Intensity Correlation Tricks

Adam Abate, Dr. Ranjini Bandyopadhyay,
Prof. Paul Dixon, Alex Gitting, Dr. Hiroaki Katsuragi,
Pierre-Anthony Lemieux, Rajesh Ojha
Douglas J. DURIAN
UCLA & University of Pennsylvania

- Good-old photon-correlation spectroscopy

  not applicable for unsteady or evolving dynamics

  - eg nearly-jammed matter

- Our extensions:

  Higher-order intensity correlations

  Speckle-Visibility Spectroscopy (SVS)

DURIAN

DLS/PCS Review

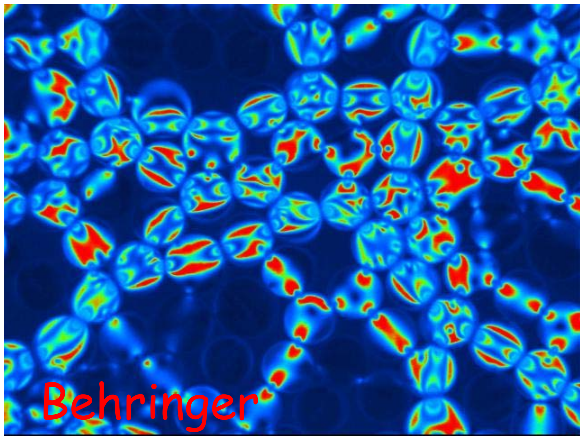
- Conventional intensity-correlation spectroscopy
  [1] Measure $\langle I(0)I(\tau)\rangle$ for ~one speckle
  [2] Equate to $\langle I\rangle^2$ $[\, 1 + \beta|\gamma(\tau)|^2 \,]$ {"Siegert relation"}
  [3] Deduce motion of scattering sites from E-field autocorrelation, $\gamma(\tau)$ = F.T. of power spectrum

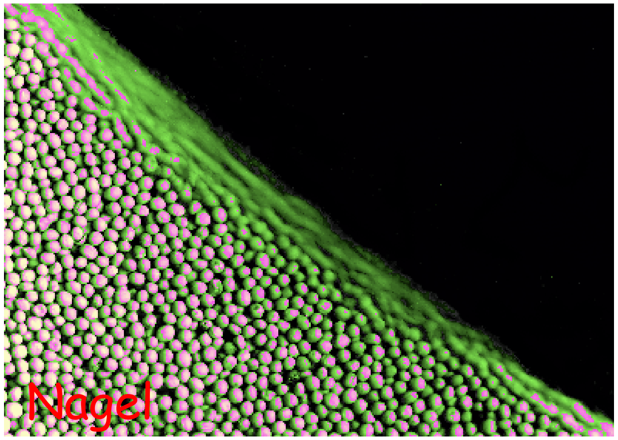
- Pitfalls
  - Unsteadiness (eg evolution, intermittency, periodicity, temperature fluctuations, etc) will be misinterpreted as slow modes in scattering site dynamics
  - NonGaussian electric field statistics (eg from number fluctuations or correlated motion) invalidate Siegert
    - these issues often arise in the most interesting systems:
      - Phase separation, glassy behavior, and…

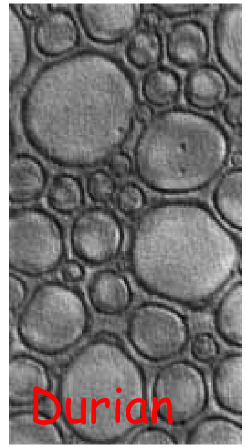
DURIAN

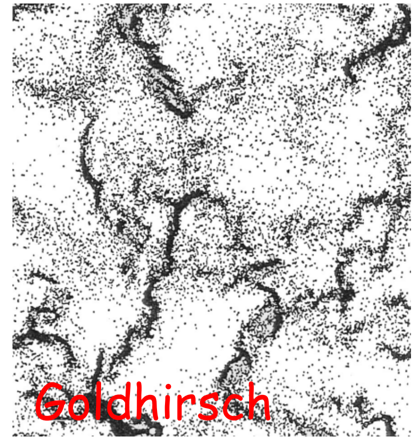
Granular Matter

- Grains, bubbles, colloids, cells, tectonic plates...









hard problems = new physics!
few engineering guidelines!

- No basis for usual intuition

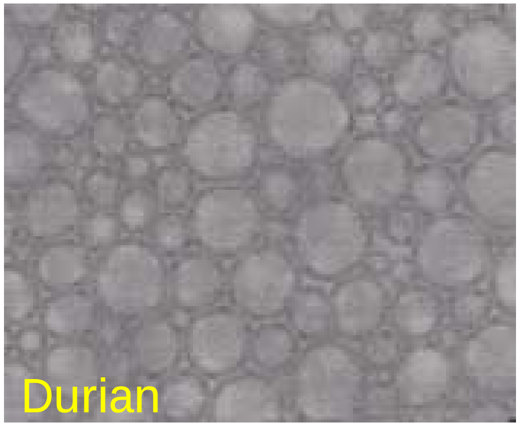
  disordered / heterogeneous: *no periodicity or symmetry*

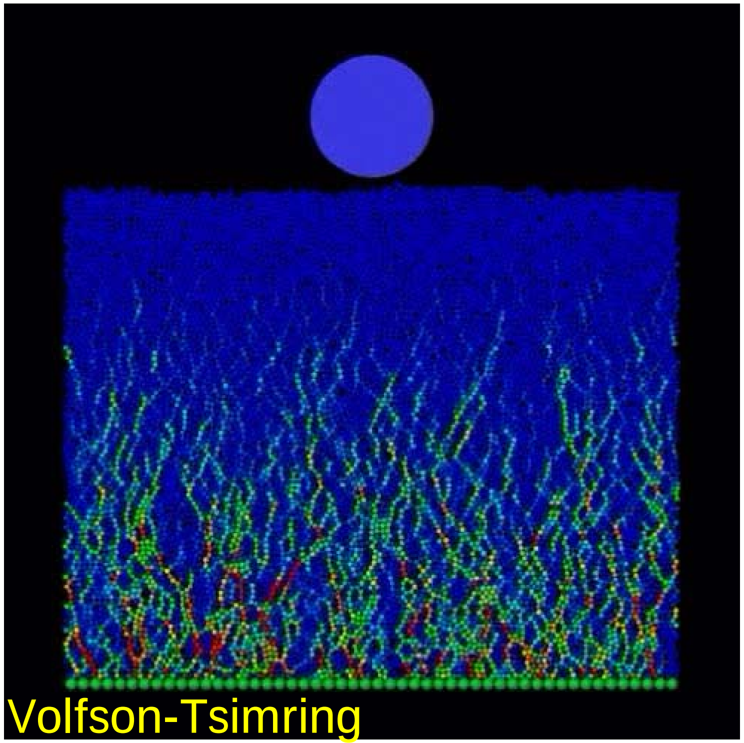
  $k_B T$<<interaction energy: *no statistical mechanics*

