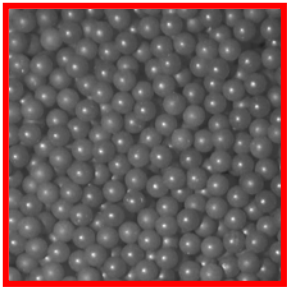# Diffusing Acoustic Wave Spectroscopy (DAWS)

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with

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We study ultrasonic waves in complex (strongly scattering) media.
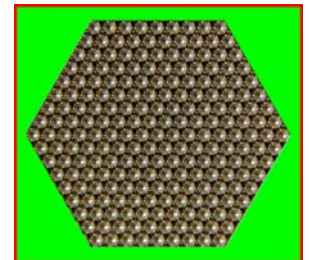
Random media:
- ballistic and diffusive wave transport
- new ultrasound scattering techniques (DSS & DAWS)

Ordered media (phononic crystals):
- ultrasound tunneling
- focusing effects

For more info and papers, see www.physics.umanitoba.ca/~jhpage
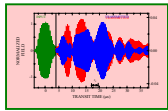
OUTLINE - Diffusing Acoustic Wave Spectroscopy (DAWS):

• Motivation: what can we learn about the dynamics of strongly scattering materials and wave phenomena using ultrasound?

• How the technique works and what it measures:
DAWS is one of two techniques in Ultrasonic Correlation Spectroscopy for measuring the dynamics of materials from the fluctuations of the speckle pattern.
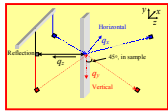
 Dynamic Sound Scattering: uses singly scattered ultrasound to determine absolute motion - analogous to Dynamic Light Scattering [Borne & Pecora, 1976].

 Diffusing Acoustic Wave Spectroscopy: uses multiply scattered ultrasound to measure relative motion - analogous to optical Diffusing Wave Spectroscopy [Maret and Wolf, Z. Phys. B **65**, 409 (1987); Pine et al., PRL **60**, 1134 (1988)].

• Using DAWS to probe fundamental properties of multiply scattered waves:
    phase statistics for temporally varying fields.
    intensity and field fluctuations: breakdown of the Siegert relation for correlated motions of the scatterers.
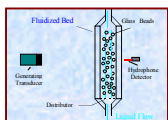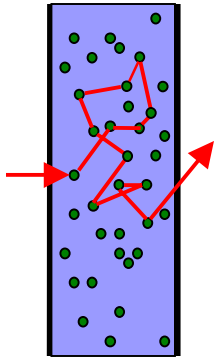
• An application:
 Studying the dynamics of fluidized suspensions of particles.

Motivation:

In strongly scattering materials...

Ultrasonic Speckle Pattern

• The scattered wave fields may be completely dominated by speckle.

⇒ direct imaging may break down
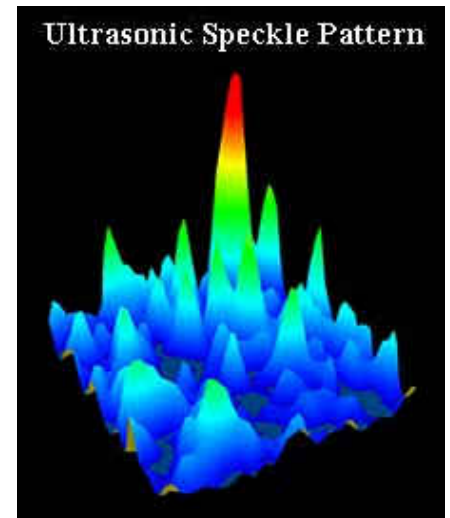
• Ultrasound may be multiply scattered.

⇒ Model the propagation using the diffusion approximation: propagation of multiply scattered ultrasonic waves is treated as a random walk. ($D = v_E \, l*/3$)

• Motion of the scattering particles may be complex.
(e.g. fluidized beds)

⇒ difficult to use Doppler ultrasound

Question: How can we use ultrasound to investigate the dynamics of such strongly scattering materials?

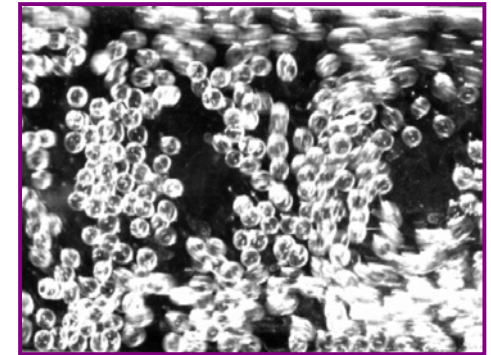Some advantages/features of correlation spectroscopy with ultrasound:

• Length and time scales for ultrasound

      set by the wavelength: $\lambda \sim 1$ mm

      &   period: $T \sim 1$ μs

$\Rightarrow$ can investigate dynamics on longer length scales than is possible with light or X-rays (very much larger for seismic applications)

• "Field fluctuation spectroscopy" - measure the scattered field, not the intensity:

    ∗ measure the field correlation function $g_1(\tau)$ directly

    ∗ can investigate phase…

• Use pulsed techniques (usually) - enables:

    ∗ near-field DSS (determine the scattering angle from the transit time)

    ∗ DAWS at fixed multiple scattering path length (much simpler analysis)

Ultrasonic Correlation Spectroscopy (DSS & DAWS) (Cowan *et al.*, *Phys Rev. Lett.* 85, 453 (2000); Cowan *et al.*, *Phys Rev. E*, in press (June 2002))

Dynamic Speckle Pattern

54.00

• When the scatterers (e.g. particles) move, the speckle pattern fluctuates.
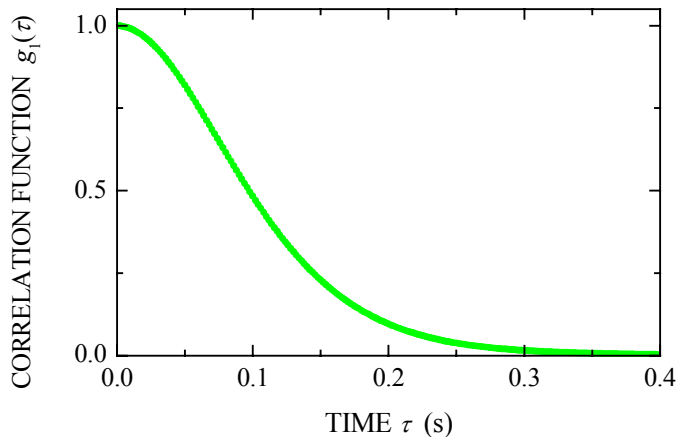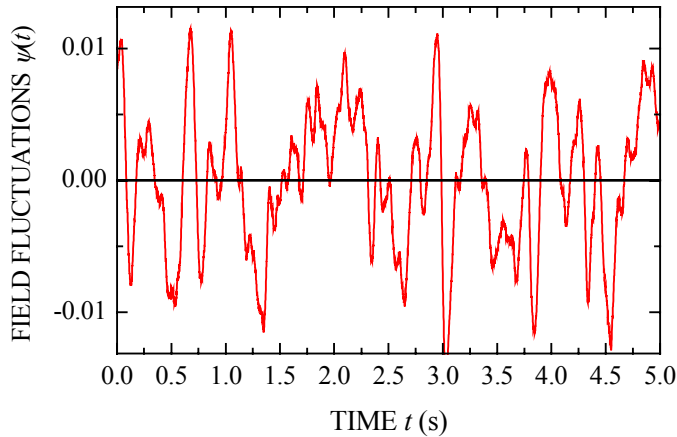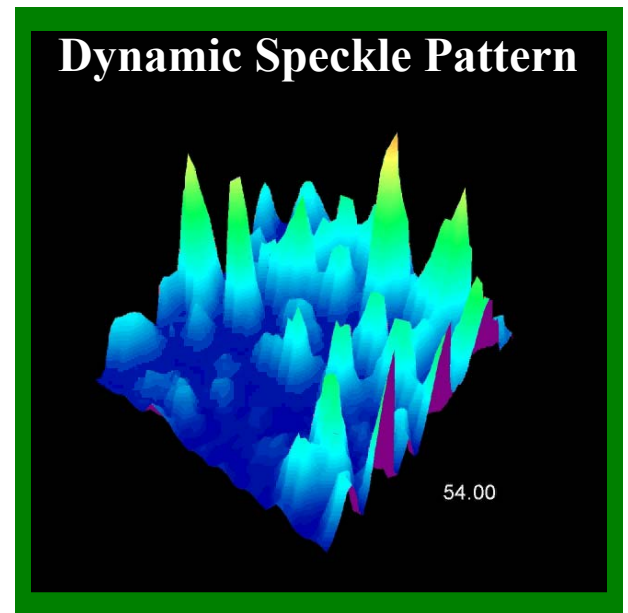
• DAWS determines the motion of the scatterers from the temporal autocorrelation function $g_1(\tau)$ of the transmitted ultrasonic field fluctuations $\psi(t)$.

$$g_1(\tau) = \frac{\int \psi(t)\, \psi^*(t+\tau)\, dt}{\int |\psi(t)|^2\, dt}$$

• The decay of $g_1(\tau)$ results from the total change in phase of the scattered ultrasonic field due to the scatterers' motion.

$$g_1(\tau) \rightarrow \ \sim\tfrac{1}{2} \ \text{ when } \Delta\phi \sim 1 \text{ rad.}$$

Fluidized suspensions - an example of a strongly scattering system with interesting dynamics:

• Challenging scientific questions

Complex behaviour due to hydrodynamic interactions between particles. *(involves the interplay of Newtonian particle dynamics and Navier Stokes fluid dynamics over a wide range of length scales)*
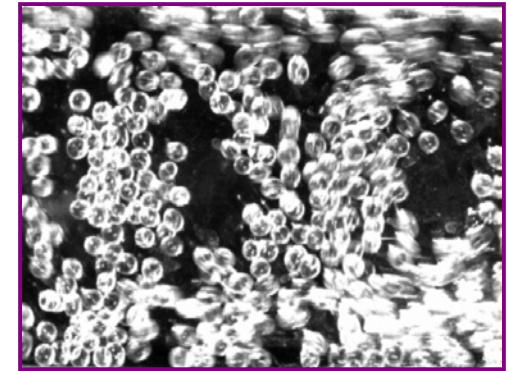
• Both fluidized and sedimenting suspensions have very large velocity fluctuations $(\Delta V \sim V_{flow})$ that are correlated over large distances ξ.

[Segrè *et al.*, PRL **79**, 2574 (1997)]

• What determines the magnitude of ΔV and ξ?

 - How does ΔV depend on volume fraction ϕ?

 - How does ΔV depend on system size L?

 - How does ΔV depend on Reynolds number Re_p ?

• Important practical applications (e.g. slurry bed reactors, flow of slurries in pipelines…)
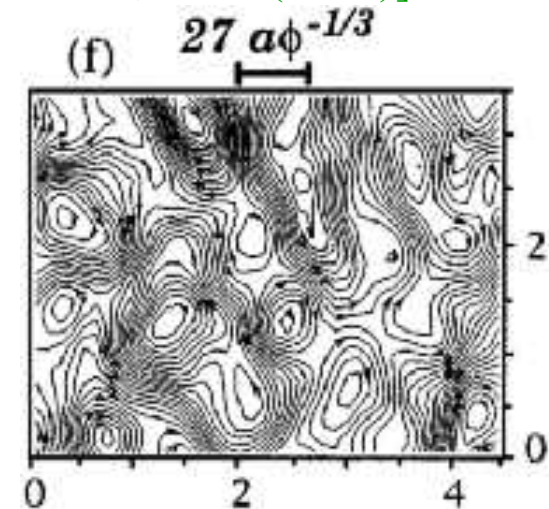
Typical DAWS experimental setup

Sounds of an aquarium...

Fluidized Bed Glass Beads

Generating
Transducer

Hydrophone
Detector

Distributor

Liquid Flow
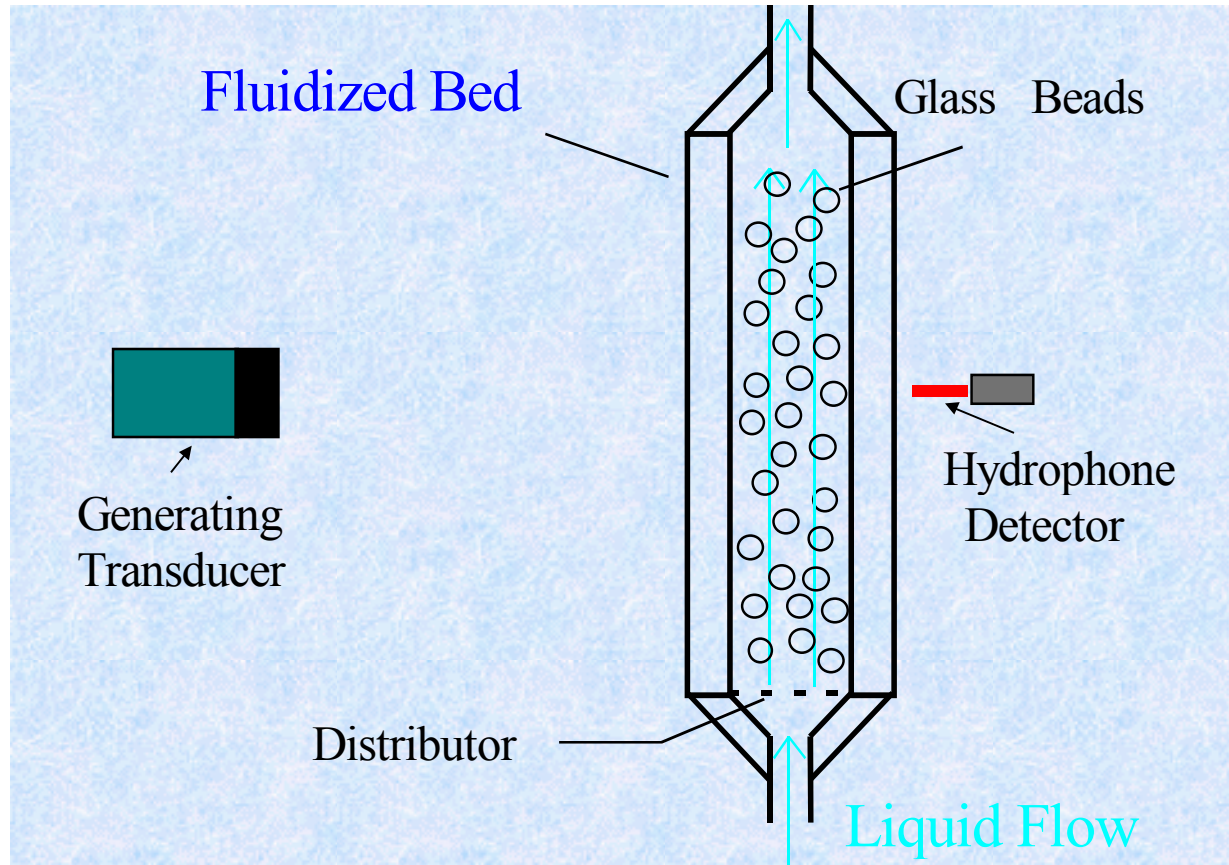
The fluidized bed has a rectangular cross section, and was immersed in a temperature regulated water tank. It contains monodisperse 0.875-mm-diameter glass beads suspended in a mixture of water and glycerol.

Dynamic Sound Scattering (DSS)

(Cowan *et al.*, *Phys Rev. Lett.* 85, 453 (2000); Cowan *et al.*, to be published)

DSS measures the mean square displacement of the particles

Singly scattered ultrasound $\Leftrightarrow$ $\lambda \gg a$

Correlation function:

$$g_1(\tau) = \frac{\left\langle \psi(t)\psi^*(t+\tau)\right\rangle_t}{\left\langle \psi(t)\psi^*(t)\right\rangle_t}$$

(from generator)

k

p $\Delta r_p^2(\tau)$ θ k'

(to detector)
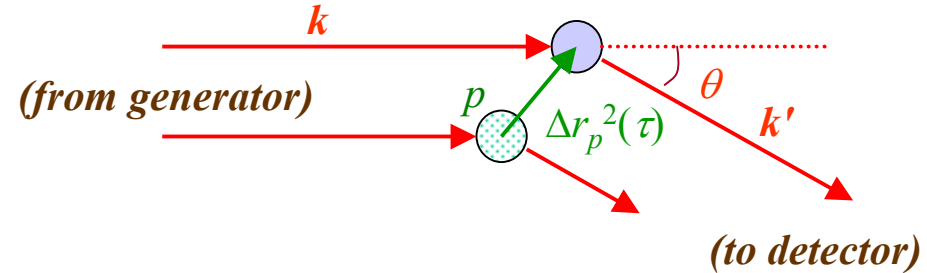
where

$$\psi(t) = \sum_p \psi_p(t) = A\sum_p \exp\left[i\phi_p(t)\right]$$

sum of scattered fields $A\exp\left[i\phi_p(t)\right]$ from each particle p.

$\therefore$

$$g_1(\tau) = \frac{\left\langle \sum_{p,p'} \exp\left[i\{\phi_p(t)-\phi_{p'}(t+\tau)\}\right]\right\rangle_t}{\left\langle \sum_{p,p'} \exp\left[i\{\phi_p(t)-\phi_{p'}(t)\}\right]\right\rangle_t}$$

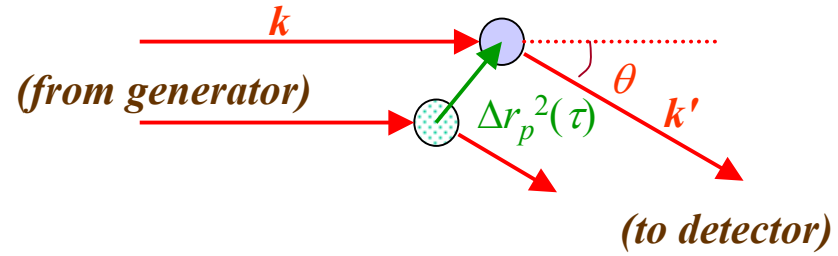For randomly positioned particles, only terms in $g_1(\tau)$, with $p = p'$ survive the time average :

$$\phi_p(t) = \vec{q} \cdot \vec{r}_p(t)$$

$$\phi_{p'}(t + \tau) = \vec{q} \cdot \left[\vec{r}_{p'}(t) + \vec{r}_{p'}(t + \tau) - \vec{r}_{p'}(t) \right]$$

$$= \vec{q} \cdot \left[\vec{r}_{p'}(t) + \Delta \vec{r}_{p'}(\tau) \right]$$

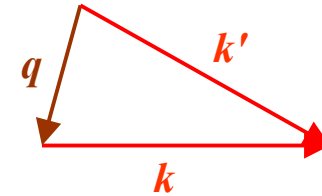$\therefore$ numerator of $g_1(\tau)$ is

$$\left\langle \sum_{p,p'} \exp\left[i\vec{q} \cdot \left\{ \vec{r}_p(t) - \vec{r}_{p'}(t) \right\} \right] \exp\left[-i\vec{q} \cdot \Delta \vec{r}_{p'}(\tau) \right] \right\rangle_t$$

$$\boxed{= 0 \text{ unless } p = p'}$$

(from generator)

k

$\Delta r_p{}^2(\tau)$

θ

k'

(to detector)

Scattering wave vector:

$$\vec{q} = \vec{k}' - \vec{k}$$

q

k'

k

$$\therefore \quad g_1(\tau) = \frac{\left\langle \sum_{p,p'} \exp\left[i\left\{ \phi_p(t) - \phi_{p'}(t + \tau) \right\} \right] \right\rangle_t}{\left\langle \sum_{p,p'} \exp\left[i\left\{ \phi_p(t) - \phi_{p'}(t) \right\} \right] \right\rangle_t} = \frac{\left\langle \sum_p \exp\left[-i\Delta\phi_p(\tau) \right] \right\rangle}{N}$$

$$= \left\langle \exp\left[-i\Delta\phi_p(\tau) \right] \right\rangle$$

$$\Rightarrow \quad \boxed{g_1(\tau) = \left\langle \exp\left[-i\vec{q} \cdot \Delta\vec{r}_p(\tau) \right] \right\rangle}$$

Evaluate ensemble and time average
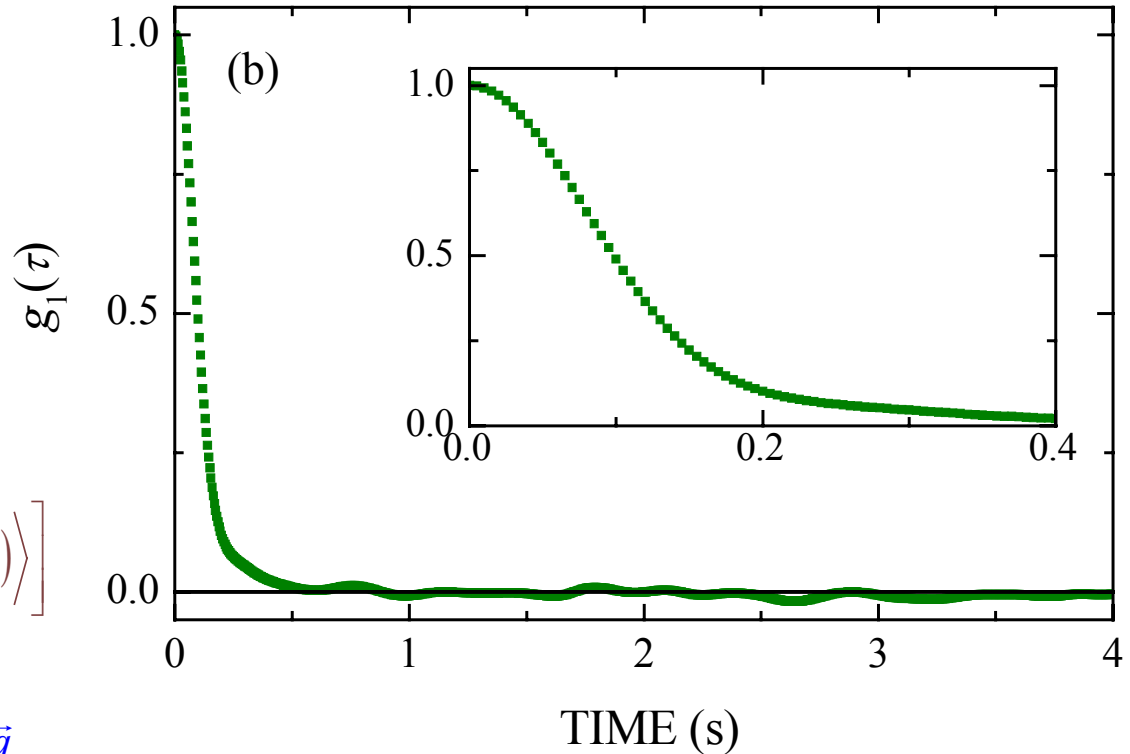using a cumulant expansion

$$g_1(\tau) = \left\langle \exp\left[-i\Delta\phi_p(\tau)\right] \right\rangle$$

$$= 1 - i\left\langle \Delta\phi_p(\tau) \right\rangle - \tfrac{1}{2!}\left\langle \Delta\phi_p^2(\tau) \right\rangle + ...$$

$$= 1 - \tfrac{1}{2}\left\langle \Delta\phi_p^2(\tau) \right\rangle + ...$$

$$\simeq \exp\left[-\tfrac{1}{2}\left\langle \Delta\phi_p^2(\tau) \right\rangle\right]$$

