COMPUTATIONAL PREDICTION OF THE STRUCTURE OF COULOMB EXPLODED MOLECULES

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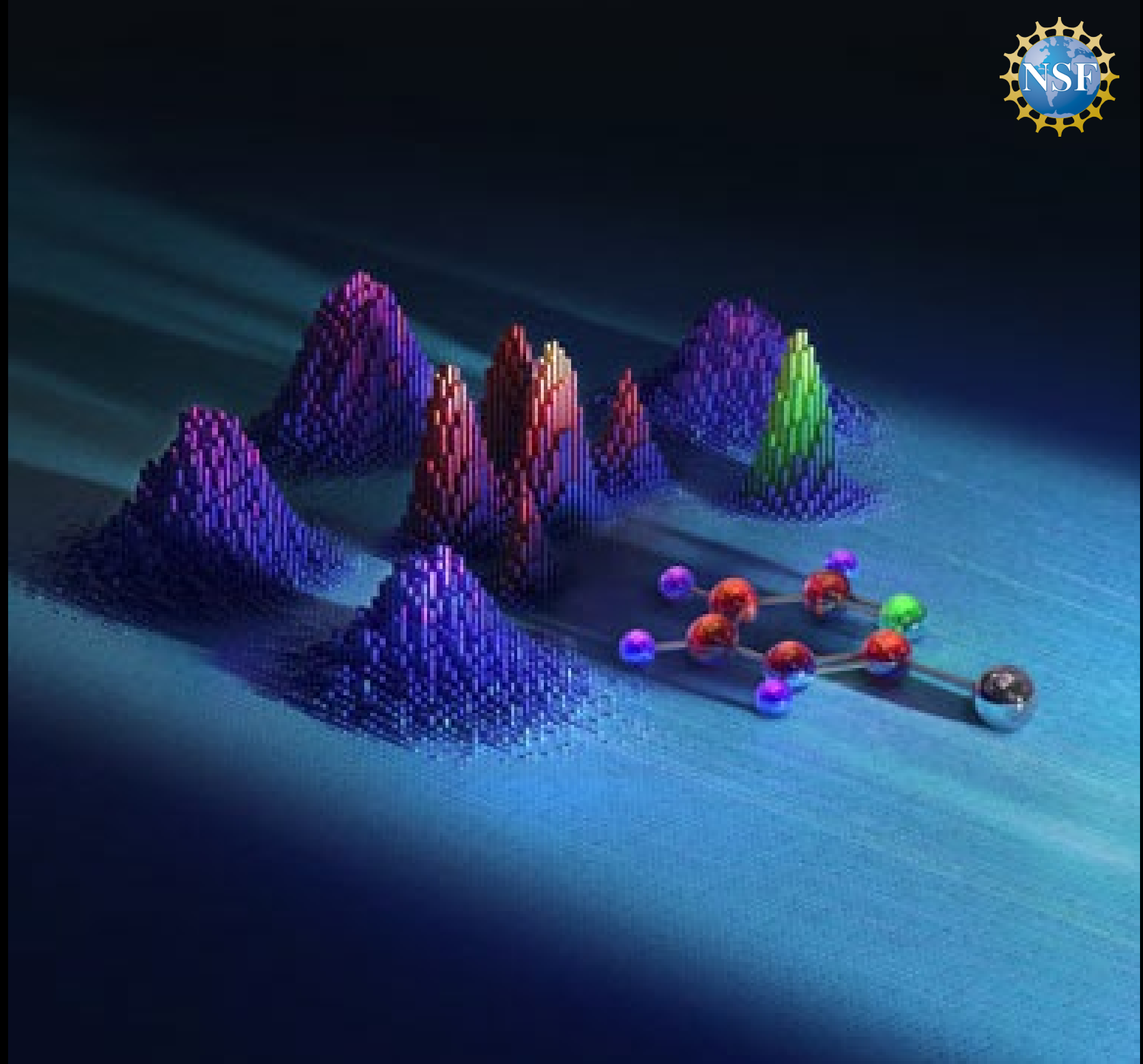


