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Micron-scale Reactive Atomistic Simulation of Void Collapse and Hotspot Growth in pentaerythritol tetranitrate (PETN)

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Sandia National Laboratories, New Mexico

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U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

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Introduction

- Material defects and heterogeneities such as dislocations, grain boundaries, entrained gas, and porosity play key roles in the shock-induced initiation of detonation in energetic materials.
- Previously, we have performed reactive NEMD simulations of weak shocks in a $60 \times 40 \times 40$ nm^3 PETN crystal containing a 20 nm spherical void using LAMMPS/ReaxFF. We observed hotspot formation and an exothermic reaction zone.

Introduction

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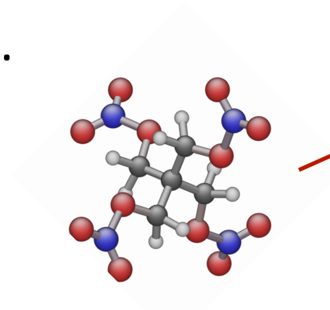
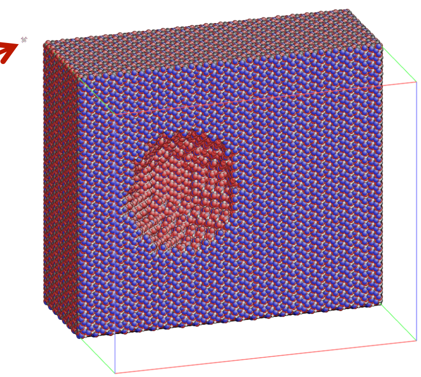
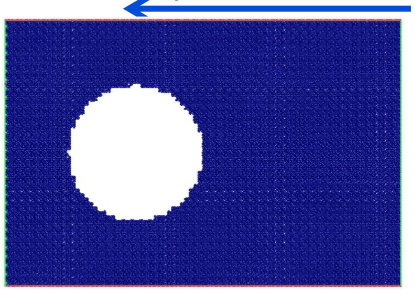
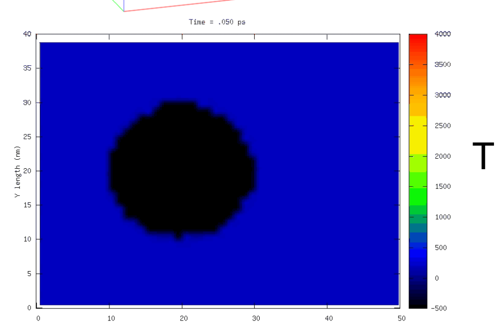
- Previously, we have performed reactive NEMD simulations of weak shocks in a 60 × 40 × 40 nm^3 PETN crystal containing a 20 nm spherical void using LAMMPS/ReaxFF. We observed hotspot formation and an exothermic reaction zone.