$\vec{q} = \vec{k}' - \vec{k}$
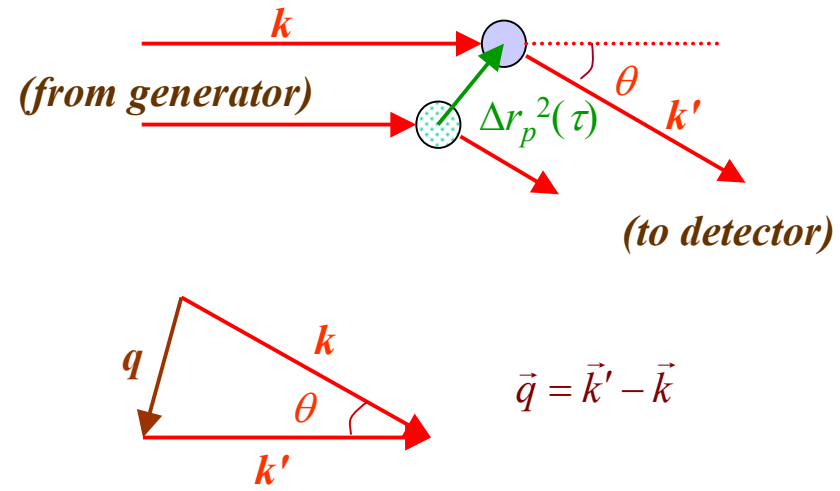
Hence the correlation function
in DSS is

$$g_1(\tau) = \left\langle \exp\left[-i\Delta\phi_p(\tau)\right] \right\rangle$$

$$= \left\langle \exp\left[-i\vec{q}\cdot\Delta\vec{r}_p(\tau)\right] \right\rangle$$

$$= \exp\left[-2k^2\sin^2(\theta/2)\left\langle \Delta r_{\vec{q}}^2(\tau) \right\rangle\right]$$

where $\left\langle \Delta r_q^2(\tau) \right\rangle$ is the mean square
displacement of the particles along $\vec{q}$

Mean square displacement

For ballistic particle motion:

$$\langle \Delta r_{\vec{q}}^2(\tau) \rangle \;=\; \frac{\langle \Delta V_{\vec{q}}^2 \rangle \tau^2}{1 + \left(\tau/\tau_c \right)^{2-m}}$$

$$\approx \; \langle \Delta V_{\vec{q}}^2 \rangle \tau^2$$

(for small τ)

By measuring $\langle \Delta r_q^2(\tau) \rangle$ versus τ, we determine:

$$V_{rms} \;=\; \sqrt{\left\langle \left\langle \Delta V_{\vec{q}}^2 \right\rangle \right\rangle}$$

(the rms particle velocity along $\vec{q}$)

$$d_c \;=\; V_{rms}\, t_c$$

(the dynamic correlation length)

τ^2
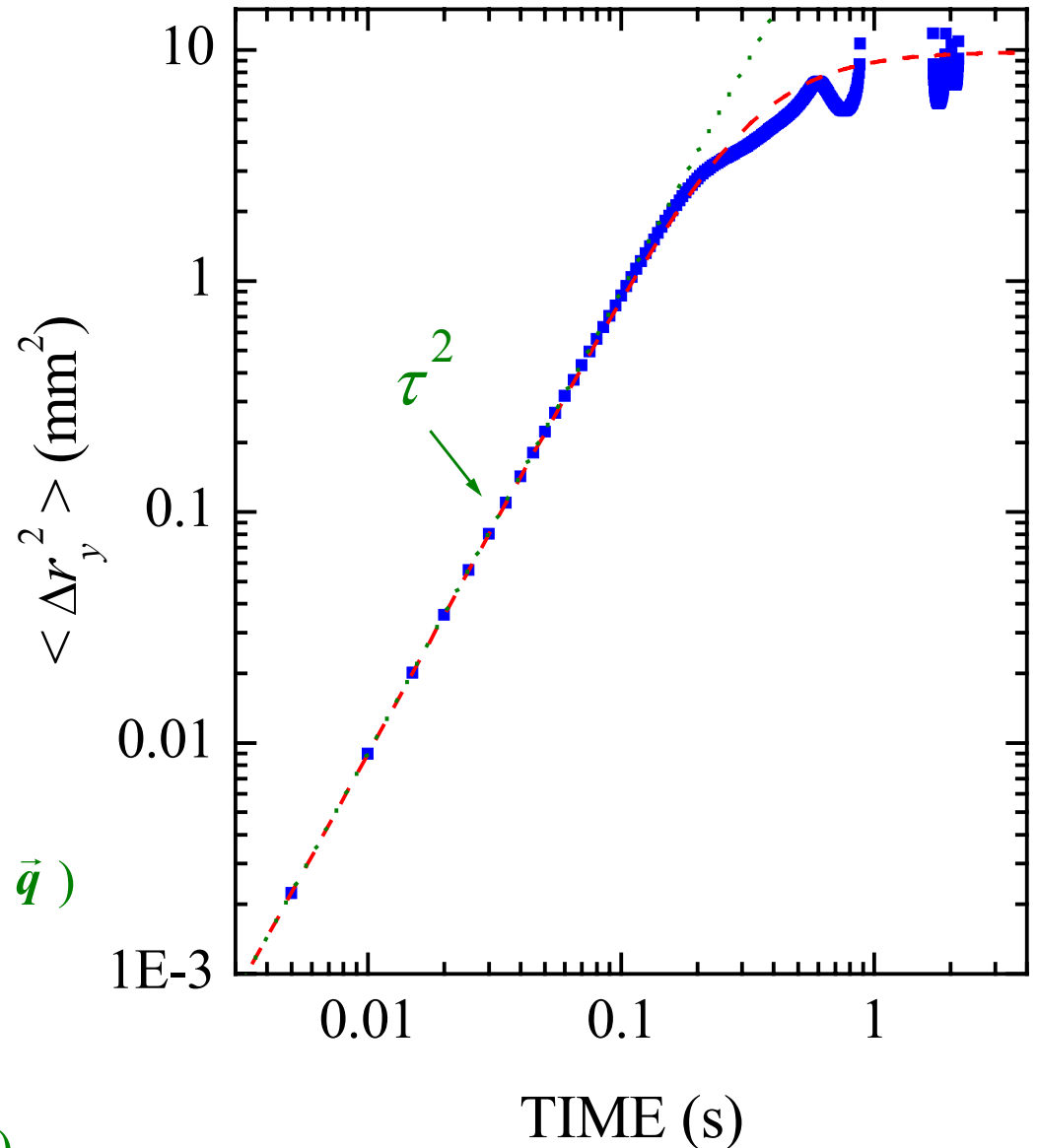
Experiment

(a) DSS far-field scattering geometries:

All three components of root mean square particle velocities V_{rms} can be measured

DSS far field measurements of the rms particle velocities in a fluidized bed, showing the dependence on volume fraction. (V_f is the average fluid velocity.)
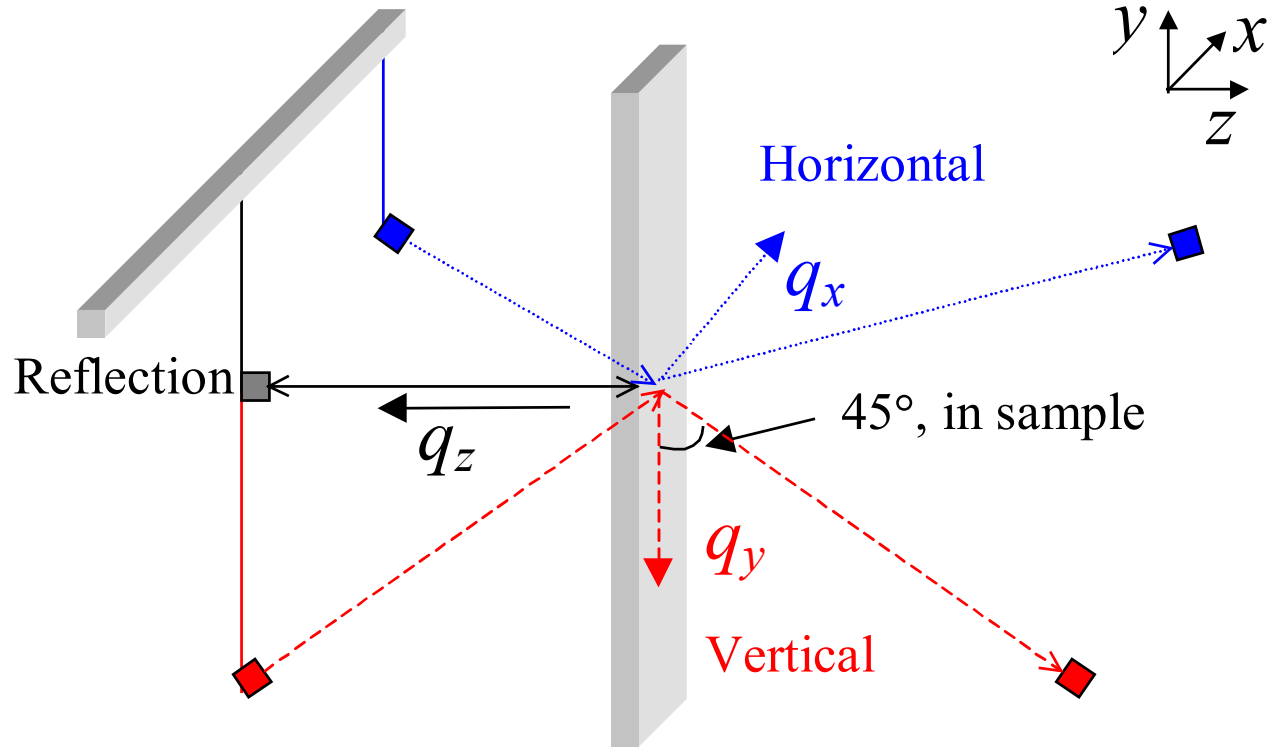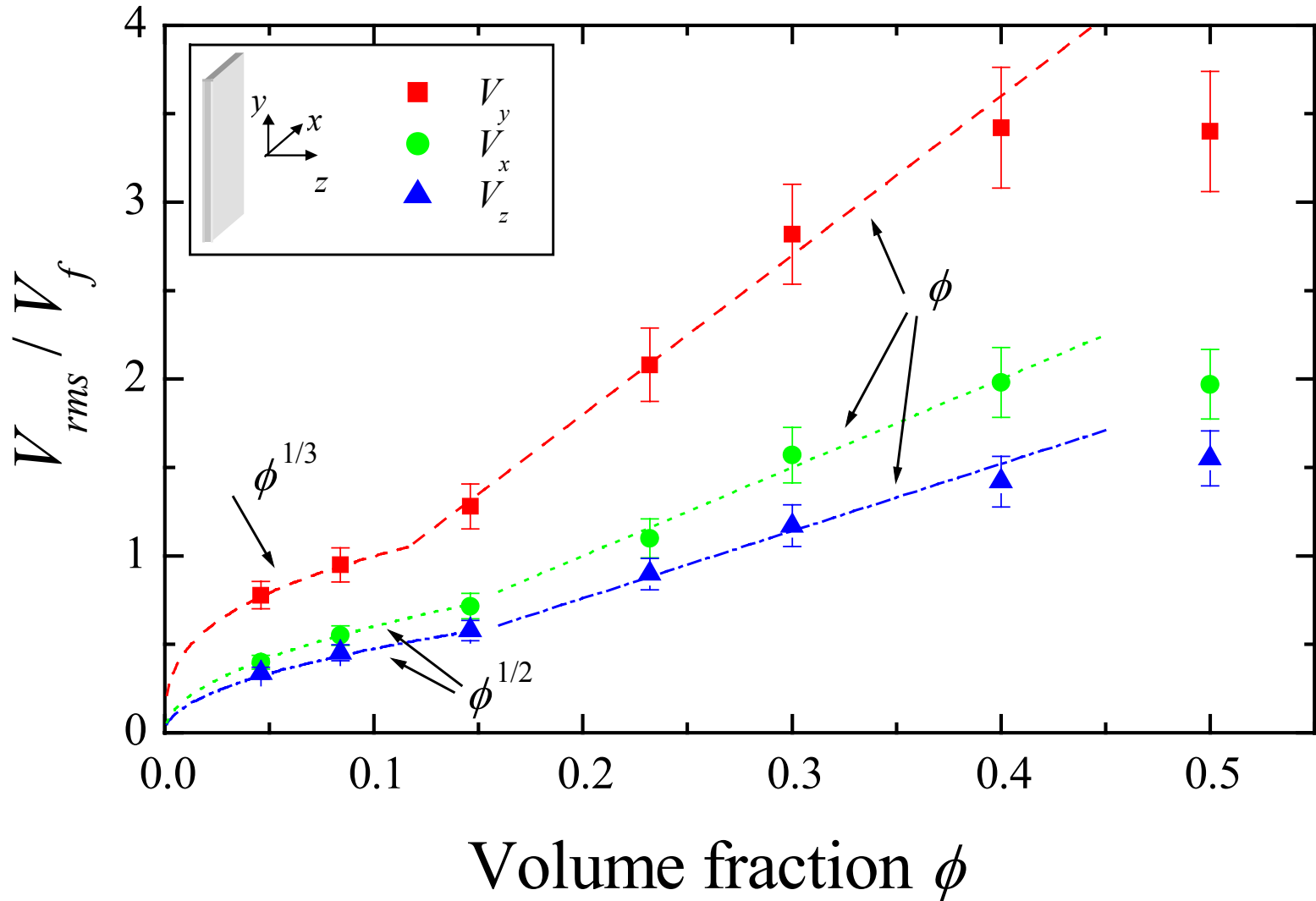
(b) DSS near-field scattering geometry:
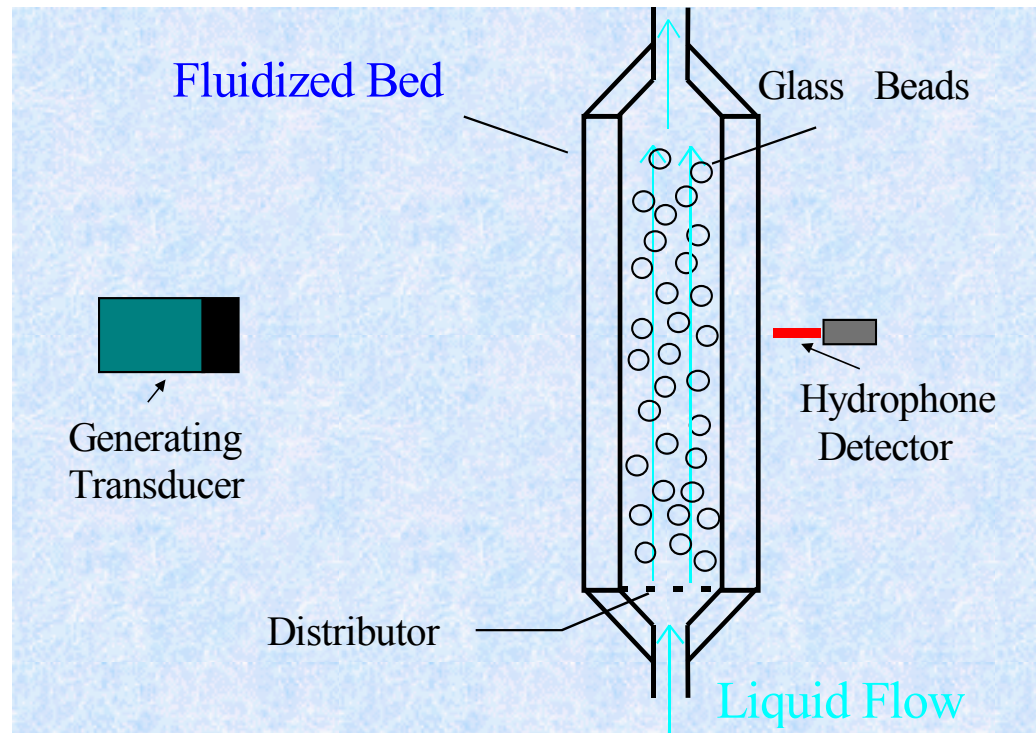
- Measure scattered field in a single near-field speckle using a miniature hydrophone $(d < \lambda)$

- Determine the scattering angle from the transit time

- Measure total rms velocity in the scattering plane

$$V_{rms} \approx \sqrt{V_{rms,x}^2 + V_{rms,y}^2}$$

Diffusing Acoustic Wave Spectroscopy (DAWS)

DAWS uses multiply scattered (diffusing) ultrasound to measure the relative motion of scattering particles that are located a transport mean free path *l** apart.

(a) Pulsed DAWS

Measure the field fluctuations for multiple scattering paths of fixed length $s = (n + 1)l^*$, where n is the number of steps.

The decay of $g_1(\tau)$ is determined by the total phase change $\Delta\phi^{(n)}(\tau)$ for paths containing n steps. Along the blue path in the figure
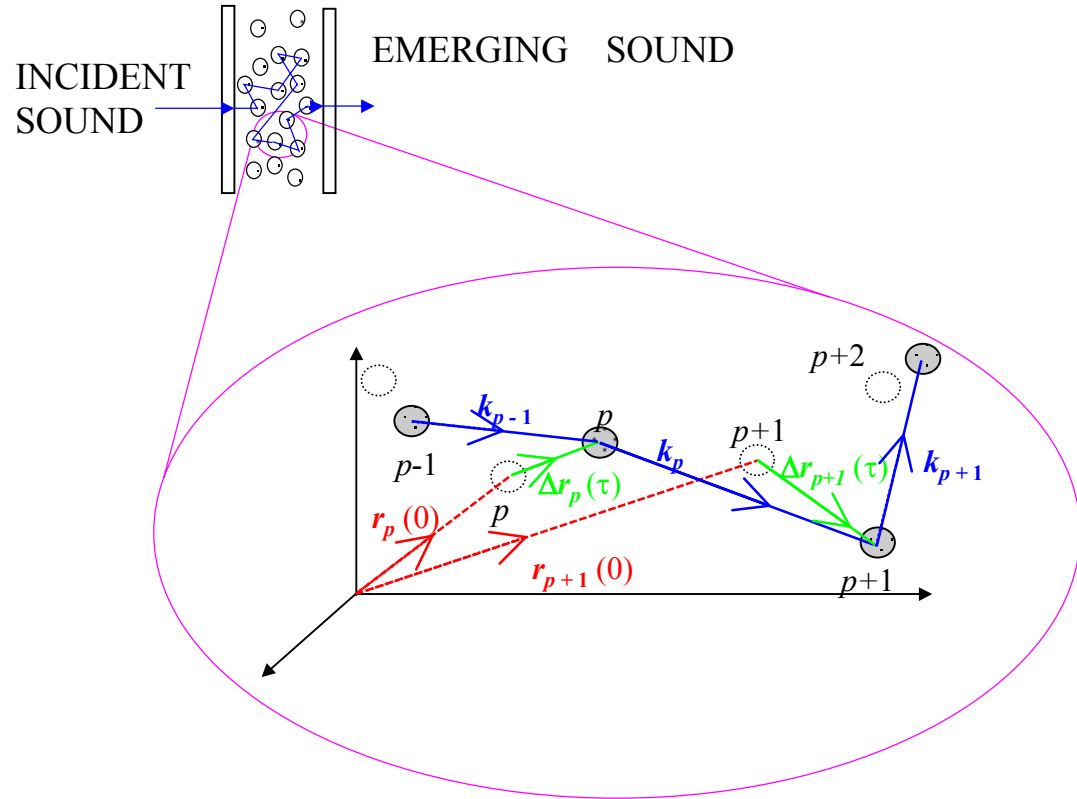
$$\Delta\phi^{(n)}(\tau) = \sum_{p}^{n} \Delta\phi_p(\tau)$$

$$= \sum_{p=0}^{n}\left[\vec{k}_p\cdot\left(\Delta\vec{r}_{p+1}(\tau)-\Delta\vec{r}_p(\tau)\right)\right] = \sum_{p=1}^{n-1}\vec{k}_p\cdot\Delta\vec{r}_{p,\,\text{rel}}(\tau,l^*) \; + \;$$

(phase change due to motion of the first and last scatterer relative to source and detector) - small contribution for large n.

The field autocorrelation function is

$$g_1(\tau) = \left\langle \exp\left[-i\Delta\phi^{(n)}(\tau)\right]\right\rangle$$

average over all paths with n steps

$$\simeq \left\langle \exp\left[-i\Delta\phi_p(\tau)\right]\right\rangle^n$$

assume successive phase shifts $\Delta\phi_p(\tau)$ are uncorrelated

$$\simeq \exp\left[-\left\langle\Delta\phi_p^2(\tau)\right\rangle n/2\right]$$

cumulant expansion

But

$$\left\langle\Delta\phi_p^2(\tau)\right\rangle = \left\langle\left(\vec{k}_p \cdot \Delta\vec{r}_{\text{rel},p}\right)^2\right\rangle = \left\langle\left(k_p\,\Delta r_{\text{rel},p}(\theta_p)\cos\theta_p\right)^2\right\rangle$$

$$= \tfrac{1}{3}k^2\left\langle\Delta r_{\text{rel}}^2\right\rangle$$

change in separation of the $(p+1)^{\text{th}}$ and p^{th} scatterers

for uncorrelated motion

N.B.: For fluidized suspensions, the spatial correlations decay rapidly, and the last expression is a good approximation for $\langle\Delta\phi_p^2(\tau)\rangle$

But for uniform shear, $\langle\Delta\phi_p^2(\tau)\rangle = 0.6 \times k^2\langle\Delta r_{\text{rel}}^2(\tau)\rangle\,/3$
and for pure rotations, $\langle\Delta\phi_p^2(\tau)\rangle = 0.$

The field autocorrelation function can be simply written as

$$g_1(\tau) \simeq \exp\left[-\frac{nk^2}{6}\left\langle \Delta r_{\text{rel}}^2(\tau, l^*)\right\rangle\right]$$

where $\left\langle \Delta r_{\text{rel}}^2(\tau, l^*)\right\rangle$ is the relative mean square displacement of particles located a distance l^* apart.

