Statistical Mechanics of Granular Materials

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I Introduction
II Density relaxation in granular compaction
III Knots in granular chains
IV Shocks in inelastic gases
V Conclusions & Outlook

Theory: Grossman, Zhou (Chicago), Krapivsky, Redner (Boston)
Simulation: Chen, Nie (Johns Hopkins)
Experiment: Daya, Ecke, Vorobief (LANL), Knight, Nowak, Jaeger, Nagel (Chicago)

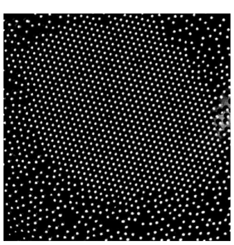
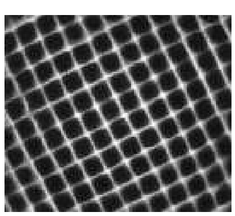
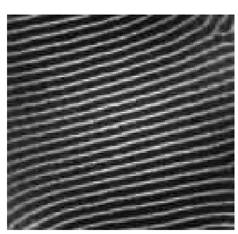
Granular Materials

Properties:

- Ensembles of macroscopic particles
- Interaction - hard core exclusion
- Collisions - dissipative

Interesting collective phenomena:

- Phase transitions — Urbach 98
- Pattern formation — Swinney 95
- Solitary waves — Umbanhowar 95
- Force chains — Coppersmith 95
- Size segregation — Knight 93

Compaction

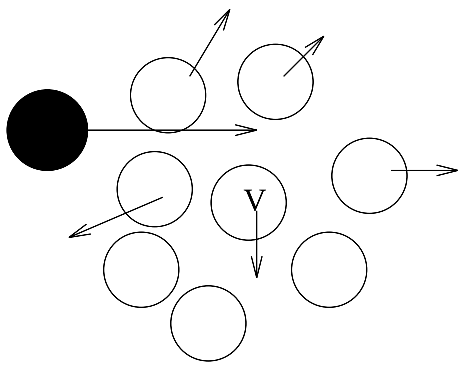
- Uniform, simple system
- Probes the density - a fundamental quantity
- Slow density relaxation Knight 95

$$\rho(t) = \rho_\infty - \frac{\rho_\infty - \rho_0}{1 + B \ln(t/\tau)}$$

- Parameters depend on Γ only
- Robust behavior - independent of grain type, grain size, container geometry, etc.

What causes logarithmic relaxation?

Heuristic picture



$$\rho = \text{volume fraction}$$
$$V = \text{particle volume}$$
$$V_0 = \text{pore volume/particle}$$

$$\rho = \frac{V}{V+V_0} \qquad \text{or} \qquad V_0 = V\frac{1-\rho}{\rho}$$

Assumption: Cooperative rearrangement

$$NV_0 = V \qquad \text{or} \qquad N = \frac{\rho}{1-\rho}$$

Assumption: Exponential rearrangement time

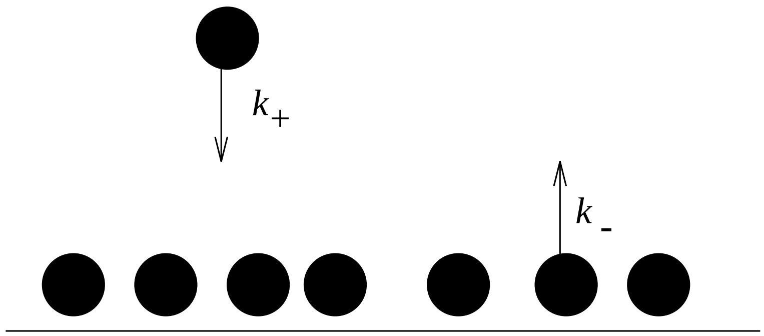
$$T = e^N = e^{\frac{\rho}{1-\rho}}$$

$$\frac{d\rho}{dt} \propto (1-\rho)\frac{1}{T} = (1-\rho)e^{-\frac{\rho}{1-\rho}}$$

$$\rho(t) \cong 1 - \frac{1}{\ln t}$$

Volume exclusion causes slow relaxation

The “parking” model



- 1D Adsorption-desorption process
- Adsorption subject to volume constrains
- Desorption not restricted
- Detailed balance satisfied
- System reaches equilibrium steady state

Ignores: mechanical stability

Realistic: excluded volume interaction

Theory

$P(x,t) =$ Density of x-size voids at time t

$$1 = \int dx(x+1)P(x,t) \qquad \rho(t) = \int dx P(x,t)$$

Master equation:

$$\frac{\partial P(x)}{\partial t} = 2k_+ \int_{x+1} dy P(y) - 2k_- P(x)$$
$$+\theta(x-1)\left[\frac{k_-}{\rho(t)}\int_0^{x-1} dy P(y) P(x-1-y) - k_+(x-1)P(x)\right]$$

Density rate equation:

$$\frac{\partial \rho(t)}{\partial t} = -k_- \rho(t) + k_+ \int_1 dx(x-1)P(x,t)$$

Convolution term assumes voids are uncorrelated (exact in equilibrium)

Exact Equilibrium Properties

Exponential void distribution

$$P_\infty(x) = \frac{\rho_\infty^2}{1-\rho_\infty} \exp\left[-\frac{\rho_\infty}{1-\rho_\infty} x\right]$$

Sticking Probability

$$S(\rho_\infty) = \exp\left[-\frac{\rho_\infty}{1-\rho_\infty}\right]$$

Gaussian Density Distribution

$$P_\infty(\rho) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left[-\frac{(\rho-\rho_\infty)^2}{2\sigma^2}\right]$$

Variance decreases with density

$$\sigma^2 = \rho_\infty (1-\rho_\infty)^2 / L \qquad \beta = 2$$

$\boxed{\text{Volume exclusion dominates at high densities}}$

Relaxation Properties

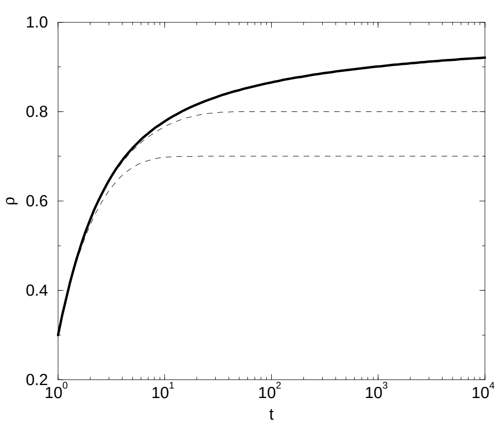
Quasistatic (near equilibrium) approximation

$$\frac{\partial \rho(t)}{\partial t} = -k_- \rho(t) + k_+ (1-\rho) \exp\left[-\frac{\rho}{1-\rho}\right]$$

