The Consortium for Enabling Technologies and Innovation

Virtual Summer Meeting for Young Researchers

Evaluation of Chemical Speciation to Enable Online Monitoring of Molten Salt Reactors

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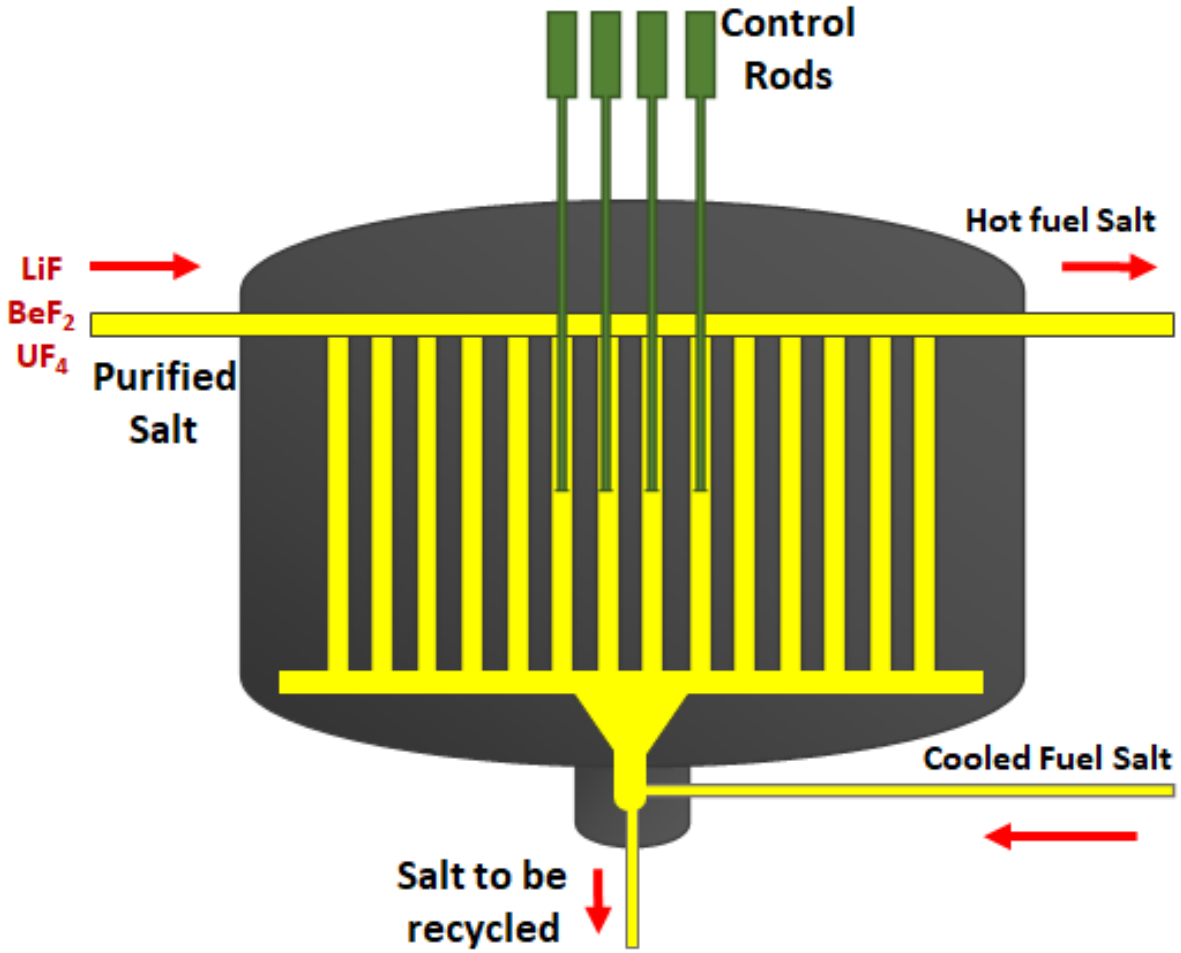
Molten Salt Reactor



Pro:

Can enable online chemical processing of fission products away from actinides

Con:

Can allow for the separation of Pu

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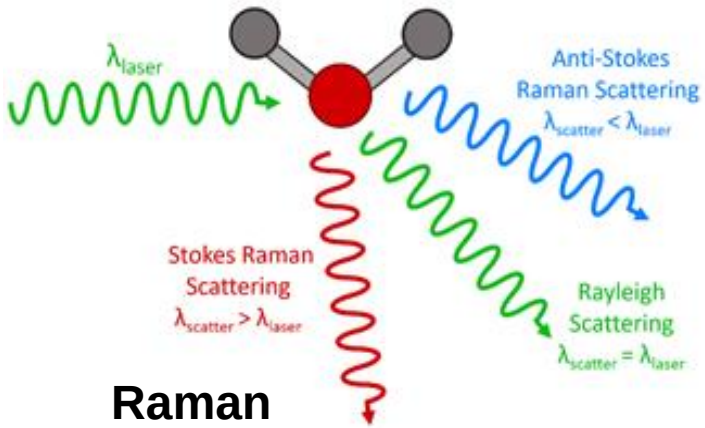
Overall Project Scope

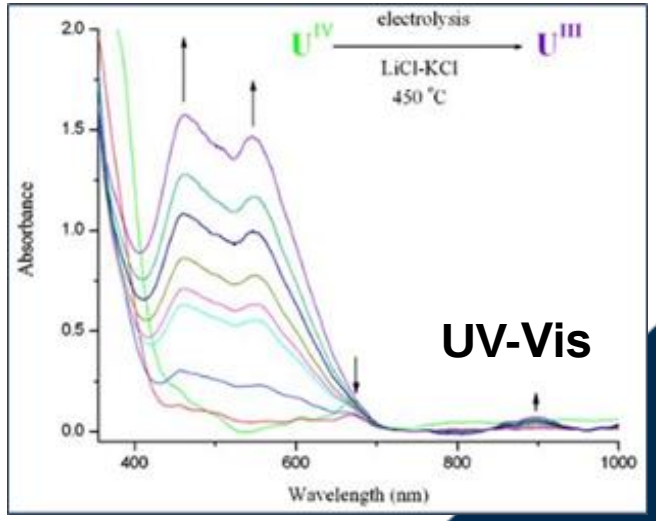
Assess MSR fuel speciation and develop online monitoring capabilities by coupling experimental and computational approaches

Online monitoring

- Uses electronic and vibrational signatures to characterize and monitor elements in the core



Raman



UV-Vis

Key Benefits

1. Decrease radiation workers dose
2. No reactor shutdowns required
3. Collect data nondestructively
4. Reduce proliferation risk

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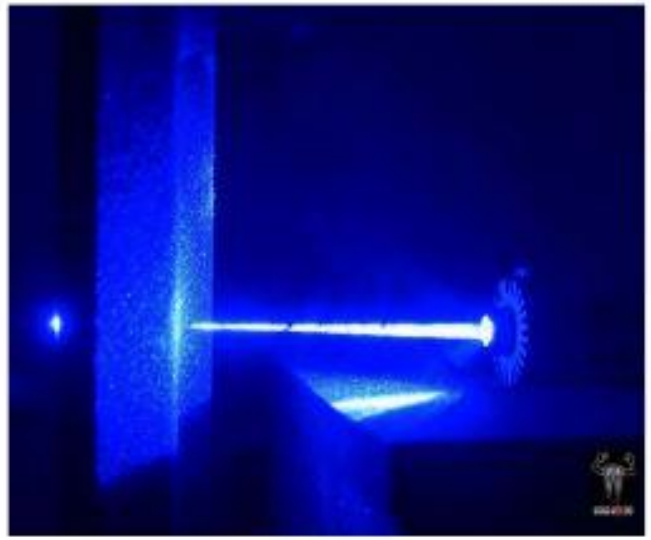
1. Tse, P.; et al. Analytica Chimica Acta. **2020**. 1101, 1-13.
2. Polovov, I.; et al. *Inorg. Chem*. **2008**, 47, 7474-7482.