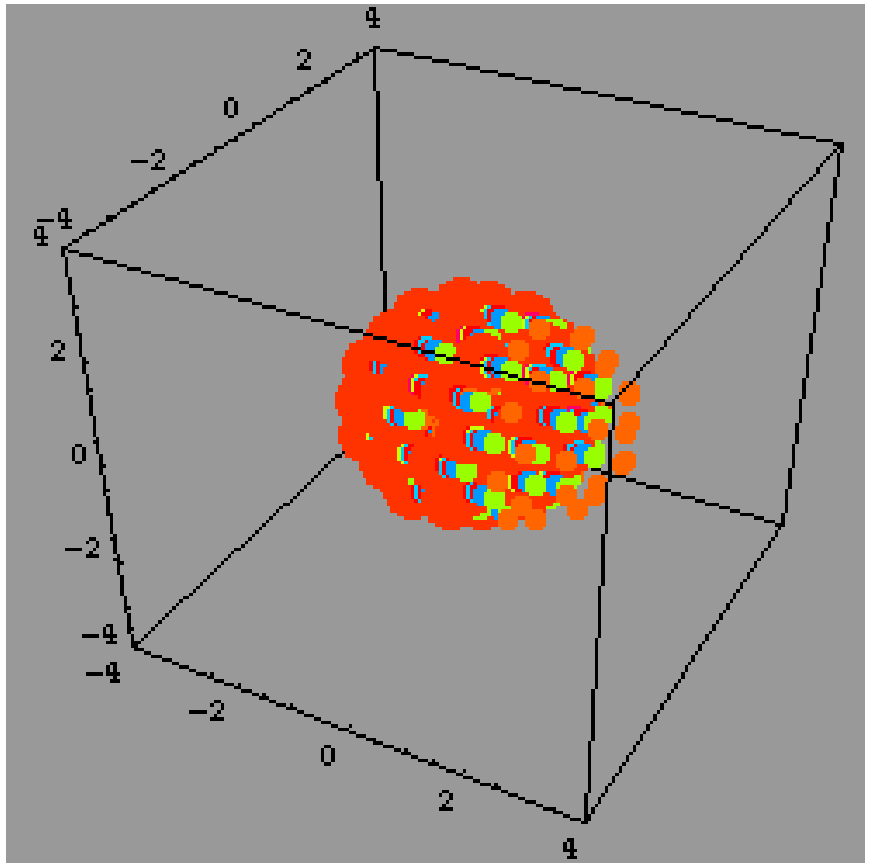
COULOMB EXPLOSION IMAGING (CEI)

CEI is performed by:

- Femtosecond X-ray free-electron (XFEL) and strong IR lasers
- Charge up of molecules through sequential multiphoton ionization
- Explosion of charged fragments!
- Final momenta of fragments recorded

- Want to understand the molecular dynamics during a pump-probe experiment ("molecular movie")

- *Current:* Geometry determined through manual analysis and indirect path from experimental data

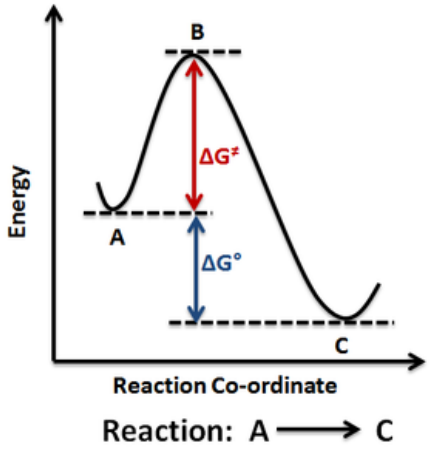


X. Li *et. al.*, Phys. Rev. Research **4** (2022)
https://doi.org/10.1103/PhysRevResearch.4.013029

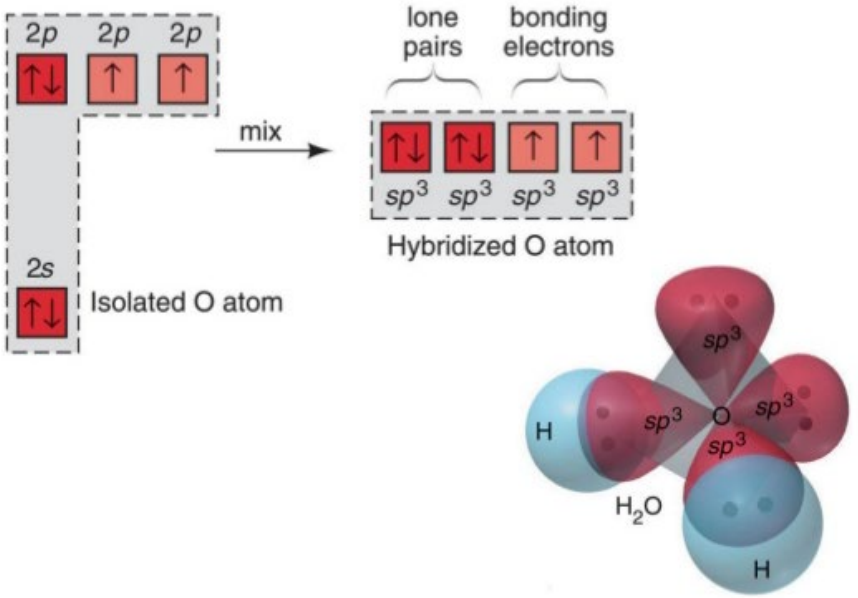
DESCRIPTION OF MOLECULES

How do we get the structure of fast-moving gas molecules that react at the attosecond timescale?

