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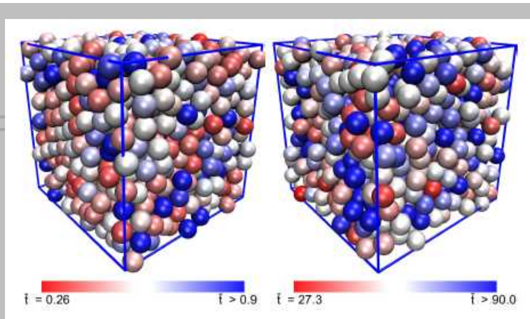

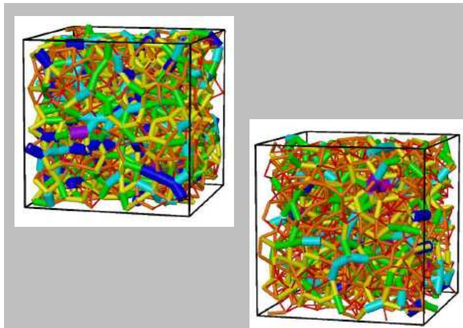

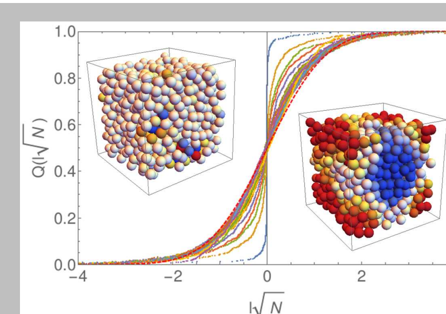

Thermal Runaway in Jammed Networks

Jeremy B. Lechman, Cole Yarrington, Dan S. Bolintineanu

Fluid and Reactive Processes

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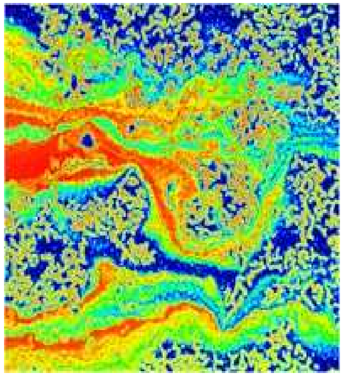
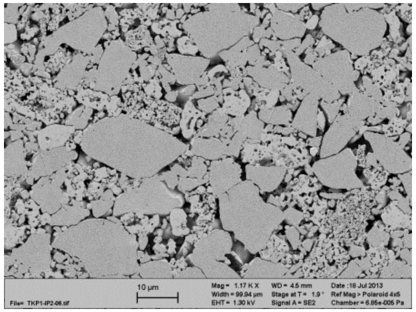
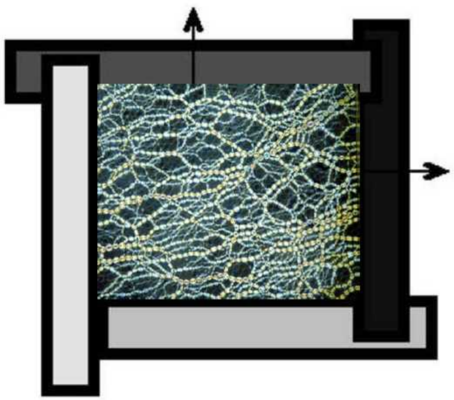
Property Prediction and Role of Microstructure in Transport



- Need better prediction and control of, e.g.
  - Thermal transport in pyrotechnic materials (particle scale)
  - Reliability of composite materials
- Heterogeneous materials
  - Inhomogeneous & "discontinuous"
    - material properties and microstructure
      - multi-phase, multi-material → interfaces



- Heterogeneous "dynamics"
  - Spatial distribution of (relaxation) timescales
    - "Anomalous" stochastic behavior
      - E.g., non-Fickian diffusion
  - Generalized Stochastic Models
    - CTRW or GME
  - How do these fluctuations couple to nonlinearities



Discretizing the Mesoscale

- Reduced-order, network modeling approach based on random walk simulations/analysis for thermal conduction in particle

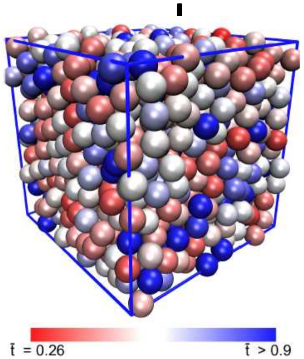
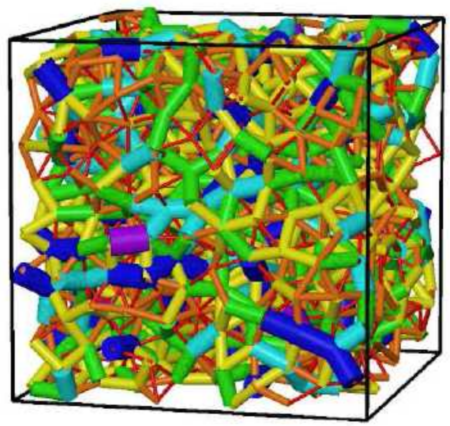
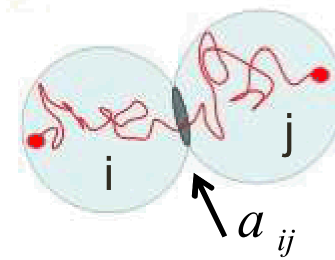


Image stack, or simulated μstructure

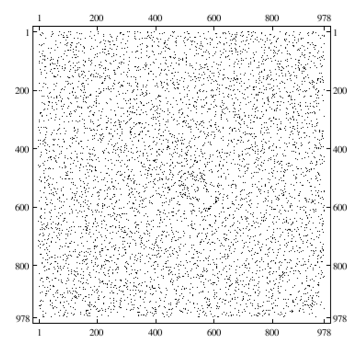
Determine segmentation: clustering & connectivity



graph of contact network



Determine: edge weights (interfacial resolution and physics models)



$$L_{ij} = \frac{1}{\tau_{ij}} \approx \frac{4 D_i a_{ij}}{V_i}$$

Graph Laplacian, Transition Rate Matrix, …

$$\frac{\partial T_i(t)}{\partial t} = \sum_{j \neq i} L_{ij} T_j(t) + \frac{Q c_0 k_0}{\rho c_v} \exp\left(-\frac{U}{R T_i}\right)$$

