Two-time correlations and coherent scattering experiments on phase-segregating materials

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Speckle intensity fluctuation spectroscopy

- Used for decades with coherent light scattering
- Commonly used to study equilibrium fluctuations
- Coherent X-rays from synchrotron sources
  $\Rightarrow$ higher penetration, finer resolution than visible light
- X-ray Intensity Fluctuation Spectroscopy (XIFS)
  recently extended to nonequilibrium systems
- Nonequilibrium speckle fluctuations are not stationary
- Fluctuation statistics reveal spatial structure and dynamics

This work

G. Brown, P.A. Rikvold, M. Sutton, and M. Grant,
Phys. Rev. E **56**, 6601 (1997); **60**, 5151 (1999)

- Numerically solved Time-Dependent Ginzburg-Landau (TDGL) equation for order-parameter field $\psi(\mathbf{r},\tau)$

$$\frac{\partial \psi(\mathbf{r},\tau)}{\partial \tau} = \left\{-\frac{1}{2}\nabla^2\right\}^{\alpha} \left[\left(1+\nabla^2\right)\psi(\mathbf{r},\tau) - \psi^3(\mathbf{r},\tau)\right]$$

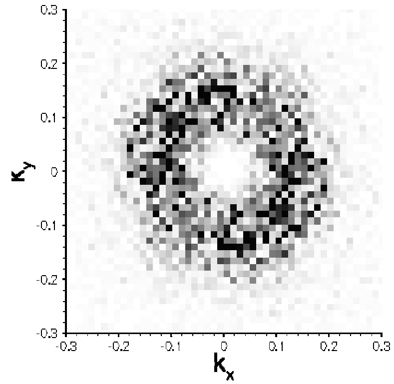
  $\alpha = 0$ for nonconserved; $\alpha = 1$ for conserved order parameter
  Dynamic scaling: Characteristic length $R(\tau) \sim \tau^n$
  $\Leftrightarrow$ Characteristic wave vector $k_c(\tau) \sim \tau^{-n}$
  $n = \frac{1}{2}$ for nonconserved; $n = \frac{1}{3}$ for conserved order parameter

- Developed analytic scaling function for intensity correlations

- Compare with recent experiments on phase separation in borosilicate glass (Malik et al.) and Al-Li alloy (Livet et al.)