- One possible energetic pathway



- Bonds and molecular orbitals



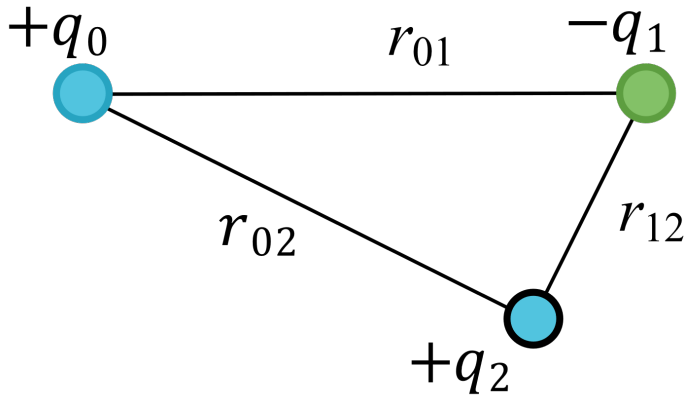
- Molecular Schrödinger equation for an *N*-atom molecule

$$\hat{H}^{ele}(\vec{R}_1, \vec{R}_2, ..., \vec{R}_N)\Psi^{ele}(\vec{R}, \vec{r_1}, \vec{r_2}, ..., \vec{r_n}) = E^{ele}\Psi^{ele}$$

NEWTONIAN DYNAMICS

$$F_i = m_i \ddot{x} = \sum_j \frac{q_i q_j}{r_{ij}^3} \boldsymbol{r}_{ij}$$

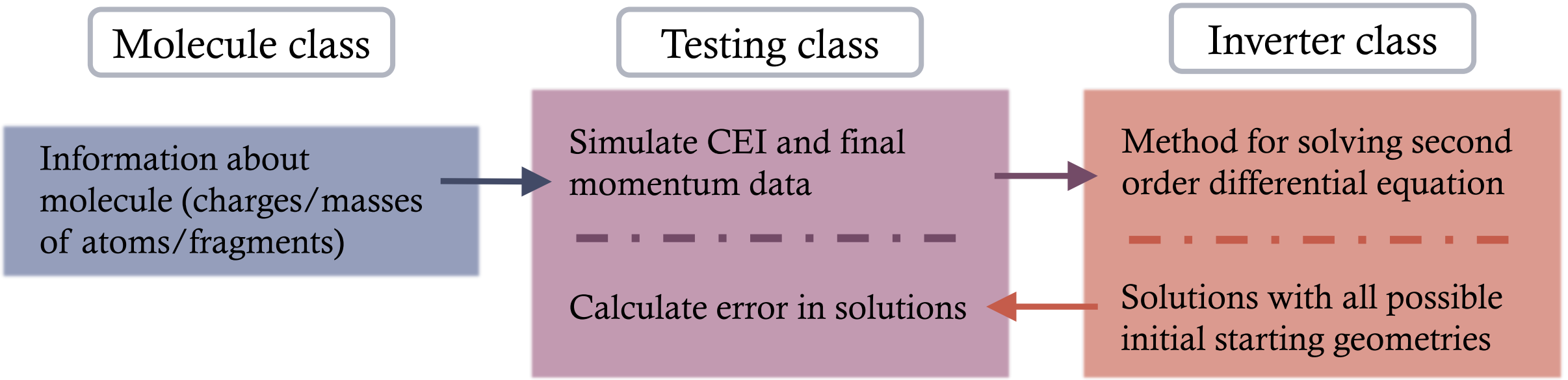
- Can use standard integration methods to solve the second order differential equations
- Initial conditions: Positions and Momenta



DEVELOPMENT OF CODE

$$F_i = m_i\ddot{x} = \sum_j \frac{q_i q_j}{r_{ij}^3}\boldsymbol{r}_{ij}$$

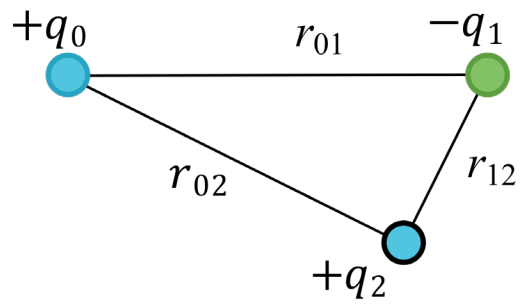
Goal: Predict molecular geometries directly from final momentum data obtained from CEI



INVERSION METHOD

$$F_i = m_i\ddot{x} = \sum_j \frac{q_i q_j}{r_{ij}^3}\boldsymbol{r}_{ij}$$

- From CEI we only have final momenta (no starting position or momenta)
- Determine the final kinetic energy
- Partition the kinetic energy into the degrees of freedom of the initial conditions