Simulate Markov Process on Contact Network

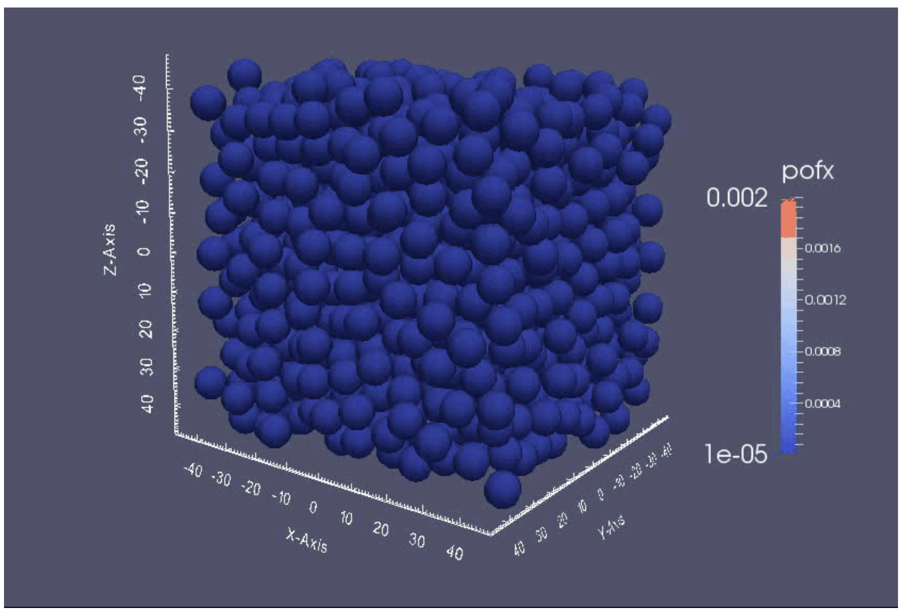
- Discretize Continuous-Time Equation

$$\frac{\partial \mathbf{T}(t)}{\partial t} = \mathbf{L}\mathbf{T}(t)$$

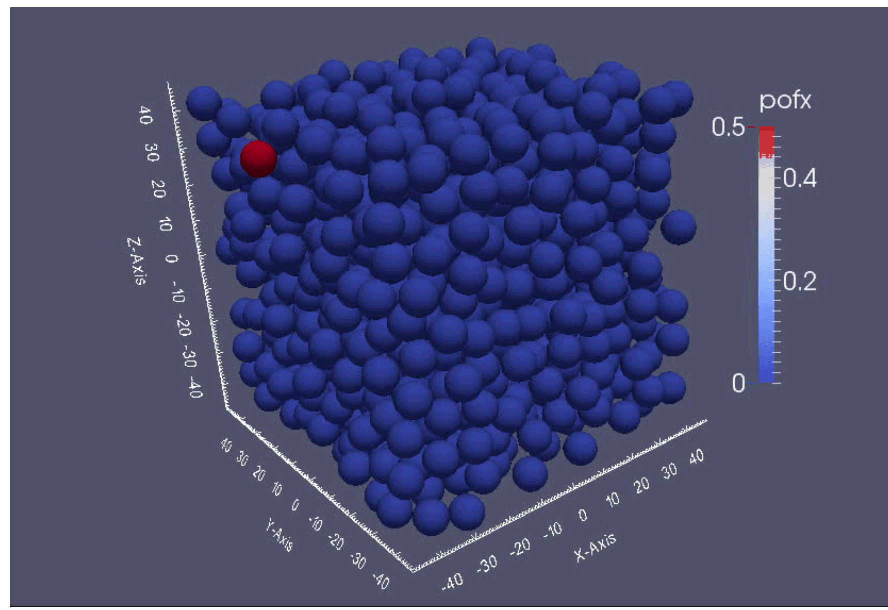
$$L_{ij} = \begin{cases} \dfrac{3D}{\pi R^2}\sqrt{\delta_{ij}/R} & i \neq j \\ -\sum_{j \neq i} L_{ij} & i = j \end{cases}$$

$$\Rightarrow \quad \mathbf{T}_{n+1} = \mathbf{M}\mathbf{T}_n \qquad \mathbf{M} = \mathbf{I} + \Delta t \mathbf{L}$$

  - I.C. $\mathbf{T}_0 = \hat{\mathbf{e}}_1 \quad \|\hat{\mathbf{e}}_1\| = 1$
  - Periodic B.C.'s



p = 0.0004

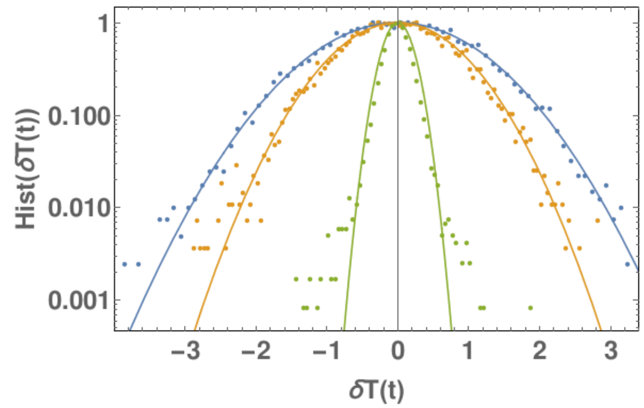


p = 0.00004

Evolution of Temperature Fluctuations

- Calculate distribution of temperature fluctuations based on Eigen decomposition

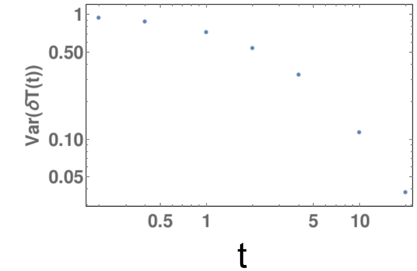
$$\delta \mathrm{T}(t) = \mathrm{T}(t) - \mathrm{T}_{eq} = \sum_{j=2}^{N} e^{-\lambda_j t}\, \mathrm{v_j} \qquad \delta T_i(t) = \sum_{j=2}^{N} e^{-\lambda_j t} \left(\mathrm{v_j}\right)_i$$



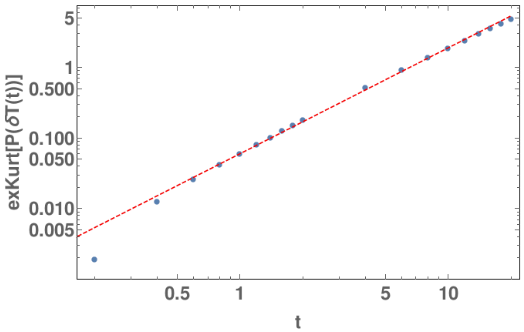
  - Fluctuations decay in time as system homogenizes
    - For sum of IID Gaussian random variables, a large deviation (LD) approximation can be obtained

$$\mathrm{P}(\delta \mathrm{T}_N = \delta \mathrm{T}\,) \sim \exp\left[-\frac{1}{2} N \left(\frac{\delta T}{\sigma(t)}\right)^2\right]$$

$$\mathrm{var}[\delta \mathrm{T}(t)] = \sigma^2(t) = \sum_{i=2}^{N} e^{-2\lambda_i t}$$



    - Initially, Gaussian seems to work
    - However, scaling of excess Kurtosis does not follow Gaussian behavior
      - FATTER TAILS! Fluctuations decay but slower than Gaussian

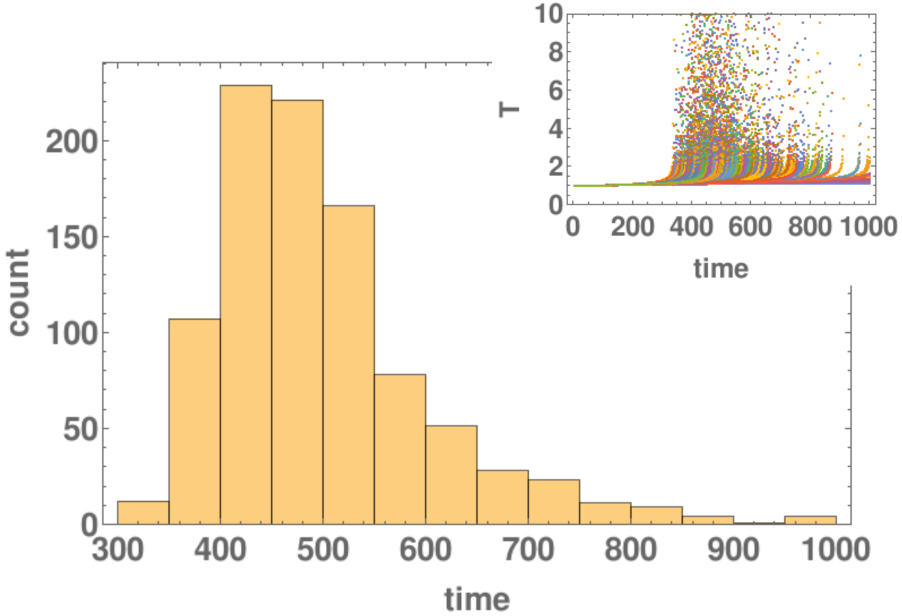


Thermal Runaway

- Add nonlinearity
  - 1st order, irreversible reactions

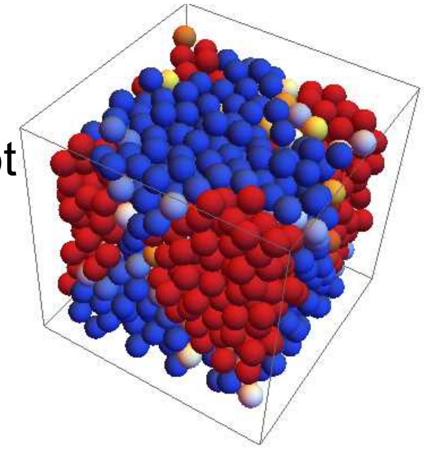
$$\frac{\partial T_i(t)}{\partial t} = \sum_{j \neq i} L_{ij} T_j(t) + \frac{Q}{\rho C_V} k c_0 \exp\left[-U/RT_i\right]$$

  - Periodic BC's
  - IC: unit impulse to particle i



- Time to thermal runaway depends on particle, i
  - Varying "sensitivity" for different particles
  - Stochastic problem due to disorder in pack
- Interaction of fluctuations (due to disordered mesostructure) and nonlinearity

Hot spot



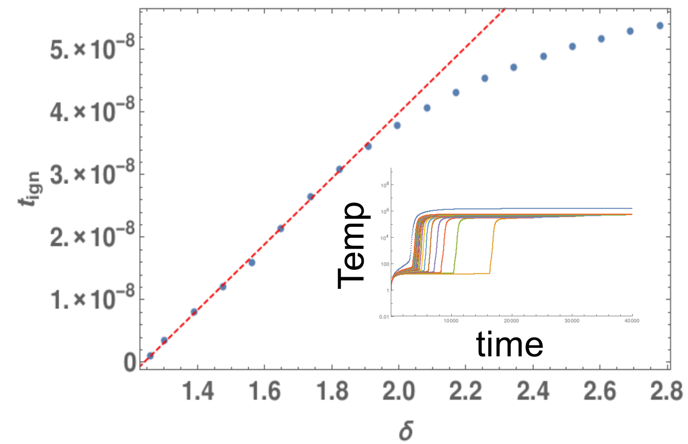
Extending spectral analysis to discrete approximation of Koopman Operator