Scientific Questions

What are the molecular structures and chemistry that govern ***optical indicators*** of molten salt reactors?



1. Establish molten salt spectroscopic monitoring capability
2. Develop and validate classical molecular dynamic (CMD) simulations
3. Leverage CMD simulations for Raman spectroscopic interpretation

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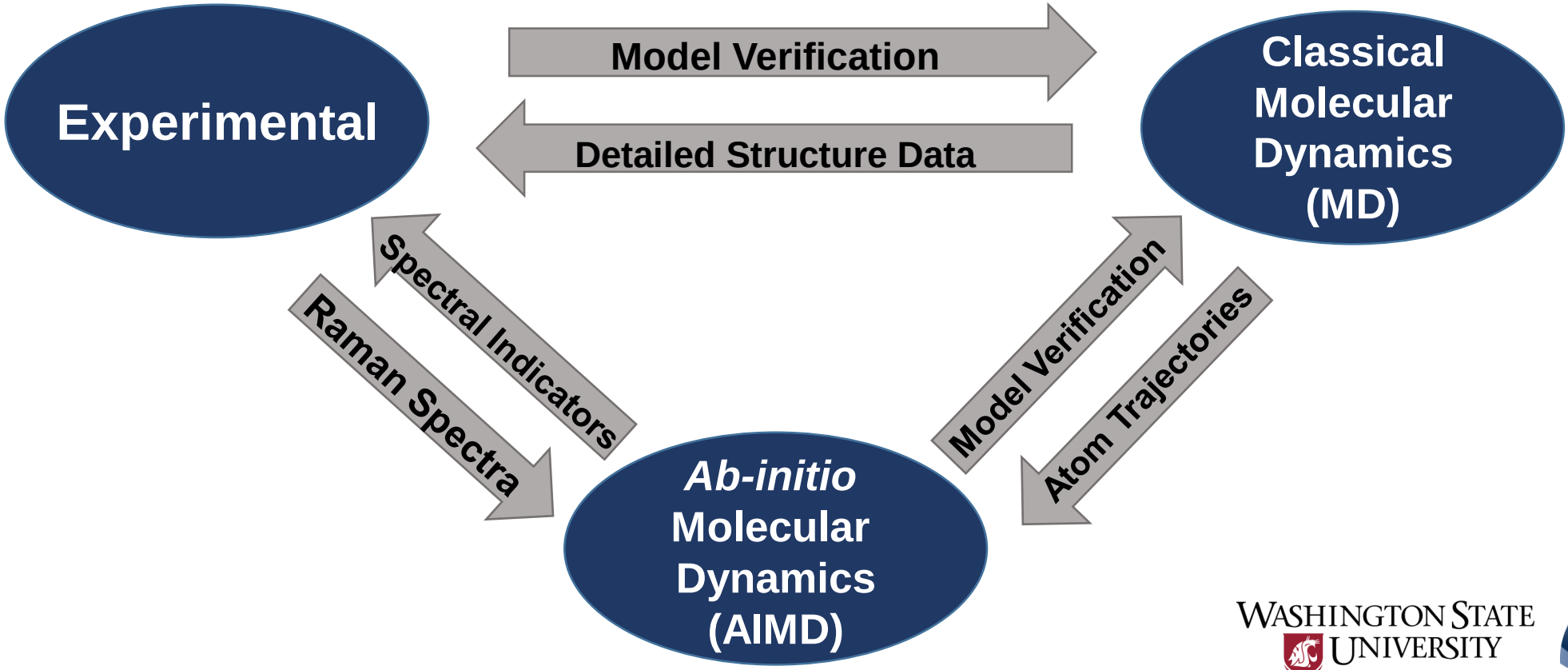
Project Integration Web



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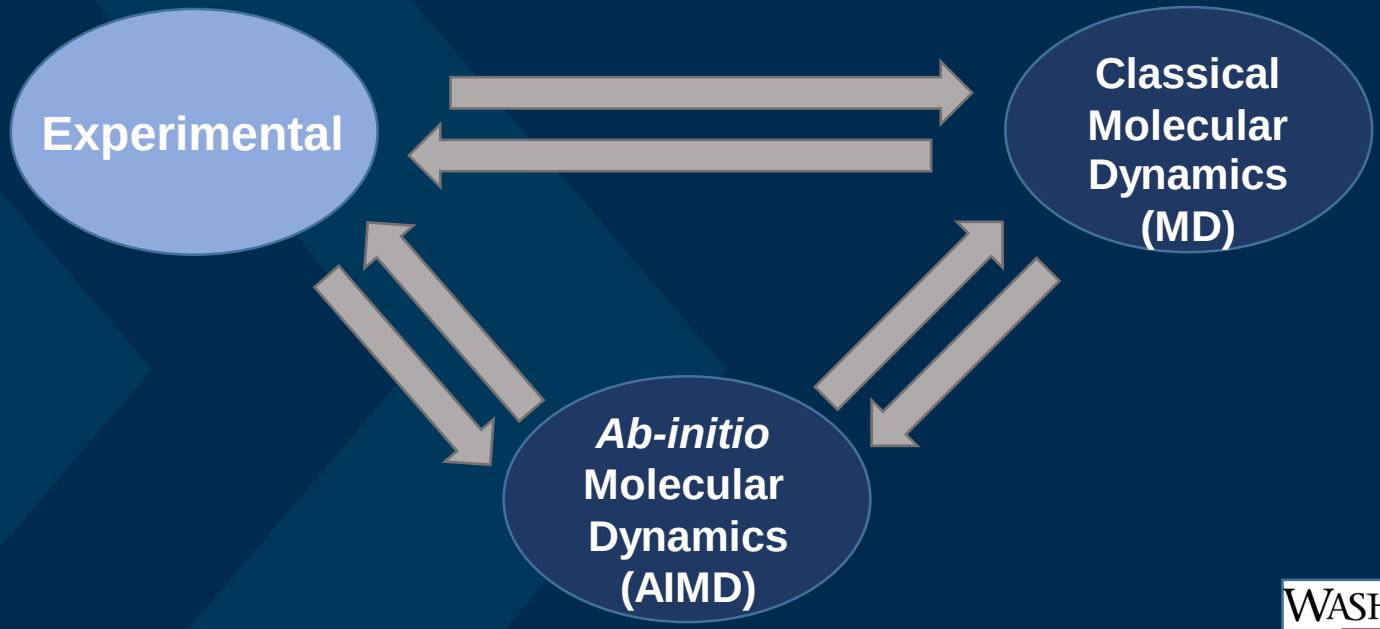
Nicole – CSM