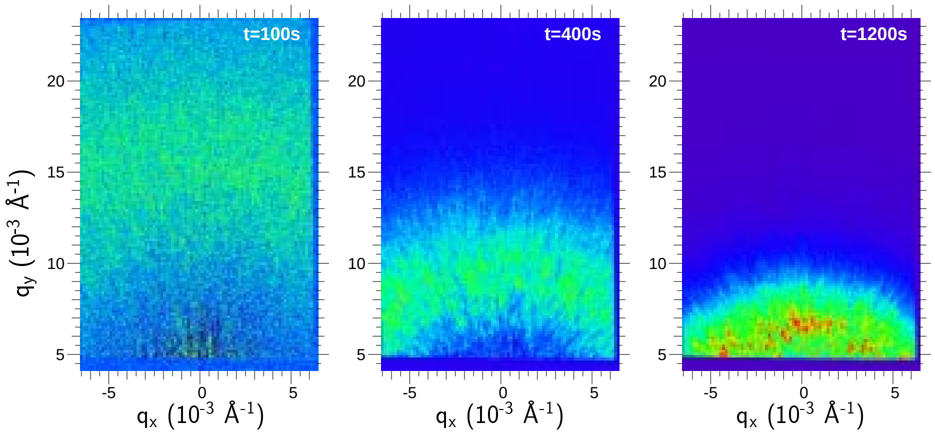
Scattering speckle

Simulated scattering intensity, conserved order parameter



Experimental scattering intensity, Sodium Borosilicate Glass

A. Malik et al., *Phys. Rev. Lett.* **81**, 5832 (1998)



Structure factor and intensity fluctuations

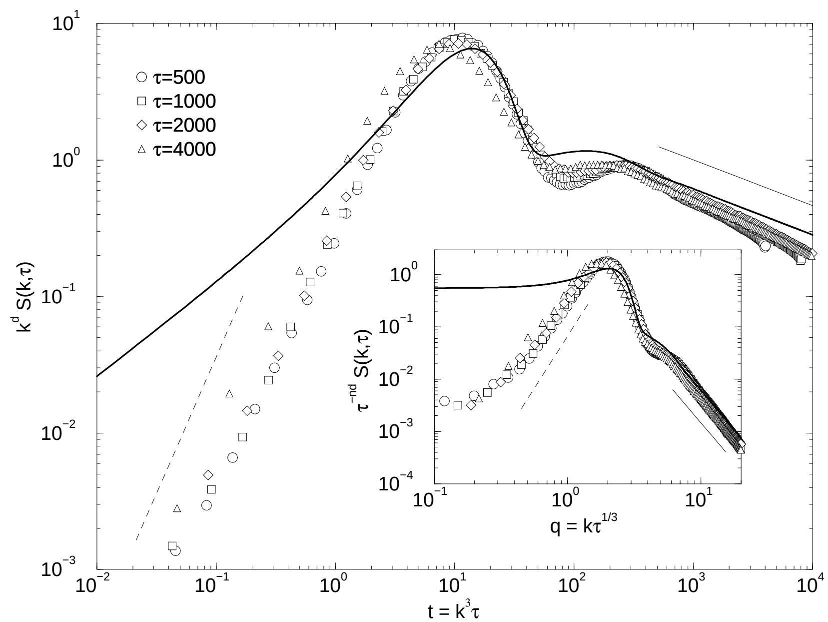
Structure factor: $S(\mathbf{k},\tau) = \langle I(\mathbf{k},\tau)\rangle$

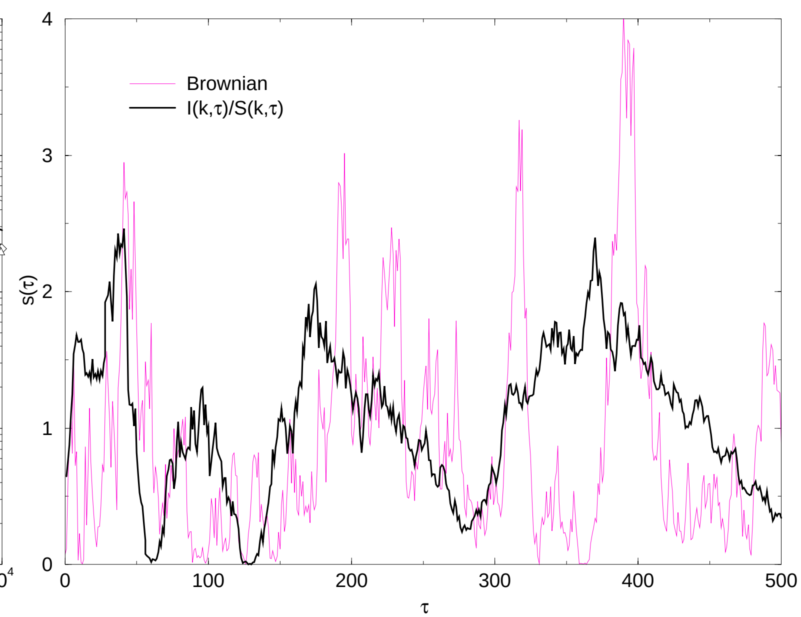
where $I(\mathbf{k},\tau) = |\hat{\psi}(\mathbf{k},\tau)|^2$ is fluctuating intensity at $(\mathbf{k},\tau)$

Characteristic wave vector $k_c \propto R(\tau)^{-1} \sim \tau^{-1/3}$

($n = 1/3$ for conserved order parameter)

Simulation results (Brown et al., PRE **60**, 5151 (1999))





