GAB Meeting

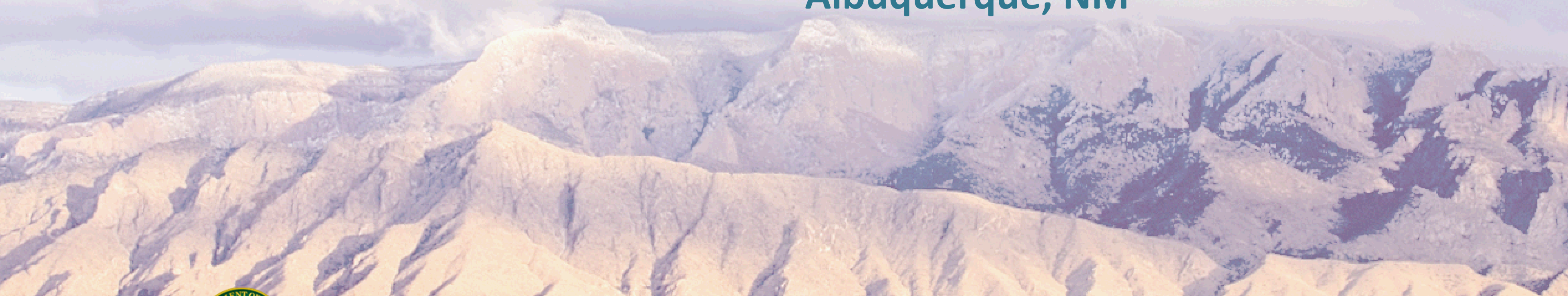
SAND2014-16970PE

June 2014

Molecular dynamics simulation of quench, stress relaxation and fracture in silica melts and glasses

J. Matthew D. Lane

Sandia National Laboratories,
Albuquerque, NM

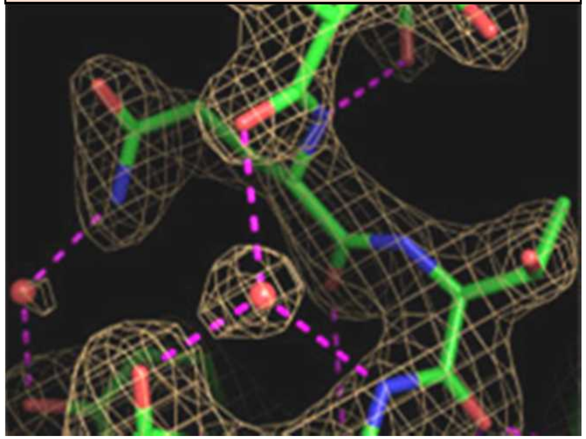


Sandia National Laboratories is a multi program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

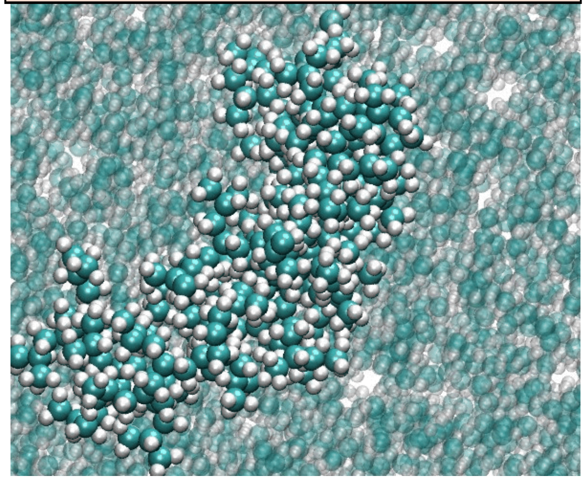
Hierarchy of modeling approaches

Fracture response in glass depends on processes and structure at several length scales:

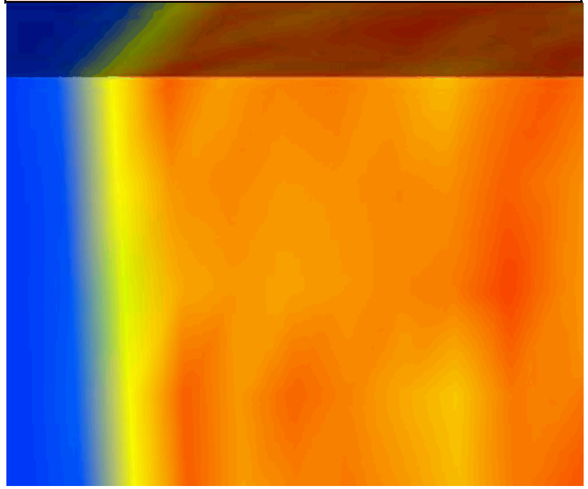
Quantum scale
fs times & Å lengths



Molecular scale
ns times & 10s nm lengths



Continuum scale
>μs times & >mm lengths



Increasing time and length scales

Molecular Dynamics (MD) simulations

- Solve Newton's equation…

$$m_i \frac{d^2\mathbf{r}_i}{dt^2} = \mathbf{F}_i$$

$$\mathbf{F}_i = -\nabla V_i$$

6*N* coupled ODEs

$$\begin{aligned}
\mathbf{v}_i^{(l+\frac{1}{2})} &= \mathbf{v}_i^l - \frac{\mathbf{F}_i^l}{2m_i}\Delta t \\
\mathbf{r}_i^{(l+1)} &= \mathbf{r}_i^l + \mathbf{v}_i^{(l+\frac{1}{2})}\Delta t \\
\mathbf{v}_i^{(l+1)} &= \mathbf{v}_i^{(l+\frac{1}{2})} - \frac{\mathbf{F}_i^{(l+1)}}{2m_i}\Delta t
\end{aligned}$$

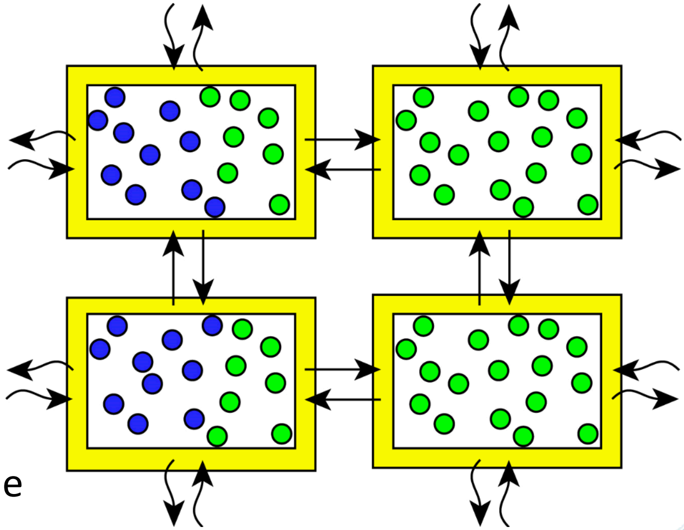
Mathematical Formulation

Classical Mechanics

Atoms are Point Masses: $\boldsymbol{r}_1, \boldsymbol{r}_2, \ldots\ldots \boldsymbol{r}_N$

