Thermal Noise and High-Schmidt Turbulent Mixing

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The Feynman Lectures on Physics, Vol. I Figure 1-1

1-2 Matter is made of atoms

If, in some cataclysm, all of scientific knowledge were to be destroyed, and only one sentence passed on to the next generations of creatures, what statement would contain the most information in the fewest words? I believe it is the *atomic hypothesis* (or the atomic *fact*, or whatever you wish to call it) that *all things are made of atoms—little particles that move around in perpetual motion, attracting each other when they are a little distance apart, but repelling upon being squeezed into one another.* In that one sentence, you will see, there is an *enormous* amount of information about the world, if just a little imagination and thinking are applied.



FIGURE: scanning tunneling microscope image of individual water molecules (Shin et al., *Nature Materials*, 2010)

(b) Classical Continuous Systems, Canonical Ensemble. In passing from the microcanonical to the canonical ensemble, the energy E is replaced as thermodynamic parameter by the *inverse temperature* $\beta > 0$. The measure defining the canonical ensemble is

$$\frac{1}{n!} \exp(-\beta H(\omega)) d\omega$$

$$= \prod_{i=1}^{n} \left[\exp\left(-\beta \frac{p_i^2}{2m}\right) dp_i \right] \times \left[\frac{1}{n!} \exp(-\beta U(x_1, \dots, x_n)) dx_1 \cdots dx_n \right] \quad (2.6)$$

This expression factorizes into a kinetic part

$$\exp\left(-\beta \frac{p_i^2}{2m}\right) dp_i \tag{2.7}$$

for each particle and a configurational part

$$\frac{1}{n!}\exp(-\beta U(x_1,\ldots,x_n))\,dx_1\cdots dx_n \tag{2.8}$$

From D. Ruelle, "Statistical Mechanics: Rigorous Results"

$$\beta = 1/k_BT$$



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Fluid Velocity and its Thermal Spectrum

The velocity in a parcel of fluid of linear size ℓ at space point x is a coarse-grained average of velocities of individual molecules, with respect to some filter kernel G:

$$\bar{\mathbf{u}}_{\ell}(\mathbf{x}) = \sum_{n} \mathbf{v}_{n} G_{\ell}(\mathbf{x} - \mathbf{x}_{n}) / \sum_{n} G_{\ell}(\mathbf{x} - \mathbf{x}_{n})$$

The statistics are Gaussian, with PDF

$$P(\bar{\mathbf{u}}_{\ell}) \propto \exp\left(-C \frac{\rho \ell^3 |\bar{\mathbf{u}}_{\ell} - \mathbf{v}|^2}{k_B T}\right), \quad \ell_{intp} \ll \ell \ll \ell_{\nabla}$$

variance

$$\langle |\delta \bar{\mathbf{u}}_{\ell}|^2 \rangle = (\text{const.}) \frac{k_B T}{\rho \ell^3} = \int_0^\infty |\widehat{G}(k\ell)|^2 E(k) \, dk$$

 $\simeq \int_0^{1/\ell} E(k) \, dk$

and energy spectrum at low Mach numbers (i.e. ignoring energy in sound waves)

$$E(k) \sim \frac{k_B T}{\rho} \frac{4\pi k^2}{(2\pi)^3}$$

corresponding to energy equipartition.

The current picture of the turbulent energy spectrum is



ISR=inertial subrange NDR=near-dissipation range FDR=far-dissipation range

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However, thermal noise should create at high-k an equilibrium spectrum $E(k) \sim \frac{k_B T}{\rho} \frac{4\pi k^2}{(2\pi)^3}$

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$$u_{\eta}^2 \eta \exp(-k\eta) \sim \frac{k_B T}{\rho} k^2$$
 or $\theta_K(k\eta)^2 \exp(k\eta) \sim 1$ or $k_c \eta \sim 2W(1/2\theta_K^{1/2})$