I Desorption-limited case $(k_- \to 0)$

$$\rho(t) \cong 1 - \frac{1}{\ln k_+ t}$$

II Finite k_- $\tau = (L/k_- \rho_\infty)\sigma^2 = (1-\rho_\infty)^2/k_-$



$$\rho(t) \cong \begin{cases} 1 - \frac{1}{\ln k_+ t} & t \ll \tau \\ \rho_\infty - A e^{-t/\tau} & t \gg \tau \end{cases}$$

Slow density relaxation

The sticking probability

Total adsorption rate

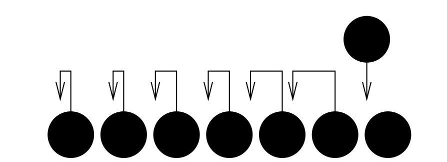
$$\int_1 dx(x-1)P_\infty(x) = k_+(1-\rho_\infty)\exp\left[-\frac{\rho_\infty}{1-\rho_\infty}\right]$$

Reduced adsorption rate $k_+ \to k_+ s(\rho)$

Sticking probability

$$s(\rho) = e^{-N} \qquad N = \frac{\rho}{1-\rho}$$

Heuristic picture is exact in 1D



Cooperative behavior in dense limit

Spectrum of density fluctuations

Definition

$$\text{PSD}(f) = \left| \int d\tau e^{if\tau} \langle \rho(t)\rho(t+\tau) \rangle \right|^2$$

Leading behavior

$$\text{PSD}(f) \cong \begin{cases} f^0 & f \ll f_L \\ f^{-\alpha} & f_L \ll f \ll f_H \\ f^{-2} & f_H \ll f \end{cases}$$

For noninteracting dilute case, linear theory, $\text{PSD}(f) \propto [1 + (f/f_0)^2]$, with $f_0 = \tau^{-1} = k_+ + k_-$

In general, still open problem. Reasonable that $f_L \approx k_-$ and $f_H \approx k_+$

Similar noise spectrum for finite system Monte Carlo and experimental data

Conclusions I

- Compaction dominated by exponentially rare grain size voids
- Growing time scales associated with cooperative bead rearrangements
- Argument is general - should hold for aspherical grains or horizontal tapping
- Gaussian density fluctuations

Vibrated Knot Experiment

- $t = 0$: knot placed at chain center
- Parameters:

— Number of monomers: $30 < N < 200$
— Minimal knot size: $N_0 = 15$

- Driving conditions:

— Frequency: $\nu = 13Hz$
— Acceleration: $\Gamma = A\omega^2/g = 2.37$

- Measurement: opening time t

Questions

1. Average opening time $\tau(N)$?
2. Survival probability $S(t, N)$?
   Distribution of opening times $R(t, N)$?

Motivation

Topological constraints, entanglements:

- Reduce accessible phase space
- Involve large relaxation time scales
- Affect dynamics, flow

$$\eta \sim \tau \sim N^3$$

Relevance:

- Polymers: melts, rubber, gels
- DNA, biomolecules

Difficulties:

- Hard to observe directly
- Slow dynamics
- Finite size effects

Granular "Polymers"

Mechanical bead-spring:

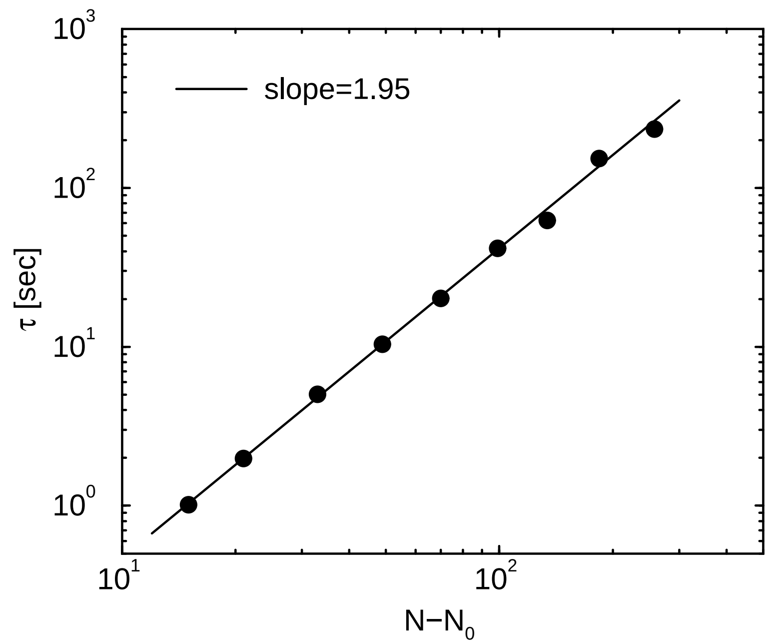
$$U(\{\mathbf{R}_i\}) = v_0 \sum_{i\neq j} \delta(\mathbf{R}_i - \mathbf{R}_j) + \frac{3}{2b^2} \sum_i (\mathbf{R}_i - \mathbf{R}_{i+1})^2$$

- Beads interact via hard core repulsions
- Rods act as springs (nonlinear, dissipative)
- Inelastic collisions: bead-bead, bead-plate
- Vibrating plate supplies energy

Advantages:

- Number of "monomers" can be controlled
- Topological constraints: can be prepared, observed directly

