HAW: Hybrid Advance Workflows

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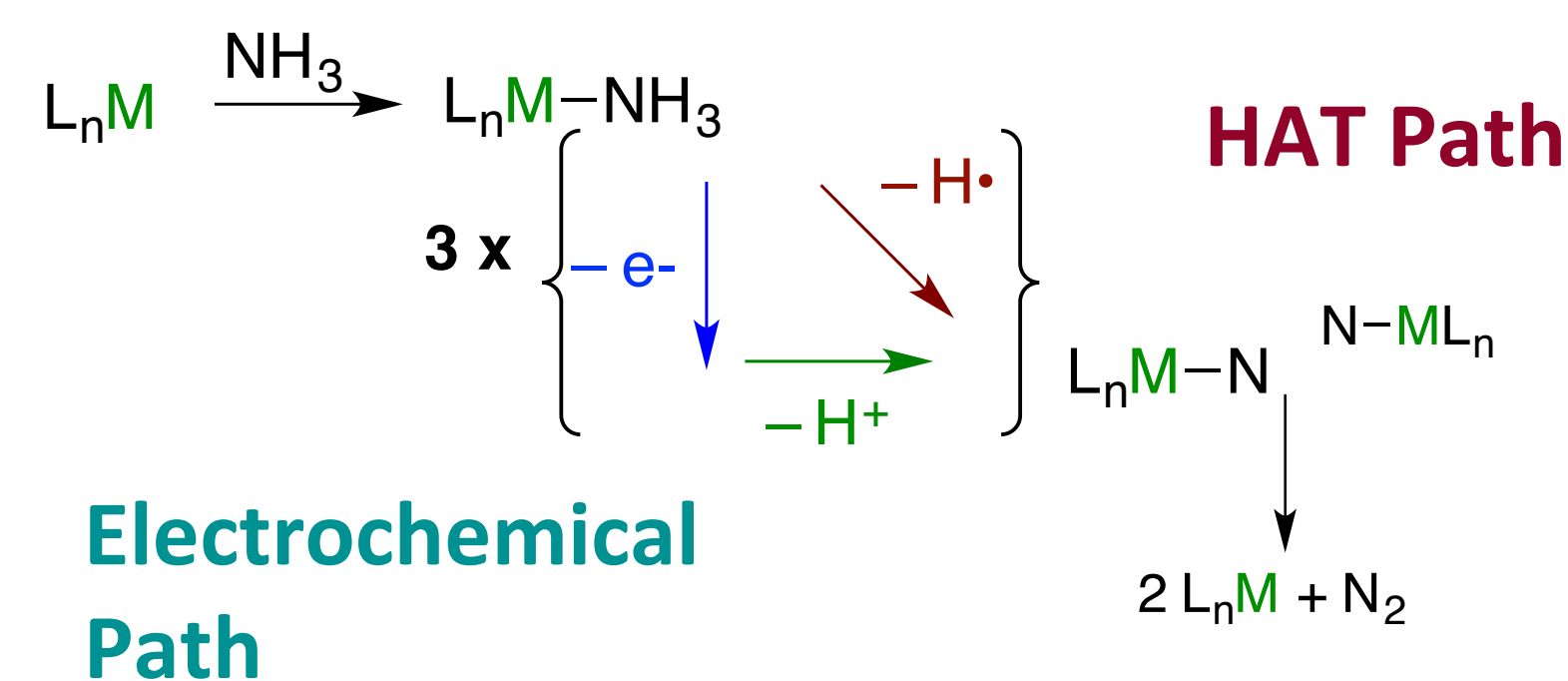
THE CHALLENGE

- Workload heterogeniety
  - Emerging applications in different domains (scientific simulations, machine learning, data analytics, signal processing, etc.) have different characteristics
- Providing a **high-performance**, **scalable**, and **versatile** solutions becomes a fundamental **requirement**
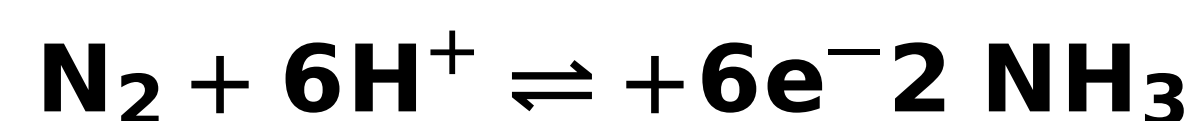
HAW'S GOALS AND SPECIFIC AIMS

- Design and develop:
  - Novel hardware/software co-design methodologies
  - Tools
  - Software programming libraries
- Efficiently perform design space exploration of future system architectures
- Implement software stacks, and applications

