Using machine-learning accelerated simulations to inform new strategies for high throughput nanocarbon synthesis

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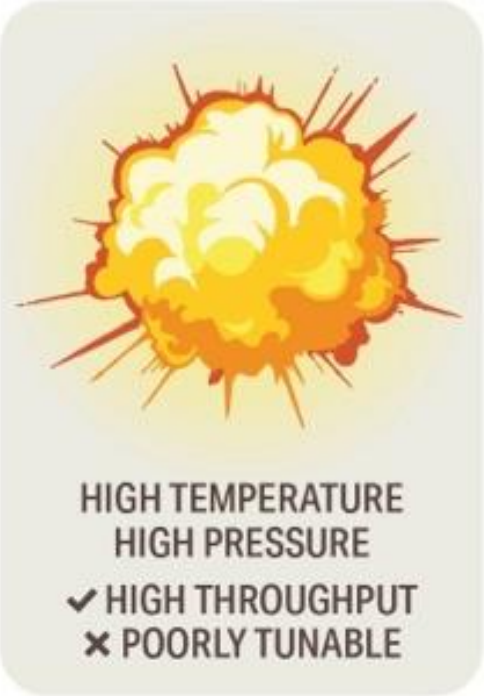
Carbon can form a manifold of technologically relevant nanomaterials

…but achieving tunable and scalable synthesis remains a grand challenge

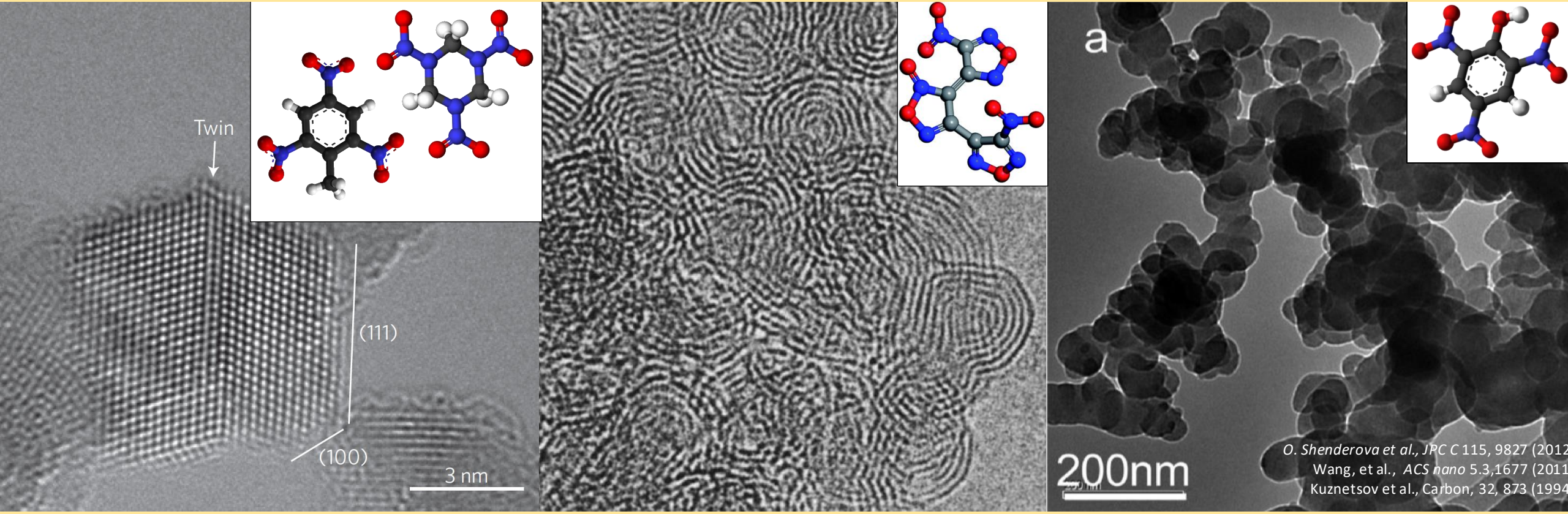
SAMPLE CNP USES	CNP TYPE		
	Amorphous	Graphitic	Diamond
Catalysis	✓	✓	
Quantum Computing		✓	
Battery	✓	✓	
Photovoltaics		✓	
Bioimaging			✓
Drug Delivery			✓



CNP: Carbon nanoparticle

Detonation holds promise as a high-throughput synthesis method

Detonation is extremely high throughput and can produce different carbon nanomaterials

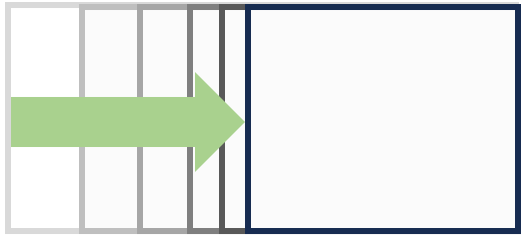


But this approach cannot be meaningfully tuned!

Hence, we developed a laser-driven, detonation inspired approach.

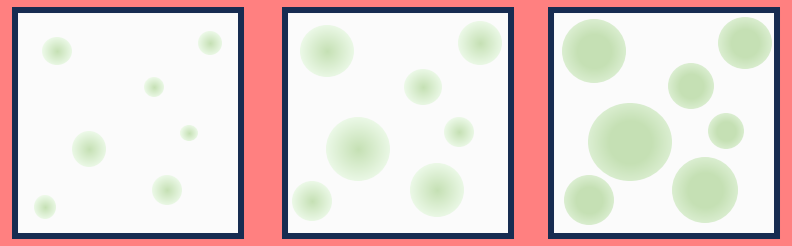
Laser-driven shock: A candidate strategy for fast, tunable CNP synthesis

Laser-driven shock compression



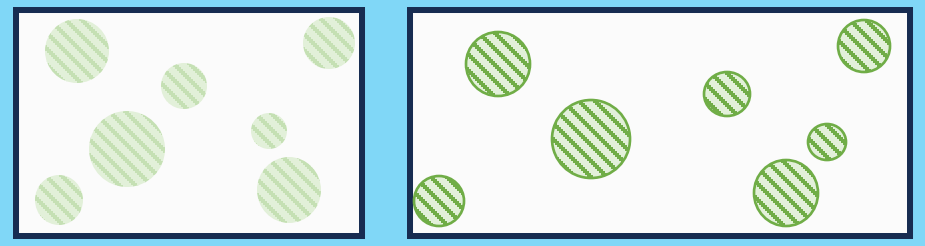
(1000s of K and 10s of GPa)

Evolution in Shocked State



(Ambient conditions)

Adiabatic Expansion



Reactive phase separation

L-CNP purification and freezing

L-CNP coarsening and purification

CNP freezing

***t* < 1 µs**

CNP: Carbon nanoparticle

L-CNP: Liquid CNP

Sounds great, but will it work?