The Average Opening Time



$$\tau(N) \sim (N - N_0)^{\nu} \qquad \nu = 2.0 \pm 0.1$$

$\boxed{\text{Opening time is diffusive}}$

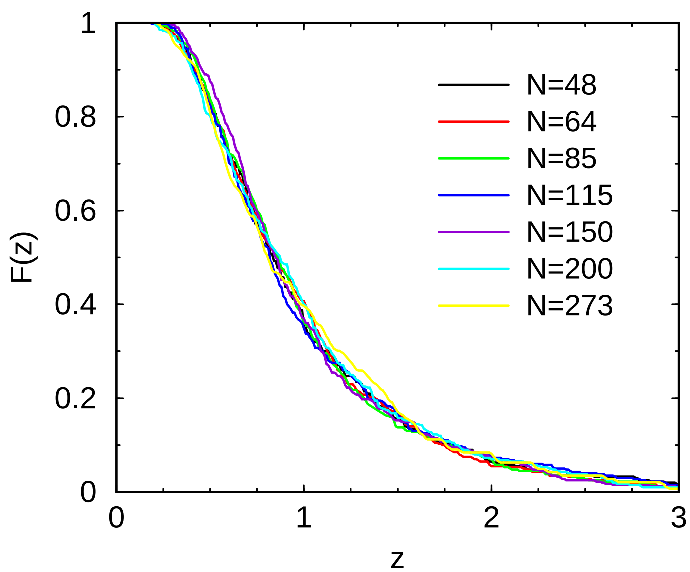
The Survival Probability

- $S(t, N)$ Probability knot "alive" at time t
- $R(t, N)$ Probability knot opens at time t

$$R(t, N) = -\frac{d}{dt} S(t, N)$$

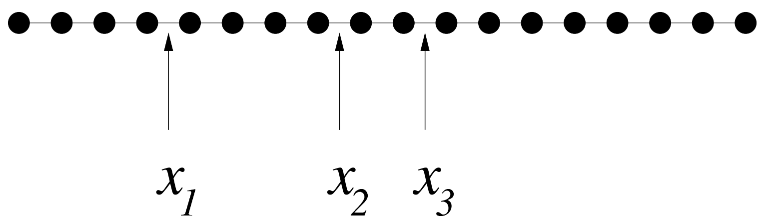
- $S(t, N)$ obeys scaling

$$S(t, N) = F(z) \qquad z = \frac{t}{\tau(N)}$$



$\boxed{\tau \text{ only relevant time scale}}$

Theoretical Model



Assumptions:

- Knot $\equiv$ 3 exclusion points
- Points hop randomly
- Points move independently (no correlation)
- Points are equivalent (size $= N_0/3$)

3 Random Walk Model:

- 1D walks with excluded volume interaction
- first point reaches boundary $\rightarrow$ knot opens

Diffusion in 3D

- Continuum limit $x_i \to \infty$, $N \to \infty$
- Dimensionless time $t \to Dt/[(N-N_0)^2]$, space $(x,y,z) = (x_1,x_2,x_3)/N$

$$1<x_1<x_2<x_3<N-N_0 \quad \longrightarrow \quad 0<x<y<z<1$$

$$\frac{\partial}{\partial t}P(x,y,z,t) = \nabla^2 P(x,y,z,t)$$

- Boundary conditions

Absorbing: $P\big|_{x=0} = P\big|_{x=1} = 0$

Reflecting: $(\partial_x - \partial_y)P|_{x=y} = (\partial_y - \partial_z)P|_{y=z} = 0$

- Initial conditions $P\big|_{t=0} = \delta(x-x_0)\delta(y-x_0)\delta(z-x_0)$

- Survival probability

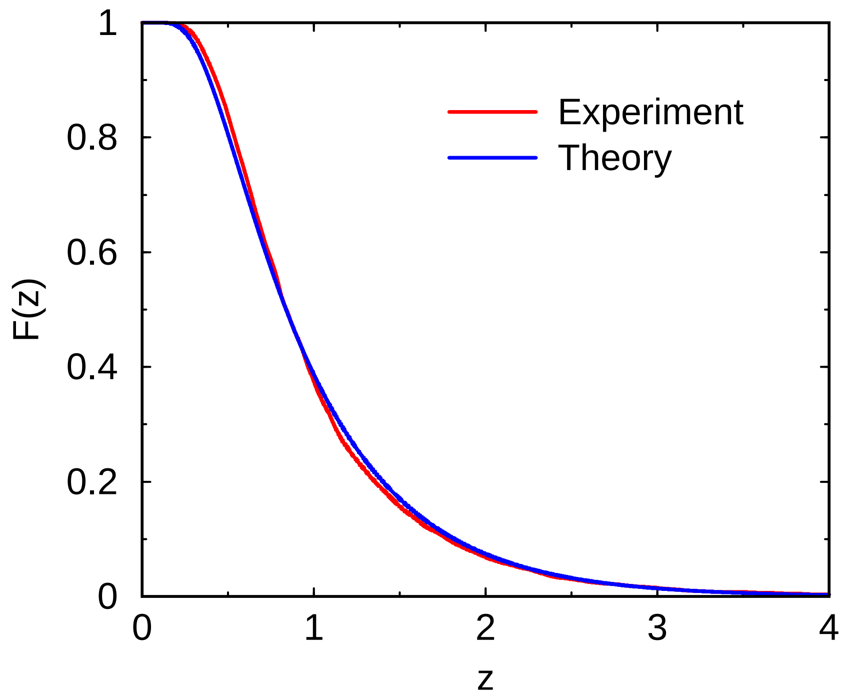
$$S_3(t) = \int_0^1 dx \int_x^1 dy \int_y^1 dz\ P(x,y,z,t)$$

$$\boxed{\textbf{3 walks in 1D} \equiv \textbf{1 walk in 3D}}$$

Experiment vs. Theory

- Work with scaling variable $z = t/\tau$ ($\langle z \rangle = 1$)
- Combine different data sets (5000 pts)
- Fluctuations $\sigma^2 = \langle z^2 \rangle - \langle z \rangle^2$

