

Excess Noise for Driven Diffusive Systems

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We investigate the steady-state scattering function for driven diffusive systems with a single conserved density. In one dimension, density fluctuations spread as $t^{2/3}$, i.e., faster than the diffusive $t^{1/2}$, for large time t . The corresponding excess noise in the current-current correlation diverges as $\omega^{-1/3}$ for small frequency ω . Monte Carlo simulation results for a driven hard-core lattice gas confirm these results. $d=2$ is the borderline dimension with marginally nondiffusive behavior; for $d > 2$, the spread is diffusive with anisotropic long-time-tail corrections.

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When trying to understand the phenomena of excess noise in stationary nonequilibrium states we made a surprising observation: In stationary driven diffusive systems at low dimensionality the spreading of density fluctuations occurs intrinsically faster than would be predicted by an ordinary diffusion law. As a consequence the excess noise diverges for small frequencies.

Let us consider diffusive systems under a constant uniform driving force and assume that on a coarse time and length scale their dynamics can be described by the nonlinear Langevin equation

$$\frac{\partial}{\partial t} c(\mathbf{r}, t) + \text{div} \mathbf{j}(\mathbf{r}, t) = 0, \quad (1a)$$

$$\mathbf{j}(\mathbf{r}, t) = -D(c(\mathbf{r}, t)) \text{grad} c(\mathbf{r}, t) + c(\mathbf{r}, t) \mathbf{u}(c(\mathbf{r}, t)) + \tilde{\mathbf{j}}_L(\mathbf{r}, t), \quad (1b)$$

with $c(\mathbf{r}, t)$ and $\mathbf{j}(\mathbf{r}, t)$ the number density and corresponding current density at position \mathbf{r} and time t . The second term on the right-hand side of (1b) represents the action of the driving force: In a system with uniform density \bar{c} it sets up a steady particle current $\bar{c} \mathbf{u}(\bar{c})$, where \mathbf{u} is the average velocity. On the length scales considered the same is assumed to be valid locally. In addition the current has a Gaussian white-noise contribution $\tilde{\mathbf{j}}_L$, which is supposed to summarize globally the effects of the fast microscopic degrees of freedom.

If $\mathbf{u} = 0$ (or $\mathbf{u} = \text{const}$ allowing for a Galilean transformation), (1) is model B of critical dynamics,¹ or the Cahn-Hilliard theory² at high temperatures. Static correlations are short ranged and the frequency spectrum of the total current $\mathbf{J}(t) = \int d^d r \mathbf{j}(\mathbf{r}, t)$ is white, i.e., $\langle \mathbf{J}(t) \mathbf{J}(s) \rangle \sim \delta(t-s)$. For the driven system, $\mathbf{u} \neq 0$, we assume to have short-range static correlations as well, an assumption which can be justified for simple model systems.³ We want to know how time correlations are modified, in particular, the truncated current correlation

$$\mathbf{C}(t-s) = V^{-1} \langle [\mathbf{J}(t) - \langle \mathbf{J}(t) \rangle - \mathbf{v} \delta N(t)] [\mathbf{J}(s) - \langle \mathbf{J}(s) \rangle - \mathbf{v} \delta N(s)] \rangle. \quad (2)$$

Here $\langle \rangle$ refers to the steady-state average with given density \bar{c} . $\langle \mathbf{J}(t) \rangle \neq 0$ is the average current, $\mathbf{v} = V^{-1} \partial \langle \mathbf{J}(t) \rangle / \partial \bar{c}$, V is the volume of the system, and $\delta N(t) = N(t) - \langle N(t) \rangle$ with $N(t) = \int d^d r c(\mathbf{r}, t)$. The subtraction of $\mathbf{v} \delta N$ is important if the particle number in the stationary ensemble is not fixed. Similar subtraction terms appear in the standard Green-Kubo expression for the bulk viscosity.⁴ The excess noise is the power spectrum of the current, i.e., the Fourier transform of (2) with its value for large frequencies subtracted.^{5,6}