N.B.: $g_1(\tau)$ can also be expressed in terms of the average local strain as

$$g_1(\tau) \simeq \exp\left[-\frac{nk^2 l^{*2}}{6}\overline{\varepsilon}^2\right]$$

where

$$\overline{\varepsilon}^2 \equiv \frac{2}{5}\left[\left\langle\left(\sum \varepsilon_{ii}\right)^2\right\rangle + 2\sum_{i,j}\left\langle \varepsilon_{ij}^2\right\rangle\right] \approx \left\langle \Delta r_{rel}^2(\tau, l^*)\right\rangle \Big/ l^*$$

and ε_{ij} is the local strain tensor

$$\varepsilon_{ij}(\tau) = \frac{1}{2}\left(\frac{\partial u_i(\tau)}{\partial r_j} + \frac{\partial u_j(\tau)}{\partial r_i}\right)$$

where $u_i(\tau)$ are the components of $\Delta\vec{r}(\tau)$

(c.f.: Bicout and Maynard, Physica A **199**, 387 (1993); Bicout and Maret, ibid **210**, 87 (1994))

A sequence of transmitted waveforms in pulsed DAWS, showing that the field fluctuates more rapidly as the path length (and n) becomes longer.
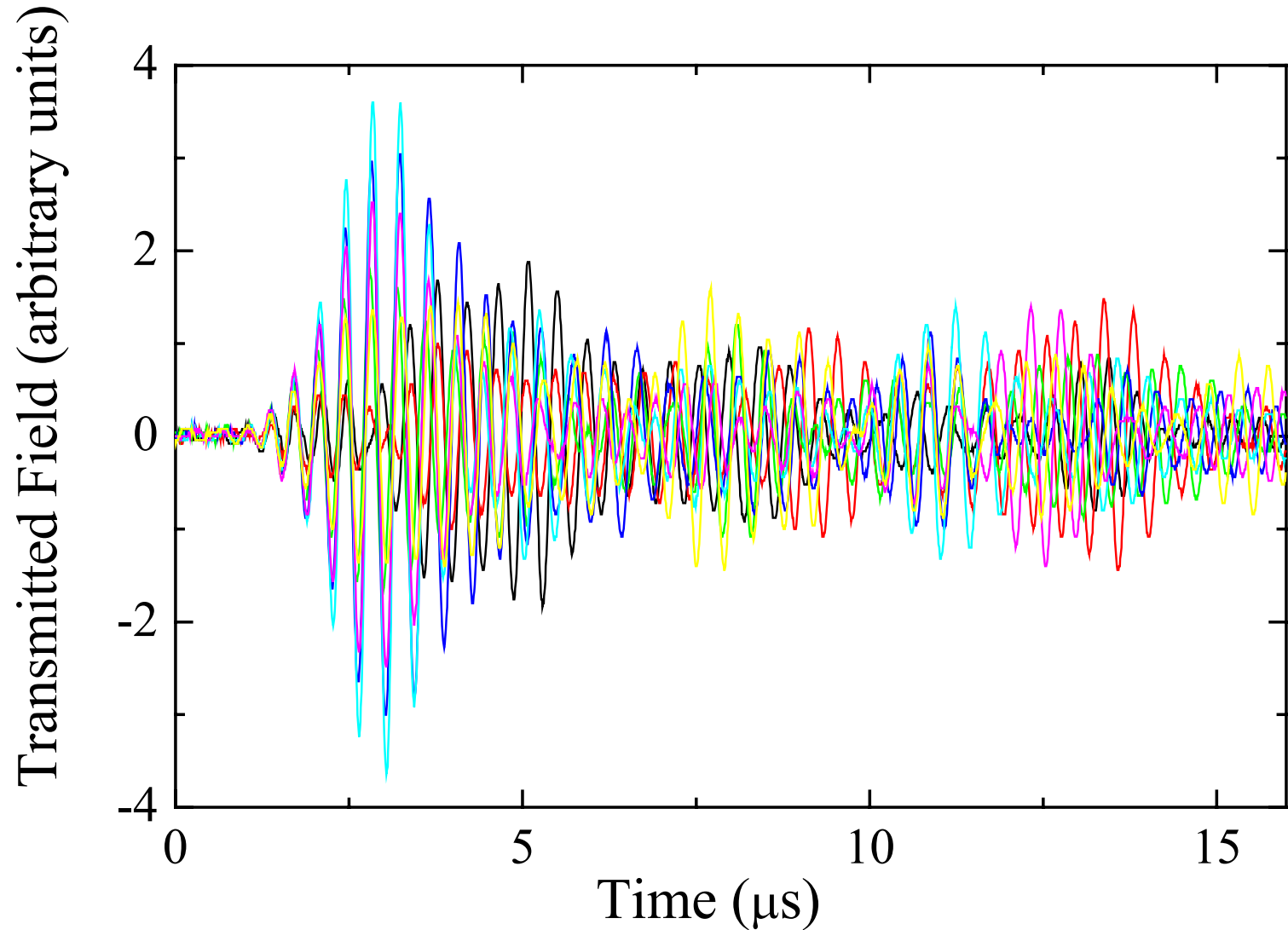
Pulsed DAWS
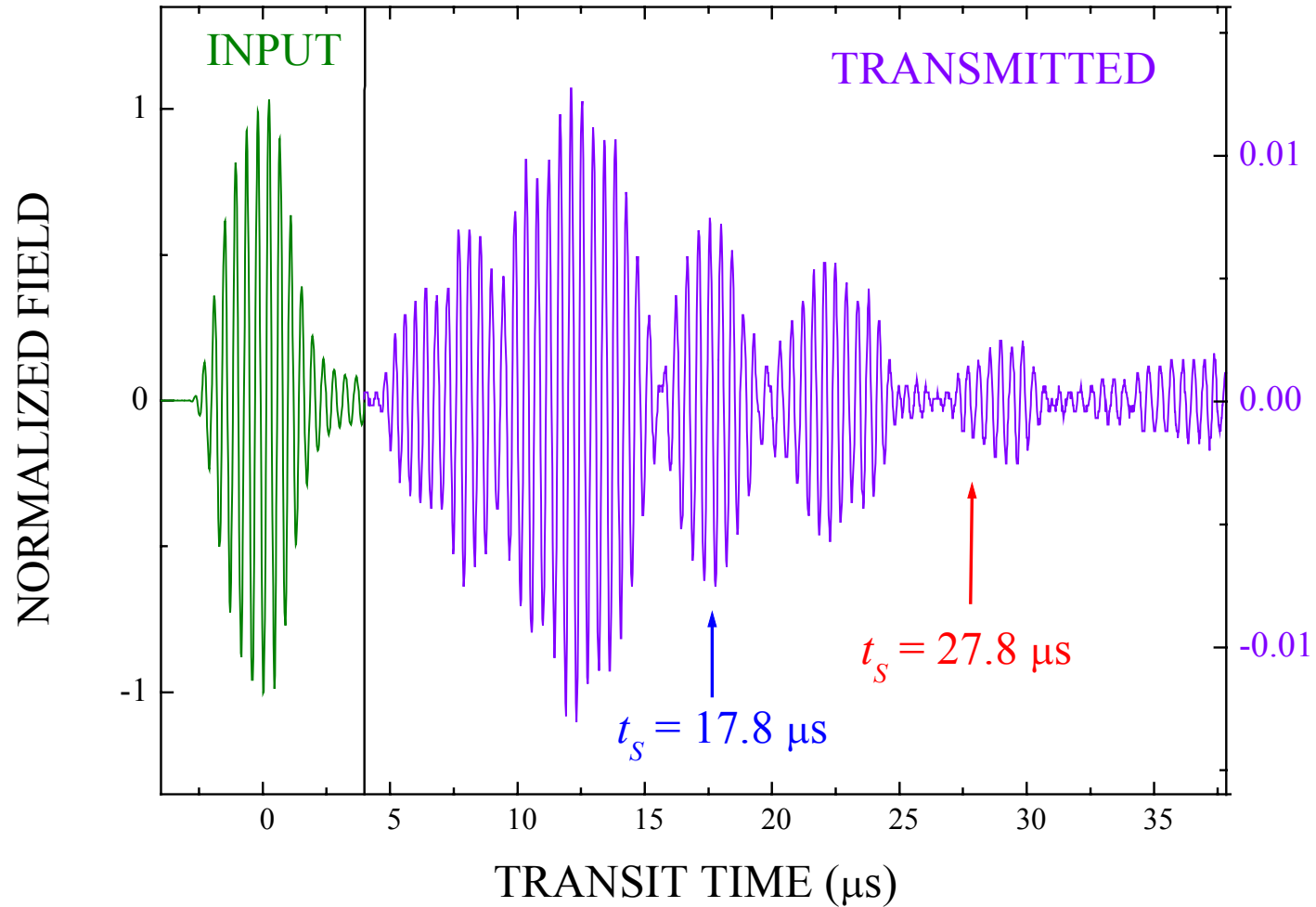
- In pulsed DAWS, a train of pulses is sent into the sample.

- The scattered field is sampled at a particular time after each pulse (using a boxcar).

- Therefore the sampling time t_s sets the average path length, $t_s = s / v_e \cong n \, l^* / v_e$.

Field fluctuations measured at the two sampling times, and the corresponding correlation functions, calculated numerically using FFTs and the correlation theorem:

$$\Im[f \otimes g] = \Im[f]\Im[g^*]$$
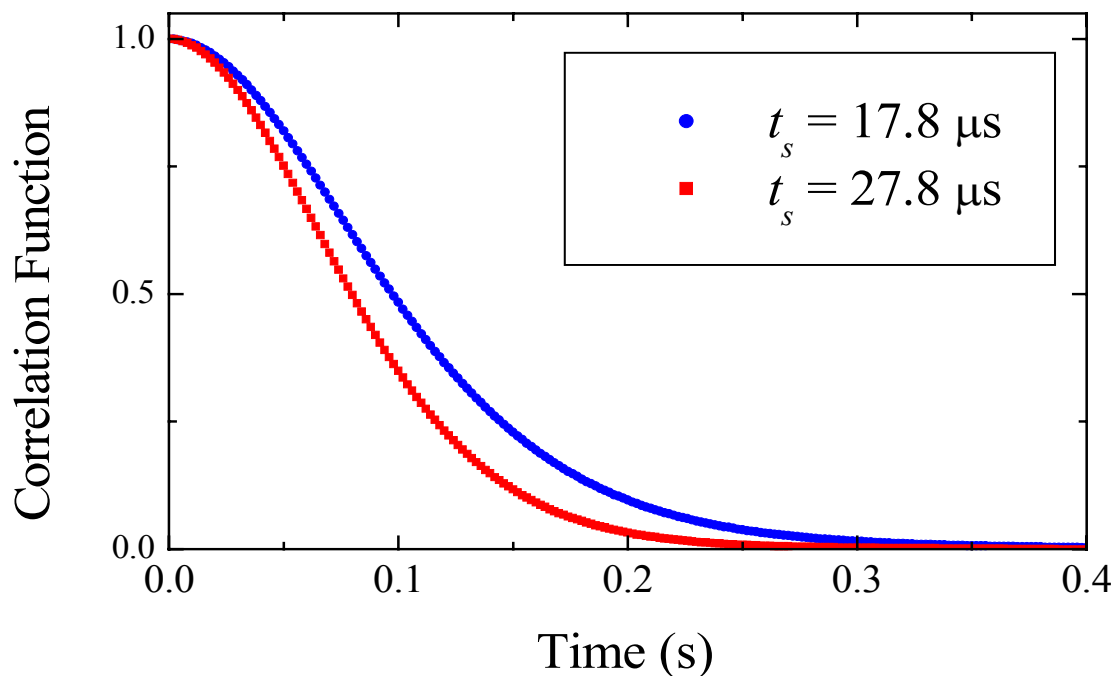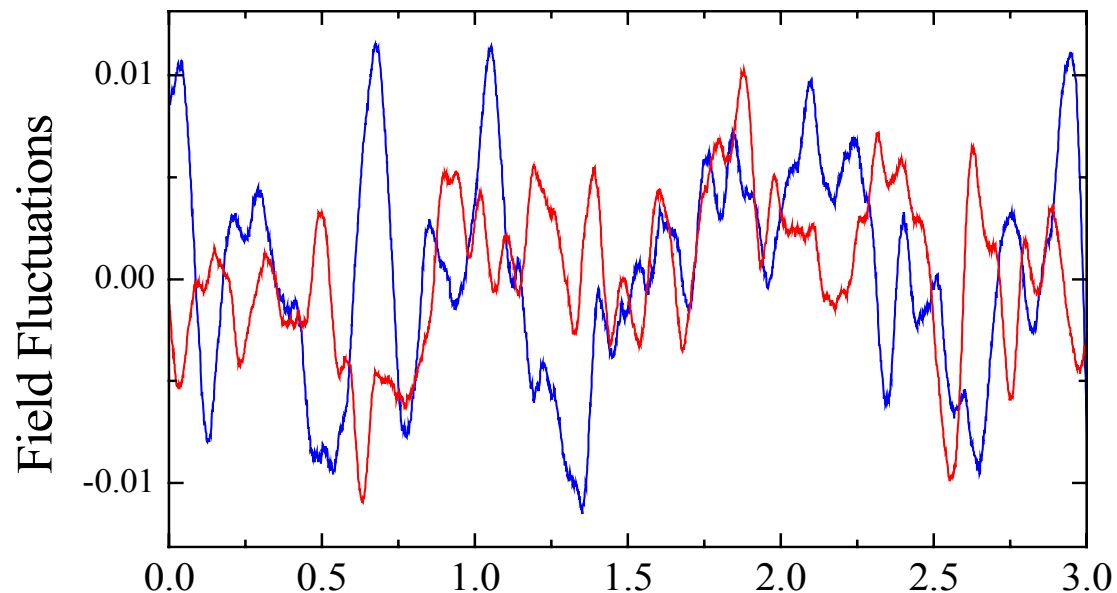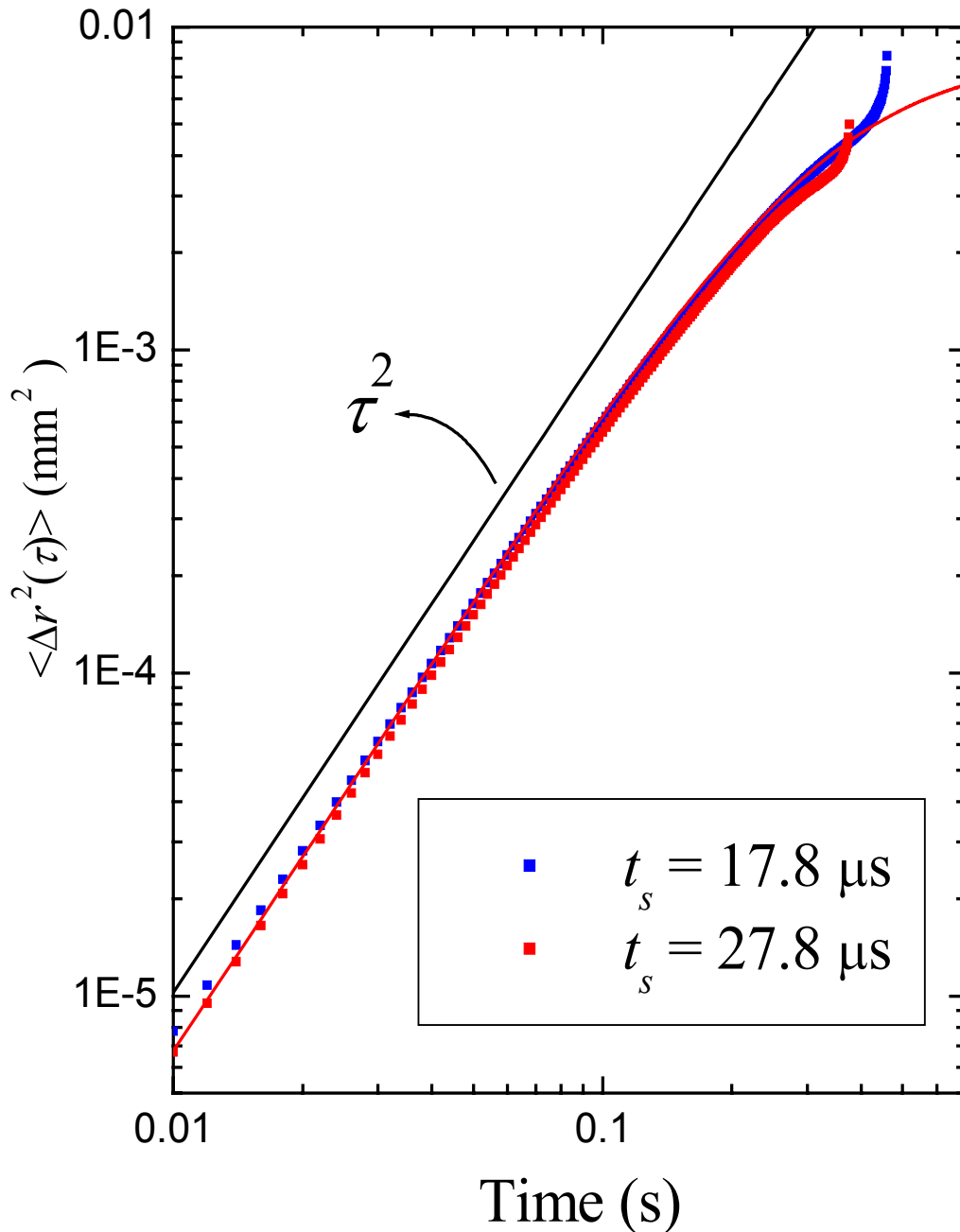
Invert the field correlation function $g_1(\tau)$

$$g_1(\tau) \approx \exp\left[-\left(s/l^*\right)k^2\langle\Delta r_{\text{rel}}^2(\tau)\rangle/6\right]$$

to determine $\langle\Delta r^2_{rel}(\tau)\rangle$ - the relative mean square displacement of particles located a distance l^* apart. (independent of the sampling time, as it should be!)

[requires measurements of l^*, v_e and $k = \omega/v_p$ - Page *et al.*, PRE **52**, 3106 (1995); Schriemer *et al.*, PRL **79**, 3166 (1997); Page et al., Science **271**, 634 (1996); Cowan et al., PRE **58**, 6626 (1998).]

Relative mean square displacement
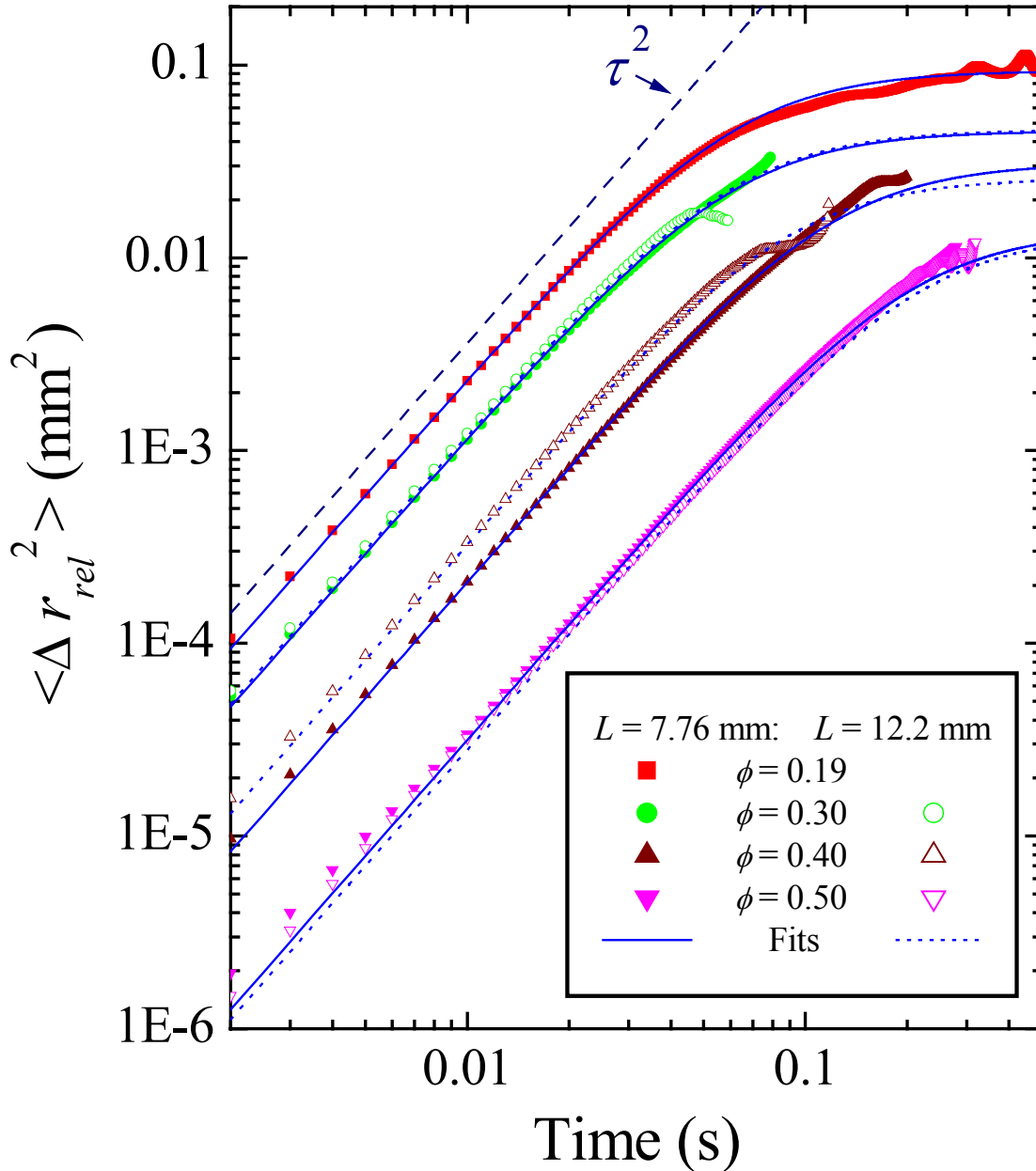
Fit with:

$$\langle \Delta r^2_{rel}(\tau) \rangle = \frac{\langle \Delta V^2_{rel} \rangle \tau^2}{1 + \left(\tau / \tau_{cl} \right)^2}$$

Measure:

$$\Delta V_{rel} = \sqrt{\langle \Delta V^2_{rel} \rangle}$$

(rms relative velocity fluctuations)

$$\Delta d_{sep} = \Delta V_{rel} \, \tau_{cl}$$

(Average change in particle separation before interactions change the particle trajectories)

Legend:

$L = 7.76$ mm:	$L = 12.2$ mm
■ $\phi = 0.19$	
● $\phi = 0.30$	○
▲ $\phi = 0.40$	△
▼ $\phi = 0.50$	▽
—— Fits	······

Axis labels:
- $\langle \Delta r^2_{rel} \rangle$ (mm^2)
- Time (s)
- τ^2

Shortest length scale on which the relative mean square displacement can be measured [determined by $l*$ at our highest frequency (2.35 MHz)] compared with the average nearest neighbour separation d_{nn}.