$$\sigma_{\rm exp} = 0.62(1),\ \sigma_{\rm theory} = 0.63047\ (< 2\%)$$



Quantitative agreement

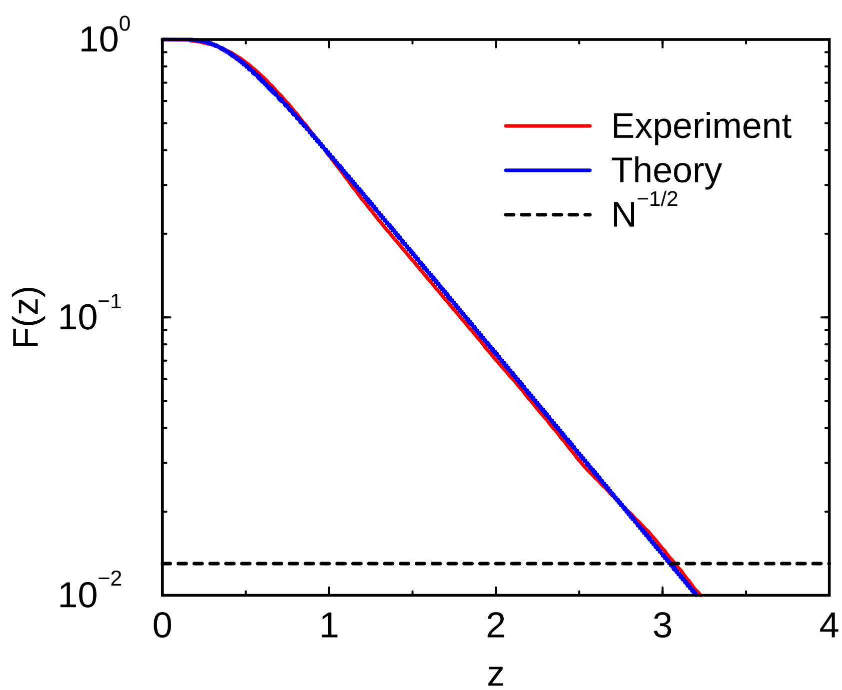
Large Exit Times

- Largest decay time dominates
- Large time tail is exponentially small

$$F(z) \sim e^{-\beta z} \qquad z \to \infty$$

- Decay coefficient

$$\beta_{\text{exp}} = 1.65(2) \qquad \alpha_{\text{theory}} = 1.66440 \qquad (1\%)$$



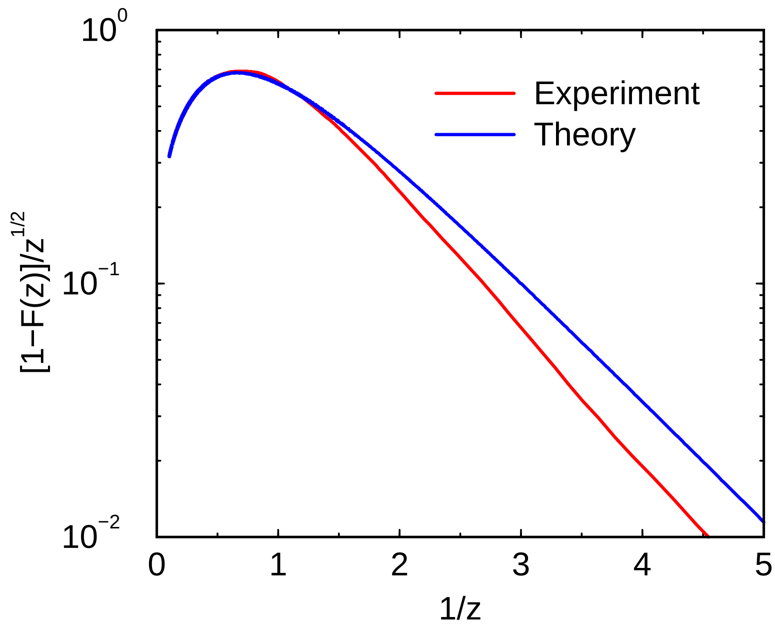
Small Exit Times

- Exponentially small (in $1/z$) tail

$$1 - F(z) \sim z^{1/2} e^{-\alpha/z} \qquad z \to 0$$

- Decay coefficient

$$\alpha_{\text{exp}} = 1.2(1),\ \beta_{\text{theory}} = 1.11184\ (10\%)$$



Larger discrepancy

Conclusions II

- Opening times are diffusive
- Distributions obey scaling
- Extreme statistics are exponential
- Macroscopic observables $(t,\ S(t))$ reveals microscopic dynamics
- Knot dynamics determined by 3 diffusing exclusion points

Outlook II

- $\tau(N)$ gives size of constraint N_0
- $S(t, N)$ gives number of constraints m
- Is motion uncorrelated?
- Is a more detailed model necessary?

Possibilities

Granular Matter

- Phase transitions in monolayers
- Compaction
- Segregation
- Stress Propagation

Polymers

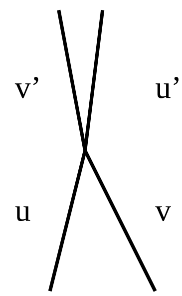
- Chain dynamics
- Entanglements
- Phase separation

Inelastic collisions

- Relative velocity reduced by $r = 1 - 2\epsilon$

$$\begin{aligned}\Delta v' &= -r\Delta v \\ v' &= v - \epsilon\Delta v\end{aligned}$$

- Energy dissipation $\Delta E \propto -\epsilon(\Delta v)^2$



Freely evolving gas

- N point particles in 1D ring.
  Random velocity distribution.
  Typical velocity v_0. Typical distance x_0.

- **Dimensionless variables** $x \to x/x_0$, $t \to tv_0/x_0$

— "Temperature" $T(t) = \langle v^2(t)\rangle - \langle v(t)\rangle^2$

— Characteristic time/length scales.

— Continuum theory?

Motivation: Granular Gases

- Applications:
  - — Granular materials: powders, grains.
  - — Geophysical flows.
  - — Large scale formation in universe.
- Characteristics:
  - — Hard sphere interactions.
  - — Dissipative collisions.
- Experimental observations (1D, 2D, 3D):
  - — **Density** inhomogeneities.
  - — **Velocity** correlations, non-Gaussian stat
  - — **Phase transitions:** order-disorder.