U_P = **2** km/s



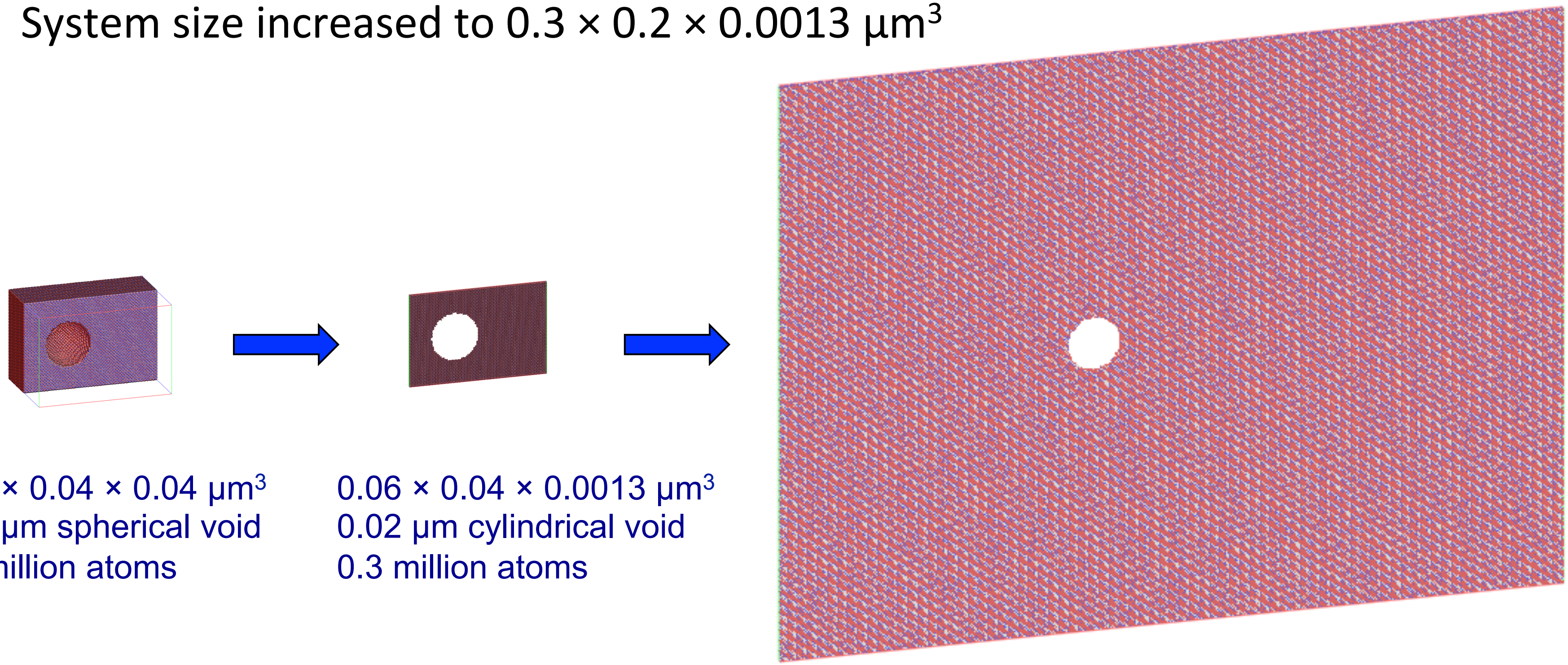


A. P. **Thompson**, T.-R. **Shan**, "*Shock-induced hotspot formation and chemical reaction initiation in PETN containing a spherical void* ", Proc. 2013 APS-SCCM

- **Formation of hotspot observed, but is it growing? Is it leading to detonation?**

Computational setup

- Reduce void dimension from 3D spherical to 2D cylindrical
- System size increased to 0.3 × 0.2 × 0.0013 µm^3

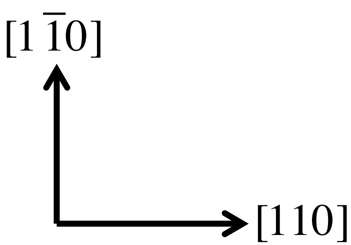


0.06 × 0.04 × 0.04 µm^3
0.02 µm spherical void
8.9 million atoms

0.06 × 0.04 × 0.0013 µm^3
0.02 µm cylindrical void
0.3 million atoms

0.3 × 0.2 × 0.0013 µm^3
0.02 µm cylindrical void
8.5 million atoms

0.1 µm

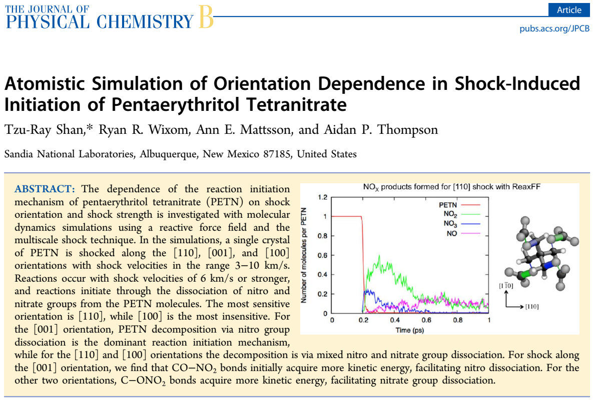


Objective: To observe hotspot growth and identify growth mechanism

Computational methods

- ReaxFF

$$
\begin{aligned}
E^{ReaxFF} = \; & E^{self} + E^{Coul} + E^{vdW} + E^{bond} + E^{angle} + E^{torsion} \\
& + E^{conjugation} + E^{H-bond} + E^{lone-pair} + E^{over} + E^{under} \\
& + E^{others}
\end{aligned}
$$


THE JOURNAL OF PHYSICAL CHEMISTRY B

Article

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Atomistic Simulation of Orientation Dependence in Shock-Induced Initiation of Pentaerythritol Tetranitrate