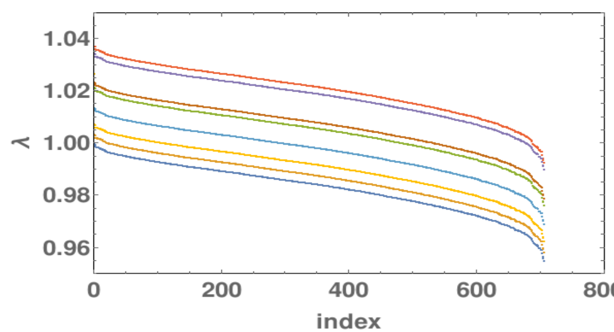
- Use homogeneous IC and Dirichlet BC's
  - Scaling of time to ignition follows classical homogeneous result; exhibiting critical slowing down

$$t_{ign} \sim (1 - \delta/\delta_c)^{-1/2}$$

where δ is the Frank-Kamenetskii parameter

- Expected that system will be sensitive to "large deviations" (i.e., fluctuations stronger than Guassian)
- DMD eigenvalues can be computed as a function of the strength of the nonlinearity
  - Interpretation and verification of analysis is ongoing





Summary & Conclusions

- Reduced-order, network-type model of thermal transport on particulate materials is possible
  - Spectral analysis of conduction matrix allows for development of macro-scale models and analysis of thermal fluctuations due to disordered microstructure
- Addition of nonlinearity due to chemical reactions can be accomplished
  - Comparison to classical Frank-Kamenetskii problem shows similar critical slowing down near critical point
  - However, details of thermal runaway time show statistical characteristics due to disorder of microstructure
  - DMD-type analysis allows for possibility of extending spectral analysis from linear to nonlinear equations through approximation of (linear) Koopman Operator

Acknowledgments

- Leo Silbert and Mike Salerno
- Gary S. Grest & Randy Schunk
- Industrial collaborators
- LDRD and ASC P&EM programs at Sandia

Backup Slides

Motivation

- Methods are needed to both inform and guide the development of models that capture the effects of microstructure at the continuum scale

- Next generation microstructure aware continuum models based on stochastic microstructures will require analysis methods capable of bridging scales from the mesoscale to the continuum

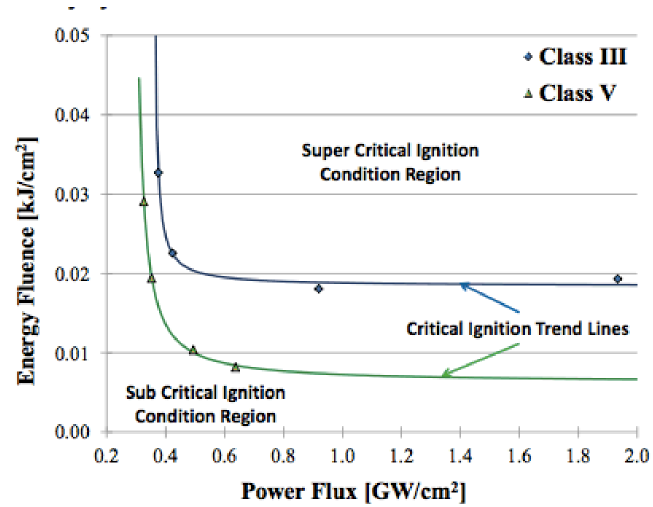


Figure 5. HMX Class III and V ignition data with fitted trend lines (*Critical Ignition Conditions*). For illustration, we have also noted the *Super* and *Sub Critical Ignition regions*.

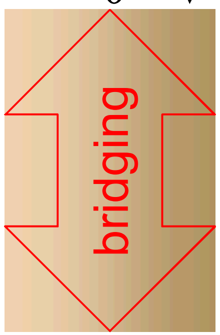
Heterogeneous Materials Awareness at Sandia

- Broad interest from Sandia’s Engineering and Materials S&T community
  - Relevance to several significant application areas
- High-level focus on issues related to behavior of heterogeneous materials
  - E.g., Meso-scale fluctuations due to random microstructure
    - What is the nature of the noise/variability in these materials
    - How to “roll-up” uncertainties from micro through meso to macro scale
    - Top-down prediction and control
  - Also, increasing recognition of role manufacturing processes play in determining the distribution of material properties
- Motivation for strategic partnerships

The Multi-scale Transport Picture *through* Particulate Media

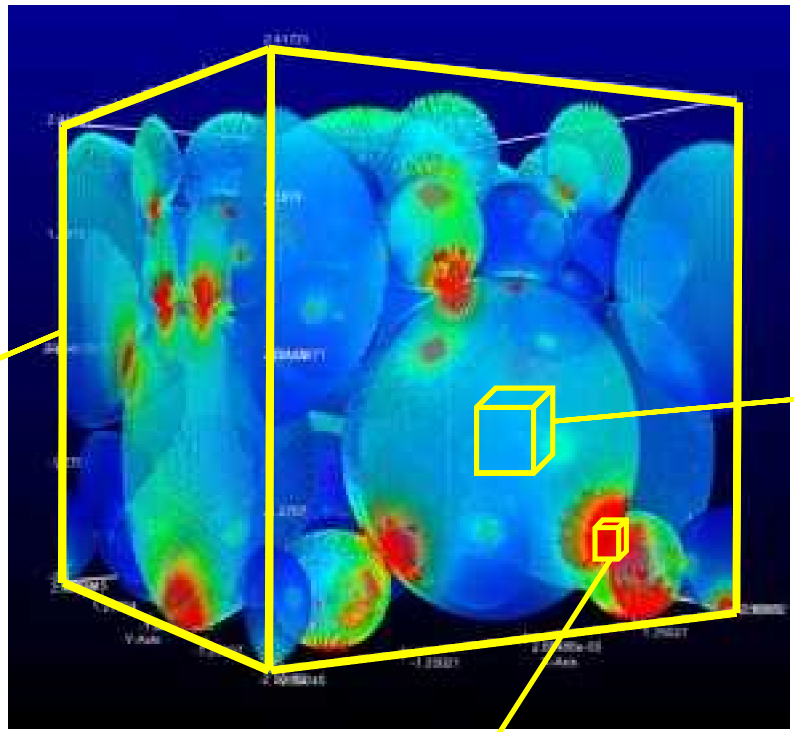
(1) Bulk, Macroscale
- Homogeneous
- "Continuum"
- Constant transport coef.

$$0 = \nabla \cdot \mathbf{q}(\mathbf{x}) = K_{eff} \nabla \cdot \langle \nabla T(\mathbf{x}) \rangle$$



(2) Particle-Particle (Meso-structure) Scale
- Inhomogeneous
- "Discrete"; Disordered
- "Anomalous" transport

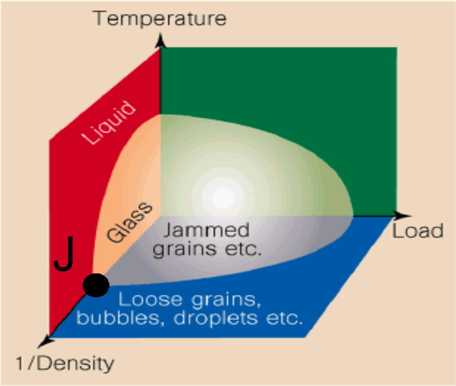
$$0 = \nabla \cdot \mathbf{q}(\mathbf{x}) = \nabla \cdot \left(K(\mathbf{x}) \nabla T(\mathbf{x}) \right)$$



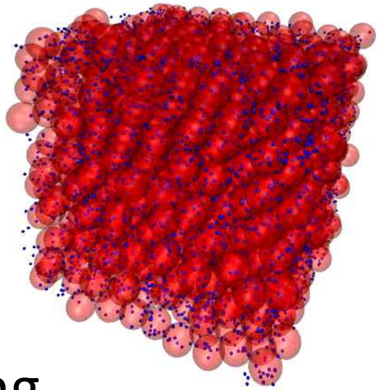
(3) Sub-particle materials structure
- Crystal structure
  - Anisotropy
  - defects, impurities, etc.
- Polycrystalline
  - Grain boundaries

