

# Quadratic Scaling Bosonic Path Integral Molecular Dynamics

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Molecular simulations



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# Bosonic Exchange

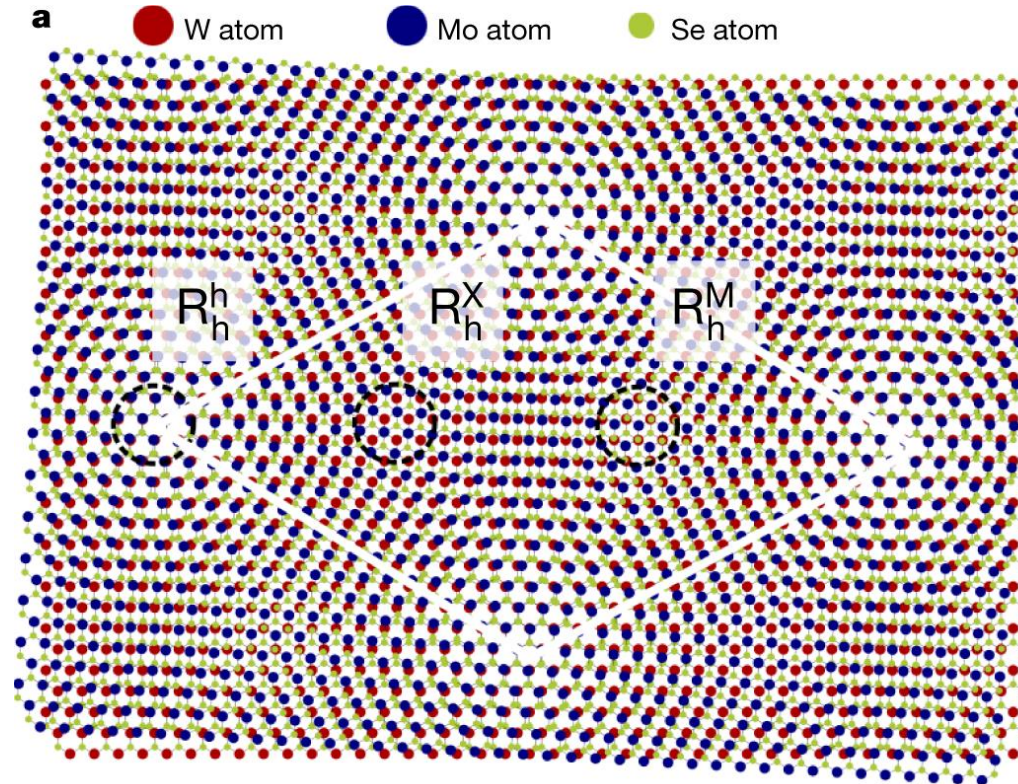
- Quantum symmetry is a fundamental property of quantum particles.
- Exchange effects at finite T are important for many systems

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

# Bosonic Exchange Effects



Netanel  
Bachar Schwartz

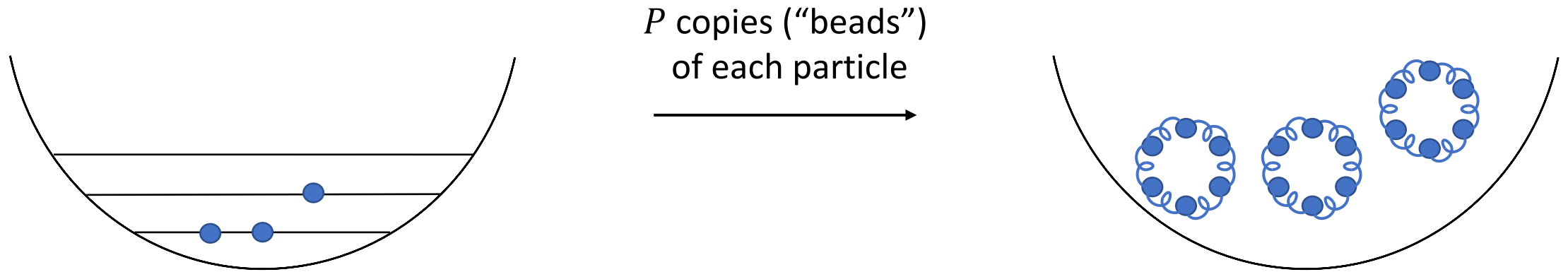


Wang et al. Nature 574, 76-80 (2019)

Tran, Moody et al. Nature 567, 71–75 (2019)



# Path-Integral Molecular Dynamics



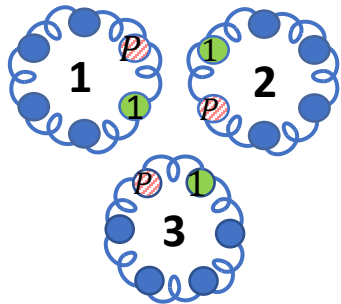
$$Z = \text{Tr}[e^{-\beta\hat{H}}] \sim \int \left( e^{-\beta E^{(1)(2)(3)}} \right) d\mathbf{q}_1 d\mathbf{q}_2 d\mathbf{q}_3$$

$$E^{(1)(2)(3)} \quad \text{1} \quad \text{2} \quad \text{3}$$

# Bosonic Path-Integral Molecular Dynamics

# Bosonic Path-Integral Molecular Dynamics

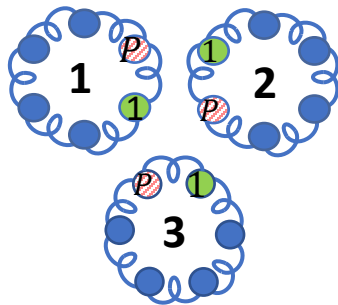
$$Z \sim \frac{1}{6} \int \left( e^{-\beta E^{(1)(2)(3)}} \right) d\mathbf{q}_1 d\mathbf{q}_2 d\mathbf{q}_3$$