Experimentation



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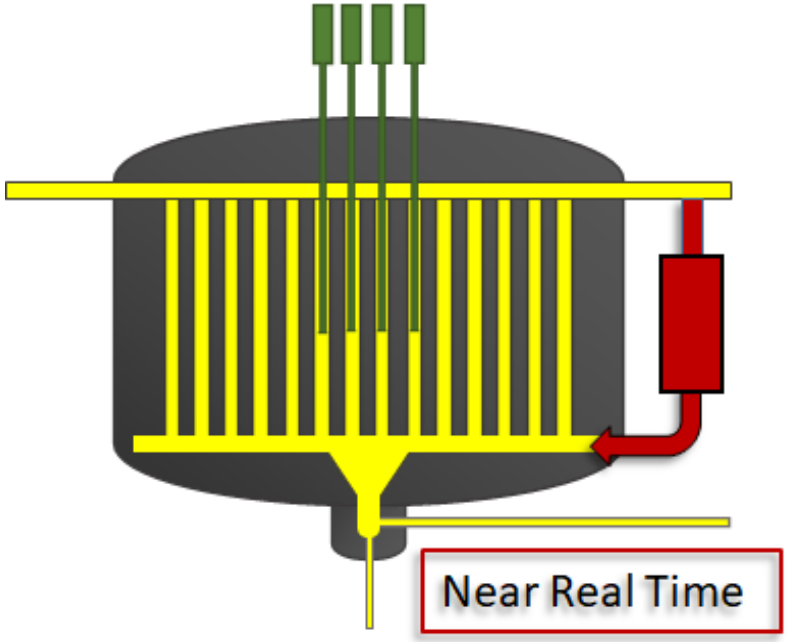
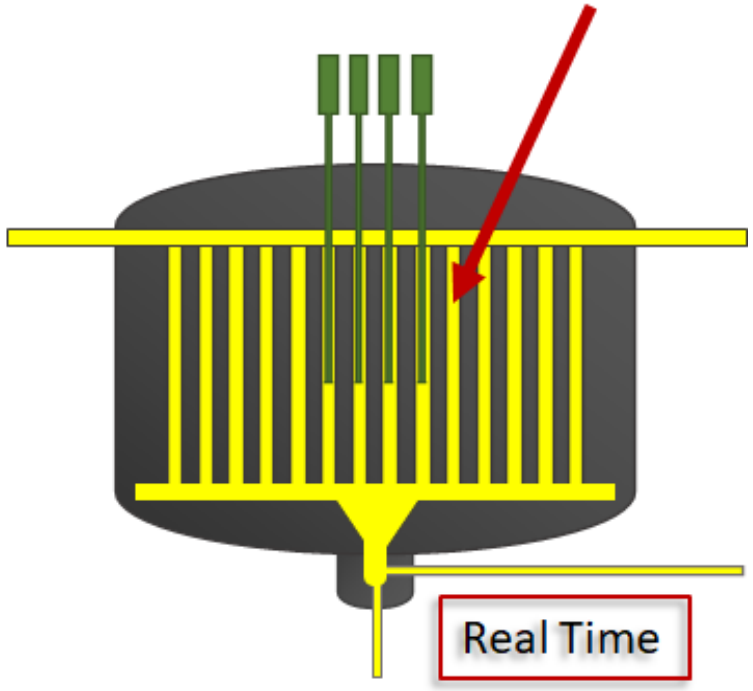
UV-Vis VS Raman

Can monitor actinides such as U, Pu



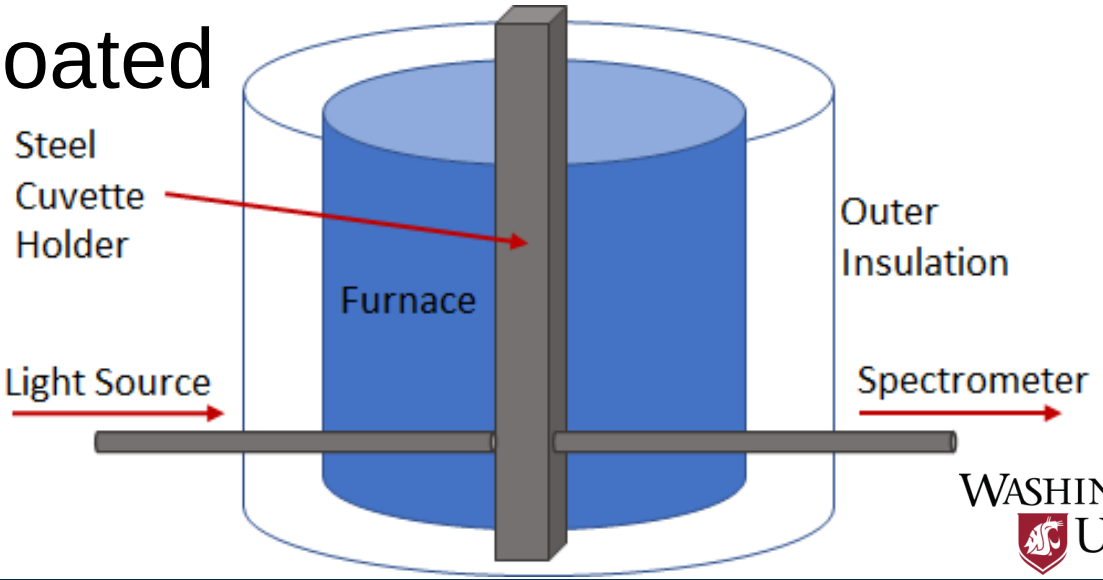


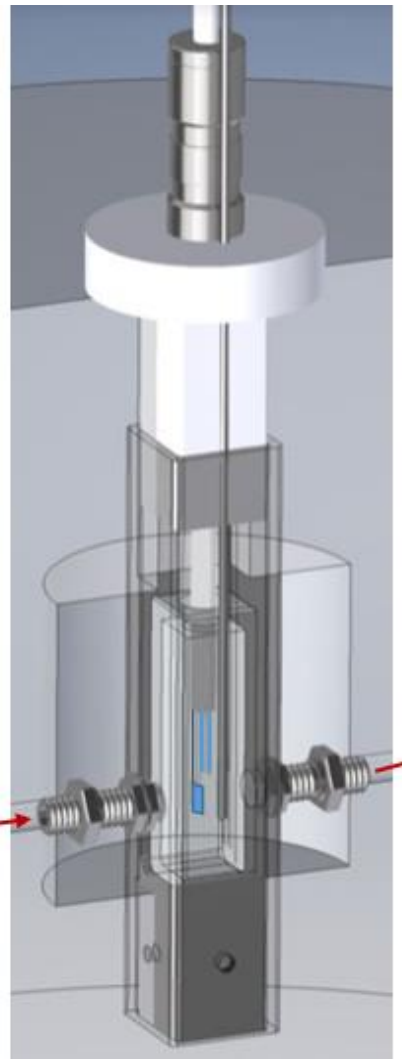
Probes the chemical environment

- Measures electronic absorbances
- Measures vibrational absorbances

Experimental Design

- Quasi-reference (Pt) electrodes
- Temperature ranges from 650-1125 K
- 316 steel, ceramic, and alumina bore tubes
- Optics (Gold-coated Fiberoptics)





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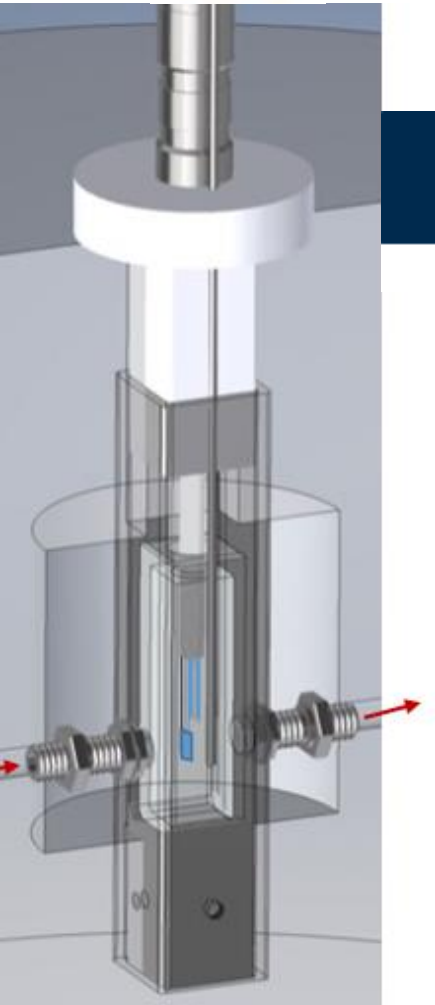
1. Schroll, C.; *et al. Anal. Chem.* **2013**, 85, 20, 9924-9931.