Mean Field Theory

- Energy dissipation

$$\Delta T \propto -\epsilon(\Delta v)^2$$

- Collision frequency

$$\Delta t \sim \ell/\Delta v \sim (\Delta v)^{-1}$$

- Assuming a uniform gas

$$\begin{aligned} v &\sim \Delta v \sim T^{1/2} \qquad \Delta\ell/\ell \ll 1 \\ \frac{dT}{dt} &\propto \frac{\Delta T}{\Delta t} \propto -\epsilon(\Delta v)^3 \propto -\epsilon\, T^{3/2} \end{aligned}$$

- Cooling law

Haff 83

$$T(t) \simeq (1 + A\epsilon t)^{-2} \sim \begin{cases} 1 & t \ll \epsilon^{-1} \\ \epsilon^{-2} t^{-2} & t \gg \epsilon^{-1} \end{cases}$$

Holds only in early homogeneous phase

The Inelastic Collapse

Bernu 90, Young 91

- 3 particles clump if $r < r_c = 7 - 4\sqrt{3} \cong 0.07$
- Finite time singularity: Cluster formation via infinite collisions when $N > N_c(r)$.
- Estimating the critical mass:

$$
\begin{aligned}
v_1 &\cong 1 - \epsilon \\
&\vdots \\
v_N &\cong 1 - N\epsilon
\end{aligned}
$$

- Particle passes through if $N < N_c(\epsilon) \sim \epsilon^{-1}$
- $N_c \sim \epsilon^{-1} \Rightarrow$ collapse always encountered in the thermodynamic limit $N \to \infty$.

Particles coalesce rather than pass through

The Sticky Gas ($r = 0$)

- Multiparticle aggregate of typical mass m
- **Momentum conservation** CPY 90

$$P_m = \sum_{i=1}^{m} P_i \quad \Rightarrow \quad P \sim m^{1/2}, \quad v \sim m^{-1/2}$$

- **Mass conservation**

$$\rho = cm = \text{const} \quad \Rightarrow \quad c \sim m^{-1}$$

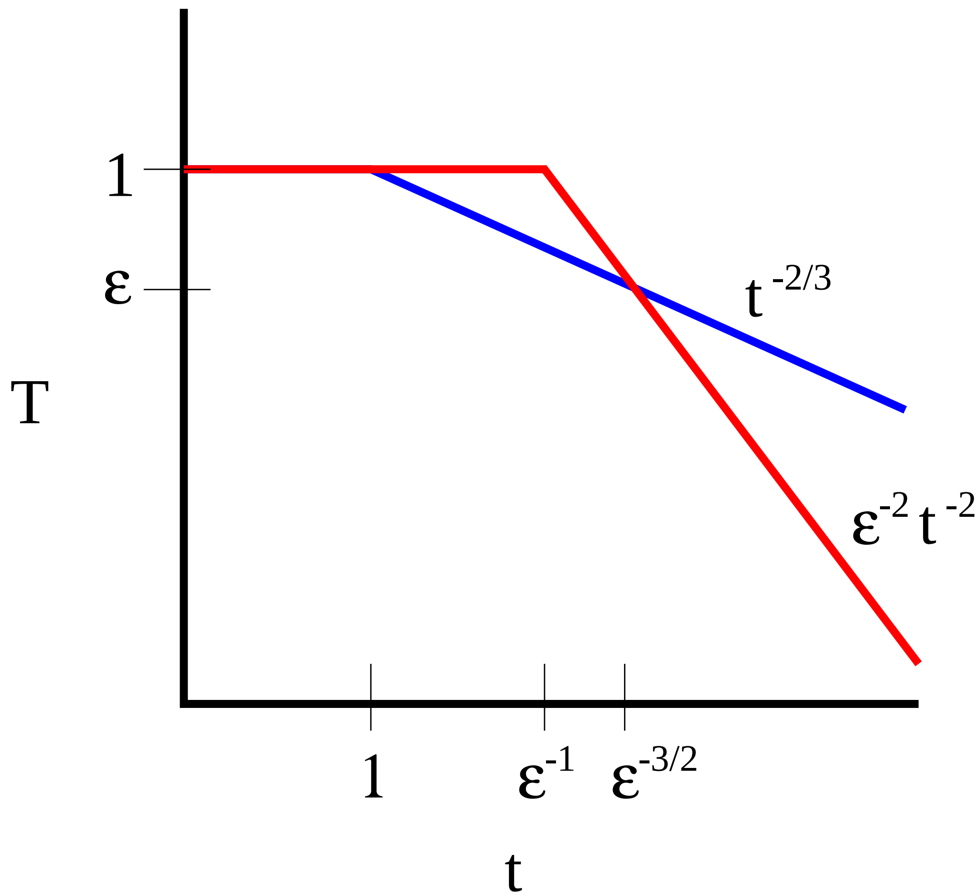
- **Dimensional analysis** $[cv] = [t]^{-1}$

$$m \sim t^{2/3} \quad v \sim t^{-1/3} \quad T \sim t^{-2/3}$$

- **Final state** 1 aggregate with $m = N$

$$T(t) \sim \begin{cases} 1 & t \ll 1; \\ t^{-2/3} & 1 \ll t \ll N^{3/2}; \\ N^{-1} & N^{3/2} \ll t \end{cases}$$

Monotonicity



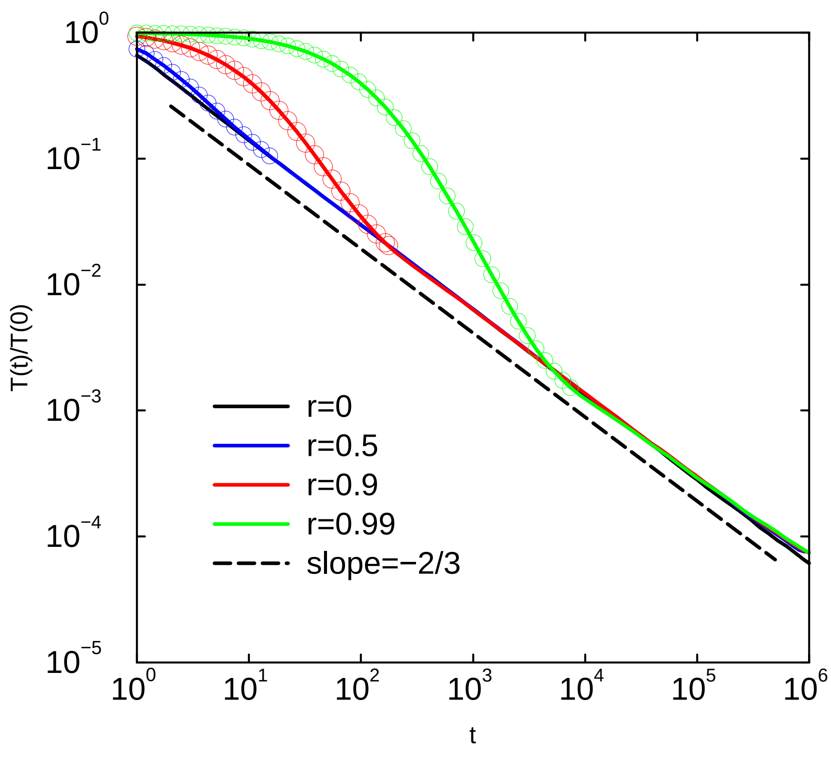
- $T(\epsilon, t)$ decreases monotonically with ϵ, t