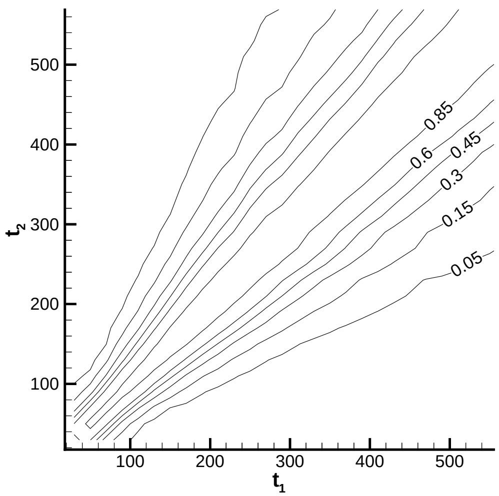
Scaled structure factor vs $\bar{t} \propto k^{1/n}\bar{\tau}$ Normalized speckle time series

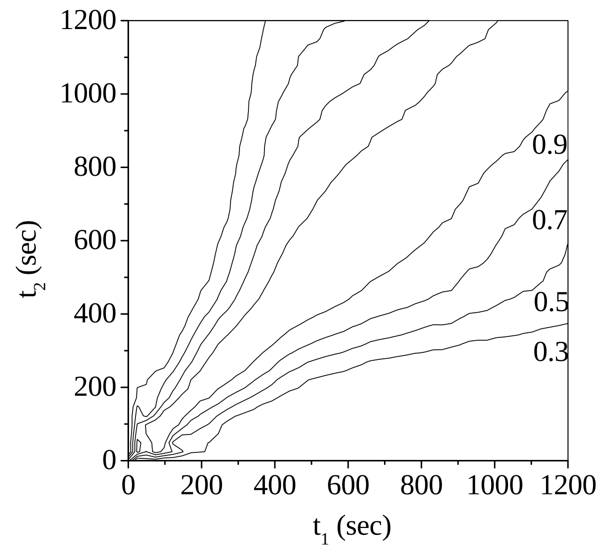
Intensity correlations

Normalized two-time intensity correlation function

$$\mathrm{Corr}(k;\tau_1,\tau_2) = \mathrm{Corr}(t_1,t_2) = \frac{\Big\langle I(\mathbf{k},\tau_1)I(\mathbf{k},\tau_2)\Big\rangle}{\Big\langle I(\mathbf{k},\tau_1)\Big\rangle\Big\langle I(\mathbf{k},\tau_2)\Big\rangle} - 1 \quad ; t_i \propto k^{1/n}\tau_i$$

Natural variables: $\delta\tau = \tau_2 - \tau_1$ and $\bar{\tau} = (\tau_2 + \tau_1)/2$





Simulated $\mathrm{Corr}(t_1,t_2)$ Experimental $\mathrm{Corr}(\tau_1,\tau_2)$ (Malik et al.)

Scaling function for Corr(t_1, t_2)

If $\langle I(\mathbf{k},\tau_1) I(\mathbf{k},\tau_2)\rangle = \langle \hat{\psi}(\mathbf{k},\tau_1)\hat{\psi}^*(\mathbf{k},\tau_1)\hat{\psi}(\mathbf{k},\tau_2)\hat{\psi}^*(\mathbf{k},\tau_2)\rangle$
breaks down into products of second moments
(Gaussian Superposition Approximation), then

$$\mathrm{Corr}(t_1, t_2) \propto \left[\int d\mathbf{r}\, e^{i\mathbf{k}\cdot\mathbf{r}} \langle \psi(\mathbf{0},\tau_1)\psi(\mathbf{r},\tau_2)\rangle\right]^2$$

Scaling for SMALL $\bar{t} \propto k^{1/n}\bar{\tau}$

In this limit, Corr(t_1, t_2) is dominated by the large-scale behavior:

Scaling with $r/R(\bar{\tau})$ and τ_2/τ_1 implies
$\langle \psi(\mathbf{0},\tau_1)\psi(\mathbf{r},\tau_2)\rangle = C\left(r/\bar{\tau}^n, \delta\tau/\bar{\tau}\right)$
To lowest order in $\bar{t}$, the Fourier transform gives

$$\mathrm{Corr}(t_1, t_2) = \mathrm{Corr}\left(\frac{\delta t}{\bar{t}}\right) \quad \text{for small } \bar{t}$$

Scaling for LARGE $\bar{t} \propto k^{1/n}\bar{\tau}$

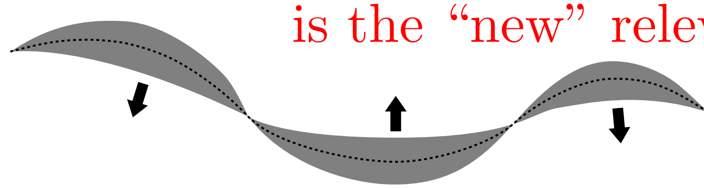
Many terms in the $\bar{t}$-expansion would contribute.

In this limit, $\mathrm{Corr}(t_1, t_2)$ is dominated by the small-scale behavior:

$$\frac{\langle \psi(\mathbf{0}, \tau_1)\psi(\mathbf{r}, \tau_2)\rangle}{\langle \psi^2 \rangle} = 1 - \frac{\delta\tau}{\bar{\tau}} G\left(\mathrm{const.}\frac{r}{\Delta R}\right)$$

$$G(x) \sim \begin{cases} 1 & \text{for } x \ll 1 \\ x & \text{for } x \gg 1 \end{cases} \quad \text{and} \quad \Delta R = \delta\tau \left.\frac{\mathrm{d}R(\tau)}{\mathrm{d}\tau}\right|_{\tau=\bar{\tau}} \propto \frac{\delta\tau}{\bar{\tau}^{(1-n)}}$$