Positions, Velocities, Forces: $\boldsymbol{r}_i, \boldsymbol{v}_i, \boldsymbol{F}_i$

Potential Energy Function = $V_i(\boldsymbol{r}^N)$



Sandia's LAMMPS code is spatially parallel

Large-scale **A**tomic/**M**olecular **M**assively **P**arallel **S**imulator

Problem: Δt tied to the fastest atomic motion t is the timescale of physical interest

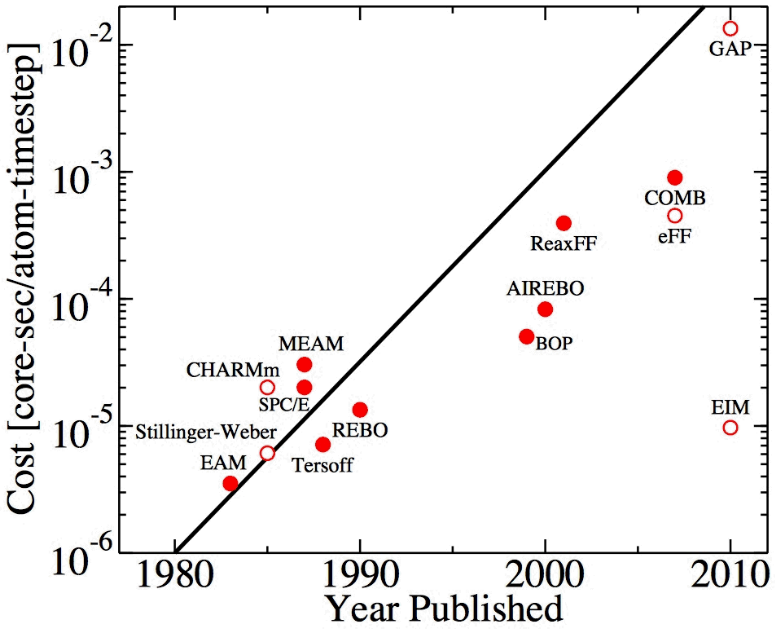
$nsteps = t / \Delta t = 10^6 \ldots 10^{15}$

Matthew Lane - jlane@sandia.gov

Strengths, weaknesses and trends

Strengths:

- Atomistic detail
  - Nanoscale Dynamics,
  - Chemistry,
  - Structure / Property
- Scales on parallel processors



Plot courtesy Aidan Thompson, Sandia

Weaknesses:

- Finite set of interatomic potentials
- Atom counts << 10^{23}
- Short (generally ns) timescales

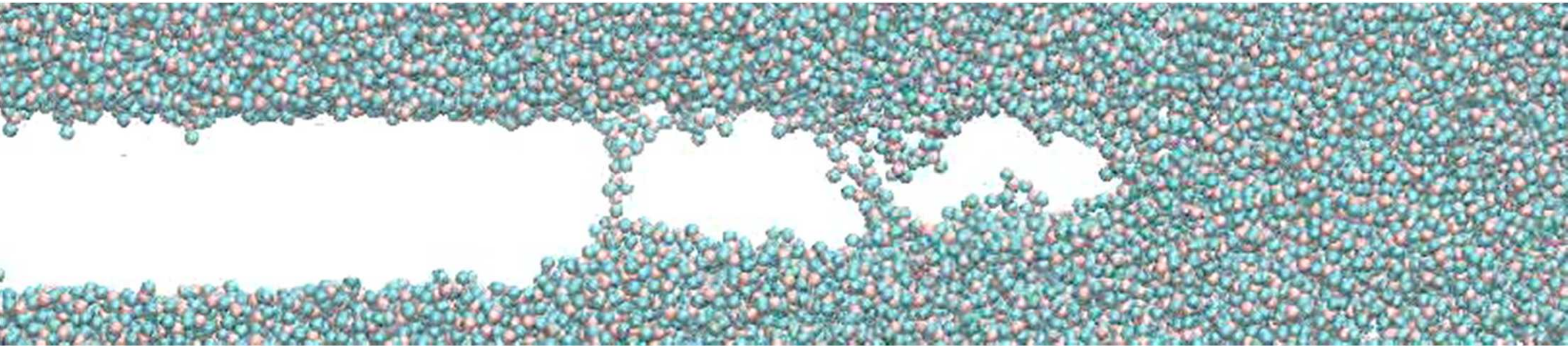
Trends in molecular dynamics:

- Complex interatomic potentials
  - Reactive & polarizable
  - Multi-elemental
- Computing for scale
  - New algorithms for new machines
  - Increased spatial parallelism
- Computing for fidelity
  - DFT-based training sets

Quantitative studies benefit from larger systems with more complex chemistry

Matthew Lane - jlane@sandia.gov

Motivation: “long-time” dynamics

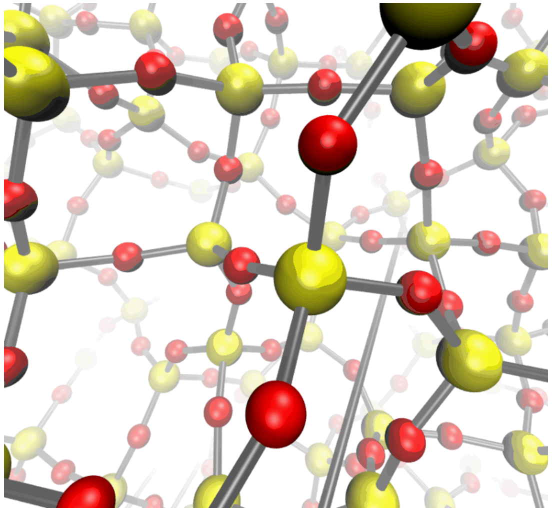


We’re interested in “long-time” dynamics in molecular dynamics for the study of fracture in glass. Particularly, we have noted that fracture response in glasses is loading rate dependent.

We’d like to take advantage of time-temperature superposition to accelerate the loading relaxation and its effect on glass structure, before crack initiation. This would allow us to investigate much slower effective loading rates.