Experimental Design



Past Design

- Quasi-reference (Pt) electrodes
- Furnace shell
- Temperature range

New Design

- Glove box
- 316 steel cuvette holder
- Center bushing/ push to connect
- Alumina bore tube
- Quartz cell
- Optics / Raman

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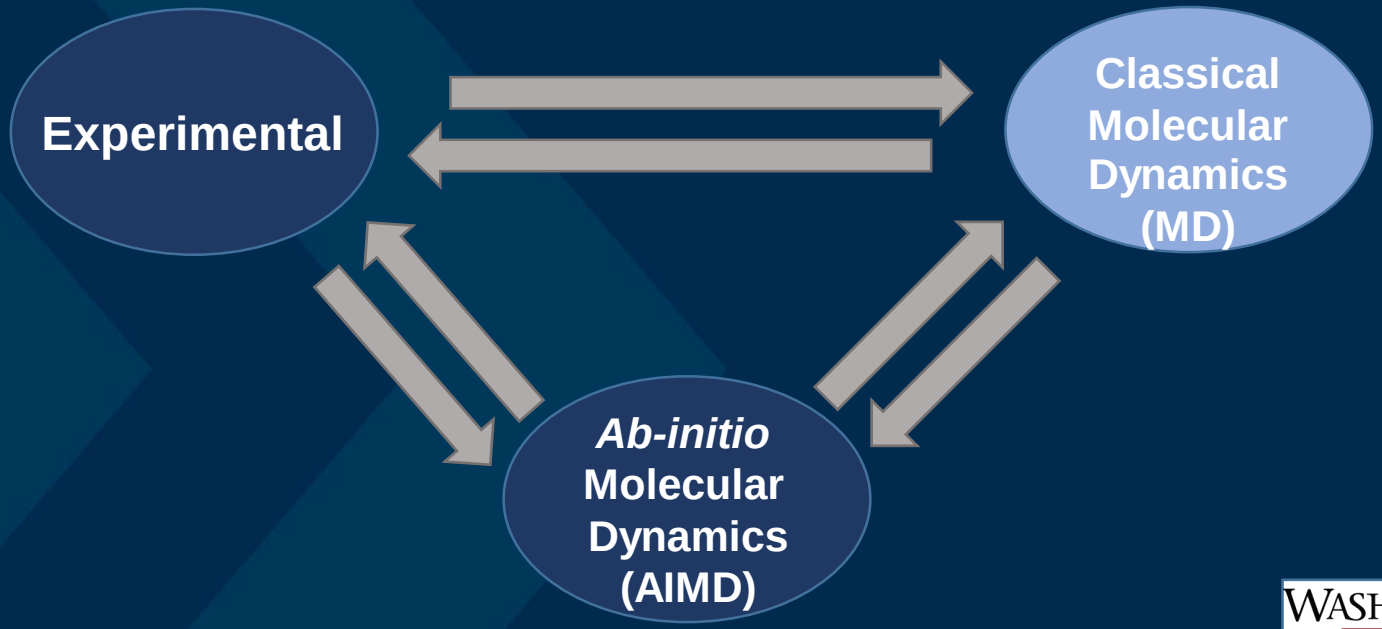
1. Schroll, C.; *et al. Anal. Chem.* **2013**, 85, 20, 9924-9931.

Jacob – CSM



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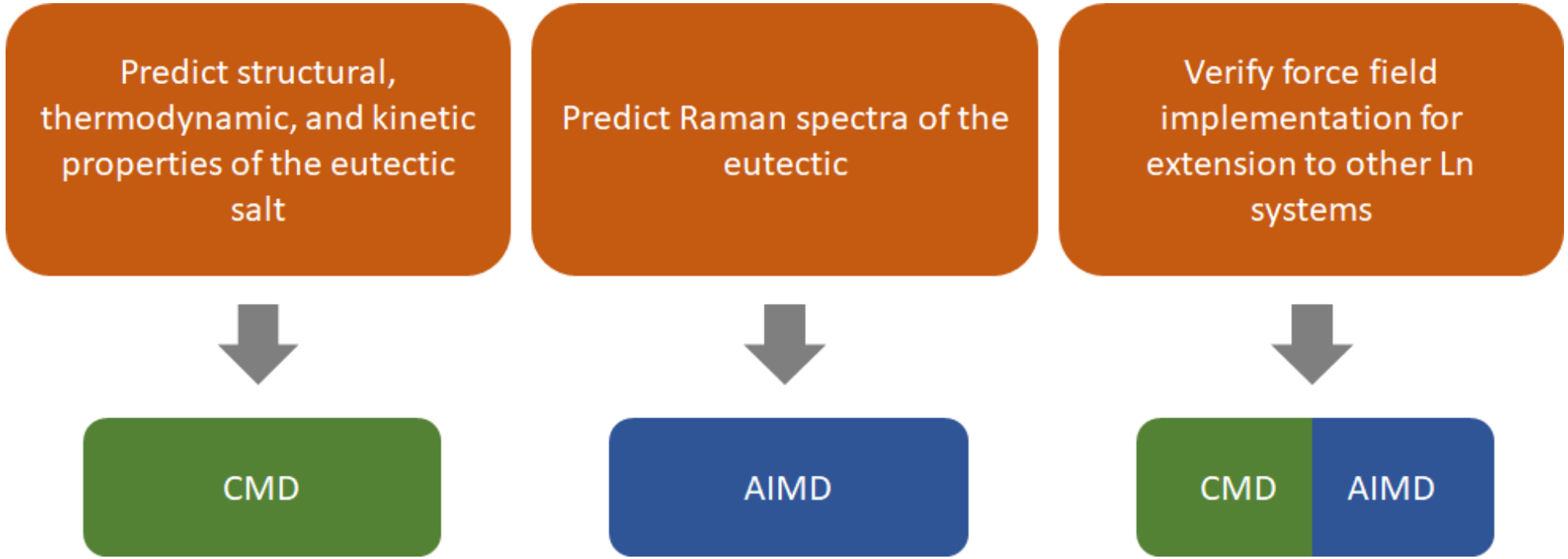
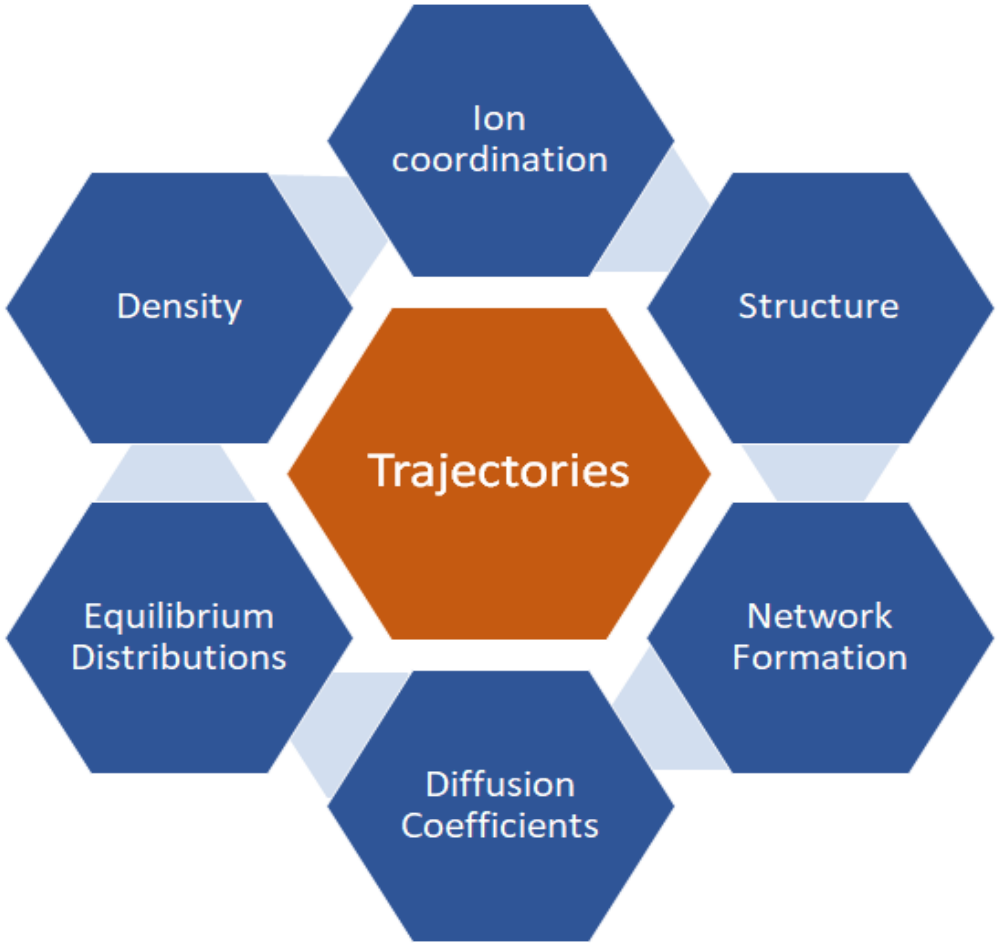
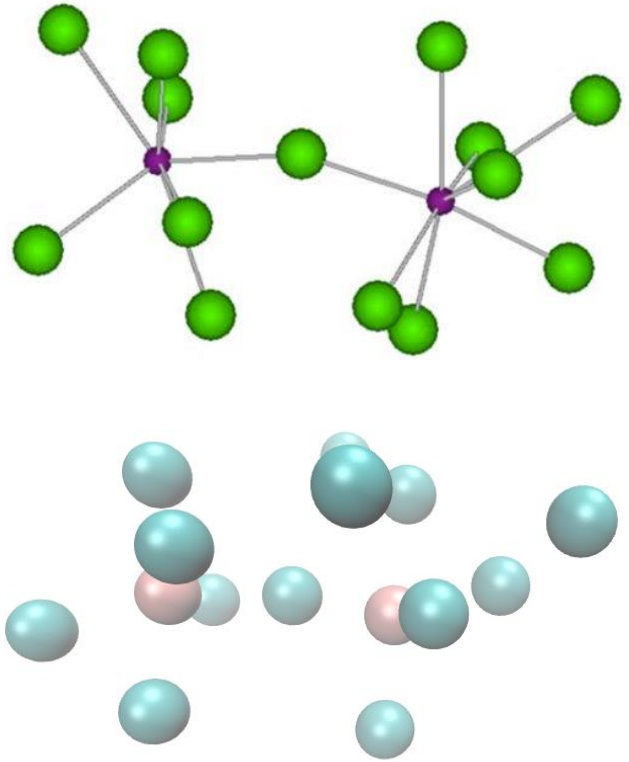
Modeling Objectives



