Quadratic Scaling Bosonic Path Integral Molecular Dynamics

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Molecular simulations

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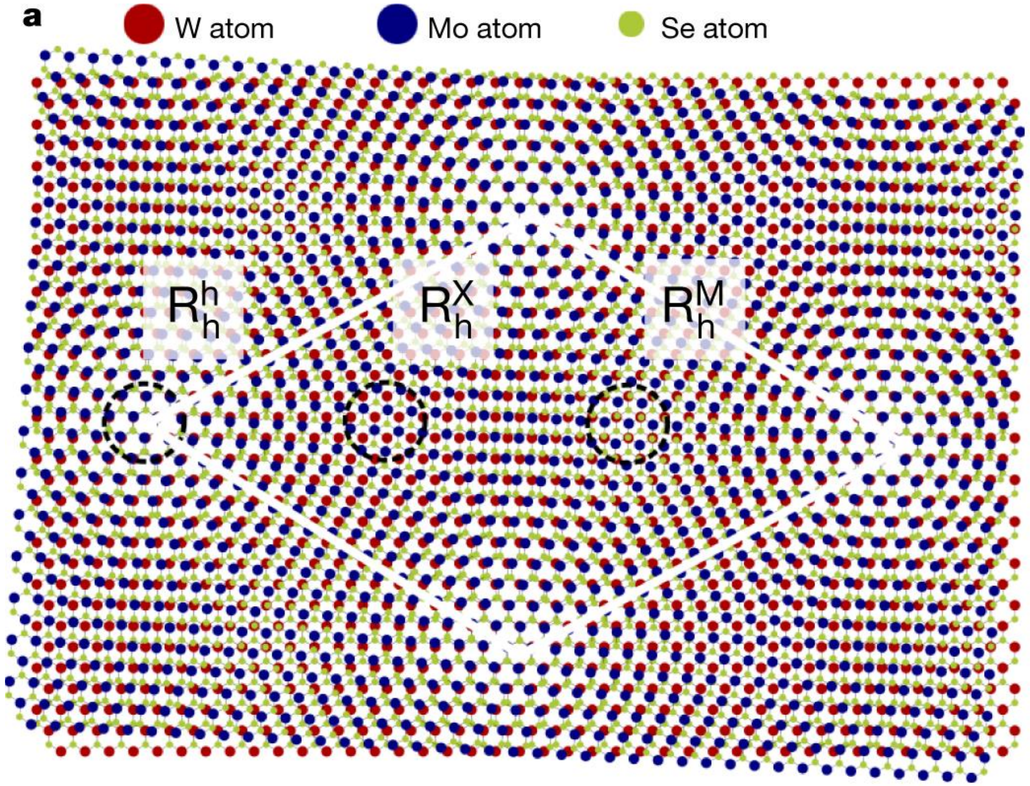
Journal of Chemical Physics, Editors’ Pick, 2023 Emerging Investigators Special Collection
doi:10.1063/5.0173749

Bosonic Exchange

- Quantum symmetry is a fundamental property of quantum particles.
- Exchange effects at finite T are important for many systems

$$\psi(q_1, q_2) = \psi(q_2, q_1)$$

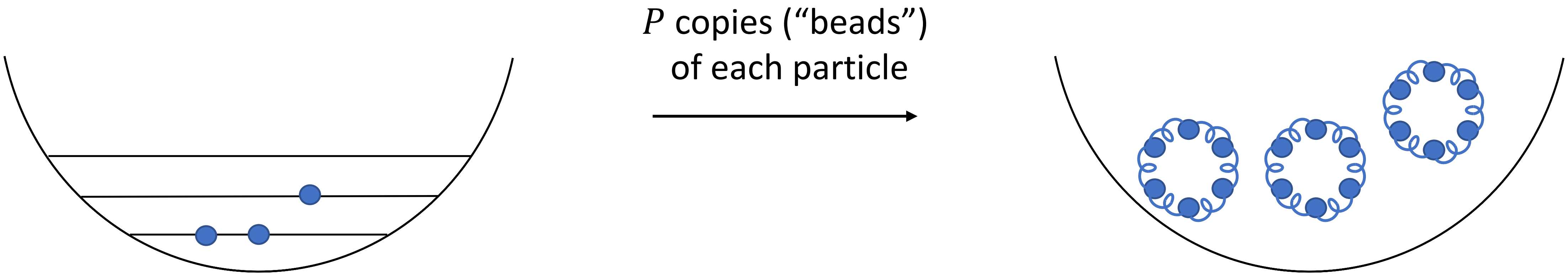
Bosonic Exchange Effects



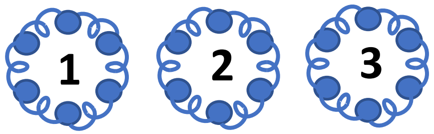
Netanel
Bachar Schwartz

Wang et al. Nature 574, 76-80 (2019)
Tran, Moody et al. Nature 567, 71–75 (2019)

Path-Integral Molecular Dynamics



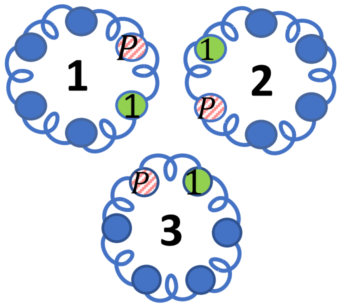
$$Z = \mathrm{Tr}\left[e^{-\beta\hat{H}}\right] \sim \int \left(e^{-\beta E^{(1)(2)(3)}}\right) d\boldsymbol{q}_1 d\boldsymbol{q}_2 d\boldsymbol{q}_3$$

$E^{(1)(2)(3)}$ 

Bosonic Path-Integral Molecular Dynamics

Bosonic Path-Integral Molecular Dynamics

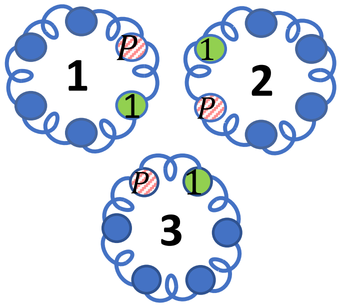
$$Z \sim \frac{1}{6} \int \left(e^{-\beta E^{(1)(2)(3)}} \right) d\boldsymbol{q}_1 d\boldsymbol{q}_2 d\boldsymbol{q}_3$$