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For von Kármán flow with water (Debue et al., 2018) $\eta = 16 \mu m$ and $\theta_K = 2.5 \times 10^{-7}$

$$k_c\eta \sim 10.5 \Longrightarrow \ell_c = 2\pi/k_c \sim 9.6 \ \mu m \gg \lambda_{mfp} = 0.25 \ nm$$

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Robert Betchov (1957, 1961, 1964), Bandak et al. (2021), Eyink et al. (2021)

Landau-Lifschitz Fluctuating Hydrodynamics

The equations which can describe thermal noise effects at scales $<\eta$ in a turbulent flow are (in the low Mach limit) incompressible fluctuating hydrodynamics:

$$\partial_t \mathbf{u} + P_{\Lambda}(\mathbf{u} \cdot \nabla) \mathbf{u} = -\nabla p + v_{\Lambda} \Delta \mathbf{u} + \nabla \widetilde{\mathbf{\tau}}, \quad \nabla \mathbf{u} = 0$$

with stochastic stress given by the fluctuation-dissipation relation

$$\langle \widetilde{\tau}_{ij}(\mathbf{x},t)\widetilde{\tau}_{kl}(\mathbf{x}',t')\rangle = \frac{2\nu_{\Lambda}k_BT}{\rho} \left(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \frac{2}{3}\delta_{ij}\delta_{kl}\right) \times \delta_{\Lambda}^3(\mathbf{x}-\mathbf{x}')\delta(t-t')$$

where viscosity v_{Λ} is Λ -dependent and the cut-off delta-function is given by

$$\delta_{\Lambda}^{3}(\mathbf{x} - \mathbf{x}') = P_{\Lambda}\delta^{3}(\mathbf{x} - \mathbf{x}') = \frac{1}{V}\sum_{|\mathbf{k}| < \Lambda} e^{i\mathbf{k}\cdot\mathbf{x}} , \quad \mathbf{u}(\mathbf{x}) = P_{\Lambda}\mathbf{u}(\mathbf{x}) := \frac{1}{V}\sum_{|\mathbf{k}| < \Lambda} e^{i\mathbf{k}\cdot\mathbf{x}}\hat{\mathbf{u}}_{\mathbf{k}}$$

These are **not** SPDE's but are better understood as an "effective field theory" valid below a UV wavenumber cut-off Λ . They are valid if the cut-off length can be chosen (arbitrarily) between the gradient length ℓ_{∇} and the mean-free-path length λ_{mfp} :

$$\ell_
abla \,\ll\, \Lambda^{-1}\,\ll\,\lambda_{mfp}$$

See Zarate & Sengers (2006) and, for the low Mach limit,

A. Donev et al. "Low Mach number fluctuating hydrodynamics of diffusively mixing liquids," Comm. Appl. Math. and Comp. Sci. 9, 47-104 (2014)

Numerical Results: Fluctuating Hydrodynamics and DSMC

<u>There are **no experiments** validating Navier-Stokes at and below Kolmogorov scale η !</u> However, computer power is now sufficient that turbulent flows can be numerically simulated by fluctuating hydrodynamics and by DSMC (an MD method)



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The dissipation range of turbulence in molecular fluids is NOT accurately described by the deterministic Navier-Stokes equations.

Breakdown of Hydrodynamic Self-Similarity

How is this conclusion consistent with the <u>scaling symmetry</u> of incompressible Navier-Stokes?

$$\mathbf{u} \to \mathbf{u}' = \lambda \mathbf{u}, \quad \mathbf{x} \to \mathbf{x}' = \lambda^{-1} \mathbf{x}, \quad t \to t' = \lambda^{-2} t, \quad Re \text{ fixed}$$

This symmetry is exploited to derive the (deterministic) Navier-Stokes equation from the Boltzmann equation (Bardos, Golse & Levermore, 1991, 1993) and from stochastic lattice gases (Quastel & Yau, 1998) in the limit $\lambda \rightarrow 0$ at any *Re*. In a turbulent flow:

$$\varepsilon \to \varepsilon' = \lambda^4 \varepsilon, \quad u_\eta \to u_\eta' = \lambda u_\eta, \quad \eta \to \eta' = \lambda^{-1} \eta,$$

so that $\theta_K \rightarrow \theta_K' = \lambda \theta_K$ and thermal noise at the Kolmogorov scale indeed vanishes!