ϕ^{-1}

$d_{nn} = 2\ a\ 0.9\ \phi^{-1/3}$

$l^* \text{ (mm)}$

ϕ
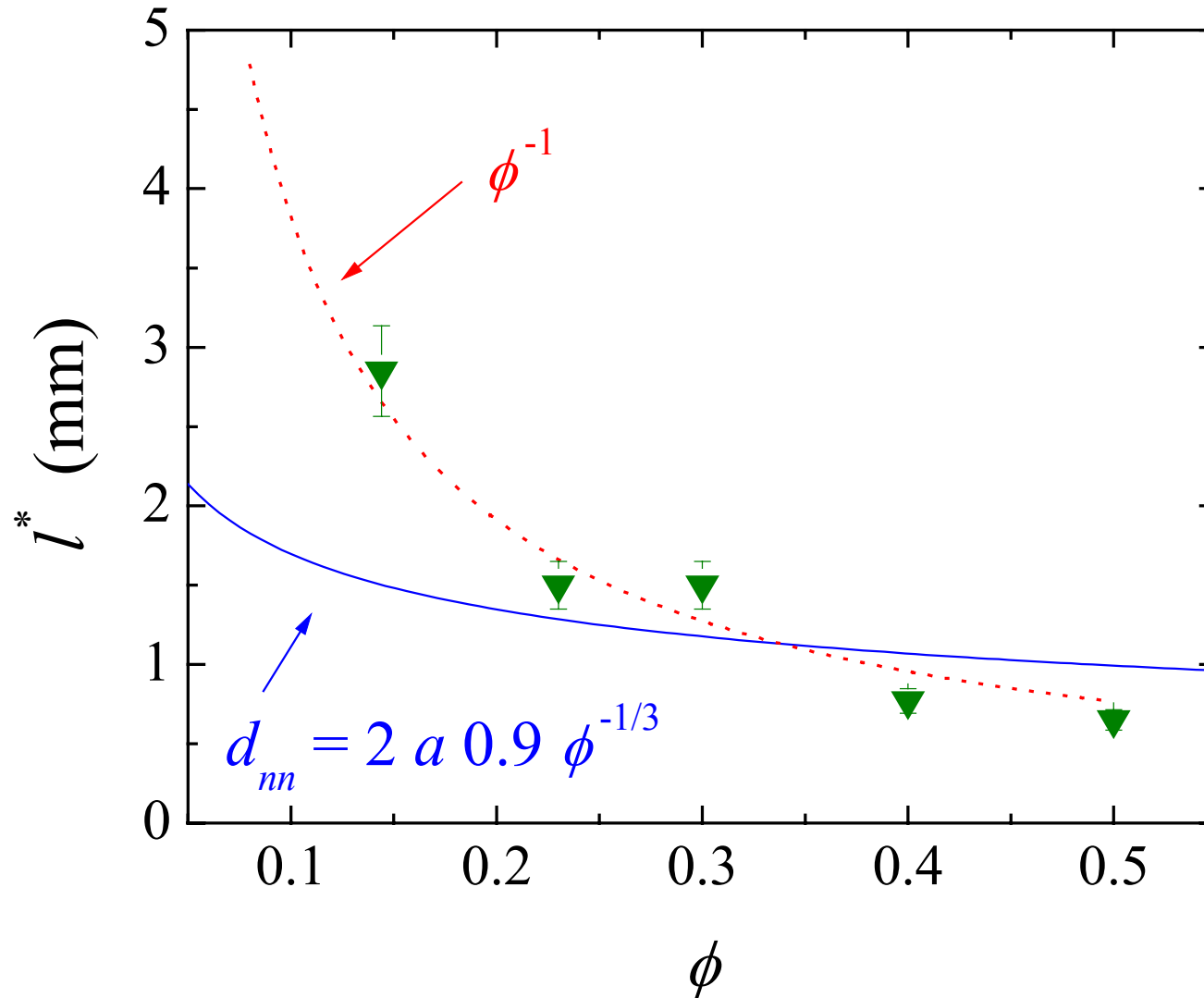
Length Scale Dependence of ΔV_{rel}

DAWS measures the relative motion (ΔV_{rel}) of scatterers separated by l^*:

$$\left\langle \Delta V_{rel}^2 \left(l^* \right) \right\rangle = \left\langle \left(\Delta \vec{V} \left(\vec{r} + l^* \right) - \Delta \vec{V} \left(\vec{r} \right) \right)^2 \right\rangle$$

Velocity correlation function

$$= 2 \left\langle \Delta V^2 \right\rangle - 2 \left\langle \Delta \vec{V} \left(\vec{r} + l^* \right) \cdot \Delta \vec{V} \left(\vec{r} \right) \right\rangle$$

$$= 2 V_{rms}^2 \left(1 - \exp\left[-l^* / \xi \right] \right)$$

- By changing the frequency l^* can be varied.

- V_{rms} can be measured using single scattering at a low frequency (DSS).

- Thus the instantaneous velocity correlation length ξ can be estimated.

Length Scale Dependence of ΔV_{rel}
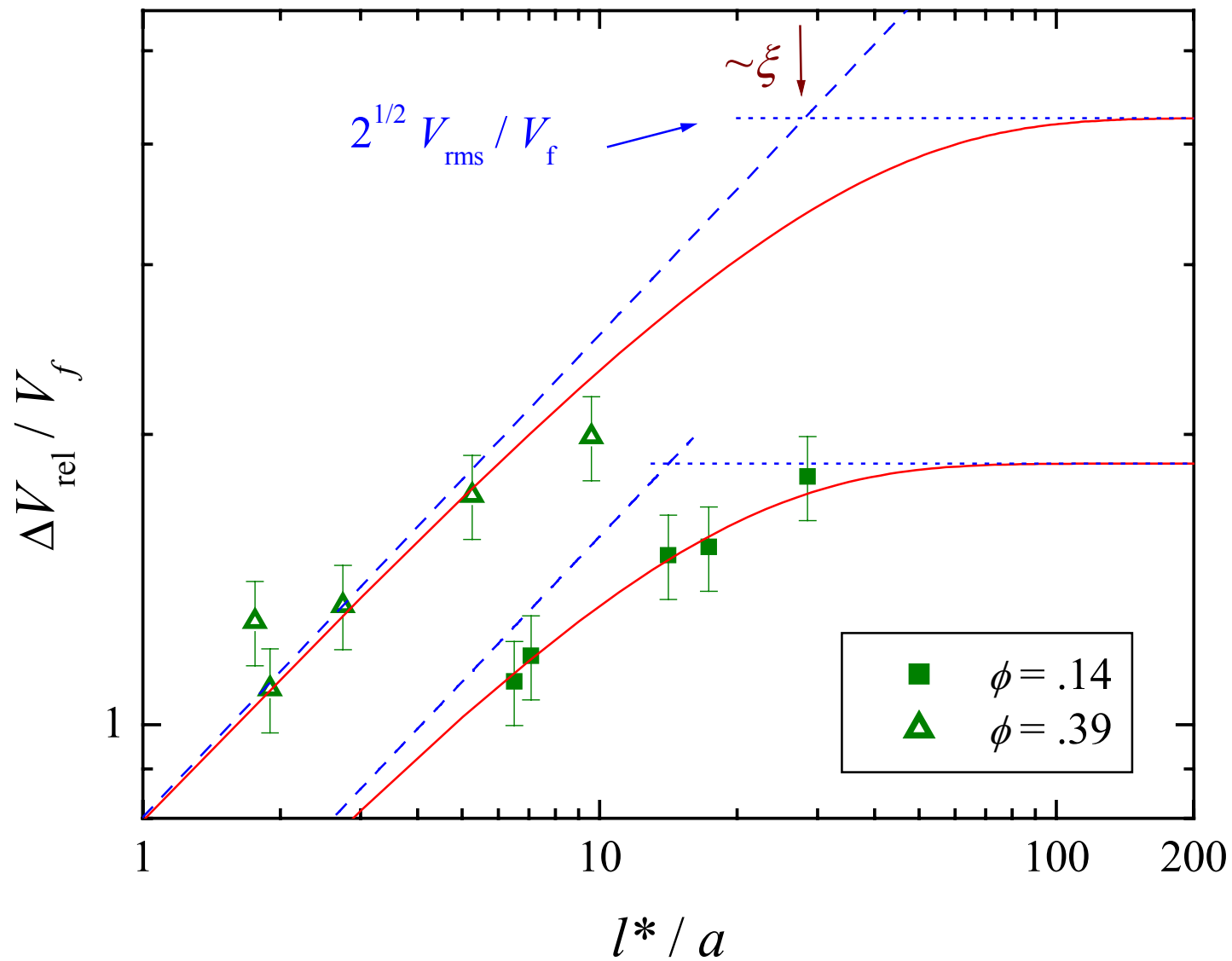
Scaled separation dependence of ΔV_{rel}

$2^{1/2}$

$2^{1/2}[1 - e^{-l^*/\xi}]^{1/2}$

$\Delta V_{\text{rel}} / V_{\text{rms}}$

l^* / ξ

$\phi = 0.080$
$\phi = 0.144$
$\phi = 0.22$
$\phi = 0.30$
$\phi = 0.40$
$\phi = 0.50$
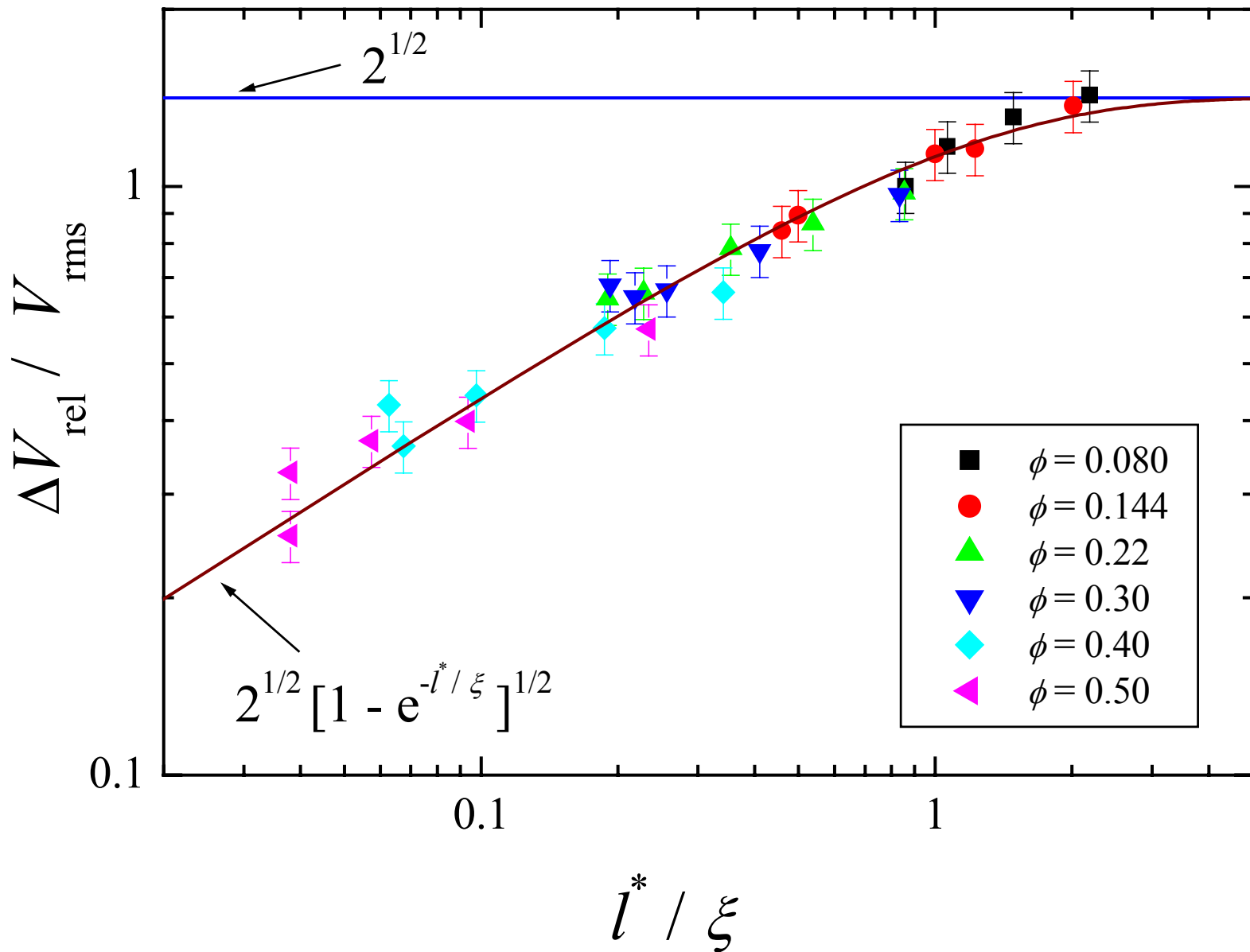
Dependence of ΔV_{rel} $(= \sqrt{\langle \Delta r^2_{rel}(\tau) \rangle} / \tau)$ on path length for short paths.
Include the contributions to $g_1(\tau)$ due to the motion of the first and last scatterer relative to the source and detector (red terms):
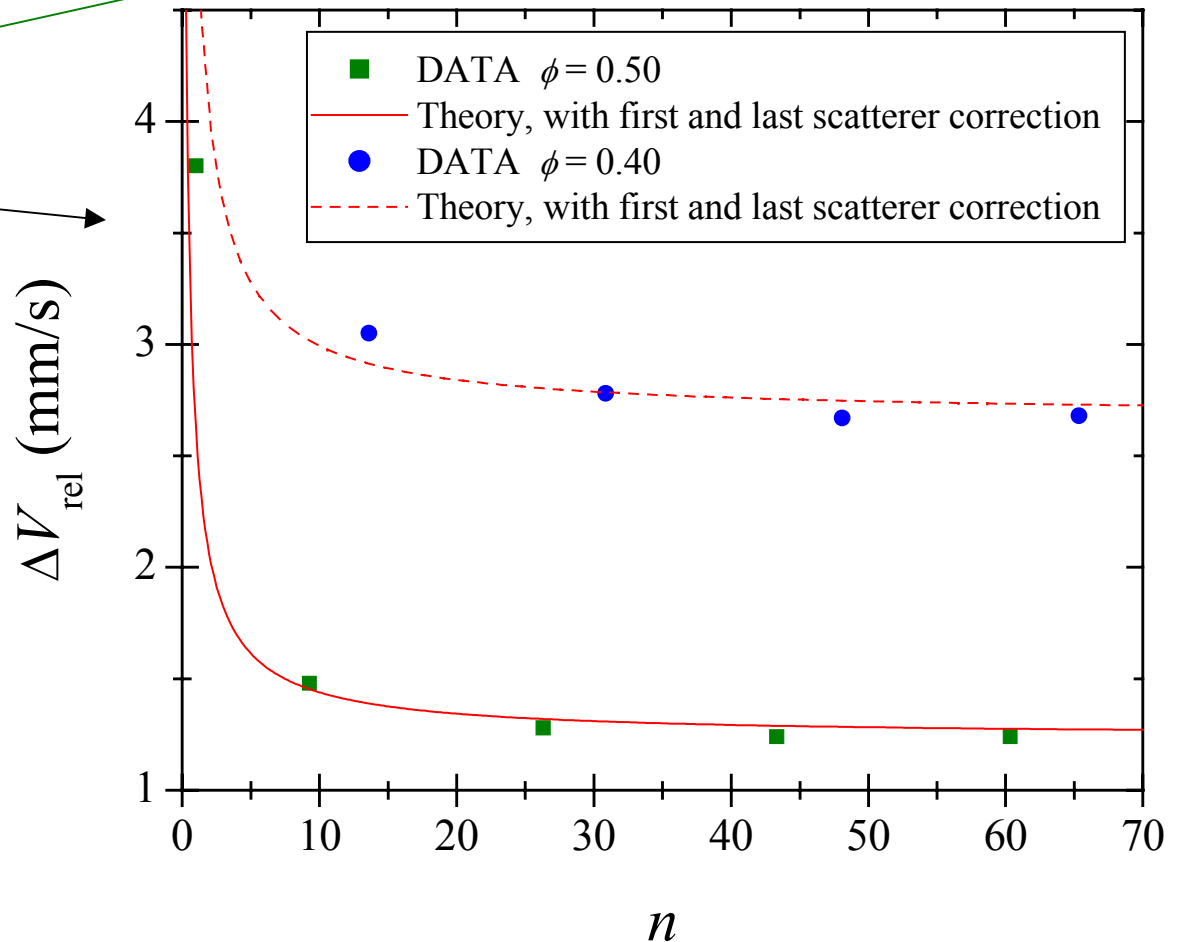
$$g_1(\tau) \approx \exp\left[-\frac{nk^2}{6}\left[\langle \Delta r_{rel}^2(\tau, l^*) \rangle + \frac{1}{n}\left\{ \langle \Delta r_{rel}^2(\tau, R) \rangle - \langle \Delta r_{rel}^2(\tau, l^*) \rangle \right\} \right] \right]$$

(R = linear distance between first and last scatterer.)

Data: ΔV_{rel} determined without correction term

Curves: Actual ΔV_{rel} plus red correction term

Conclude: The simple expression for $g_1(\tau)$, neglecting the motion of the first and last scatterer relative to the source and detector, is accurate for $n > 20$.

(b) Continuous wave (cw) DAWS

Potential advantages

• monochromatic ultrasonic waves - can avoid possible complications if there is a strong frequency dependence of v_e and $l*$ because of dispersion.

• can measure faster dynamics (no delay due to the pulse repetition rate).

But $g_1(\tau)$ is more complicated for cw DAWS (need to include absorption and correct boundary conditions for diffusing sound):

$$g_1^{(cw)}(\tau) \simeq \int P(s) \exp\left[-\frac{s}{l*}k^2\left\langle\Delta r_{rel}^2(\tau)\right\rangle/6\right] ds$$

$P(s)$: fraction of diffusing sound transmitted for each path length s.

$$= \frac{(L+2h)\big/(z_0+h)\left\{\sinh\left(z_0\sqrt{q^2+\alpha^2}\right)+h\sqrt{q^2+\alpha^2}\cosh\left(z_0\sqrt{q^2+\alpha^2}\right)\right\}}{\left[1+h^2\left(q^2+\alpha^2\right)\right]\sinh\left(L\sqrt{q^2+\alpha^2}\right)+2h\sqrt{q^2+\alpha^2}\cosh\left(L\sqrt{q^2+\alpha^2}\right)}$$

where $q^2 = k^2\left\langle\Delta r_{rel}^2(\tau)\right\rangle/2l*^2$, $\alpha^2 = 1/D\tau_a$ and z_0 and h are the penetration depth and extrapolation length of diffusing sound.