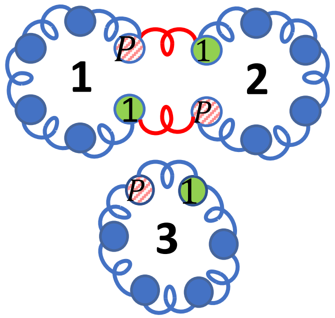
$E^{(1)(2)(3)}$

Bosonic Path-Integral Molecular Dynamics

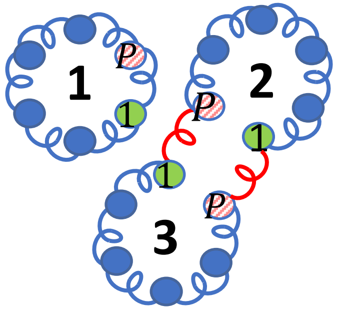
$$Z \sim \frac{1}{6} \int \left(e^{-\beta E^{(1)(2)(3)}} + e^{-\beta E^{(12)(3)}} + e^{-\beta E^{(1)(23)}} + e^{-\beta E^{(13)(2)}} \right) d\boldsymbol{q}_1 d\boldsymbol{q}_2 d\boldsymbol{q}_3$$



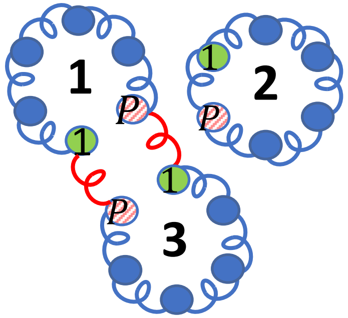
$E^{(1)(2)(3)}$



$E^{(12)(3)}$



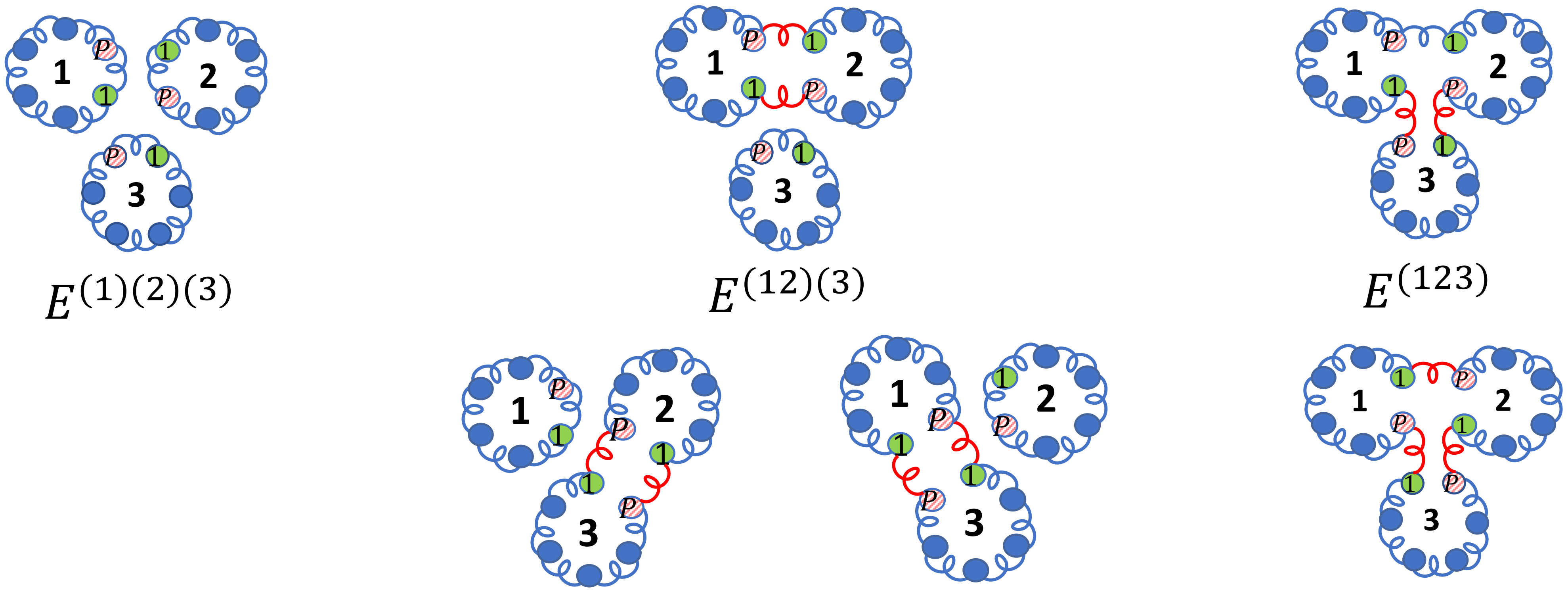
$E^{(1)(23)}$



$E^{(13)(2)}$

Bosonic Path-Integral Molecular Dynamics

$$Z \sim \frac{1}{6} \int \left(e^{-\beta E^{(1)(2)(3)}} + e^{-\beta E^{(12)(3)}} + e^{-\beta E^{(1)(23)}} + e^{-\beta E^{(13)(2)}} + e^{-\beta E^{(123)}} + e^{-\beta E^{(132)}} \right) d\boldsymbol{q}_1 d\boldsymbol{q}_2 d\boldsymbol{q}_3$$