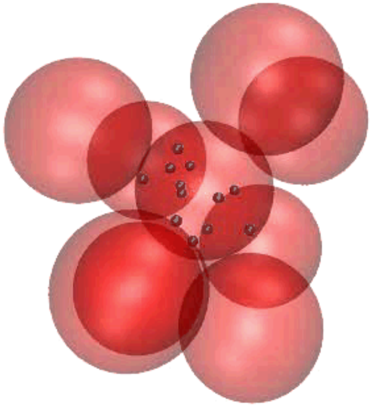
(4) Interfacial Scale
- Contact area, roughness, inter-diffusion
- Material types (e.g., phonon, electron dominated)

Background: Random Walks in Particle Packs



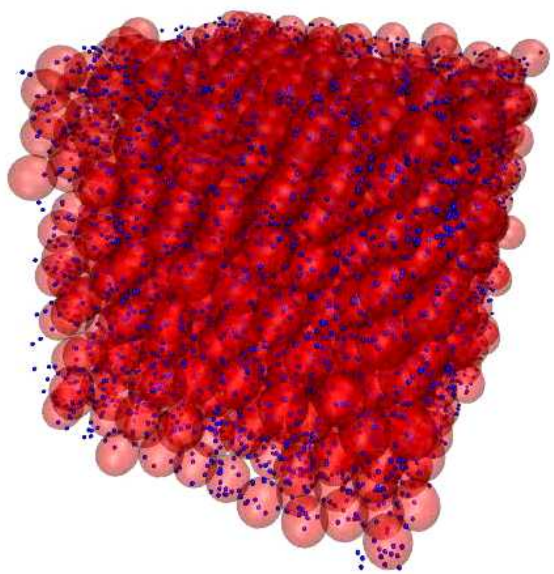
- Jammed particles near "Point J"
  - Critical-like "point" of marginal mechanical stability
    - Control of apparent microstructural length scale
  - Well defined process for creating packs
    - Remove "rattlers"
- Random Walker Simulations
  - Random walkers initially uniformly distributed within particles
  - Particles conducting; voids insulating
    - Reflecting (specular) BC at interface
      - Neumann-like, no-flux
  - Global periodic simulation domain

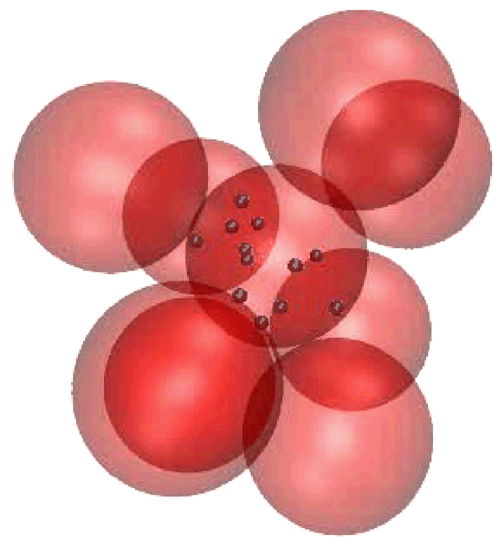




Example Simulation

- Random walk through interior of particles, where diffusion coefficient $D_0 = 1$
- Similar to method of Kim and Torquato[1](“walk on spheres”), but modified to yield time-dependent behavior
- Random walker displacement relates to material properties
- “Narrow escape” hopping between neighboring particles requires long simulation times, but accounts for small contacts explicitly and accurately

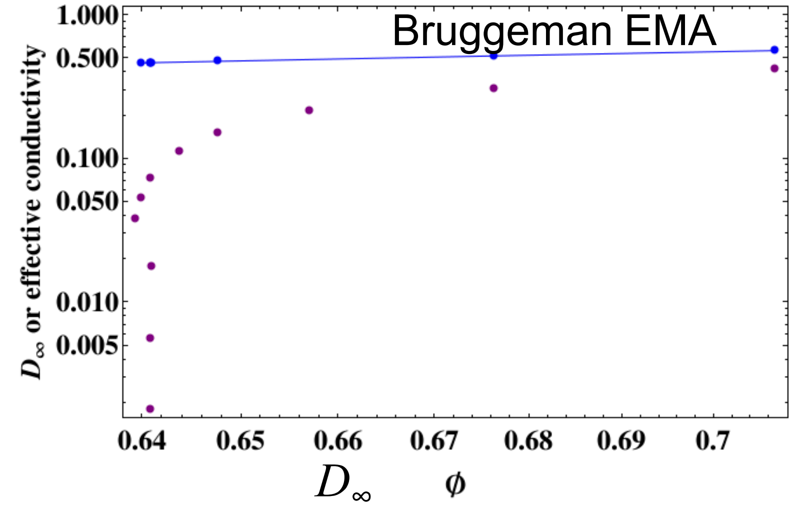


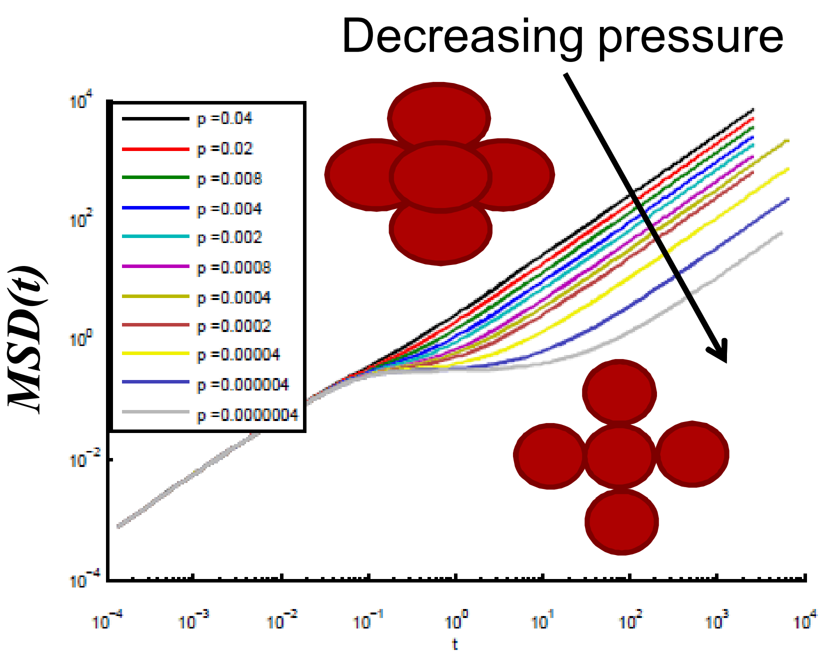


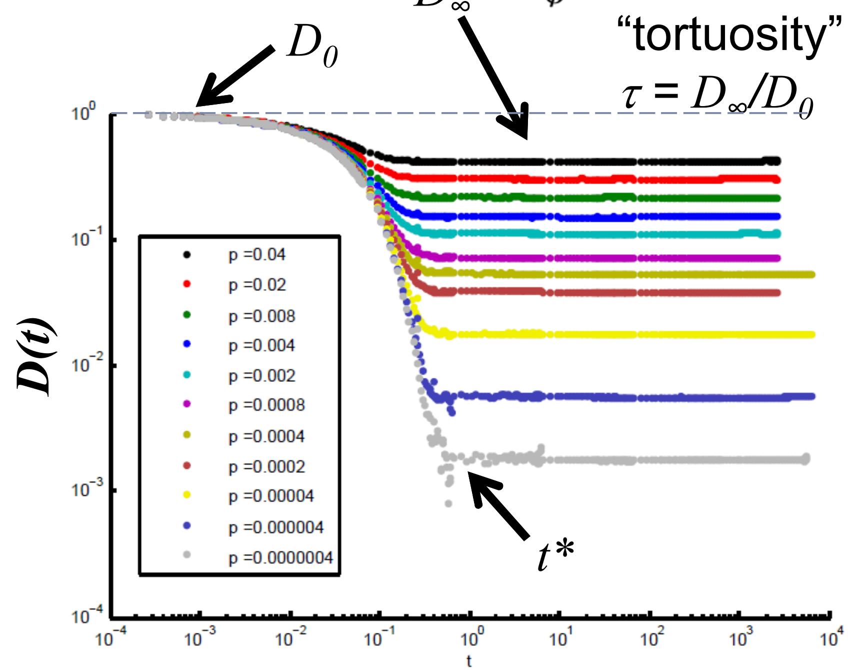
1. Kim IC and Torquato S., J. Appl. Phys. 68 (1990): 3892; Kim IC and Torquato S., J. Appl. Phys. 69 (1991): 2280

Conductivity of Particulate Microstructures

- Results
  - Late-time D_∞ function of pressure
  - Controlled by particle contact radius
  - Apparently, single relevant timescale