Mechanics of Classical MD





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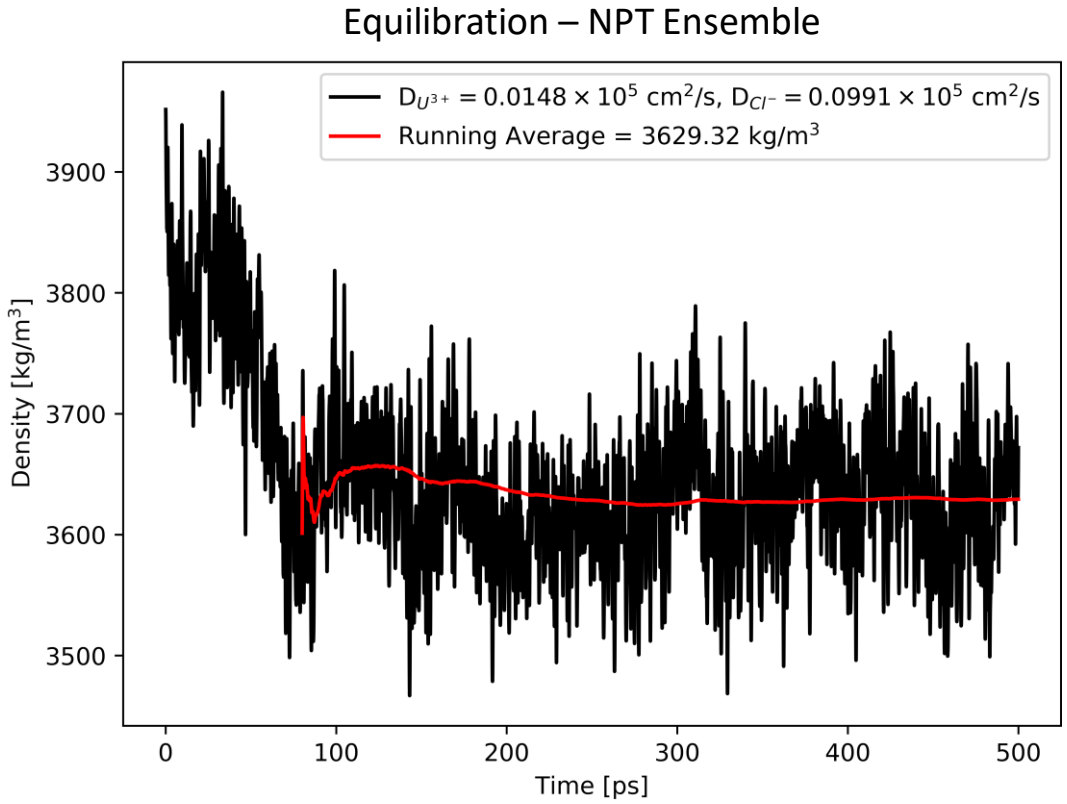
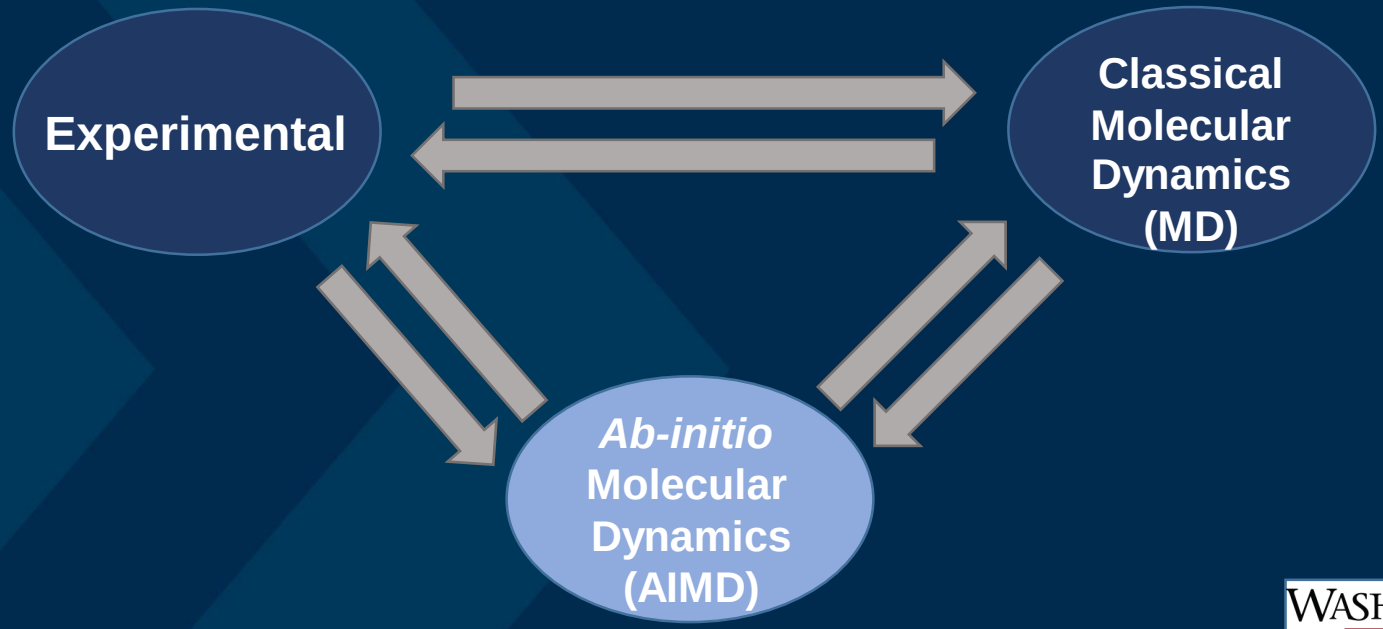
Y. Okamoto, et al. *Journal of Nuclear Materials*, 2005, *334*, 109-114.