Comparison of pulsed (open green symbols) and cw (closed red symbols) DAWS measurements of the relative mean square displacement at two different volume fractions.
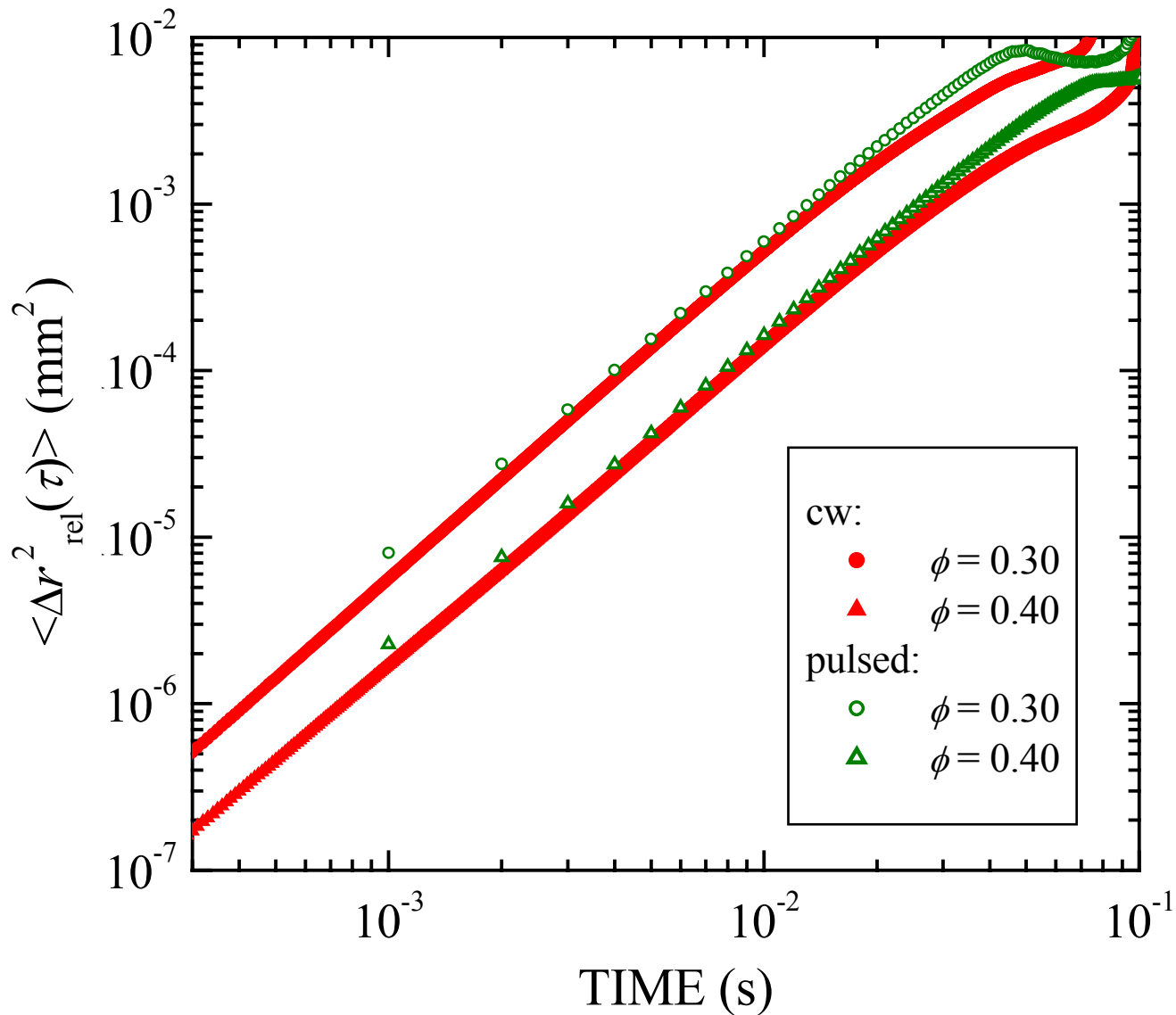
Some typical results for DAWS in fluidized suspensions.
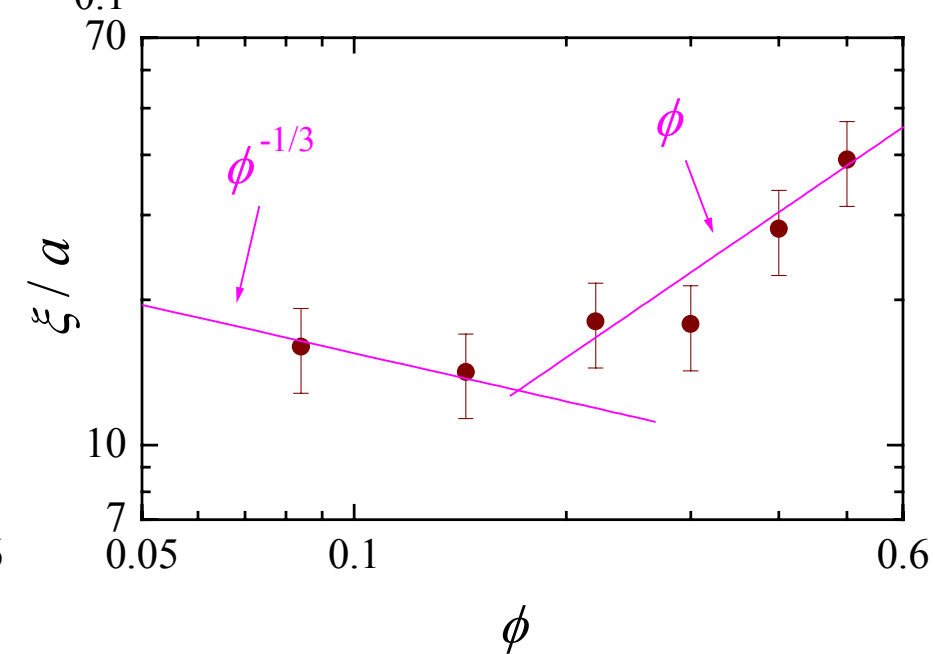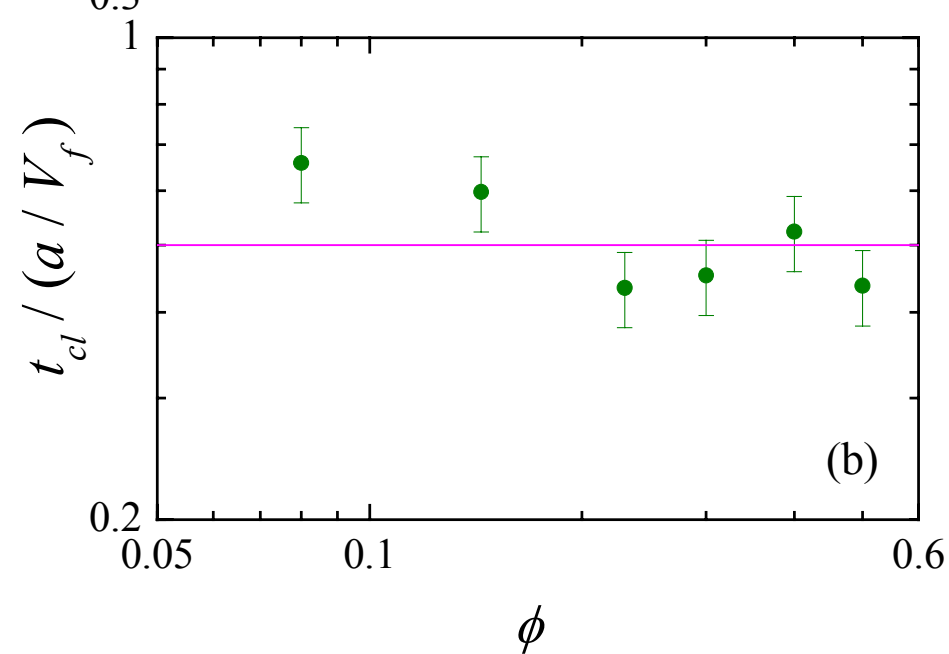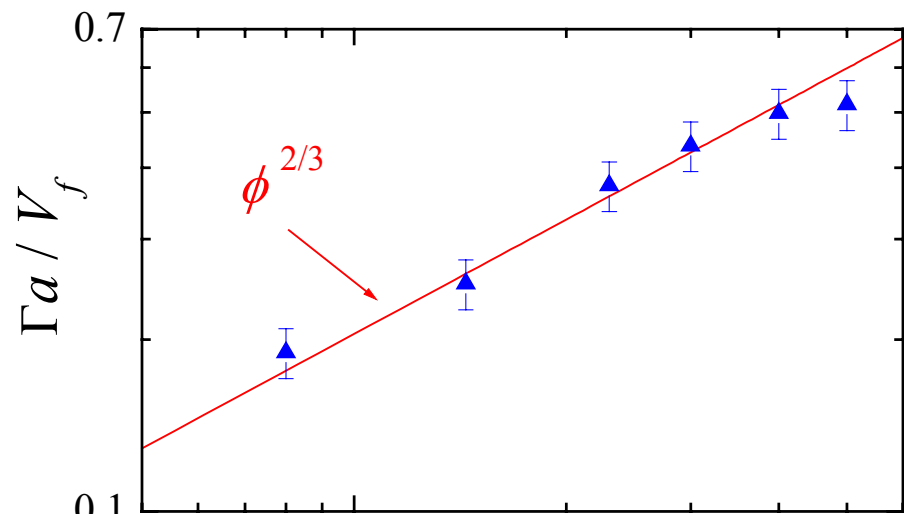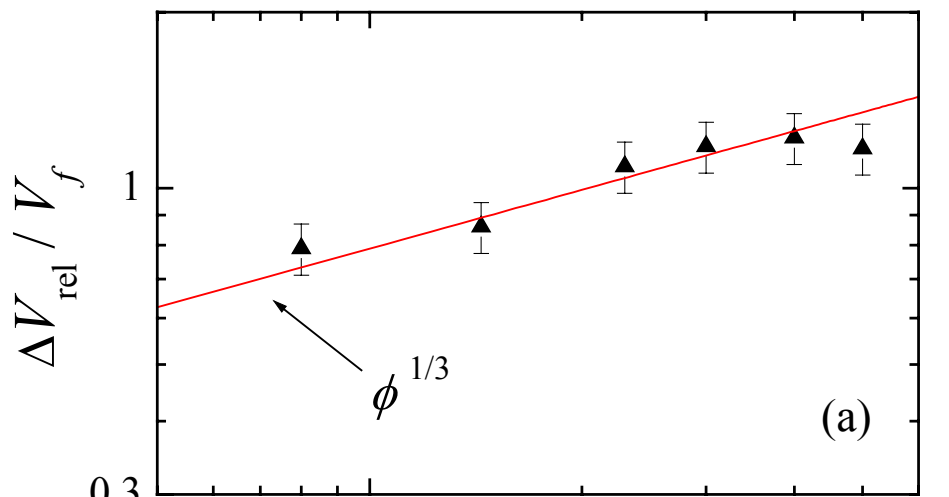
Summary: what field fluctuation spectroscopy measures in fluidized suspensions

DSS measures

$$V_{rms} = \sqrt{\left\langle \Delta V_{\vec{q}}^{\,2} \right\rangle}$$ the rms particle velocity along $\vec{q}$

$$d_c = V_{rms}\, t_c$$ the dynamic correlation length

DAWS measures

$$\Delta V_{rel} = \sqrt{\left\langle \Delta V_{rel}^2 \right\rangle}$$ local rms relative velocity fluctuations

$$\Delta d_{sep} = \Delta V_{rel}\, \tau_\Delta$$ Average change in particle separation before interactions change the local particle trajectories:

Combine DSS and DAWS to determine

$$\xi$$ instantaneous velocity correlation length

Question: What can we learn from the amplitude and phase fluctuations of multiply scattered ultrasonic waves?
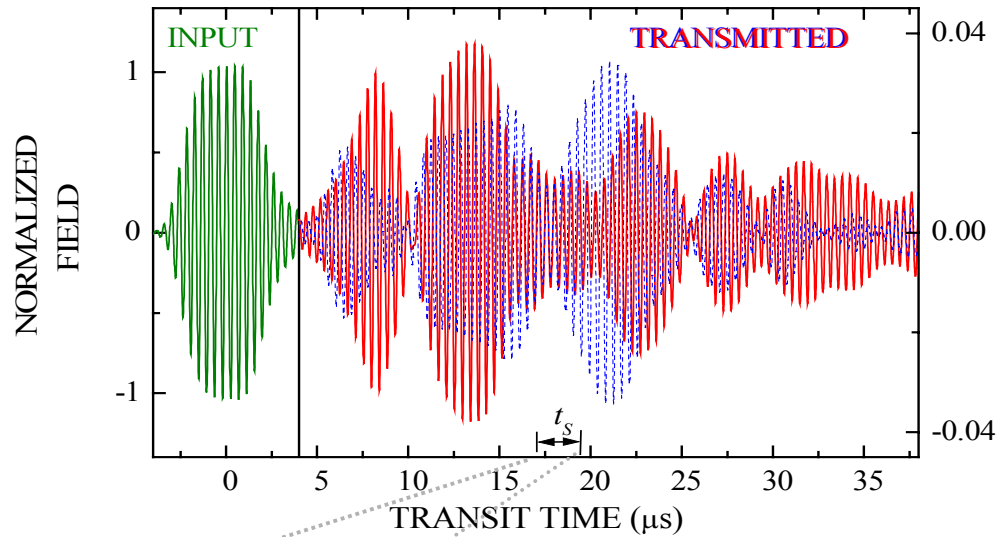
Experiment:

- Pulsed technique

- Record a short segment of the scattered wave for each input pulse. ⇒ Extract the phase and amplitude fluctuations.

FIELD · AMPLITUDE · PHASE

$\tau = 0$ s

$\tau = .75$ s

$\tau = 1.5$ s

TRANSIT TIME (µs)
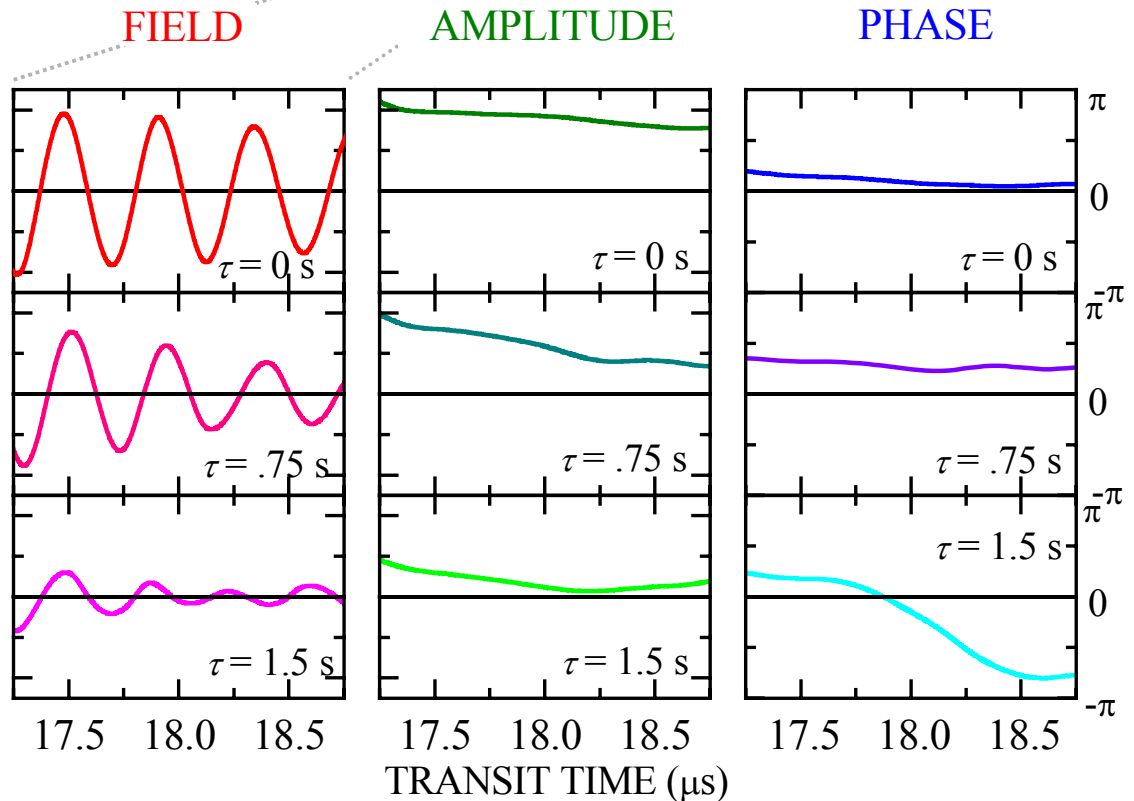
Amplitude and wrapped phase fluctuations

Our experiments measure the amplitude and the "wrapped phase" [- π : π], for multiple scattering paths of fixed length, as function of time.
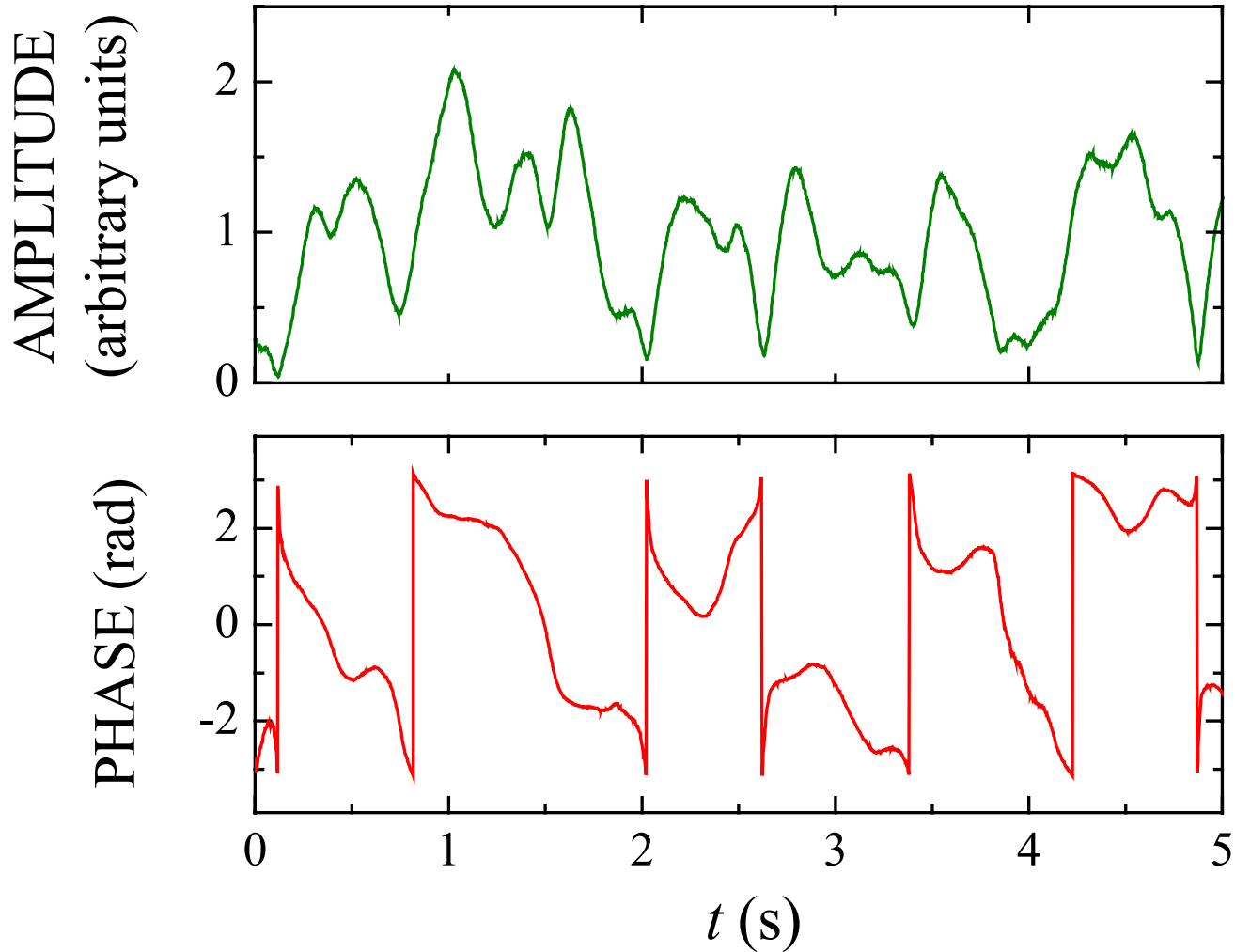
First-order speckle statistics (scattered field: complex Gaussian random variable)

[Goodman, J. Opt. Soc. Am. **66** 1145 (1976)]

Intensity:

$$P(I) = \frac{1}{I_{ave}} \exp\left(-\frac{I}{I_{ave}}\right)$$
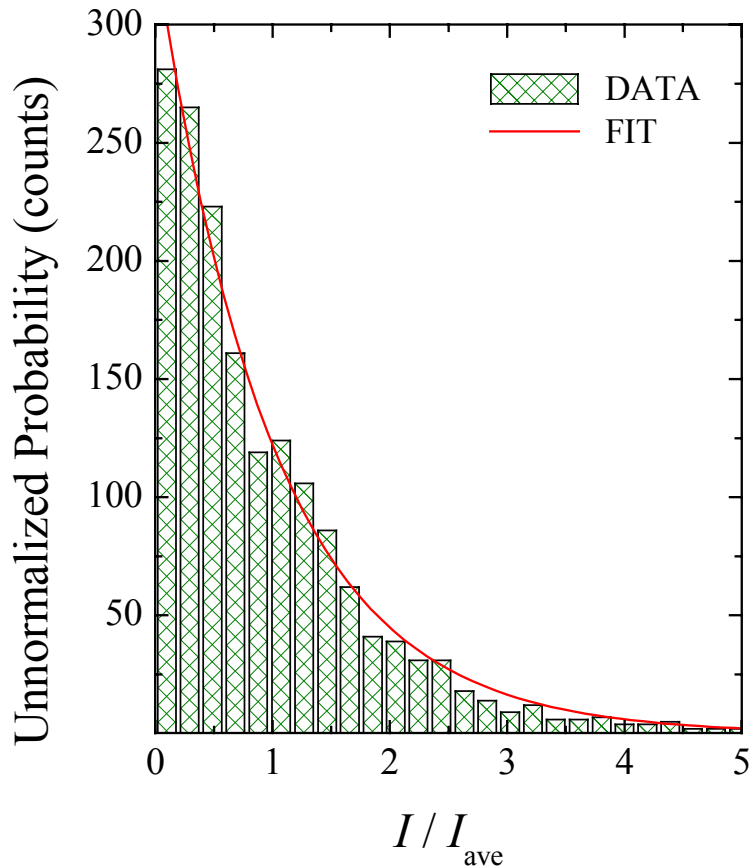
Phase:

$$P(\varphi) = \frac{1}{2\pi} \qquad -\pi < \varphi < \pi$$
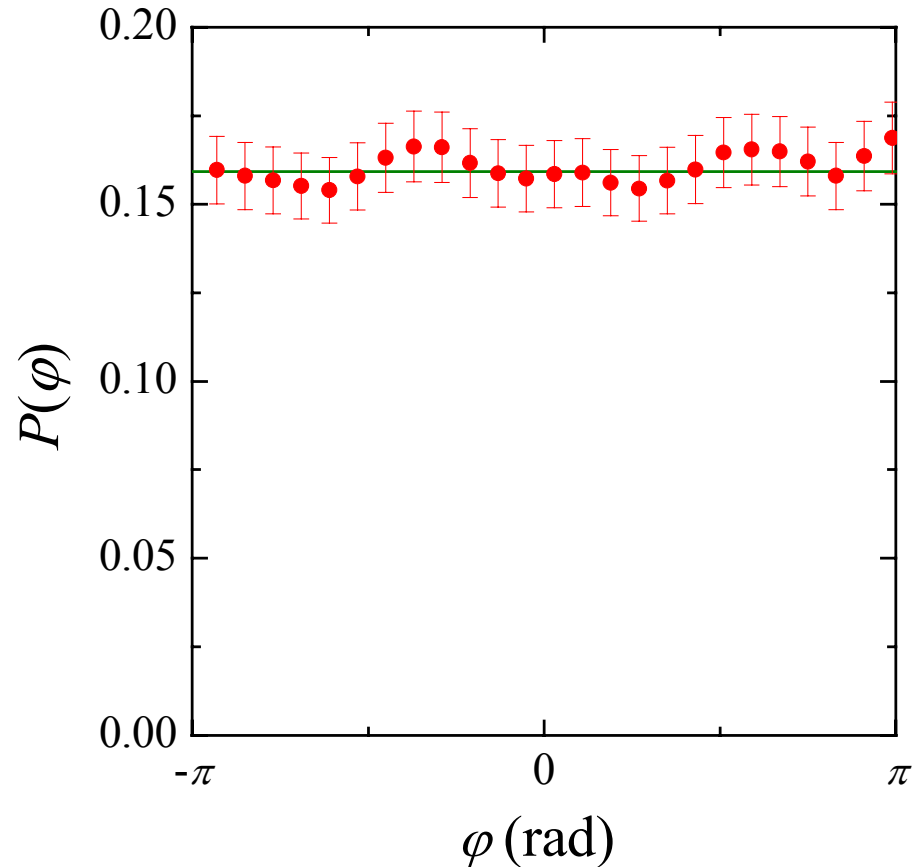
Phase information: The Wrapped Phase Difference Probability Distribution

Information on the particle dynamics is contained in the phase difference $\Delta\varphi(\tau) = \varphi(t+\tau) - \varphi(t)$.

The scattered ultrasonic field

$$\psi(t) = A_t e^{i\varphi_t} = \sum_{paths\ p} a_p(t) e^{i\phi_p(t)}$$

is a complex Gaussian random variable. (C_1 approximation.)

The statistics of the phase difference can be obtained from the joint probability distribution $P(\psi_t, \psi_{t'})$ of the fields at times t and $t' = t+\tau$ (for useful background, see Goodman, *Statistical Optics* (1985); van Tiggelen *et al.* Phys. Rev. E **59**, 7168 (1999)). For a complex Gaussian process, $P(\psi_t, \psi_{t'})$ can be written

$$P(\psi_t \psi_{t'}) = \frac{1}{\pi^2 \det \mathbf{C}} \exp\left(-\sum_{i,j=t,t'} \psi_i C_{ij}^{-1} \psi_j \right)$$

where $C_{ij} = \langle \psi_i \psi_j^* \rangle$ is the covariance matrix.

Normalize the fields so that $\langle \psi_i \, \psi_i^* \rangle = \langle |\psi(t)|^2 \rangle = 1$. Then C_{ij} is

$$C = \begin{pmatrix} 1 & g_1(\tau) \\ g_1(\tau) & 1 \end{pmatrix}$$

where $g_1 = \langle \psi_i \, \psi_j^* \rangle = \langle \psi(t) \, \psi^*(t') \rangle$ is the field autocorrelation function. Express $P(\psi_t, \psi_{t'})$ in terms of amplitude and phase:

$$P(A_t, A_{t'}, \varphi_t, \varphi_{t'}) = \frac{A_t A_{t'}}{\pi^2 (1 - g_1^2)} \exp\left\{ -\left[A_t^2 + A_{t'}^2 - 2 g_1 A_t A_{t'} \cos(\varphi_t - \varphi_{t'}) \right] / (1 - g_1^2) \right\}$$

Next, integrate out A_t, $A_{t'}$ and φ_t at constant $\Delta\varphi(\tau)$ to obtain the wrapped phase difference probability distribution:

$$P(\Delta\varphi) = \frac{2\pi - |\Delta\varphi|}{4\pi^2} \left[\frac{1 - g_1^2}{1 - g_1^2 \cos^2(\Delta\varphi)} \right] \left[1 + \frac{g_1 \cos(\Delta\varphi) \arccos\{ -g_1 \cos(\Delta\varphi) \}}{\sqrt{1 - g_1^2 \cos^2(\Delta\varphi)}} \right]$$

The wrapped phase difference probability distribution

$$P(\Delta\varphi) = \frac{2\pi - |\Delta\varphi|}{4\pi^2} \left[\frac{1 - g_1^2}{1 - g_1^2 \cos^2(\Delta\varphi)} \right] \left[1 + \frac{g_1 \cos(\Delta\varphi) \arccos\{-g_1 \cos(\Delta\varphi)\}}{\sqrt{1 - g_1^2 \cos^2(\Delta\varphi)}} \right]$$

gives information on the particle dynamics through its dependence on $g_1(\tau)$.