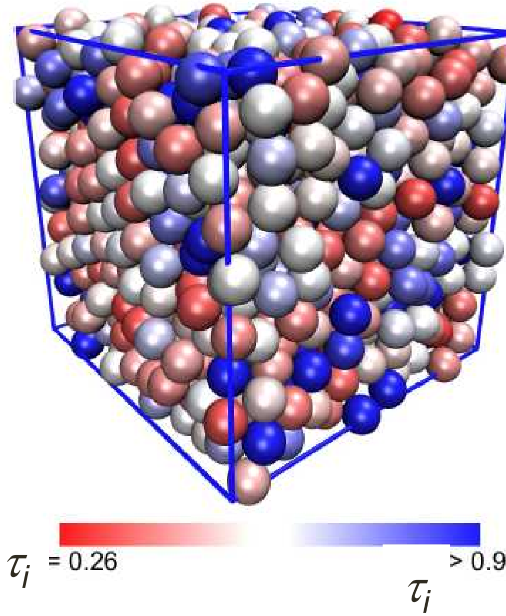




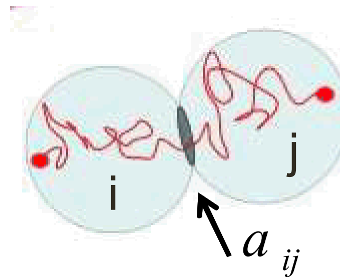


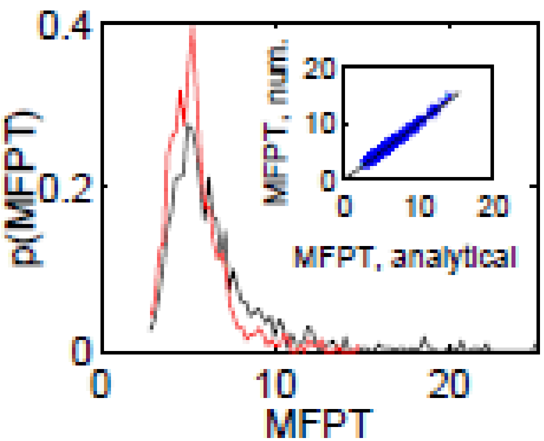
Bulk Thermal Conductivity

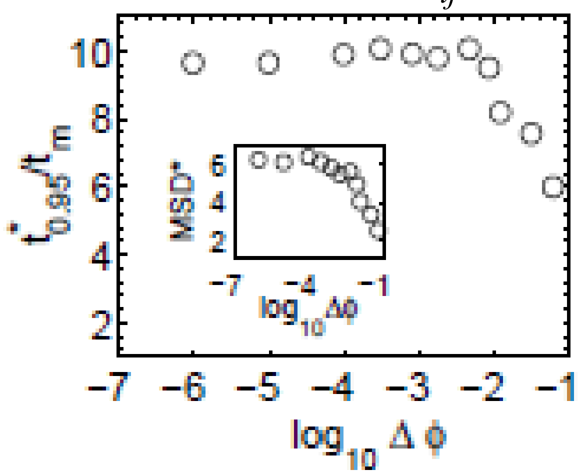
- Volume averaged MFPT per particle
  - Narrow Escape
    - **Small, well separated contacts** ($a_{ij} << d$, $r_{ij} << d$)
  - Largest Eigen value of Laplace operator in sphere with mixed BC's



Cheviakov et al. (2010), Multiscale Model. Simul., v.8, pp.836–870







$$\lambda_i = \frac{1}{\tau_i} \approx \sum_{j=1}^{z_i} \frac{4 D_i a_{ij}}{V_i}$$

$$\bar{\tau} = \frac{1}{N_p} \sum_{i=1}^{N_p} \tau_i$$

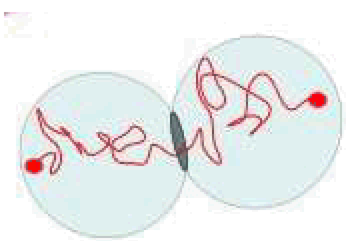
- Particle averaged, volume averaged MFPT ~ bulk conductivity

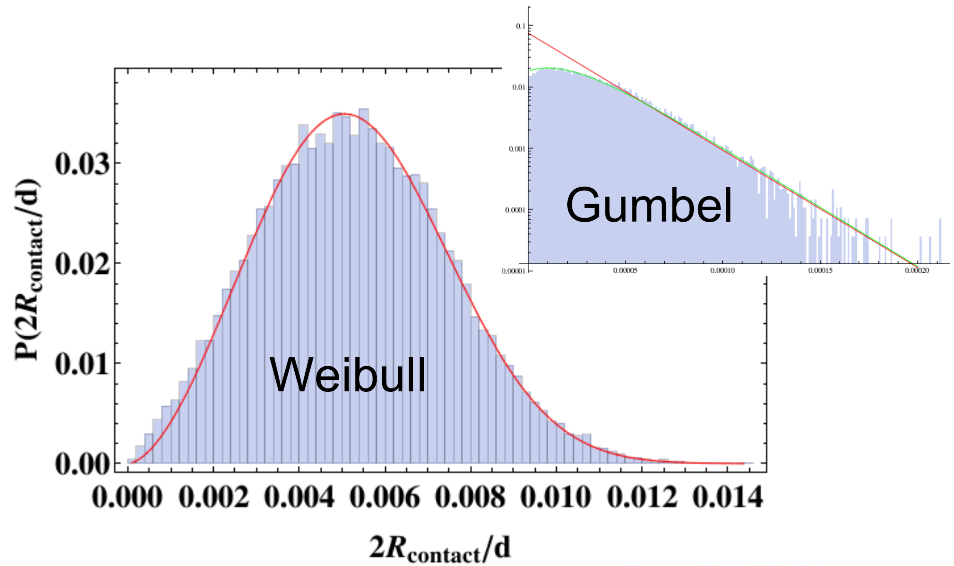
Microstructural Details: Particle-Particle Interfaces

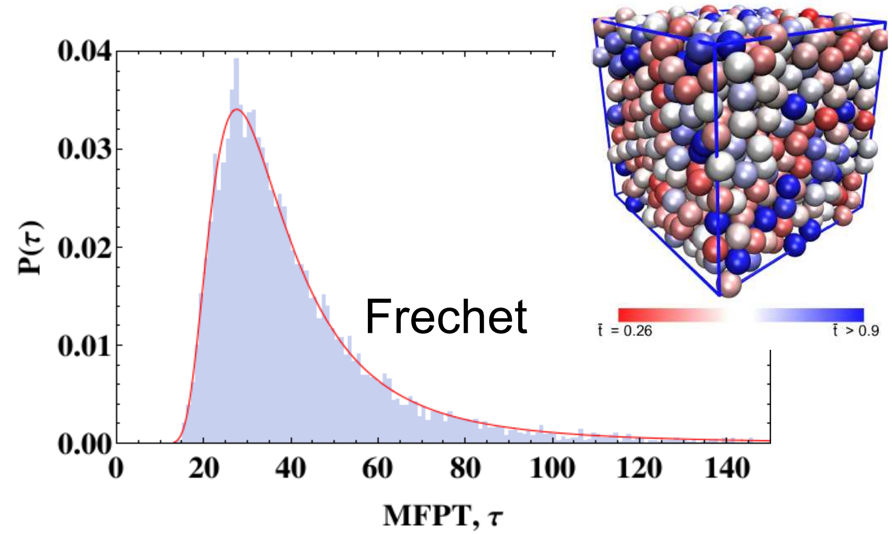
- **Difference from, say, SC lattice**
  - Disordered graph
  - Distribution of coord. #'s
  - Distribution of forces/"overlaps"
    - Distribution of contact radii
    - Distribution of volume-averaged MFPT
      - Narrow Escape
        - » Single and multiple contacts in well separated limit ($a << d$)
        - » Largest Eigen value of Laplace Operator in spherical domain with mixed (Dirichlet and Neumann) BC's

$$\bar{\tau} \sim \frac{1}{a}$$

$$\bar{\tau}_{z_i} \sim \sum_{j=1}^{z_i} \frac{1}{a_{ij}}$$







Homogenized Models: Bridging particle meso-scale to Bulk scale

- Consider Continuous-Time Random Walk a la Montroll and Wiess

  cf. Chaudhuri et al. (2010) PRL, v.99 , p.060604

  - Conditional probability of walker being at position r at time t

$$G_s(k,s) = f_{vib}(k)\left[\frac{1-\phi_1(s)+f(k)\left(\phi_1(s)-\phi_2(s)\right)}{s(1-\phi_2(s)f(k))}\right]$$

$$f(k) = f_{vib}(k) f_{jump}(k)$$

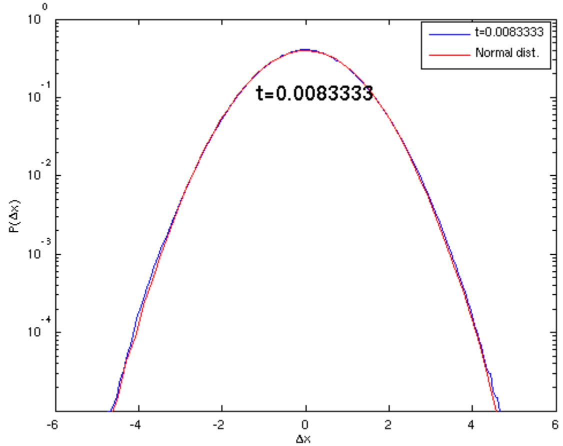
$$f_{vib}(k) = \left(2\pi\ell^2\right)^{-3/2}\exp\left(-r^2/2\ell^2\right)$$

$$f_{jump}(k) = \left(2\pi\lambda^2\right)^{-3/2}\exp\left(-r^2/2\lambda^2\right)$$