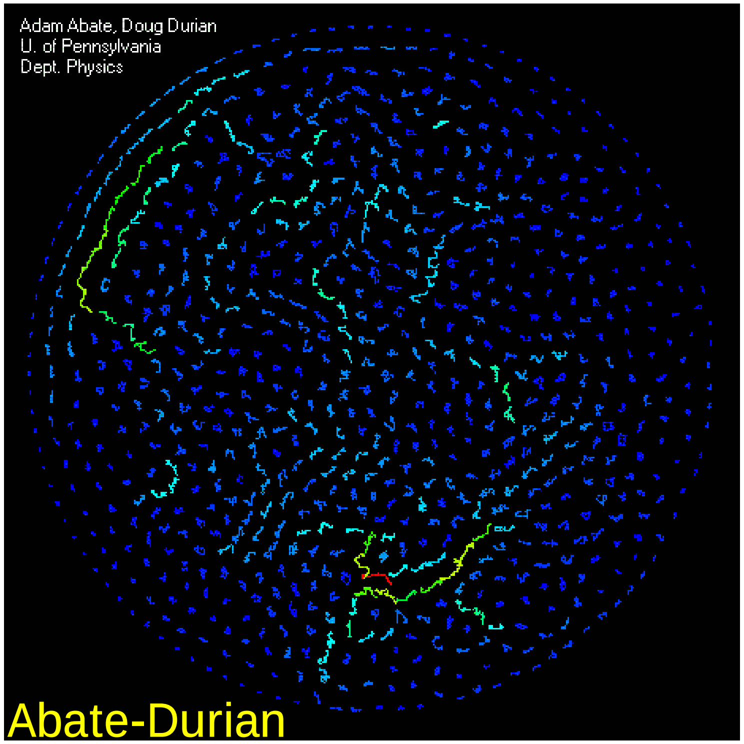
  flow beyond threshold: *no linear response*

Selected granular unsteadiness

- Avalanches in sand
- Rearrangements in foam
- Heterogeneities near jamming
- Aging near glass transition







Temporal intensity statistics

- Normalized nth-order intensity correlations:
  - I=EE* so this is really a (2n)th-order field correlation

$$g^{(n)}(\tau_1, \tau_2, \ldots, \tau_{n-1}) = \langle I(0)I(\tau_1)I(\tau_2)\ldots I(\tau_{n-1})\rangle / \langle I\rangle^n$$

- Experiment:
  - $g^{(2)}(\tau)$ is routinely measured with a digital correlator
  - We have used a home-build digital delay device (T) and a commercial correlator (τ) to measure:
    - $g^{(3)}(T,\tau) = \langle I(0)I(T)I(\tau)\rangle / \langle I\rangle^3$
    - $g^{(4)}(T,\tau,\tau+T) = \langle I(0)I(T)\, I(\tau)I(\tau+T)\rangle / \langle I\rangle^4$

- Theory: analyze in terms of $\gamma(\tau)=\langle E(0)E^*(\tau)\rangle / \langle EE^*\rangle$
  - The Fourier transform of the power spectrum; if spectrum is symmetric then $\gamma(\tau)=|\gamma(\tau)|\mathrm{Exp}[i\omega_o\tau]$, ω_o=central frequency

DURIAN

Ordinary systems

- If there are many independent scattering sites:

  E is complex Gaussian random variable

  - phasor diagram for total E at some point is a random walk
  - "speckle" pattern with spatial and temporal correlation lengths

  $g^{(n)}(\tau_1,\tau_2,\ldots)$ can be computed in terms of $\gamma(\tau)$:

  - sum of products of all possible two-time field correlations
  - spatially average over detector area

    – For example $\langle I(0)I(t)\rangle$ is

$$\langle E_0E_0^*E_1E_1^*\rangle = \langle E_0E_0^*\rangle\langle E_1E_1^*\rangle + \langle E_0E_1\rangle\langle E_0^*E_1^*\rangle + \langle E_0E_1^*\rangle\langle E_1E_0^*\rangle$$

$$= \langle I\rangle^2 + 0 + \langle I\rangle^2\,\gamma(\tau)\gamma(\tau)^*$$

which, after averaging over $1/\beta$ speckles, is $\langle I\rangle^2\,[\,1 + \beta|\gamma(\tau)|^2\,]$

Correlations for Gaussian fields

Lemieux and DJD, JOSA-A (1999)

$$g^{(2)}(\tau_1) = 1 + \beta|\gamma_{01}|^2,$$

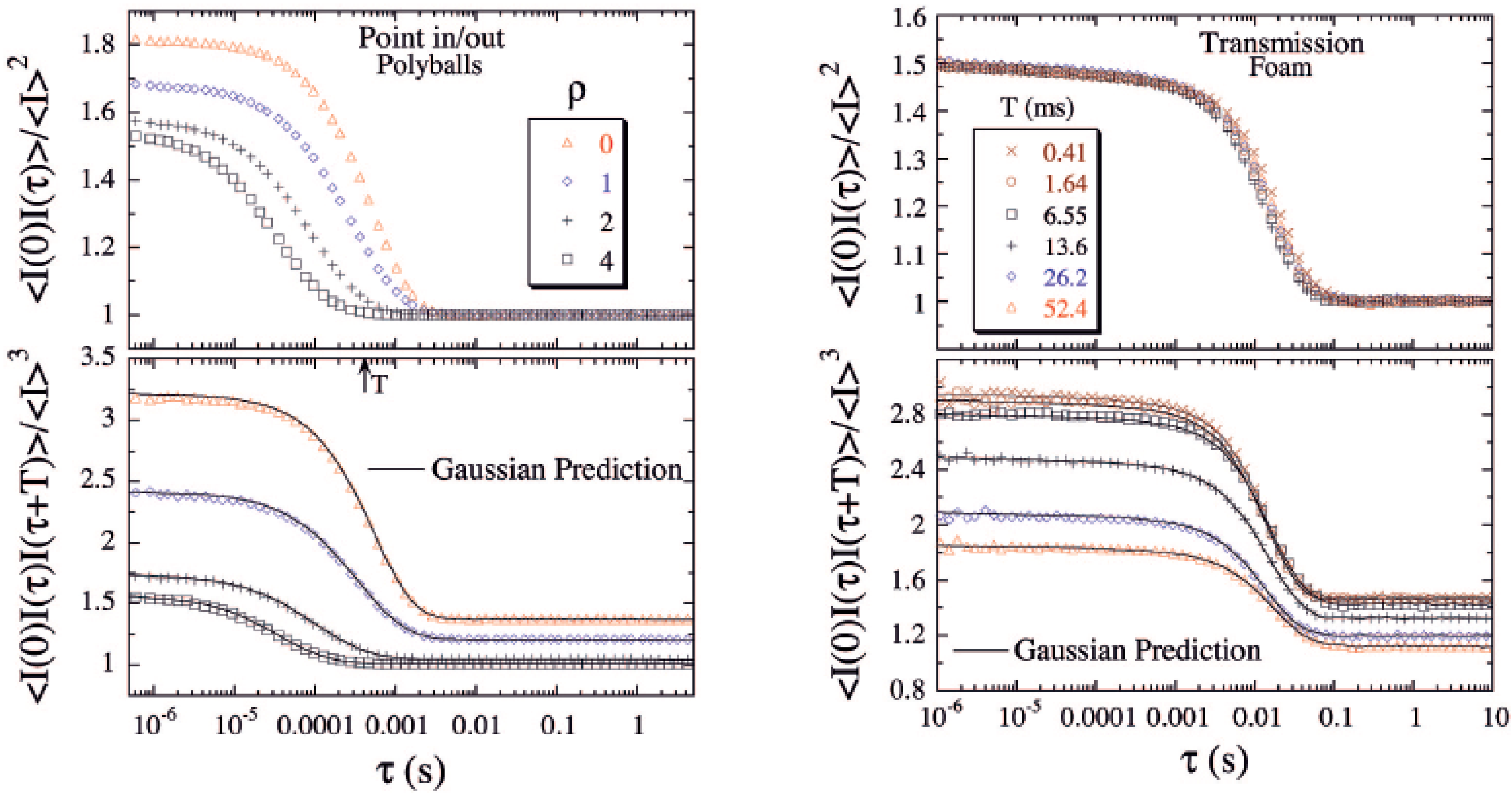
$$g^{(3)}(\tau_1, \tau_2) = 1 + \beta(|\gamma_{01}|^2 + |\gamma_{12}|^2 + |\gamma_{20}|^2) + 2\beta^2 \operatorname{Re}(\gamma_{01}\gamma_{12}\gamma_{20}),$$

