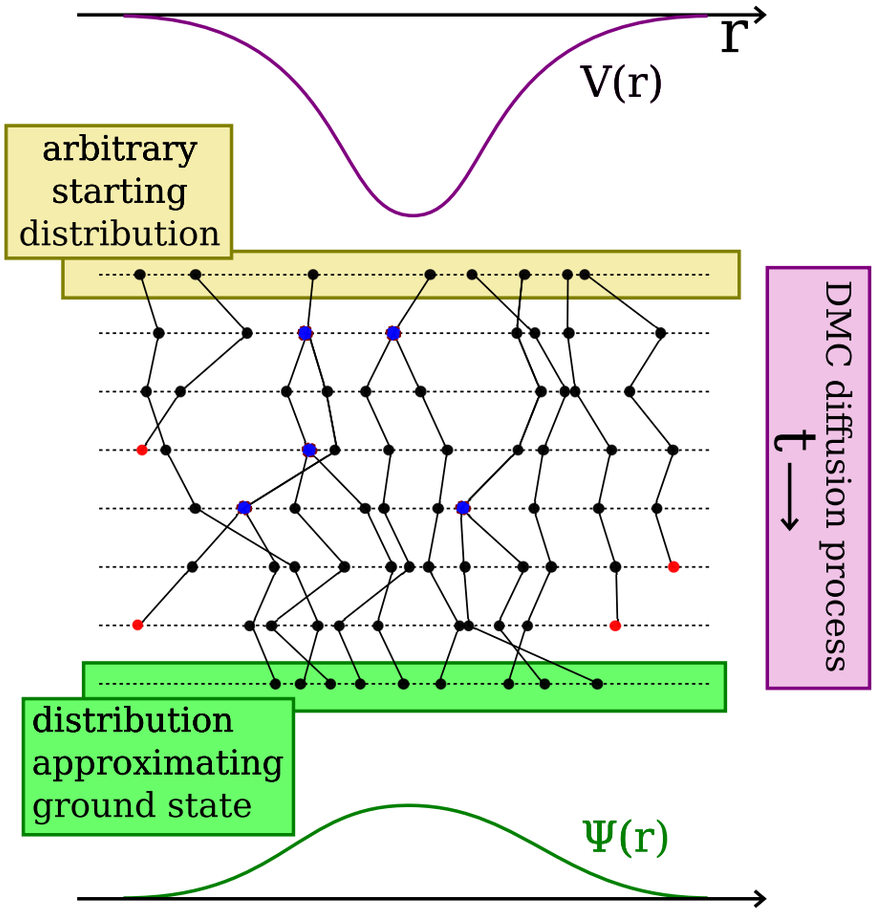
with $\boldsymbol{r} = (\boldsymbol{r}_1, \boldsymbol{r}_2 \ldots \boldsymbol{r}_N)$.

As diffusion equation in imaginary time:

$$\partial_t \Psi(\boldsymbol{r}) = \underbrace{\frac{1}{2}\nabla^2\Psi(\boldsymbol{r})}_{\text{diffusion-term}} - \underbrace{V(\boldsymbol{r})\Psi(\boldsymbol{r})}_{\substack{\text{source-term} \\ (\text{branching}/\text{killing})}}$$

Compute (bosonic!) ground state energy:

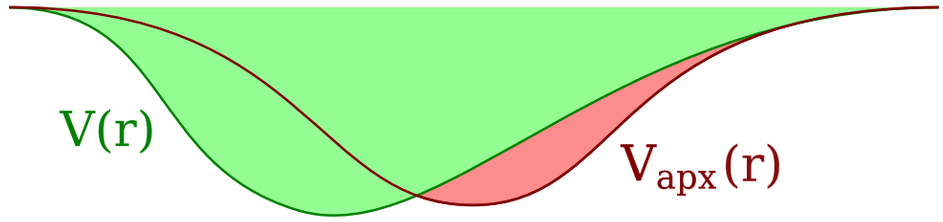
$$E_0 = \langle \Psi_{\text{DMC}} | \mathcal{H} | \Psi_{\text{apx}} \rangle \approx \sum_{\boldsymbol{r} \in \{\text{cfg}\}} \mathcal{H}\Psi_{\text{apx}}(\boldsymbol{r})$$



Fermionic DMC (Fixed node approximation)

approximate ("trial") wave function Ψ_{apx} (e.g. from DFT)
defines nodal surface through modified **drift-diffusion equation**:

$$\partial_t f(\boldsymbol{r}) = \underbrace{\frac{1}{2}\nabla^2 f(\boldsymbol{r})}_{\mathrm{diffusion-term}} - \underbrace{\nabla\cdot\left(\frac{\nabla\psi_{\mathrm{apx}}}{\psi_{\mathrm{apx}}}\right)f(\boldsymbol{r})}_{\mathrm{drift-term}} + \underbrace{[E-E_{\mathrm{loc}}(\boldsymbol{r})]f(\boldsymbol{r})}_{\substack{\mathrm{source-term}\\(\mathrm{branching}/\mathrm{killing})}}$$

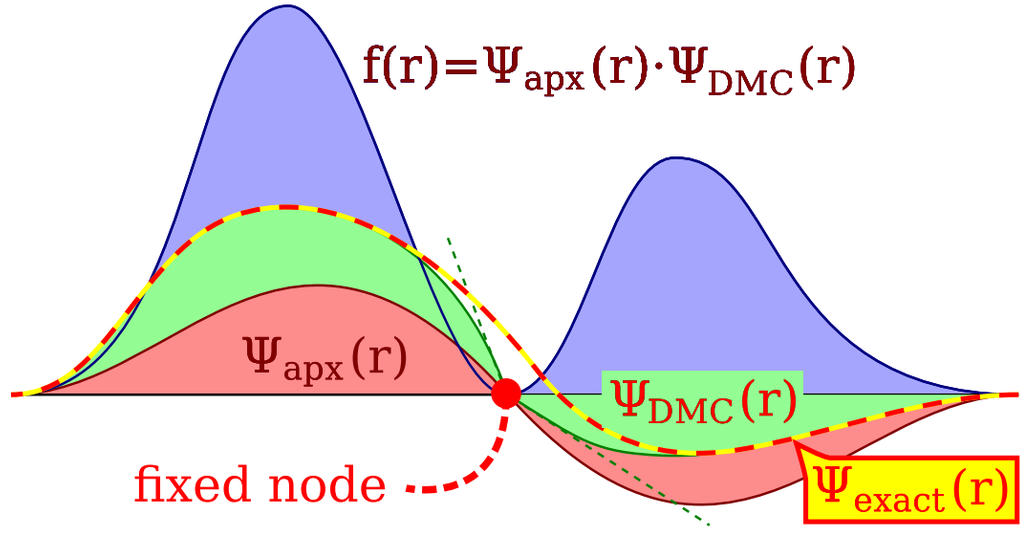


Mixed distribution:

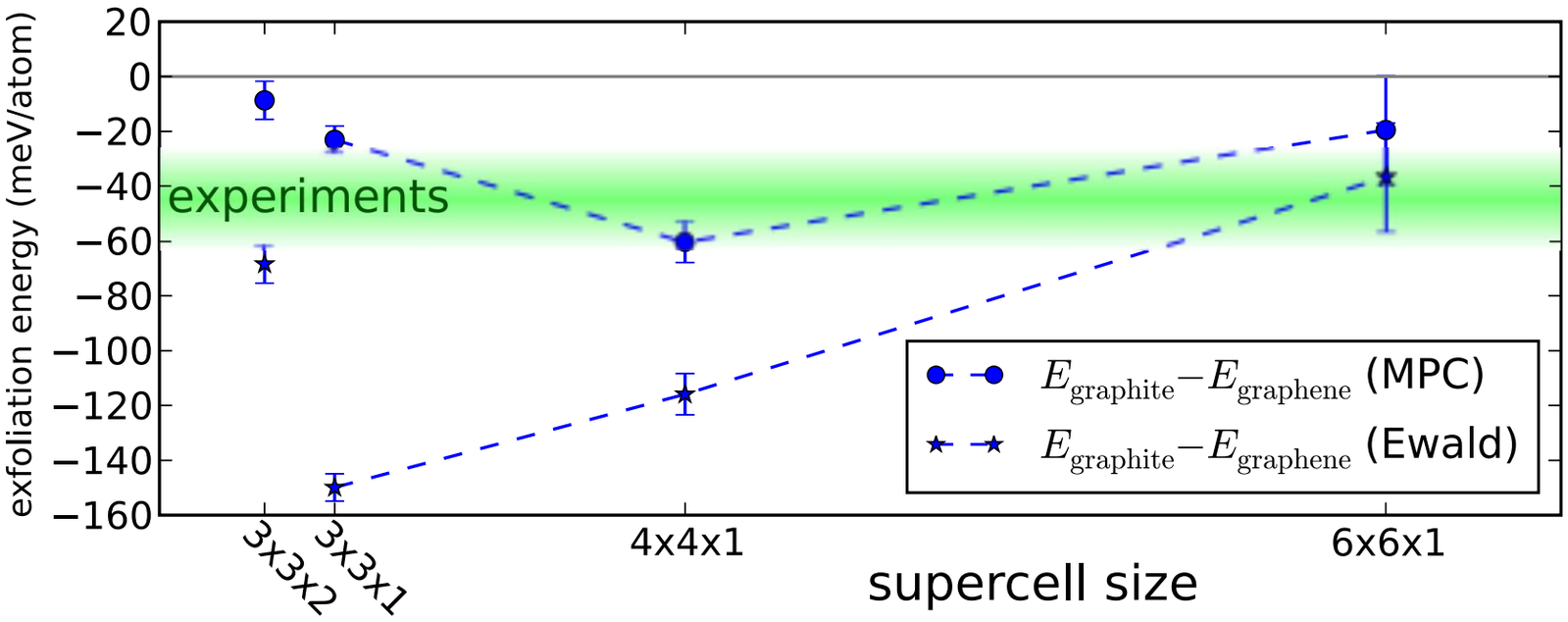
$$f(\boldsymbol{r}) = \Psi_{\mathrm{apx}}(\boldsymbol{r})\Psi_{\mathrm{DMC}}(\boldsymbol{r})$$

Local energy:

$$E_{\mathrm{loc}} = -\frac{\nabla^2\Psi_{\mathrm{apx}}(\boldsymbol{r})}{\Psi_{\mathrm{apx}}(\boldsymbol{r})} + V(\boldsymbol{r})$$



Results: finite size convergence



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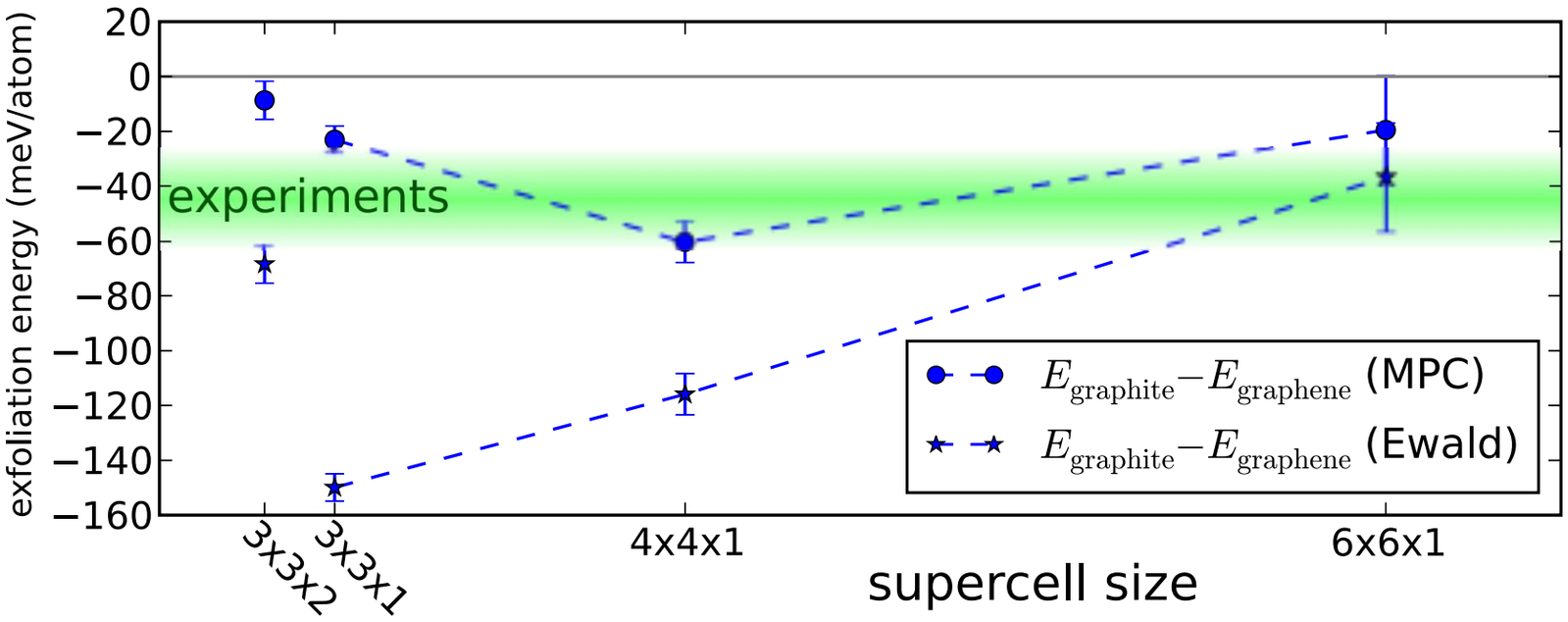
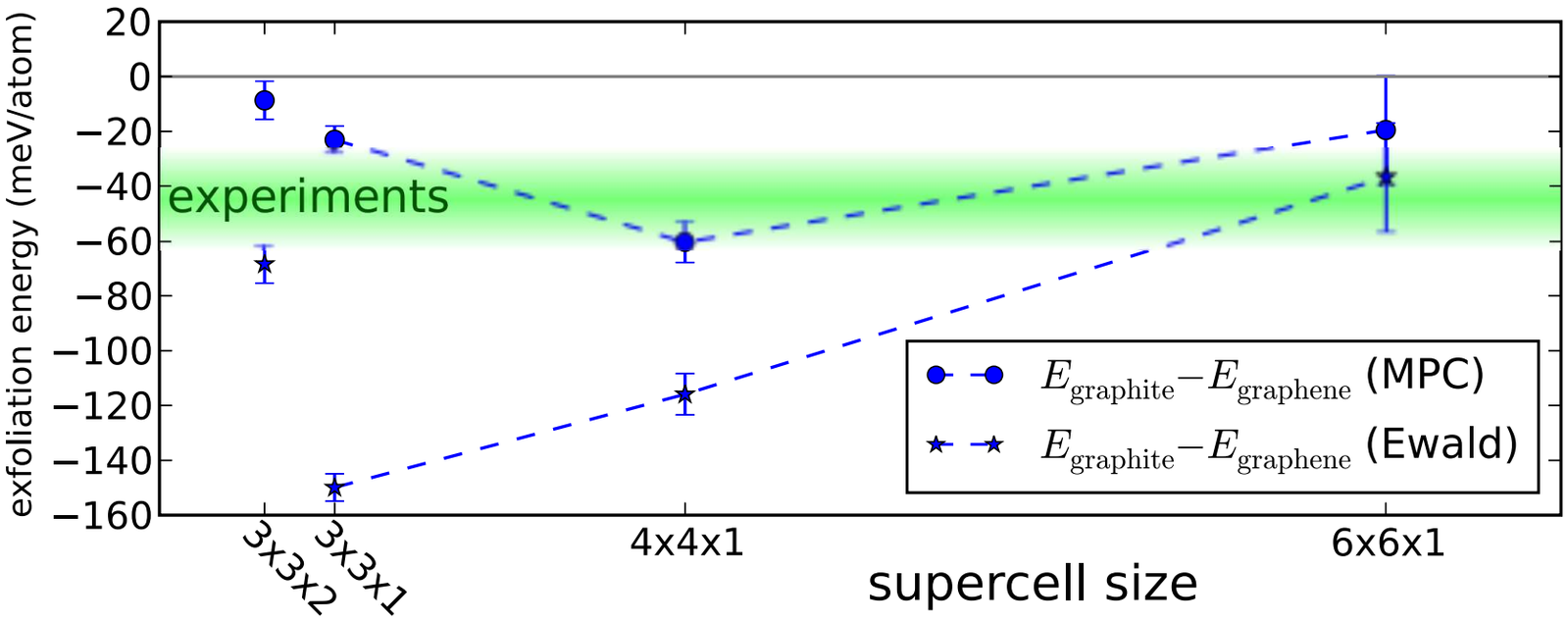


