Granular Matter I
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Outline

• Brief intro to granular matter

• Granular jets & freely falling streams

• Dense suspension droplet break-up

• Shear thickening in suspensions

What is Granular Matter?

Lots of internal degrees of freedom \( \rightarrow \) integrate / average / ignore

Action happens at interfaces & contacts: local, short-ranged

Overall, aggregate behavior can be strikingly different from that of individual grain

1960s – 80s: granular systems = percolating thin metal films (M-I or I-I transitions)

1990s – today: granular systems = aggregates of macroscopic particles; but also co-polymer domain patterns, perhaps fluctuating high % quidles, and more recently (again) arrays of nanoparticles

(Macroscopic) Granular matter

Solid-, liquid-, gas-like states
\( \Box \) and transitions between them

Behavior far from equilibrium

• non-Brownian (kT irrelevant, mgd huge)

• friction and dissipation during collisions (cannot easily relax)

• free volume important (jamming, dilation)
Unique consequences for granular matter

- Typically stuck in amorphous metastable state once energy is removed
- Inherently heterogeneous structure (not just in static case but even if driven)
- Friction: non-linear, due to surfaces plus particle geometry
- Dilation: can work like positive feedback that localizes shear (shear band formation, avalanching)
- Time scales $10^6 \ldots 10^9$ s, length scales nm $\ldots >10$ particle diameters;
  - Issues: How to separate relevant scales? How to perform averages?

Inhomogeneous force paths
- Characteristic probability distribution of forces, $P(f)$

Jamming
- Geometrical confinement such that relative particle movement is suppressed and a yield stress develops

Hull is flexible!
- Rigidity comes from increasing packing density by just a few %
Jamming Phase Diagram

3 parameters determine jamming:
- Temperature
- Available free volume per particle (i.e., packing density)
- Shear stress

Sid Nagel (Chicago), Andrea Liu (UPenn)

Use jamming to increase # DoF of actuators


Currently Hot Research Topics for Granular Materials
(my obviously biased list)

- Effect of particle shape on properties of jammed state
- Jamming/unjamming transition under shear, away from point I
- Nature of the granular fluid state (incl. interactions of particles with interstitial medium)
- Effect of attractive interactions (cohesion, "wet" granular material)

These lectures: look at examples for items 2-4
- What keeps the jet so collimated? OR: How does a collectively liquid-like state emerge from a bunch of macroscopic, individually solid particles that interact via short-range (contact) forces?

- For dry, freely flowing grains, how does the emerging granular liquid differ from ordinary liquids?

- What can we learn about local grain-grain interactions from analyzing the structures formed by granular liquids?

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**Granular Jets**

- Fine loose powder (<200μm)
- Impact velocity 0-10m/s


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**Looking inside the bed material....**

- 1" steel sphere
- Penetrating bed at ~3m/s
- Stitch together 14 independent recordings

High-speed radiographs, >5000 full frames per second
Collaboration with CARS at Argonne National Lab’s Advanced Photon Source

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**Track Bed Density Changes Quantitatively**

- Atmospheric
- 3 Torr
During the time scale of impact, interstitial gas is effectively trapped:

- Presence of gas opposes changes in packing fraction
- Loose packing with air behaves almost like incompressible fluid

\[
k = \frac{d^2(1 - \phi)^2}{180\phi^2} \\
D = \frac{k}{\eta(1 - \phi)}
\]

Fine-grained, loosely-packed bed
- \(d = 50\mu m\)
- \(\phi = 0.50\ldots0.58\)
- \(D = 400\ldots850\text{ cm}^2/\text{s} \at 1014\text{ Pa}\)

\(t_0 = L/D \approx 90\ldots180\text{ ms} = \) time to diffuse depth of bed

\(t_{	ext{max}} = 60\text{ ms} = \) time for sphere to travel through bed

\(t_{	ext{rise}} = 30\text{ ms} = \) time for bed to rise

What about dense packing?