Tzu-Ray Shan,* Ryan R. Wixom, Ann E. Mattsson, and Aidan P. Thompson

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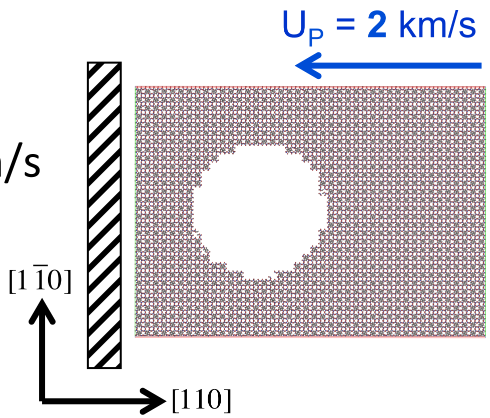
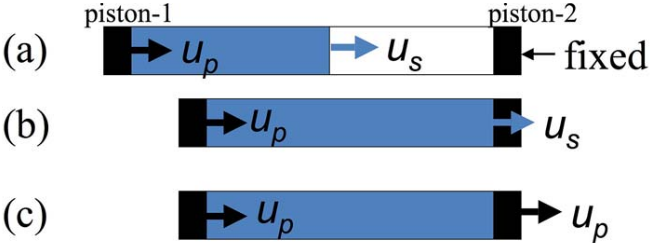
ABSTRACT: The dependence of the reaction initiation mechanism of pentaerythritol tetranitrate (PETN) on shock orientation and shock strength is investigated with molecular dynamics simulations using a reactive force field and the multiscale shock technique. In the simulations, a single crystal of PETN is shocked along the [110], [001], and [100] orientations with shock velocities in the range 3−10 km/s. Reactions occur with shock velocities of 6 km/s or stronger, and reactions initiate through the dissociation of nitro and nitrate groups from the PETN molecules. The most sensitive orientation is [110], while [100] is the most insensitive. For the [001] orientation, PETN decomposition via nitro group dissociation is the dominant reaction initiation mechanism, while for the [110] and [100] orientations the decomposition is via mixed nitro and nitrate group dissociation. For shock along the [001] orientation, we find that CO−NO_2 bonds initially acquire more kinetic energy, facilitating nitro dissociation. For the other two orientations, C−ONO_2 bonds acquire more kinetic energy, facilitating nitrate group dissociation.

T.-R. **Shan**, R. R. **Wixom**, A. E. **Mattsson**, A. P. **Thompson**, J. Phys. Chem. B, 117, 3 928-936 (2013)

- NEMD
  - Impact velocity = 2 km/s
  - Impact velocity = 1.25 km/s



- Shock-front absorbing boundary condition (ABC)

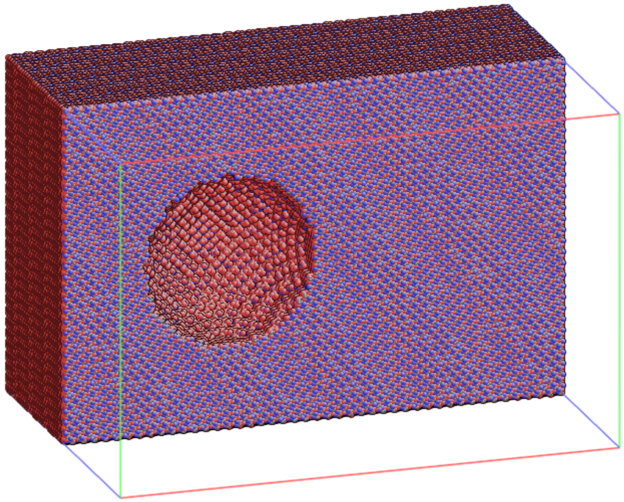


A. V. **Bolesta**, L. **Zheng**, D. L. **Thompson**, and T. D. **Sewell**, *Phys. Rev. B* 76, 224108 (2007).

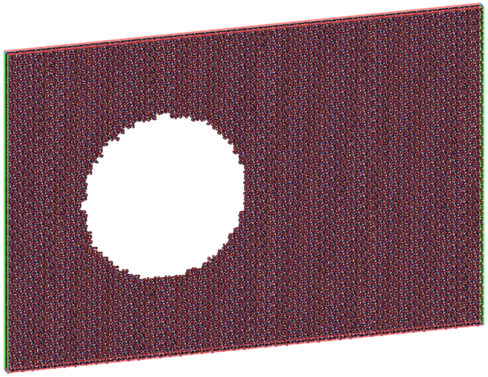
Results: spherical vs. cylindrical voids