In order to investigate (1) for $\mathbf{u} \neq 0$ we split the density as $c(\mathbf{r}, t) = \bar{c} + \phi(\mathbf{r}, t)$ and expand $c \mathbf{u}(c)$ through quadratic order as

$$c \mathbf{u}(c) = \bar{c} \mathbf{u}(\bar{c}) + \mathbf{v} \phi(\mathbf{r}, t) + \mathbf{w} \phi^2(\mathbf{r}, t) + \dots$$

with $\mathbf{w} = \frac{1}{2} \partial \mathbf{v}(\bar{c}) / \partial \bar{c}$. In principle, also $D(c)$ should be expanded, but the nonlinearities resulting from this ex-

pansion are irrelevant compared to those in $c\mathbf{u}(c)$. The linear term is taken care of by the Galilean transformation $\phi(\mathbf{r}, t) = \tilde{\phi}(\mathbf{r} - \mathbf{v}t, t)$. Then (1) is approximated as

$$\frac{\partial}{\partial t} \phi(\mathbf{r}, t) = -\mathbf{w} \cdot \text{grad} \phi^2(\mathbf{r}, t) + D \Delta \phi(\mathbf{r}, t) - \text{div} \mathbf{j}_L(\mathbf{r}, t), \quad (3)$$

with $D = D(\bar{c})$ and $\mathbf{j}_L(\mathbf{r}, t) = \tilde{\mathbf{j}}_L(\mathbf{r} - \mathbf{v}t, t)$. Physically the nonlinearity $-\mathbf{w} \cdot \text{grad} \phi^2$ expresses the fact that the velocity at which a density fluctuation travels depends on its magnitude.

Let us consider the intermediate scattering function $S(\mathbf{k}, t) = \langle \hat{\phi}(-\mathbf{k}, 0) \hat{\phi}(\mathbf{k}, t) \rangle$, where $\hat{\phi}(\mathbf{k}, t)$ is the spatial Fourier transform of $\phi(\mathbf{r}, t)$. From it the dispersion of a density fluctuation obtains as a function of time proportional to

$$\lim_{k \rightarrow 0} V^{-1} (2/k^2) [S(\mathbf{k}, 0) - S(\mathbf{k}, t)],$$

and the current-current correlation function as

$$\hat{\mathbf{k}} \cdot \mathbf{C}(t) \cdot \hat{\mathbf{k}} = \frac{\partial^2}{\partial t^2} \lim_{k \rightarrow 0} \frac{1}{V} \left[\frac{1}{k^2} \right] [S(\mathbf{k}, 0) - S(\mathbf{k}, t)], \quad (4)$$

with $\hat{\mathbf{k}} = \mathbf{k}/k$.

$S(\mathbf{k}, t)$ may be calculated approximately by employing the mode-coupling formalism.⁷⁻⁹ In the simplest approximation this yields the integrodifferential equation

$$\frac{\partial}{\partial t} S(\mathbf{k}, t) = -Dk^2 S(\mathbf{k}, t) - 2(\mathbf{w} \cdot \mathbf{k})^2 [(2\pi)^d V S(\mathbf{k})]^{-1} \int_0^t ds S^* S(\mathbf{k}, t-s) S(\mathbf{k}, s). \quad (5)$$

Here d is the spatial dimension, the center star denotes convolution in \mathbf{k} space, and $S(\mathbf{k}) = S(\mathbf{k}, t=0)$. In (5) we employed the two-mode approximation for the self-energy operator appearing in the mode-coupling propagator, assuming this to be dominant for large t and small k . Strictly speaking, this assumption is justified only in dimensions $d > 2$, but it seems to be a good approximation also in low dimension.