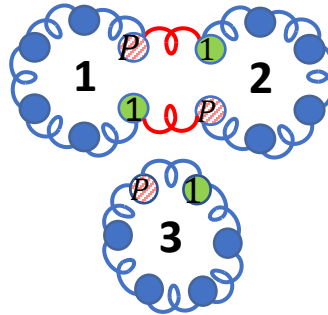
$E^{(1)(2)(3)}$

# Bosonic Path-Integral Molecular Dynamics

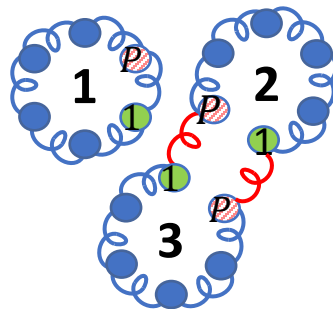
$$Z \sim \frac{1}{6} \int \left( e^{-\beta E^{(1)(2)(3)}} + e^{-\beta E^{(12)(3)}} + e^{-\beta E^{(1)(23)}} + e^{-\beta E^{(13)(2)}} \right) d\mathbf{q}_1 d\mathbf{q}_2 d\mathbf{q}_3$$



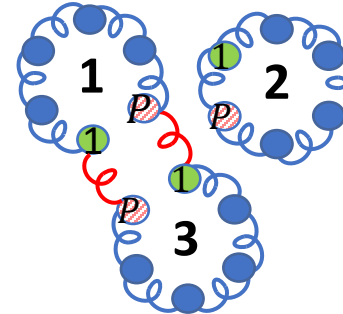
$E^{(1)(2)(3)}$



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



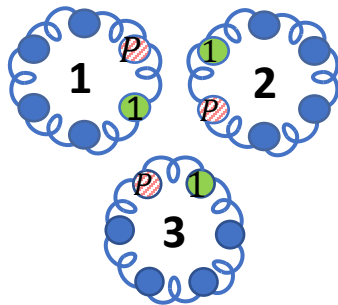
$E^{(1)(23)}$



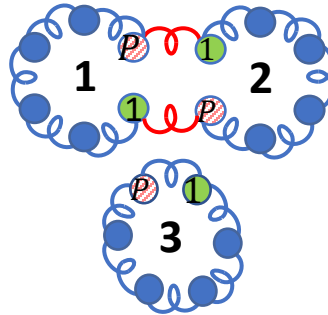
$E^{(13)(2)}$

# Bosonic Path-Integral Molecular Dynamics

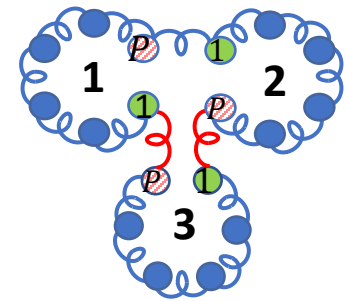
$$Z \sim \frac{1}{6} \int \left( e^{-\beta E^{(1)(2)(3)}} + e^{-\beta E^{(12)(3)}} + e^{-\beta E^{(1)(23)}} + e^{-\beta E^{(13)(2)}} \right. \\ \left. + e^{-\beta E^{(123)}} + e^{-\beta E^{(132)}} \right) d\mathbf{q}_1 d\mathbf{q}_2 d\mathbf{q}_3$$