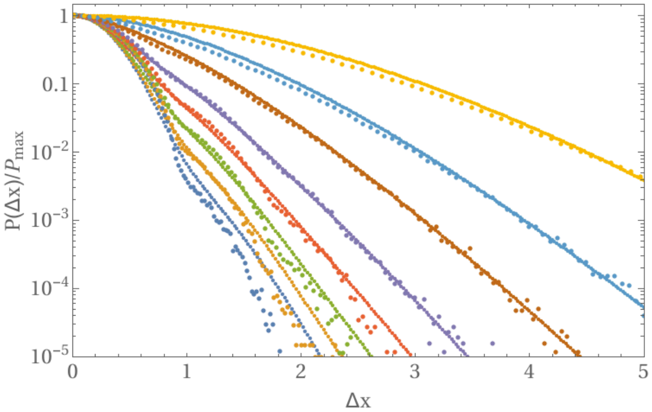
$$\phi_1 = \tau_1^{-1}\exp\left(-t/\tau_1\right)$$

$$\phi_2 = \tau_1^{-1}\exp\left(-t/\tau_2\right)$$

$p = 0.002$

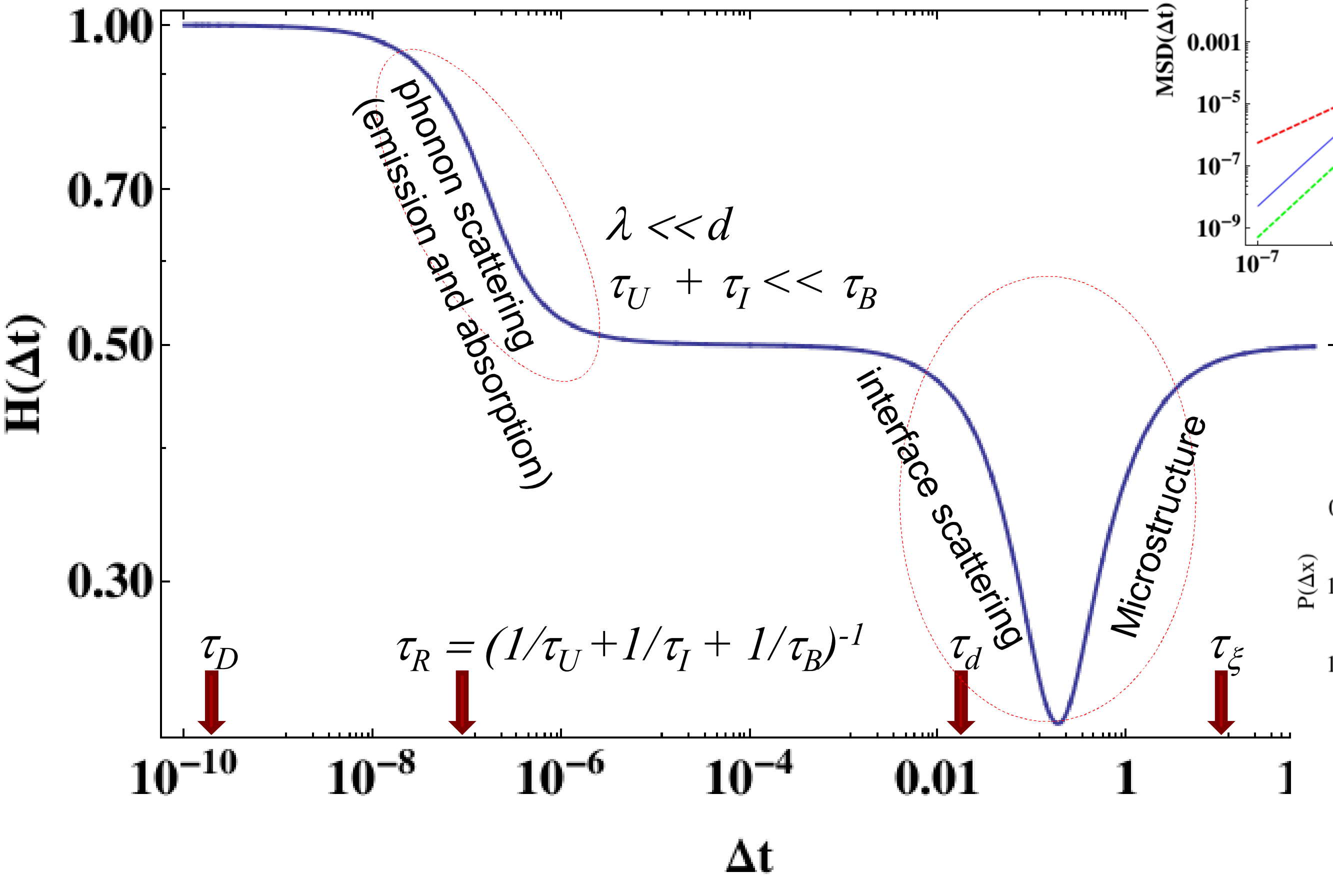


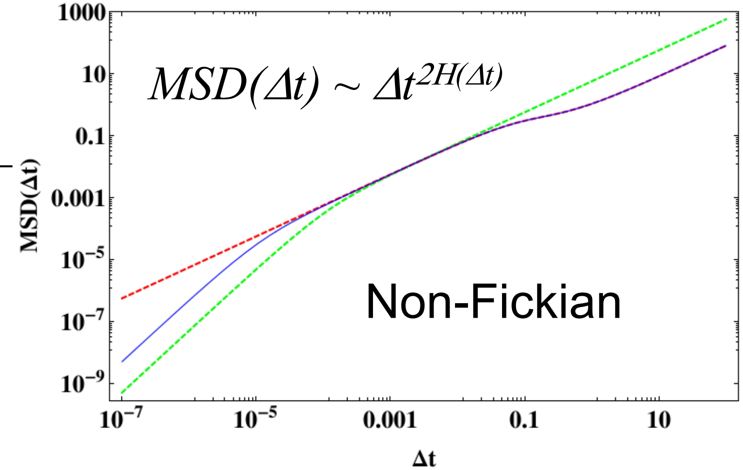
$p = 0.0004$

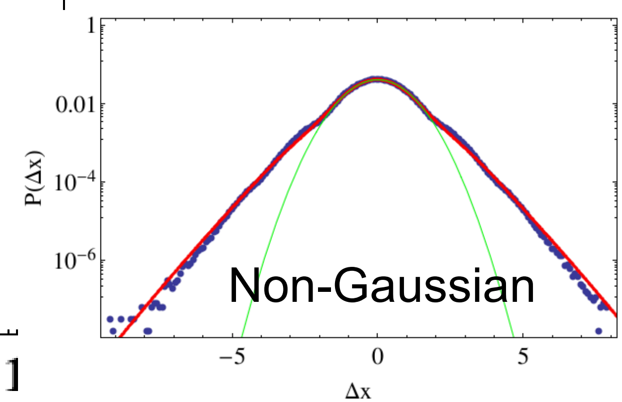


  - Equivalent to Generalized Master Equation

Transport Heterogeneity: Crossing Scales







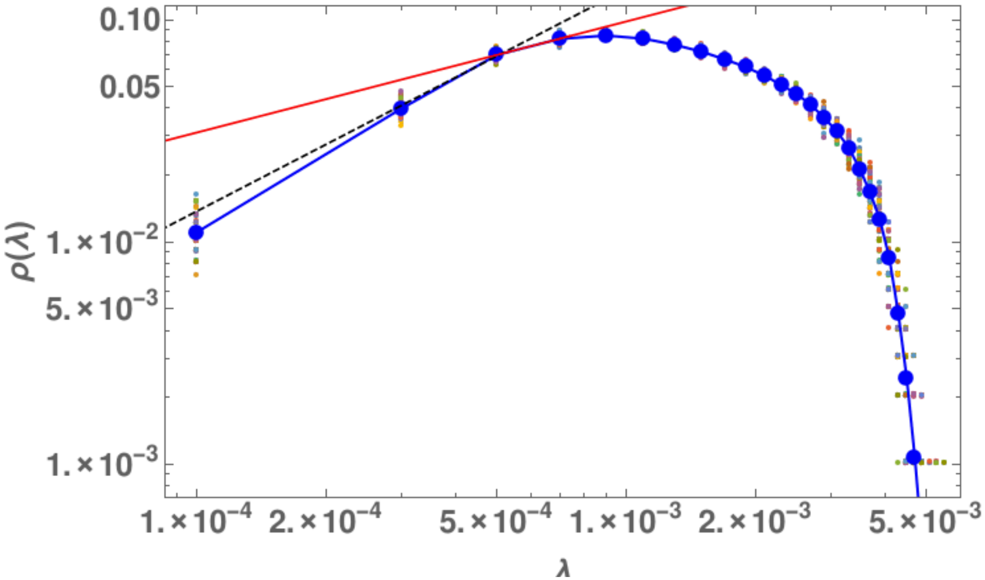
cf. K. Razi Naqvi and S. Waldenstrom (2005) *PRL* **95**, 065901