Matthew Lane -
jlane@sandia.gov

Silica glass structure and methodology



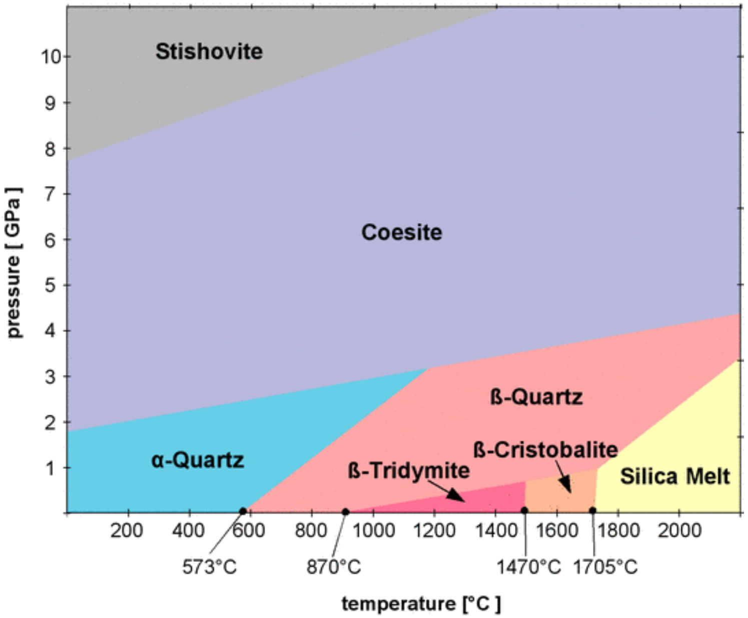
Silica, is a strong random network glass former with

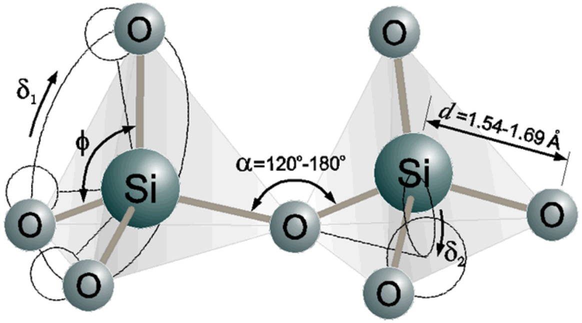
- Tetrahedal cores about the **silicon**
- Bridging **oxygen** linking tetraheda (Si-O-Si-O)

Well ordered on short length scales, disordered on longer length scales

SiO_2 has several metastable phases at room temperature, leading to easy structure frustration during rapid cooling from melt

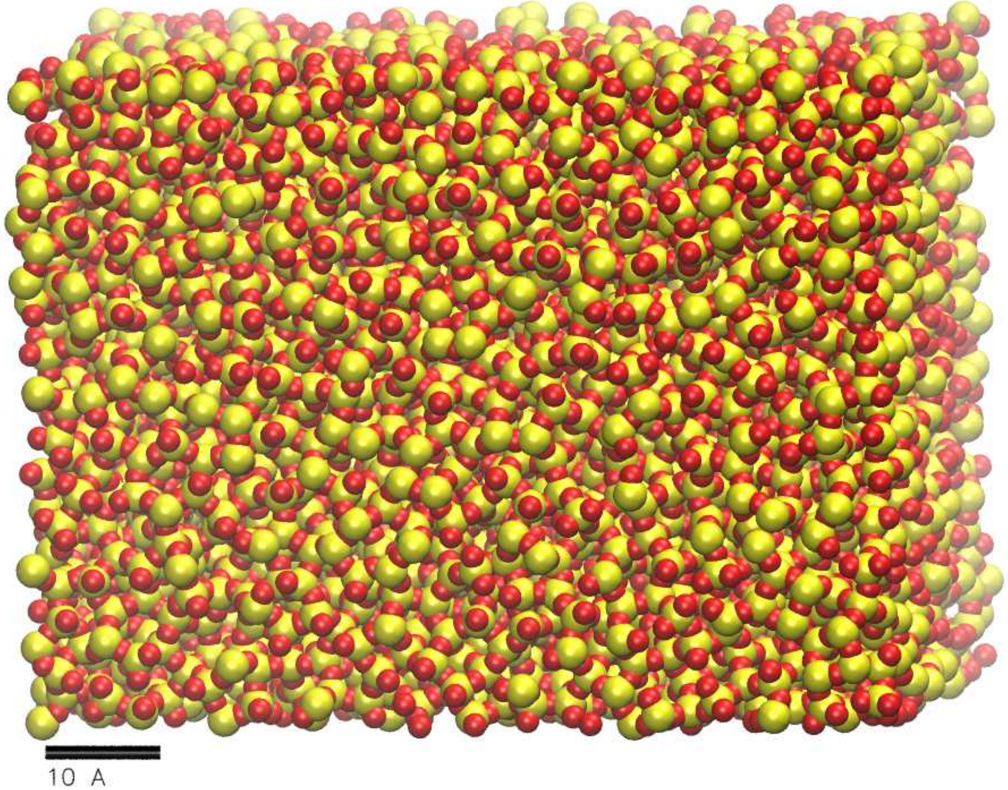
Glasses are formed by a melt/quench process with experimental quench rates of a few Kelvin/minute





Matthew Lane - jlane@sandia.gov

Simulation methods and models



Van Beest, Kramer, and Van Santen. *Phys. Rev. Lett.* **64**, 1955 (1990).
Vollmayr, Kob and Binder, *PRB*, **54**, 15808 (1996)

Sandia's LAMMPS molecular dynamics code http://lammps.sandia.gov

BKS potential, modified to prevent core collapse at high temperatures
With long-range coulomb interaction

Thermostat and barostat with Nose-Hoover, NPT and NVT in LAMMPS

45 x 25 x120 Angstrom cell with periodic boundaries with 13824 atoms

Formation from β-cristobalite crystal melted to 8000 K, then cooled at a linear rate to 300 K