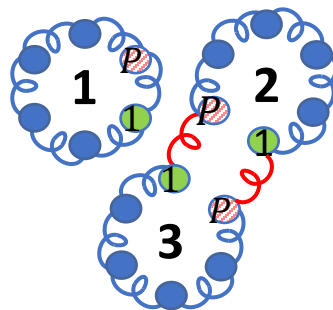
$E^{(1)(2)(3)}$



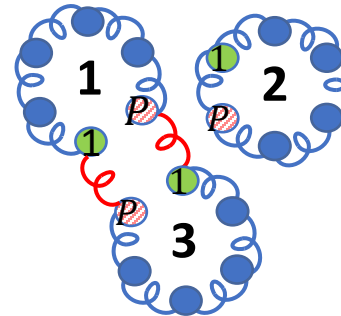
$E^{(12)(3)}$



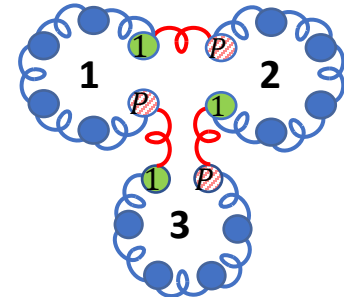
$E^{(123)}$



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



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

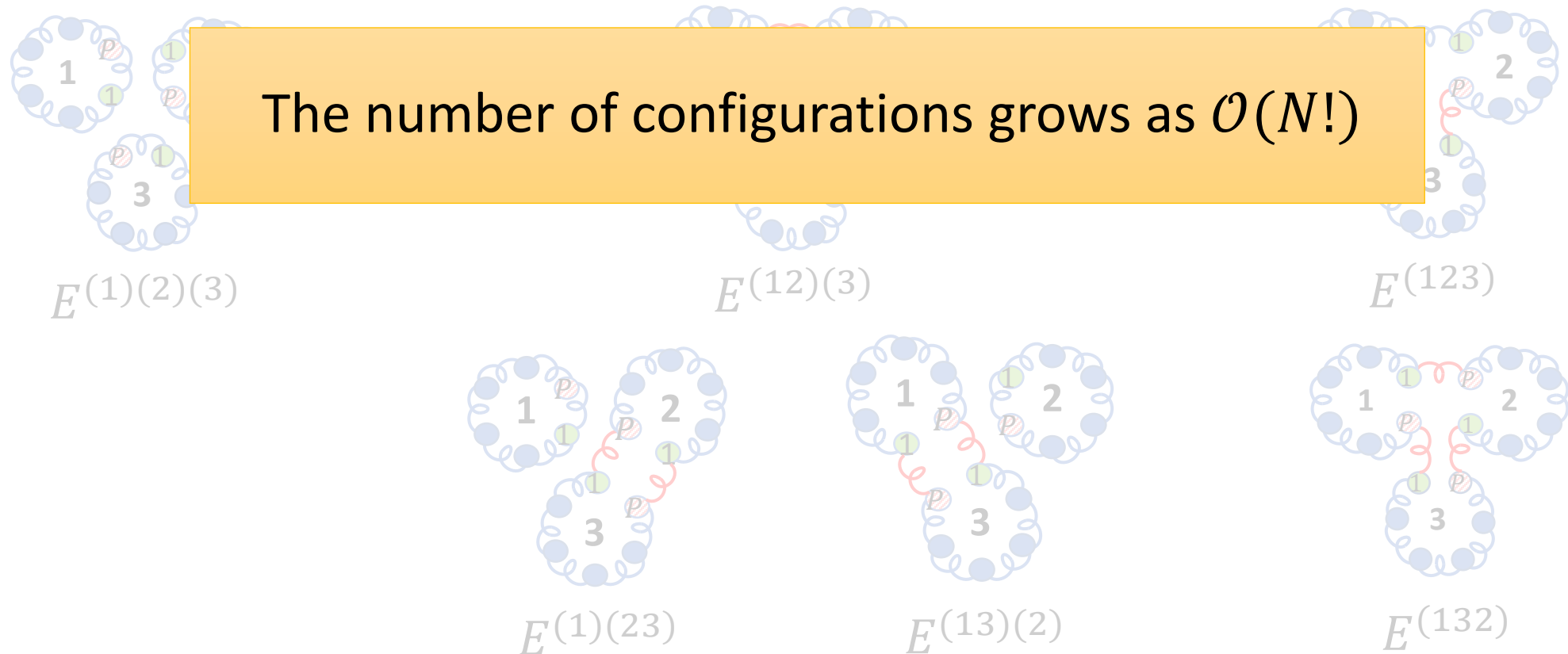


$E^{(132)}$



# Bosonic Path-Integral Molecular Dynamics

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



# Scaling of Bosonic Path-Integral MD

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

All permutations:

$$\mathcal{O}(PN!)$$

Hirshberg et al.:

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

← ~ 100 bosons

**This work:**

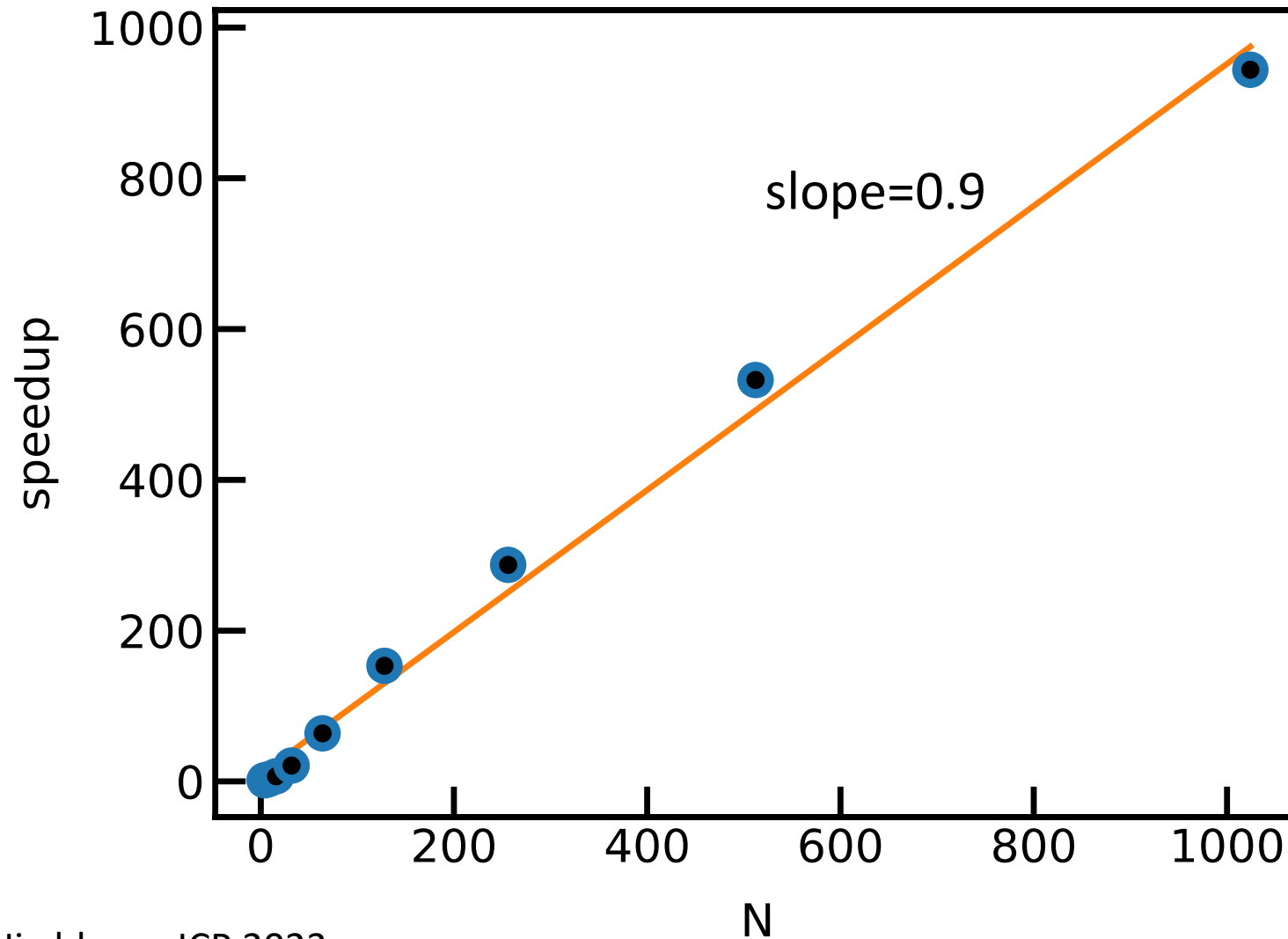
$$\mathcal{O}(N^2 + PN)$$

← > 1000 bosons

Hirshberg et al. PNAS 2019

Feldman and Hirshberg. JCP 2023

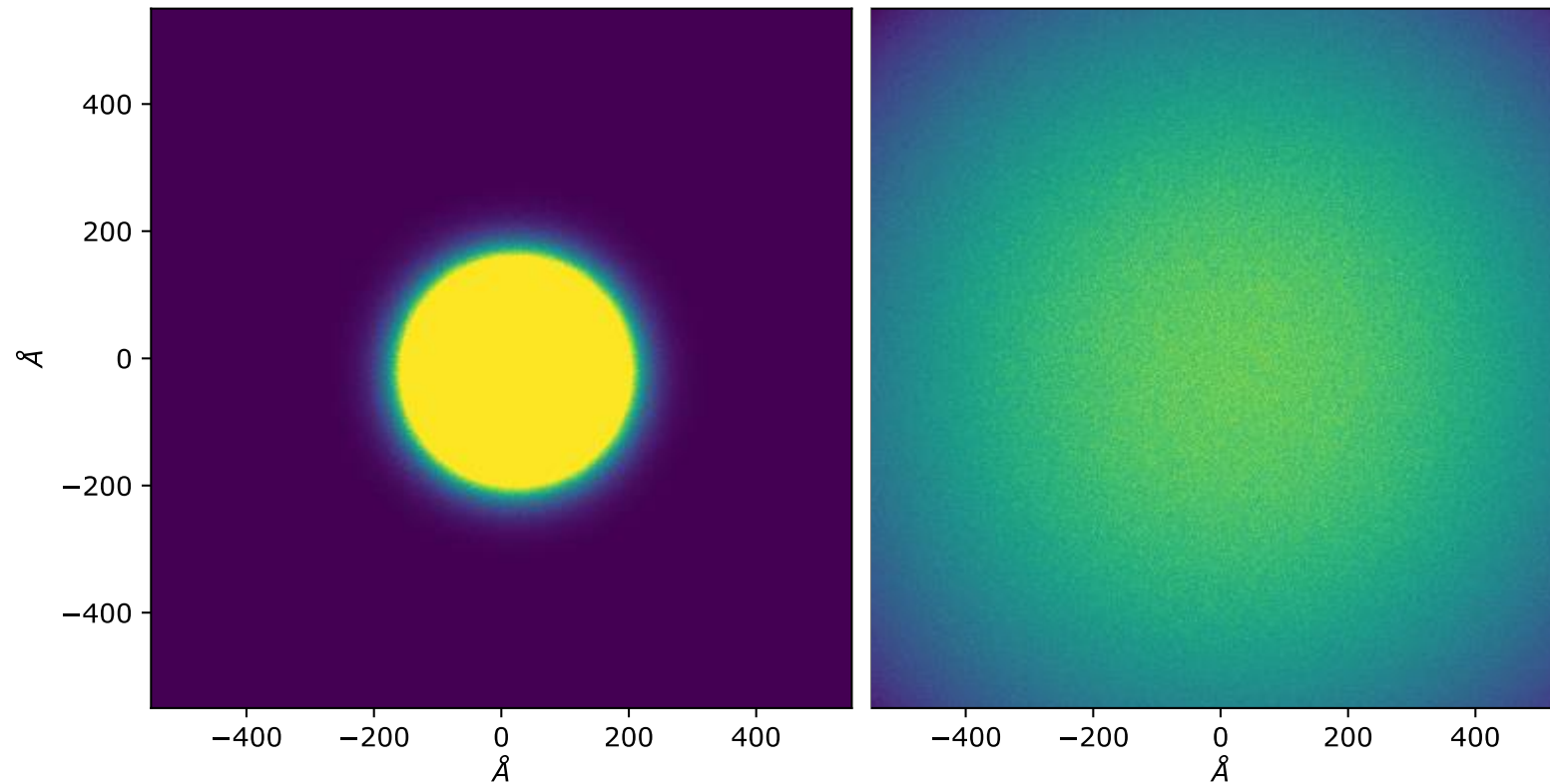