However, the relation $x^2e^{x}=1/\theta_K$ that determines the crossover wavenumber k_c by $x_c = k_c \eta$ implies that $1/\lambda$ is <u>unattainably</u> large. E.g. x'=2x requires $1/\lambda = \theta_K/\theta_K'=4e^x$. In the water experiment of Debue et al. (2018) a doubled crossover $k_c'=2k_c$ would require a $4e^{10.5}=145,262$ times larger system!

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A "far-dissipation range" with exponentially decaying energy spectrum is a virtual-reality construct that does not exist in Nature. What occurs is a thermal equipartition spectrum!

Batchelor-Kraichnan Theory of High-Schmidt Turbulent Mixing

Scalar concentration, solving $\partial_t c + \mathbf{u} \cdot \nabla c = D\Delta c$ for $Sc = \nu/D \gg 1$, below the **Kolmogorov length** $\ell_K = \nu^{3/4} \varepsilon^{-1/4}$ is subject to strain rate $\gamma = (\varepsilon/\nu)^{1/2}$ which drives a scalar cascade with flux χ down to the **Batchelor length** $\ell_B = (D/\gamma)^{1/2} = \ell_K/\sqrt{Sc}$



Fig. 2. Schematic of scalar spectrum for Sc = 1 (red). Sc < < 1 (areen), and Sc > > 1 (purple).





Batchelor (1959) assumed velocity-gradients constant both in space and time:

$$E_c(k) \sim C_B \frac{\chi}{\gamma} k^{-1} \exp(-C_B (k\ell_B)^2/2)$$



Kraichnan (1968, 1974) took velocity-gradients <u>constant in space</u> but <u>rapidly varying in time</u>:

$$E_c(k) \sim C_B(\chi/\gamma k)(1 + \sqrt{6C_B}k\ell_B) \exp\left(-\sqrt{6C_B}k\ell_B\right)$$

viscous-convective range: observed in several experiments (e.g., Gibson & Schwartz 1963; Grant et al. 1968; Nye & Brodkey 1967; Jullien et al. 2000; Iwano et al. 2021).



viscous-diffusive range: observed only in simulations (e.g., Yeung et al. 2004; Donzis et al. 2010; Gotoh et al. 2014, Clay 2017)



Effects of Thermal Noise



FIG. 1: Our predicted scalar concentration spectrum (red solid line, -) and the prediction of Kraichnan [25] (Kr74; green dashed line, ---), for a water-glycerol solution at temperature $T = 25^{\circ}$ C, pressure p = 1 bar and mean concentration of glycerol $\bar{c} = 0.5$, with $\gamma = 10^2 \text{ s}^{-1}$ and $\chi = 10^2 \text{ s}^{-1}$. Distinct ranges of the concentration spectrum are labelled: Batchelor's k^{-1} spectrum (Ba59); k^{-2} power-law associated to giant concentration fluctuations (GCF); k^2 equipartition spectrum (EQ).

letters to nature

Giant fluctuations in a free diffusion process

Alberto Vailati & Marzio Giglio

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Eyink & Jafari (2021) arXiv:2112.13115 [physics.flu-dyn]

$$F(k) = F_{\mathcal{A}}(k) + F_{\mathcal{B}}(k) + F_{\mathcal{C}}(k),$$
$$F_{\mathcal{A}}(k) = \frac{\chi}{3\pi\Gamma} \mathcal{A} \frac{1}{k} \operatorname{Re} \left(\operatorname{fi}(k(a+ib)) \right),$$
$$F_{\mathcal{B}}(k) = -\frac{\chi}{3\pi\Gamma} \frac{\mathcal{B}r_1}{h} \frac{1}{k} \operatorname{Im} \left(\operatorname{fi}(k(a+ib)) \right),$$

$$F_{\mathcal{B}}(k) = -\frac{\chi}{3\pi\Gamma} \frac{\mathcal{D}T_{1}}{b} \frac{1}{k} \operatorname{Im}\left(\operatorname{fi}(k(a+ib))\right)$$