$$\begin{aligned} g^{(4)}(\tau_1, \tau_2, \tau_3) = 1 &+ \beta(|\gamma_{01}|^2 + |\gamma_{02}|^2 + |\gamma_{03}|^2 + |\gamma_{12}|^2 \\ &+ |\gamma_{13}|^2 + |\gamma_{23}|^2) + \beta^2(|\gamma_{01}|^2|\gamma_{23}|^2 \\ &+ |\gamma_{02}|^2|\gamma_{13}|^2 + |\gamma_{03}|^2|\gamma_{12}|^2) \\ &+ 2\beta^2 \operatorname{Re}(\gamma_{01}\gamma_{12}\gamma_{20} + \gamma_{01}\gamma_{13}\gamma_{30} \\ &+ \gamma_{02}\gamma_{23}\gamma_{30} + \gamma_{12}\gamma_{23}\gamma_{31}) \\ &+ 2\beta^3 \operatorname{Re}(\gamma_{01}\gamma_{12}\gamma_{23}\gamma_{30} + \gamma_{02}\gamma_{23}\gamma_{31}\gamma_{10} \\ &+ \gamma_{02}\gamma_{21}\gamma_{13}\gamma_{30}). \end{aligned}$$

- Subscripts denote time delay differences
- Expressions for n={3,4} are not in previous literature

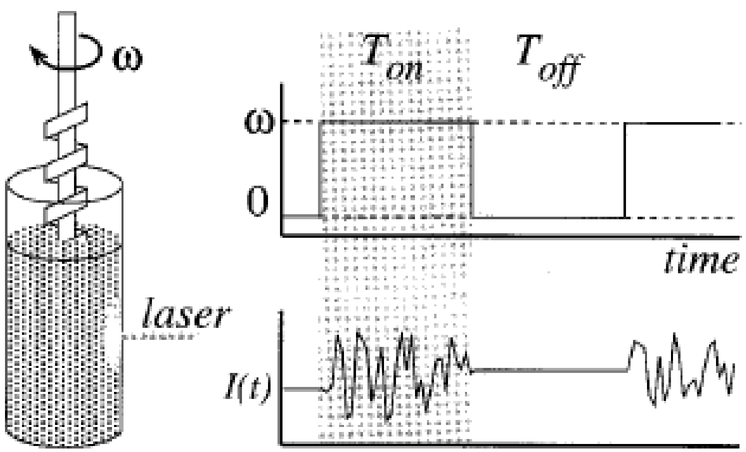
Verify health of data

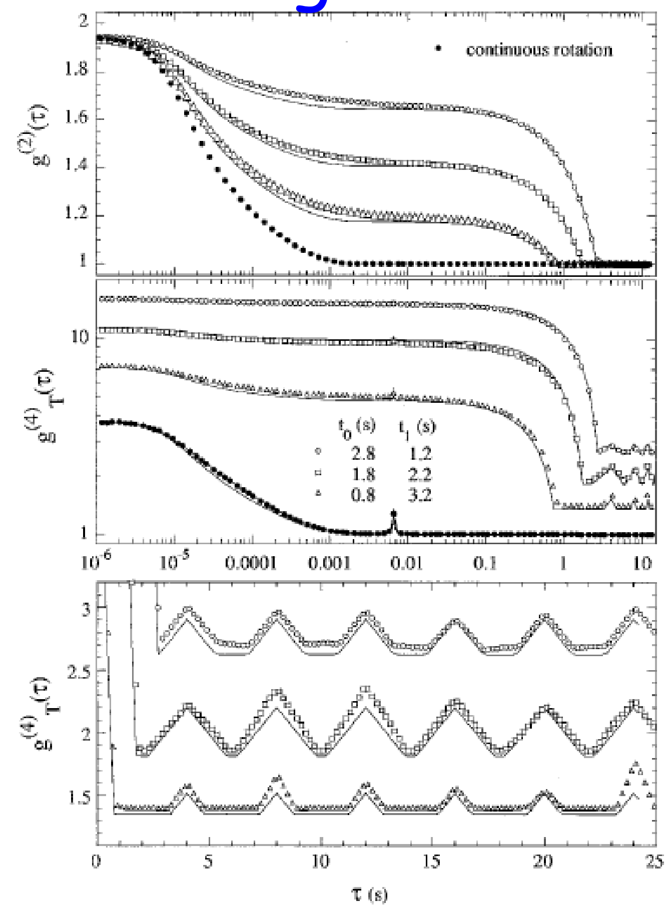
- Generate $g^{(n)}(\tau_1,\tau_2,\ldots)$ predictions from $g^{(2)}(\tau)$ data:
  - agreement means Gaussian field statistics and hence that field autocorrelation may be extracted from $g^{(2)}(\tau)$ using Siegert
  - this check ought to be routinely performed!



Failure of Gaussian predictions

- Nature of discrepancy indicates the problem:
  - eg number fluctuations, correlations in dynamics, source drift, source incoherence, static/heterodyning component,…
- Extra information is available in higher orders
  - eg intermittent flow of sand:
    (*stationary* unsteadiness)