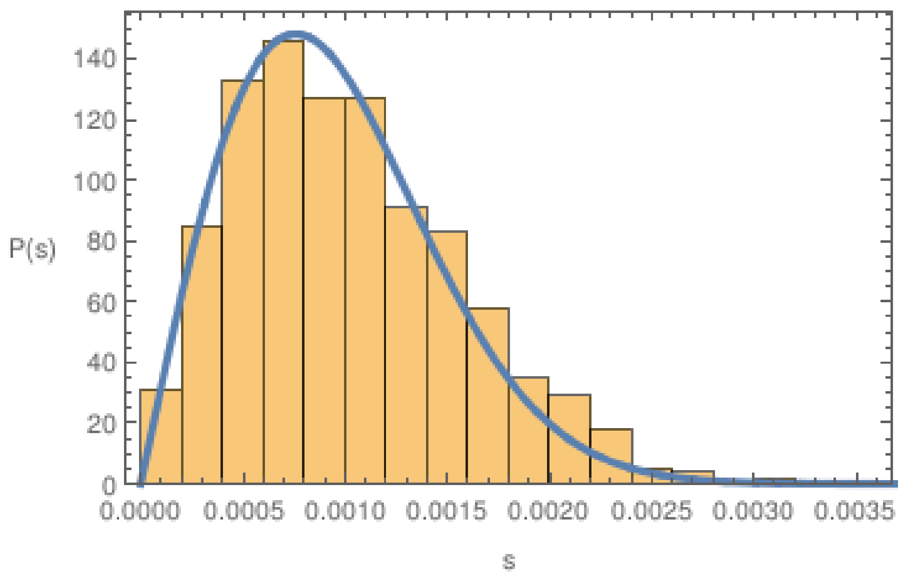
Spectral Analysis

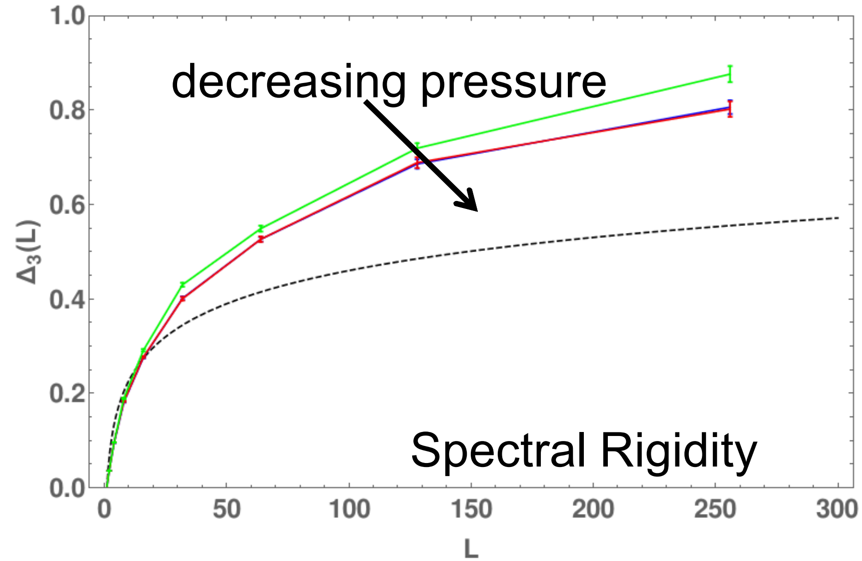
- **Transition Rate Matrix**

$$W_{ij} = \begin{cases} \dfrac{3D}{\pi R^2}\sqrt{\delta_{ij}/R} & i \neq j \\ -\sum_{j\neq i} W_{ij} & i = j \end{cases}$$

$$\delta_{ij} = 2R - \|\mathbf{r}_j - \mathbf{r}_i\| \geq 0$$







Meso-Macro Model Development

- Temperature distribution in isotropic, homogeneous, 3-dimensional, infinite medium classically modeled by heat equation; heated by an instantaneous point source at r=0

$$T(r,t) = \frac{Q\exp(-r^2/4Dt)}{8\pi\rho C(Dt)^{3/2}}$$

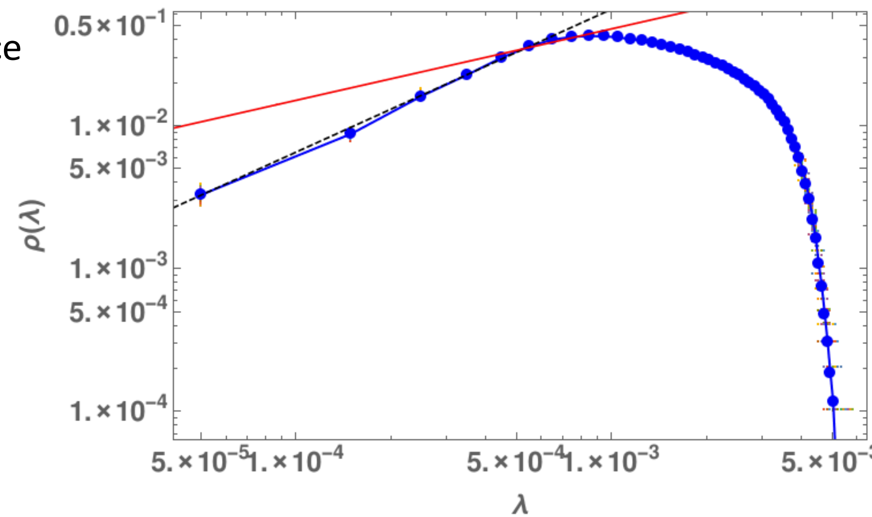
- Hence, T(0,t) scales as $T(0,t) \sim t^{-3/2}$
- Discrete case (transport on a graph) return probability

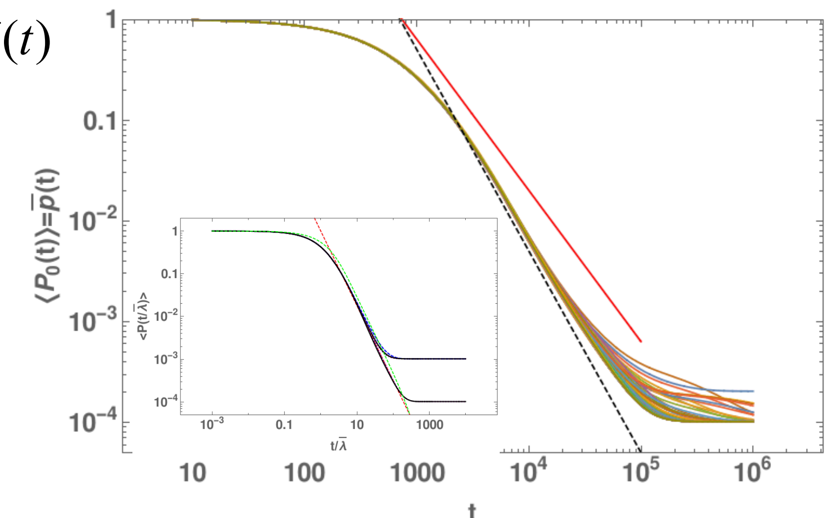
$$\overline{p}_{discr}(t) = \frac{1}{N}\sum_{n=1}^{N}\exp(-\lambda_n t)$$