and

$$F_{\mathcal{C}}(k) = -\frac{\chi}{3\pi\Gamma} \mathcal{C}\frac{1}{k} \Big(\mathrm{fi}(kr_1) - \pi \cos(kr_1) \Big),$$

where $f_i(z)$ denotes the auxiliary sine integral function

$$fi(z) = \int_0^\infty \frac{\sin t}{t+z} dt = \int_0^\infty \frac{e^{-zt}}{1+t^2} dt, \quad Re(z) > 0.$$

the low-*q* fluctuation

amplitude does not depend on any relevant fluid parameter. So the orders-of-magnitude increase of the fluctuations above the equilibrium value (the most prominent feature that can be captured experimentally) is to be expected for any non-equilibrium fluid that has macroscopic concentration variations comparable to those in this experiment.

Fluctuating hydrodynamics of a binary fluid mixture:

See Morozov (1984), Nonaka et al. (2015):

$$\partial_{t} \mathbf{u} = \mathscr{P} \Big[-\mathbf{u} \cdot \nabla \mathbf{u} + \nu \Delta \mathbf{u} + \nabla \cdot \left(\sqrt{2\nu k_{B} T \rho^{-1}} \boldsymbol{\eta}(\mathbf{x}, t) \right) \Big]$$
$$\partial_{t} c + \mathbf{v} \cdot \nabla c = \nabla \cdot \left(D_{0} \nabla c + \sqrt{2m D_{0} \rho^{-1} c(1-c)} \boldsymbol{\eta}_{c}(\mathbf{x}, t) \right)$$

with $\mathbf{v} = G_{\sigma} \star \mathbf{u}$ removing scales < σ =radius of solute molecules (Donev et al. 2014)

$$\langle \eta_{ij}(\mathbf{x},t)\eta_{kl}(\mathbf{x}',t')\rangle = (\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \frac{2}{3}\delta_{ij}\delta_{kl}) \times \delta^3_{\Lambda}(\mathbf{x} - \mathbf{x}')\delta(t - t') \qquad \langle \eta_{c_i}(\mathbf{x},t)\eta_{c_j}(\mathbf{x}',t')\rangle = \delta_{ij}\delta^3_{\Lambda}(\mathbf{x} - \mathbf{x}')\delta(t - t')$$

Linearization around the turbulent Navier-Stokes solution:

$$\partial_t \mathbf{u}_T = \mathscr{P} \Big[-\mathbf{u}_T \cdot \nabla \mathbf{u}_T + \nu \Delta \mathbf{u}_T \Big]$$

Decompose $\mathbf{u} = \mathbf{u}_T + \mathbf{u}_{\theta}$ so that

$$\partial_t \mathbf{u}_{\theta} = \mathscr{P} \Big[-\mathbf{u}_T \cdot \nabla \mathbf{u}_{\theta} - \mathbf{u}_{\theta} \cdot \nabla \mathbf{u}_T - \mathbf{u}_{\theta} \cdot \nabla \mathbf{u}_{\theta} + \nu \Delta \mathbf{u}_{\theta} + \nabla \cdot \left(\sqrt{2\nu k_B T \rho^{-1}} \boldsymbol{\eta}(\mathbf{x}, t) \right) \Big]$$

The crossed term is negligible because

$$\frac{|\mathbf{u}_{\theta} \cdot \nabla \mathbf{u}_{\theta}|}{|\nu \Delta \mathbf{u}_{\theta}|} \sim \frac{\ell u_{\theta,\ell}}{\nu} \sim \frac{\ell}{\nu} \frac{c_{th}}{\sqrt{n\ell^3}} \sim \left(\frac{\lambda_{intp}^3}{\lambda_{mfp}^2 \ell}\right)^{1/2}$$

assuming $\nu \sim c_{th} \lambda_{mfp}$. The ratio is small for $\ell > \lambda_{intp}^3 / \lambda_{mfp}^2$ (Eyink et al. 2021)