CPU times:

graphene	3x3 cell	(72 electrons):	500 CPU hours	$\rightarrow \pm 3.1\,\mathrm{meV/atom}$
graphite	3x3x1 cell	(144 electrons):	1000 CPU hours	$\rightarrow \pm 2.3\,\mathrm{meV/atom}$
graphite	3x3x2 cell	(288 electrons):	4000 CPU hours	$\rightarrow \pm 4.4\,\mathrm{meV/atom}$
graphite	6x6x1 cell	(576 electrons):	4000 CPU hours	$\rightarrow \pm 9.9\,\mathrm{meV/atom}$

Results: finite size convergence



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... 10x10x1 (1600 electrons), $\pm 5.0\,\text{meV/atom} \rightarrow$ 120,000 CPU hours (??)

DMC algorithm

$$\partial_t f(\boldsymbol{r}) \;=\; \underbrace{\frac{1}{2}\nabla^2 f(\boldsymbol{r})}_{\text{diffusion-term}} - \underbrace{\nabla\cdot\left(\frac{\nabla\psi_{\text{apx}}}{\psi_{\text{apx}}}\right) f(\boldsymbol{r})}_{\text{drift-term}} + \underbrace{[E - E_{\text{loc}}(\boldsymbol{r})]\, f(\boldsymbol{r})}_{\substack{\text{source-term}\\ \text{(branching/killing)}}}$$

time step $\mathrm{d}\tau$ in imaginary time τ:

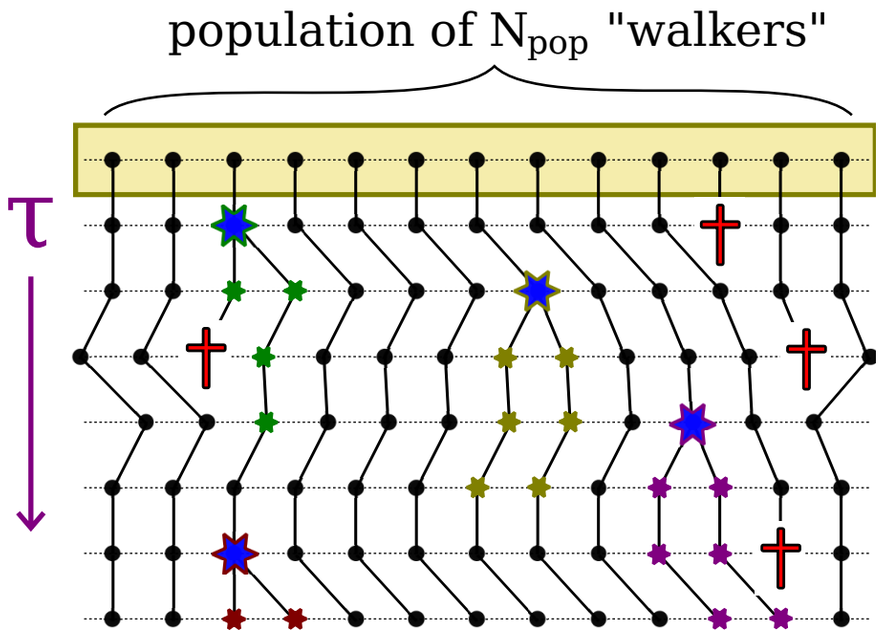
1. drift-diffusion step
2. "growth" of each walker

$$w^{(p)} \;=\; \exp\left(E_{\text{ref}} - E_{\text{loc}}^{(p)}\right)$$

3. integer multiplicity of walker:

$$\left\langle N_{\text{mult}}^{(p)} \right\rangle \;=\; w^{(p)}$$

4. $N_{\text{mult}}^{(p)} \geqslant 2$: clone walker (p) *
   $N_{\text{mult}}^{(p)} = 0$: kill walker (p) †
5. start over for next time step



$$\underbrace{\tau_{\text{branch}}^{-1}}_{\text{(branching rate)}} \times \underbrace{\tau_{\text{corr}}}_{\text{(correlation time)}}$$

$\Longrightarrow$ correlation within population

Computational cost of DMC

$$€_{\text{total}} = N_{\text{step}} \times N_{\text{pop}} \times €_{\text{step}}$$

N_{step} number of (imaginary) time steps

N_{pop} average population size

$€_{\text{step}}$ CPU time needed for one single step per configuration

$\rightarrow$ "Linear scaling" QMC: $€_{\text{step}} \propto N_{\text{atom}}$

[A. J. Williamson, R. Q. Hood, and J. C. Grossman, PRL 87, 246406 (2001)]

(Reblocked) Statistical standard error

Standard error in average over $N_{\mathrm{step}} \times N_{\mathrm{pop}}$ *correlated* data points:

$$\sigma(E_{\mathrm{total}}) \;=\; N_{\mathrm{atom}} \times \sigma(E_{\mathrm{atom}}) \;=\; \sqrt{\left(\frac{\tau_{\mathrm{corr}}}{\tau_{\mathrm{step}}}\frac{1}{N_{\mathrm{step}}}\right)\left(\frac{\chi_{\mathrm{pop}}}{N_{\mathrm{pop}}}\right)\sigma^2_{\mathrm{dmc}}}$$

For a perfect insulator: N independent, identical atoms

σ^2_{dmc}	raw DMC variance ($\approx \sigma^2_{\mathrm{vmc}} = \langle\Psi\vert\mathcal{H}^2\vert\Psi\rangle - \langle\Psi\vert\mathcal{H}\vert\Psi\rangle^2 \propto N_{\mathrm{atom}}$)
τ_{corr}	integrated serial correlation time (independent of N_{atom})
$\chi_{\mathrm{pop}} \geqslant 1$	**population correlation factor** (yet unknown)

$$€_{\mathrm{total}} \;\propto\; \frac{\chi_{\mathrm{pop}}}{\sigma^2(E_{\mathrm{atom}})}$$

$\longrightarrow$ Scaling of χ_{pop} with N_{atom} ???

