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Shock Hugoniot Relationships for Crystalline and Amorphous HNAB: Insights from Atomistic Simulations and Virtual Diffraction Calculations

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Motivation and Objectives

Facts on Crystalline and Amorphous HNAB:

- Amorphous energetic materials, such as hexanitroazobenzene (HNAB), have been experimentally observed to undergo crystallization when subjected to sufficient shock compression.
- Experimentally characterizing the shock-induced crystallization of A-HNAB is difficult:
  - HNAB has three (3) crystalline polymorphs.
  - Equation-of-state (EOS) not well studied for crystalline structures and unknown for the amorphous state.
  - Crystallization occurs at the nanosecond timescales.

Left: Vapor deposited A-HNAB. **Right:** Shocked A-HNAB samples via flyer plate experiments illustrating shock induced crystallization.

- Simulating shock behavior and microstructural changes under shock compression for HNAB is suitable for atomistic simulations:
  - Length- and time-scales appropriate for molecular dynamics (MD) to study transition pathways for all HNAB polymorphs.

Objectives of this work:

1. **Calculate shock Hugoniot EOS for all HNAB polymorphs** using ReaxFF-MD to probe the role of atomic configuration on macro-scale shock properties. Is an amorphous Hugoniot feasible?
2. Perform virtual diffraction calculations to **elucidate possible structural changes occurring during simulated shock compression** that may also provide a one-to-one comparison with shock experiments.

References

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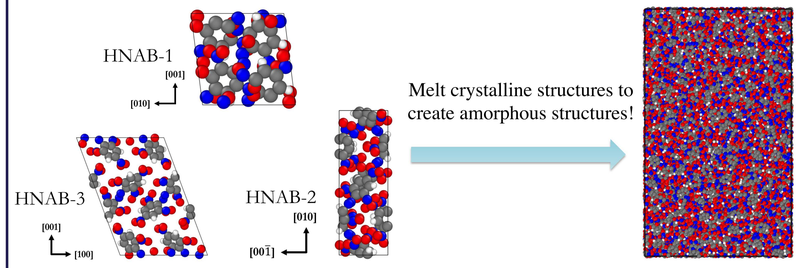
Finding the Hugoniot State (P, T, E) for Any V

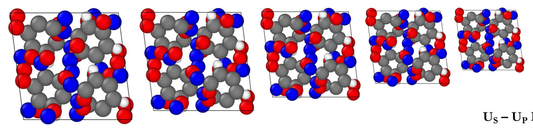
HNAB Reference Structures at Experimental Densities



Approach: 1. Set $V < V_0$ and equilibrate to ~ 300 K
2. Ramp system T and track pressure, energy, density
3. Solve for T where Rankine-Hugoniot condition is true

Series of compressed states for all polymorphs

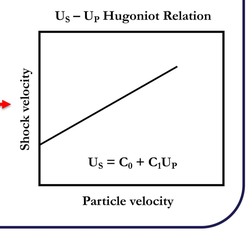
$V = V_0$ → $V < V_0$



Mass Conservation: $\rho_0 D = \rho_1 (D - u_1)$

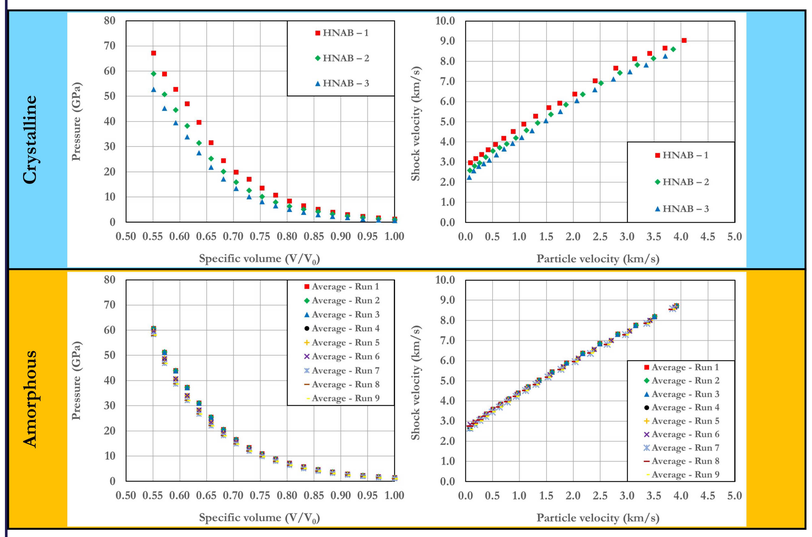
Momentum Conservation: $P_1 = \rho_0 D u_1$