Matthew Lane -
jlane@sandia.gov

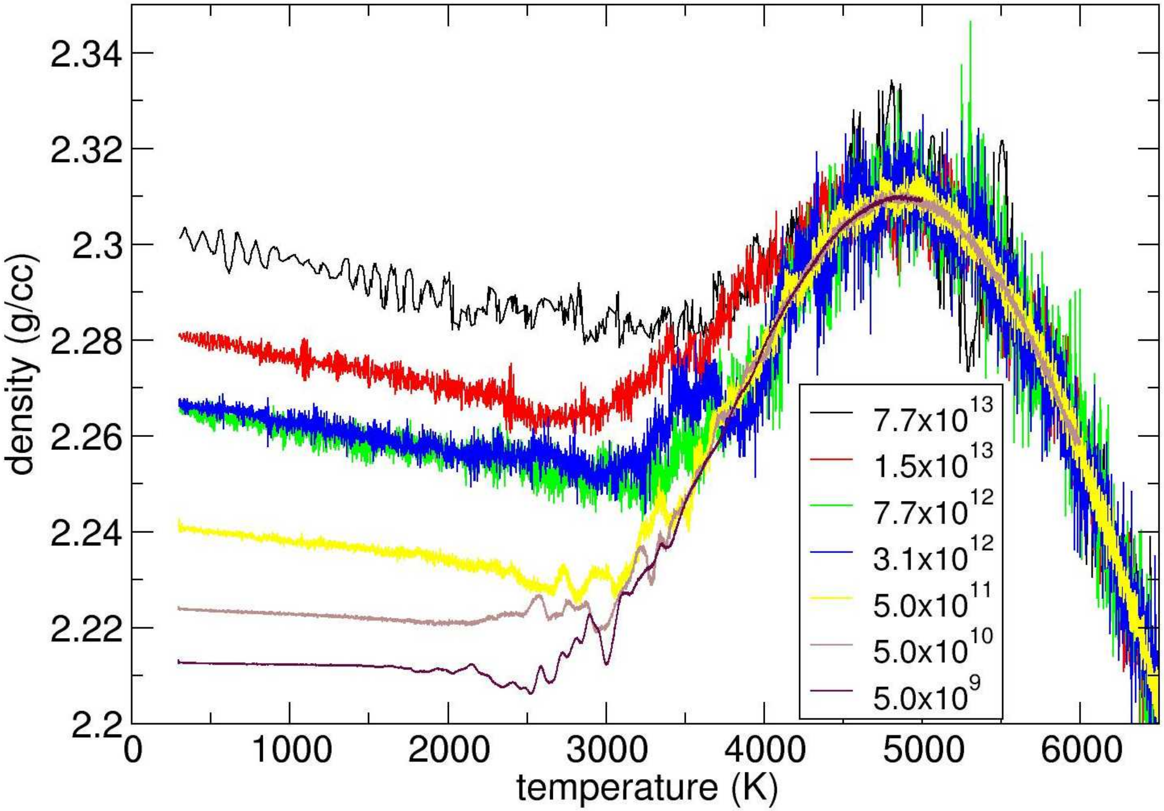
Glass formation and cooling-rate dependence

Both experiment and model silica has an anomalous density maximum in temperature near T_g

Strong (~5%) density dependence on cooling rate

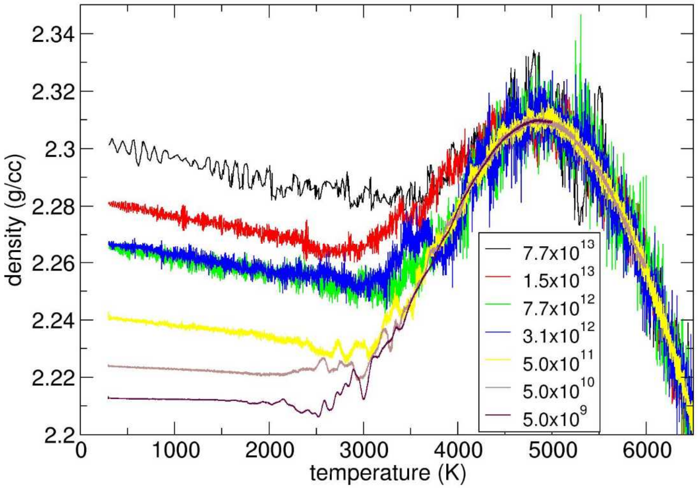
MD cooling rates are many orders of magnitude larger than experiments

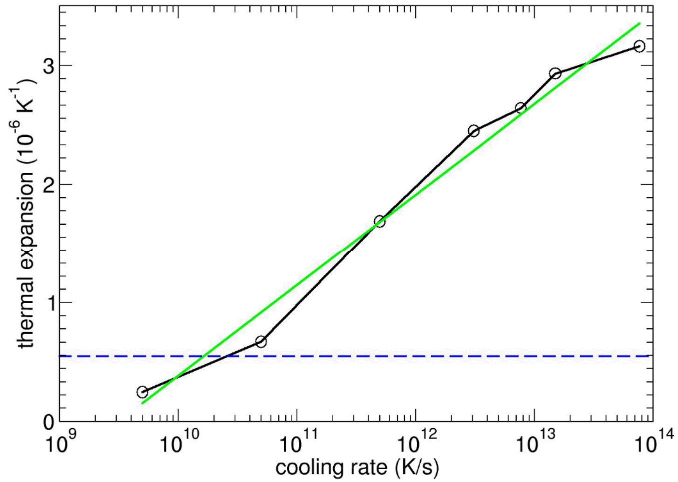
Thermal expansion properties of final glass appear to depend on quench rate.

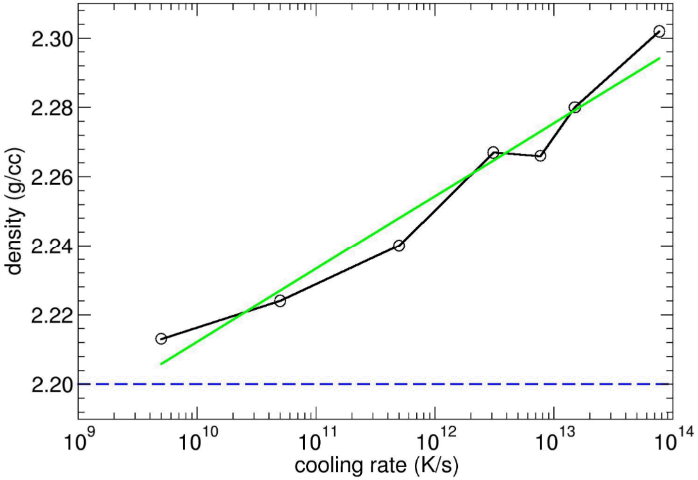


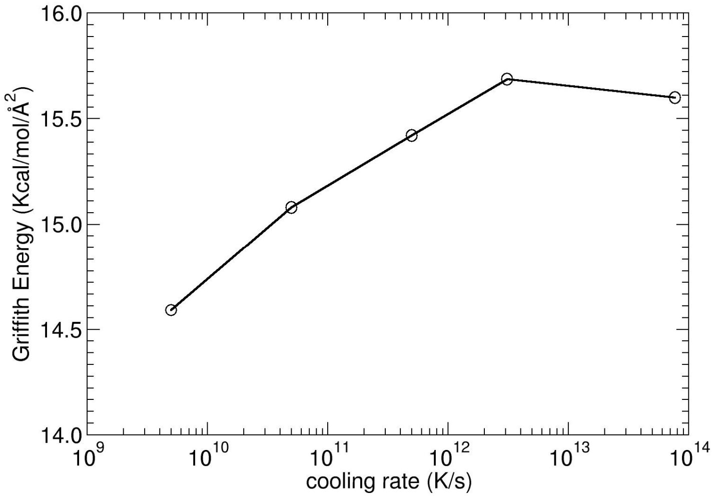
Excellent previous work studied glass behavior down to 4.44 x 10^{12} K/s. They extrapolated out an experimental glass transition temperature T_g = 2525 K compared to experimental values of 1446 K. Vollmayr, Kob and Binder, *PRB*, **54**, 15808 (1996)