Some formalism

random variable X, expectation value $\langle X \rangle$

variance: $\sigma_X^2 = \mathrm{var}[X] = \langle X^2 \rangle - \langle X \rangle^2$

standard deviation $\sigma_X = \mathrm{std}[X] = \sqrt{\mathrm{var}[X]}$

covariance: $\mathrm{cov}[X, Y] = \langle XY \rangle - \langle X \rangle \langle Y \rangle$ $\Rightarrow \mathrm{cov}[X, X] = \mathrm{var}[X]$

correlation: $\mathrm{corr}[X, Y] = \mathrm{cov}[X, Y] / \sigma_X \sigma_Y$ $\Rightarrow \mathrm{corr}[X, X] = 1$

homogeneous series of random variables X_i, $i = 1 \ldots N$, $\mathrm{var}[X_i] = \sigma_X^2$

variance of correlated sum: $\mathrm{var}[\sum X_i] = \sum_{i,j} \mathrm{cov}[X_i, X_j]$

variance of series average: $\mathrm{var}[\sum X_i / N] = \sum_{i,j} \mathrm{cov}[X_i, X_j] / N^2$

effective sample size: $$N_{\mathrm{eff}} := \frac{\sigma_X^2}{\mathrm{var}[\sum X_i / N]} = \frac{N^2}{\sum_{i,j} \mathrm{corr}[X_i, X_j]}$$

uncorrelated series: $\mathrm{corr}[X_i, X_j] = \delta_{i,j}$ $\Rightarrow$ $N_{\mathrm{eff}} = N$

non-negative correlation: $\mathrm{corr}[X_i, X_j] \geqslant 0$ $\Rightarrow$ $1 \leqslant N_{\mathrm{eff}} \leqslant N$

Integrated autocorrelation factor

homogeneous series of random variables X_i with $\mathrm{var}[X_i] = \sigma_X^2$

Autocorrelation:

$$a_j = \mathrm{corr}[X_i, X_{i+j}]$$

Series average:

$$\langle X \rangle_\tau = \frac{1}{N_{\mathrm{step}}} \sum_{i=1}^{N_{\mathrm{step}}} X_i$$

$$\mathrm{var}\big[\langle X \rangle_\tau\big] = \frac{\sigma_X^2}{N_{\mathrm{step}}^2} \sum_{i,j} \mathrm{corr}[X_i, X_j]$$

$$\overset{N_{\mathrm{step}} \to \infty}{\approx} \frac{\sigma_X^2}{N_{\mathrm{step}}} \sum_{j=-\infty}^{\infty} a_j$$

Integrated autocorrelation factor:

$$\chi_{\mathrm{autocorr}} = \frac{N_{\mathrm{step}}}{N_{\mathrm{step}}^{\mathrm{eff}}} = 1 + 2 \sum_{j=1}^{\infty} a_j$$

Integrated autocorrelation time τ_{corr}

- "physical" process: drift diffusion in continuous time τ
- computer simulation: small, finite time step τ_{step}
- time-step extrapolation $\tau_{\mathrm{step}} \to 0$
- physical quantities must be independent of τ_{step}
- integrated autocorrelation factor must scale as

$$\chi_{\mathrm{autocorr}} = \frac{\tau_{\mathrm{corr}}}{\tau_{\mathrm{step}}}$$

Integrated correlation time τ_{corr}:

$\to$ independent of τ_{step} when close enough to $\tau_{\mathrm{step}} \to 0$

$\to$ characteristic of physical system in DMC process

$\to$ independent of N_{atom} for sufficiently independent atoms

Measuring population correlation

N_{step} time steps (index i)
population size N_{pop} (index p)
local energy of individual configuration: E_p^i

(assuming constant N_{pop}, neglecting weight fluctuations!)

Raw DMC variance:

$$\sigma^2_{\mathrm{dmc}} := \left\langle \left\langle \left(E_p^i\right)^2 \right\rangle_p \right\rangle_i - \left\langle \left\langle E_p^i \right\rangle_p \right\rangle_i^2$$

Variance of population average:

$$\sigma^2_{\mathrm{pop}} := \left\langle \left\langle E_p^i \right\rangle_p^2 \right\rangle_i - \left\langle \left\langle E_p^i \right\rangle_p \right\rangle_i^2$$

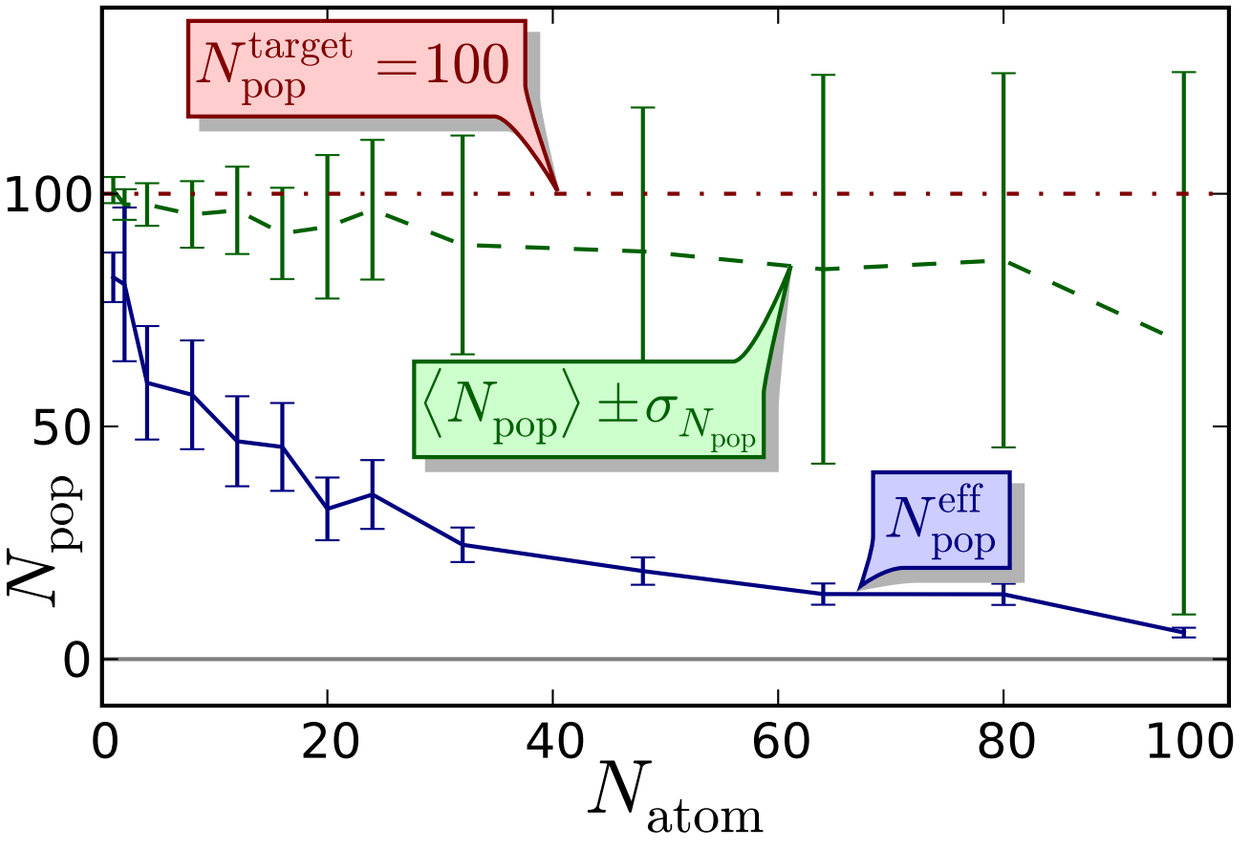
Effective population size:

$$N^{\mathrm{eff}}_{\mathrm{pop}} = \sigma^2_{\mathrm{dmc}} / \sigma^2_{\mathrm{pop}}$$