[Recall that for pulsed DAWS, $g_1 = \left\langle \exp\left[-i\Delta\phi_p(\tau)\right] \right\rangle \cong \exp\left[-\frac{1}{2}\left\langle \Delta\phi_p^2(\tau) \right\rangle\right]$

where $\left\langle \Delta\phi_p^2(\tau) \right\rangle \simeq n\, k^2 \left\langle \Delta r_{rel}^2(\tau) \right\rangle / 3$ is the average change in the phase of all paths containing n scattering events, over time τ.]

Short times ($\tau \to 0$ [small $\Delta\phi$]):

$$P(\Delta\varphi) \cong \frac{\frac{1}{2}\left\langle \Delta\phi_p^2(\tau) \right\rangle}{\left[\left\langle \Delta\phi_p^2(\tau) \right\rangle + \Delta\varphi^2\right]^{3/2}}$$

Power law
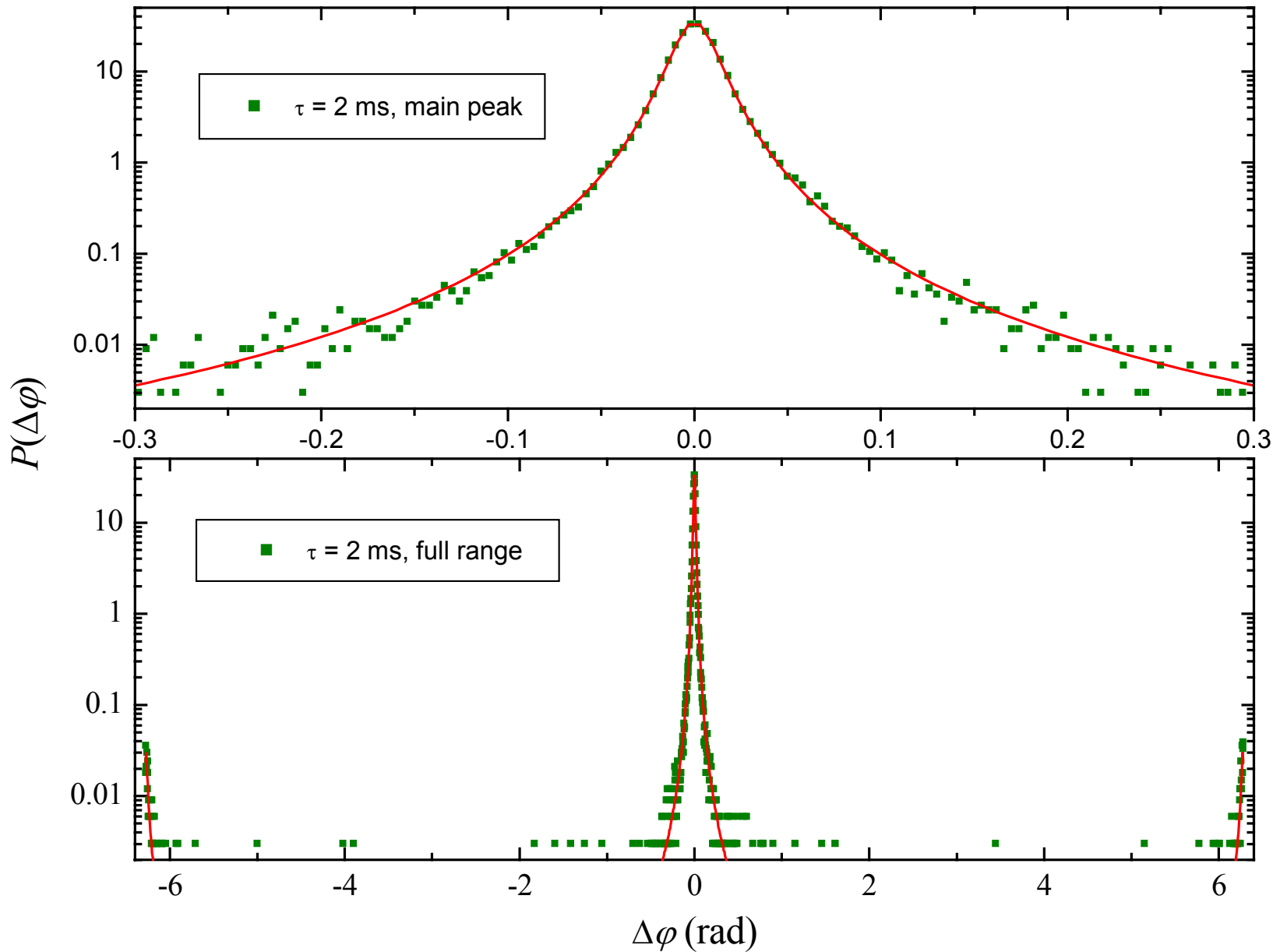- width gives $\left\langle \Delta\phi_p^2(\tau) \right\rangle$
- equivalent to $P\left(\frac{d\varphi}{dt}\right)$
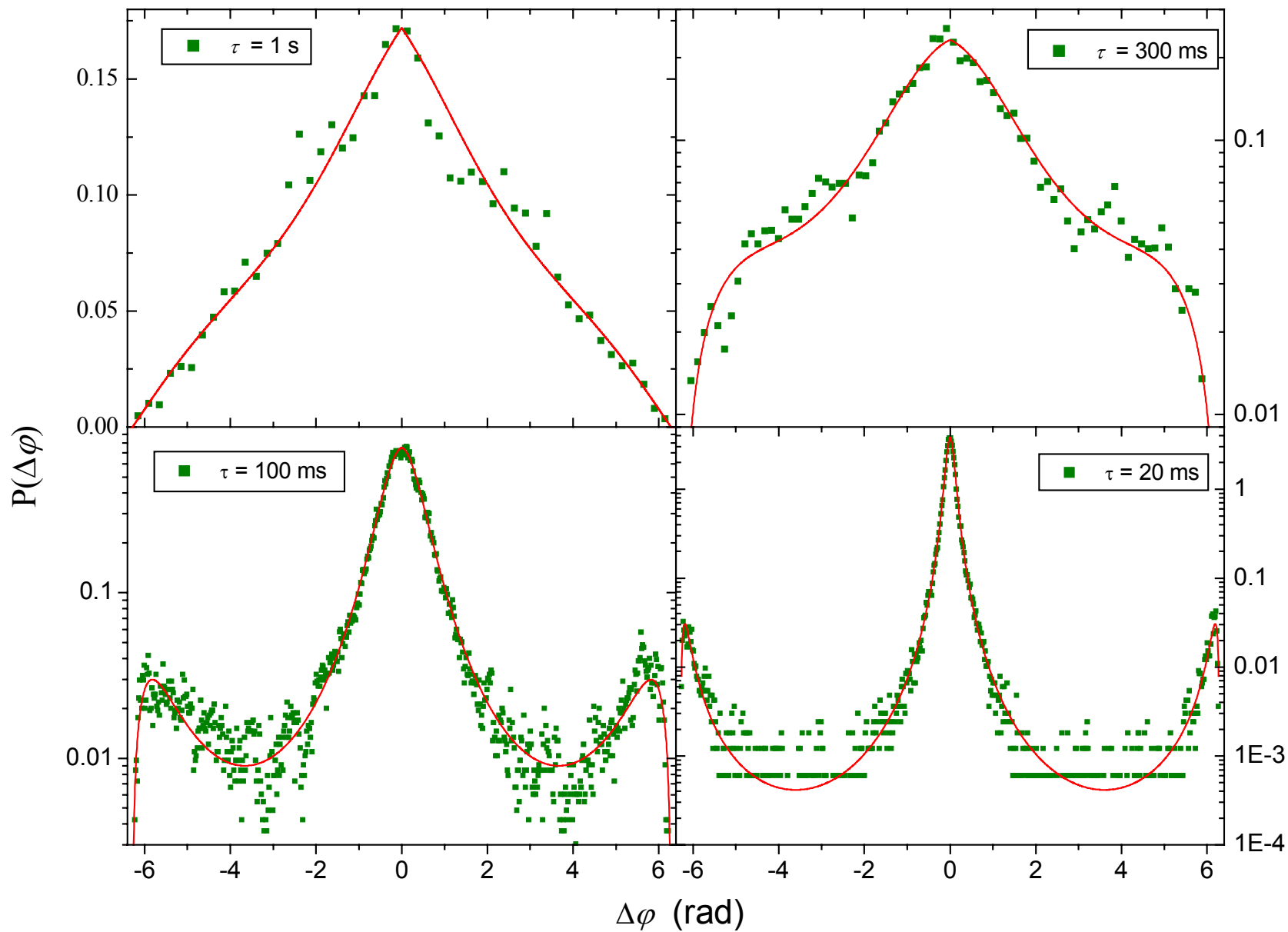
Long times ($\tau \to \infty$):

$$P(\Delta\phi) = \frac{2\pi - |\Delta\phi|}{4\pi^2}$$

Triangle function - no information on the particle dynamics

Wrapped phase difference probability distribution at short times τ

Wrapped phase difference probability distribution as τ increases

Relationship between the wrapped phase moment $\langle \Delta \varphi^2 \rangle$ and the particle motion.

From $P(\Delta \varphi)$, calculate numerically $\langle \Delta \phi_p^2 \rangle$ (the variance of the phase change for paths p of length n) as a function of $\langle \Delta \varphi_{wrap}^2 \rangle$ (the variance of the wrapped phase).
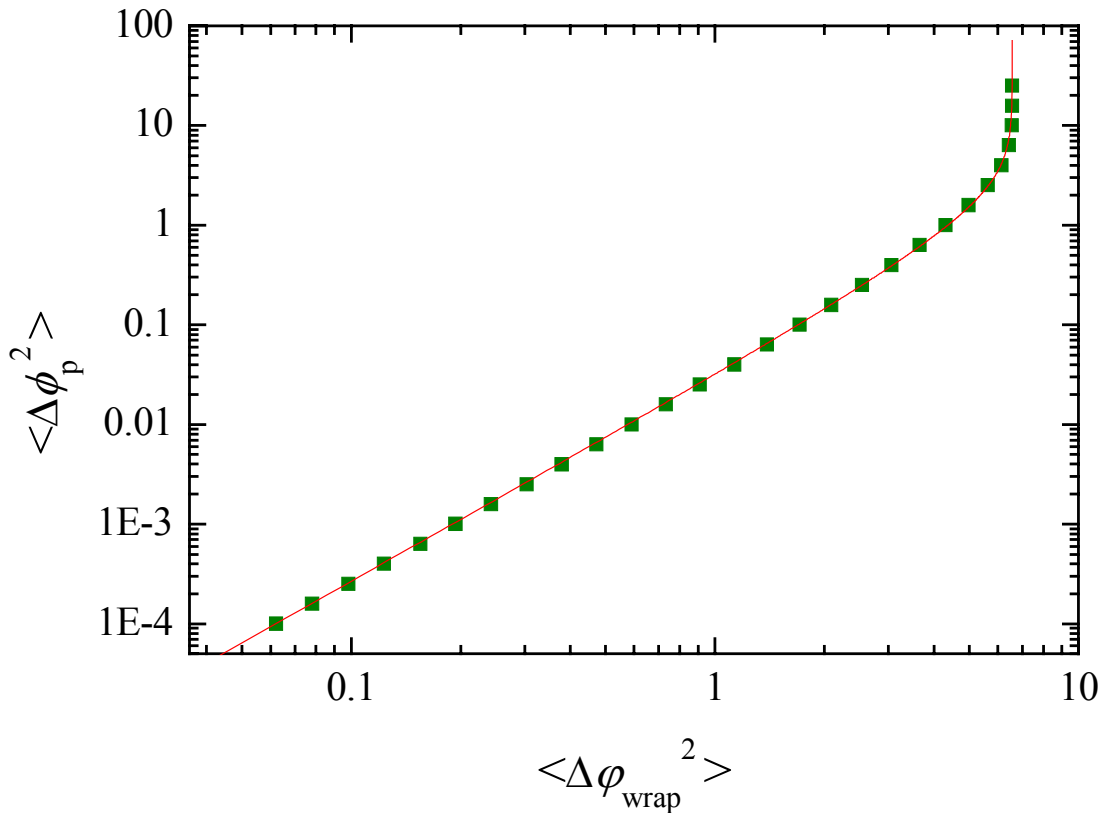
Allows the relative motion of the scattering particles, $\langle \Delta r_{rel}(\tau)^2 \rangle$ to be determined directly from the wrapped phase moment $\langle \Delta \varphi_{wrap}^2 \rangle$.

Particle dynamics from the temporal evolution of the phase difference $\Delta\phi(\tau)$
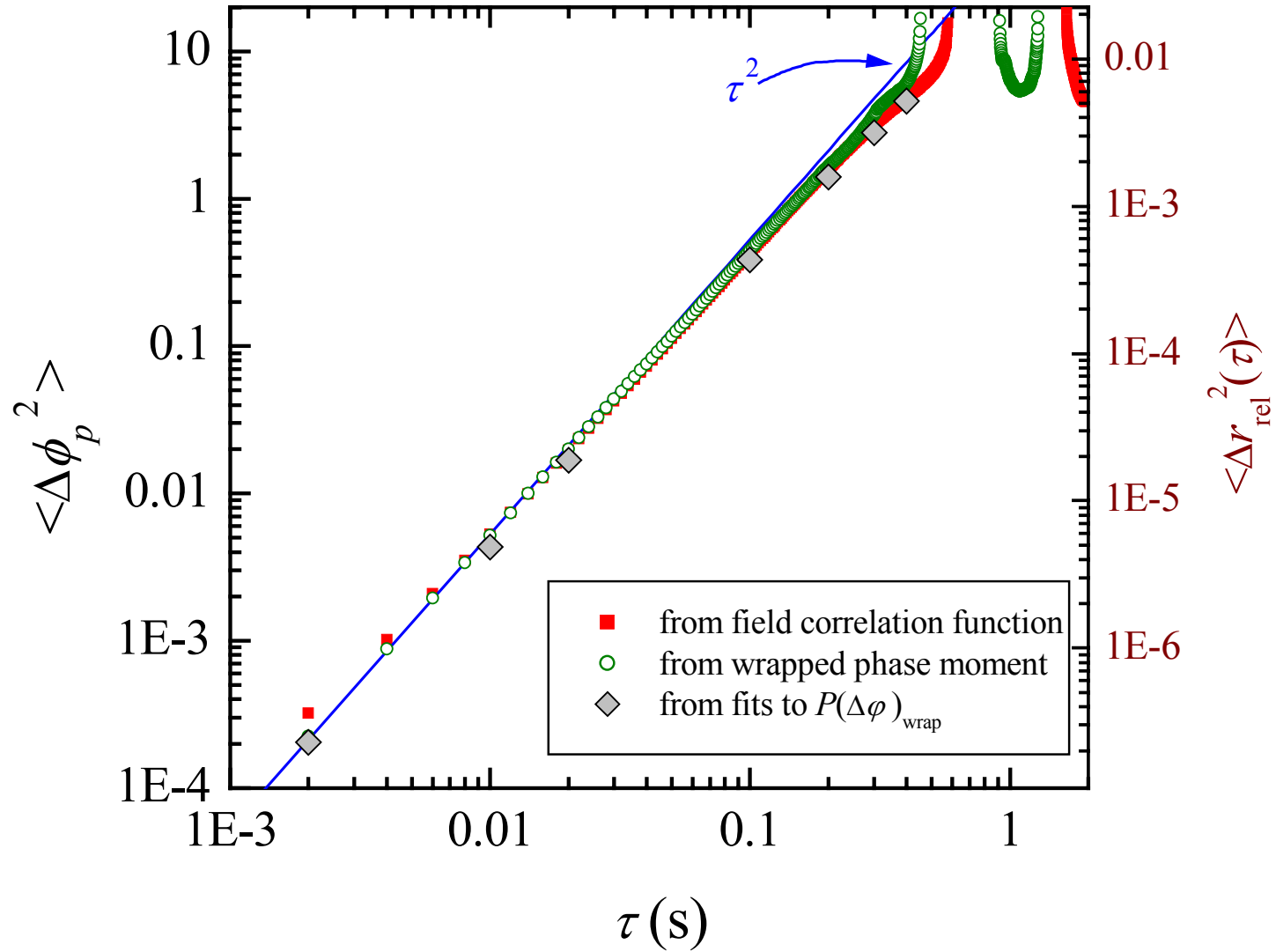
Excellent agreement with measurements of the relative mean square displacement $\langle\Delta r_{rel}^2(\tau)\rangle$ from the field correlation function in DAWS

Cumulative phase

- By unwrapping the phase, the jumps of 2π can be removed, giving the cumulative phase.

- The cumulative phase allows the phase changes to be followed for longer times.
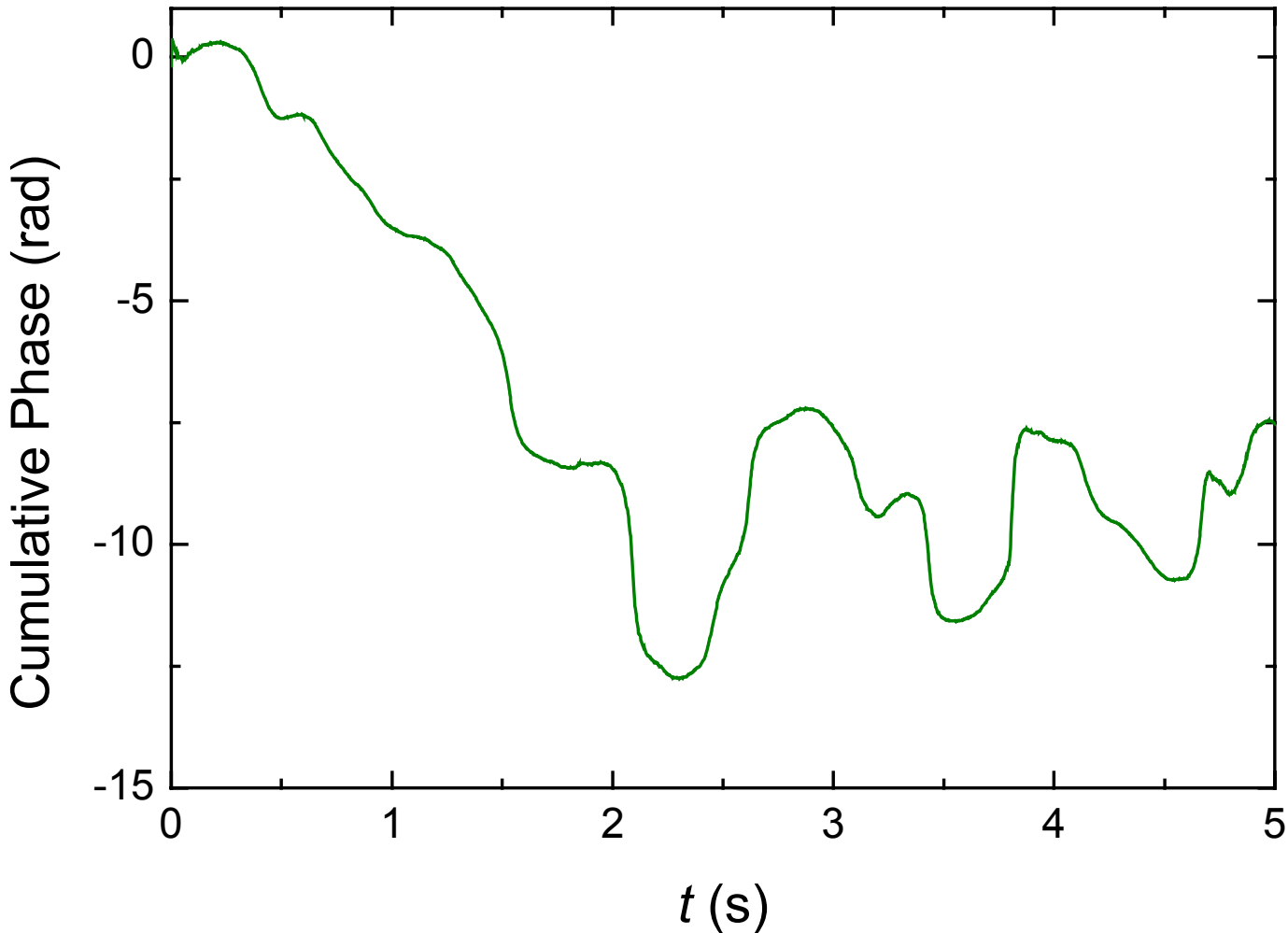
The wrapped and cumulative phase difference probability distributions.