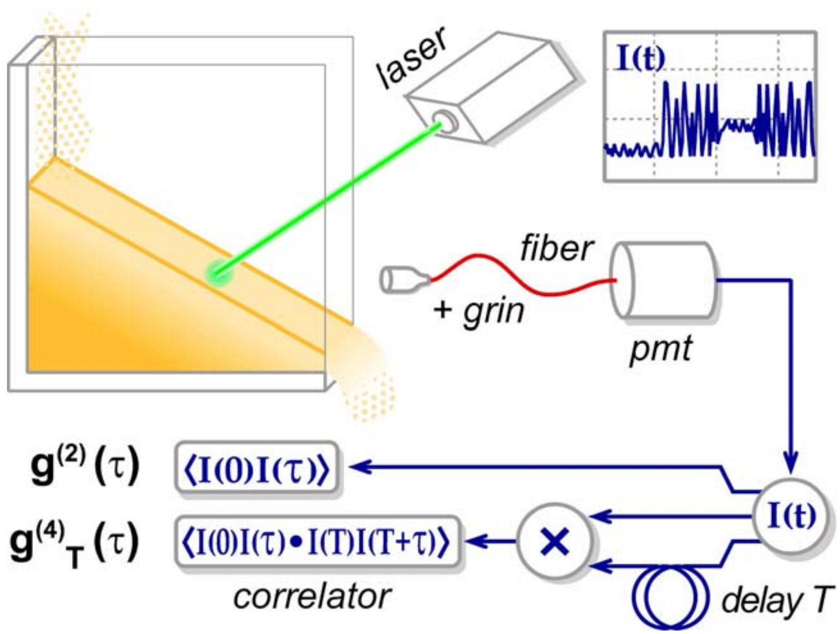


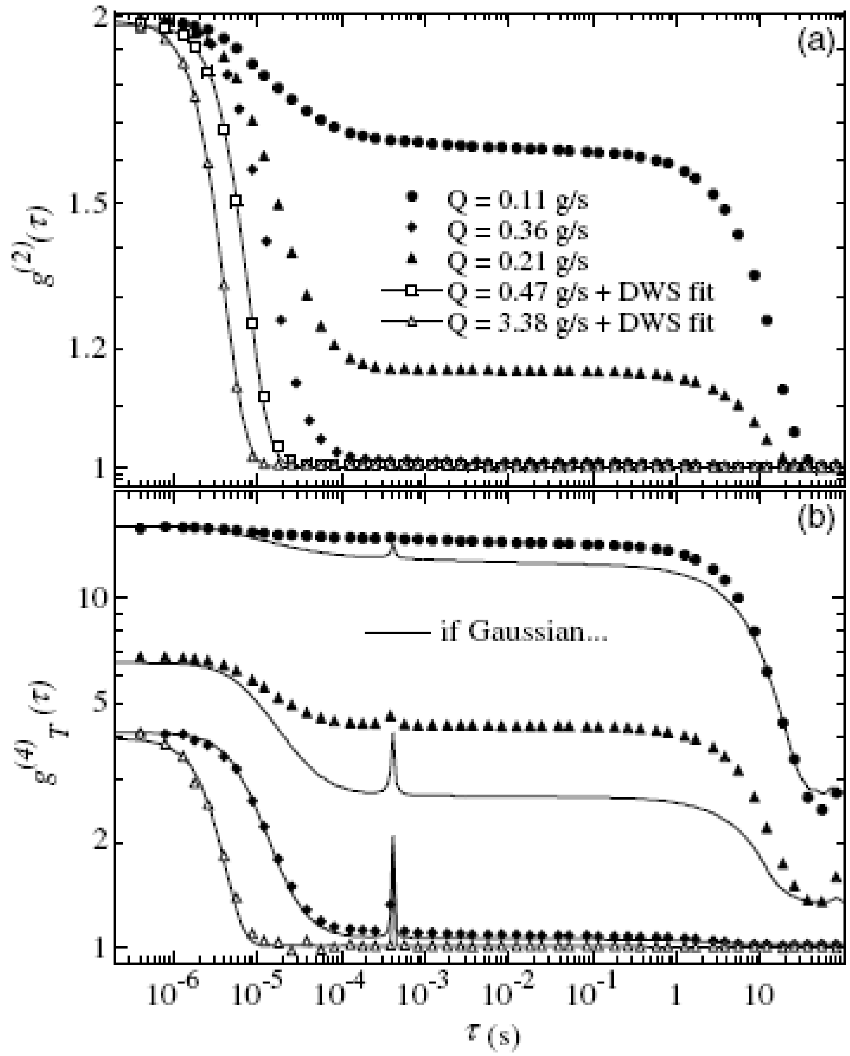


Intermittent granular avalanches

Lemieux and DJD, PRL (1999) and Appl.Opt. (2001)

- Deduce on/off times and switching prob functs

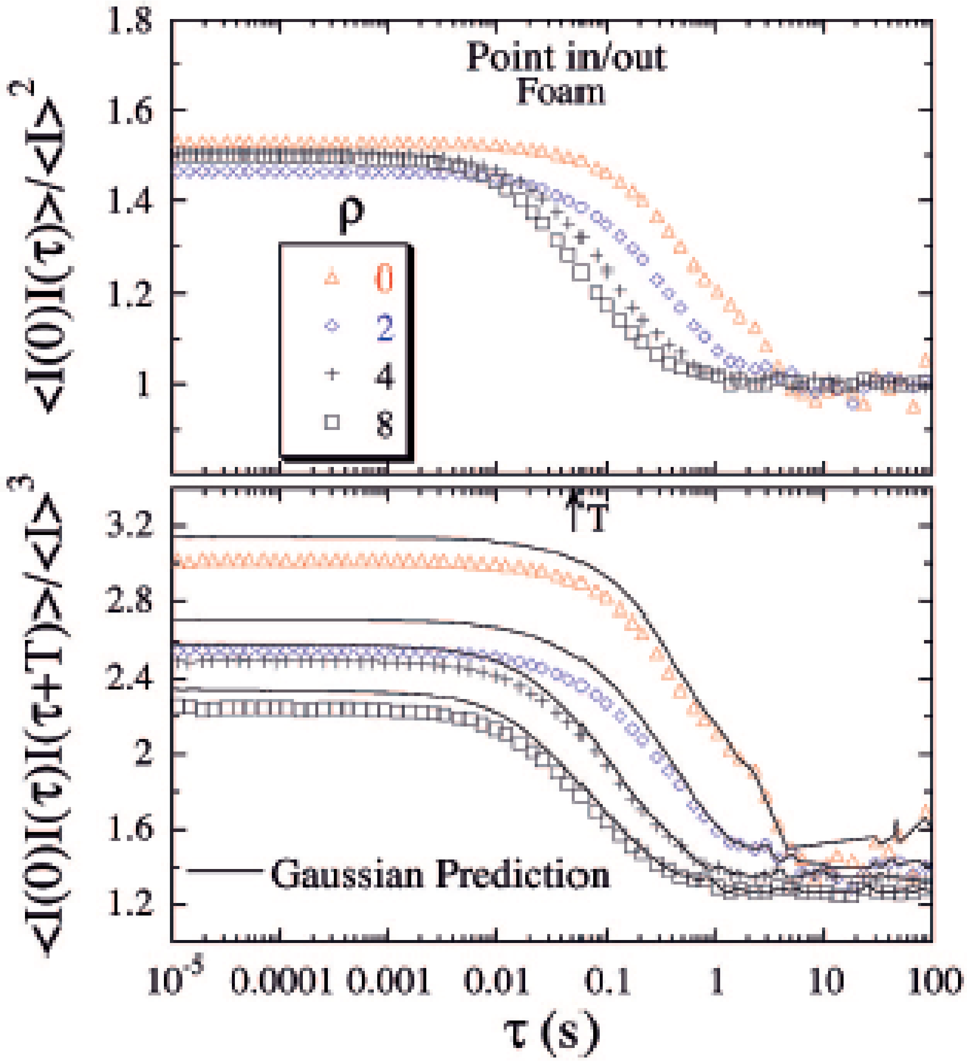




Intermittent bubble rearrangements

Gittings and DJD (preprint)

- Deduce event frequency and rearrangement size



Time-resolved DLS

- If unsteadiness in dynamics is not stationary, then a good way to capture the physics is
  $\langle I(t_o)I(t_o+\tau)\rangle$ vs τ ←→ power spectrum at age t_o
  - sequence of start-times t_o
  - <…> is ensemble average of many speckles by use of area detector like CCD camera

Wiltzius; Sillescu; Sutton; Mochrie; Weitz; Pine-Lequeux; Cipelletti

Low frame rates; extensive storage and post-processing; short evolution

- We propose an alternative…

Speckle-visibility spectroscopy

Dixon & DJD, PRL (2003); Bandyopadhyay, Gittings, Suh, Dixon & DJD, RSI (2006)