# Speedup $\mathcal{O}(PN)$



# 1600 Particles

9 days of simulation rather than  $> 20$  years

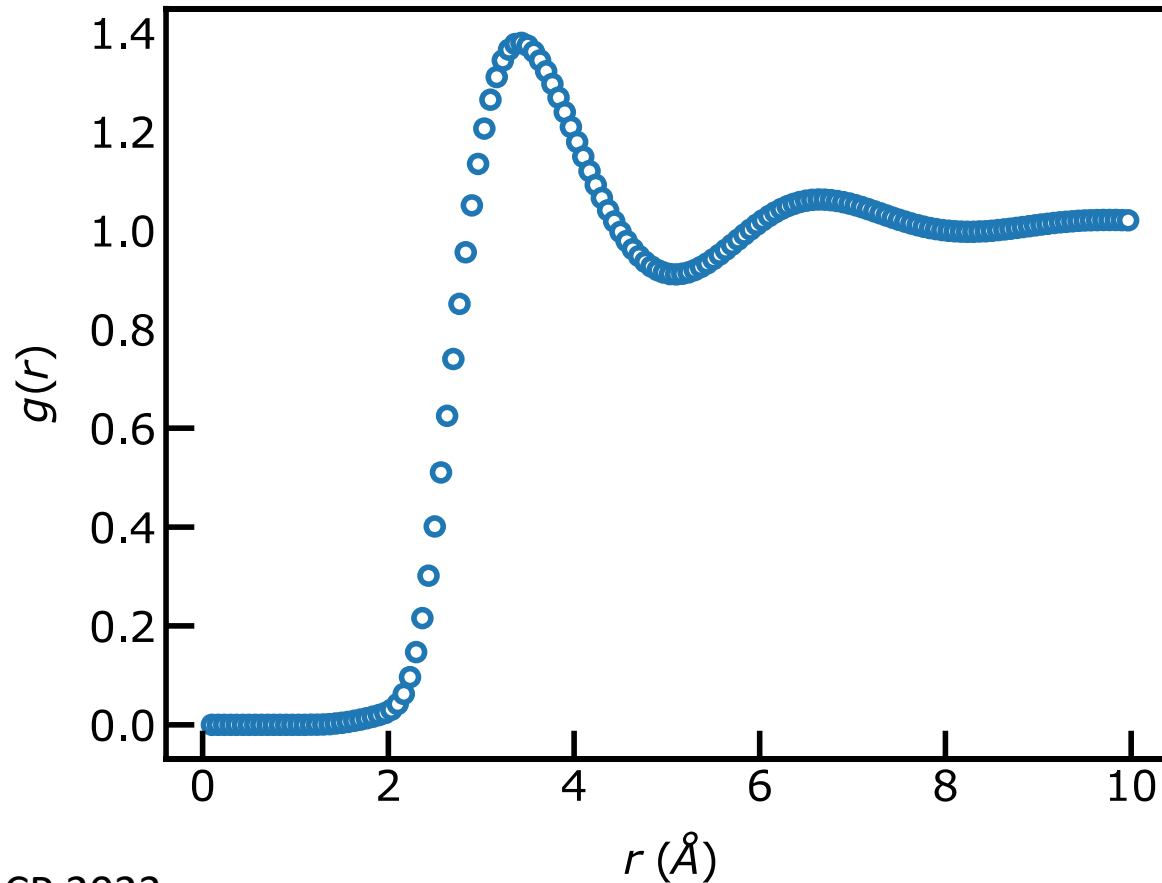
2D density of  $N = 1600$  trapped bosons  
without and with Gaussian repulsive interaction