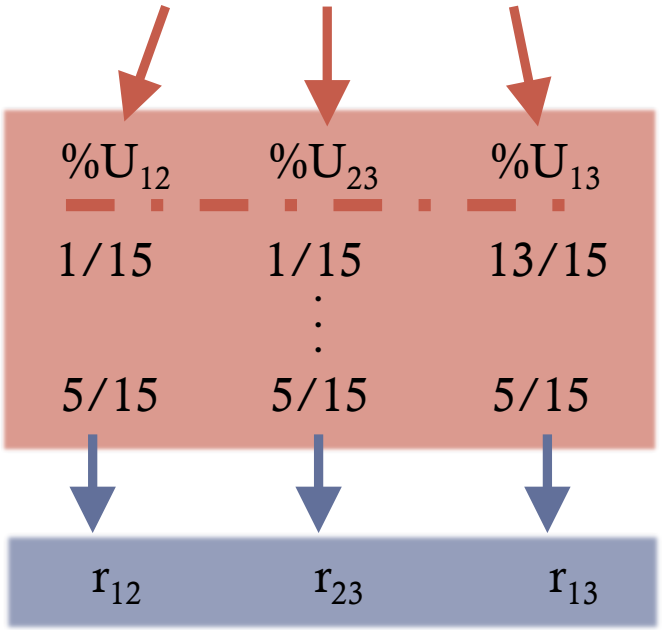
(Simulated) CEI

Final momenta

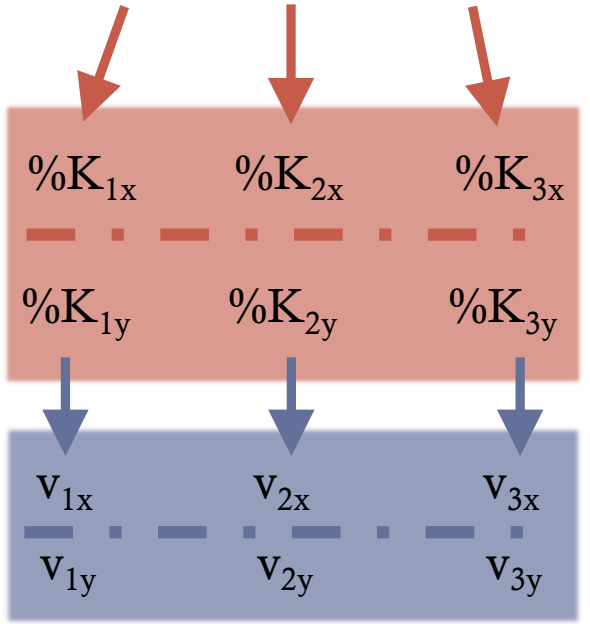
Final kinetic energy
100%

Partition final into initial energy

$$U = k\frac{q_i q_j}{r_{ij}}$$

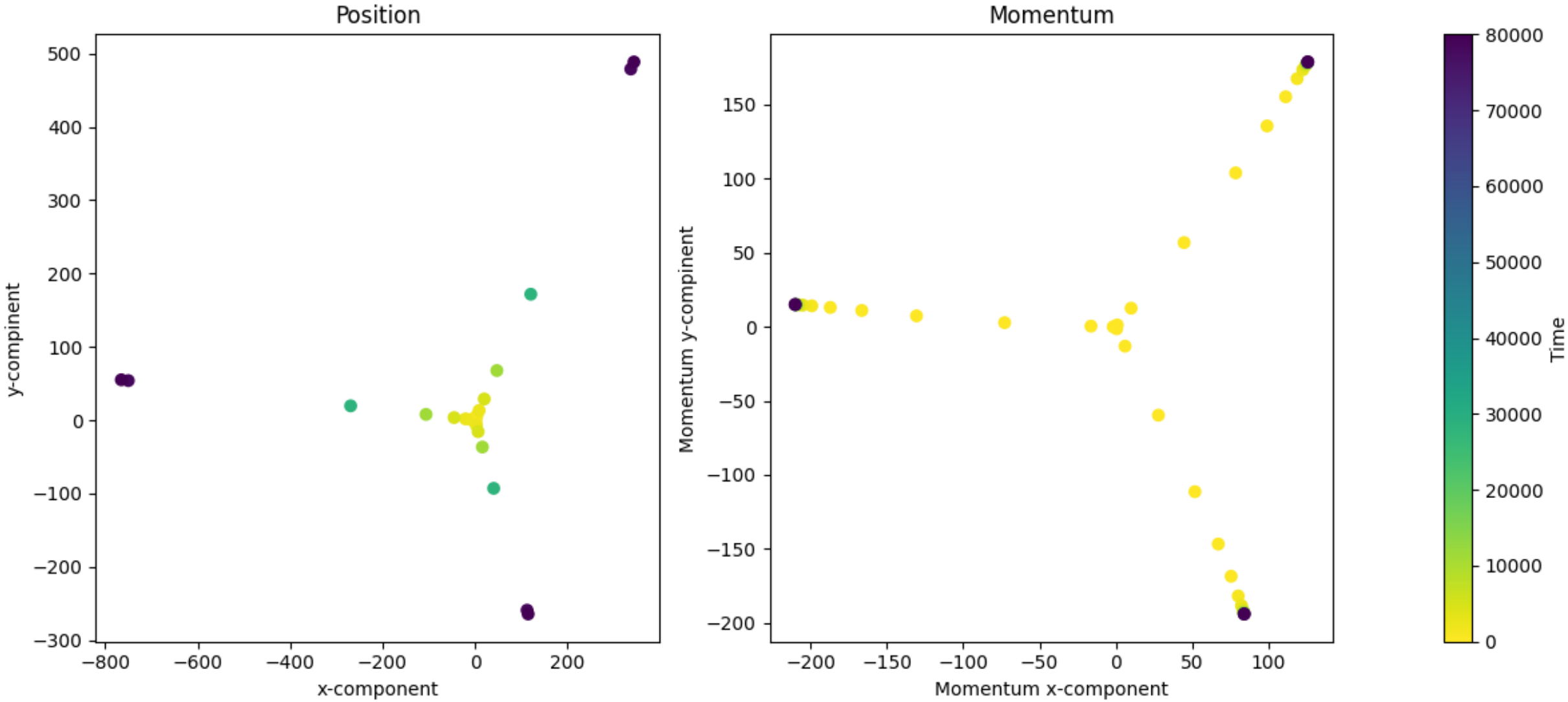