- Comparing particle and shock velocity

spherical



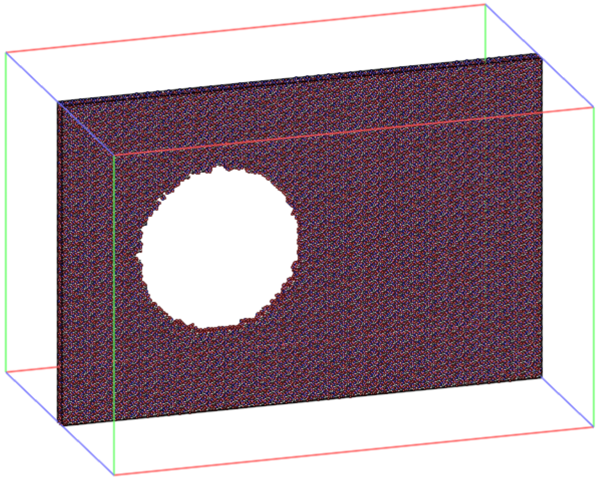
cylindrical



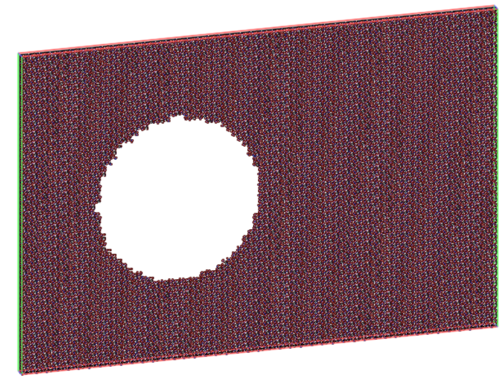
Results: spherical vs. cylindrical voids

- Co ity

spherical



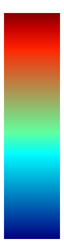
cylindrical

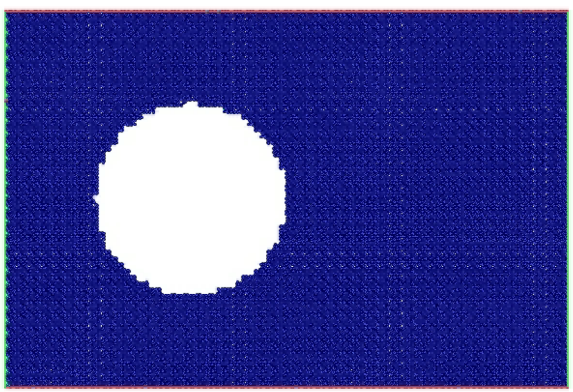


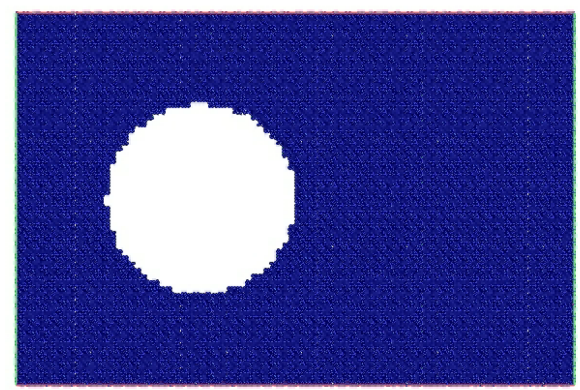
viewing one slice

$U_S \approx 5.7$ km/s

$U_S \approx 5.6$ km/s





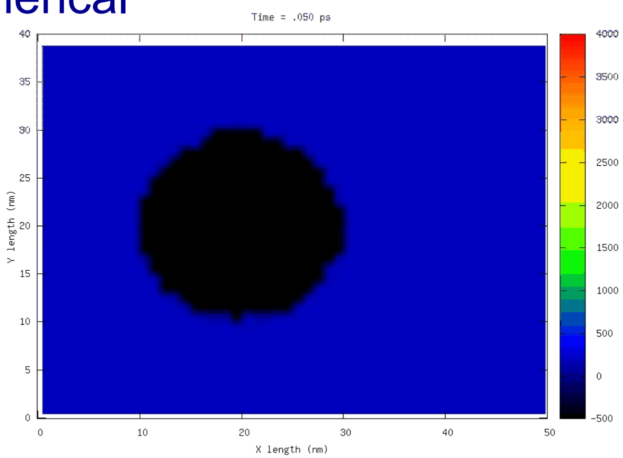


- **Cylindrical void with slightly slower shock velocity**
  - **Due to weaker ejecta focusing effect**