Population correlation of a real system

- N_{atom} independent hydrogen atoms (nuclear charge Z, one electron)
- STO wave functions with detuning parameter α (exact for $\alpha = 1$):

$$\Psi(r) = \alpha e^{-\alpha Z r} + (1-\alpha)\mathrm{e}^{-(\alpha+1)Zr}$$



$$\begin{aligned} \alpha &= 1.5 \\ Z &= 1 \end{aligned}$$

$$\begin{aligned} \sigma_{\mathrm{dmc}}^2 &\approx 0.08 \times N_{\mathrm{atom}} \\ \tau_{\mathrm{corr}} &\approx 2.0 \end{aligned}$$

$\rightarrow$ significant correlation for $\sigma_{\mathrm{dmc}} \times \tau_{\mathrm{corr}} \gtrsim 1$

Weight fluctuations

Assume weighted average over N_{step} time steps:

$$\langle X \rangle_N = \frac{1}{\sum w_i} \sum_{i=1}^{N_{\mathrm{step}}} w_i X_i$$

(assuming uncorrelated, homogeneous series! $\rightarrow \mathrm{var}[X_i] = \sigma_X^2$*)*

Variance of average:

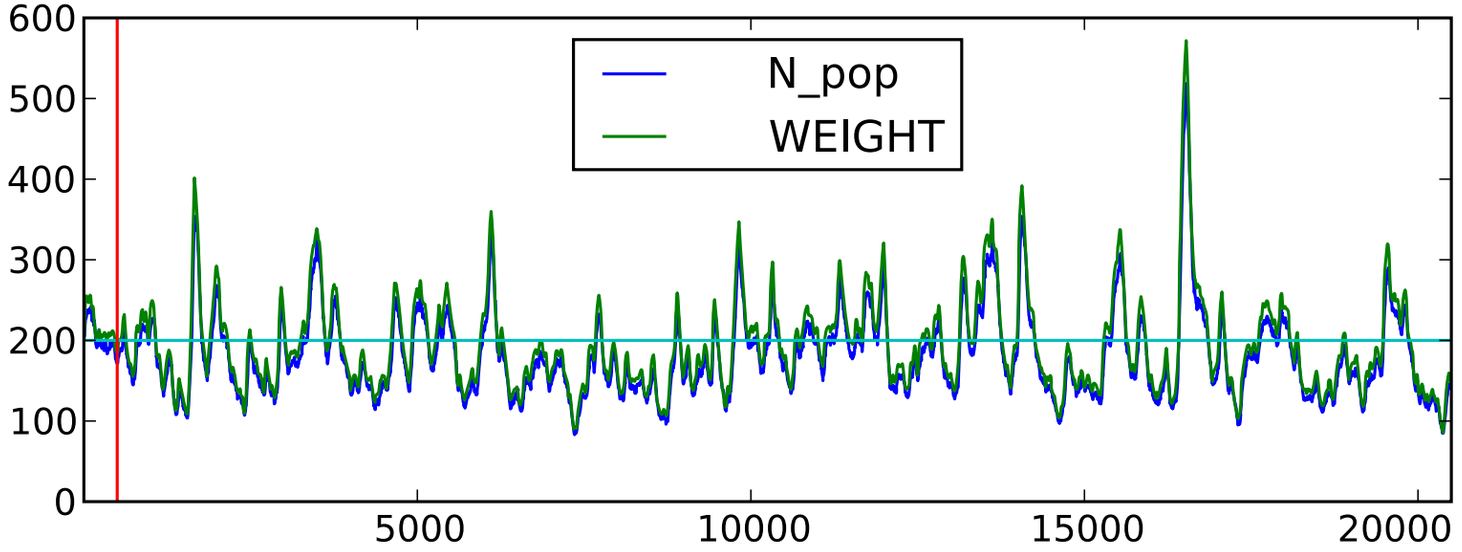
$$\mathrm{var}\left[\langle X \rangle_N\right] = \mathrm{var}\left[\frac{1}{\sum w_i} \sum_{i=1}^{N_{\mathrm{step}}} w_i X_i\right]$$

$$= \frac{1}{\left(\sum w_i\right)^2} \sum_{i=1}^{N} \left(w_i\right)^2 \mathrm{var}[X_i] = \frac{\sum \left(w_i\right)^2}{\left(\sum w_i\right)^2} \sigma_X^2$$

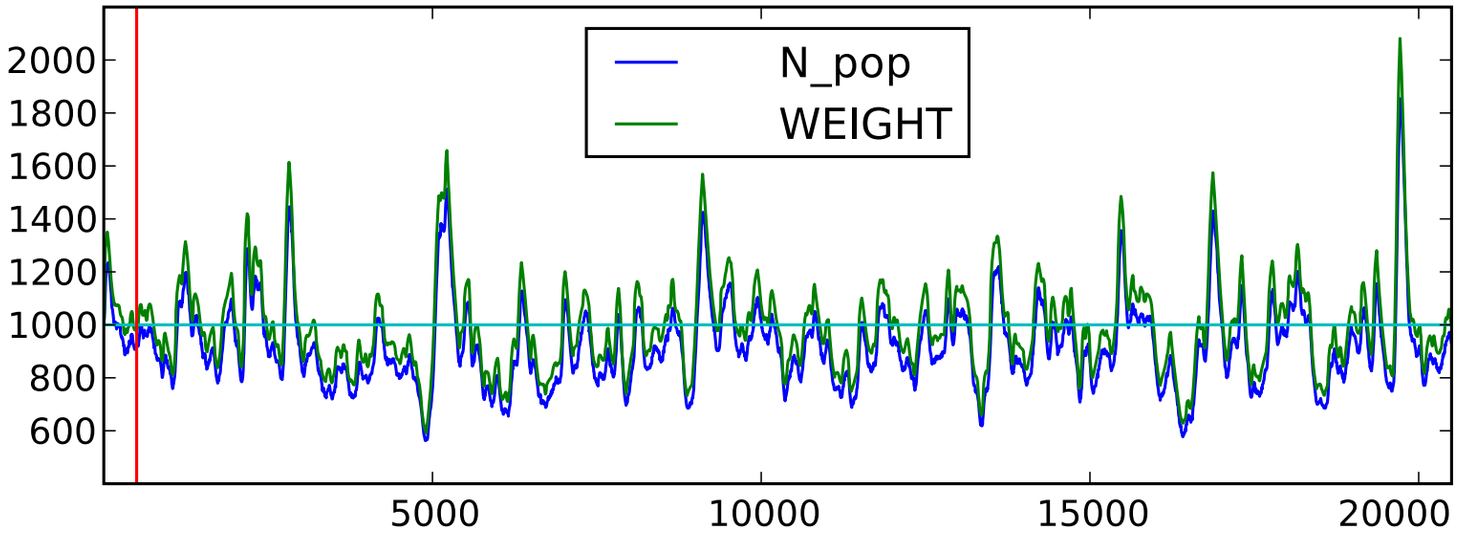
$$N_{\mathrm{step}}^{\mathrm{eff}} = \frac{\sigma_X^2}{\mathrm{var}\left[\langle X \rangle_N\right]} = \frac{\left(\sum w_i\right)^2}{\sum \left(w_i\right)^2}$$

Weight fluctuations in a real system

independent H-atoms: $N_{\mathrm{atom}} = 12,\ \alpha = 3.0\ (\rightarrow \sigma^2_{\mathrm{dmc}} \approx 2.4,\ \tau_{\mathrm{corr}} \approx 1.1)$



$$N_{\mathrm{pop}}^{\mathrm{target}} = 200$$
$$N_{\mathrm{step}} = 20000$$
$$N_{\mathrm{pop}}^{\mathrm{avg}} \approx 180$$
$$N_{\mathrm{pop}}^{\mathrm{eff}} \approx 45$$
$$N_{\mathrm{step}}^{\mathrm{eff}} \approx 18300$$
$$\Rightarrow \chi_{\mathrm{pop}} \approx 4.3$$



$$N_{\mathrm{pop}}^{\mathrm{target}} = 1000$$
$$N_{\mathrm{step}} = 20000$$
$$N_{\mathrm{pop}}^{\mathrm{avg}} \approx 910$$
$$N_{\mathrm{pop}}^{\mathrm{eff}} \approx 148$$
$$N_{\mathrm{step}}^{\mathrm{eff}} \approx 19400$$
$$\Rightarrow \chi_{\mathrm{pop}} \approx 6.3$$