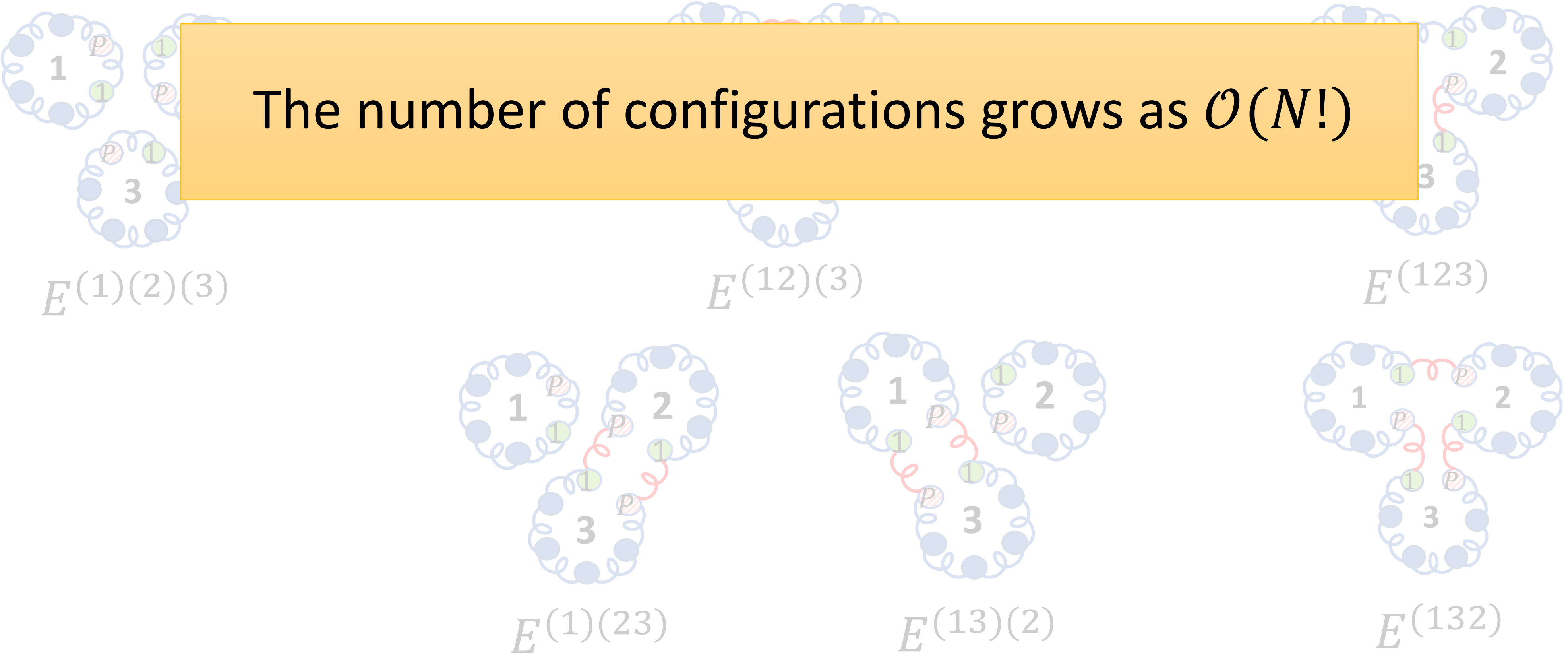


$E^{(1)(2)(3)}$ $E^{(12)(3)}$ $E^{(123)}$

$E^{(1)(23)}$ $E^{(13)(2)}$ $E^{(132)}$

Bosonic Path-Integral Molecular Dynamics

$$Z \sim \frac{1}{6} \int \left(e^{-\beta E^{(1)(2)(3)}} + e^{-\beta E^{(12)(3)}} + e^{-\beta E^{(1)(23)}} + e^{-\beta E^{(13)(2)}} + e^{-\beta E^{(123)}} + e^{-\beta E^{(132)}} \right) d\boldsymbol{q}_1 d\boldsymbol{q}_2 d\boldsymbol{q}_3$$



The number of configurations grows as $\mathcal{O}(N!)$

Scaling of Bosonic Path-Integral MD

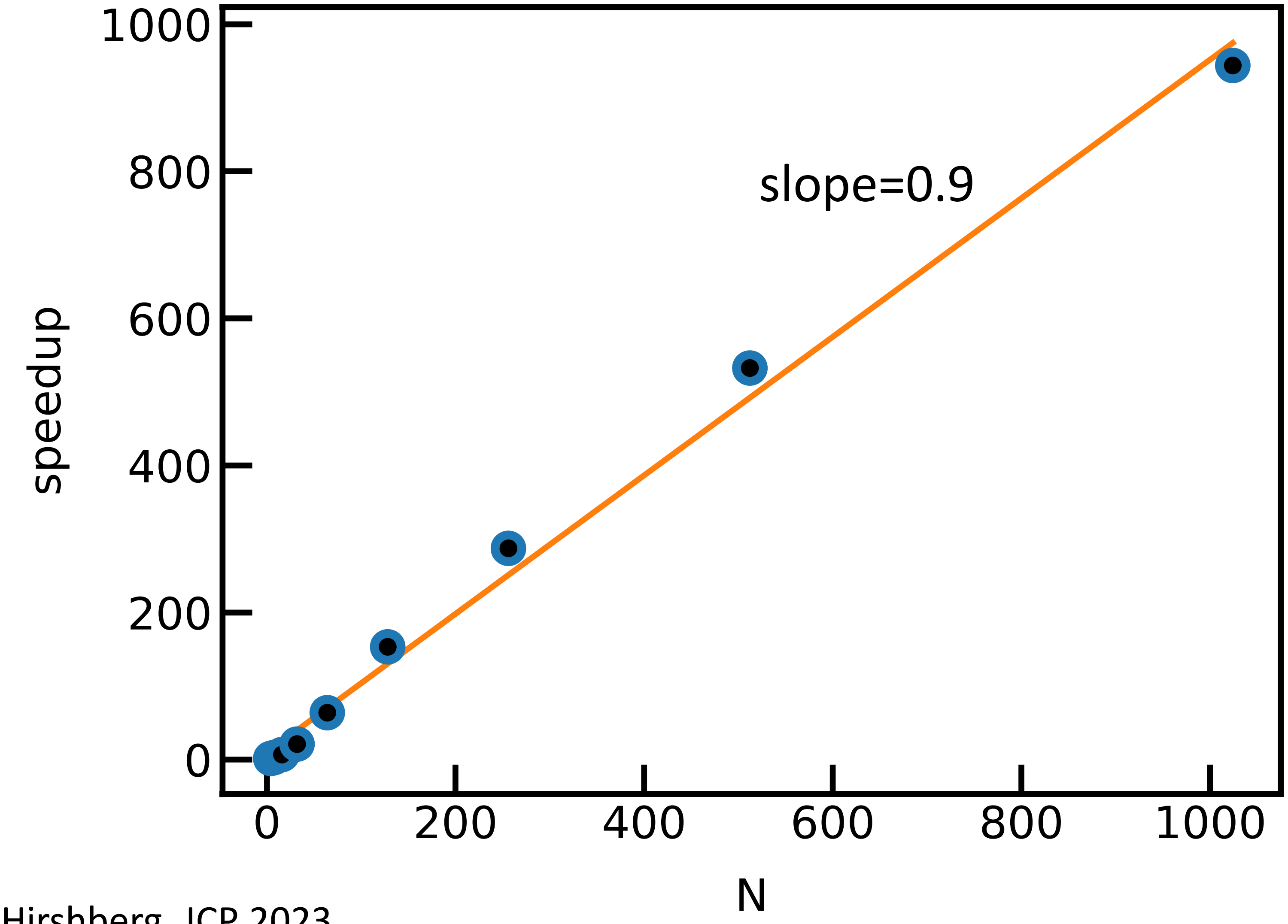
$N = \#\text{bosons},\ P = \#\text{beads}$