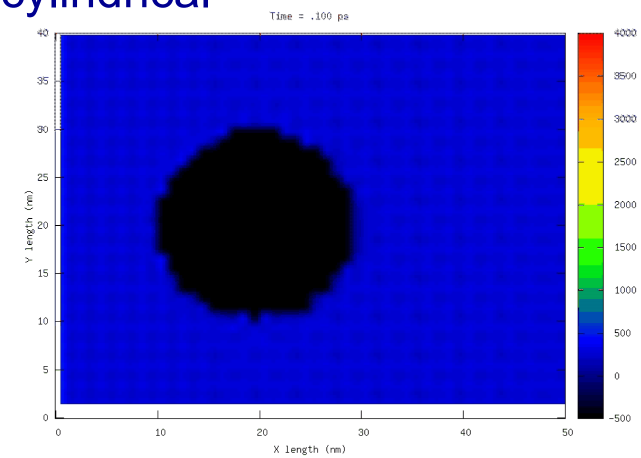
Results: spherical vs. cylindrical voids

- Comparing system and hotspot temperature

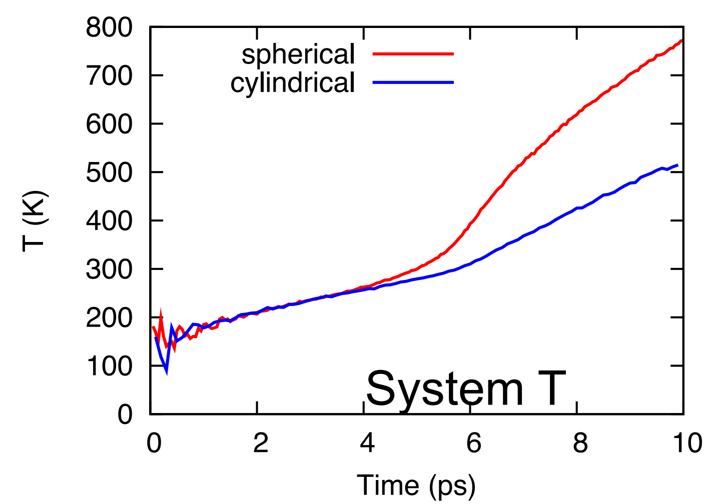
spherical



cylindrical



- Cylindrical void with lower temperature and smaller hotspot
  - Can be mitigated with a stronger impact velocity
- Despite these differences, qualitative void collapse and hotspot formation mechanisms are similar