$$K = \frac{1}{2}mv^2$$



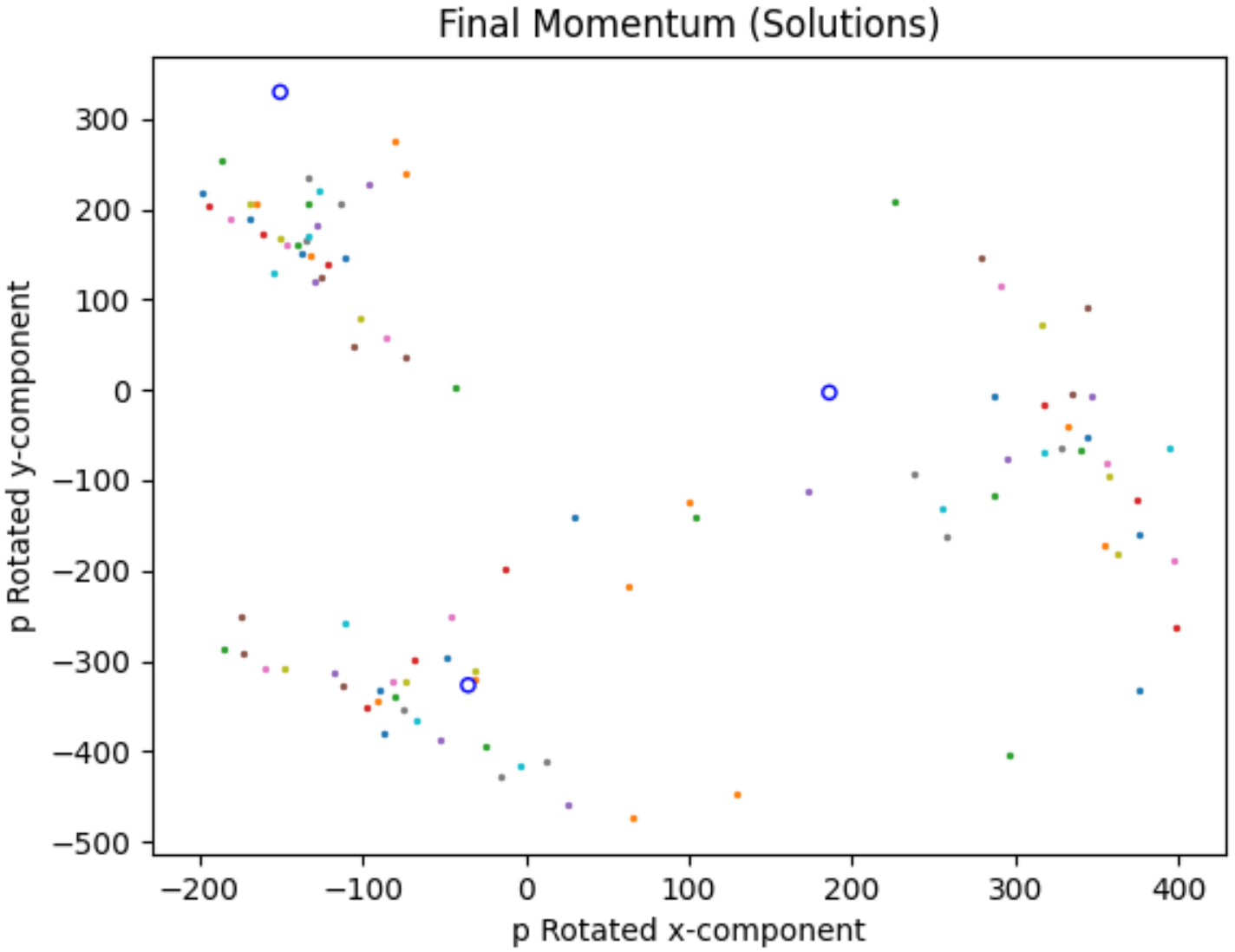
SIMULATIONS (O+C+S+ CHANNEL)

$$O = C = S$$



INVERTER SOLUTIONS

$$O = C = S$$



Assumptions:

No initial KE

→ No KE partition terms

Partitions:

n = 15 → 91 solutions

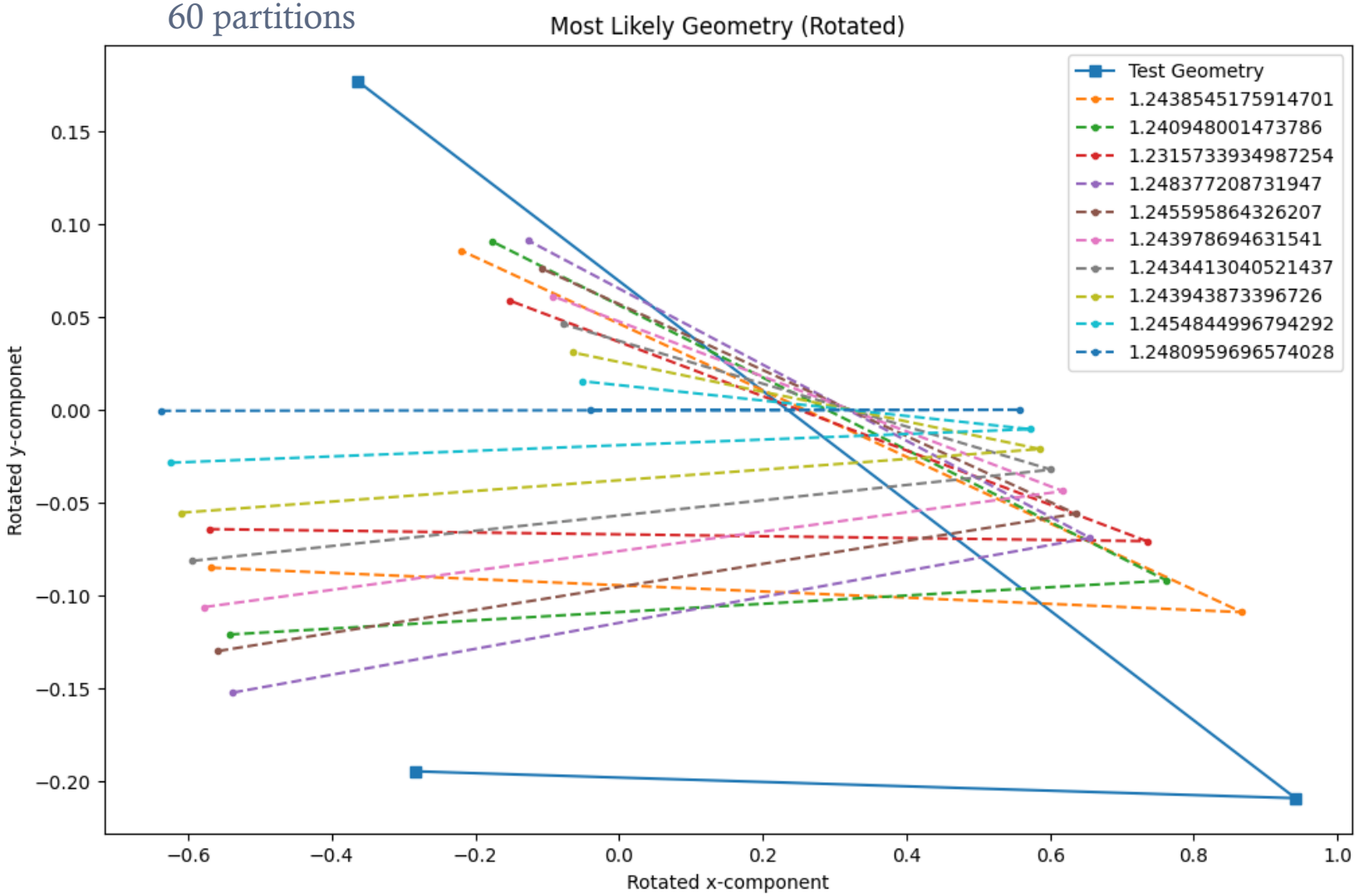
n = 30 → 406 solutions

n = 60 → 1711 solutions

SIMULATION GEOMETRY

$$O = C = S$$

60 partitions

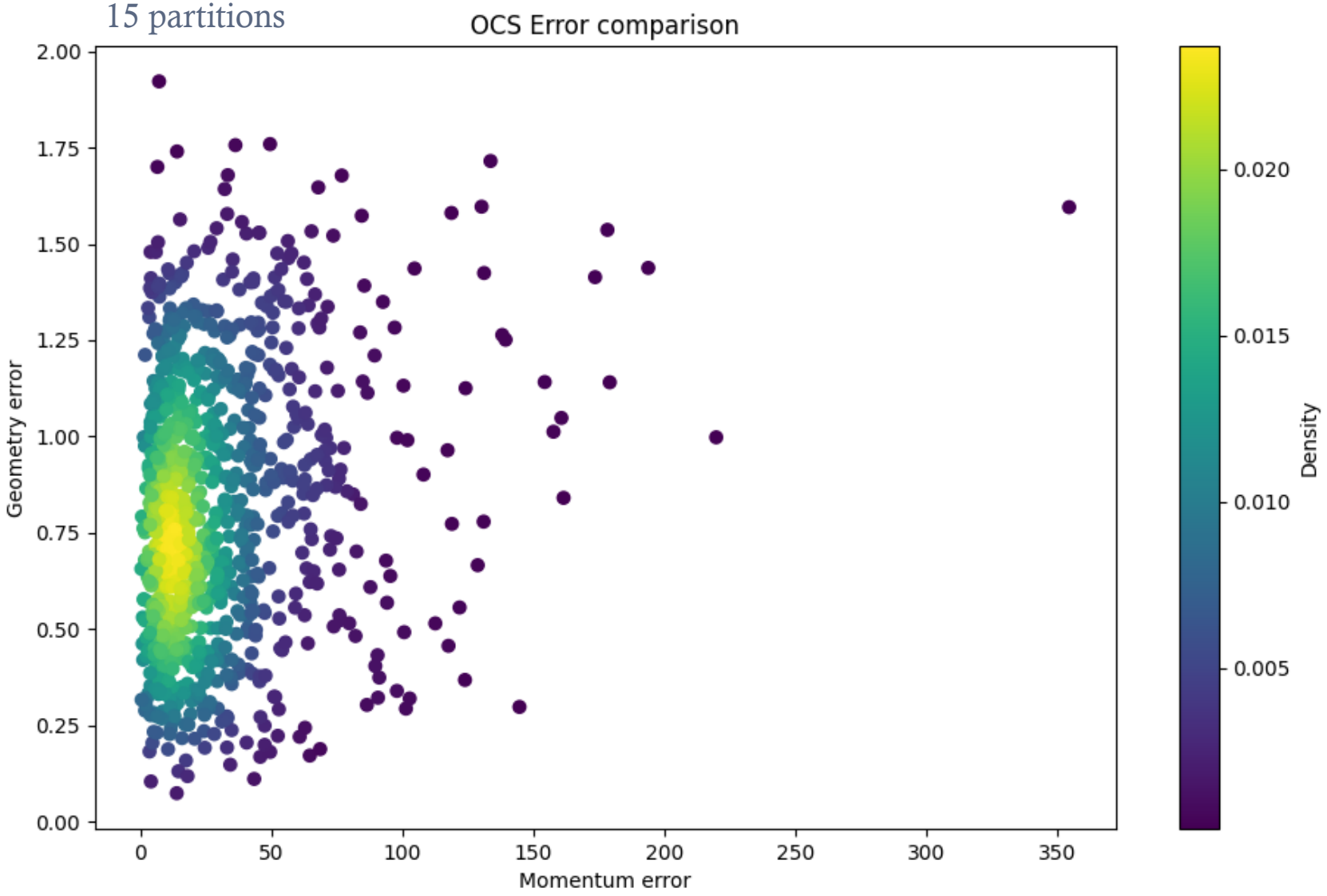


Partitions	Non-normalized RMS error in the geometry
15	1.244
60	1.232

ERROR COMPARISON

$O = C = S$

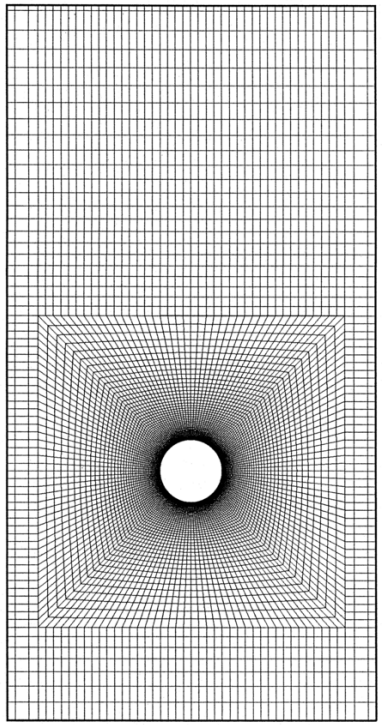
15 partitions



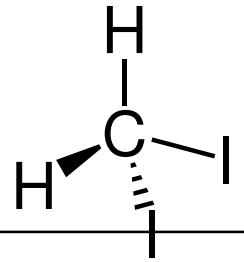
1000 simulations of OCS

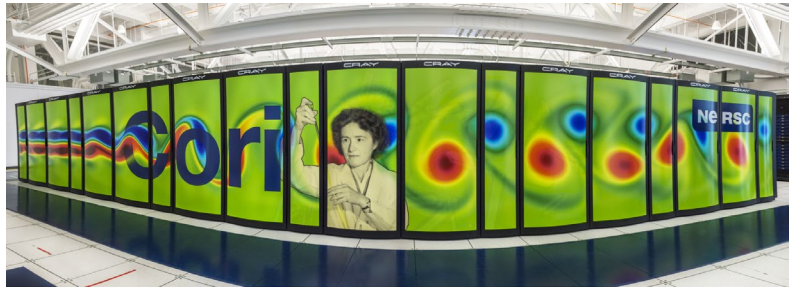
High density of points at low momentum error suggests that there is a promise to this methodology

LIMITATIONS AND NEXT STEPS



- Non-unique solutions of the initial geometry
- Preciseness of energy partitions?
  - Do we need to increase the computation power?
- Generalization to *N*-fragment molecule
- Generalization to 2 and 3 dimensional spaces
- Comparison to CEI experimental data
  - What does the laser do to the molecule while it is charging up?
  - Error in method?