Kraichnan model of the turbulent solution

We take $\mathbf{u}_T(\mathbf{x}, t)$ to be a Gaussian, random (incompressible) velocity field, white-noise in time, with

$$\langle u_{T,i}(\mathbf{x},t)u_{T,j}(\mathbf{x}',t')\rangle = \mathcal{U}_{T,ij}(\mathbf{x}-\mathbf{x}')\delta(t-t')$$

and zero mean, where $\mathscr{U}_{T,ij}(\mathbf{r}) = \mathscr{U}_{T,ij}(\mathbf{0}) - 2\Gamma\left(2r^2\delta_{ij} - r_ir_j\right)$ and $\mathscr{U}_{T,ij}(\mathbf{0}) = 2\mathscr{U}_{T0}\delta_{ij}$. Then

$$\partial_t \mathbf{u}_{\theta} = \mathscr{P} \Big[-\mathbf{u}_T \odot \nabla \mathbf{u}_{\theta} - \mathbf{u}_{\theta} \odot \nabla \mathbf{u}_T + \nu \Delta \mathbf{u}_{\theta} + \nabla \cdot \left(\sqrt{2\nu k_B T \rho^{-1}} \boldsymbol{\eta}(x, t) \right) \Big]$$
(S

(Stratonovich)

$$\partial_t c = -\mathbf{v}_T \odot \nabla c - \mathbf{v}_\theta \cdot \nabla c + \nabla \cdot \left(D_0 \nabla c + \sqrt{2mD_0 \rho^{-1} c(1-c)} \, \boldsymbol{\eta}_c(\mathbf{x}, t) \right)$$

Scalings for high-Schmidt asymptotic limit:

Following Donev, Fai & vanden-Eijnden (2014) we take

$$\nu \to \epsilon^{-1} \nu , D_0 \to \epsilon D_0$$

so that $D_0 \nu \sim e^0$ and $Sc_0 = \frac{\nu}{D_0} \sim e^{-2}$. Furthermore, we consider long times of mass diffusion $t \to e^{-1}t$ Finally, since $\frac{d}{dt} \frac{1}{2} \langle |\mathbf{u}|^2 \rangle = -\nu \langle |\nabla \mathbf{u}|^2 \rangle \sim \nu \Gamma^2$, we take $\Gamma \to \epsilon \Gamma$

so that a finite amount of kinetic energy is dissipated in a diffusive time.

Rescaled equations

$$\partial_{t} \mathbf{u}_{\theta} = \mathscr{P} \Big[-\mathbf{u}_{T} \odot \nabla \mathbf{u}_{\theta} - \mathbf{u}_{\theta} \cdot \nabla \mathbf{u}_{T} + \nu \epsilon^{-2} \Delta \mathbf{u}_{\theta} + \nabla \cdot \left(\sqrt{2\nu \epsilon^{-2} k_{B} T / \rho} \ \boldsymbol{\eta}(\mathbf{x}, t) \right) \Big]$$

$$\partial_{t} c = -\mathbf{v}_{T}(\mathbf{x}, t) \odot \nabla c - \epsilon^{-1} \mathbf{v}_{\theta}(\mathbf{x}, \epsilon^{-1} t) \cdot \nabla c + D_{0} \Delta c + \nabla \cdot \left(\sqrt{2m D_{0} \rho^{-1} c (1 - c)} \ \boldsymbol{\eta}_{c}(\mathbf{x}, t) \right)$$

High-Sc limit equations as $\epsilon \to 0$

To leading order \mathbf{u}_{θ} satisfies the linear equation $\partial_t \mathbf{u}_{\theta} = \mathscr{P} \Big[\nu \epsilon^{-2} \Delta \mathbf{u}_{\theta} + \nabla \cdot \left(\sqrt{2\nu \epsilon^{-2} k_B T / \rho} \, \boldsymbol{\eta}(\mathbf{x}, t) \right) \Big]$, the same as for a fluid in thermal equilibrium, at rest!