Our Goal: Use molecular dynamics (MD) to assess feasibility

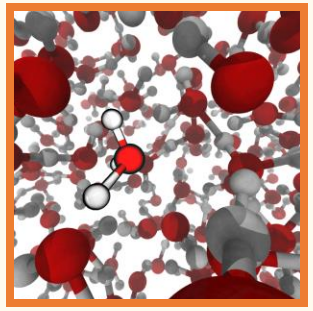
MD in a Nutshell

$$F = ma = -\frac{d\,E(r)}{dr}$$

Input

Initial atom coordinates

Interatomic model



Output

Dynamics
Equation of state
Thermochemistry
Microscopic response

The utility of atomistic simulations relies critically on interatomic model fidelity

… but how should these models be selected?

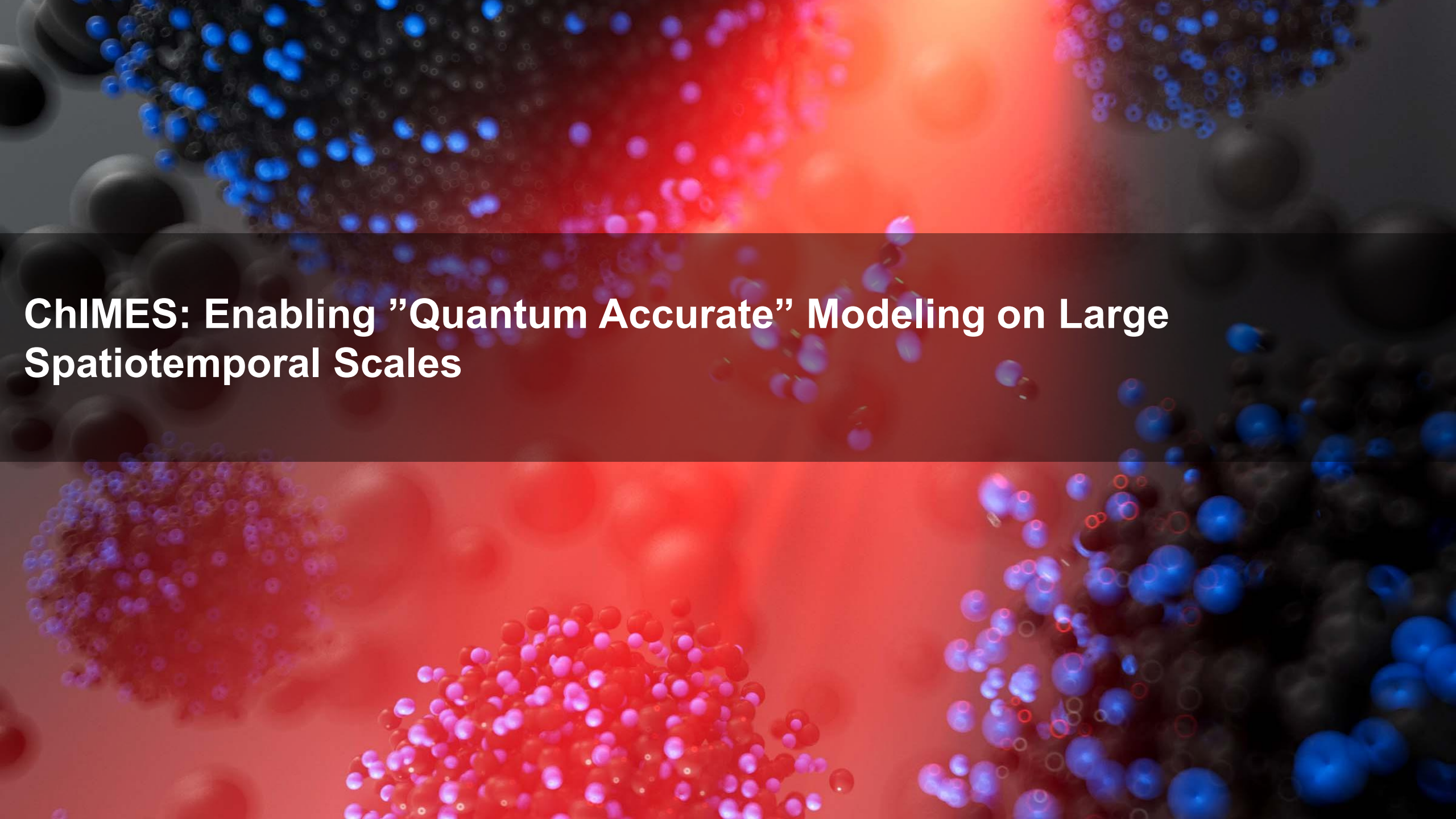
Simulating shockwave synthesis requires models that are fast *and* accurate

We want to enable predictive atomistic simulation of the **structure**, **dynamic response**, **speciation**, and **kinetics** associated with **materials and chemicals** under non-equilibrium conditions

Desired Model Features	QM	Classic Reactive IAMs
Can describe chemistry	Yes	Yes
Accurate at high T & p	Yes	Maybe
Can access large ℓ and t scales	No	Yes
Transferable/rapid and automated fit	–	No

IAM: Interatomic model

We develop machine-learned IAMs to bridge this capability gap



ChIMES: Enabling ”Quantum Accurate” Modeling on Large Spatiotemporal Scales

We developed ChIMES[1,2] to meet our unique needs

The Chebyshev Interaction Model for Efficient Simulation

*Corr. auth.