All permutations:	$\mathcal{O}(PN!)$		
Hirshberg et al.:	$\mathcal{O}(PN^3)$	$\leftarrow$	~ 100 bosons
This work:	$\boldsymbol{\mathcal{O}(N^2 + PN)}$	$\leftarrow$	> 1000 bosons

Hirshberg et al. PNAS 2019

Feldman and Hirshberg. JCP 2023

Speedup $\mathcal{O}(\boldsymbol{PN})$

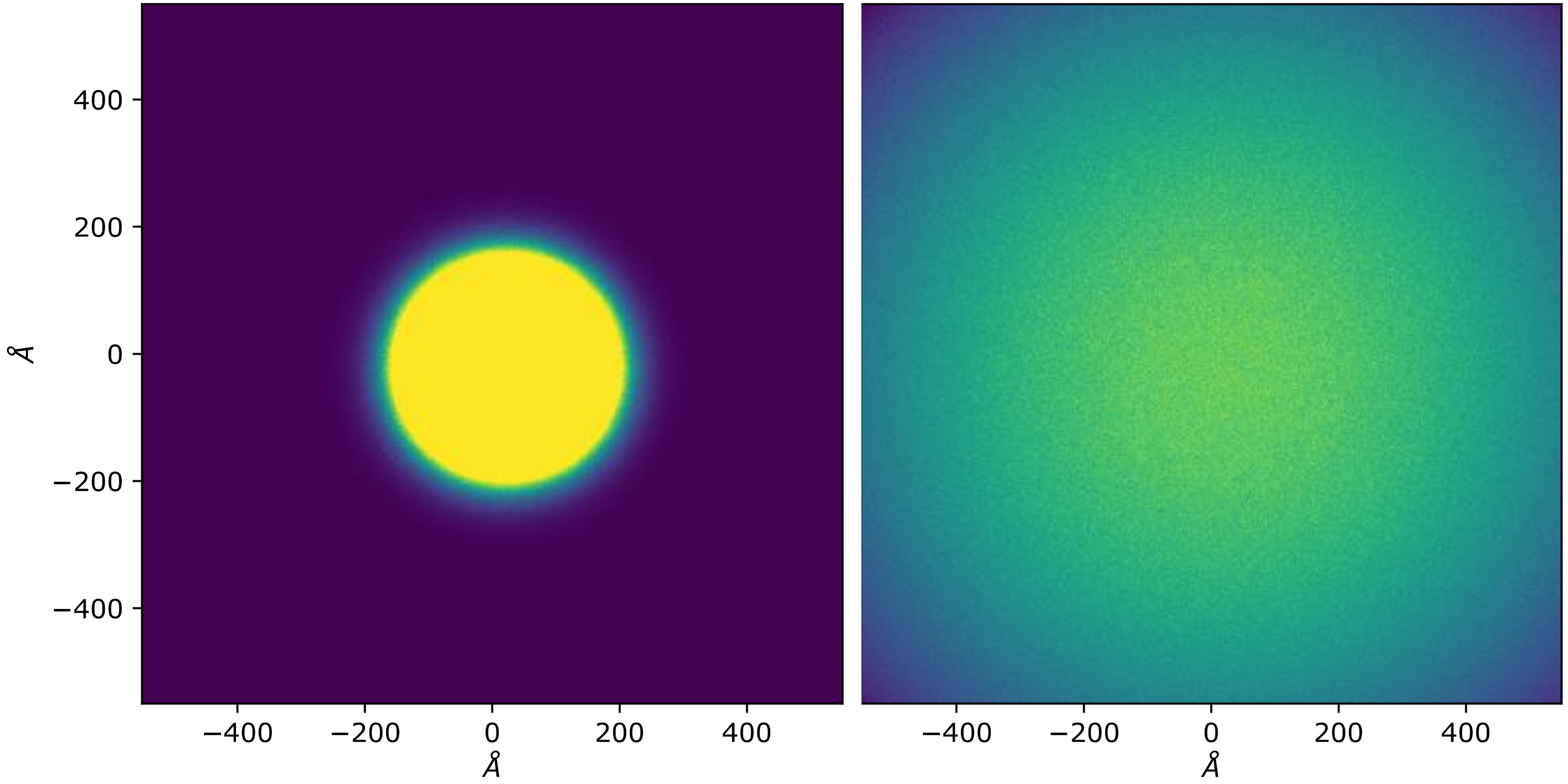


Feldman and Hirshberg. JCP 2023

1600 Particles

9 days of simulation rather than > 20 years

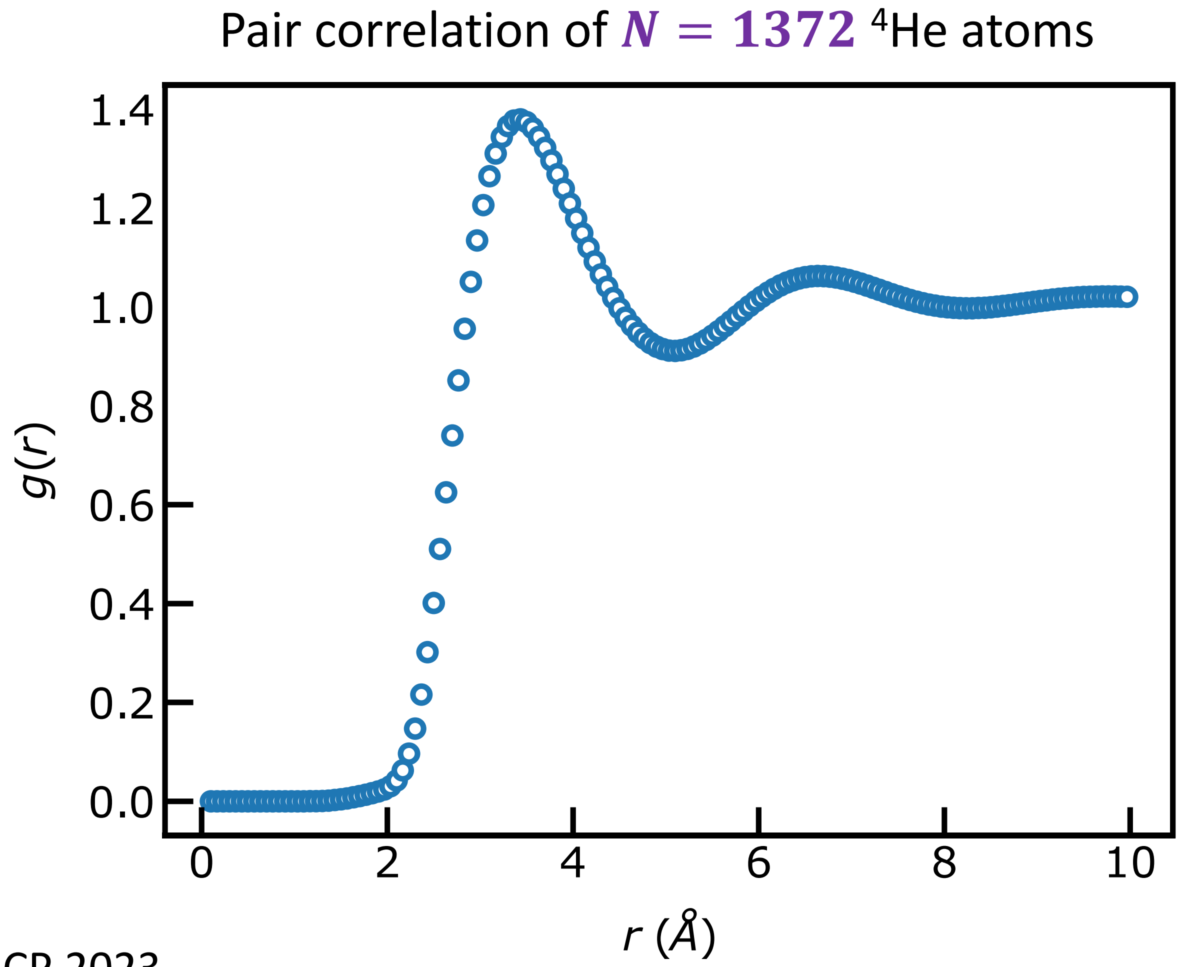
2D density of $N = 1600$ trapped bosons
without and with Gaussian repulsive interaction