Sticky gas $r = 0$ ($\epsilon = 1/2$) is a lower bound

Crossover Picture

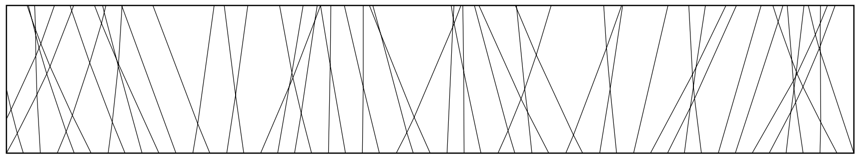
- Universal Cooling law $T(t) \sim t^{-2/3}$

$$T(t) \sim \begin{cases} 1 & t \ll \epsilon^{-1}; \\ \epsilon^{-2} t^{-2} & \epsilon^{-1} \ll t \ll \epsilon^{-3/2}; \\ t^{-2/3} & \epsilon^{-3/2} \ll t \ll N^{3/2}; \\ N^{-1} & N^{3/2} \ll t \end{cases}$$

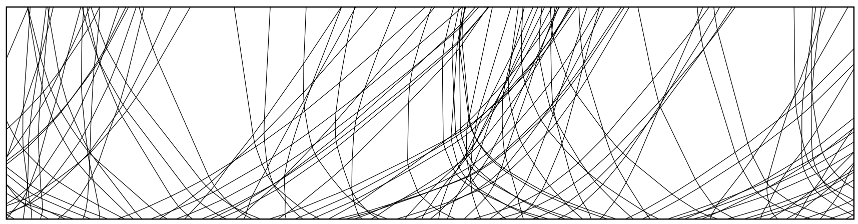


Asymptotic behavior is independent of r

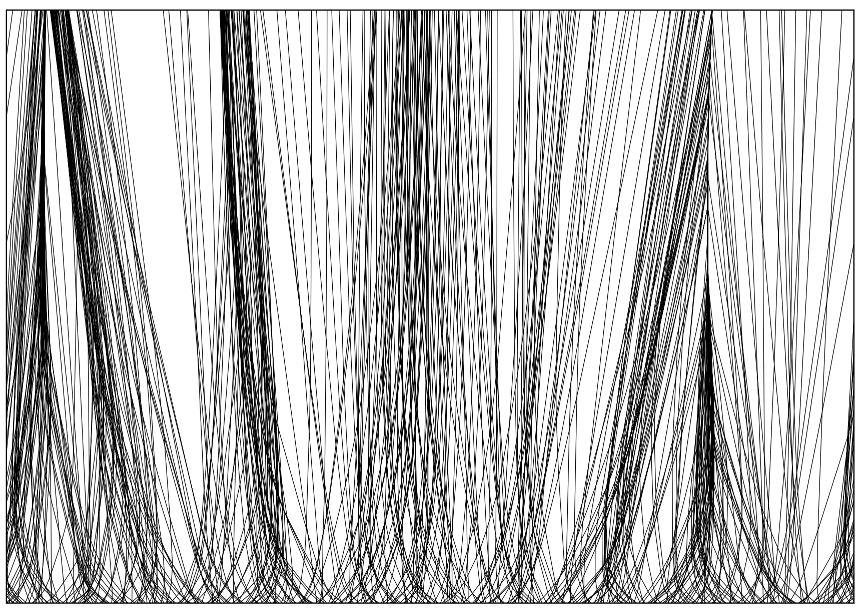
Early = Elastic gas ($r = 1$)



Intermediate = Inelastic gas ($r = 0.9$)



Late = Sticky gas ($r = 0$)