**Sphere Impact Into Dense Bed**

\(\phi = 0.6\)

- atmospheric
- 3 Torr

**Freely falling granular stream**

50 micron glass spheres flow through funnel (5mm nozzle diam.)

Mathias Milbich, PRE 2006
Track evolution of stream in co-moving frame

- vary air pressure
  - 101 kPa to 0.05 kPa (factor of 6 in drag force)
- grain material:
  - glass 50µm to 310 µm
  - copper 100µm
  - coated glass (hydrophobic, silver, ...)

Air Drag

- Increased air drag has little effect on cluster formation
- Instead, rips clusters apart
- Can use drag to estimate inter-particle forces:
  - grains ripped off when air drag ~ cohesion
  - gives nN cohesive forces

Electrostatics

- E-field 10⁴ - 10¹ V/m across stream (parallel plate capacitor)
- Stream = neutral, charge distribution P(σ) nearly symmetric
- Maximum attractive force only 0.1 nN: too small
- Clusters also observed with conductive, silver coated glass spheres

Inelasticity?

\[ \varepsilon = \frac{v'}{v} \leq 1 \]

- d = 130µm copper
- Otherwise identical conditions (vacuum, 4 mm nozzle)
- Lower coefficient of restitution
  - e ~ 0.90 vs. ~0.97 for glass
- Clustering not simply due to inelasticity alone
Particle Agglomeration Mechanisms


Controlling Clustering by Altering Nanoscale Surface Roughness

Clean Glass

with Aerosil

5 µm

5 µm

Estimate energy loss from AFM data:

\[ W_{\text{coh}} = \int F' (\zeta) d\zeta \sim 10^{-15} \text{ J} \]

Clustering due to ‘Sticky’ Collisions

Add small (~ 10 nm) layer of oil

increasing cohesion

Scenario for Cluster Formation

Near nozzle: large velocity fluctuations ~ 1 cm/s (high granular temperature) particles collide, lose energy to inelasticity

\[ \frac{1}{2} m (\delta u)^2 \gg W_{\text{coh}} \]

Further downstream: temperature comparable to \( W_{\text{coh}} \) particles collide, stick

\[ \frac{1}{2} m (\delta u)^2 \sim W_{\text{coh}} \]

Clustering due to attractive potential

Analog to surface tension in normal liquids

\[ \gamma \sim W_{\text{coh}} / d^2 \sim 0.1 \mu \text{N/m} \]

Granular Temperature ~ 3 m/s

"Hot" near nozzle
collisions, grain stretching short-ranged attractions

"cooler"

3D MD simulations with ~100,000 particles, using Isaca PFCSd
Scott Waltukaitis, Helge Grütjen
Model: Linear contact + Hysteretic Attractive Force

- **Repulsive part:** linear contact model for normal & tangential components
- **Attractive part:** Hysteretic "step" force commonly used to model liquid bridges
- PFC3d, ~100,000 particles


Granular temperature not a sensitive diagnostic

No Cohesion: Particles Always Drift Apart in velocity gradient (gravitational stretching)

Force-Inelasticity Phase Space

Wide range of behaviors:
- Spraying
- Clustering
- Drop-forming
- Dripping

No droplets for $F_{out} = 0$

Both $e$ & $F_{coh}$ affect cluster size, necking behavior

Diagnostic of grain-grain attractions

Use like molecular beams?
**Neck Width Scaling During Breakup**

Different from thermal exponent (0.418), instead > 1/2

Consistent with

\[ w_{\text{min}} = \left[ \frac{4}{3} (l_0 - l)^{1.5} \right] \]

Same scaling as inviscid liquid break up (i.e. water in air)

In liquid case

\[ a = 1.4 \left( \frac{\mu}{\rho} \right) \]

**Effective Surface Tension**

\( \approx 1 \mu \text{N/m} \)

4 orders of magnitude below standard liquids

**To sum up Jets & Streams:**

- Tiny attractive interactions \((N, nm)\) drive clustering and droplet formation...same as for molecules, but here acting between macroscopic constituents, therefore often masked by gravity

- Corresponding effective surface tension 4-5 orders lower than water \(\Rightarrow\) ultra-low regime not reachable with ordinary liquids under ambient conditions; break-up neither Rayleigh-Plateau nor thermal: where do aspect ratio, neck shape and 2/3 power law come from?