Feldman and Hirshberg. JCP 2023

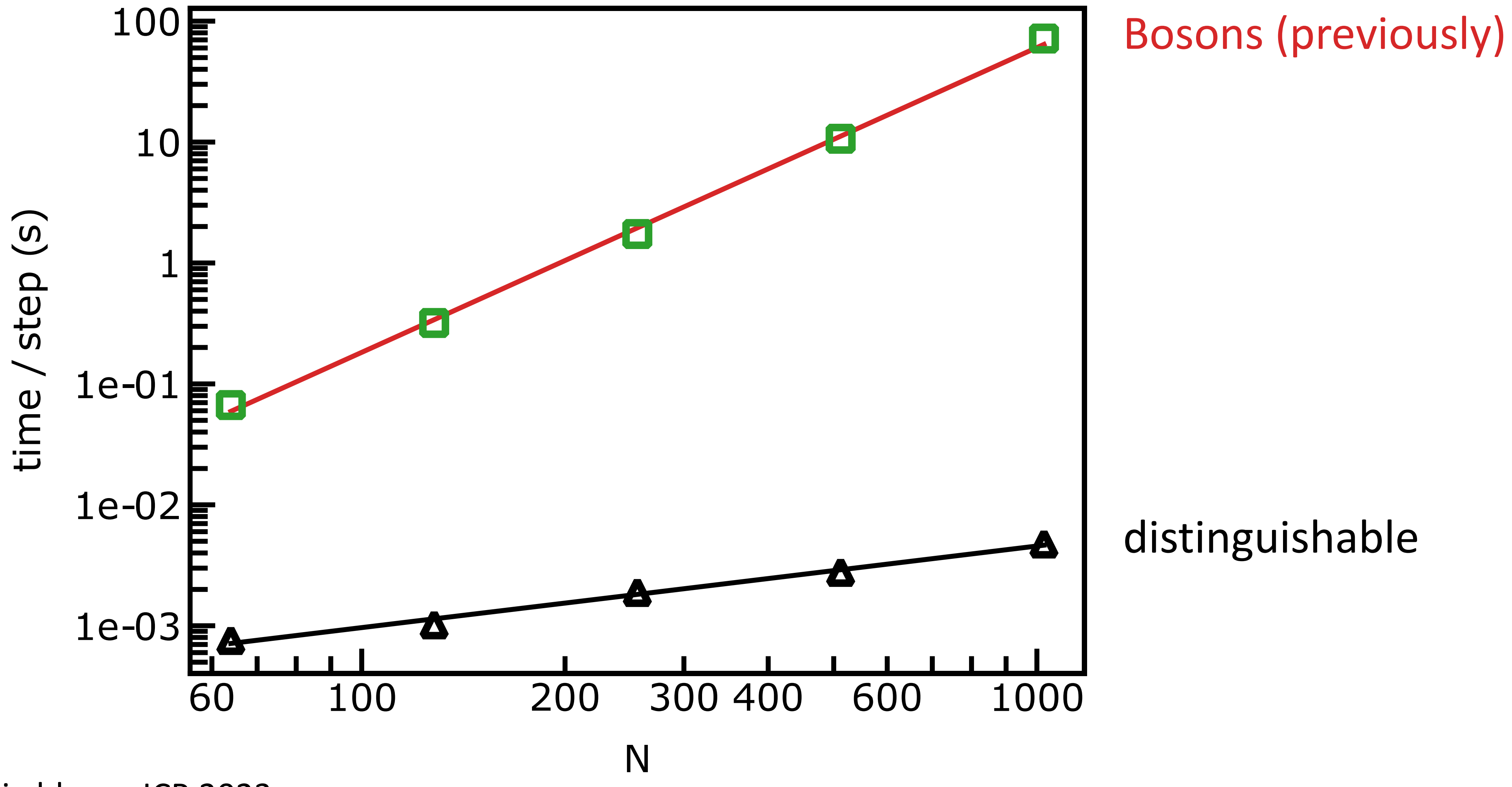
1372 Particles

7 days of simulation rather than > 14 years



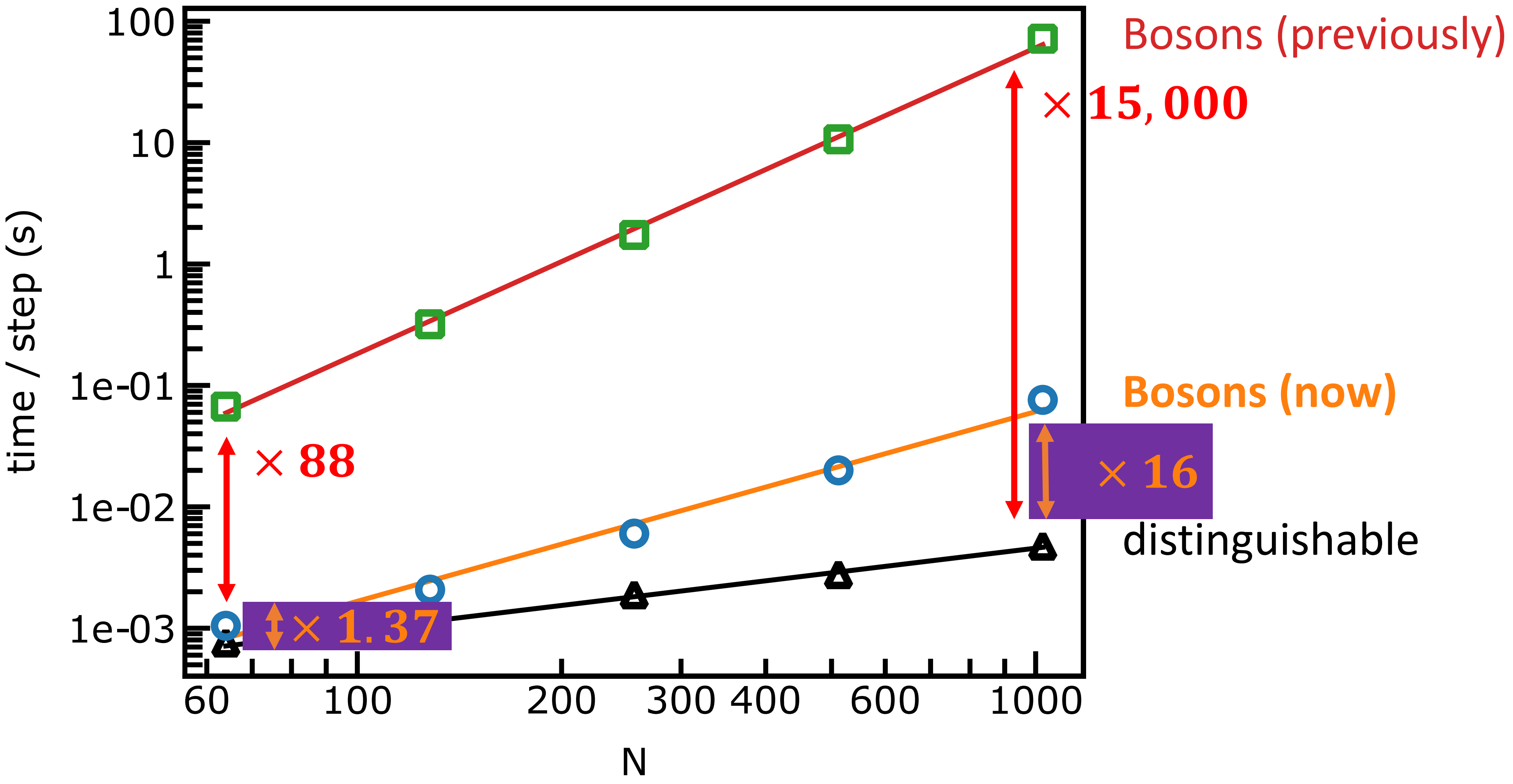
Feldman and Hirshberg. JCP 2023

The Cost of Bosonic Exchange



Feldman and Hirshberg. JCP 2023

The Cost of Bosonic Exchange



Feldman and Hirshberg. JCP 2023

Hirshberg et al.'s Algorithm

Potential:

$$e^{-\beta V^{[1,N]}} = \frac{1}{N}\sum_{k=1}^{N} e^{-\beta\left[V^{[1,N-k]}+E^{[N-k+1,N]}\right]}$$