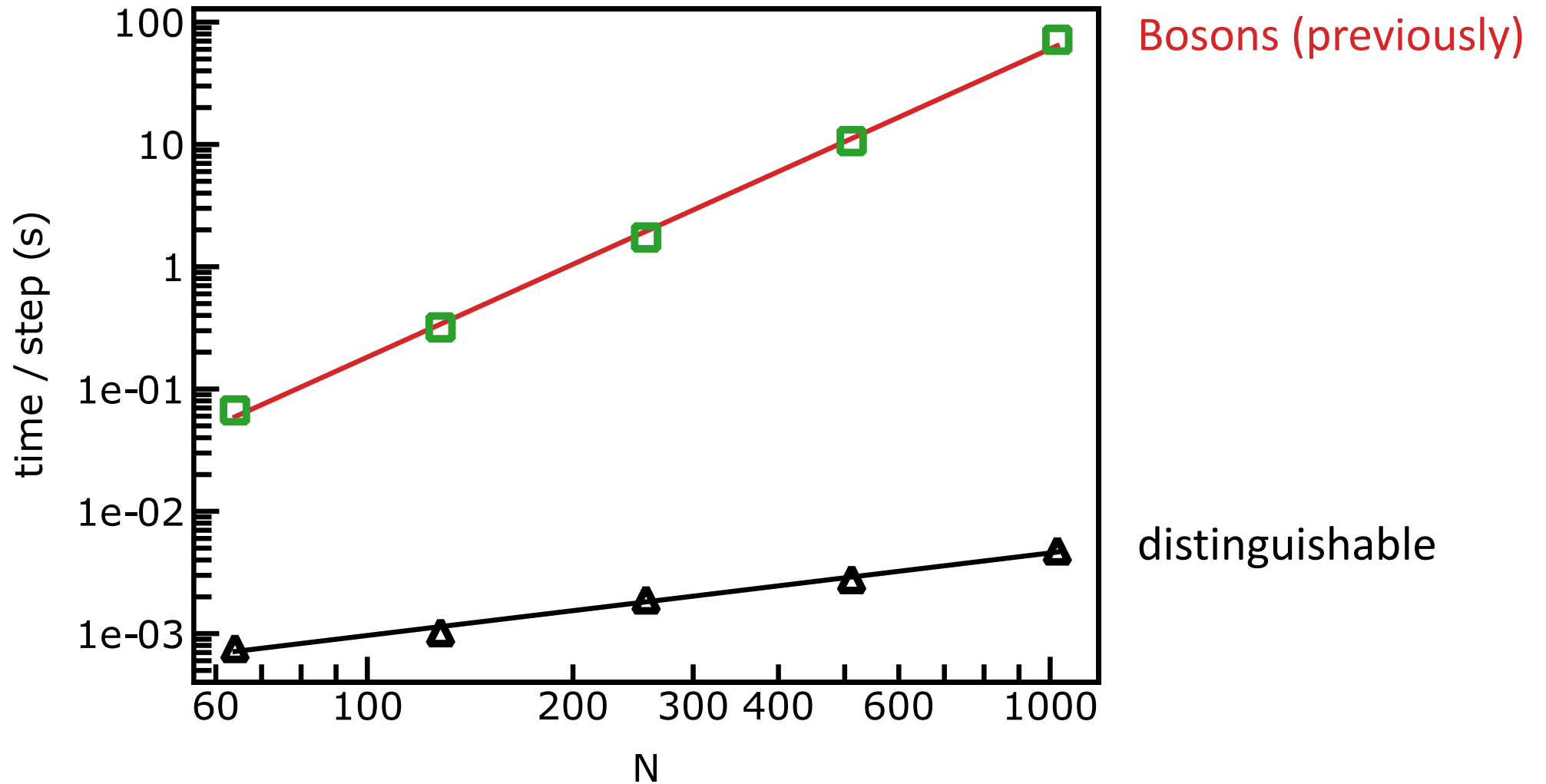
# 1372 Particles

7 days of simulation rather than  $> 14$  years

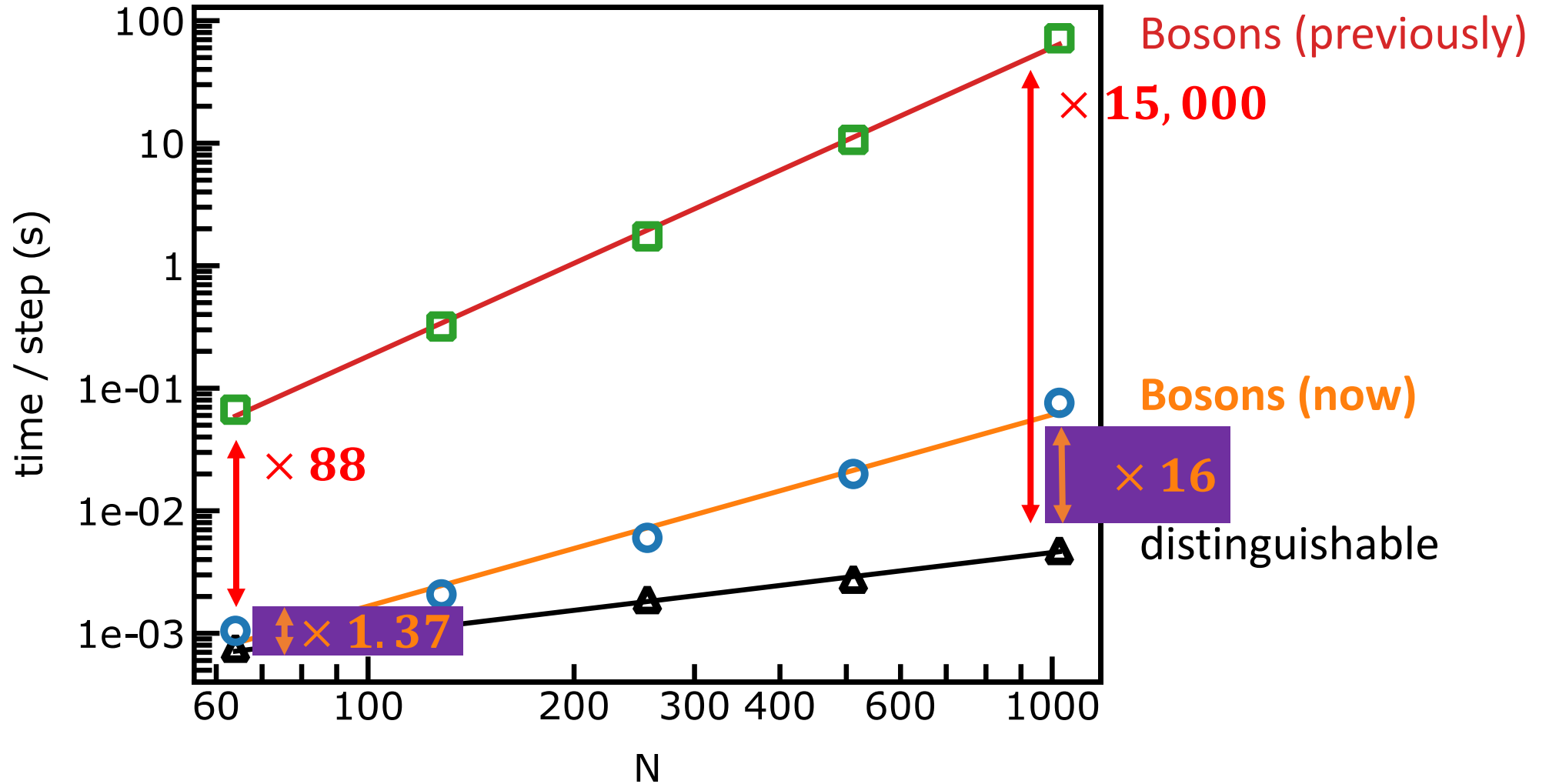
Pair correlation of  $N = 1372$   $^4\text{He}$  atoms