$\boxed{r = 0 \textbf{ is fixed point}}$

The Velocity Distribution

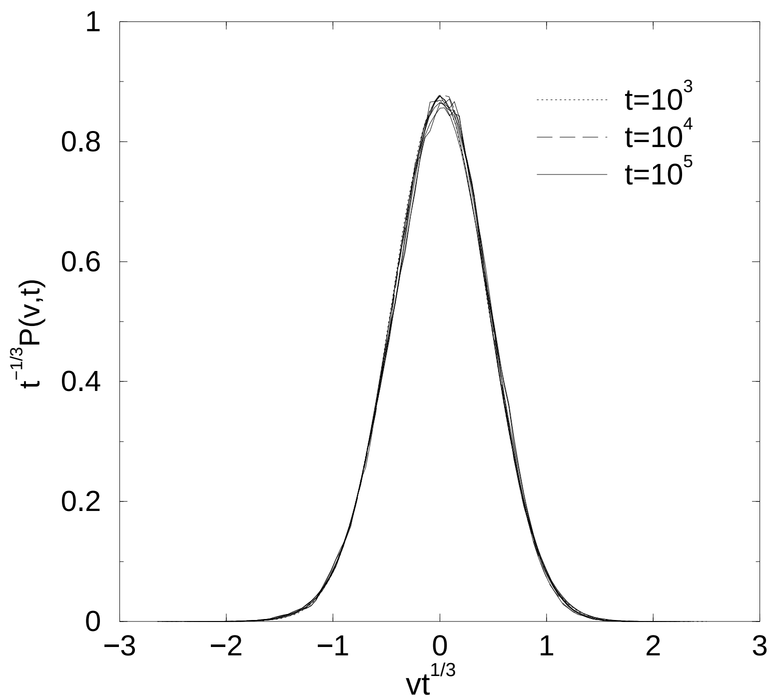
- Self similar distribution

$$P(v,t) \sim t^{1/3}\Phi(vt^{1/3})$$

- Large velocity tail

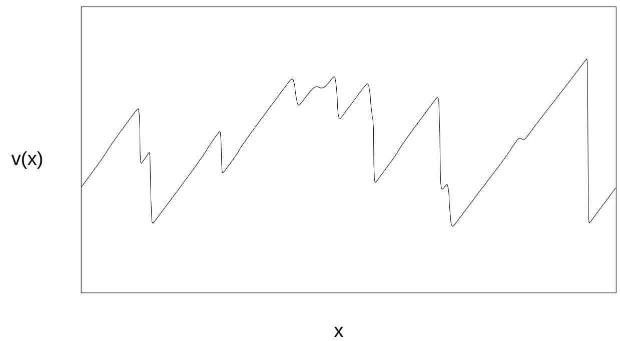
$$\Phi(z) \sim \exp(-\text{const.} \times z^3) \qquad z \gg 1$$

- Simulation results $r = 0,\ 0.5,\ 0.9$



$\boxed{P(r,v,t) \textbf{ is function of } z = vt^{1/3} \textbf{ only}}$

The Inviscid Burgers Equation



- Nonlinear diffusion equation

$$v_t + v v_x = \nu v_{xx} \qquad \nu \to 0$$

- Transform to linear diffusion equation

$$u_t = \nu u_{xx} \qquad \Leftarrow \qquad v = -2\nu(\ln u)_x$$

- Sawtooth (shock) velocity profile

$$v(x,t) = \frac{x - q(x,t)}{t}$$

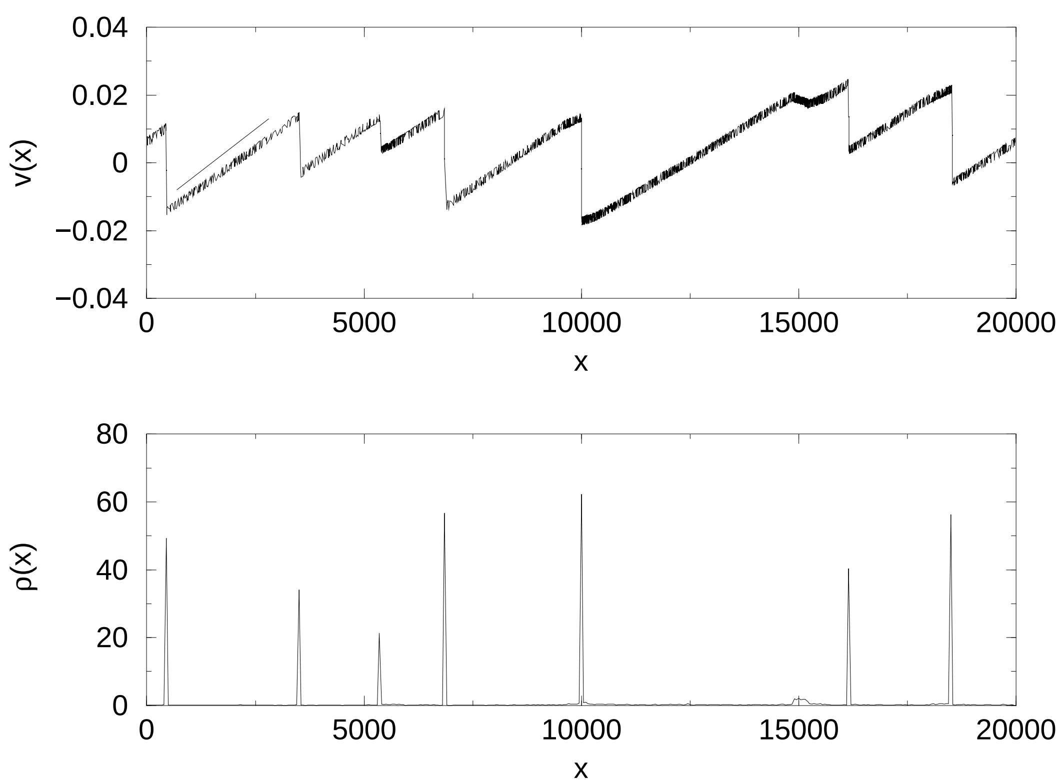
- Shock collisions conserve mass & momentum

- Describes "sticky gas" $r = 0$ Zeldovich RMP 89

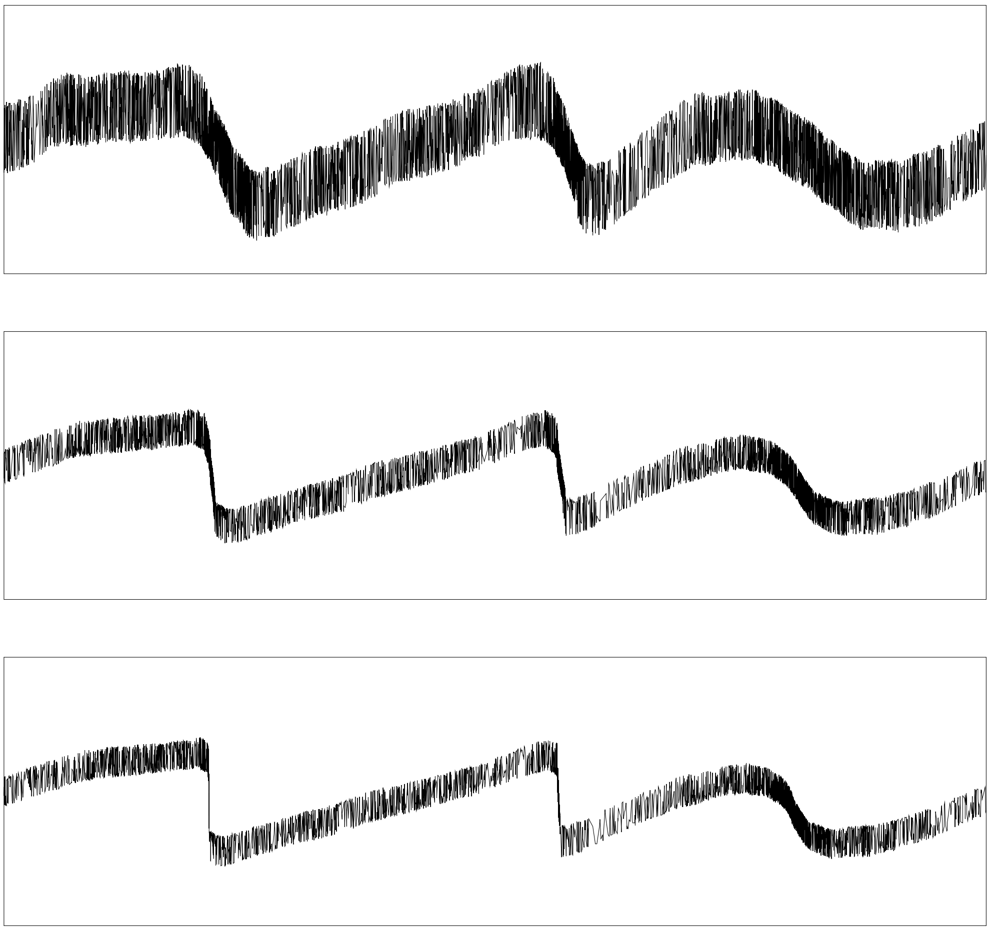
Burgers equation ≡ sticky gas ≡ inelastic gas