$\mathcal{O}(PN^3)$ to evaluate

Forces:

$$-\nabla_{q_\ell^j} V^{[1,N]} = \cdots$$

$\mathcal{O}(PN^3)$ to evaluate

Our Algorithm

Potential:

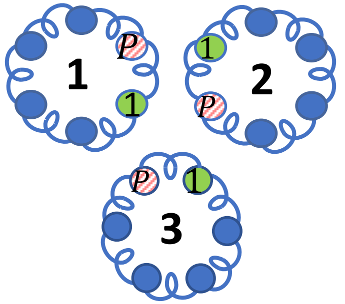
$$e^{-\beta V^{[1,N]}} = \frac{1}{N}\sum_{k=1}^{N} e^{-\beta\left[V^{[1,N-k]}+E^{[N-k+1,N]}\right]}$$

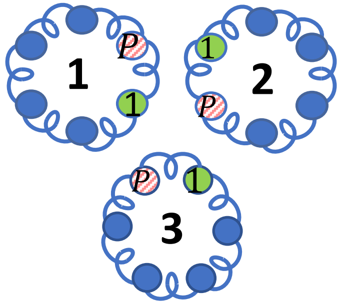
$\mathcal{O}(N^2 + PN)$ **to evaluate**

Forces:

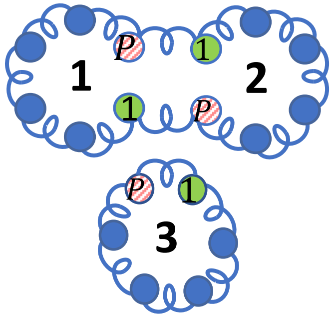
$$-\nabla_{q_\ell^j} V^{[1,N]} = \cdots$$