Energy Conservation: $E - E_0 = \frac{1}{2}(P + P_0)(V_0 - V)$



Predicting Shock Hugoniot Relationships

Computational Point-of-view | Experimental Point-of-view



- **Crystalline:** Each HNAB polymorph has its own unique shock relationship and behavior at high pressures.
- **Amorphous:** Non-negligible macro-scale variations in results from both points-of-view due to atomic configuration only.
- Calculated behavior provides EOS input and sensitivity for device-level calculations and guide to experimental analysis.

Simulating Virtual Diffraction from MD

1. MD simulations of hydrostatic compression for HNAB polymorphs
2. Create a mesh of reciprocal space within simulation domain
   - 3D rectilinear mesh with fine resolution eliminates any need for prior knowledge of the crystal structure
3. Compute diffraction intensities at each point on the reciprocal space mesh using the structure factor equations
   - Diffraction conditions satisfied with nodes located on Ewald sphere
4. Analysis and visualization of diffraction intensities to produce Selected Area Electron Diffraction (SAED) patterns.

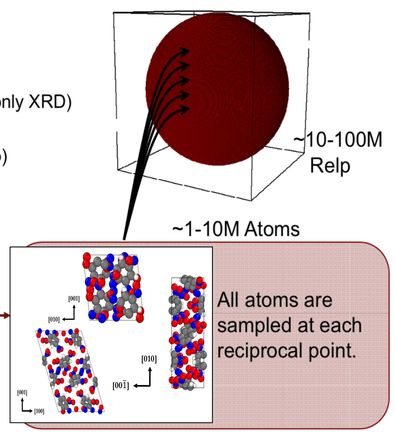
$$I(\mathbf{K}) = \mathrm{Lp} F(\mathbf{K}) F^*(\mathbf{K})$$

Lp = Lorentz-Polarization Factor (only XRD)
F = Structure Facture
$\mathbf{K}$ = Reciprocal Lattice Point (Relp)

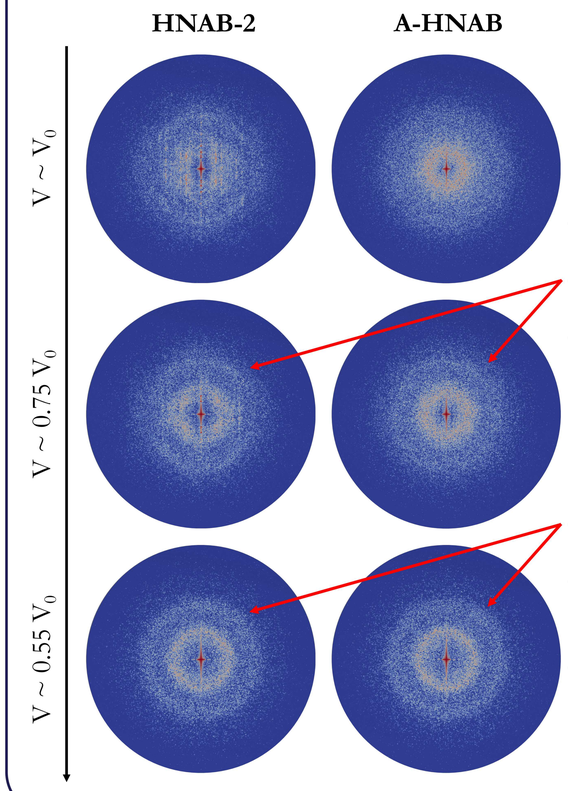
Structure Factor:

$$F(\mathbf{K}) = \sum_{j=1}^{N} f_j \exp(2\pi i \, \mathbf{K} \cdot \mathbf{r}_j)$$

N = Number of Atoms in Simulation
f_j = Atomic Scattering Factor
$\mathbf{r}_j$ = Atom Position



Elucidating Structural Changes during Shock



- Crystalline "peaks" becoming smeared.
- Formation of secondary rings.
- Outward expansion due to compression.
- Similar final "double ring" patterns.
- Possibly indicating a reordering of the compact structure.
- XRD and RDF calculations may help clarify this observation.

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