For large τ,   wrapped phase: $P(\Delta\phi) \rightarrow$ triangle function
cumulative phase: $P(\Delta\phi) \rightarrow$ Gaussian
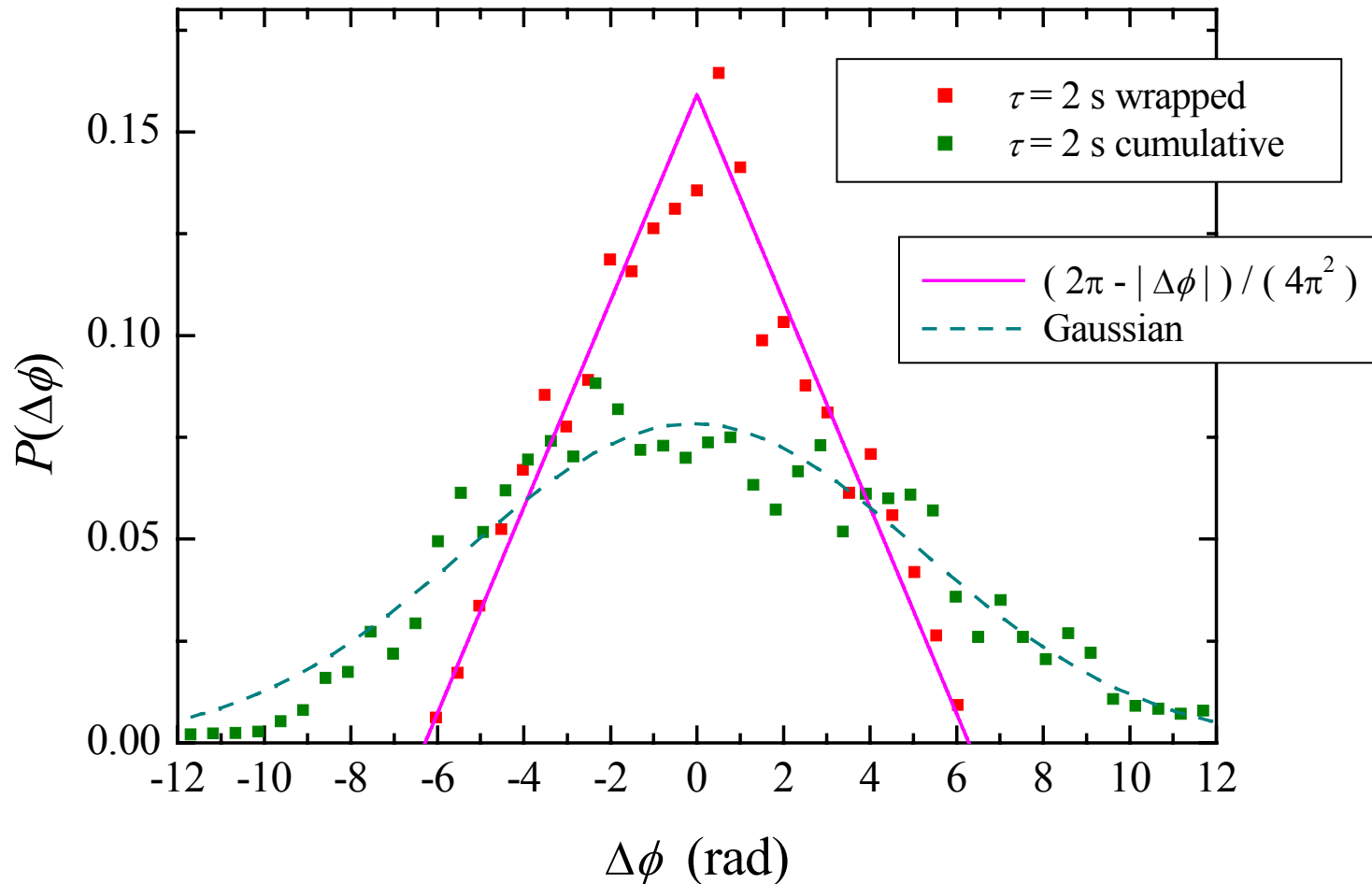
Can additional information on the dynamics be obtained from the cumulative phase?

Summary - phase statistics

Experiments and theory for the temporal fluctuations of the phase of multiply scattered ultrasonic waves in fluidized suspensions:

• a beautiful example of mesoscopic wave phenomena - excellent agreement between theory and experiment for the wrapped phase difference probability distribution $P(\Delta\phi, \tau)$.

• an alternative approach for investigating the dynamic behaviour of multiply scattering systems - excellent agreement with "traditional" Diffusing Acoustic Wave Spectroscopy (DAWS).

Amplitude information $\rightarrow$ measure simultaneously the scattered intensity and field to investigate the validity of the Siegert Relation.

Determine the intensity correlation function from the amplitude fluctuations, $[I(t) \propto A^2(t)]$:

$$G_2(\tau) = \frac{\langle I(t)I(t+\tau)\rangle}{\langle I(t)\rangle^2} = 1 + \beta g_2(\tau)$$

Siegert relation:

$$g_2(\tau) = \left| g_1(\tau) \right|^2$$

• relates the intensity correlation function to the field correlation function

• valid for complex random Gaussian fields

• often used in light scattering to relate measurements of G_2 to theory for g_1

Question: Does the Siegert relation break down when the motions of the scatterers are correlated?

cw DAWS: Siegert relation is valid

AUTOCORRELATION FUNCTIONS vs TIME (s)

$\phi = 0.245$
■ $|g_1|^2$
○ g_2 / β
$\phi = 0.50$
▲ $|g_1|^2$
▽ g_2 / β
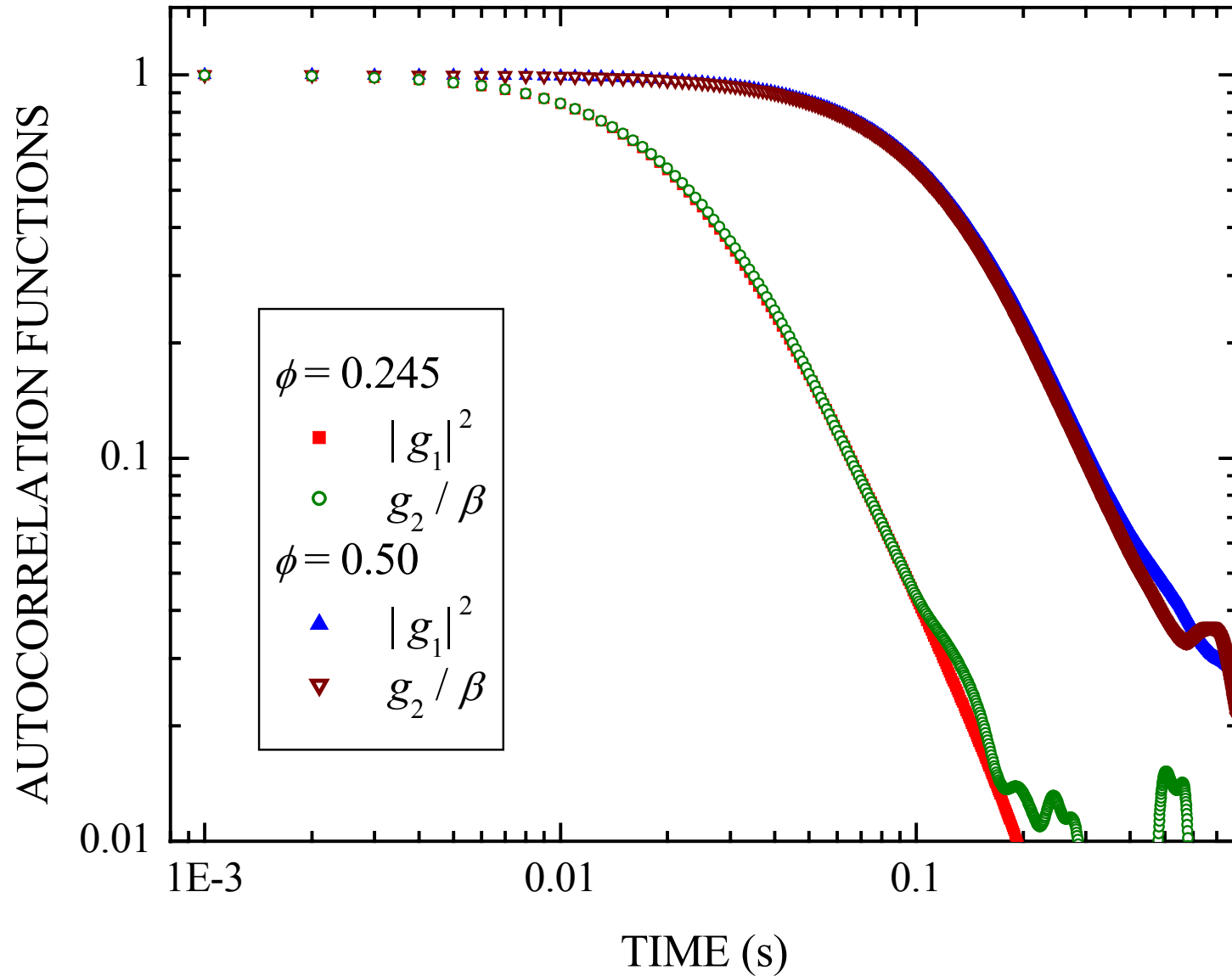
Pulsed DAWS: Siegert relation breaks down for short paths!
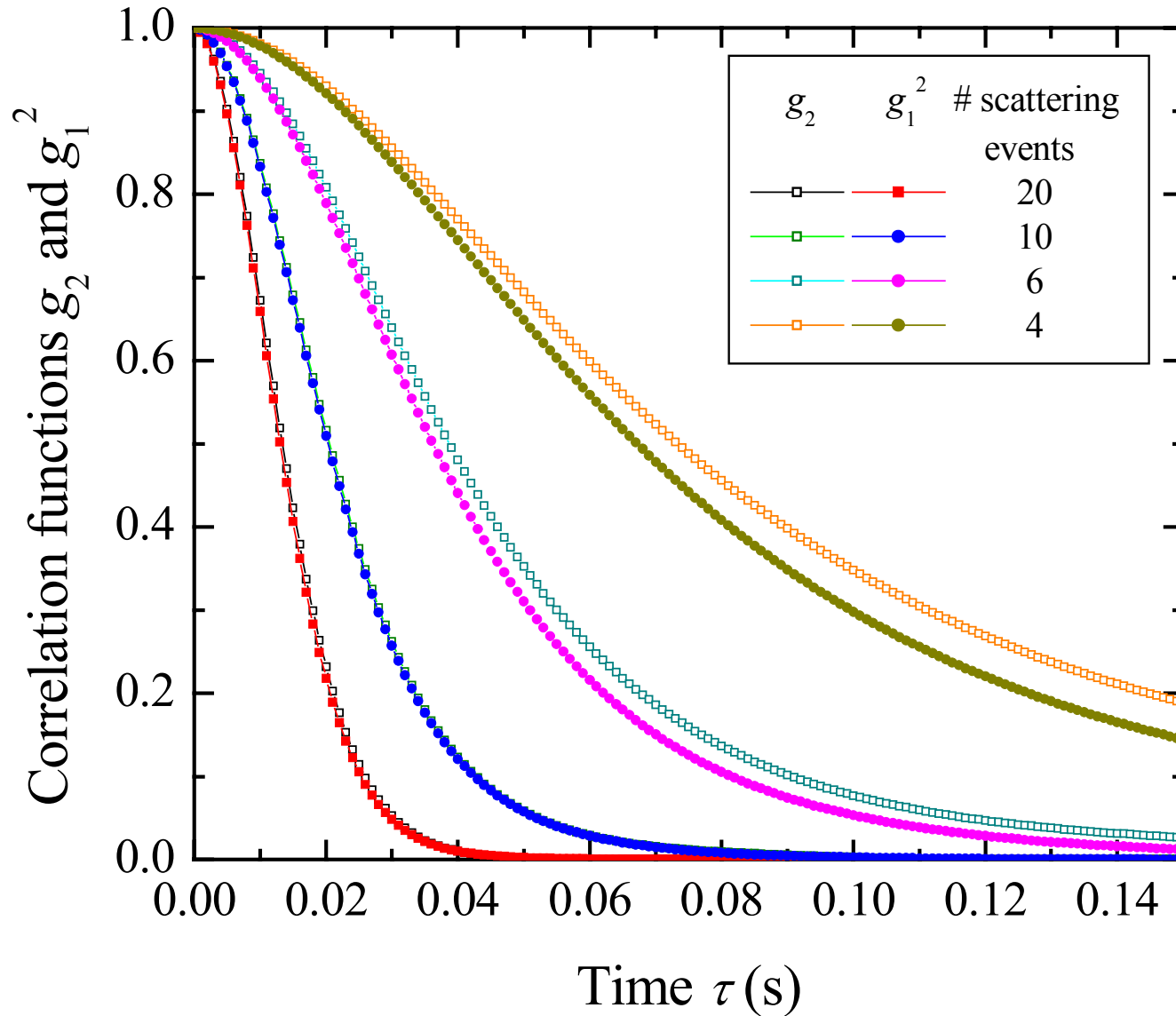
Pulsed DAWS: Deviation from the Siegert relation as a function of path length - significant for $n < 10$.

• Motion of the particles in a path containing 10 steps is strongly correlated:

Particle velocity correlation volume
$500\ (\phi=.25) \leq \xi^3 \leq 5000\ (\phi=.5)$ mm^3

Typical volume probed by diffusing sound for 10 scattering events
$130\ (\phi=.25)$ to $17\ (\phi=.5)$ mm^3

$\Rightarrow$ Siegert relation breaks down for short paths

• Siegert relation is valid for $n > 10$ because the phases of multiply scattered waves along different scattering paths are no longer correlated for $n > 10$.
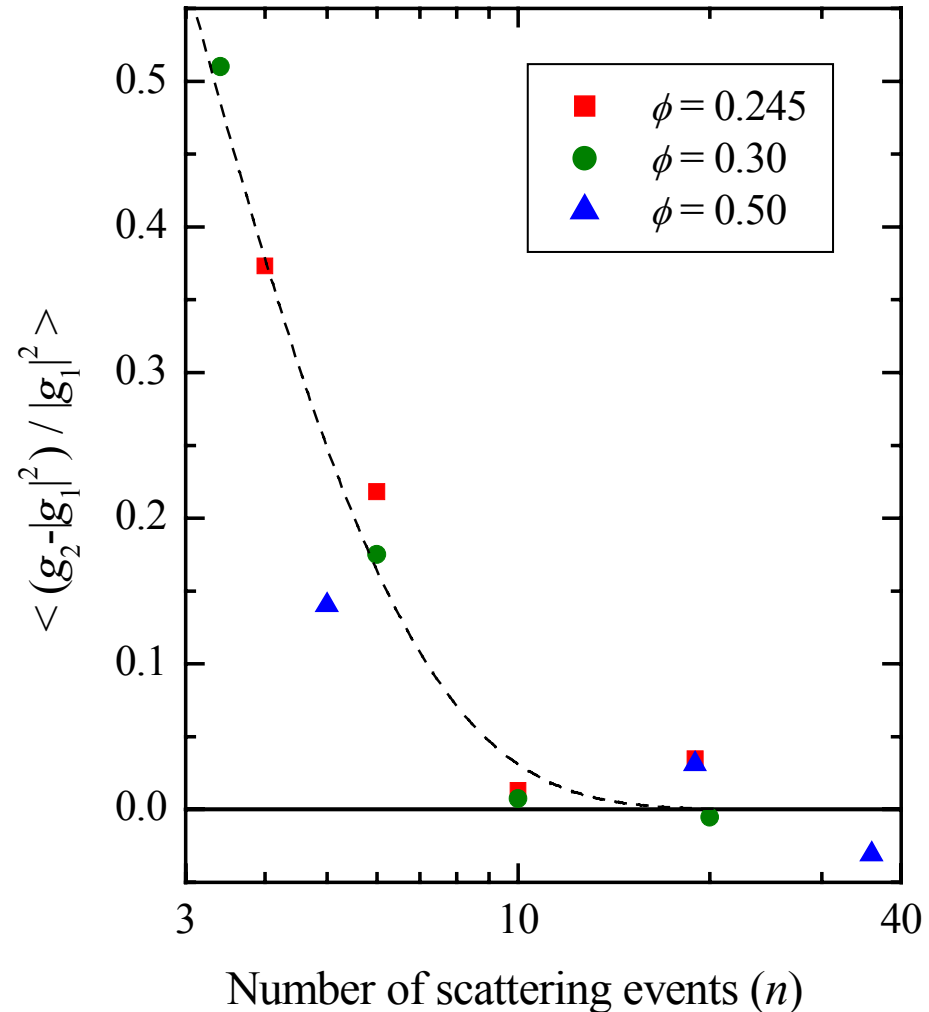
Origin of the breakdown of the Siegert relation for short paths - examine single scattering (DSS)

$$G_2(\tau) = \frac{\langle \psi(t)\psi^*(t)\psi(t+\tau)\psi^*(t+\tau) \rangle}{\langle \psi(t)\psi^*(t) \rangle^2}$$

$$= \frac{\left\langle \sum_{i,j,k,l} \exp\left[i\{\phi_i(t) - \phi_j(t) + \phi_k(t+\tau) - \phi_l(t+\tau)\}\right] \right\rangle}{\left\langle \sum_{i,j} \exp\left[i\{\phi_i(t) - \phi_j(t)\}\right] \right\rangle^2}$$

Random particle positions, scattered phases from different particles are uncorrelated:

only terms $i{=}j$ with $k{=}l$ and $i{=}l$ with $j{=}k$ survive

only $i{=}j$ terms survive

Hence

$$g_2(\tau) \equiv (G_2(\tau) - 1)/\beta = \int_V h(R)\exp\left[-\frac{q^2}{2}\langle \Delta r_{rel,\vec{q}}^2(\tau, R) \rangle_t\right] dR$$

where $h(R)$ gives the fraction of particles in scattering volume separated by R.

Bonus: compare $g_2(\tau)$ and $|g_1(\tau)|^2$ to measure the spatial-temporal correlation function

$$C(\tau, R) = \frac{\langle \Delta r_{\vec{q}}(\tau, \vec{x}) \Delta r_{\vec{q}}(\tau, \vec{x} + \vec{R}) \rangle}{\langle \Delta r_{\vec{q}}^2(\tau, \vec{x}) \rangle}$$

$\Rightarrow$ Can measure $C(\tau, R)$ on longer length scales, determined by the scattering volume, than are accessible to DAWS.

Breakdown of the Siegert relation in single scattering (DSS)
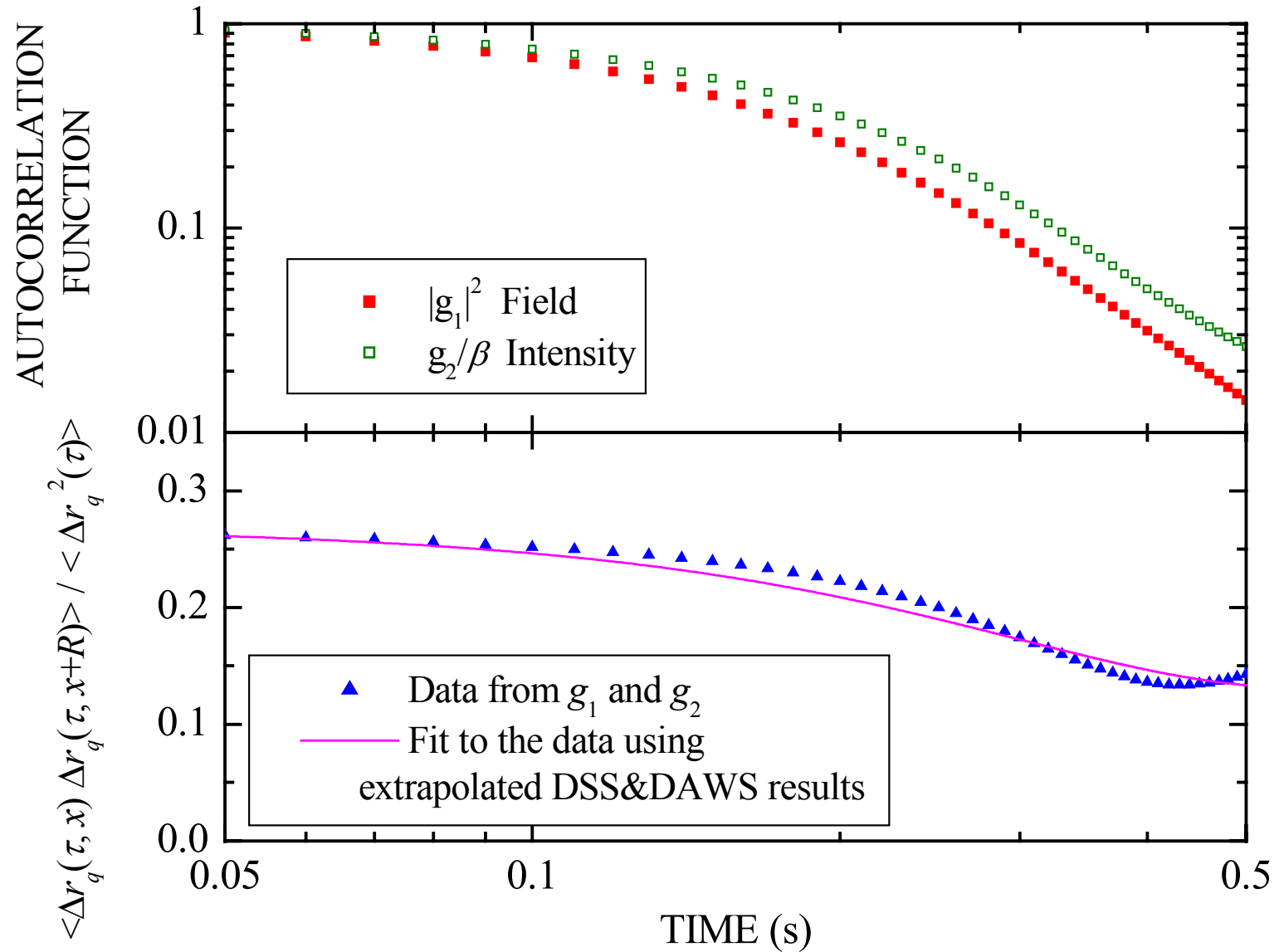
Results for the dynamics of fluidized suspensions.

Volume fraction dependence at Re = 0.9.

Volume Fraction Dependence - Blob Model

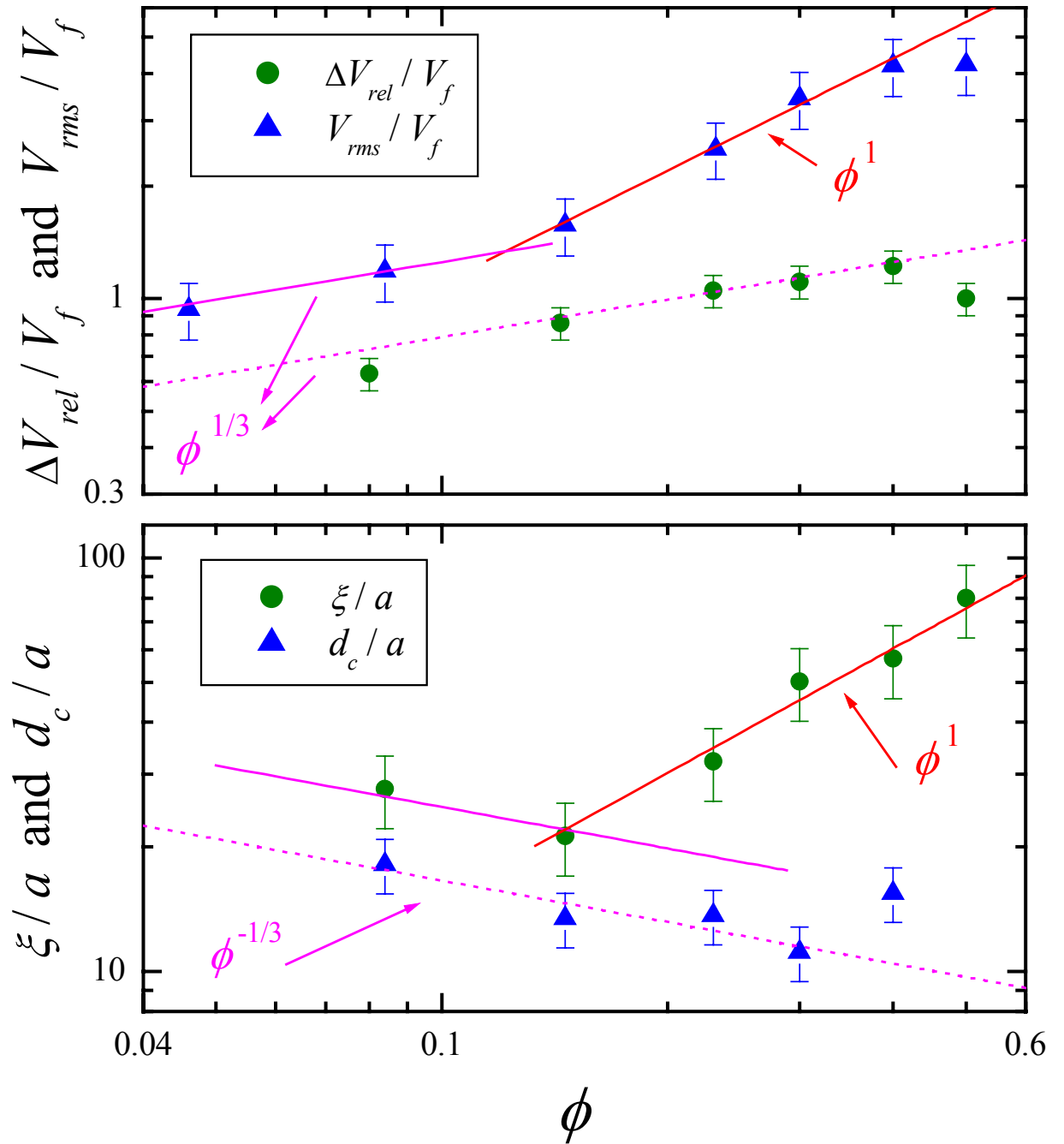Consider a "blob" with N particles in a volume $V \sim \xi^3$, with a deficit (or surplus) of ΔN particles.