- Freely Falling Granular Streams: can probe wide range of behaviors from gas to liquid to plastically deforming solid; granular analog of molecular beam to probe subtle grain-grain interactions in situ

Next: add water...
4.106 ± 4, 0, 4.1060

40, 60, 80, 100

q = (-1 ± 10) x 10^4

e = (1.58 ± 0.04) x 10^6

Charge distribution for 265 ± 15 m ZrO2

Thanks to:

Matthias Middus  first work on clustering freely falling streams
John Royer  jets from impact, freely falling streams
Scott Wiltukaitis  stream simulations, streams as probes of granular self-charging
Marc Minin  break-up of suspensions
Sid Nagel, Wendy Zhang

http://j.uchicago.edu/~jaeger/group/

More Permeable Bed

scale up by factor ~20

drop shot put from several stories into 55 gallon drum filled with 1mm diameter particles

Tracking the Bed Response

Bed particle flow:

Interaction of interstitial gas with grains ➔ granular bed behaves like incompressible liquid

du/dt = ±0.1
Force varied
$F = 5 \text{mN}$

Contact Number

$F_{coh} = 100 \text{ nN}$
$\epsilon = 0.80$

$d_s = 200 \mu \text{m}$
$D_N = 3.0 \text{ mm}$

$F_{coh} = 1000 \text{ nN}$
$\epsilon = 0.40$

Collide and Capture

Parabolic Orbits

Particle Type & Interactions Important
(1) Thin films are 250 nm thick, 200 ml of 10 Molar NaOH fell from an
apertured nozzle (0.15 mm) which was held at an angle of 45 degrees to the
substrate. (2) Gases were the same as the control and the 130 μm
horizontal white strips (first white strips in a reflectance mode) mean of
drained portions is zero (within error) and sign is +1.2 million electrons (6). Pressure was 0.53 kPa
Fluidized for 0.3 hour beforehand (6)
All from single batch.
\[ \frac{1}{2} m(v_c)^2 = W_{coh} \]

\[ W_{coh} = \int F_{coh}(x) dx \]

Near nozzle, fluctuations ~10mm/s quench T by factor 10 in 1/4s
Effective surface tension: \( \gamma \approx \frac{\varepsilon^2}{d^2} \sim \frac{W_{coh}}{d^2} = 0.1\mu\text{m} \)

\( H_2O: 70\mu\text{m} \)

AFM cannot provide complete picture!

Coarse sand: \( F_{coh} \) nearly identical to glass spheres, but clustering very different
AFM: only simple, head on collisions, measure max. force
Need total energy loss from cohesive & dissipative forces (including sliding and rolling motion)

Simplify: \( \text{sticky} \) collisions where head-on collisions dominate

Granular Fluids

Remarkable similarities to conventional liquids.

- How does liquid-like behavior emerge for an aggregate of grains?
- What features come simply from large number of rapid collisions?
- When are attractive interactions between grains important?

other possible influences

Role of surrounding gas?
(1) air vs. He;
(2) small particles & small # density
(4) simulations, w/o gas (T. Pöschel)
more possibilities...

Inelastic nature of grain-grain collisions
(1) copper particles;
(2) rough sand;
(3) simulations (T. Pöschel).