Classical MD Verification

- Successful reproduction of
  - Density
  - Coordination
  - Uranium diffusion coefficients
- Quasi-successful reproduction of
  - Chlorine diffusion coefficients
  - Ion pair distributions



Will – WSU



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Why AIMD?

Use classical MD trajectory as the initial starting coordinates for DFT-based *ab initio* MD

Advantages: QM modeling of electrons to measure change in polarization and predict Raman spectra

Disadvantages: increased computational cost, limitations on system size and simulation length

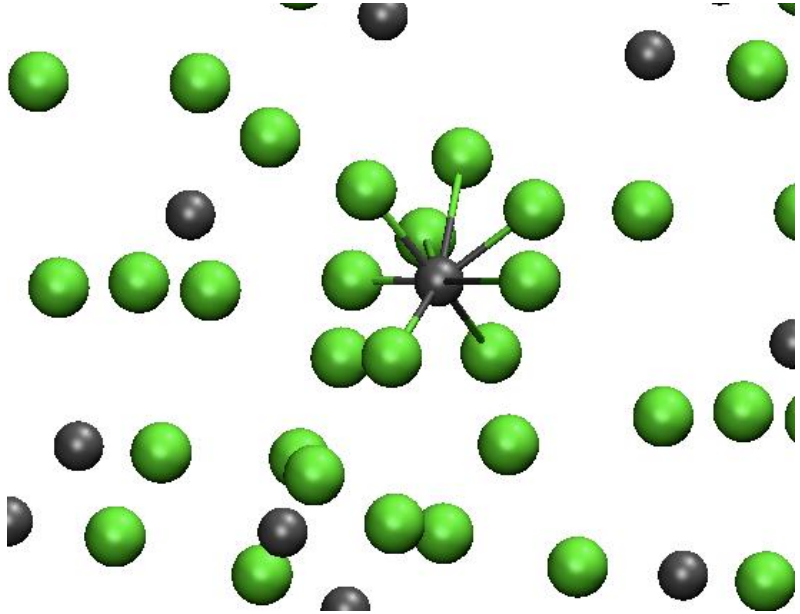
Important that starting configuration is near equilibrium