Glass formation and cooling-rate dependence









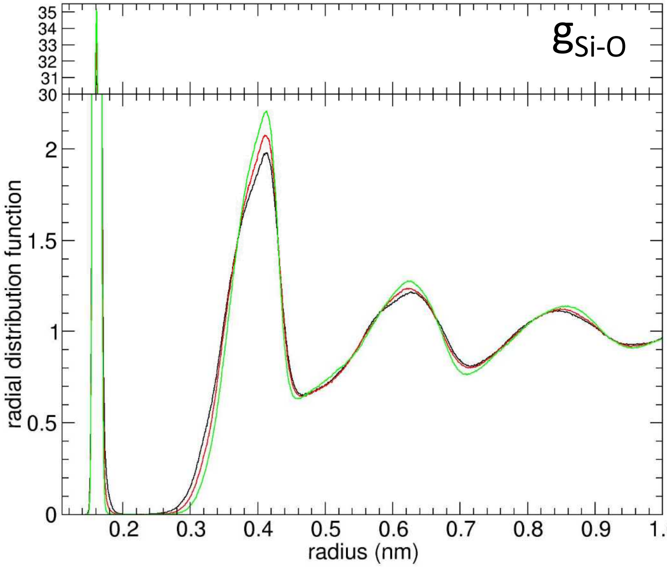
Matthew Lane - jlane@sandia.gov

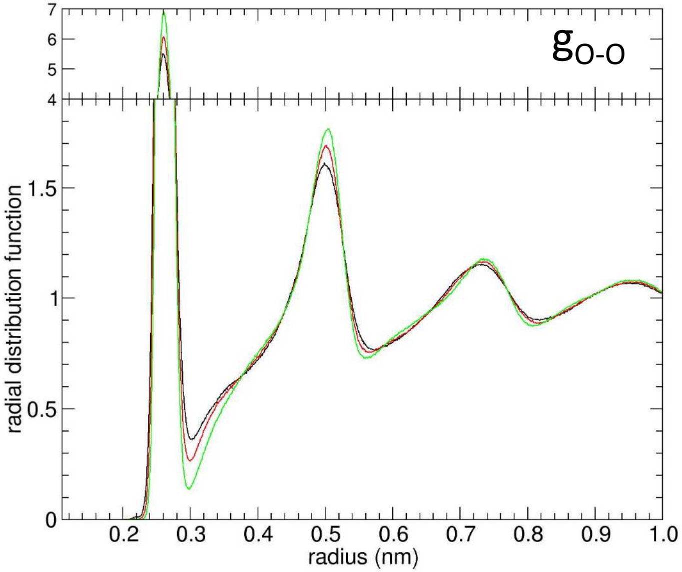
Cooling-rate dependence - structure

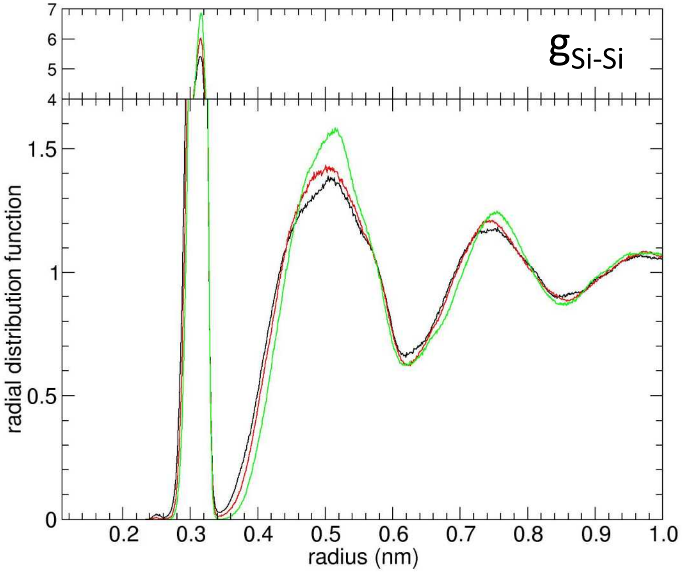
Radial distribution function analysis of the glass structure from **fast 10^{13}**, **moderate 10^{12}** and **slow 10^{9}**

Significant additional structural relaxation occurs with reduced cooling rate. More ordered structures result when more time is allowed to cool.

The 2nd neighbor structure is most significantly impacted with lesser effects of 1st and 3rd neighbors, indicating ordering of tetrahedra rather than bonds.



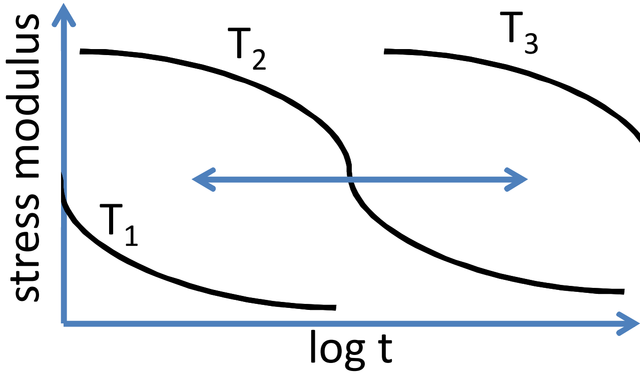




Matthew Lane - jlane@sandia.gov

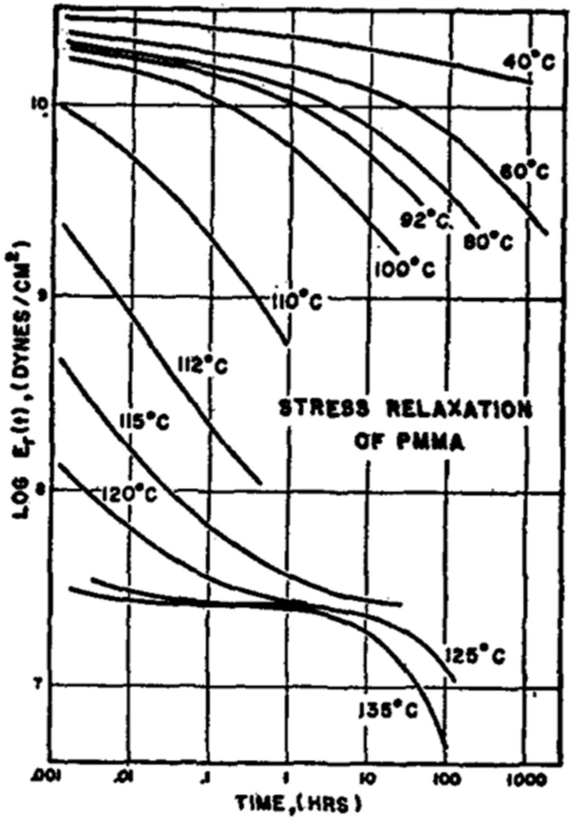
Time-Temperature Superposition

Time-temperature superposition is a property of viscoelastic glass forming liquids which simply states that the relaxation dynamics follow a master curve which can be shifted in log(time) to account for response at different temperature.