Furthermore, $e^{-1}\mathbf{v}_{\theta}(\mathbf{x}, e^{-1}t) \rightarrow \mathbf{w}_{\theta}(\mathbf{x}, t)$, a white-in-time velocity field:

$$\langle \mathbf{w}_{\theta}(\mathbf{x},t) \otimes \mathbf{w}_{\theta}(\mathbf{x}',t') \rangle = \mathbf{R}(\mathbf{x}-\mathbf{x}')\delta(t-t')$$

whose spatial realizations satisfy $\mathscr{P}[\nu\Delta w_{\theta} + \nabla \cdot [(2\nu k_B T/\rho)^{1/2}\eta_{\sigma}] = 0$, $\eta_{\sigma} = G_{\sigma} \star \eta$ and thus

$$\mathbf{R}(\mathbf{r}) = \frac{2k_B T}{\eta} (G_{\sigma} \star \mathbf{G} \star G_{\sigma})(\mathbf{r}), \quad \eta = \nu \rho \quad \text{(shear viscosity)}$$

with the Oseen tensor $G^{ij}(\mathbf{r}) = \frac{1}{8\pi r} \left(\delta^{ij} + \frac{r^i r^j}{r^2} \right)$. Thus, the concentration field in the limit satisfies

$$\partial_t c = -\left(\mathbf{v}_T + \mathbf{w}_\theta\right) \odot \nabla c + D_0 \Delta c + \nabla \cdot \left(\sqrt{2mD_0\rho^{-1}c(1-c)} \ \boldsymbol{\eta}_c(\mathbf{x},t)\right)$$

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Neglecting molecular noise, this is a Kraichnan white-noise advection model!

Solution of the Model

Converting from Stratonovich to Itō and adding a source term to drive a statistical steady-state:

"the ratio $l = kT/D\eta$ is a length of the order of magnitude of molecular dimensions, normally smaller than the value $6\pi a$... From the point of view of molecular theory, viscous flow and diffusion present parallel problems. It would seem that for an exact theory of either, we should have to analyze the cooperative character of the molecular motion involved; but this difficult analysis has not yet been developed further than the hydrodynamic approximation." — L. Onsager, *Theories and Problems of Liquid Diffusion* (1945)

Closed Equations for Correlation Functions:

$$\partial_t C(\mathbf{r}, t) = \left[\mathcal{V}_{ij}(\mathbf{0}) - \mathcal{V}_{ij}(\mathbf{r}) \right] \partial_i \partial_j C + 2D_0 \Delta C + S\left(\frac{\mathbf{r}}{L}\right)$$
$$C(r) = \int_r^\infty \frac{\int_0^\rho S\left(\frac{\bar{\rho}}{L}\right) \bar{\rho}^{d-1} d\bar{\rho}}{\rho^{d-1} [2D_0 - (d-1)(J(0) - J(\rho))]} d\rho$$

where $J(r) = -\frac{1}{r^d} \int_0^r K(\rho) \rho^{d-1} d\rho$ with $\mathcal{V}_{ij}(\mathbf{r}) = \mathcal{P}_{ij}K(r)$. The rest is an exercise in analysis!

Conclusions

- 1. The dissipation range of turbulent flows is argued to be described by Landau-Lifschitz fluctuating hydrodynamics and not by the deterministic Navier-Stokes equations. This conclusion is supported by simulations.
- 2. In prior work (Eyink et al. 2021, Bell et al. 2021), it has been shown that thermal noise at sub-Kolmogorov scales erases far dissipation-range intermittency and modifies extreme events due to inertial-range intermittency.
- 3. We have presented here a theory of effects of thermal noise on the Batchelor-Kraichnan regime of high Schmidt-number turbulent mixing of passive concentration fields, predicting that exponential decay of the scalar spectrum in the viscous-diffusive range is replaced by "giant concentration fluctuations" which are well-observed for diffusion in laminar flows.
- 4. Similar thermal noise effects can be expected for other physical processes at sub-Kolmogorov scales of turbulent flows, such as combustion, formation of droplets and bubbles, locomotion of micro-organisms, etc.

THANKS!