Unique molecular configurations

Within molten salts there are unique clusters of ions

Local organization –

- The chemical environment within the first coordination shell



UCl_3 molten salt

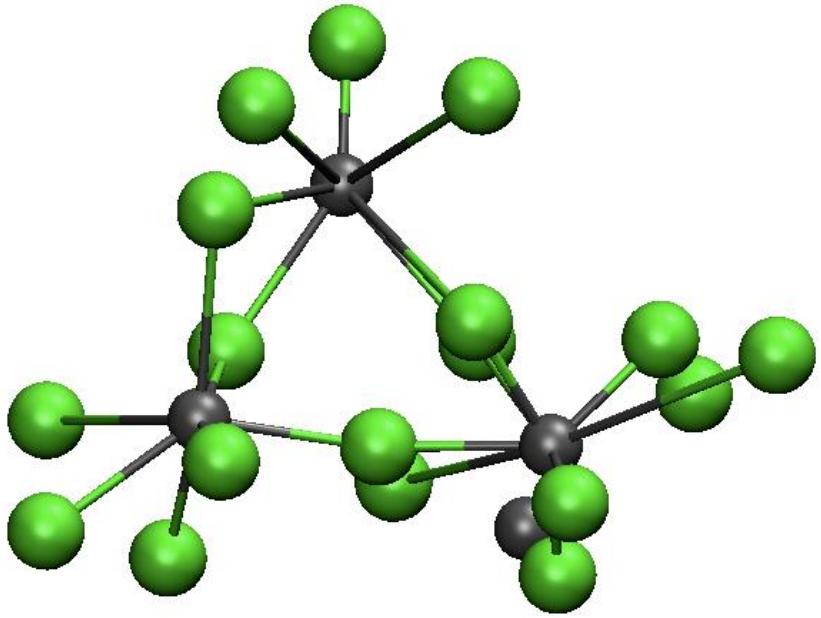
grey: U atoms; green: Cl atoms

Unique molecular configurations

Within molten salts there are unique clusters of ions

Chemical Network–

- The chemical environment beyond the first coordination shell



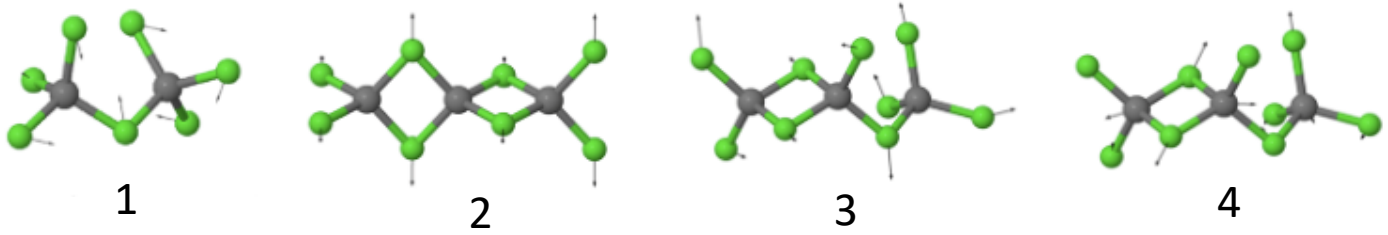
UCl_3 molten salt

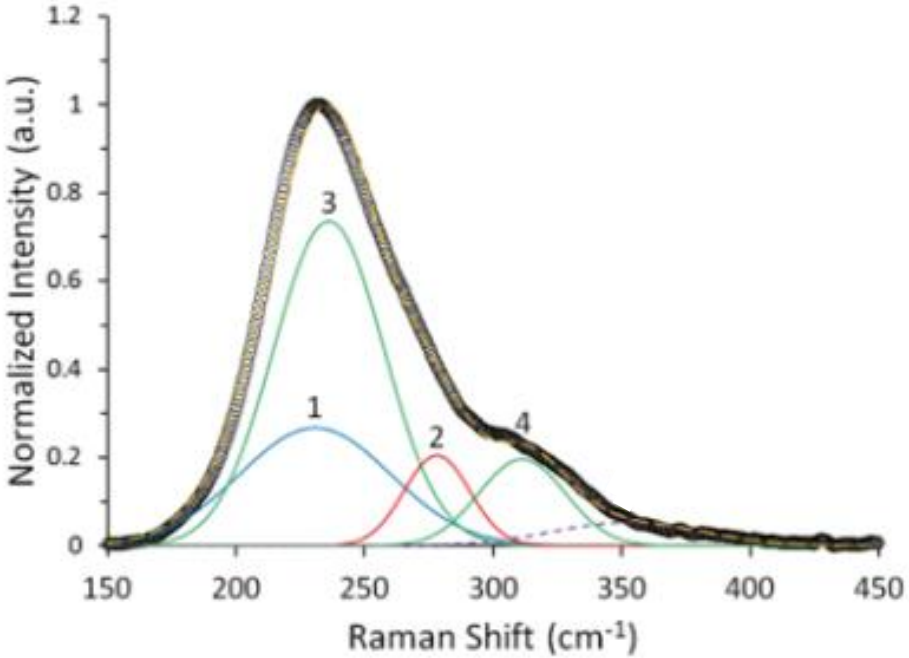
grey: U atoms; green: Cl atoms

Individual signatures leading to total Raman spectra

Experimental Raman spectra is a combination of all species in the sample

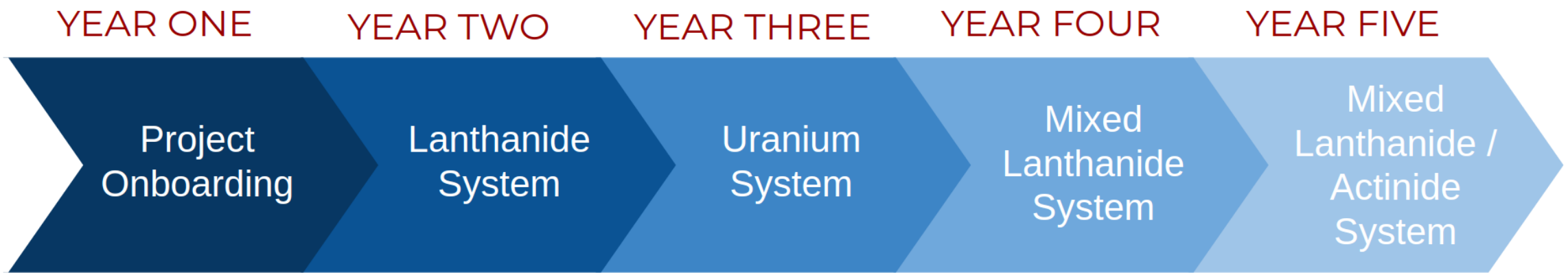
Computational chemistry allows for identification of unique species and their signatures





Alsayoud, A.; et al. *The Journal of Physical Chemistry B* 2016, *120* (17), 4174-4181.

Project Timeline



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Acknowledgement

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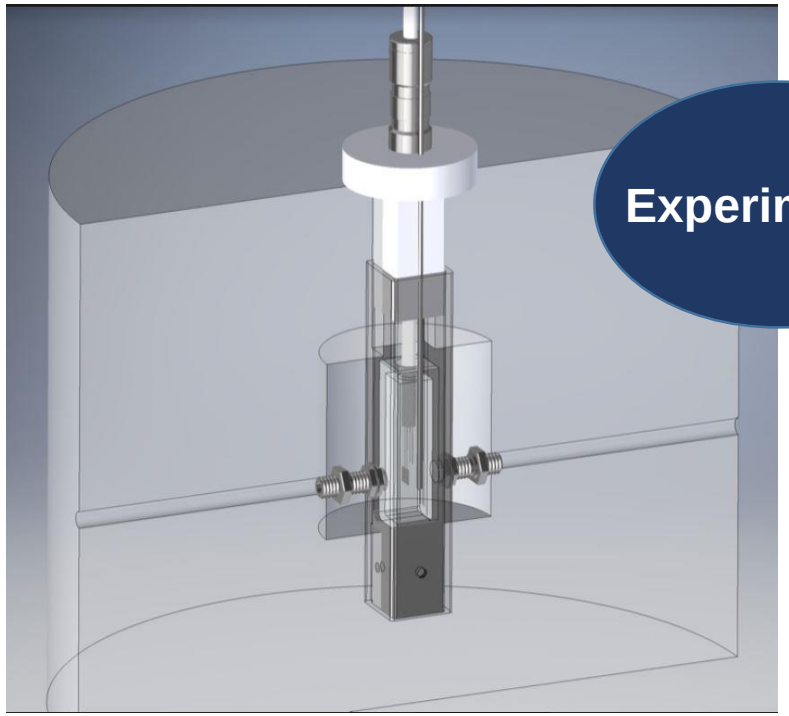
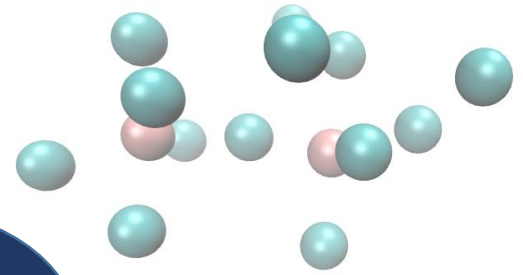
Summary

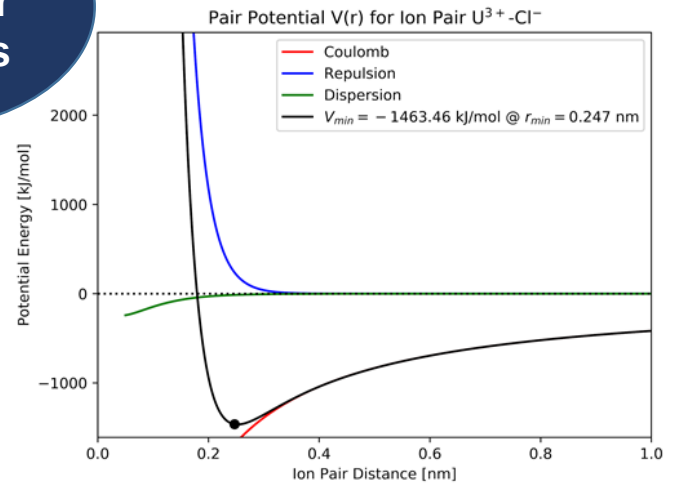


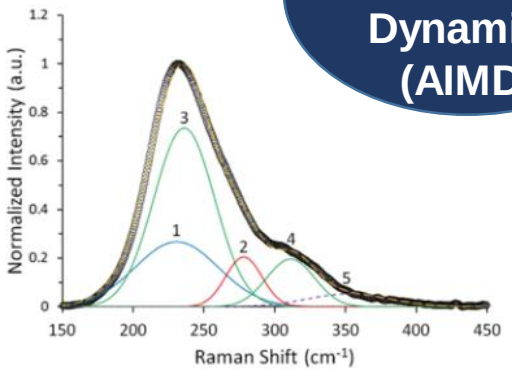
Experimental

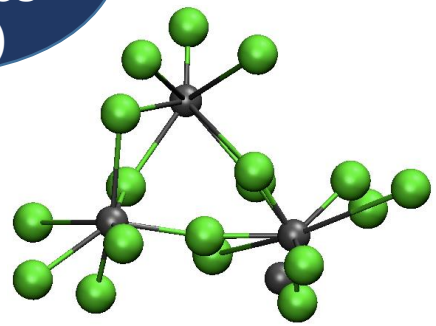
Classical Molecular Dynamics (MD)