Predictions verified in 1D

- Velocity statistics $v \sim t^{-1/3}$
- Discontinuous (shock) velocity profile
- Slope $= t^{-1}$ (simulation with $r = 0.99$)



Formation of Singularity



$\boxed{\textbf{Collapse} \equiv \textbf{finite time singularity in } v_t + v v_x = 0}$

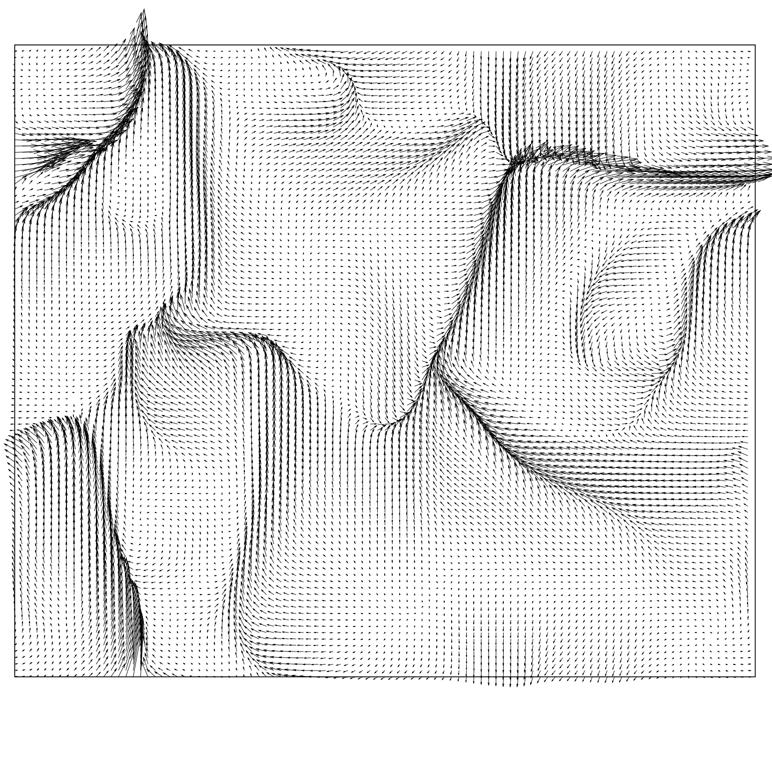
Higher Dimensions

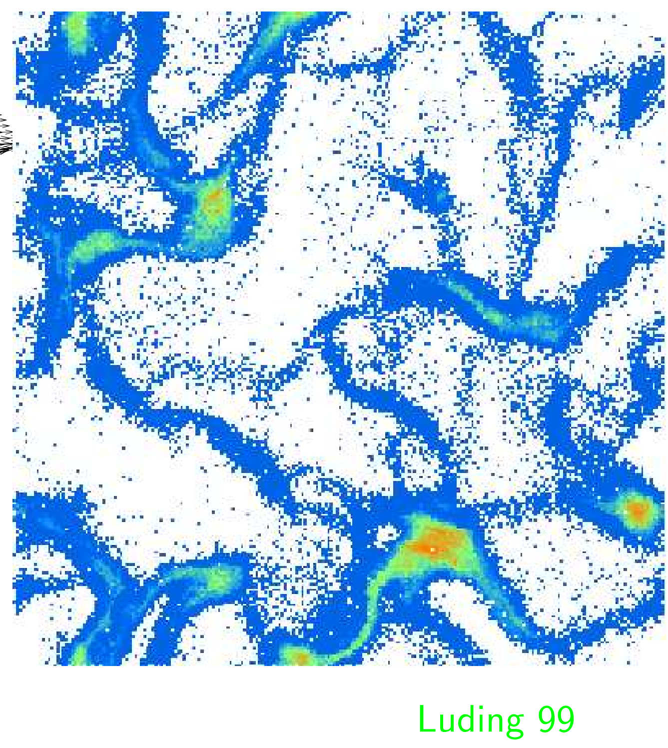
If $r_{\text{eff}} = 0$, then $\mathbf{v}_t + \mathbf{v} \cdot \nabla \mathbf{v} = \nu \nabla^2 \mathbf{v}$ predicts:

- Cooling law for $2 \leq d \leq 4$

$$T(t) \sim \begin{cases} 1 & t \ll \epsilon^{-1}; \\ \epsilon^{-2} t^{-2} & \epsilon^{-1} \ll t \ll \epsilon^{-4/(4-d)}; \\ t^{-d/2} & \epsilon^{-4/(4-d)} \ll t \ll N^{2/d}; \\ N^{-1} & N^{2/d} \ll t \end{cases}$$

- Critical size for collapse $N_c(\epsilon) \sim \epsilon^{-2d/(4-d)}$





Luding 99

Conclusions III

Asymptotic behavior:

- **Governed by cluster-cluster coalescence**
- **Independent of restitution coefficient**
- **Described by inviscid Burgers equation**

Outlook

- **Velocity & spatial correlations**
- **Predictions in higher dimensions**