[1]**RKL***, et al., *JCTC* (2017)
[2]**RKL***, et al., *JCTC* (2019)

The generalized ChIMES equation

$$E_{n_B} = \sum_{i_1>i_2}^{n_a} {}^2E_{i_1 i_2} + \sum_{i_1>i_2>i_3}^{n_a} {}^3E_{i_1 i_2 i_3} + \cdots + \sum_{i_1>i_2\cdots i_{n_B-1}>i_{n_B}}^{n_a} {}^{n_B}E_{i_1 i_2 \cdots i_{n_B}}$$

***n*-body energy**

$${}^nE_{i_1 i_2 \cdots i_{n_B}} \propto \sum_{O}^{\mathcal{O}_n^*} c_O^E \prod_{y \in P} T_{m_3(y)}\left(s_{m_1(y)}^{m_2(y)}\right)$$

2-body energy (Single atom type)

$${}^2E_{ij} \propto \left[c_0 T_0^{ij}(s_{ij}) + c_1 T_1^{ij}(s_{ij}) + c_2 T_2^{ij}(s_{ij}) + \ldots + c_{\mathcal{O}_{2B}} T_{\mathcal{O}_{2B}}^{ij}(s_{ij})\right]$$

Fitting parameter

Transformed pair distance

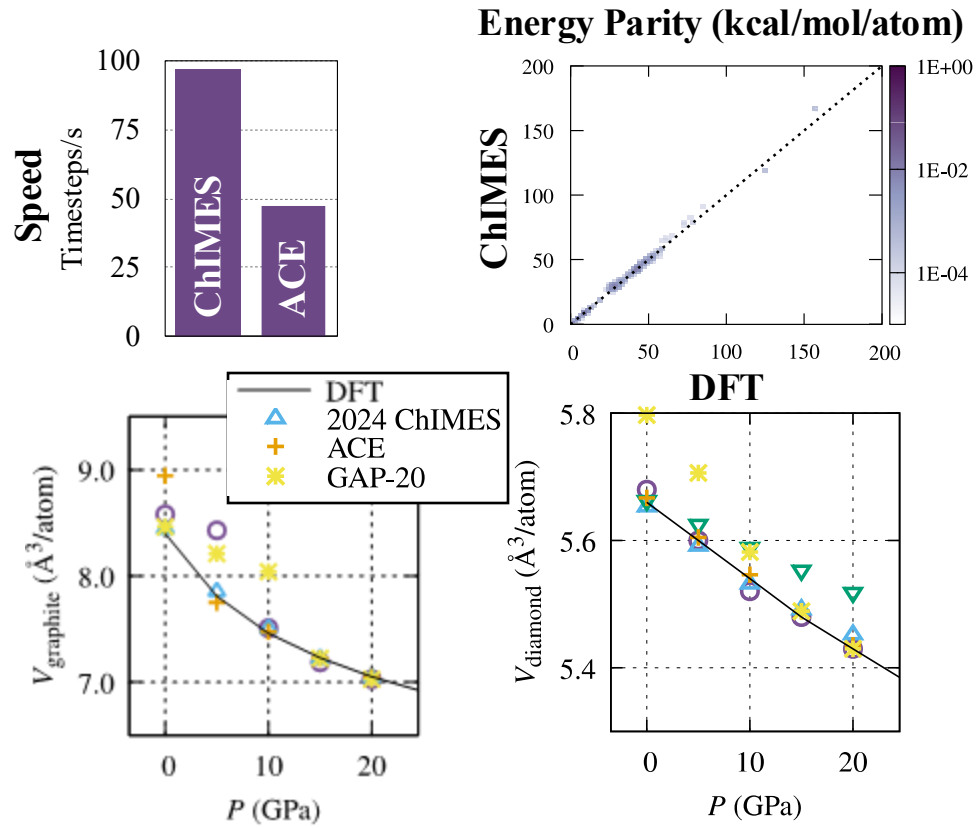
Polynomial of subscripted order

The Basic ChIMES Fitting Procedure

1. Build a training database from QM-MD simulations
2. Generate parameters via $\mathbf{min}\left|\boldsymbol{\varepsilon} = \frac{\partial \boldsymbol{F}^{\mathrm{ChIMES}\{c\}}}{\partial \boldsymbol{c}}\boldsymbol{c} - \boldsymbol{F}^{\mathrm{QM}}\right|$

RKL*, *et al.*, *npj Comput. Mater.* (2025)

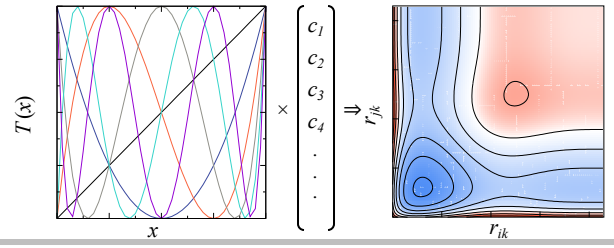
Sample Model Performance: ChIMES Carbon 2.0



ChIMES models are rapidly parameterizable and capable of "quantum accuracy" by design

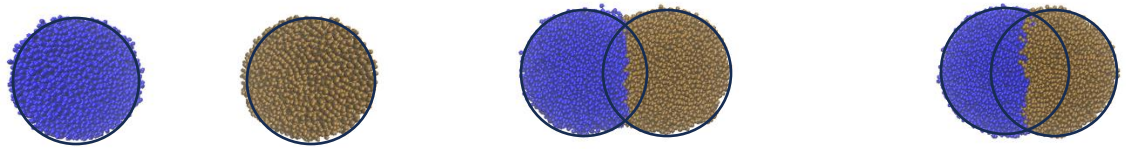
ChIMES is surprisingly versatile
Selected Use Cases and Applications