Cori "Haswell" supercomputer with peak performance of 30 petaflops (Floating Point Operations per Second)

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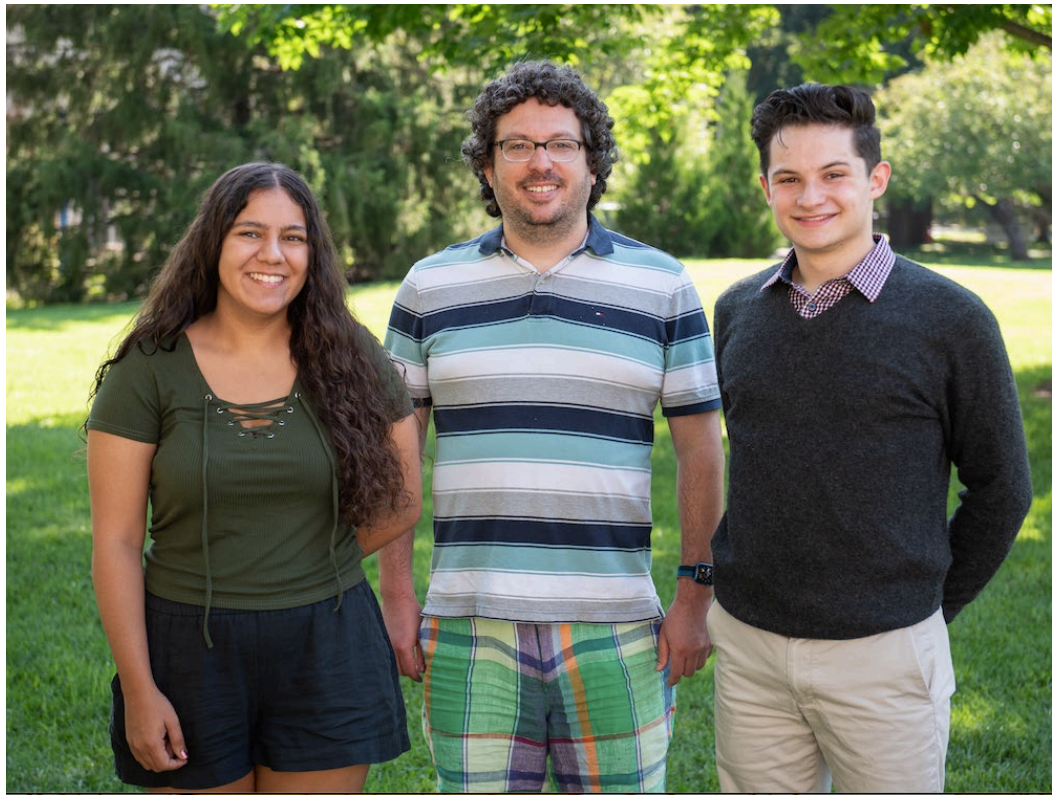
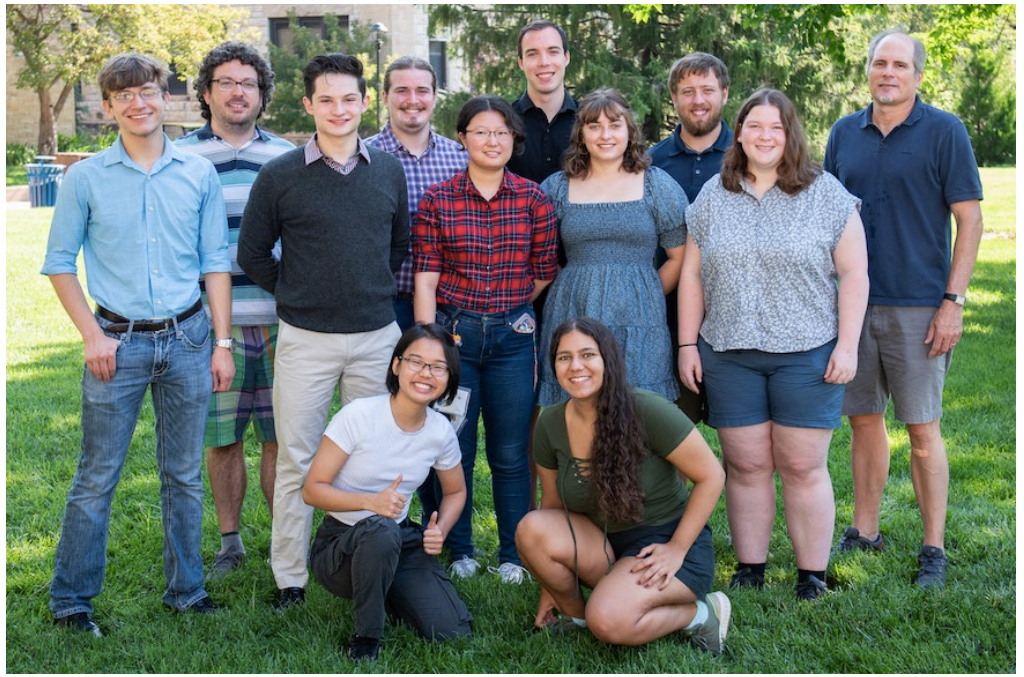
NSF Research Experiences For Undergraduates



For advising this research project:

Loren Greenman, Ph.D.



For running of of the KSU Physics REU 2022

Loren Greenman, Ph.D.

Bret Flanders, Ph.D.

Kim Coy

PICTURE CREDITS

- Source: K-State physicists publish on X-ray lasers taking pictures of complex molecules
- Source: “Chemical bonding of water”, Wikipedia
- Source: “Energy profile (chemistry)”, Wikipedia
- Source: “Coulomb explosion”, Wikipedia
- Source: “Electric Force”, Khan Academy
- Source: “Flow past a rotating cylinder”, ResearchGate
- Source: “Cori”, NERSC