- balance buoyant force and viscous drag on the "blob":

$$\frac{V_{rms}}{V_f}\sqrt{\frac{a}{\xi}} \simeq \phi^{1/2}\left[\frac{\eta(0)}{\eta(\phi)}\right]\left[\frac{\Delta N_\xi}{\sqrt{\langle N_\xi \rangle}}\right]\left[\frac{V_o}{V_f(\phi)}\right]$$

- η = viscosity of the suspension
   $\Delta N = N^{1/2}$ for random particle positions.

Experiments of Lei *et al*., PRL 2001 : number fluctuations suppressed at long length scales, cut off V_{rms}.
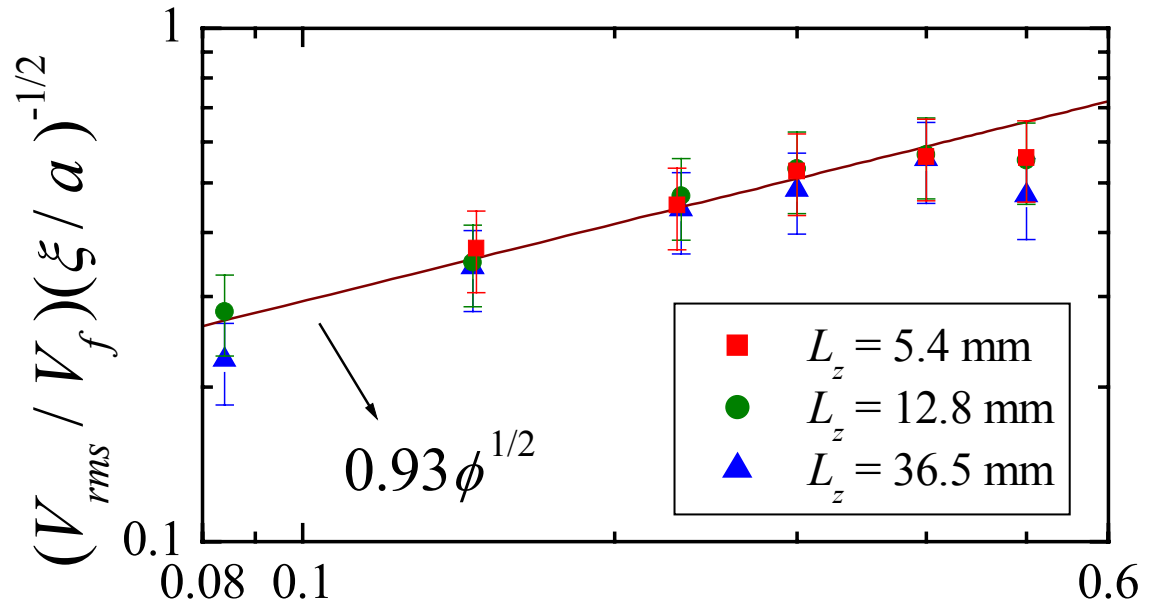
Segrè *et al*., Nature (2001): number fluctuations also suppressed at high ϕ by volume exclusion effects.

Question: Do number fluctuations set the correlation length?

Blob model scaling

$$\frac{V_{rms}}{V_f}\sqrt{\frac{a}{\xi}}$$

$$\simeq \phi^{1/2}\left[\frac{\eta(0)}{\eta(\phi)}\right]\left[\frac{\Delta N_\xi}{\sqrt{\langle N_\xi\rangle}}\right]\left[\frac{V_o}{V_f(\phi)}\right]$$

Estimate the viscosity:

$$\frac{\eta(\phi)}{\eta(0)}=\left[1-\frac{\phi}{0.63}\right]^{-2}$$

$\Rightarrow$ Suppression of number fluctuations is constant in the correlation volume.

$(V_{rms}/V_f)(\xi/a)^{-1/2}$ vs ϕ

$0.93\phi^{1/2}$

- $L_z = 5.4$ mm
- $L_z = 12.8$ mm
- $L_z = 36.5$ mm

$\Delta N_\xi/\langle N\rangle_\xi^{1/2}$ vs ϕ

Extrapolation of the $\phi = 0.004$ measurement [Lei et al. 2001]

Prediction for a random distribution of Brownian spheres
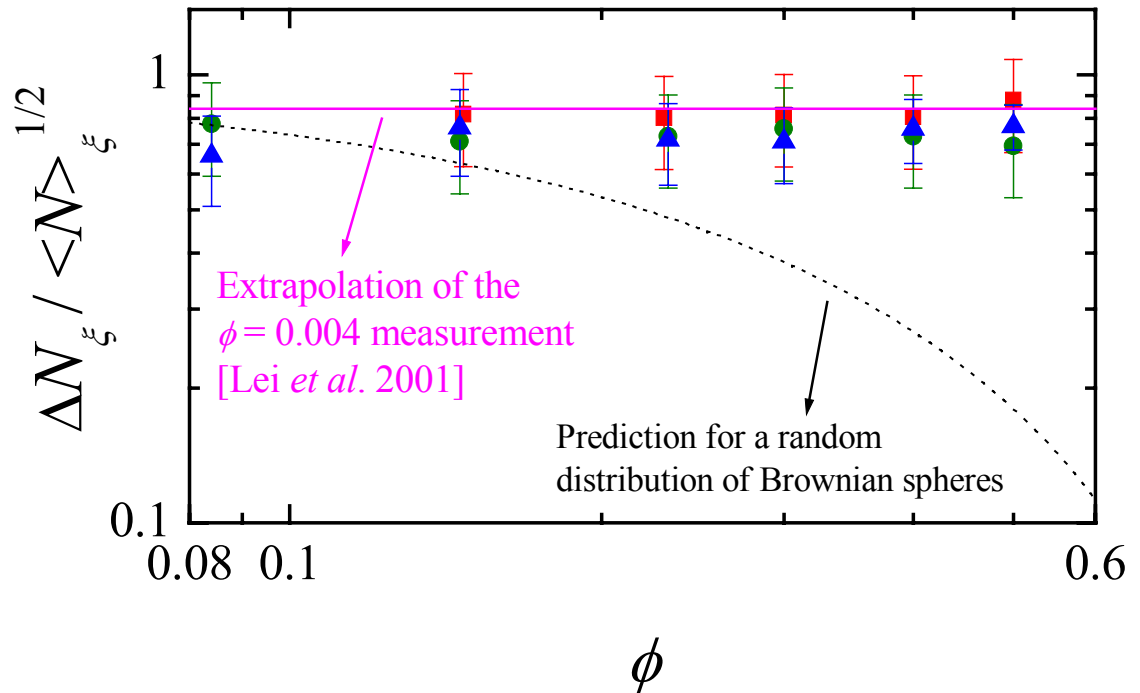
Sample Size Dependence at $Re_p = 1$

Vary only the smallest cell dimension (L_z)

RMS Velocity Components (at $\phi = 0.08$)

• weak dependence on thickness (not $L_z^{1/2}$)

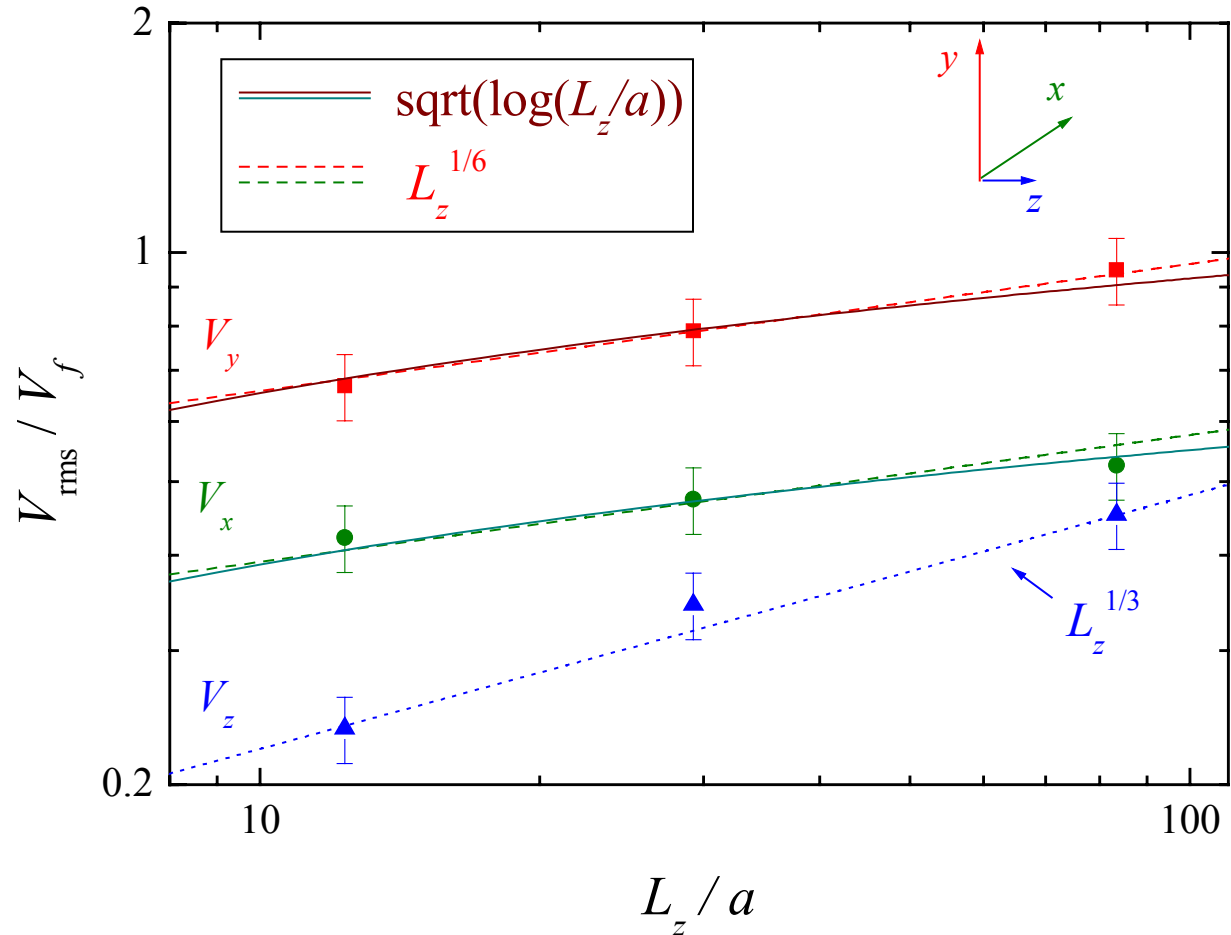$$V_y / V_f \sim \sqrt{\log\left(L_z/a\right)}$$

• variation is greatest along the smallest dimension

Legend:
— sqrt(log(L_z/a))
- - - $L_z^{1/6}$

V_y, V_x, V_z, $L_z^{1/3}$

Axes: V_{rms} / V_f versus L_z / a

Comparison with experiments at low Re$_p$

1.04 $L_z^{1/6}$

1.5sqrt(log(L_z/a))

$[V_y / V_f]/\phi^{1/3}$

L_z/a

■ Our data (Re$_p \sim 1$; $\phi \sim 0.1$)
○ From Nicolai & Guazzelli [1995]
and Nicolai *et al.* [1995] (Re$_p \sim 10^{-3}$; $\phi \sim 0.05$)
✳ From Segre *et al.* [1997] (Re$_p \sim 10^{-4}$; $0.0001 < \phi < 0.05$)
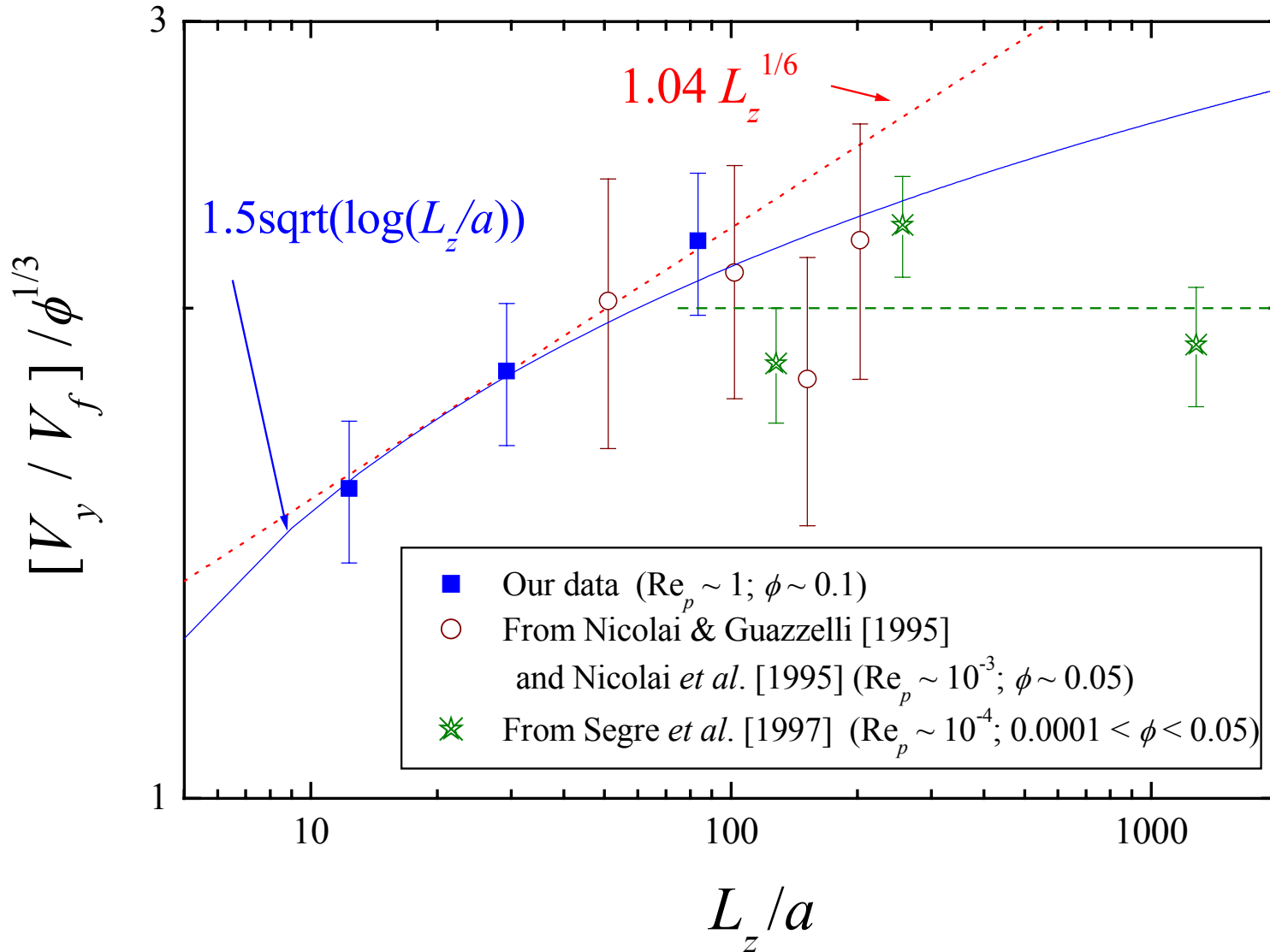
Local fluctuation picture

Question:

Do local fluctuations set the magnitude of V_{rms} ?

Our data indicate:

$\Delta V_{rel}(r_{nn})$ is independent of L_z , while V_{rms} and ξ are not.

$\Delta V_{rel}(r) \propto r^{1/2}$ until the divergence is cut off at ξ (by wall, inertial or other intrinsic effects).

But

$$V_{rms} \approx \Delta V_{rel}\left(r_{nn}\right) \frac{1}{\sqrt{2}} \sqrt{\frac{\xi}{r_{nn}}}$$
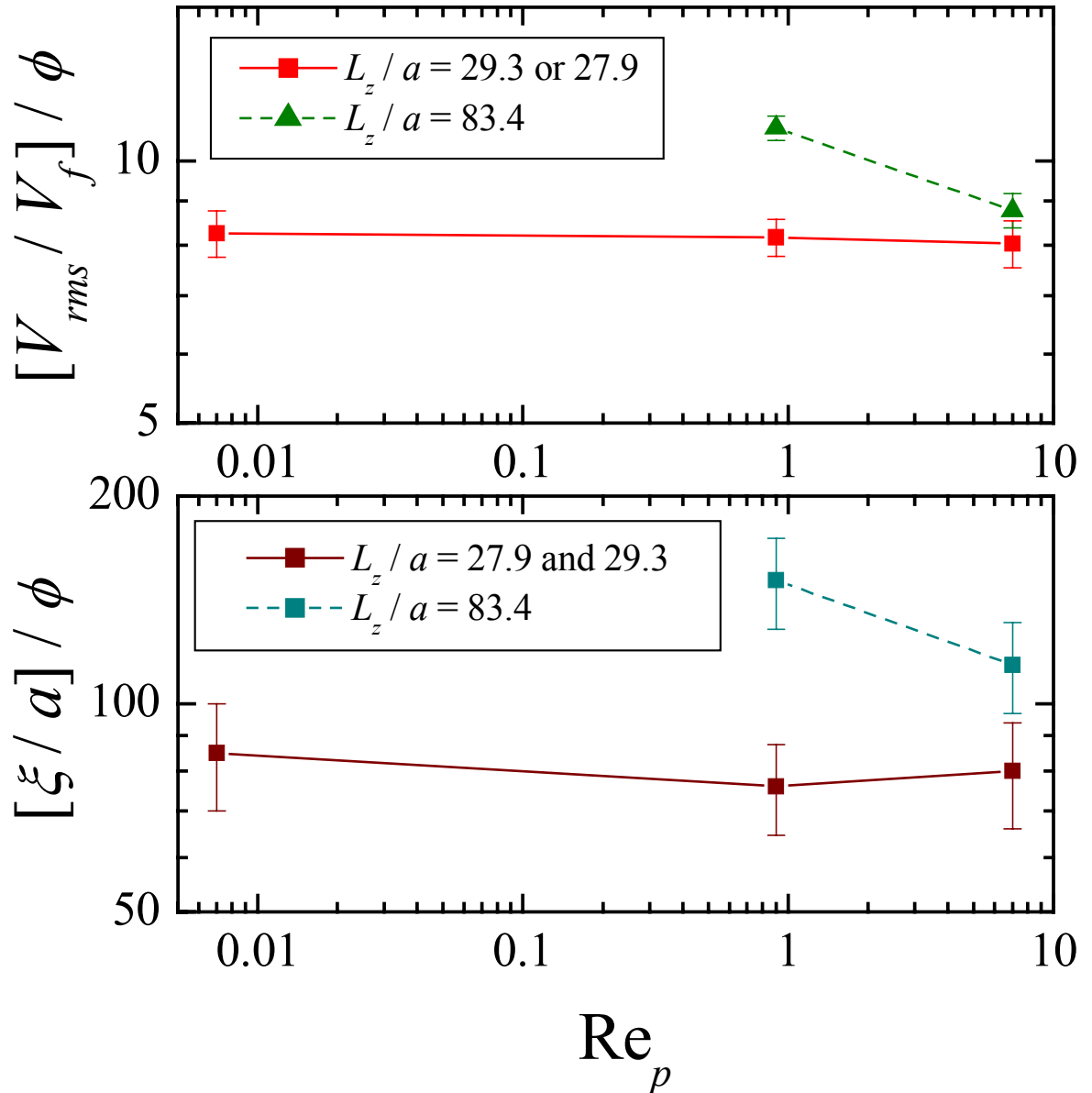
Suggests

$\Delta V_{rel}(r_{nn})$ may be the fundamental quantity that sets $V_{rms} / \sqrt{\xi}$.

Reynolds Number Dependence

• For thin samples, both V_{rms}, ξ are independent of Reynolds number up to $\text{Re}_p = 7$. (Wall effects dominate.)

• In our thickest sample, inertial screening decreases V_{rms}, ξ at $\text{Re}_p = 7$. ($\xi_{\text{screening}} < \xi_{\text{walls}}$)

Conclusions

I have described two new ultrasonic techniques :

• Dynamic Sound Scattering (DSS)

• Diffusing Acoustic Wave Spectroscopy (DAWS)

$\Rightarrow$ powerful new approaches for investigating the dynamics of strongly scattering media where direct imaging fails.

e.g. in fluidized suspensions:

DSS measures $\begin{cases} V_{rms} & \text{rms particle velocity (all 3 components)} \\ d_c & \text{dynamic correlation length} \end{cases}$

DAWS measures $\begin{cases} \Delta V_{rel} & \text{local relative velocity fluctuations} \\ \Delta d_{sep} & \text{local dynamic correlation length} \end{cases}$

Combine DAWS and DSS to determine the instantaneous velocity correlation length ξ..

Some applications of DSS and DAWS...

- fundamental studies of particle dynamics

e.g. understanding the large velocity fluctuations in fluidized beds -
dependence on volume fraction ϕ, system size L and Reynolds number Re:

- surprisingly large increase in V_{rms} and ξ at high ϕ - our data suggest that suppression of number fluctuations is important.

- Weak dependence of V_{rms} and ξ on the smallest cell dimension at $Re_p \sim 1$. (Walls cut off ξ and hence V_{rms} (via the Oseen wake?); ΔV_{rel} is unaffected.)

- Inertial screening of V_{rms} and ξ only seen at $Re_p = 7$ in our thickest sample.

- fundamental studies of wave phenomena in strongly scattering media.

e.g. Phase statistics of temporally fluctuating multiply scattered fields
Breakdown of the Siegert relation due to particle velocity correlations

- new applications in the nondestructive evaluation of strongly scattering media

e.g. monitoring fish in a cavity (de Rosny & Fink)
measuring velocity changes with temperature (Weaver, Gret)
seismic monitoring (CWI - Colorado group; Paris-Grenoble group)
process control...