is the "new" relevant length scale

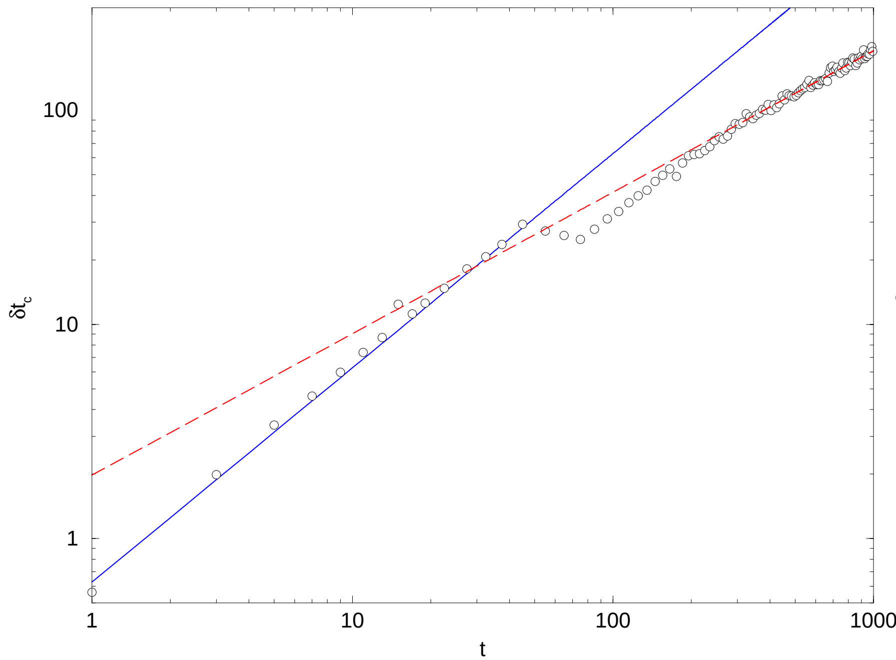


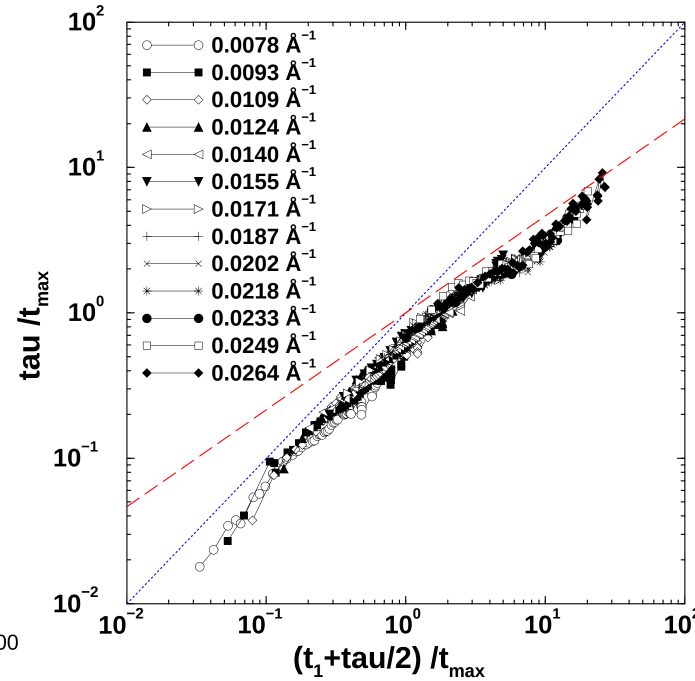
Fourier transforming we obtain:

$$\mathrm{Corr}(t_1, t_2) = \mathrm{Corr}\left(\frac{\delta t}{\bar{t}^{(1-n)}}\right) \quad \text{for large } \bar{t}$$

Characteristic time difference (correlation time), δt_c

Define $\delta t_c(\bar{t})$ by $\mathrm{Corr}(\delta t_c, \bar{t}) = 1/2$





Simulated δt_c vs $\bar{t}$

(Brown et al., PRE **60**, 5151 (1999))

Experimental δt_c vs $\bar{t}$

$Al_{0.91}Li_{0.09}$

(Livet et al., unpublished)

Speckle-intensity correlation scaling function for large $\bar{t}$

Analytical result

$$C_d\,(z) \;=\; \left[\frac{2}{\Gamma\left[\frac{d+1}{2}\right]}\left(A\frac{z}{2}\right)^{\frac{d+1}{2}} K_{\frac{d+1}{2}}\,(Az)\right]^2$$

$$\sim\; \begin{cases} 1 - O\left((Az)^2\right) & \text{for } Az \ll 1 \text{ (persistence)} \\ (Az)^d \exp[-2Az] & \text{for } Az \gg 1 \end{cases}$$