- match speckle size to pixel size
- measure the speckle pattern's visibility by variance of detected intensity levels for a single exposure of duration T:
  - speckle is less visible for faster dynamics and vice-versa

$$V_2(T) \equiv \left[\left\langle I^2 \right\rangle_T \Big/ \left\langle I \right\rangle^2 - 1\right] \Big/ \beta \;=\; \int_0^T 2\left(1 - t/T\right)\left|g_1(t)\right|^2 \mathrm{d}t/T$$

$$\rightarrow \begin{cases} 1 & \text{short exposure / slow dynamics} \\ 0 & \text{long exposure / fast dynamics} \end{cases}$$

- ***Just ONE exposure gives information on microscopic motion***
- ***OK if motion varies from exposure to exposure***
- ***Remove β by computing variance ratio $V_2(2T)/V_2(T)$***

- This opens up a world of new experiments…

DURIAN

Eg: sand shaken at 10 Hz

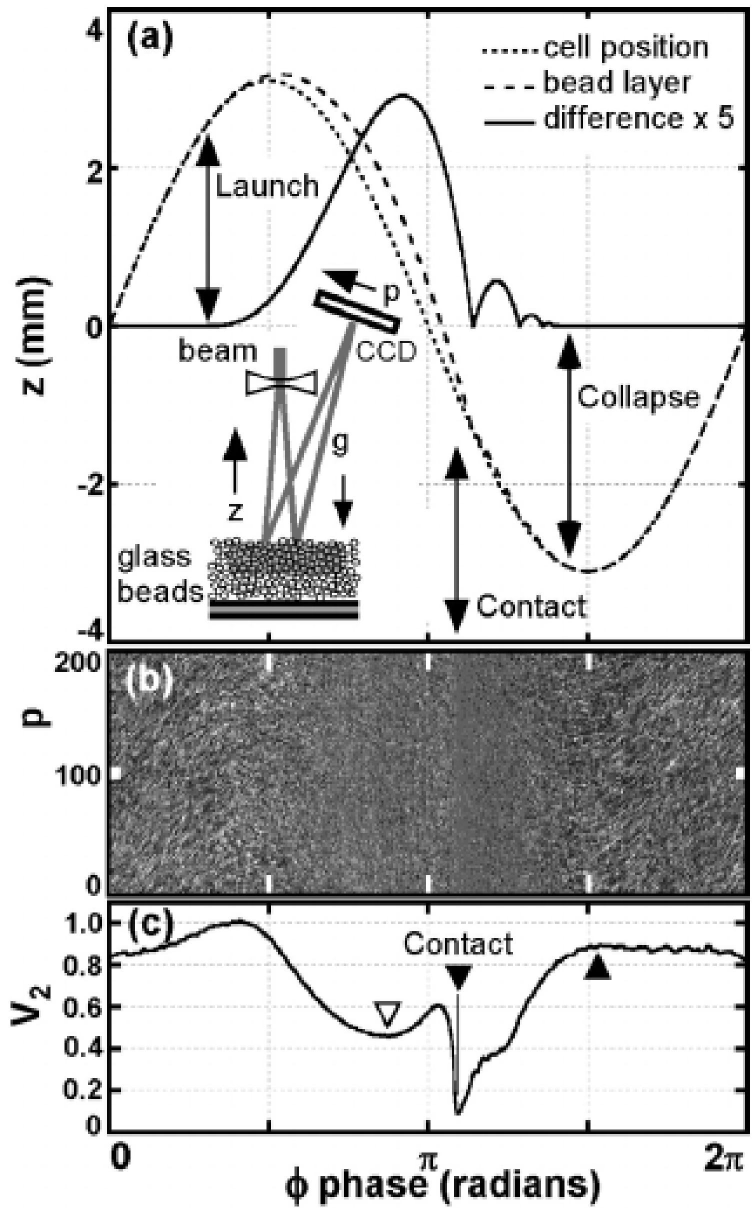
(Dixon & DJD, PRL 2003)

Source:

- NdYAG laser
- 535 nm
- 1 W

Camera:

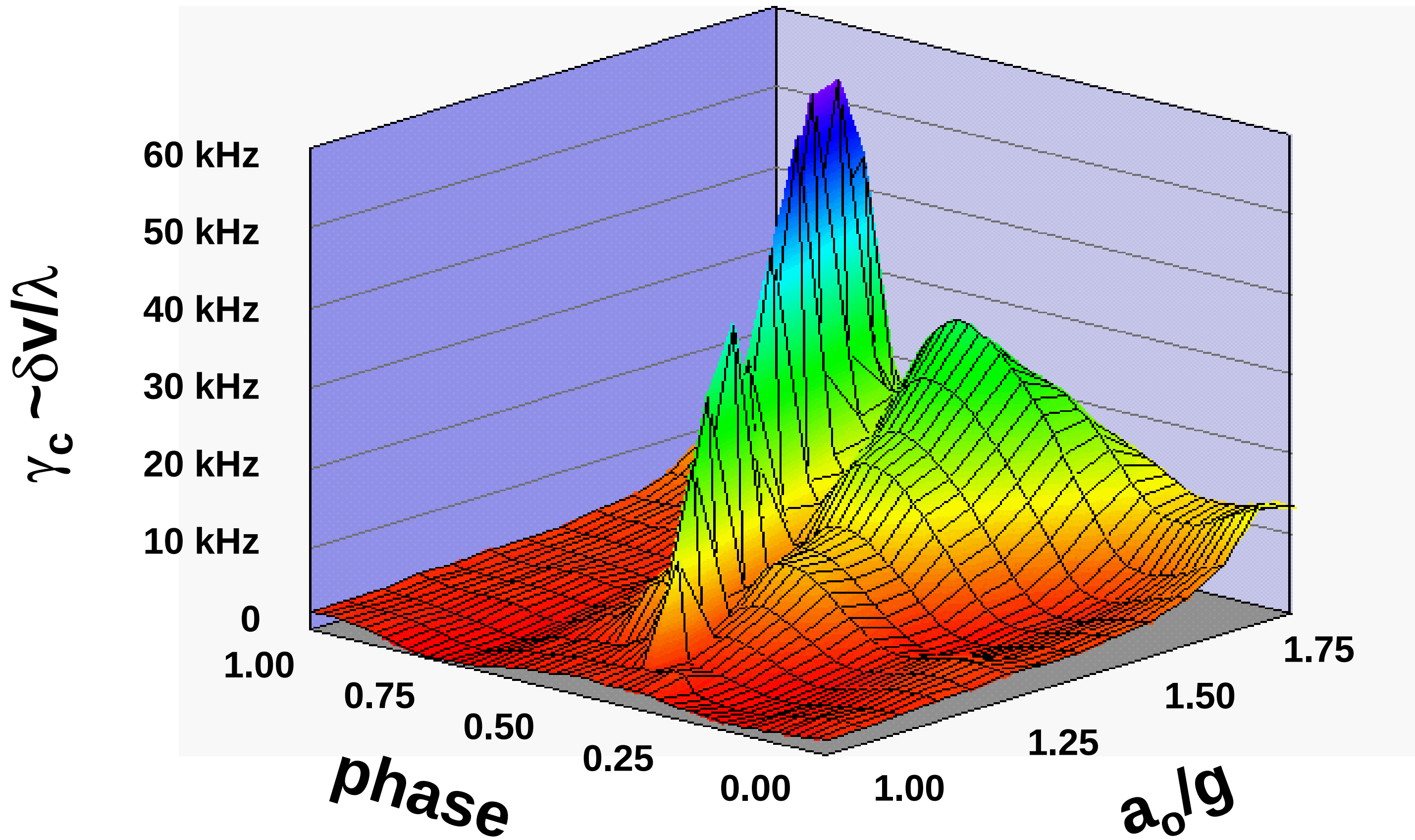
- Linescan CCD
- 1024 pixels
- 8 bits
- 58 kHz