We investigate the solution to (5). For $d > 2$ the diffusive term dominates. Following standard procedures⁹ we obtain from (5) and (4) a long-time tail in the current-current correlation as

$$\mathbf{C}(t) \cong 2(\mathbf{w}\mathbf{w}) [S(0)/V]^2 (8\pi D t)^{-d/2}, \quad (6)$$

and the excess noise

$$\mathbf{P}(\omega) = 4(\mathbf{w}\mathbf{w}) [S(0)/V]^2 (8\pi D)^{-d/2} [\Gamma(d/2)]^{-1} \times \begin{cases} \text{Re}\{i\pi(i\omega)^{(d-2)/2}\} & (d \text{ odd}) \\ \text{Re}\{(i\omega)^{(d-2)/2} \ln(i/\omega)\} & (d \text{ even}) \end{cases} \quad (7)$$

for small frequencies, where $\Gamma(z)$ denotes Euler's gamma function. Note that the long-time tail appears only for the current in the direction of \mathbf{w} .

In one dimension the mode-coupling term dominates. To extract its predictions we first rewrite (5) in terms of the dimensionless variables $\tau = \alpha t$, $\kappa = \beta k$, and $\Sigma(\kappa, \tau) = S(\kappa/\beta, \tau/\alpha)/S(0)$, with $\alpha = w^4 S^2(0)/8D^3 V^2$ and $\beta = 4D^2 V/w^2 S(0)$. In addition, we assume $S(\mathbf{k}) = S(0)$ for the range of \mathbf{k} values of interest to us. Then, for $d = 1$, Eq. (5) takes the form

$$\frac{\partial \Sigma(\kappa, \tau)}{\partial \tau} = -\frac{1}{2} \kappa^2 \left[\Sigma(\kappa, \tau) + \frac{2}{\pi} \int_0^\tau d\sigma \Sigma^* \Sigma(\kappa, \sigma) \Sigma(\kappa, \tau - \sigma) \right]. \quad (8)$$

Next we look for a scale-invariant solution of the form $\Sigma(\kappa, \tau) = h(\kappa\tau^{2/3})$ which should hold in the limit $\kappa \rightarrow 0$, $\tau \rightarrow \infty$ while $\kappa\tau^{2/3}$ remains constant. Indeed, in this limit the diffusive term $-\frac{1}{2} \kappa^2 \Sigma$ vanishes as $\tau^{-1/3}$ relative to the mode-coupling term. As a result the scaling function h satisfies

$$\frac{d}{dx} h(x) = -\frac{9}{4\pi} x \int_0^1 d\lambda \lambda^{-1/2} h^* h(\lambda x) h(x(1-\lambda^{3/2})^{2/3}), \quad (9)$$

where we substituted $x = \kappa\tau^{2/3}$ and $\lambda = (\sigma/\tau)^{2/3}$. From this equation one readily derives

$$\lim_{\kappa \rightarrow 0} (2/\kappa^2) [\Sigma(\kappa, 0) - \Sigma(\kappa, \tau)] = -h''(0) \tau^{4/3} \quad (10)$$

implying that a density fluctuation spreads *faster* than diffusively.