Measuring statistical inefficency χ_{pop}

Total energy of DMC run – a weighted average over N_{step} values:

$$E_{\mathrm{tot}} = \frac{1}{\sum_i w_i} \sum_{i=1}^{N_{\mathrm{step}}} w_i \left\langle E_p^i \right\rangle_{\mathrm{pop}}$$

View E_p^i as random variables for fixed w_i and N_{pop}^i:

$$\begin{aligned} \mathrm{var}[E_{\mathrm{tot}}] &= \frac{\tau_{\mathrm{corr}}}{\tau_{\mathrm{step}}} \frac{1}{\left(\sum_i w_i\right)^2} \sum_{i=1}^{N_{\mathrm{step}}} w_i^2 \mathrm{var}\left[\left\langle E_p^i \right\rangle_{\mathrm{pop}}\right] \\ &=: \frac{\tau_{\mathrm{corr}}}{\tau_{\mathrm{step}}} \times \frac{1}{N_{\mathrm{step}}^{\mathrm{eff}}} \times \frac{1}{N_{\mathrm{pop}}^{\mathrm{eff}}} \times \sigma_{\mathrm{dmc}}^2 \end{aligned}$$

$$\frac{1}{N_{\mathrm{step}}^{\mathrm{eff}}} \times \frac{1}{N_{\mathrm{pop}}^{\mathrm{eff}}} = \frac{\sum_i \left(w_i^2\right)}{\left(\sum_i w_i\right)^2} \times \left(1 - \frac{1}{\sigma_{\mathrm{dmc}}^2} \frac{\sum_i w_i^2 \left(\left\langle \left(E_p^i\right)^2 \right\rangle_{\mathrm{pop}} - \left\langle E_p^i \right\rangle_{\mathrm{pop}}^2 \right)}{\sum_i w_i^2} \right)$$

$$\chi_{\mathrm{pop}} = \frac{N_{\mathrm{step}} \times N_{\mathrm{pop}}}{N_{\mathrm{step}}^{\mathrm{eff}} \times N_{\mathrm{pop}}^{\mathrm{eff}}}$$

Variance of local energy

$$\partial_t f(\boldsymbol{r}) \;=\; \frac{1}{2}\nabla^2 f(\boldsymbol{r}) - \nabla\cdot\left(\frac{\nabla\psi_{\mathrm{apx}}}{\psi_{\mathrm{apx}}}\right) f(\boldsymbol{r}) + \left[E - E_{\mathrm{loc}}(\boldsymbol{r})\right] f(\boldsymbol{r})$$

$$E_{\mathrm{loc}} \;=\; -\frac{\nabla^2\Psi_{\mathrm{apx}}(\boldsymbol{r})}{\Psi_{\mathrm{apx}}(\boldsymbol{r})} + V(\boldsymbol{r})$$

VMC-variance σ^2_{vmc} – physical property of trial wave function Ψ_{apx}:

$$\sigma^2_{\mathrm{vmc}} \;=\; \left\langle \Psi_{\mathrm{axp}} \middle| \mathcal{H}^2 \middle| \Psi_{\mathrm{apx}} \right\rangle - \left\langle \Psi_{\mathrm{axp}} \middle| \mathcal{H} \middle| \Psi_{\mathrm{apx}} \right\rangle^2 = \mathrm{var}_{\Psi^2_{\mathrm{apx}}}[E_{\mathrm{loc}}]$$

- depends on quality of trial wfn: $\Psi_{\mathrm{apx}} \to \Psi_0 \Rightarrow \sigma^2_{\mathrm{vmc}} \to 0$
- typically scales with system size: $\sigma^2_{\mathrm{vmc}} \propto N_{\mathrm{atom}}$

DMC-variance σ^2_{dmc} ("raw" variance of local energy during DMC run):

$$\sigma^2_{\mathrm{dmc}} \;=\; \mathrm{var}_{\Psi_{\mathrm{DMC}}(\boldsymbol{r})\cdot\Psi_{\mathrm{apx}}(\boldsymbol{r})}[E_{\mathrm{loc}}(\boldsymbol{r})]$$

- typically $\sigma^2_{\mathrm{dmc}} \approx \sigma^2_{\mathrm{vmc}}$ (since $\Psi_{\mathrm{DMC}} \approx \Psi_{\mathrm{apx}} \approx \Psi_0$)

Asymptotic estimate of χ_{pop} for $\tau_{\mathrm{corr}} \ll \tau_{\mathrm{branch}}$

Branching rate:

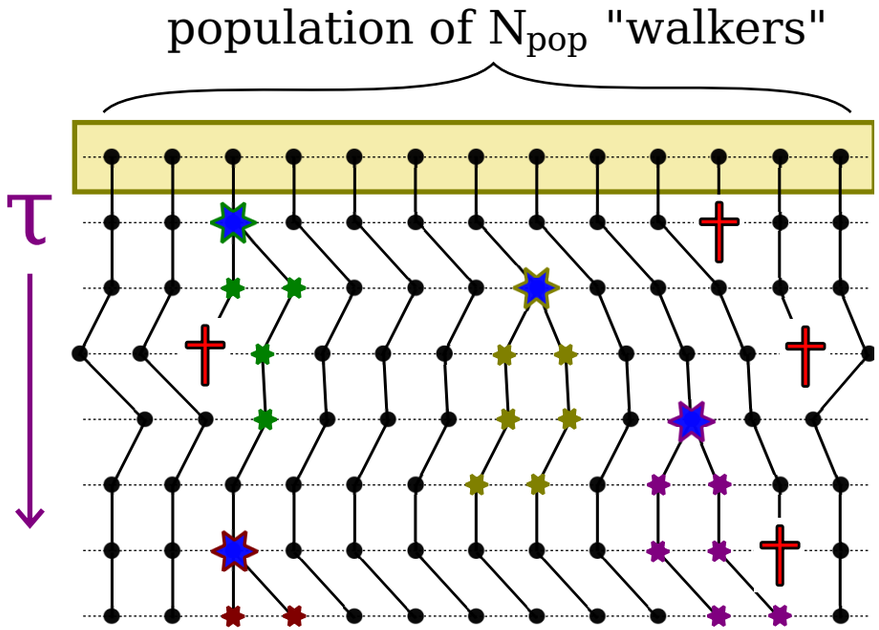
$$\tau_{\mathrm{branch}}^{-1} = \frac{\sigma_{\mathrm{dmc}}}{\sqrt{2\pi}}$$

Killing rate:

$$\tau_{\mathrm{kill}}^{-1} = \tau_{\mathrm{branch}}^{-1}$$

Time to decorrelate two identical copies:

$$\tau_{\mathrm{corr}}/2$$



after killing event ($N'_{\mathrm{pop}} = N_{\mathrm{pop}} - 1$):

$$N_{\mathrm{pop}}^{\mathrm{eff}\,\prime} = N_{\mathrm{pop}}^{\mathrm{eff}} - 1$$

after branching event ($N'_{\mathrm{pop}} = N_{\mathrm{pop}} + 1$):

$\rightarrow$ $N_{\mathrm{pop}} - 1$ uncorrelated walkers, 2 identical copies

$$N_{\mathrm{pop}}^{\mathrm{eff}\,\prime} = \frac{\left(\sum w_i\right)^2}{\sum \left(w_i\right)^2} = \frac{\left(\left(N_{\mathrm{pop}} - 1\right) \times 1 + 1 \times 2\right)^2}{\left(\left(N_{\mathrm{pop}} - 1\right) \times 1^2 + 1 \times 2^2\right.} = \frac{\left(N_{\mathrm{pop}} + 1\right)^2}{N_{\mathrm{pop}} + 3} \overset{N_{\mathrm{pop}} \gg 1}{\approx} N_{\mathrm{pop}} - 1$$