Analysis of visibility data

- Deduce grain fluctuation speed, δv, vs phase in cycle and for different shaking amplitudes:

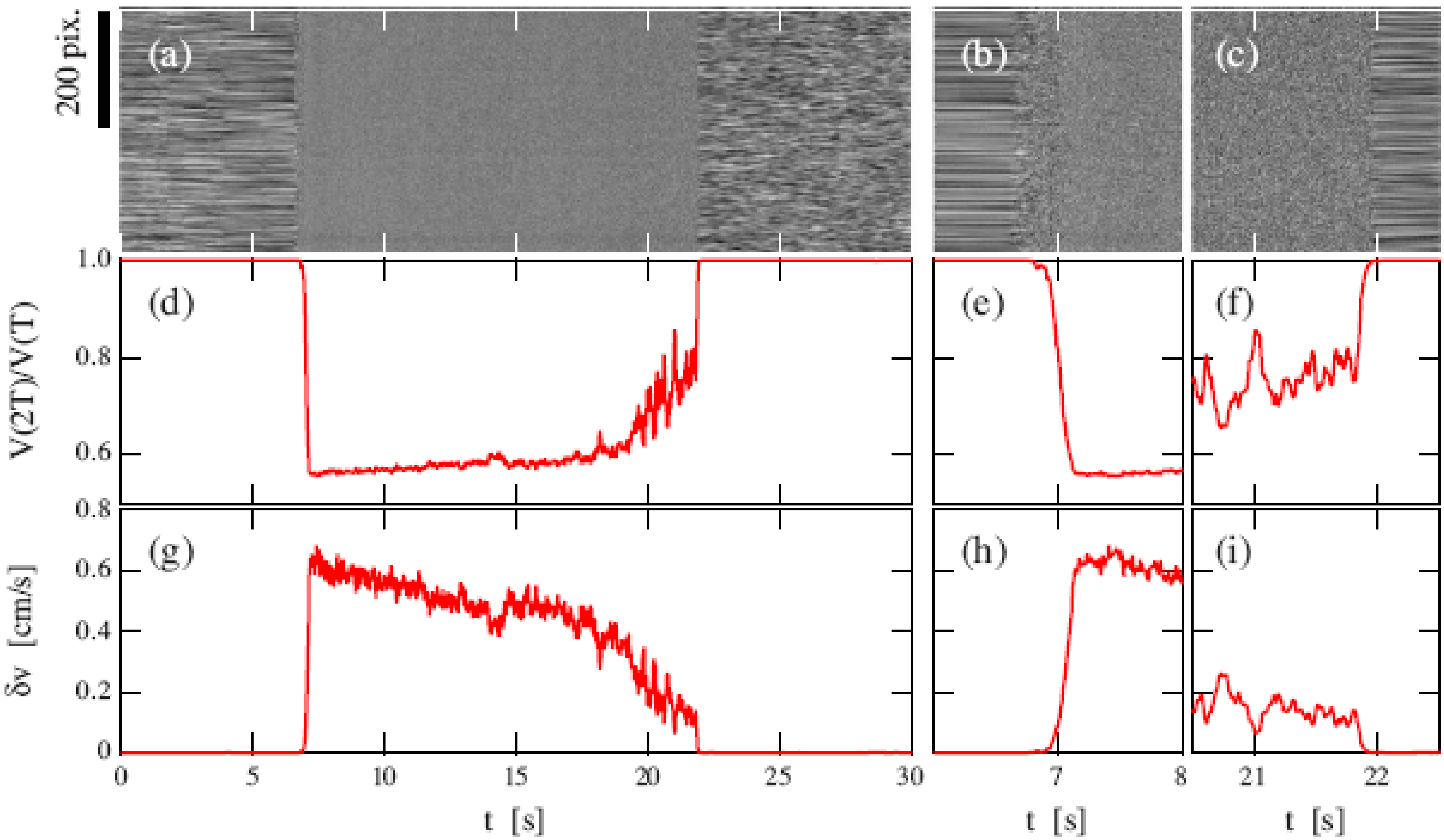
[m δv^2 / 2 is granular temperature]



Eg: SVS for granular avalanches

- Flow down a heap

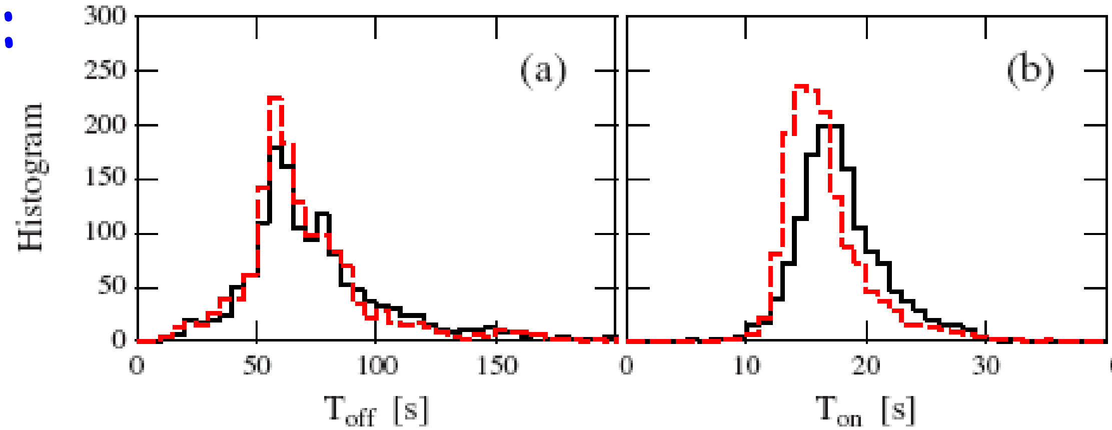
Abate, Katsuragi, & DJD, PRE (2007)