# The Cost of Bosonic Exchange



# The Cost of Bosonic Exchange



# Hirshberg et al.'s Algorithm

Potential:

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

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

Forces:

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

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



# Our Algorithm

Potential:

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

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

Forces:

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

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

$$\begin{aligned}
-\nabla_{q_1^j} V^{[1,N]} = & \Pr[ \text{Diagram 1} ] \cdot -\nabla_{q_1^j} ( \text{Diagram 2} ) \\
& + \Pr[ \text{Diagram 3} ] \cdot -\nabla_{q_1^j} ( \text{Diagram 4} ) \\
& + \Pr[ \text{Diagram 5} ] \cdot -\nabla_{q_1^j} ( \text{Diagram 6} ) \\
& + \Pr[ \text{Diagram 7} ] \cdot -\nabla_{q_1^j} ( \text{Diagram 8} )
\end{aligned}$$

The diagrams consist of three clusters of blue particles labeled 1, 2, and 3. Each cluster contains a green particle labeled '1' and a red particle labeled 'p'.
   
 - Diagram 1: Three separate circular clusters. Cluster 1 has '1' and 'p' on the right. Cluster 2 has '1' and 'p' on the left. Cluster 3 has '1' and 'p' on the left.
   
 - Diagram 2: Three separate circular clusters. Cluster 1 has '1' and 'p' on the right. Cluster 2 has '1' and 'p' on the left. Cluster 3 has '1' and 'p' on the left.
   
 - Diagram 3: Clusters 1 and 2 are connected by a horizontal bridge. Cluster 3 is separate below. Cluster 1 has '1' and 'p' on the right. Cluster 2 has '1' and 'p' on the left. Cluster 3 has '1' and 'p' on the left.
   
 - Diagram 4: Clusters 1 and 2 are connected by a diagonal bridge. Cluster 3 is separate below. Cluster 1 has '1' and 'p' on the right. Cluster 2 has '1' and 'p' on the left. Cluster 3 has '1' and 'p' on the left.
   
 - Diagram 5: Clusters 1 and 2 are connected by a vertical bridge. Cluster 3 is separate below. Cluster 1 has '1' and 'p' on the right. Cluster 2 has '1' and 'p' on the left. Cluster 3 has '1' and 'p' on the left.
   
 - Diagram 6: Clusters 1 and 2 are connected by a horizontal bridge. Cluster 3 is separate below. Cluster 1 has '1' and 'p' on the right. Cluster 2 has '1' and 'p' on the left. Cluster 3 has '1' and 'p' on the left.

$$-\nabla_{q_1^j} V^{[1,N]} = \Pr[ \text{Diagram 1} ] \cdot -\nabla_{q_1^j} ( \text{Diagram 2} )$$

**Weight of a configuration:**

$$\Pr[ \text{Diagram 1} ] \propto \exp(-\beta \cdot E( \text{Diagram 2} ))$$

$$+ \Pr[ \text{Diagram 3} ] \cdot -\nabla_{q_1^j} ( \text{Diagram 4} )$$

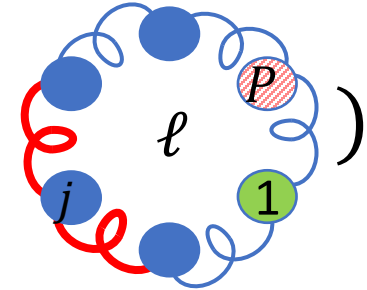
$$\begin{aligned}
-\nabla_{q_1^j} V^{[1,N]} = & \Pr[ \text{Diagram 1} ] \cdot -\nabla_{q_1^j} ( \text{Diagram 2} ) \\
& + \Pr[ \text{Diagram 3} ] \cdot -\nabla_{q_1^j} ( \text{Diagram 4} ) \\
& + \Pr[ \text{Diagram 5} ] \cdot -\nabla_{q_1^j} ( \text{Diagram 6} ) \\
& + \Pr[ \text{Diagram 7} ] \cdot -\nabla_{q_1^j} ( \text{Diagram 8} )
\end{aligned}$$

The diagrams consist of three clusters of blue particles, labeled 1, 2, and 3. Each cluster contains a green particle labeled '1' and a red particle labeled 'p'.
   