Asymptotic estimate of χ_{pop} for $\tau_{\mathrm{corr}} \ll \tau_{\mathrm{branch}}$

Branching rate:

$$\tau_{\mathrm{branch}}^{-1} = \frac{\sigma_{\mathrm{dmc}}}{\sqrt{2\pi}}$$

Killing rate:

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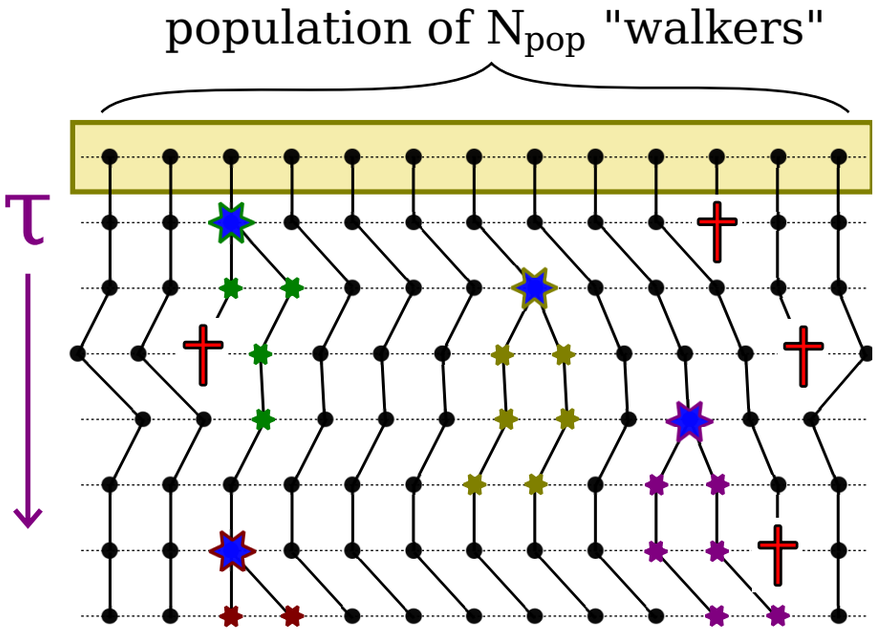
$$\tau_{\mathrm{corr}}/2$$

after branching event:

$$N'_{\mathrm{pop}} = N_{\mathrm{pop}} + 1 \qquad N_{\mathrm{pop}}^{\mathrm{eff}\,\prime} = N_{\mathrm{pop}}^{\mathrm{eff}} - 1$$

after killing event:

$$N'_{\mathrm{pop}} = N_{\mathrm{pop}} - 1 \qquad N_{\mathrm{pop}}^{\mathrm{eff}\,\prime} = N_{\mathrm{pop}}^{\mathrm{eff}} - 1$$



$\Rightarrow$ asymptotically for $\tau_{\mathrm{corr}} \ll \tau_{\mathrm{branch}}$:

$$\left\langle N_{\mathrm{pop}}^{\mathrm{eff}} \right\rangle = N_{\mathrm{pop}} - 2N_{\mathrm{pop}}\, \tau_{\mathrm{branch}}^{-1} \times \tau_{\mathrm{corr}} \,/2$$

$$\chi_{\mathrm{pop}} = 1 + \tau_{\mathrm{corr}}\sigma_{\mathrm{dmc}} \Big/ \sqrt{2\pi}$$

Model DMC process

Uncorrelated Gaussian random sequence ξ_i:

$$\langle \xi_i \rangle = 0 \ ; \ \mathrm{var}[\xi_i] = \sigma^2$$

Defined correlated diffusion process:

$$E_0 := X_0 \ , \ E_i := \alpha E_{i-1} + \sqrt{1-\alpha^2}\, \xi_i$$

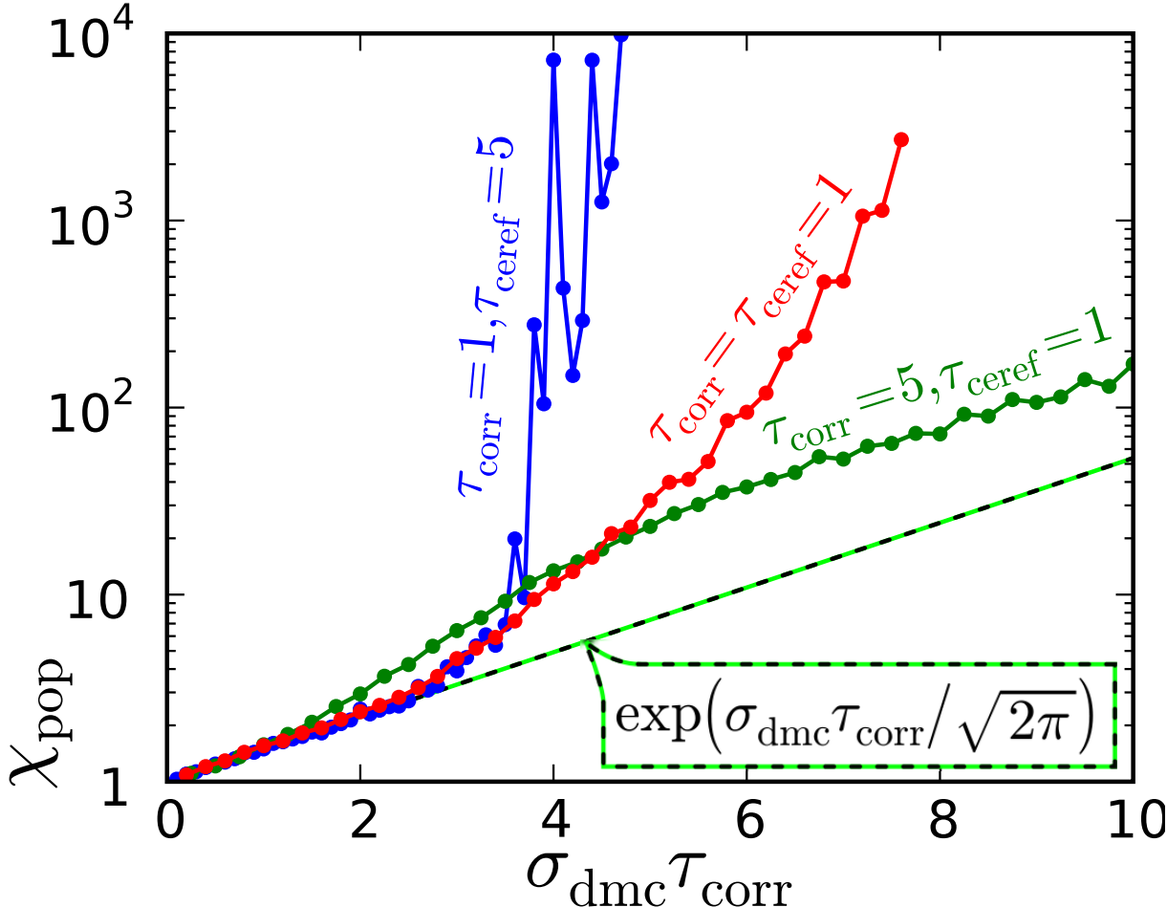
$$\Rightarrow \mathrm{corr}[E_i, E_j] \ = \ \alpha^{|i-j|}$$

$$\Rightarrow \frac{\tau_{\mathrm{corr}}}{\tau_{\mathrm{step}}} \ = \ \frac{1+\alpha}{1-\alpha}$$