*Corresponding author(s)



Molten Carbon

R.K. Lindsey*, L.E. Fried, N. Goldman
JCTC, ***13*** *6222 (2017)*

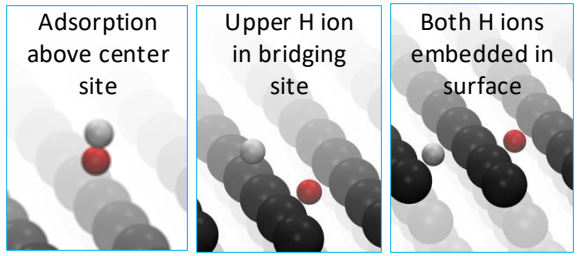


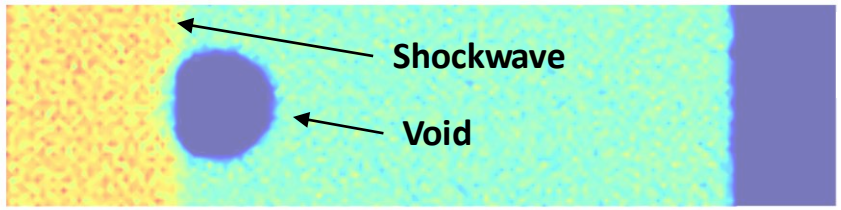
Inverse Design for Self-Assembling Nanomaterials (Coarse Grained & Alchemical ChIMES)

M. Zhang, S.C. Glotzer, **R.K. Lindsey*** (Under review: *JCTC*)

Pu Hydrides (ChIMES-Corrected DFTB)

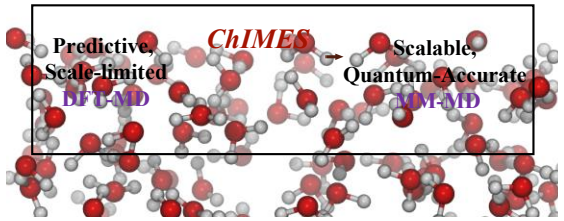
N. Goldman*, B. Aradi, **R.K. Lindsey**, L.E. Fried
JCTC, **14** 2652 (2018)





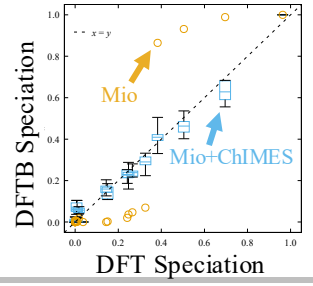
Hydrazoic Acid

H. Pham*, **R.K. Lindsey**, L.E. Fried, N. Goldman
JCP, ***153*** *224102 (2020)*



Ambient Water

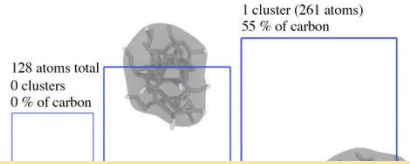
R.K. Lindsey*, L.E. Fried, N. Goldman
JCTC, **15** 436 (2019)



3,4-bis(3-nitrofurazan-4-yl)furoxan (ChIMES-Corrected DFTB)

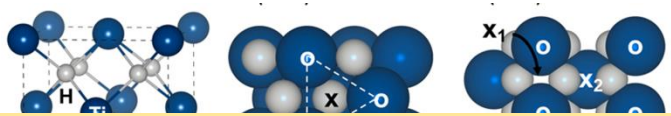
R.K. Lindsey*, S. Bastea, N. Goldman, L.E. Fried,
J. Chem. Phys. **154** 164115 (2021)

C/O Under Extreme Conditions



Titanium Hydrides

N. Goldman*, K.E. Kweon, B. Sadigh, T. W. Heo, **R.K. Lindsey**, C.H. Pham,

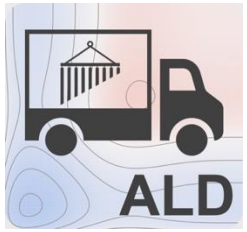


ChIMES is helping us tackle real problems and foray into previously inaccessible problem spaces

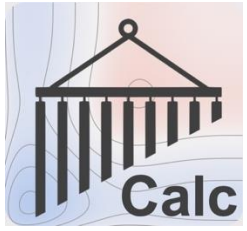
Model development efforts are enabled through the ChIMES software suite

[1]**RKL*** *et al., JCP* (2020)
[2]**RKL*** *et al., JCP* (2021)
[3]**RKL*** *et al., npj Comput. Mater.* (2025)
[4]**RKL*** *et al., npj Comput. Mater.* (2025)
[5]B.R. Laubach and **RKL***, Under Review: *JCIM*
**Corr. auth.*