 - **Diagram 1:** Three separate circular clusters. Cluster 1 has '1' at the top and 'p' at the bottom. Cluster 2 has '1' at the top and 'p' at the bottom. Cluster 3 has '1' at the top and 'p' at the bottom.
   
 - **Diagram 2:** Three separate circular clusters. Cluster 1 has '1' at the top and 'p' at the bottom. Cluster 2 has '1' at the top and 'p' at the bottom. Cluster 3 has '1' at the top and 'p' at the bottom.
   
 - **Diagram 3:** Clusters 1 and 2 are connected by a horizontal bridge. Cluster 3 is separate below. Cluster 1 has '1' at the top and 'p' at the bottom. Cluster 2 has '1' at the top and 'p' at the bottom. Cluster 3 has '1' at the top and 'p' at the bottom.
   
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 - **Diagram 5:** Clusters 1 and 2 are connected by a diagonal bridge. Cluster 3 is separate below. Cluster 1 has '1' at the top and 'p' at the bottom. Cluster 2 has '1' at the top and 'p' at the bottom. Cluster 3 has '1' at the top and 'p' at the bottom.
   
 - **Diagram 6:** Clusters 1 and 2 are connected by a diagonal bridge. Cluster 3 is separate below. Cluster 1 has '1' at the top and 'p' at the bottom. Cluster 2 has '1' at the top and 'p' at the bottom. Cluster 3 has '1' at the top and 'p' at the bottom.
   
 - **Diagram 7:** Clusters 1 and 2 are connected by a horizontal bridge. Cluster 3 is connected to the bridge by a vertical line. Cluster 1 has '1' at the top and 'p' at the bottom. Cluster 2 has '1' at the top and 'p' at the bottom. Cluster 3 has '1' at the top and 'p' at the bottom.
   
 - **Diagram 8:** Clusters 1 and 2 are connected by a horizontal bridge. Cluster 3 is connected to the bridge by a vertical line. Cluster 1 has '1' at the top and 'p' at the bottom. Cluster 2 has '1' at the top and 'p' at the bottom. Cluster 3 has '1' at the top and 'p' at the bottom.

# Evaluating the Forces

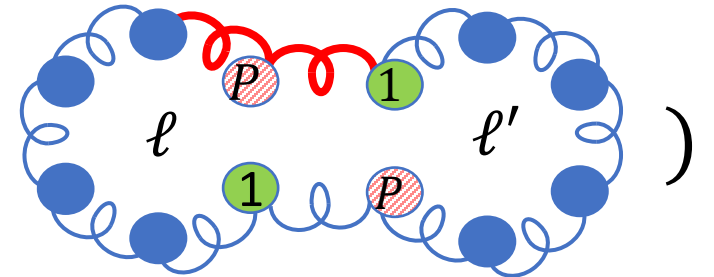
$$-\nabla_{q_\ell^j} V[1, N] = -\nabla_{q_\ell^j} \left( \text{Diagram} \right)$$

( $j \neq 1, P$ )



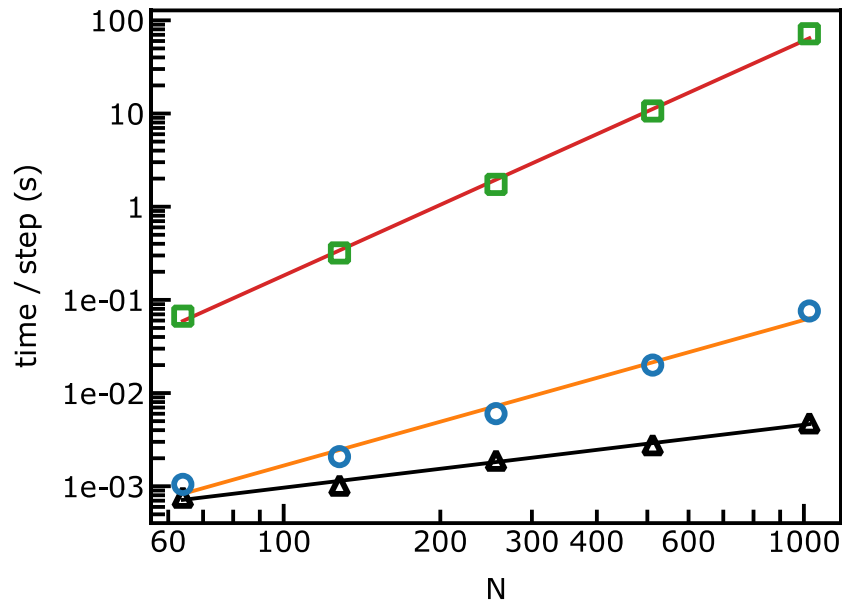
$\mathcal{O}(PN)$  to evaluate for  $N(P - 2)$  interior beads

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



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

# Conclusion



Bosons  
(previously)

Bosons (now)

distinguishable

- 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