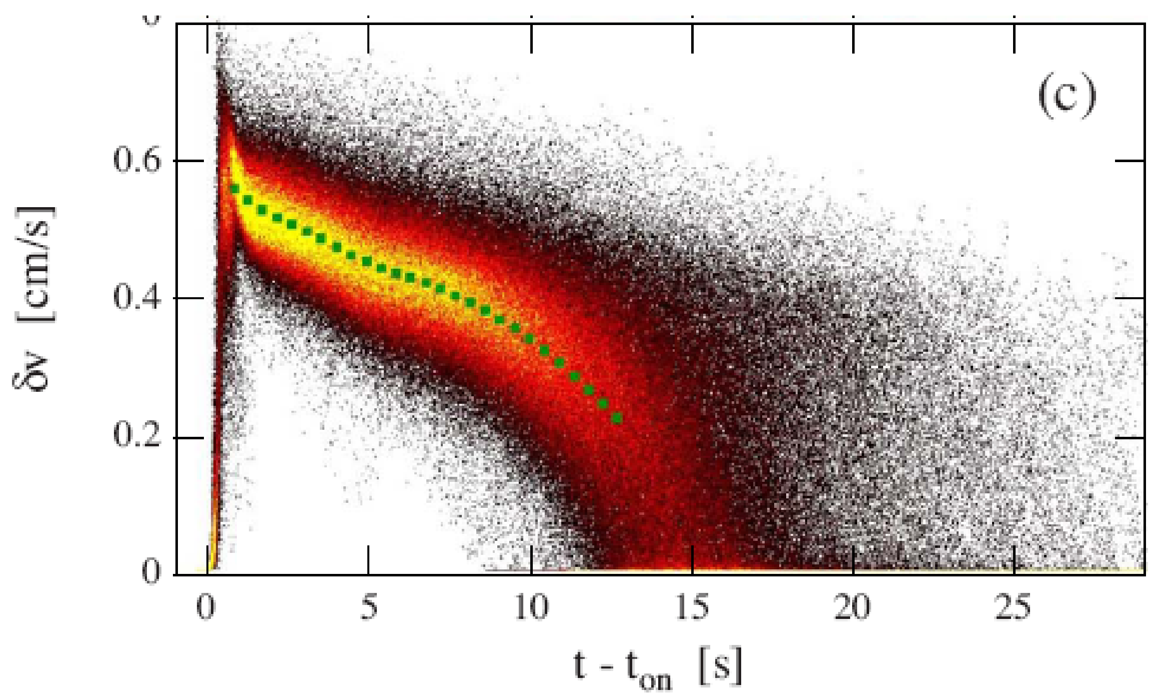
DURIAN

Deduce avalanche statistics

- On/off durations:

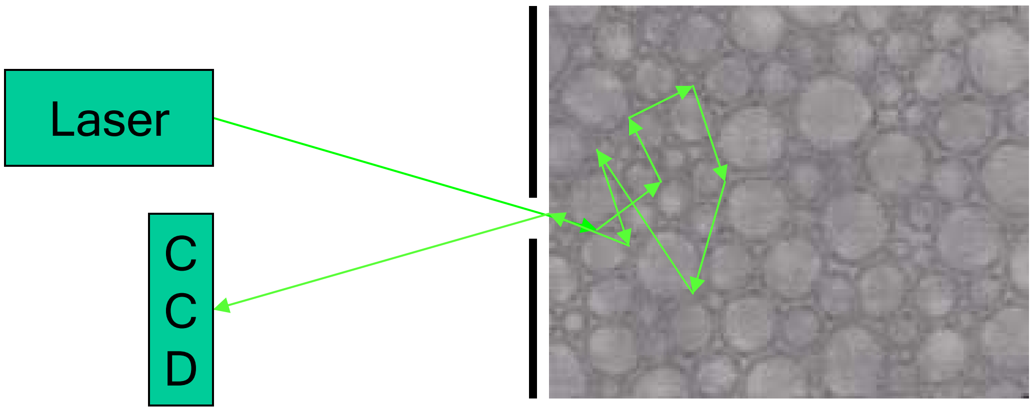


- Speeds vs time:



SVS on Foam

- Illuminate / detect through 1 mm aperture

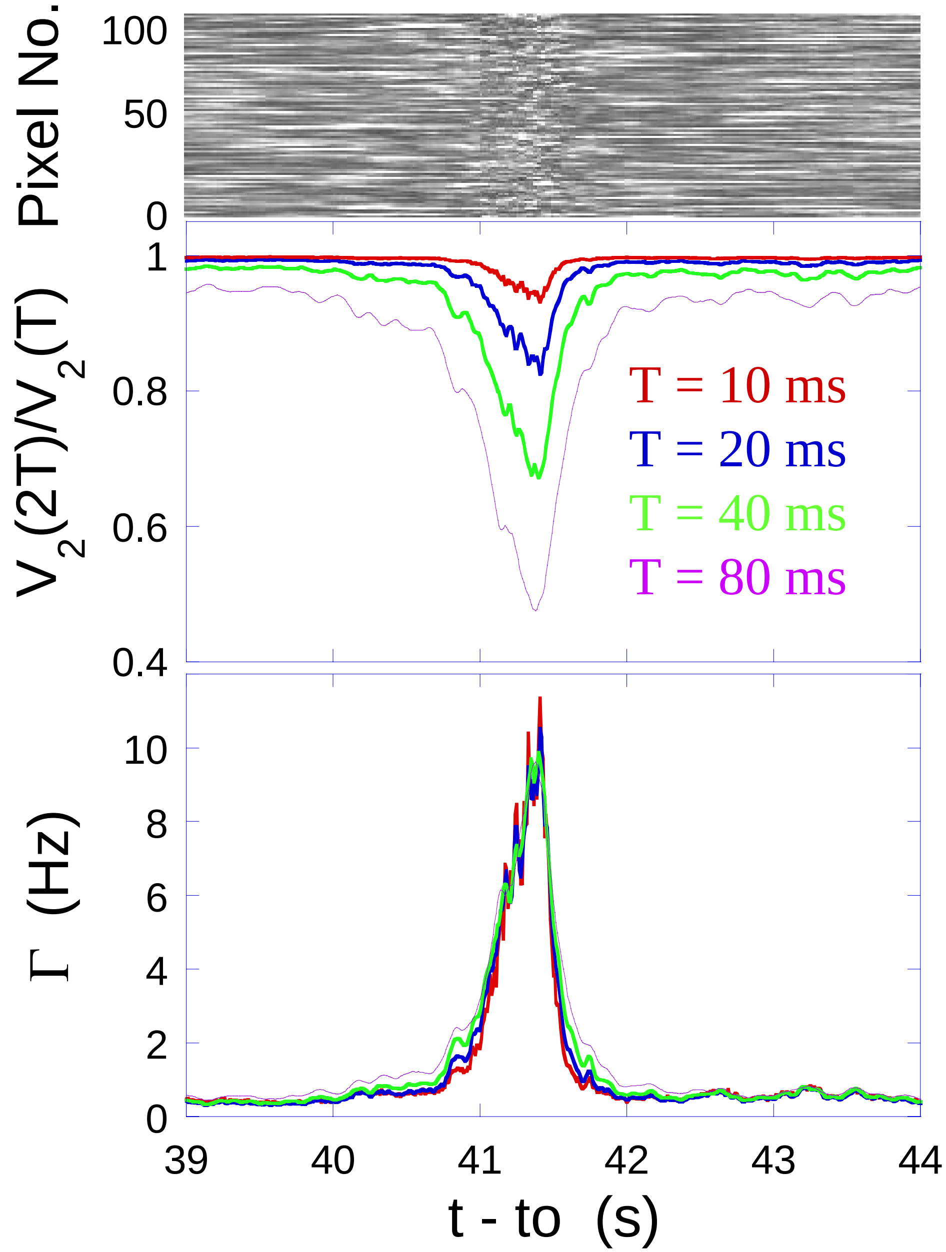


A typical rearrangement event

- Raw image data:
- Variances vs time:
- Linewidths:

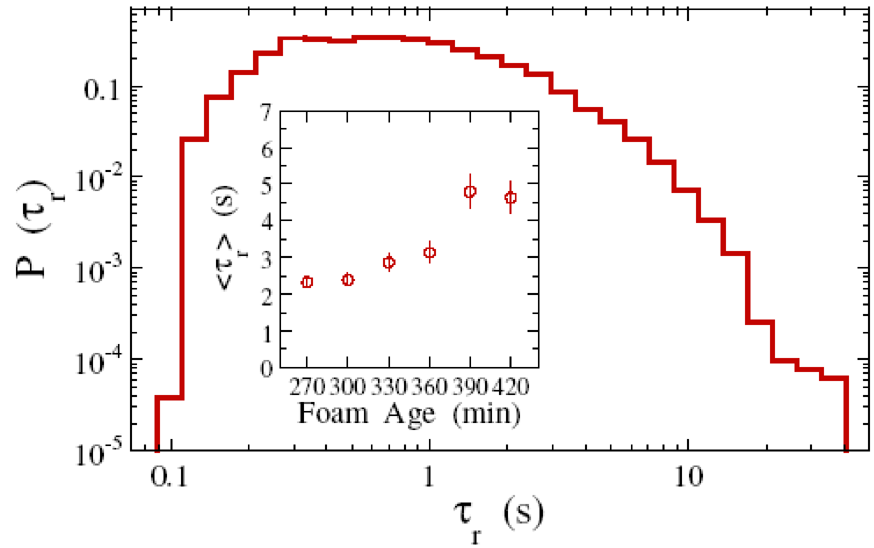
  $\Gamma=4\pi v/\lambda$

  v=bubble speed IF all bubbles are in motion

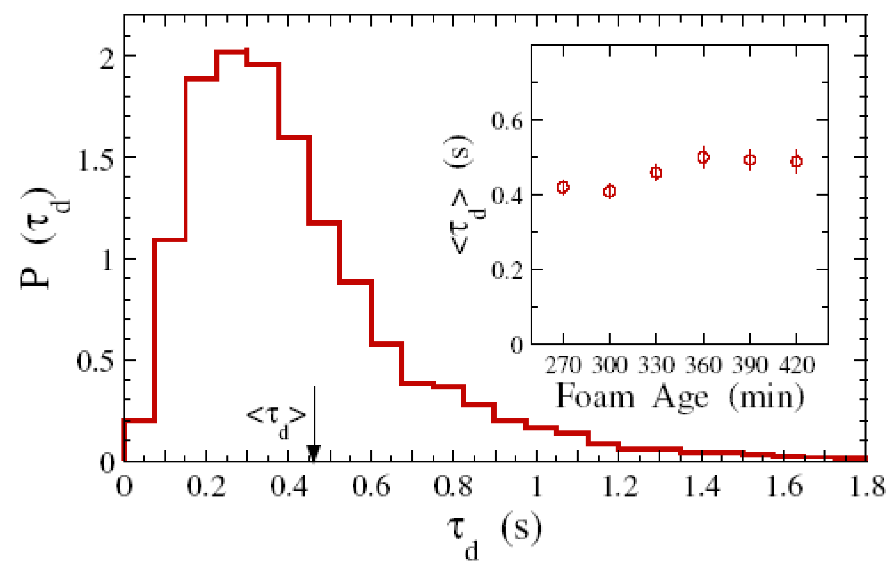


Events Times

- Between events:



- Event durations:



Conclusion

HIGHER-ORDER INTENSITY CORRELATIONS:

- Assess health of data for ordinary systems
  - ought to be routine feature in commercial correlators:

    $\langle I(0)I(t)\rangle$ $\langle I(0)I(t)I(2t)\rangle$ $\langle I(0)I(t)I(2t)I(3t)\rangle$
- Deduce experimental artifacts
- Characterize stationary unsteadiness

SPECKLE-VISIBILITY SPECTROSOPY:

- A time-resolved dynamic light scattering method
- Application to problems of current interest
- Advantages over $\langle I(t_o)I(t_o+\tau)\rangle$:
  - Short times
  - Long runs
  - No storage issues
  - No post-processing

Granular physics at Penn

- Five faculty: DJD, Gollub, Kamien, Liu, Lubensky
- DJD & group:

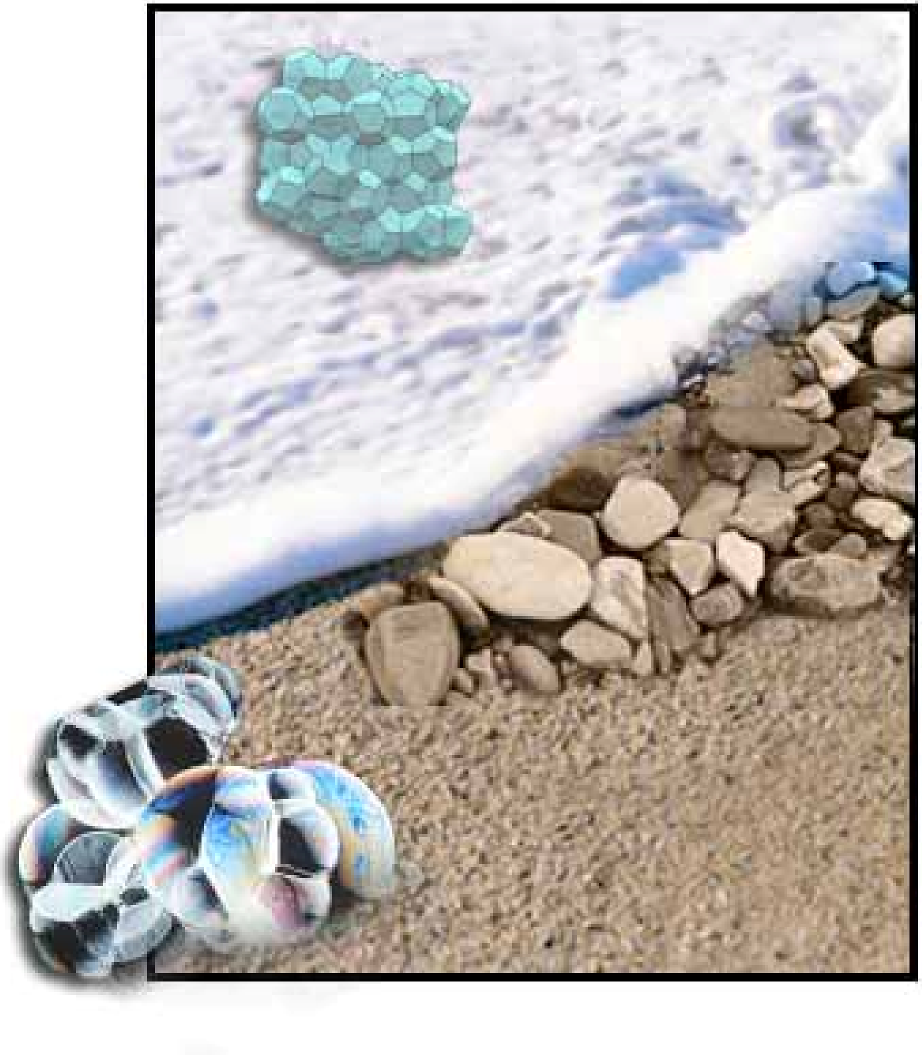
  "sand", foam, microfluidics; novel optical spectroscopies

TODAY'S TALK:

Adam Abate, Ranjini Bandyopadhyay, Paul Dixon, Alex Gittings, Hiroaki Katsuragi, Pierre-Anthony Lemieux, Rajesh Ojha

THE END

- Thank you for your interest.



DURIAN