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Zacros

Software Package Development: Pushing the Frontiers of Kinetic Monte Carlo Simulation in Catalysis

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Introduction to Zacros

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Zacros:

Zacros

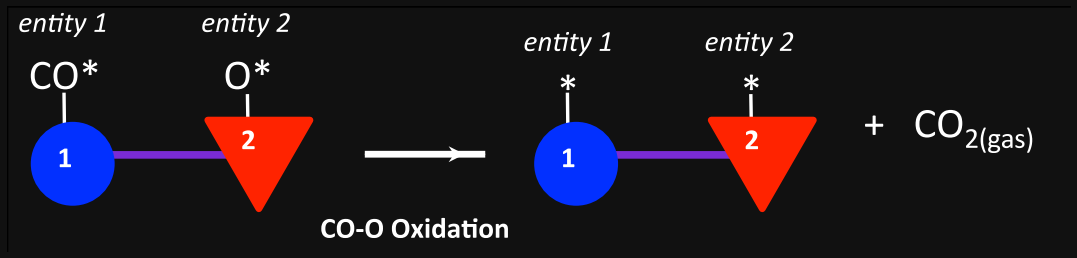
Advanced lattice-KMC made easy

- Kinetic Monte Carlo (KMC) simulation of surface chemistry
- Graph theoretical framework
  - Handle complex surface patterns

For more info visit: zacros.org

Reaction patterns

- Possible reactions get a time assigned based on:
  - kinetic rate constants (Propensities)
  - A random number



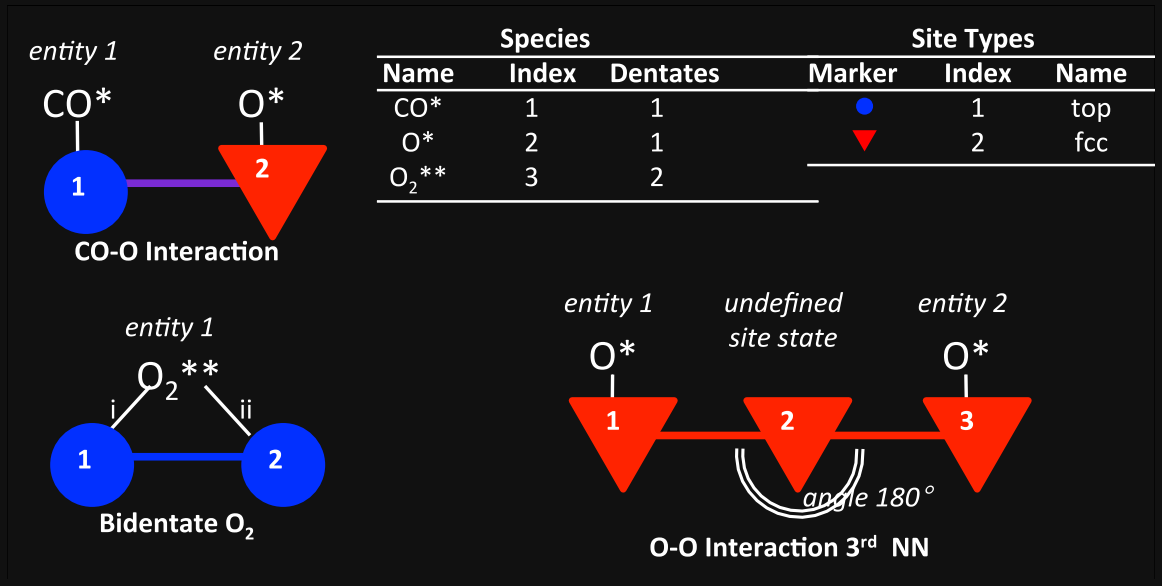
Example of simple reaction pattern

See Stamatakis and Vlachos. 2011. J. Chem. Phys. 134(21): 214115
and Nielsen et. al. 2013. J. Chem. Phys. 139(22): 224706

Surface energy

To calculate rates, the surface energy is needed:

- Surface energy is given as a cluster expansion
- I.e. an expansion in simple surface patterns



Species			Site Types		
Name	Index	Dentates	Marker	Index	Name
CO*	1	1	●	1	top
O*	2	1	▼	2	fcc
O_2**	3	2			

Example of cluster expansions

Pseudo code

- Select most imminent process
- Remove adsorbates from lattice
- Remove clusters with reactants
- Remove reactions involving the reactants
- Add product adsorbates
- Find new energy clusters
- Find existing processes that need update
- Update rates of existing processes
- Add new processes