- Provide heterogeneous support for DMC applications dependent on both novel hardware and software designs for peak performance
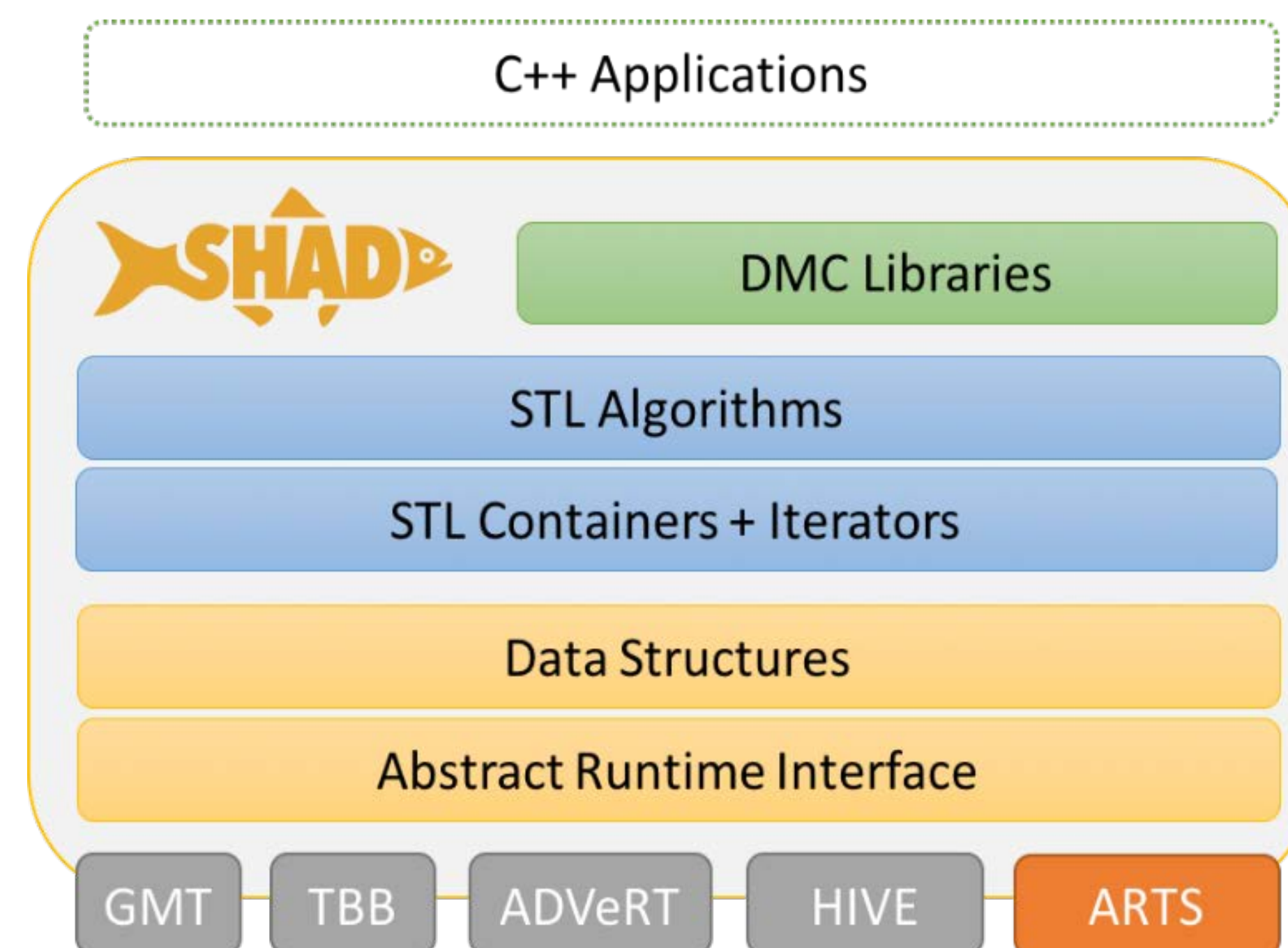
METHOD

$$L_nM \xrightarrow{NH_3} L_nM-NH_3$$

Electrochemical Path

HAT Path

$$3 \times \left\{ -e^- \quad -H^+ \quad -H\cdot \right\} \quad L_nM-N \quad \xrightarrow{N-ML_n} \quad 2\,L_nM + N_2$$

- Want to remove H atoms via hydrogen atom transfer (HAT) or electrochemical steps.
- Can calculate thermodynamics using DFT.

$$N_2 + 6H^+ \rightleftharpoons +6e^- \, 2\,NH_3$$

PROPOSED SOFTWARE STACK AND FEATURES

Correlation exists: Available $\pi*$ orbittal on the nitride (making nitride more electrophilic) leads to more favorable coupling.

KEY TAKEAWAY: We predict a series of MO-based complexes with potential to perform NH_3 oxidation and N-N coupling. Future work involves designing less oxophilic systems and exploring other modes of N-N bond formation.
S.I. Johnson et al. Chem. Commun., 2019, **55**, 5083–5086

https://github.com/pnnl/SHAD

DFT: B3LYP//6-31G**/SDD on M in Gaussian 09 and ORCA, SMD Solvent: MeCN, energies in kcal/mol, 2,4,6-tri-tert-butylphenoxyl redical (Ar0·) as HAT agent, followed by NBO analysis

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Sample table with alternating colored lines

ΔG for coupling and N_2 Release		
Cr	Mn	Fe
-108.6	-184.1	-194.4
Mo	Tc	Ru
-35.5	–	167.4
W	Re	Os
-13.7	–	-143.7

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