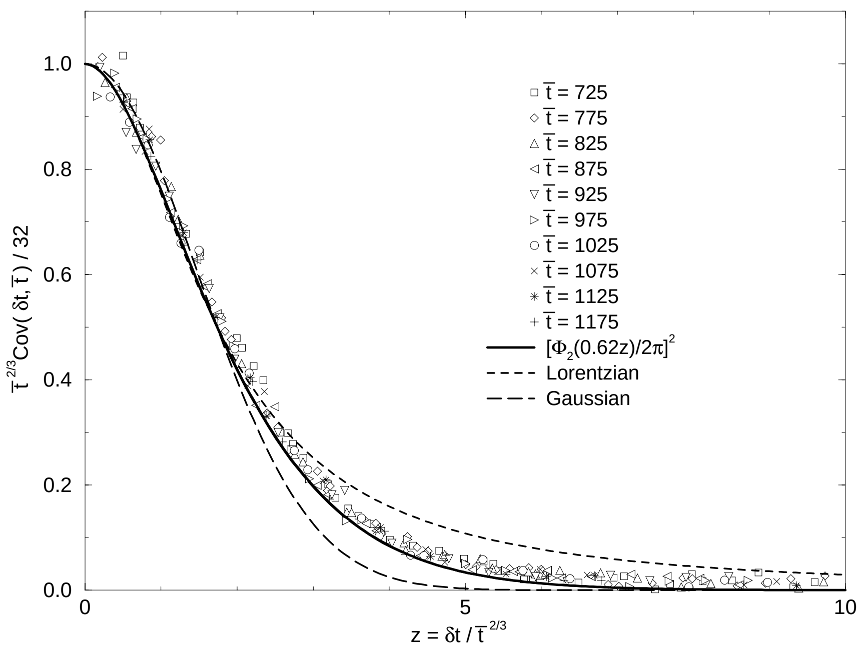
where $z = \delta t/\bar{t}^{(1-n)}$

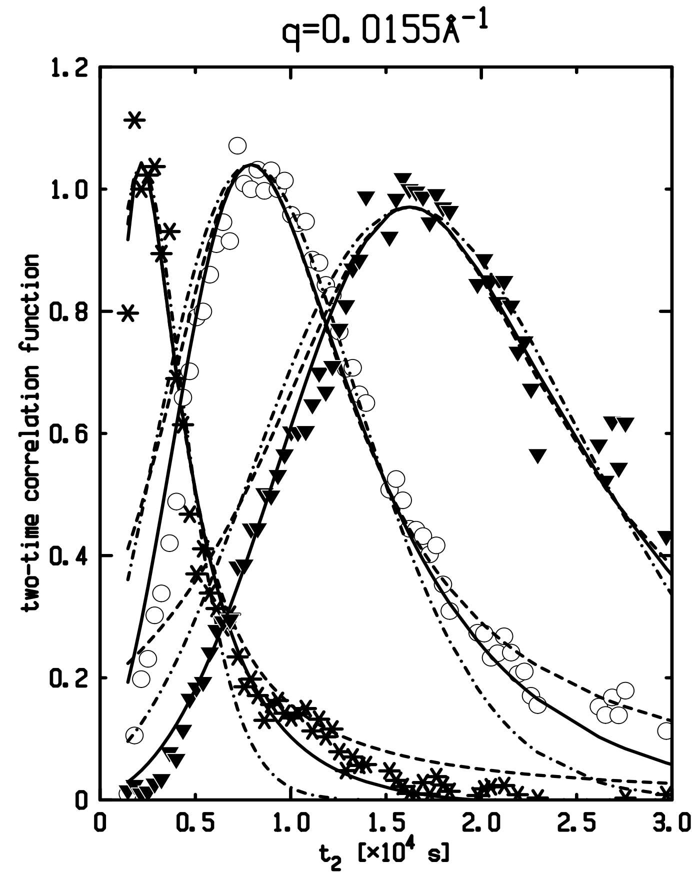
K_ν is a modified Bessel function, d is the spatial dimension, and A is a numerical constant

Valid for conserved *and* nonconserved order parameter and in 2 *and* 3 dimensions!

Speckle-intensity correlation scaling function for large $\bar{t}$

Comparison with numerical and experimental results





Simulated Corr($\delta t, \bar{t}$) vs $z = \delta t / \bar{t}^{(1-n)}$

(Brown et al., PRE **60**, 5151 (1999))

$Al_{0.91}Li_{0.09}$ (Livet et al.)

$\tau_1 = 2163$ s, 7920 s, 16320 s

Conclusions

- Speckle-intensity fluctuations in phase-segregating nonequilibrium systems
  - Give nonstationary, persistent time series
  - At early times reflect large-scale dynamics and structure
    Speckle correlation time $\delta t_c \sim \bar{t}$
  - At late times reflect local interface dynamics and structure
    Speckle correlation time $\delta t_c \sim \bar{t}^{(1-n)}$
  - Are observable in XIFS experiments
- Analytic large-$\bar{t}$ scaling function involving modified Bessel function valid for systems with both conserved and nonconserved order parameter, and in 2 and 3 dimensions
- Good agreement between analytic results, simulations, and XIFS experiments on Sodium Borosilicate Glass and Al-Li alloy