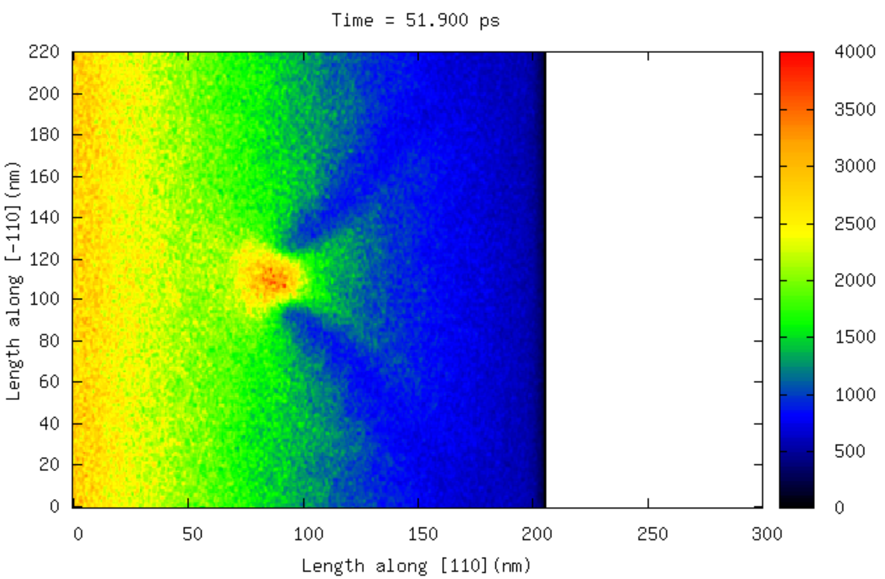


Results: micron-scale system

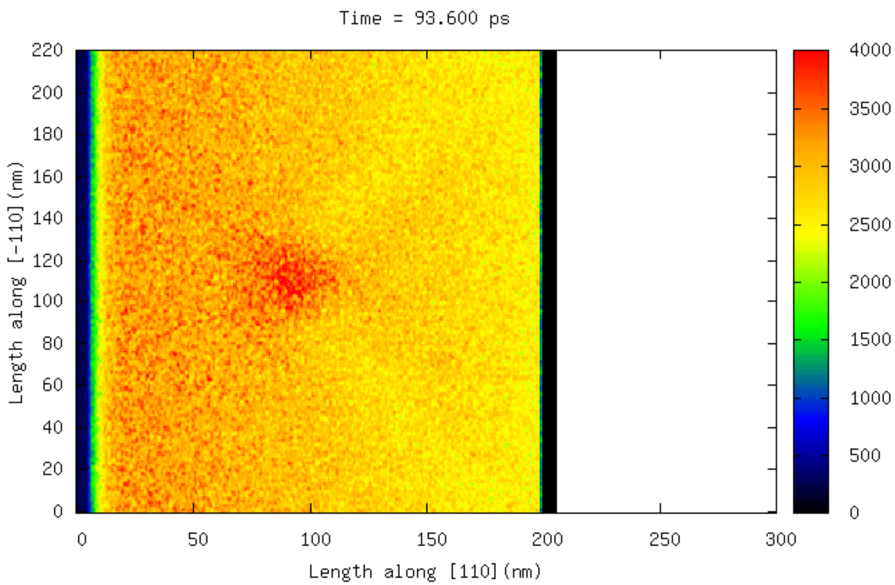
- **Impact velocity = 2 km/s**
- Normal NEMD shock run from 0 – 52 ps
  - Observed hotspot formation due to void collapse
- Shock-front ABC run from 52 – 94 ps:
  - Observed hotspot growth due to coupling to exothermic chemical reactions
  - However, heating from the wall catch up with the growing hotspot