Moreover, the log(time) shift factor for all temperatures can be determined from the measurement of only a few.

Once time-temperature superposition is demonstrated then the master curve can be used to infer response times which are inaccessible (too long or too short)

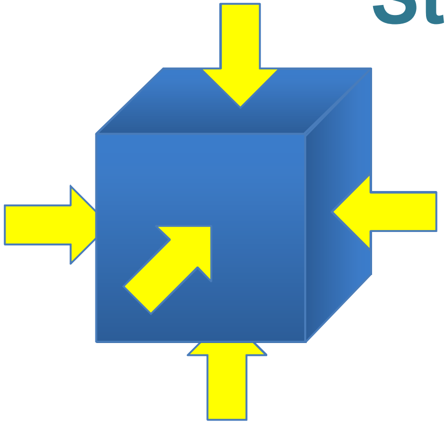


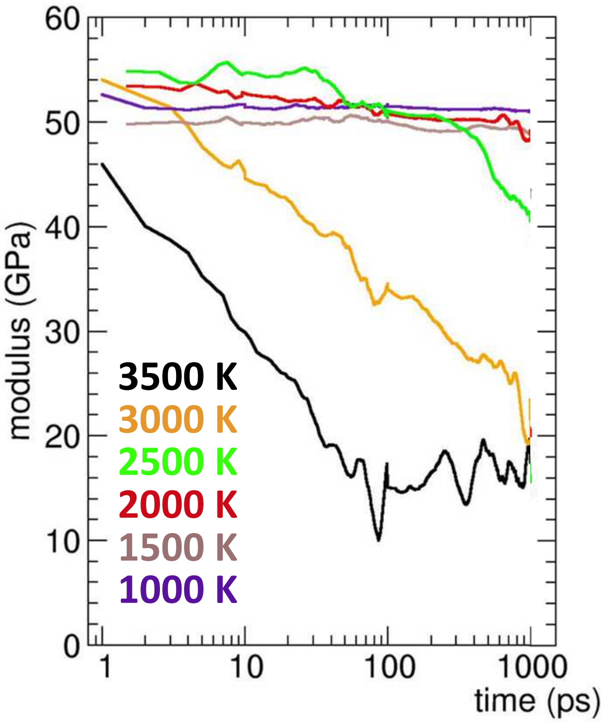
This has been used successfully in polymer experiment since the 1940s.

Plot from A.V.Tobolsky, *J. Appl. Phys.*, **27**, 627, (1956).

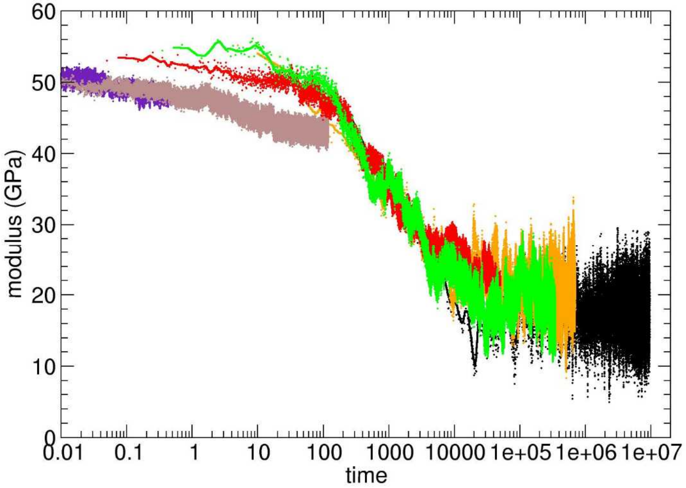
Matthew Lane - jlane@sandia.gov

Stress relaxation results





- Imposed and held a 3% volumetric strain imposed on equilibrated glass
- Measured modulus relaxation times at various temperatures



- Time-temperature superposition is shown to hold for relaxation curves down to 2000 K
- Two lowest temperatures do not appear to be consistent with the master curve – not unexpected below Tg

Matthew Lane - jlane@sandia.gov

Stress relaxation results

The shift distance along the logarithmic time axis is the time-temperature shift factor a_T,

$$a_T = \frac{t_T}{t_0}$$

Two fits are generally made to the shift factor, the Williams-Landel_Ferry (WLF) fit near the glass transition,

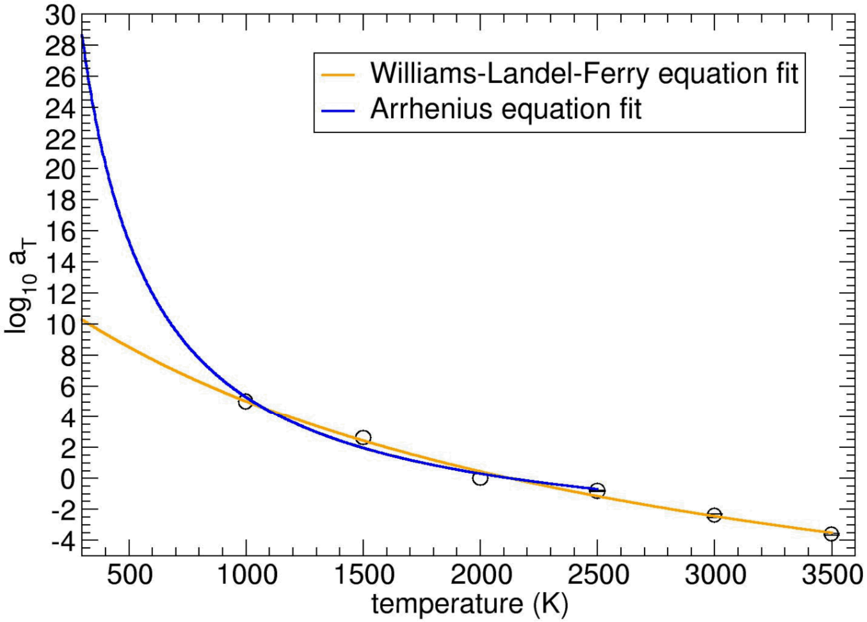
$$\log_{10} a_T = \frac{-C_1 \times (T - T_0)}{C_2 + T - T_0}$$

$$C_1 = 15.11, \quad C_2 = 4511, \quad T_0 = 2128$$

and an Arrhenius fit well below T_g.

$$\log_{10} a_T = A\left(\frac{1}{T} - \frac{1}{T_0}\right)$$

$$A = 9933, \quad \mathrm{T}_0 = 2128$$



Full relaxation at room temperature will take simulations between 10^{10} and 10^{28} times longer to capture.