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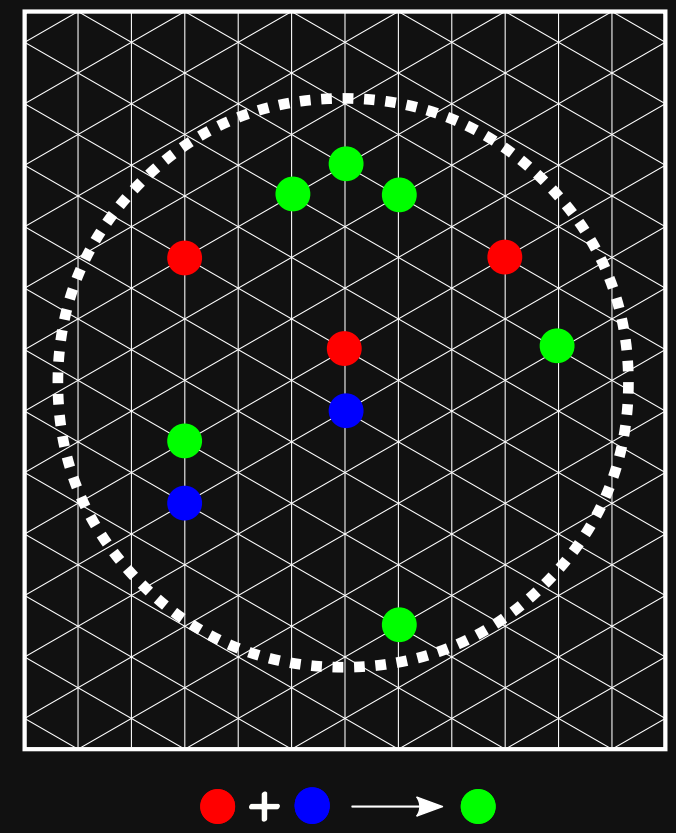
Technical

- Fortran 95/2003 code
- Originally fully serial
- Two performance issues with serial code identified

Cluster Expansion

- Long range interactions requires large cluster expansions
- Larger cluster expansions => More processes to update
- This is the first performance issue



Interaction length affected by reaction

Lattice size

Large lattice for accurate simulations

- Update time is independent of lattice size
- But reaction rate is not
- KMC time / CPU time depends linearly on the number of sites
- Large lattices are time consuming to simulate
- The second performance issue

Reaction updates:

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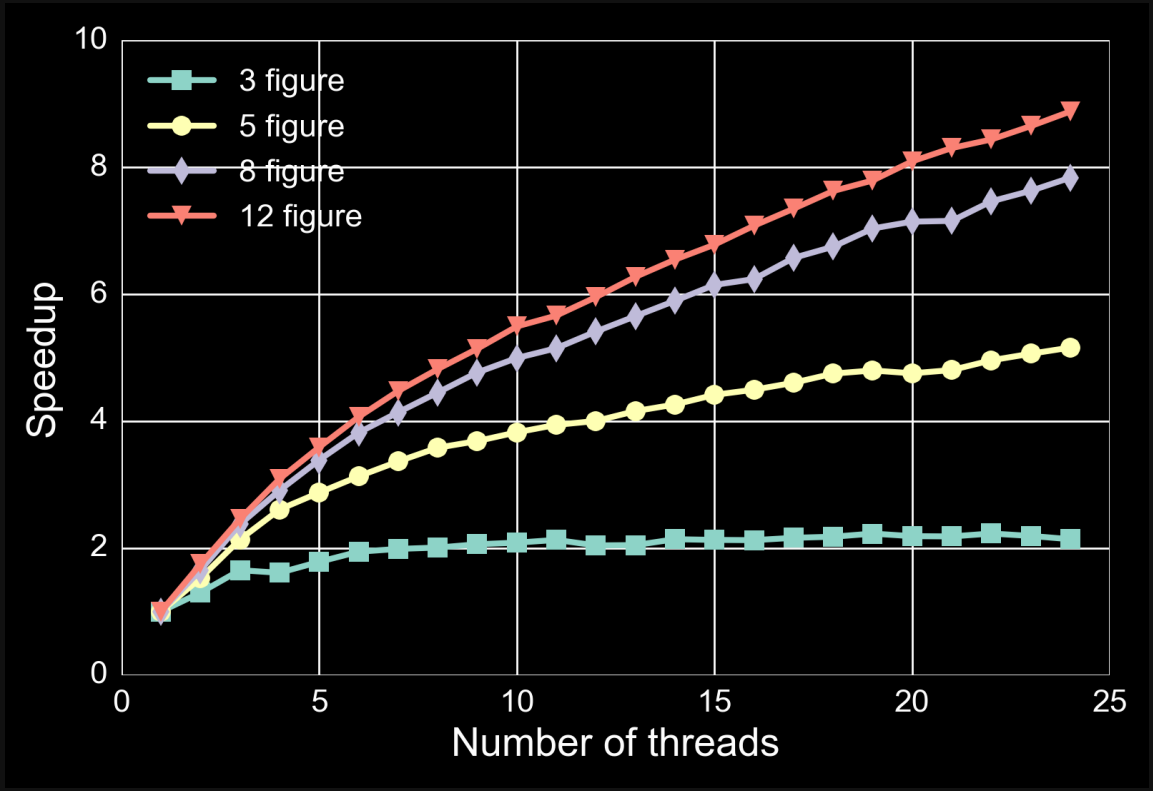
OpenMP:

- Profiling shows bottleneck in update rates ...
- Many processes are affected
  - Especially for large cluster expansion
- Do loop of independent processes to update
- OpenMP parallization of this loop

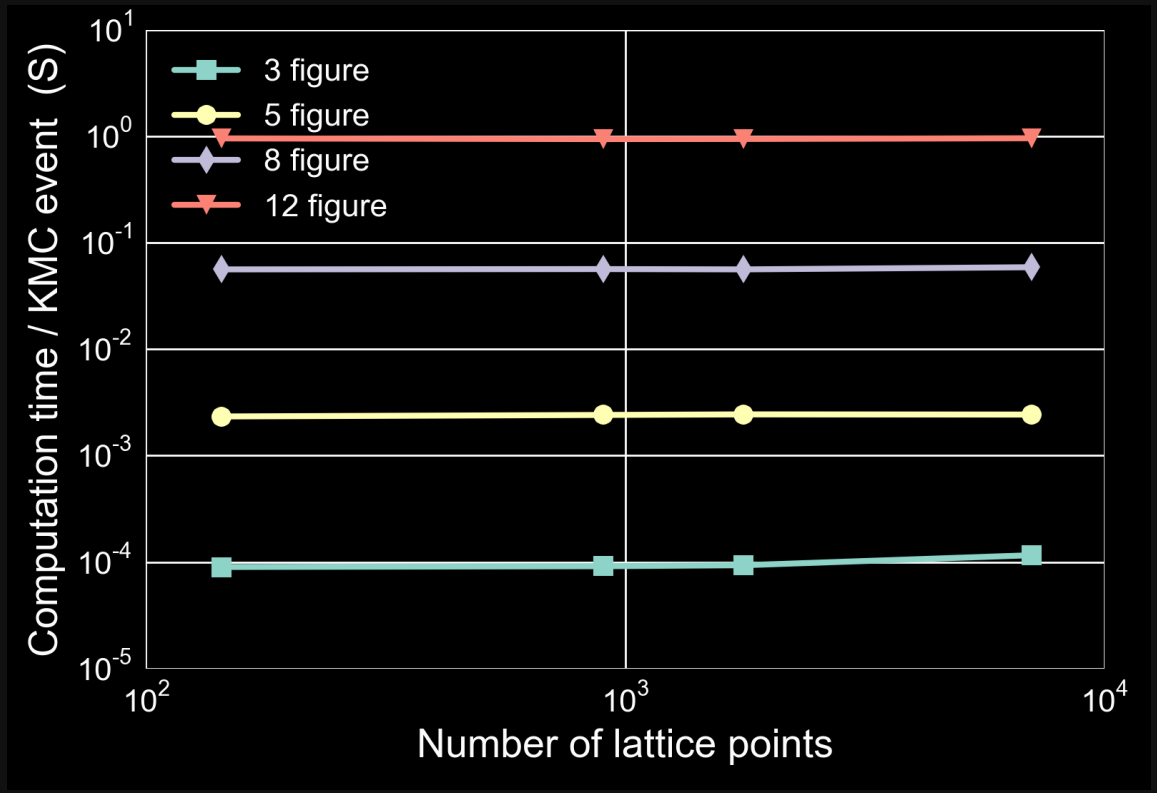
See Nielsen et. al. 2013. J. Chem. Phys. 139(22): 224706