Input: Training set
Output: Untested model



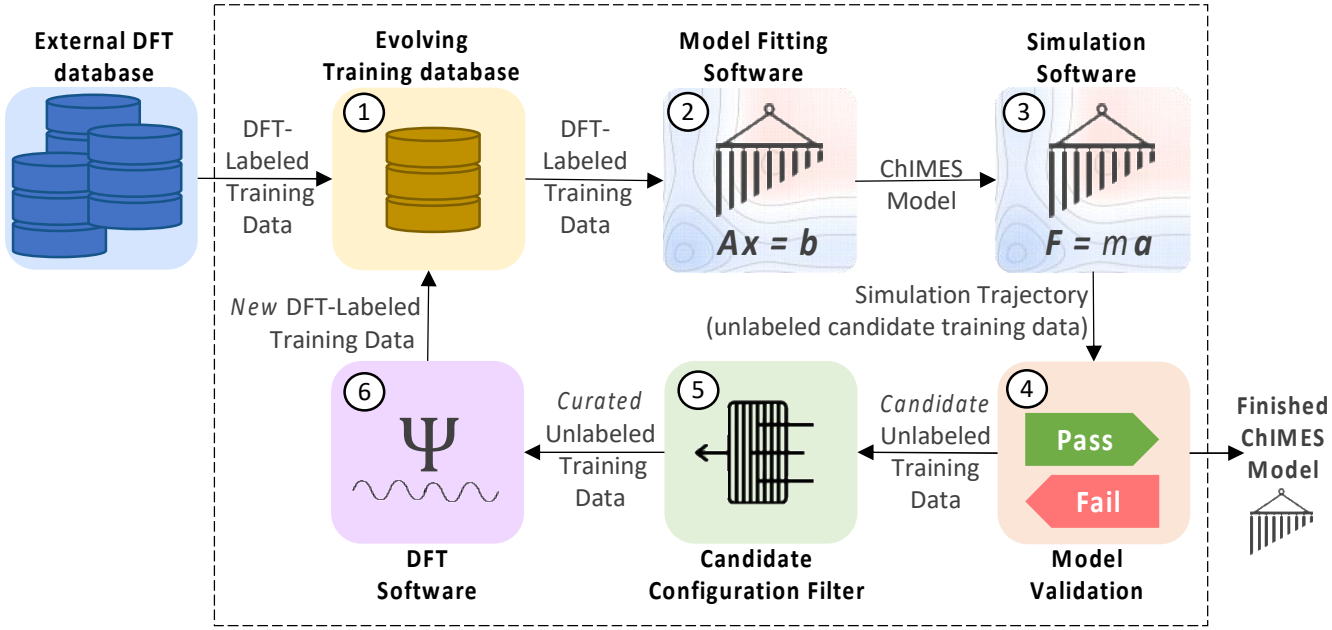
Input: Initial training set
Output: Stable/tested model



Input: ChIMES Model
Output: Forces/Energies/Stresses
Also: LAMMPS integration*

`(stable)` `github.com/rk-lindsey`
`(development)` `github.com/LindseyLab-umich`

The ChIMES Active Learning Driver
(a modular ChIMES development workflow tool)

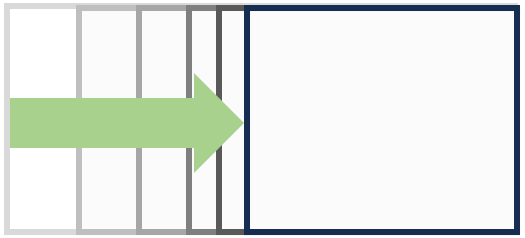


- **Features:**
  - Parallel learning
  - Transfer learning
  - Delta learning
  - Active learning
  - UQ (coming soon!)
- **Support for:**
  - VASP
  - CP2K
  - DFTB+
  - LAMMPS
  - ChIMES MD

Back to our application:
Understanding Shockwave-Driven Nanocarbon Synthesis

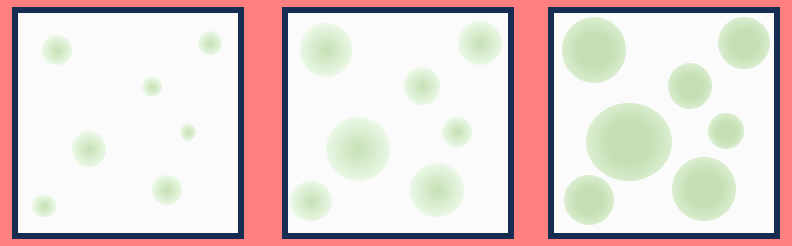
Our goal: Deploy ChIMES in a reductionist study to elucidate this phenomenon

Laser-driven shock compression



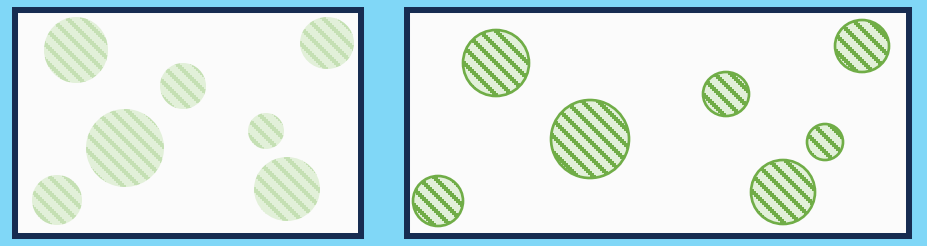
(1000s of K and 10s of GPa)

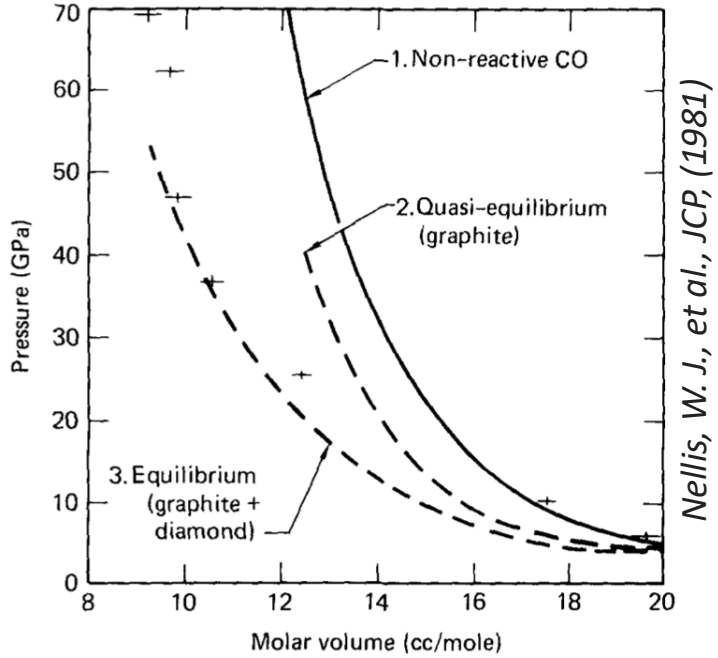
Evolution in Shocked State



(Ambient conditions)

Adiabatic Expansion





Nellis, W. J., et al., JCP, (1981)