Matthew Lane -
jlane@sandia.gov

Summary and conclusions

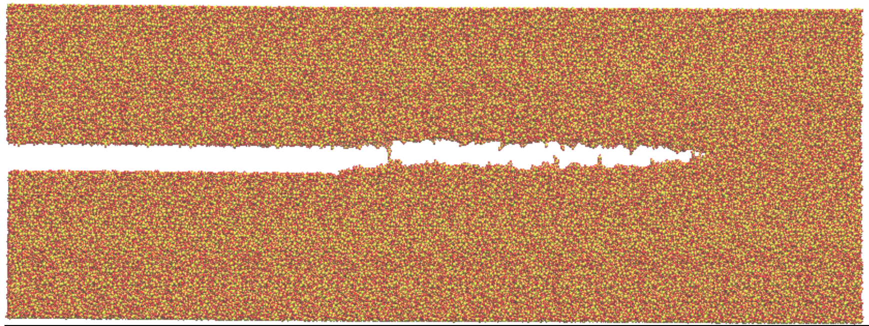
- We've studied dynamics of a simple, but realistic, glass model out to microsecond times – beginning to overlap experiment. An increase in times of 2 to 3 orders of magnitude.
- In slow quench cooling rate simulations we see significant additional relaxation from previously reported as would be expected. The thermal expansion of the set glass is significantly lower than previously reported for faster cooled glasses.
- BKS model glass demonstrates well characterized viscoelastic behavior, including time-temperature superposition which may allow well characterized accelerated stress relaxation for fracture simulations initial loadings.

FY14 ASC P&EM

Brittle fracture models from molecular to component scale

J. Matthew D. Lane, Org. 1814

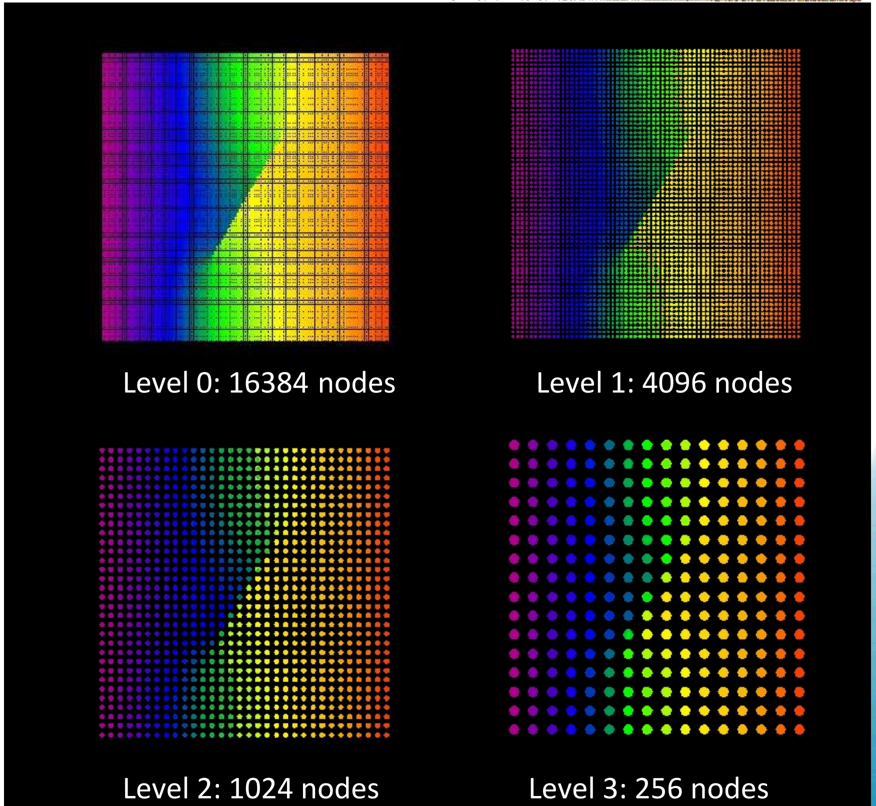
Stewart Silling and David Littlewood, Org. 1444



Fracture is a multiscale problem in which response at different length scales interact to determine crack propagation

Link molecular dynamics simulation to peridynamics, which has a property called upscaling to coarsen the problem to allow scaling to component geometry

MD is used to calibrate the material model and fracture properties of the peridynamic model



Why Multiscale with Peridynamics?

THE KEY POINT IN THE UPSCALING IS NONLOCALITY

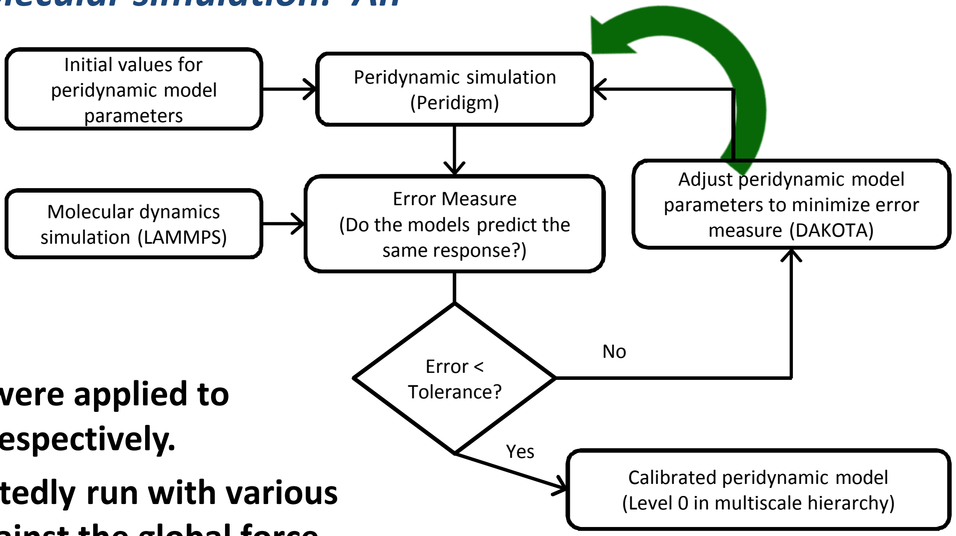
- **Nonlocality greatly expands the set of deformation-force relationships that can be captured in the model**
  - Nonlocal kinematic information provides a rich, vivid, highly-resolved description of deformation this is not possible in a local model
  - Nonlocal forces allow for direct incorporation of heterogeneous lower-length-scale mechanisms in the higher-scale system by explicitly modeling material point interactions over a range of length scales
- **This provides a means to incorporate lower-scale mechanisms into the higher-scale model *without smearing***
- **The response of individual bonds at the component scale is determined (hierarchically) by analysis of submodels at lower length scales**
  - Small-scale features are resolved only in the lower-scale models
  - Approach circumvents the need for full system analysis at the lower length scale
- **There is no analogy to this approach with classical local models**

Matthew Lane - jlane@sandia.gov

Peridynamic material models from MD

Extracting peridynamic material models from molecular simulation: An optimization-based approach

- A generalized parameter extraction routine was developed in DAKOTA to determine parameters for the material model of amorphous silica based on molecular dynamics (MD) simulation.