Scaled performance of OpenMP



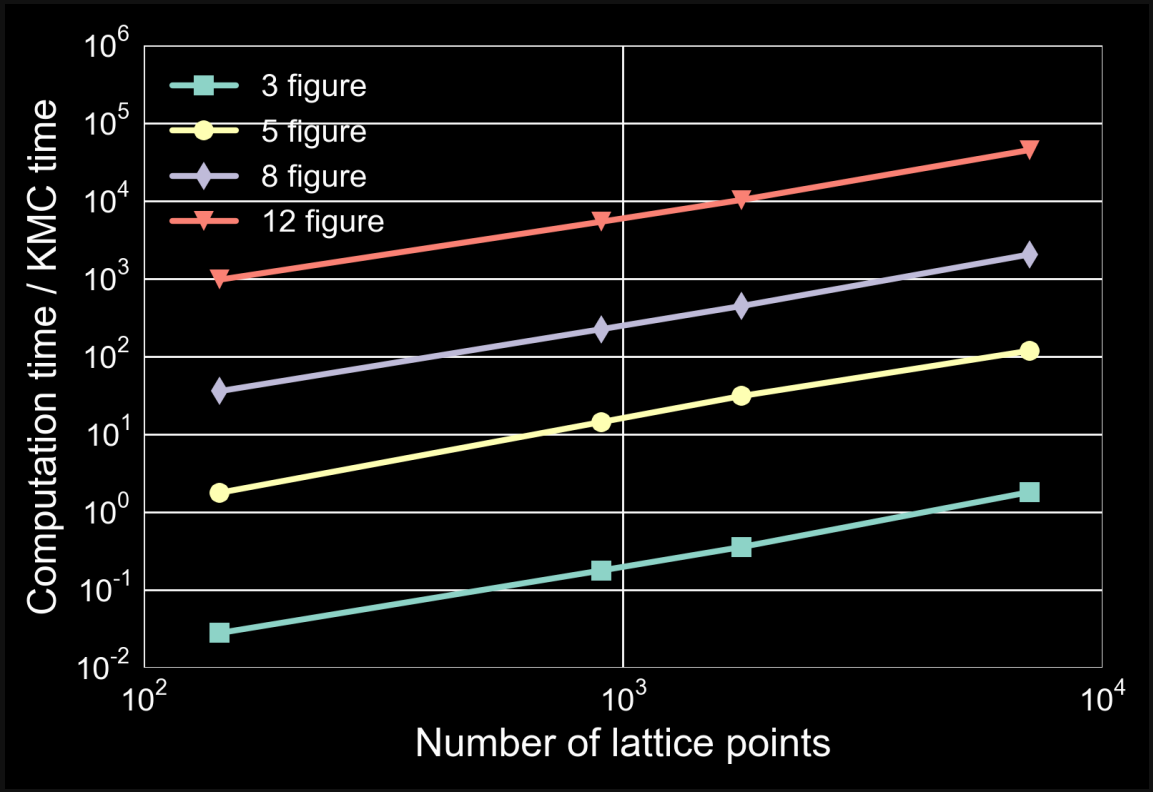
NO oxidation model with 4 different cluster expansions on Archer

Computational time per event



Time per KMC event is independent of lattice size.

Computational time / KMC time



- But number of events per simulated second is not
- 12 figure expansion at 7056 lattice points (12 threads):
  - $> 10^4$ seconds per simulated second

Limitations

Decent speed up for large cluster expansions but:

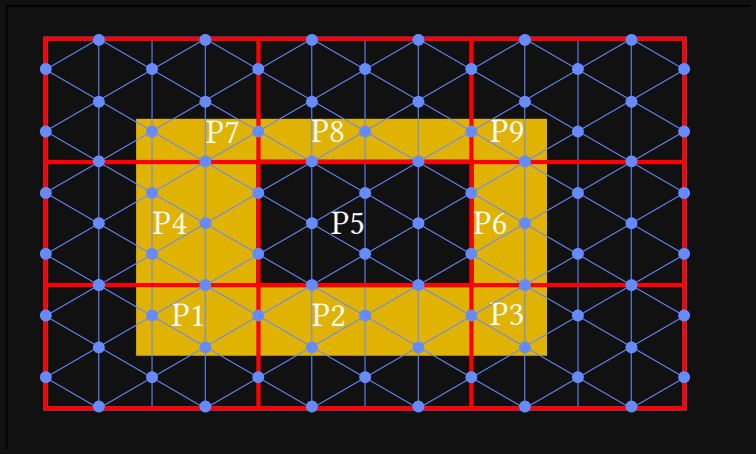
- OpenMP limited to one computational node
- Simulations are still too slow for large lattices and clusters