Ab-initio Molecular Dynamics (AIMD)









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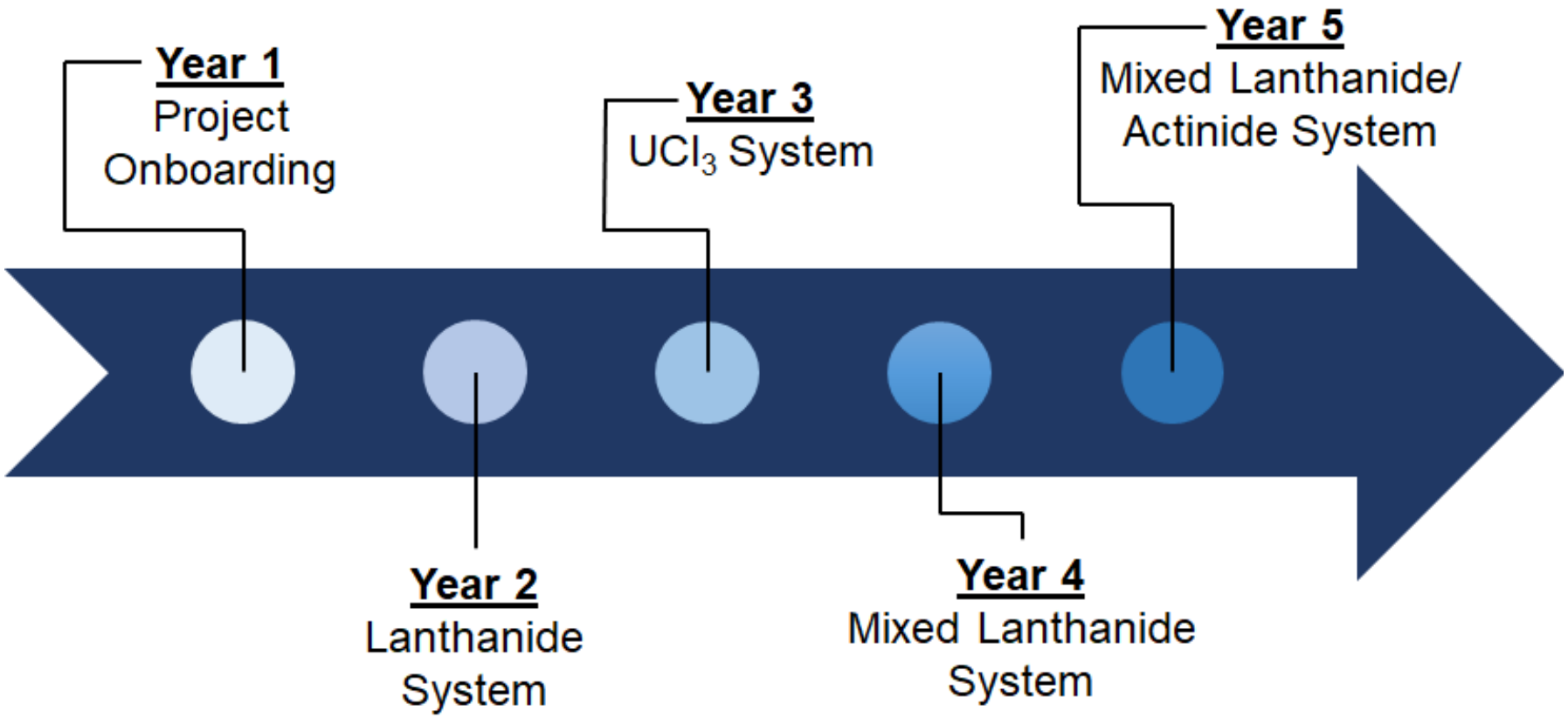

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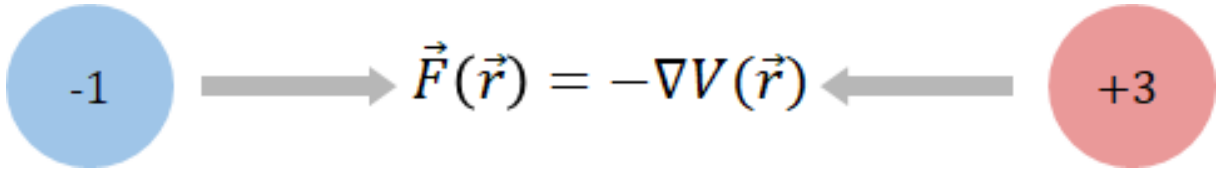
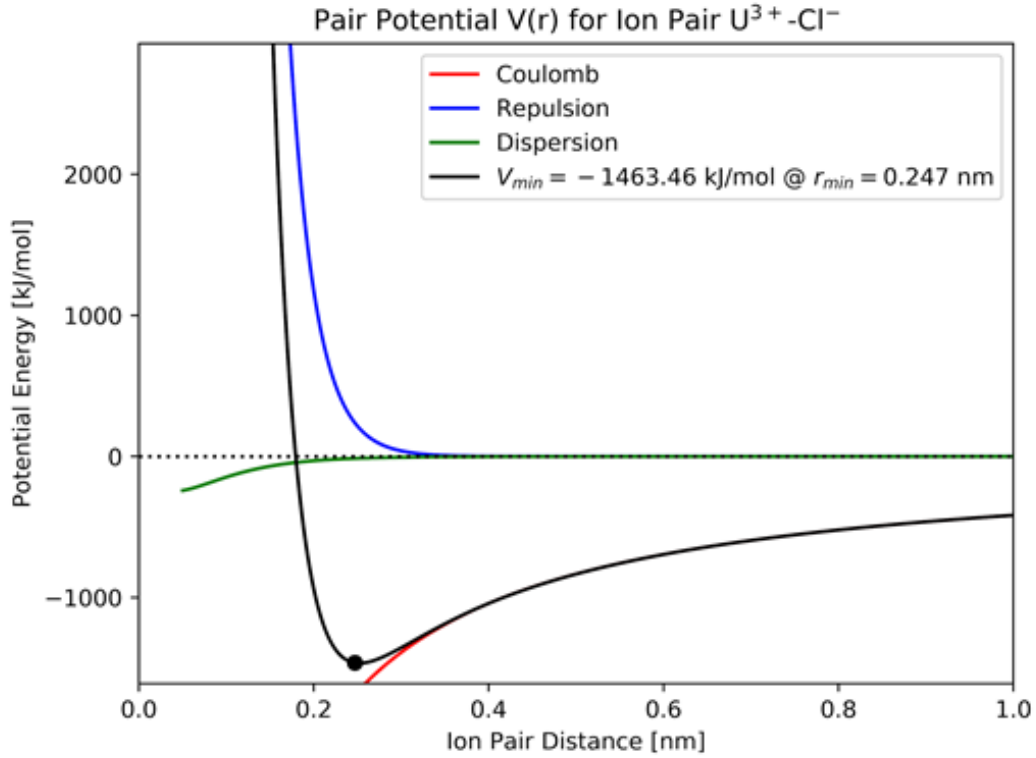
Mechanics of Classical MD

- Method of CMD stems from Newtonian mechanics
  - Particles have mass and velocity
  - Potentials (forces) define the interaction of each pair in the system





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