Drawing inspiration from previous shock experiments, we used ChIMES to

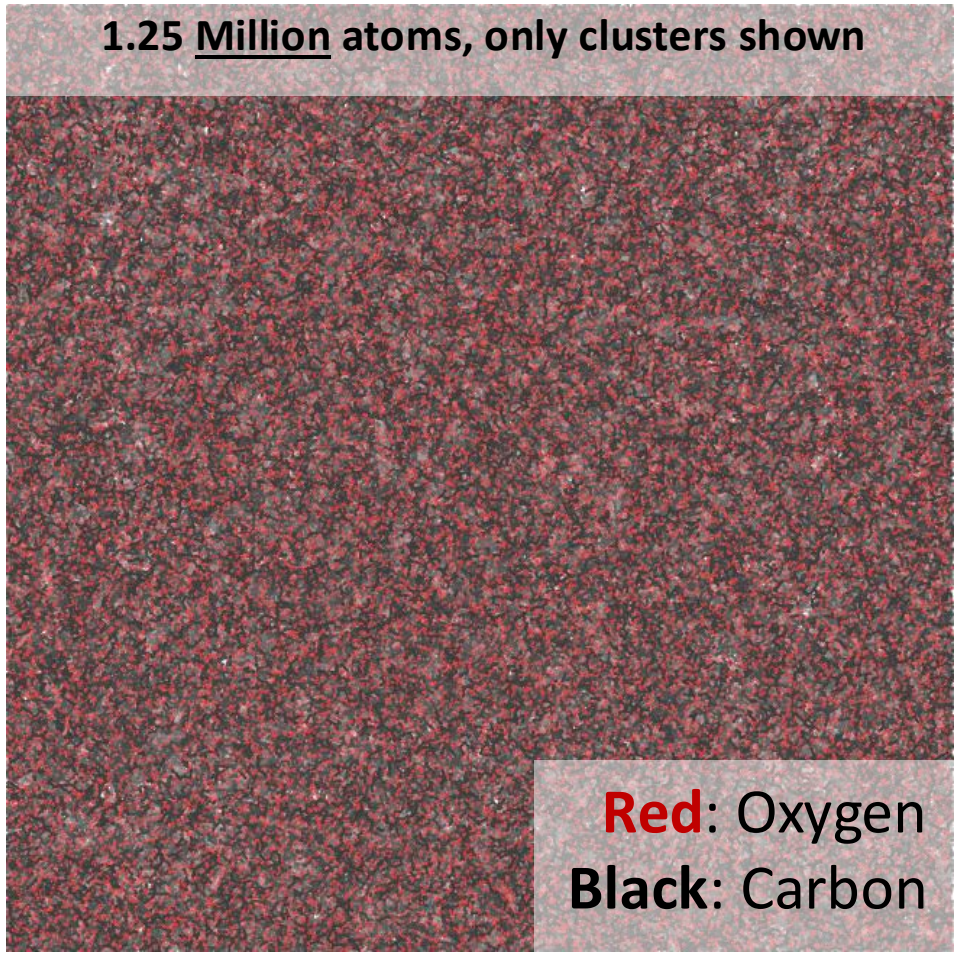
(1) Predict whether carbon nanoparticles can form from shocked cryogenic CO

(2) Investigate the underlying phenomena

ChIMES enabled atomically resolved simulation shockwave-driven CNP synthesis from C/O on scales overlapping with experiment

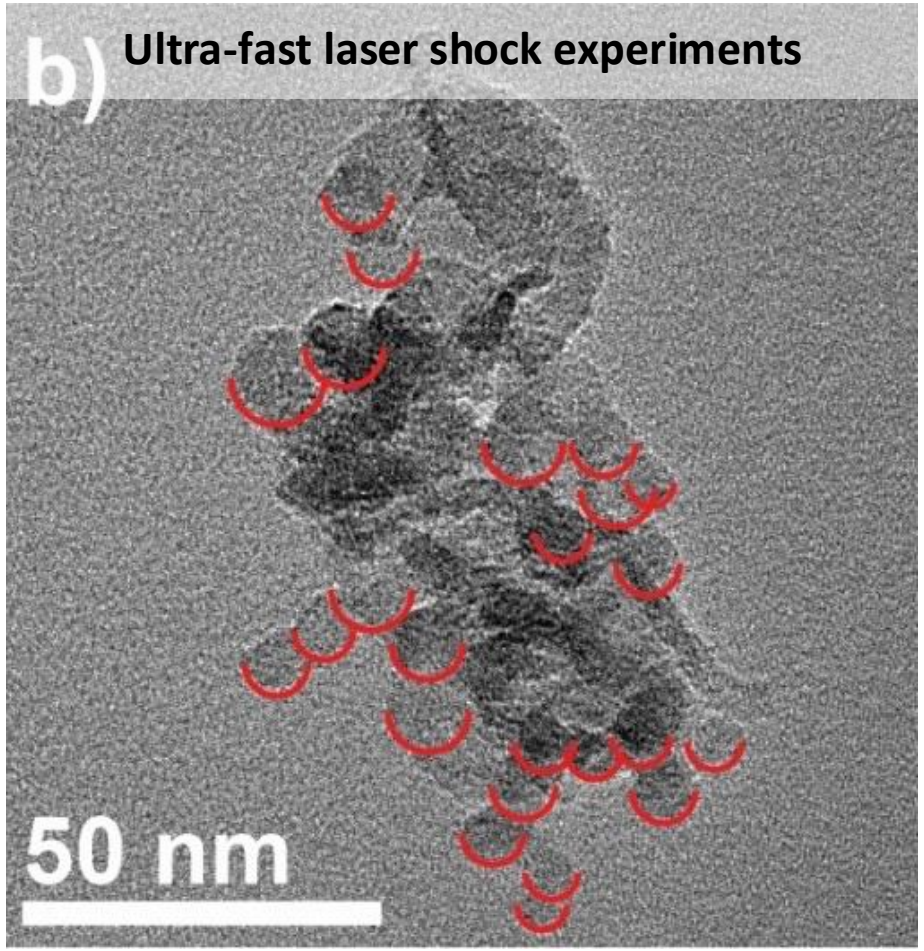
*Corr. auth.