Imaging the bed interior at Argonne’s APS
John Roger et al., Nature Physics (105); PRL (107);
PRE (138)

Impact Depth vs. Bed Density
(for fixed $v_f$)

Momentum Conservation $\rightarrow$ Cone Angle

Permeability Determines Bed Response

Patterns at Different Pressures

Different from ordinary liquids:
Shape of fingers & pressure dependence
Roths, Thoms, Mitare-Wilkinson, and Steen, PRE '96
Near Yielding: Fractal Dimension as Expected for Zero Surface Tension Limit

Granular Analog to Water Bell

- Dense plug of grains
- Compressed air \( \Delta P \)
- Vary:
  1. Granular material: Glass beads, Copper beads.
  2. Bead diameter, \( d = 50 \mu m \) to 2mm.
  3. Impact velocity: \( U_i = 1.3 m/s \) to 16m/s.
  4. Target size: \( D_{target} = 0.13 cm \) to 2.8cm.
- Fix \( D_p = 0.75 cm \), \( L = 40 cm \) and \( w = 2.54 cm \).

Granular Liquids: “Zero” Surface Tension?

- Granular Hele-Shaw system:
  - patterns and cusps as predicted for vanishing surface tension
- Granular jets impacting targets:
  - quantitative agreement with high-We results for water
  - with increasing particle \# density, transition from particulate to liquid-like behavior (thin sheets, “water bells”)
  - “liquid” produced by brief interval of rapid collisions right in front of target; sheet is “finger print” of this interval, but no longer a liquid (density too low)

Cone angle depends on Weber number: \( We = \frac{\rho U L_{jet}^2}{\gamma} \)

- \( \rho \) density of water; \( U_L \) velocity of jet; \( \gamma \) surface tension of water

Data for water C. Clanet, J. Fluid Mech. 430, 111 (2001)

Viscous Fingering at Liquid Interfaces

- Fractal patterns & DLA-like growth
- Finite time singularities

Granular Fingering in a Hele-Shaw Cell

- Xiang Cheng et al., Nature Physics [08]
- Compressed air
- Low pressure (\( \Delta P = 0.24 \) atm)
- Sand diameter, \( d_s = 150 \mu m \), gas is \( L = 2.5 \) mm
- Video frame rate 250 frames/s

For the zero-surface-tension limit, theory predicts:

In ordinary liquids: zero tension limit approximated by increasing the flow rate

What happens for dry granular material forced by gas (air)?
Singular Dynamics for Vanishing Surface Tension: Cusps in Granular Fingering

Near threshold pressure for yielding:

Scale bars: 0.5mm

Cusps different from any finger structure found in normal liquids, where surface tension rounds off sharp features.

Liquid fingers:

Cusp Shape Scaling

Shape predicted for Laplacian growth in limit of zero surface tension:

\[ y = A(x - x_0)^{1/2} \]

Consistent with profile of finger tips as they become sharper, i.e., near the finite time singularity.

Water Bells

How about a fluid with vanishing surface tension?

Water bell formed.

water jet from faucet hitting a spherical target

Cone Angle

From (a) to (h): \( \theta_{\text{cone}} / \theta_{\text{tip}} = 4.0, 3.7, 2.0, 1.6, 1.3, 1.2, 1.0 \) and 0.85.
Grains: Cone Angle Independent of Velocity

Shoot jet upward to avoid asymmetry due to gravity.

Black grains
Red: water

We = \( \rho U_0 D_\text{p}^2 / \gamma \)

Large We limit: grains behave like \( \text{H}_2\text{O} \)

For non-cohesive granular material, \( \gamma \to 0 \) ⇒ We = \( \rho U_0 D_\text{p}^2 / \gamma \), independent of \( U_0 \)

From Particles to Liquids

How does liquid-like behavior emerge from the particle nature of a granular material?

10μm diam. glass beads striking aluminum target

What does it take to create a liquid?

- Sufficiently many / strong interactions
- Attractions? ⇔ Confinement?

\[
\text{cold stream} \\
\downarrow \\
\text{"hot" collision volume in front of target (~ 1 particle deep; >30 particles across)} \\
\downarrow \\
\text{cold gas}
\]