Solution: MPI Parallelization over lattice

Spatial Parallelization

MPI based parallelization

- Reactions on individual domains
- Halo for
  - Reactants
  - Products
  - Energetic clusters



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Original plan:

Implement algorithm proposed by Lubachevsky

- Algorithm is developed for Ising spin model
- Each domain keeps track of a local time
- Global time is min(localTimes)
- Updates in a MPI domain is allowed if:
  - Local time is smaller than all neighbours

Lubachevsky. 1988. J. Comp. Phys. 75 (1): 103

Algorithm

- Perform spin flip if time is smallest among neighbours
- Select a site and either:
  - Perform spin flip
  - Perform null event
- Advance local time by a random interval
- Repeat

Time advancement is independent of whether a spin is flipped

Energetics affect the relative probability of null events

Algorithm in Zacros

Same principle:

- If local time < neighbours time:
  - Advance local time
  - Perform reaction
  - Send halo and new local time
- Else:
  - Wait to receive halo and time

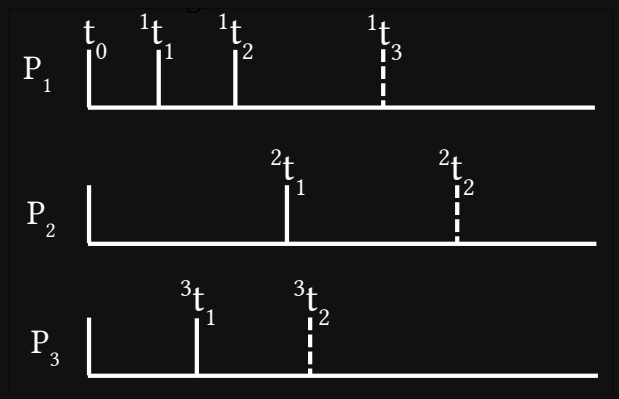
Issues in Zacros

- Future reactions have a wait time associated with them
- Wait time is random but determined by reaction rates
- Most imminent reaction is performed
- Reaction happen after wait time
- Wait time can differ by several orders of magnitude

Example

Assume that we have 3 MPI nodes in a 1D array

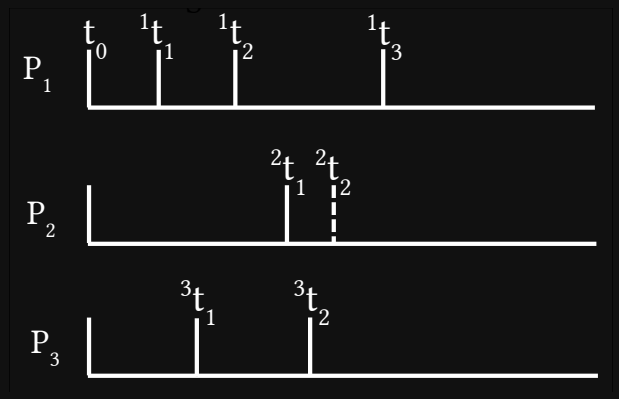
Both P_1 and P_3 are free to perform reactions



Example

Assume that we have 3 MPI nodes in a 1D array

Conflict P_1 should not have performed a reaction



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In summary

- Can't change the condition to smallest among $T_{local} + T_{wait}$
  - The reactions that T_{wait} on neighbours represent have not happened:
  - In fact they may never happen
  - Reactions may propagate across domains

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Alternative strategies

An alternative proposed by Jefferson

- Each node propagates its reactions without synchronization
- Stores a list of anti reactions to performed reactions
- When a reaction is performed messages are sent to relevant neighbours

Jefferson. 1985. ACM Trans. Program. Lang. Syst. 7 (3): 404

Alternative strategies

- If conflicts arise neighbours will roll-back
  - Sending anti messages to their neighbours
  - With further potential roll-back
- The "slowest" node determine a virtual time horizon (Global time)
  - No roll-backs beyond this the are needed

Conclusion

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Conclusion

- Good performance improvement for long range interactions
- Spatial parallelization is on-going work
  - Change of algorithm