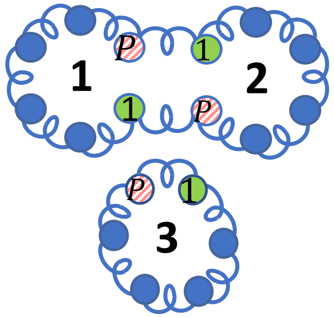
$\mathcal{O}(N^2 + PN)$ **to evaluate**

$-\nabla_{q_1^j} V^{[1,N]} = \Pr[$ 

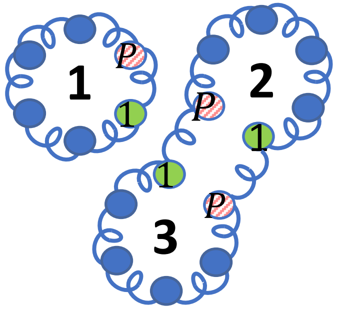
$] \cdot -\nabla_{q_1^j}($ 

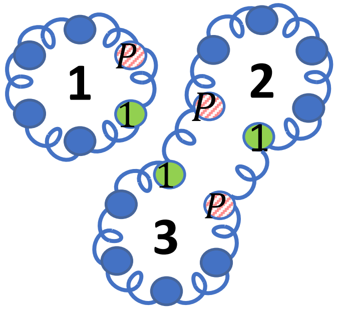
$)$

$+ \Pr[$ 

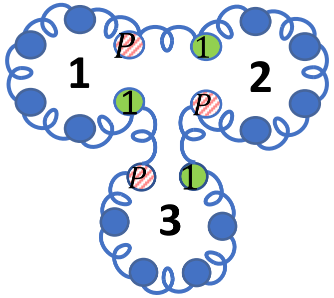
$] \cdot -\nabla_{q_1^j}($ 

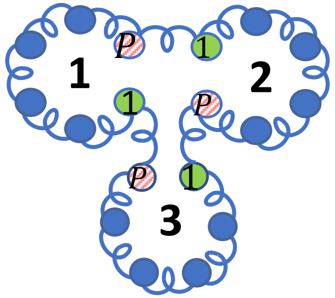
$)$

$+ \Pr[$ 

$] \cdot -\nabla_{q_1^j}($ 

$)$

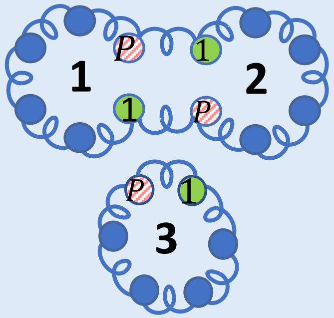
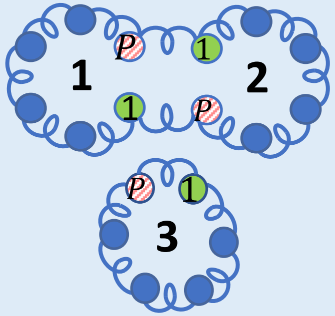
$+ \Pr[$ 

$] \cdot -\nabla_{q_1^j}($ 

$)$

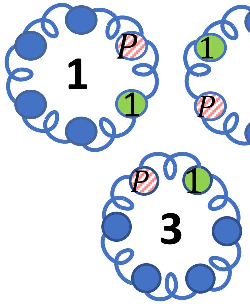
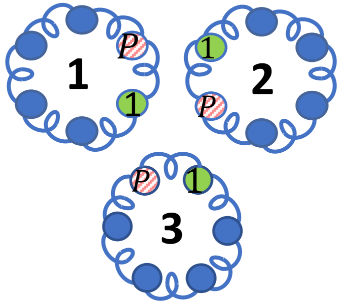
$-\nabla_{q_1^j} V^{[1,N]} = \Pr[$ $] \cdot -\nabla_{q_1^j}($ $)$

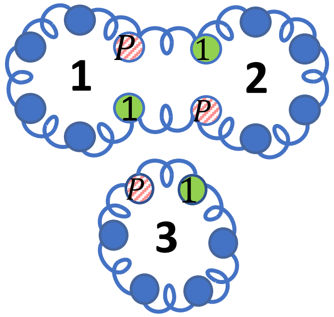
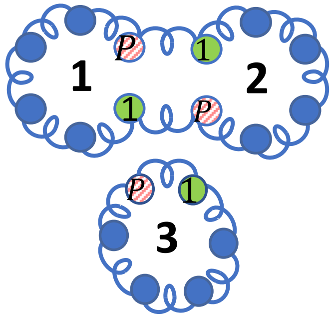
Weight of a configuration:

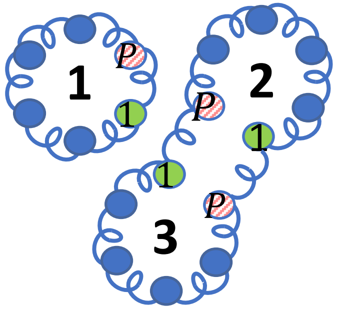
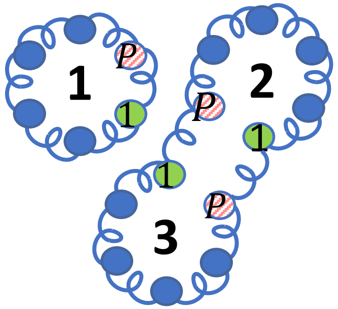
$\Pr[$  $] \propto \exp(-\beta \cdot E($  $))$

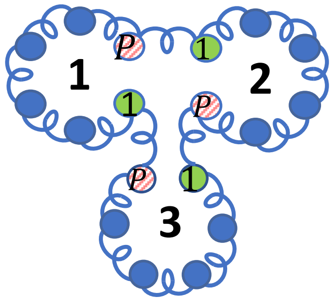
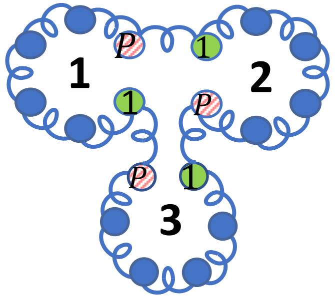
 q_1