- In "thermodynamic" (continuum) limit, N → ∞, $T(0,t) = \overline{p}(t)$

$$\overline{p}(t) = \int \rho(\lambda)\exp(-\lambda t)d\lambda$$

- Thus, if $\rho(\lambda) \sim \lambda^{\nu}$, $\overline{p}(t) \sim t^{-(1+\nu)}$ and $\nu = d/2-1$, with d = 3 for the homogeneous, isotropic case above
- Hence, scaling is anomalous with respect to classical descriptions
- Could be measured…





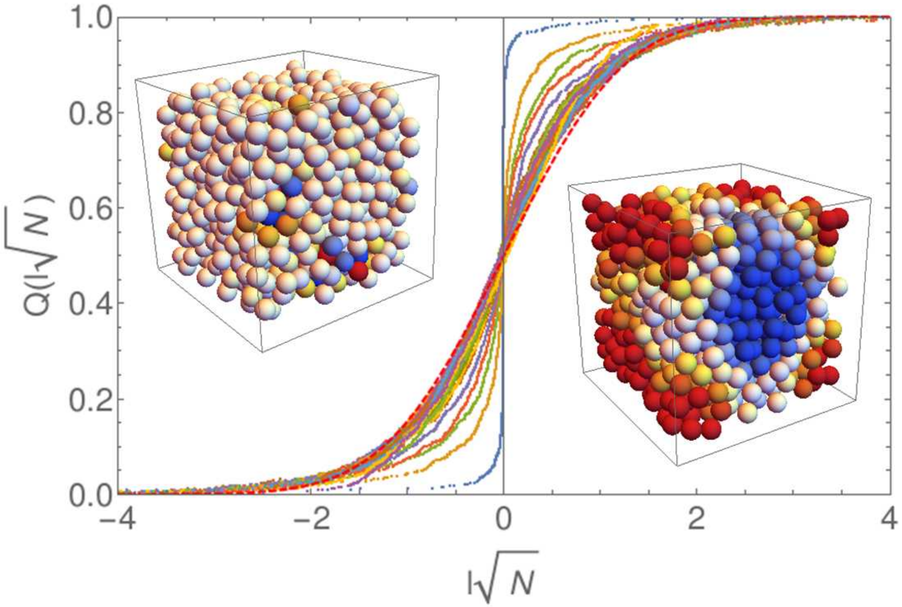
Eigenvectors and Statistics

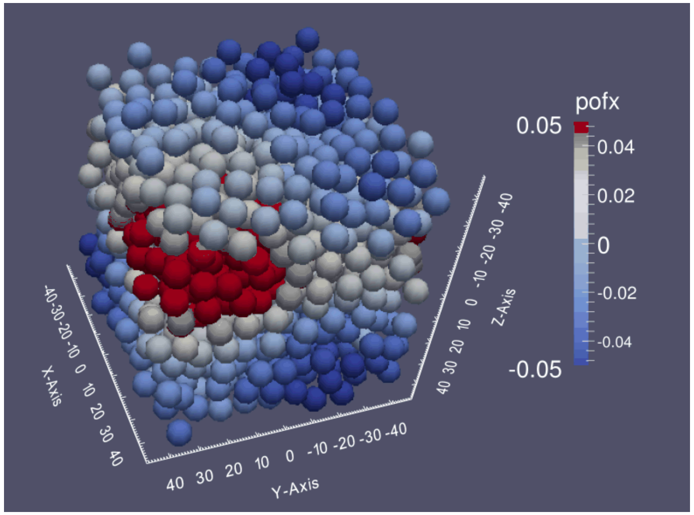
- Eigenvectors
  - small eigenvalues show plane

Cf. Silbert et al. (2009), PRE v.79, p.021308

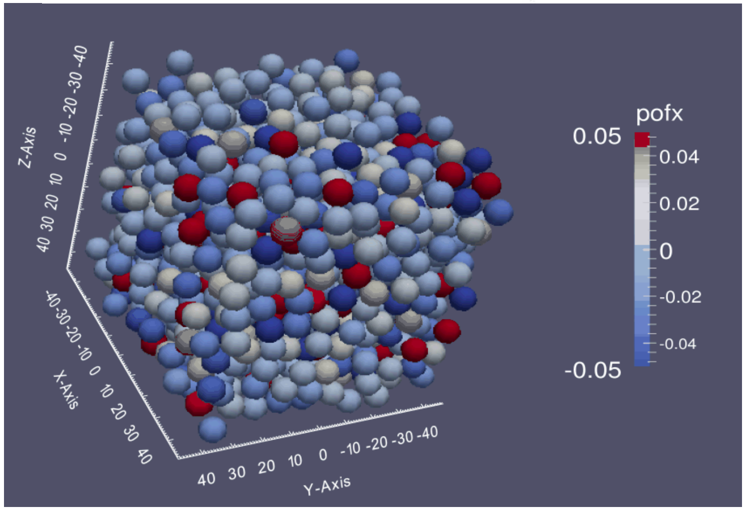
- Close to Porter-Thomas distribution
  - But, not quite

cf. Manning and Liu (2015), EPL v.109, p.36002





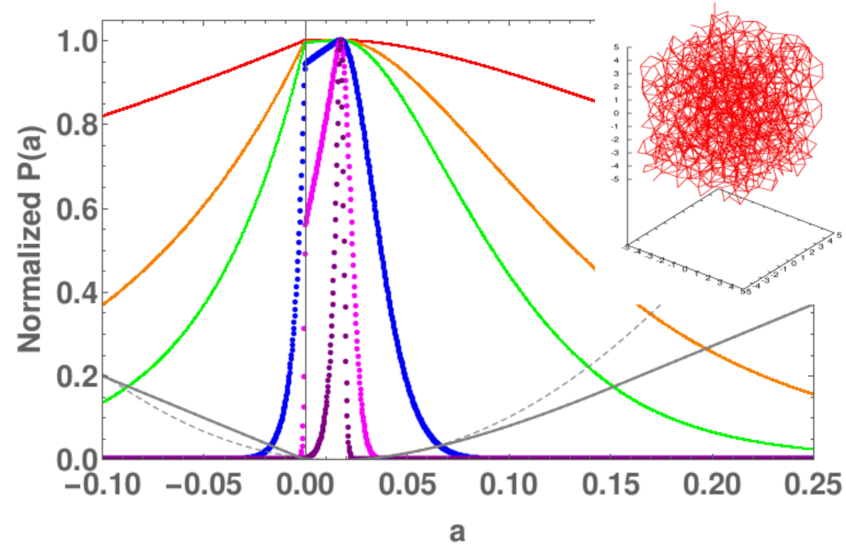
Eigenvector for large λ

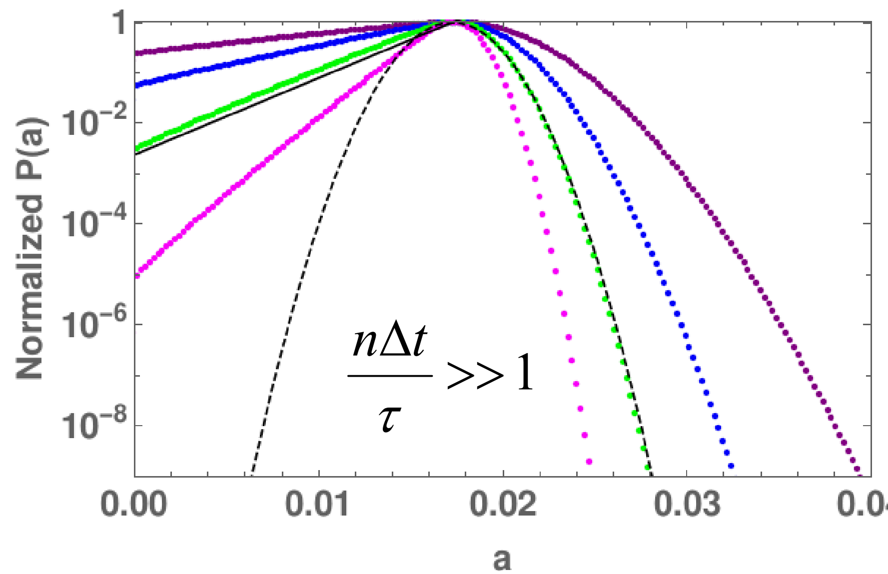


Eigenvector for small λ

Large Deviations in Disordered Networks

- Statistical Mechanics of "Trajectories"
- Use thermodynamic formalism for systems with Markovian dynamics
  - Largest Eigen value of *modified* transition rate matrix is dynamical free energy
  - The negative of the rate function can be viewed as a dynamical entropy
- Obtain convergence (in distribution) of fluctuations in diffusion coefficient
- Distributions reminiscent of "Extreme Value Statistics" (e.g., Gumbel distribution)





Large Deviation Function

- SC lattice vs. Jammed network
  - Dynamic Phase Transition?