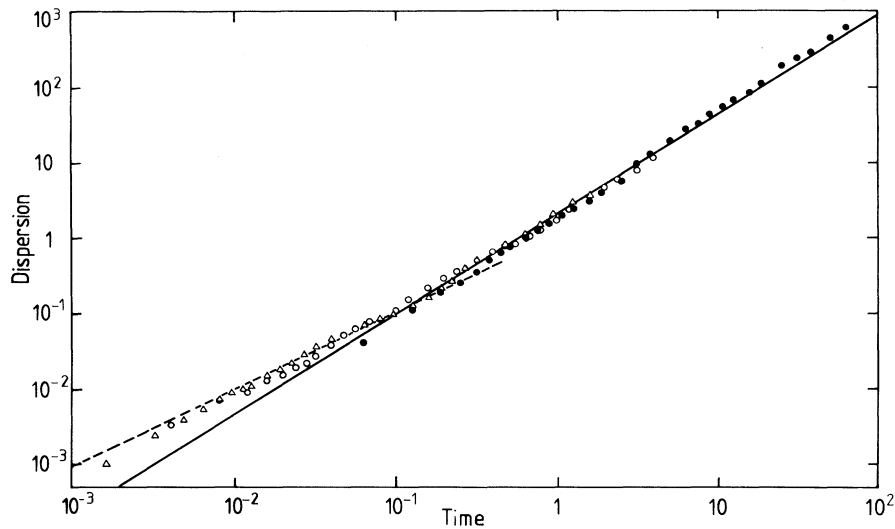


FIG. 1. The scaled center-of-mass dispersion as a function of the scaled time τ . Monte Carlo results are shown for $c = 0.503$, $p = 1$ (closed circles); for $c = 0.503$, $p = 0.75$ (open circles); and for $c = 0.802$, $p = 0.75$ (triangles). Solid line: fit by the eye with a $\tau^{4/3}$ law of the Monte Carlo results for large τ . Dashed line: theoretical short-time behavior.

We were not able to solve (9) analytically. Yet a few properties of the scaling function can be deduced: h is even and, provided it is positive, it is decreasing for $x > 0$ and bounded from above by $\exp(-Cx^{3/2})$. Also $h''(0) = (-9/2\pi)h(0)h'(0)$. So for small x , h has to be Gaussian. If one makes the *Ansatz* $h(x) = \exp(-\gamma x^2)$, which cannot be correct for large x , and determines γ from the above expression for $h''(0)$, then $-h''(0) \approx (9/2\sqrt{\pi})^{2/3}$. Transforming back to the original variables we predict that the excess noise diverges for small ω as

$$P(\omega) \approx (2/9\pi)^{1/3} \sqrt{3} \Gamma(\frac{1}{3}) [S(0)/V]^{5/3} w^{4/3} \omega^{-1/3}, \quad (11)$$

where the $-\frac{1}{3}$ power is exact and the coefficient is approximate.

The borderline dimension is two. Here the mode-coupling term still dominates for long times, but the enhancement of the diffusion in the field direction is only logarithmic. Assuming

$$S(\mathbf{k}, t) = S(0) \exp[-Dk^2 t - \delta(\mathbf{k} \cdot \mathbf{w})^2 t (\ln t)^\zeta]$$

for t large and $k \rightarrow 0$ and solving for δ and ζ by identifying both sides of (5) for $k \rightarrow 0$, one obtains $\zeta = \frac{2}{3}$ and $\delta = [3S(0)/8\pi V w]^{2/3} D^{-1/3}$, yielding for the current correlation

$$\mathbf{C}(t) = \mathbf{w}\mathbf{w} [S(0)/V] D^{-1/3} [S(0)/2\sqrt{6}\pi w V]^{2/3} / t (\ln t)^{1/3}. \quad (12)$$

The crossover in the dispersion from linear to $t(\ln t)^{2/3}$ behavior occurs for times of the order $(D\bar{c})^{-1} \exp[8\pi V D^2 / 3w^2 S(0) \cos^3 \Phi]$, where Φ is the angle between \mathbf{k} and \mathbf{u} .

We checked our predictions for one-dimensional systems by Monte Carlo simulations on a hard-core lattice gas: particles hop on a one-dimensional lattice with jump rate $p\Gamma$ [(1-p)\Gamma] for hops to unoccupied neighboring sites on the right (left). Double occupancy of sites is forbidden. In the steady state particles are distributed at random with density c , $0 \leq c \leq 1$. Then, choosing the lattice spacing as the unit of length, one has $v(c) = (1-2c)(2p-1)\Gamma$, $w = -(2p-1)\Gamma$, and $S(0) = c(1-c)V$, with V the number of sites in the system. Hence the scaling parameters α and β follow. On a lattice of 20 000 sites we simulated systems with the following combinations of values for c and p : $c = 0.503$, $p = 0.75$; $c = 0.503$, $p = 1$; $c = 0.802$, $p = 0.75$. We recorded the first and second moment of the center-of-mass position $X(t)$, which yields directly the dispersion $\lim_{k \rightarrow 0} V^{-1} (2/k^2) [S(k, 0) - S(k, t)]$. The results are shown in Fig. 1, where we have plotted the scaled center-of-mass dispersion,