Initial values for peridynamic model parameters

Peridynamic simulation (Peridigm)

Molecular dynamics simulation (LAMMPS)

Error Measure (Do the models predict the same response?)

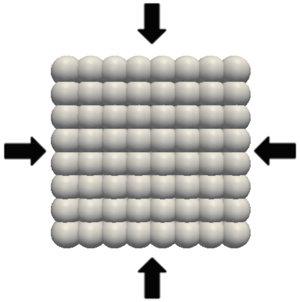
Adjust peridynamic model parameters to minimize error measure (DAKOTA)

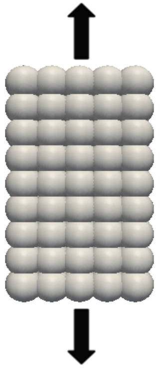
Error < Tolerance?

No

Yes

Calibrated peridynamic model (Level 0 in multiscale hierarchy)



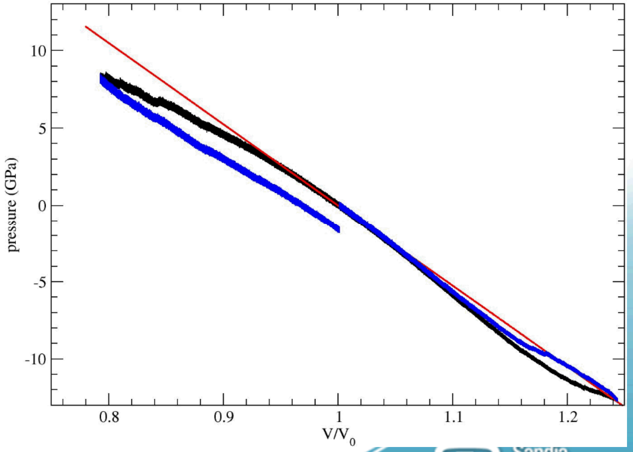


Strain response optimization

- **Both volumetric and uniaxial strains were applied to measure bulk and Young's modulus, respectively.**
- **Peridynamics simulations were repeatedly run with various material properties and optimized against the global force-displacement response**

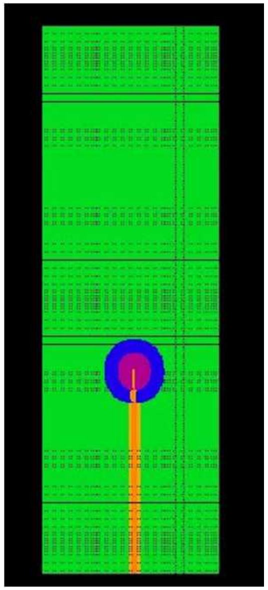
Stress-strain results

- The plot to the right shows the volumetric stress-strain response to approximately 20% strain in compression and extension. Black is the MD material response. Blue is the MD response during cycling back to zero strain. Red is the peridynamic model response.
- The glass is elastic in extension in this strain range, but nonlinear at strains over 10%. In compression, the glass deforms plastically, becoming more dense after release.



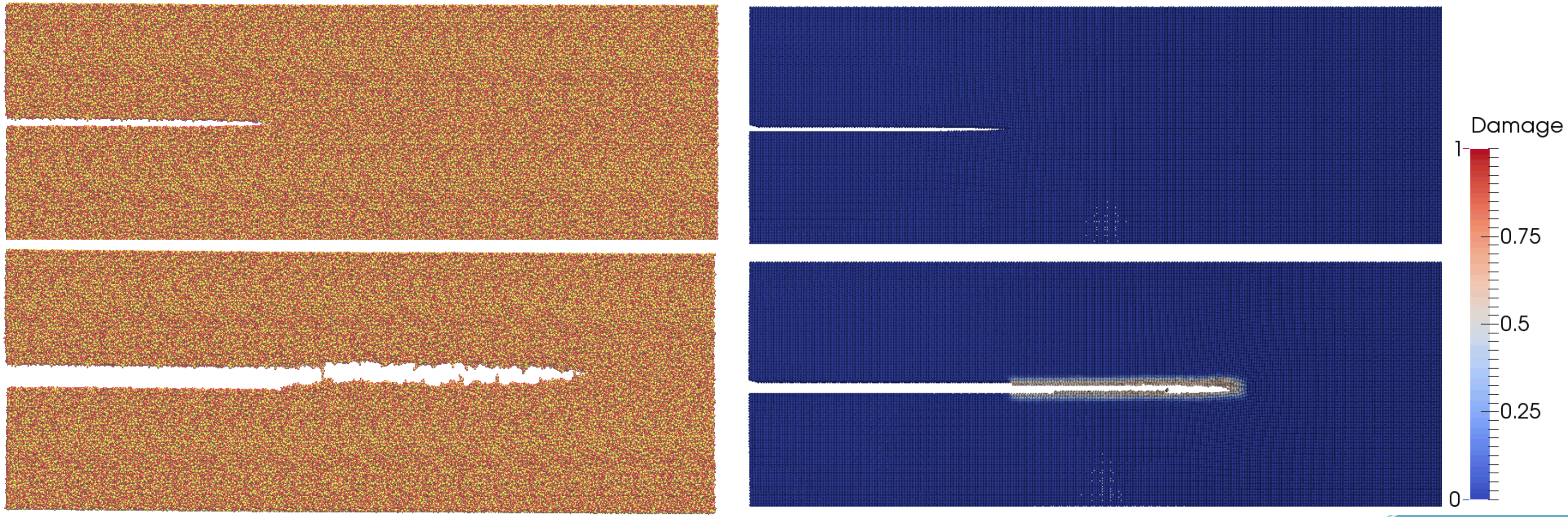
Matthew Lane - jlane@sandia.gov

Peridynamic fracture models from MD



- Peridynamic damage is modeled through bond breaking criteria and the suitable selection of a parameter called the horizon, which characterizes the degree of nonlocality in the model. Initial qualitative work to capture fracture properties has been promising.
- A reformulation of the peridynamic upscaling, which coarse grains the system by constrained optimization, is being evaluated in plane-strain 2D fracture geometries.

Preliminary results demonstrating qualitative agreement in glass fracture (on the left, atomistic simulation, and on the right, peridynamics modeling.)



Matthew Lane - jlane@sandia.gov