Model DMC process:

- each walker defined by single variable E
- correlated diffusion step according to $E \rightarrow \alpha E + \sqrt{1-\alpha^2}\,\xi$
- branching according to weight $w = \exp(E_{\mathrm{ref}} - E)$
- algorithm (incl. population control) similar to CASINO ($\rightarrow \tau_{\mathrm{ceref}}$)

Model DMC process – results



simulated DMC process
based on correlated series
of gaussian random numbers

parameter τ_{ceref}:
$\rightarrow$ population control

confirmed for $\sigma_{\mathrm{dmc}}\tau_{\mathrm{corr}} \ll 1$:

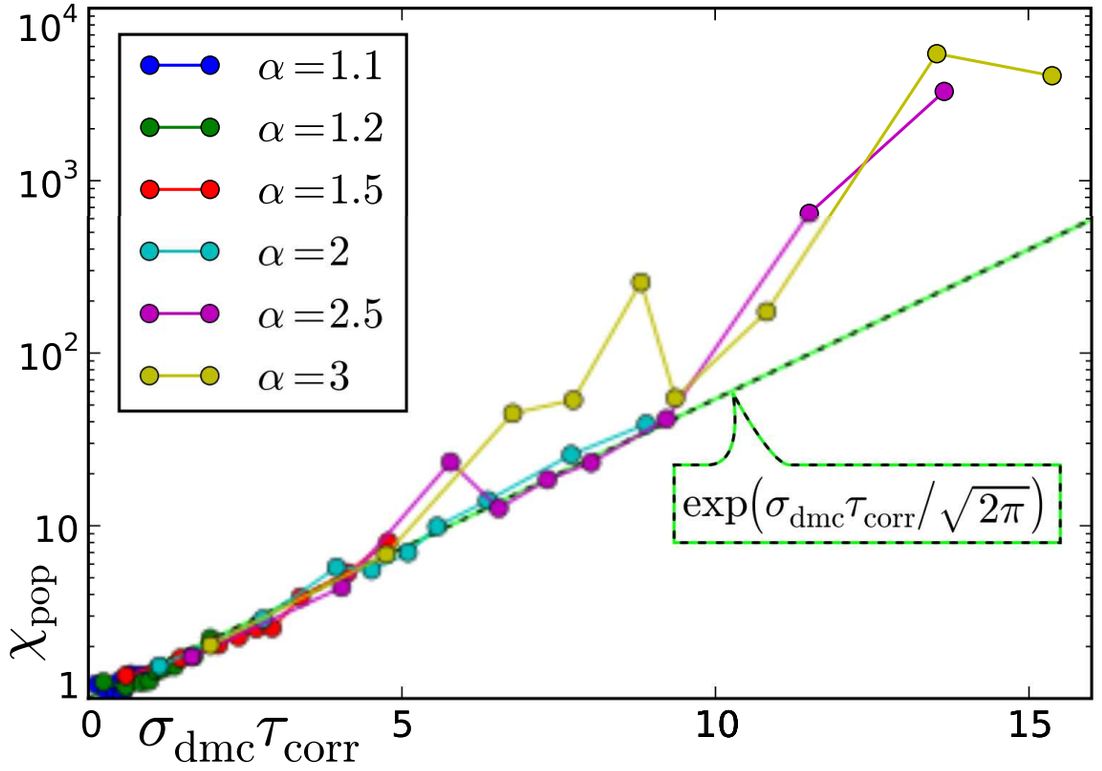
$$\chi_{\mathrm{pop}} = 1 + \frac{\tau_{\mathrm{corr}}\sigma_{\mathrm{dmc}}}{\sqrt{2\pi}}$$

found extrapolation:

$$\chi_{\mathrm{pop}} \gtrsim \exp\left(\frac{\tau_{\mathrm{corr}}\sigma_{\mathrm{dmc}}}{\sqrt{2\pi}}\right)$$

Scaling of χ_{pop} in sample system

- N_{atom} independent hydrogen atoms with detuned wave function



- $N_{\mathrm{atom}} \in \{1, \ldots, 64\}$
- $N_{\mathrm{pop}} = 200$
- $Z = 1$

$\longrightarrow$ $\tau_{\mathrm{corr}} \approx 2.55 \ldots 1.06$

$\longrightarrow$ $\sigma^2_{\mathrm{dmc}} \approx \sigma^2_{\mathrm{vmc}}$

confirmed for $\sigma_{\mathrm{dmc}}\tau_{\mathrm{corr}} \ll 1$:

$$\chi_{\mathrm{pop}} = 1 + \frac{\tau_{\mathrm{corr}}\sigma_{\mathrm{dmc}}}{\sqrt{2\pi}}$$

found extrapolation:

$$\chi_{\mathrm{pop}} \gtrsim \exp\left(\frac{\tau_{\mathrm{corr}}\sigma_{\mathrm{dmc}}}{\sqrt{2\pi}}\right)$$

Quantitative analysis of various real systems

atoms (ae)	τ_{corr}	$\sigma^2_{\mathrm{dmc}}/\mathrm{atm}$	$\chi_{\mathrm{pop}}=2$
He	0.5	0.0044	2700 atoms
C	0.4	0.16	140 atoms
Ar	0.04	8.0	250 atoms
molecules (ae)		$\sigma^2_{\mathrm{dmc}}/\mathrm{mlc}$	
$\mathrm{H_2O}$	0.1	0.58	550 molec.
$\mathrm{CH_4}$	0.3	0.24	120 molec.
$\mathrm{C_2H_4}$	0.4	0.51	38 molec.
$\mathrm{SO_2}$	0.06	7.5	105 molec.
crystals		$\sigma^2_{\mathrm{dmc}}/\mathrm{atom}$	
diamond (pp)	0.15	0.23	630 atoms
diamond (ae)	0.1	2.3	133 atoms
graphite (pp)	0.3	0.20	135 atoms
silicon (pp)	0.4	0.052	328 atoms
electron gas		$\sigma^2_{\mathrm{dmc}}/\mathrm{elec.}$	
3d crystal ($r_s=1$)	0.2	0.26	193 elec.
3d fluid ($r_s=5$)	5	0.00042	330 elec.
3d fluid ($r_s=10$)	16	0.000051	242 elec.
2d crystal ($r_s=1$)	0.4	0.038	570 elec.
2d fluid ($r_s=1$)	0.3	0.033	1154 elec.

The big picture

Conjecture: exponential scaling of χ_{pop} is fundamental property of DMC.

"Diffusion Monte Carlo":

- not really a Monte Carlo *integration*
- rather the simulation of the time evolution of a chaotic process
- as such exponentially instable in simulation time/system size

$\rightarrow$ other QMC methods do not suffer from this kind of scaling:
variational MC, path integral MC, generally: proper MC integrations

($\rightarrow$ very different from the fermion sign problem!)

Summary

- CPU cost of DMC simulation:

$$€_{\text{total}} \propto \frac{\chi_{\text{pop}}}{\sigma^2(E_{\text{atom}})}$$

- Measurement of statistical inefficiency:

$$\chi_{\text{pop}} = \frac{N_{\text{step}}}{N_{\text{step}}^{\text{eff}}} \times \frac{N_{\text{pop}}}{N_{\text{pop}}^{\text{eff}}}$$

- Asympotics + extrapolation based on numerical data:

$$\chi_{\text{pop}} \gtrsim \exp\left(\frac{\tau_{\text{corr}}\sigma_{\text{dmc}}}{\sqrt{2\pi}}\right)$$

- Statistical inefficiency becomes relevant for typically $N_{\text{atom}} \gtrsim 100$
- Beyond this point, DMC scales as

$$€_{\text{total}} \propto \exp\left(X\sqrt{N_{\text{atom}}}\right)$$