$$(Vc)^2 \langle \{X(t) - X(0) - \langle X(t) - X(0) \rangle\}^2 \rangle / \beta^2 S(0),$$

as a function of $\alpha t = \tau$ on a doubly logarithmic scale (note that $\delta N = 0$ in our simulations). The solid line shows a $C\tau^{4/3}$ law; hence our theoretical prediction for the long-time behavior appears to be well confirmed. The coeffi-

cient is found as $C_{MCS} \approx 2.1$ from the simulations, which is to be compared to our approximate prediction $C_{th} \approx (9/2\sqrt{\pi})^{2/3} \approx 1.86$.

Some comments are in order:

(i) Equation (3) in one dimension *without* fluctuating current is known as the Burgers equation and used as a simple model for one-dimensional gas flow. It can be solved exactly. An initial disturbance spreads diffusively. The (deterministic) Burgers equation with random initial data has been studied extensively.^{10,11}

(ii) As we remarked already, in one dimension the mode-coupling equation (8) does not suffice to determine the scaling function completely. Instead the full self-energy operator of the mode-coupling propagator should be used in this equation. The scaling behavior $\Sigma(\kappa, \tau) = h(\kappa\tau^{2/3})$ would not be affected, but the explicit form of the scaling function h would be altered as well as the coefficient of the $\tau^{4/3}$ dispersion.

(iii) The $t^{-2/3}$ decay of current correlation functions will also be found in equilibrium fluids in one-dimensional geometry (e.g., in a narrow tube), provided that momentum cannot dissipate away through the walls. Notice, however, that real one-dimensional systems fall outside this class, as they do not satisfy the ordinary hydrodynamic equations.

(iv) In Eq. (8) the diffusive term $-\frac{1}{2}\kappa^2\Sigma$ dominates the mode-coupling term for small τ . As a result the mean square displacement of the center of mass relative to the average drift is linearly proportional to τ in this time regime. In Fig. 1 the theoretical prediction of scaled dispersion equal to τ is shown by the dashed line. The crossover from this initial behavior to the asymptotic $\tau^{4/3}$ behavior appears to occur rather sharply around $\tau \approx 0.1$, but a better resolution of the Monte Carlo data in this region would be needed.

(v) The anomalous diffusive behavior found for fluctuations of the bulk density does not occur for tracer density fluctuations.^{12,13} The reason is that tracer currents couple only to products of a tracer and a bulk density mode. The drift velocities of these modes in general differ from each other and therefore correlations between them decay exponentially. For certain specific choices of parameters in a given system one may be able to find equal drift velocities for bulk and tracer fluctuations. In that case the truncated velocity autocorrelation function of a tracer particle will exhibit the same type of long-time behavior as the collective current correlation function discussed here.

(vi) It would be of great interest to find experimental confirmation of the phenomena described here, especially in the one-dimensional case. The main

problem seems to be realizing conditions under which the crossover frequency $\approx 10\alpha$, cf. above Eq. (8), lies within the accessible range. To our guess the most promising systems for showing $\omega^{-1/3}$ behavior in electrical current noise would be hydrogen in metals near a critical point (so the sample diameter can be made comparable to the correlation length) and quasi-one-dimensional systems, such as TCNQ salts. For the type of experiment as discussed, for instance, by Scofield and Webb¹⁴ our theory predicts a constant noise below a corner frequency $\omega_0 \sim \alpha^{1/4}(2\pi^2 D/L^2)^{3/4}$, and a noise spectrum proportional to $\omega^{-5/3}$ for $\omega_0 < \omega < \omega_1 \approx 10\alpha$ and to $\omega^{-3/2}$ for $\omega > \omega_1$. One could also search for the effect in semimacroscopic systems, e.g., polystyrene suspensions flowing through capillary tubes, or in real macroscopic systems, such as pellets dropping through tubes or traffic flows on highways.

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