$+ \Pr[$ $] \cdot -\nabla_{q_1^j}($ $)$

$-\nabla_{q_1^j} V^{[1,N]} = \Pr[$  $] \cdot -\nabla_{q_1^j}($  $)$

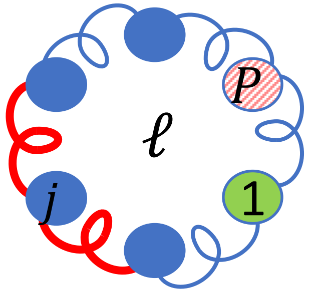
$+ \Pr[$  $] \cdot -\nabla_{q_1^j}($  $)$

$+ \Pr[$  $] \cdot -\nabla_{q_1^j}($  $)$

$+ \Pr[$  $] \cdot -\nabla_{q_1^j}($  $)$

Evaluating the Forces

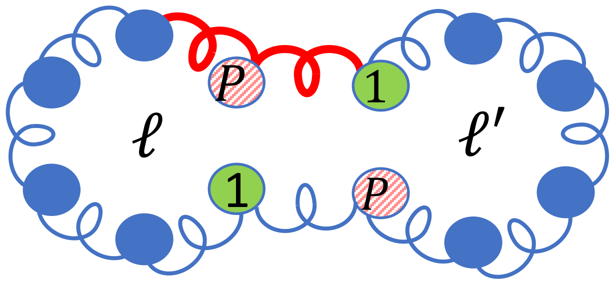
$$-\nabla_{q_\ell^j} V^{[1,N]} = -\nabla_{q_\ell^j} \left(\quad \right)$$



$(j \neq 1, P)$

$\boldsymbol{\mathcal{O}(PN)}$ to evaluate for $N(P-2)$ interior beads

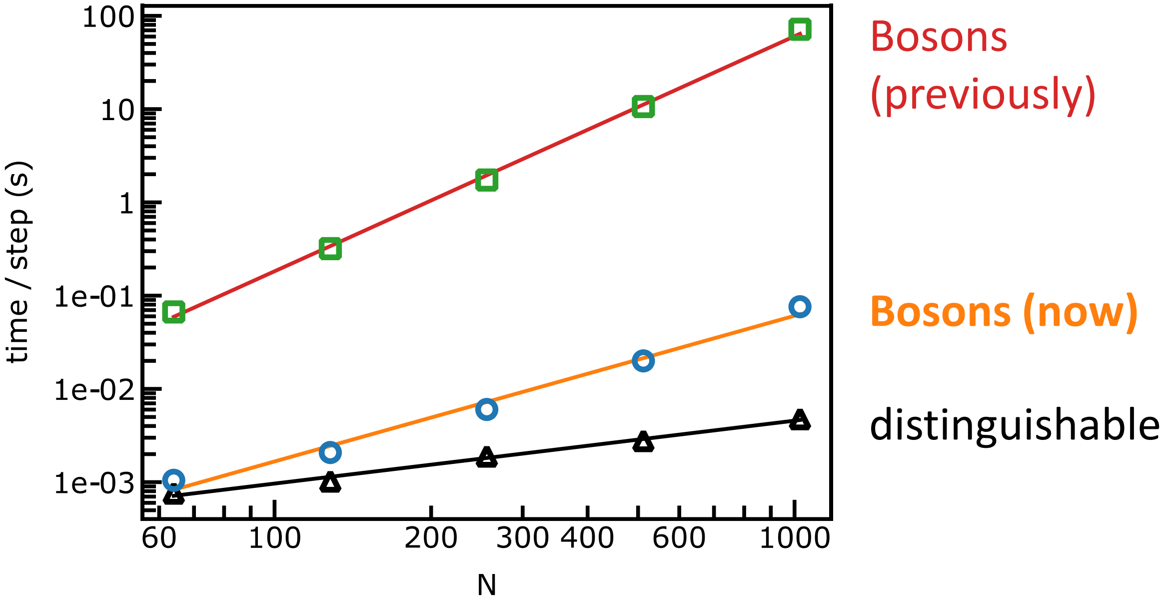
$$-\nabla_{q_\ell^P} V^{[1,N]} = \sum_{\ell'=1}^{N} \Pr[\ell \to \ell'] \cdot -\nabla_{q_\ell^P} \left(\quad \right)$$



$\boldsymbol{\mathcal{O}(N^2)}$ to evaluate for $2N$ exterior beads

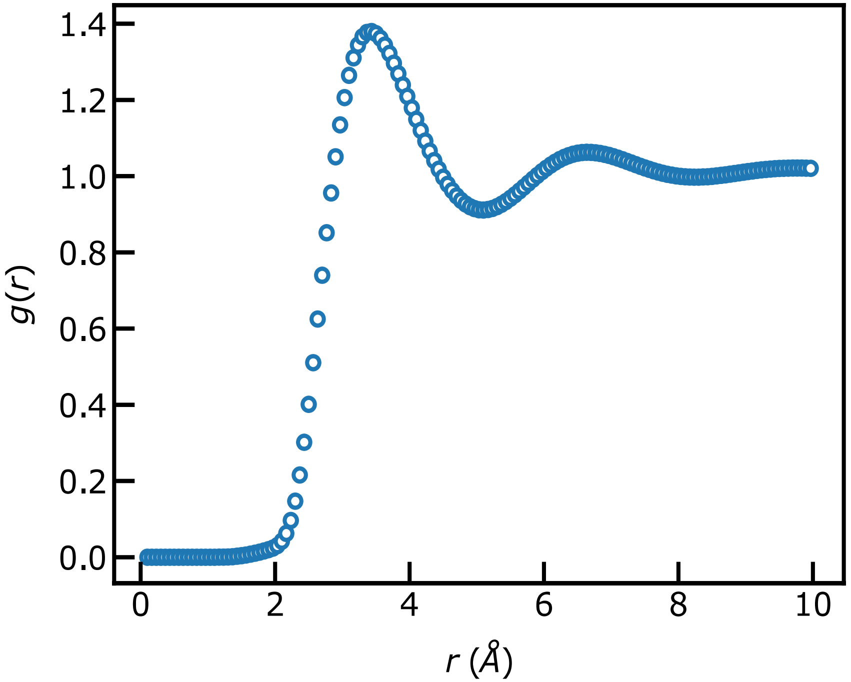
Feldman and Hirshberg. JCP 2023

Conclusion



- Bosonic path integral molecular dynamics in quadratic time
- Same potential, forces & trajectories – only faster

- > 1000 bosons in days rather than decades
- Eliminates most of the overhead of bosonic exchange in PIMD



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