0.3 × 0.2 × 0.0013 μm^3
0.02 μm cylindrical void
8.5 million atoms

52 ps



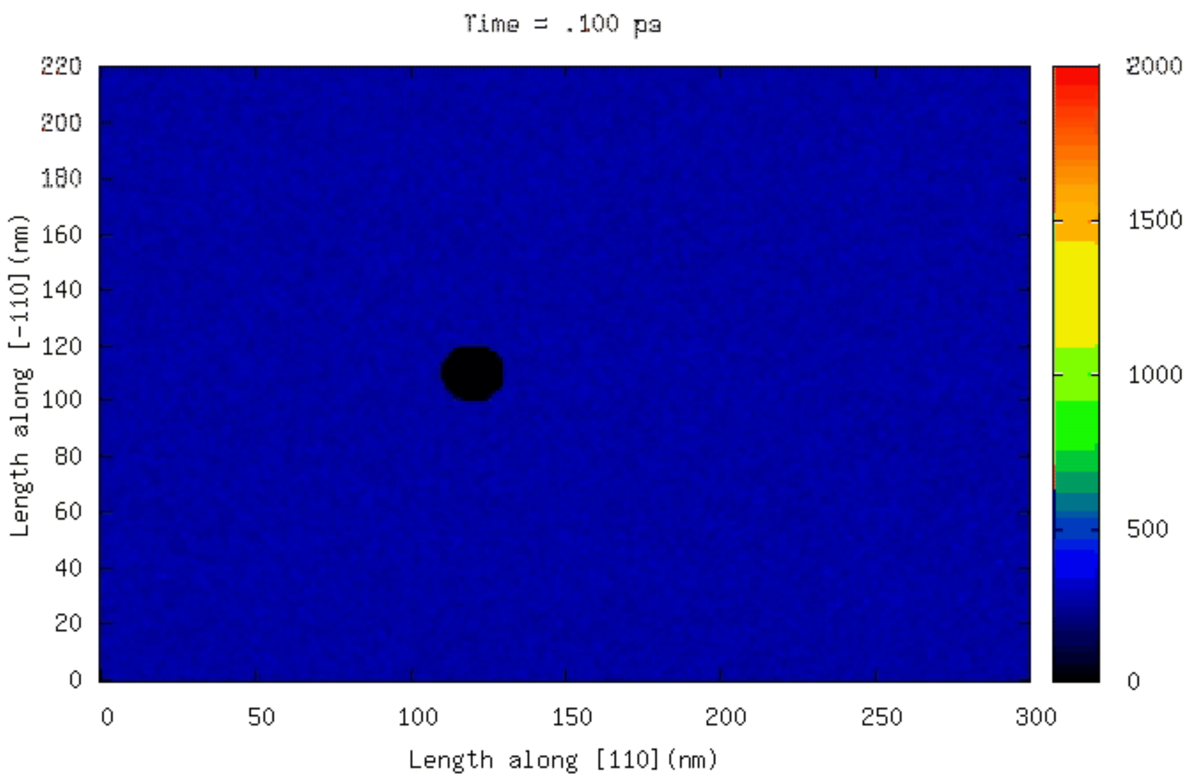
94 ps



Results: micron-scale system

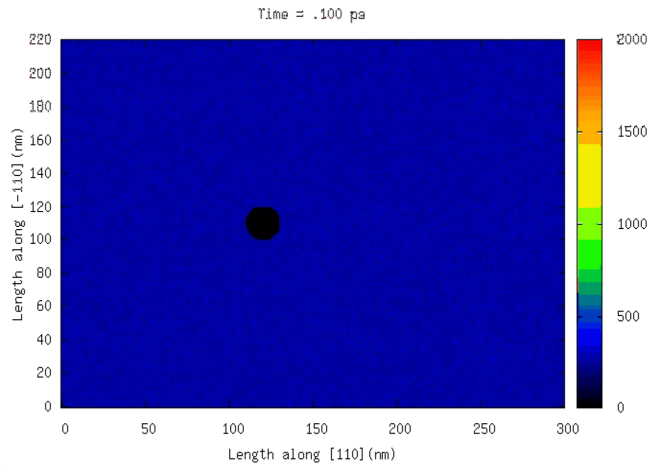
- **Impact velocity reduced to 1.25 km/s**
- Normal NEMD shock run from 0 – 64 ps
  - Observed hotspot formation due to void collapse
- Shock-front ABC run from 64 – 195 ps:
  - Observed hotspot growth due to coupling to exothermic chemical reactions

0.3 × 0.2 × 0.0013 μm^3
0.02 μm cylindrical void
8.5 million atoms

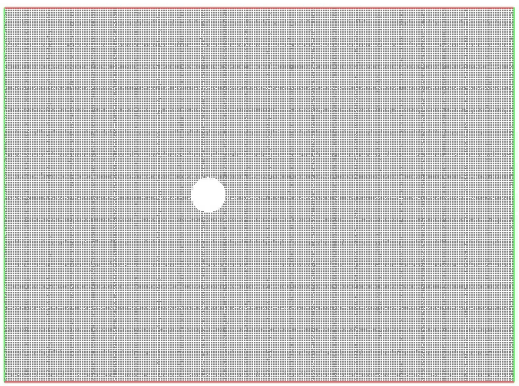


Coupling between hotspot growth and exothermic chemical reactions

Temperature



PETN



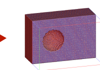
H_2O

NO_2

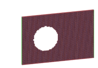
CO_2

Conclusions

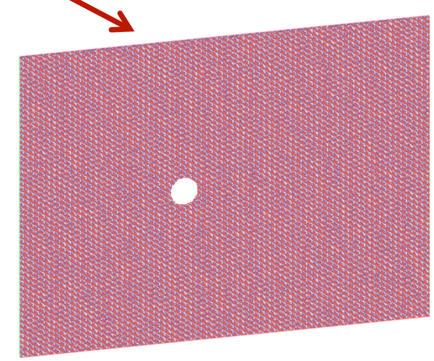
- Performed NEMD shock simulations with shock-front ABC using LAMMPS/ReaxFF
- Observed formation of hotspot due to void collapse in the 0.06 × 0.04 × 0.04 μm^3 cell containing a 20 nm spherical void



  - Similar qualitative void collapse and hotspot formation behaviors observed in the 0.06 × 0.04 × 0.0013 μm^3 cell containing a 20 nm cylindrical void



  - Smaller hotspot and lower temperature due to weaker ejecta focusing effect
- In the 0.3 × 0.2 × 0.0013 μm^3 cell containing a 20 nm cylindrical void



  - Observed hotspot formation and hotspot growth
  - Self-sustained hotspot growth coupled to exothermic chemical reactions