5 nm



QM limit

1.25 × 10² Atoms
(1.1 nm, 0 Clusters)

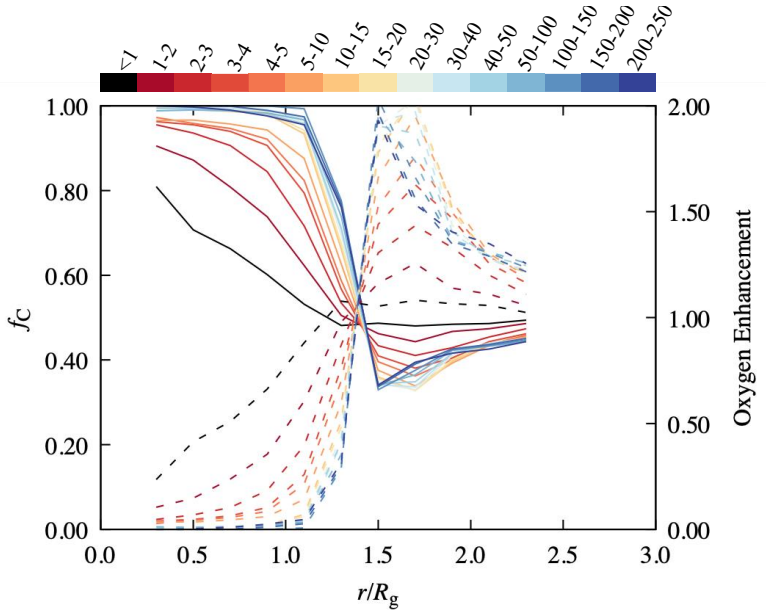


RKL*, et al. *Nat. Commun.* **13**, 1424 (2022).
RKL*, et al. *JCP,* **153** 054103 (2020).
M.R. Armstrong*, **RKL***, … , S. Bastea* *Nat. Commun.* **11**, 353 (2020).

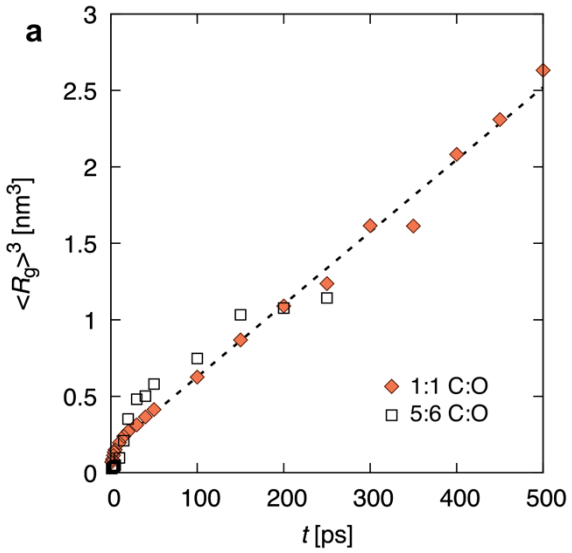
Our simulations also provided a critical, atomistically-resolved glimpse into underlying chemical mechanisms and kinetics

*Corr. auth.

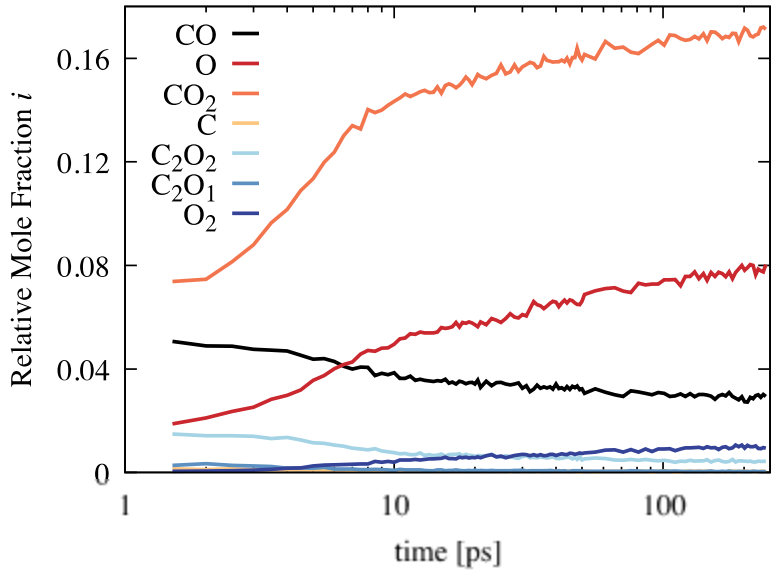
RKL*, N. Goldman, L.E. Fried, S. Bastea, *Nat. Commun.* (2022)



Time-resolved cluster composition



Cluster size growth kinetics



Time-resolved composition of surrounding fluid

Our simulations provide insight into strategies for *tunable* shock synthesis

To make CNP shock-synthesis a *tunable* process, we need to enhance understanding of the underlying phenomena

Open questions include:

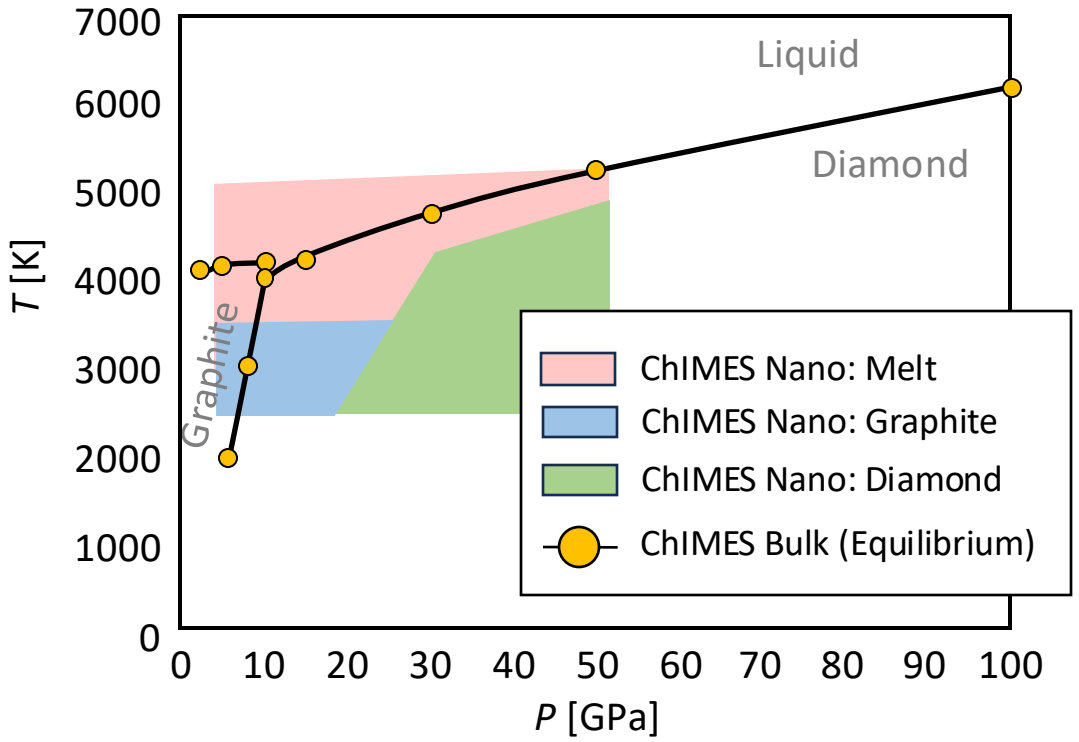
1. Where are the carbon phase boundaries?
2. What are the kinetics for transformation across them?
3. How to these boundaries/kinetics change for finite system sizes?

4. What is the fundamental behavior of elemental systems under extreme T/P?
5. How does chemical composition mediate this behavior?

Guiding question:
How do phase boundaries shift for finite-sized systems?

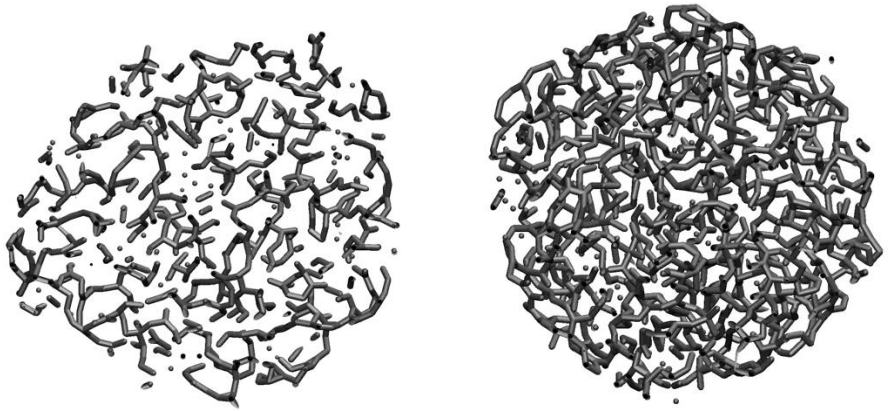
*Corr. auth.

Carbon Phase Diagram



Solidification of Liquid Carbon Nanoparticles at Constant *T* and *P*

4 nm nanoparticles (cross-sections)



3500 K, 15 GPa **3500 K, 50 GPa**

RKL* et al, npj Comput. Mater. (2025)
Y. Lyu, …, **RKL***, *in prep*

Our simulations are allowing us to construct size-dependent carbon nanoparticle phase diagrams

Guiding question:
What are the mechanisms and kinetics of solidification?

3500 K, 50 GPa, 0.5 ns

4 nm CNP
(cross section)

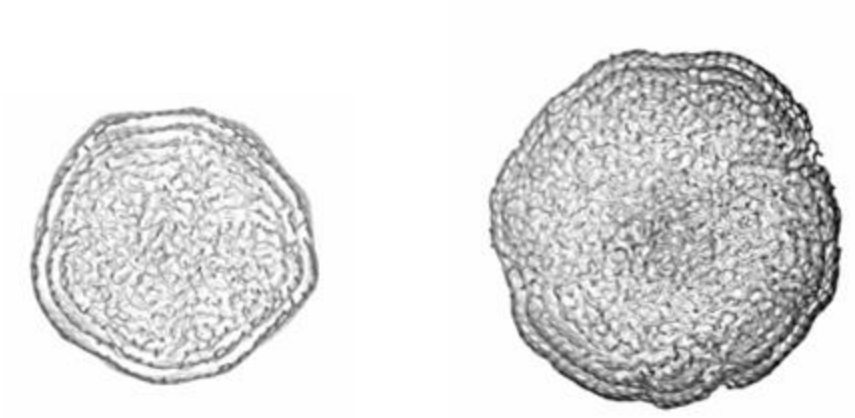
10 nm CNP
(whole particle)

Classical nucleation

3000 K, 50 GPa, 0.5 ns

6 nm CNP
(cross section)

8 nm CNP
(cross section)



Non-classical nucleation (2-step)

Small changes in temperature and pressure can drive large changes in the solidification process

Funding

U.S. DEPARTMENT OF ENERGY
National Nuclear Security Administration
LDRD
GRFP

Collaborators

Sorin Bastea (LLNL)
Sebastien Hamel (LLNL)
Nir Goldman (LLNL)
Huy Pham (LLNL)
Larry Fried (LLNL)
Vince Lordi (LLNL)

Software

```
chimes_lsq
chimes_calculator
al_driver
```

Concluding Remarks

- ChIMES enables “quantum accurate” atomically resolved simulation on unprecedented scales
- Using these tools, we are establishing a comprehensive description of coupling between chemistry, miscibility, and phase transformation in carbon-rich materials under extreme conditions

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https://lindseylab.engin.umich.edu

LINDSEY AB