Fluctuating Hydrodynamics for Quasi2D Diffusion

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1 Nonlinear FHD Equations

The concentration (number density) of the particles can be described by [1]

$$\begin{aligned}\partial_t c(\boldsymbol{r},t) &= -\boldsymbol{\nabla}\cdot\left(\boldsymbol{w}\left(\boldsymbol{r},t\right)c(\boldsymbol{r},t)\right)+\boldsymbol{\nabla}\cdot\left(\chi\boldsymbol{\nabla}c(\boldsymbol{r},t)\right)\\ &+\left(k_BT\right)\boldsymbol{\nabla}\cdot\left(c(\boldsymbol{r},t)\int\boldsymbol{\mathcal{R}}(\boldsymbol{r}-\boldsymbol{r}')\boldsymbol{\nabla}'c(\boldsymbol{r}',t)\,d\boldsymbol{r}'\right)\\ &+\boldsymbol{\nabla}\cdot\left(\sqrt{2\chi c(\boldsymbol{r},t)}\,\boldsymbol{\mathcal{W}}(\boldsymbol{r},t)\right)\end{aligned} \tag{1}$$

Here $\boldsymbol{w}\left(\boldsymbol{r},t\right)$ is a random velocity field that is white in time and has a spatial covariance $\sim\boldsymbol{\mathcal{R}}$,

$$\left\langle\boldsymbol{w}\left(\boldsymbol{r},t\right)\otimes\boldsymbol{w}\left(\boldsymbol{r}',t'\right)\right\rangle=\left(\frac{2k_BT}{\eta}\right)\boldsymbol{\mathcal{R}}\left(\boldsymbol{r}-\boldsymbol{r}'\right)\delta\left(t-t'\right), \tag{2}$$

and has a clear physical interpretation in *fluctuating hydrodynamics* [2]. For ordinary diffusion in 2D and 3D, the hydrodynamic kernel is divergence free, $\boldsymbol{\nabla}\cdot\boldsymbol{\mathcal{R}}(\boldsymbol{r})=0$, and so the nonlinear term in (1) disappears and we obtain a *linear* fluctuating advection-diffusion equation that can easily be solved numerically [2].

If we color the particles into red and green particles, then we get a system of two coupled nonlinear nonlocal diffusion equations,

$$\begin{aligned}\partial_t c_{R/G}\left(\boldsymbol{r},t\right) &= \boldsymbol{\nabla}\cdot\left(-\boldsymbol{w}\left(\boldsymbol{r},t\right)c_{R/G}(\boldsymbol{r},t)+\chi\boldsymbol{\nabla}c_{R/G}\left(\boldsymbol{r},t\right)+\sqrt{2\chi c_{R/G}(\boldsymbol{r},t)}\,\boldsymbol{\mathcal{W}}^{(R/G)}(\boldsymbol{r},t)\right)\\ &+ \left(k_BT\right)\boldsymbol{\nabla}\cdot\left(c_{R/G}\left(\boldsymbol{r},t\right)\int\boldsymbol{\mathcal{R}}\left(\boldsymbol{r},\boldsymbol{r}'\right)\boldsymbol{\nabla}'c\left(\boldsymbol{r}',t\right)\,d\boldsymbol{r}'\right),\end{aligned} \tag{3}$$

where $c=c_R+c_G$. If we add these two equations we get back equation (2), as we must. Note that here the noise terms proportional to $\sqrt{2\chi c}$ are separate and independent in the two equations, but the velocity $\boldsymbol{w}$ is the *same* in both equations (there is only one velocity).

As explained in [1], for compressible velocity fields we have not had success solving the nonlinear FHD equation (3) and instead we solve numerically the much simpler equation

$$\partial_t c_{R/G}=-\epsilon^{\frac{1}{2}}\boldsymbol{w}^{\perp}\cdot\boldsymbol{\nabla}c_{R/G}+\chi\boldsymbol{\nabla}^2c_{R/G}, \tag{4}$$

where $\boldsymbol{w}^{\perp}$ is the incompressible or vortical component of $\boldsymbol{w}$. More precisely, $\left\langle\boldsymbol{w}^{\perp}\left(\boldsymbol{r},t\right)\otimes\boldsymbol{w}^{\perp}\left(\boldsymbol{r}',t'\right)\right\rangle=\left(2k_BT\right)\boldsymbol{\mathcal{R}}^{\perp}\left(\boldsymbol{r}-\boldsymbol{r}'\right)\delta\left(t-t'\right)$, where in Fourier space $\hat{\boldsymbol{\mathcal{R}}}_{\boldsymbol{k}}^{\perp}=\left(c_2\left(ka\right)/\eta k^3\right)\,\boldsymbol{k}_{\perp}\otimes\boldsymbol{k}_{\perp}$ is the incompressible component of the Quasi2D hydrodynamic kernel, see formulas below.

1.1 Hydrodynamic kernel

We will use the hydrodynamic kernel given in Fourier space by

$$\hat{\boldsymbol{\mathcal{R}}}_{\boldsymbol{k}} = \frac{1}{\eta k^3} \left(c_2 \left(ka \right) \, \boldsymbol{k}_\perp \boldsymbol{k}_\perp^T + c_1 \left(ka \right) \, \boldsymbol{k} \boldsymbol{k}^T \right), \tag{5}$$

where we singled out a $1/k^3$ prefactor but this is not required.

In **Quasi2D (q2D)** (particles on a fluid-fluid interface) we have:

$$\begin{aligned} c_1 \left(K \right) &= \frac{1}{2\pi} \left(-K \, \exp\left(-\frac{K^2}{\pi} \right) - \left(K^2 + \frac{\pi}{2} \right) \left(\operatorname{erf}\left(\frac{K}{\sqrt{\pi}} \right) - 1 \right) \right) \\ c_2 \left(K \right) &= \frac{1}{2} \left(1 - \operatorname{erf}\left(\frac{K}{\sqrt{\pi}} \right) \right), \end{aligned} \tag{6}$$

where $K = ka$ and a is the hydrodynamic radius of the particles. An important point is that both expression decay exponentially like $\exp\left(-a^2 k^2 \right)$ in Fourier space, which is crucial for pseudospectral methods to work. I

For **True2D (t2D)** (thin films in vacuum) we have:

$$\begin{aligned} c_1 \left(K \right) &= 0 \\ c_2 \left(K \right) &= \frac{a}{K} \, \exp\left(-\frac{K^2}{\pi} \right), \end{aligned} \tag{7}$$

For **(2+1)D** (diffusion in a thin membrane surrounded by a viscous unbounded liquid) we have the Saffman kernel:

$$\begin{aligned} c_1 \left(K \right) &= 0 \\ c_2 \left(K = ka \right) &= \frac{1}{k + k_c} \, \exp\left(-\frac{K^2}{\pi} \right), \end{aligned} \tag{8}$$

where k_c is a roll-over wavenumber that depends on the ratio of the viscosity of the surrounding fluid to the two-dimensional viscosity η of the membrane itself. When the membrane or thin film is suspended in vacuum, $k_c \to 0$ and (8) becomes the same as (7).

In terms of the particle simulations, we can also generically write the diffusion coefficient as

$$\chi = f \, \frac{k_B T}{\eta},$$

where the factor f is [3]

$$\begin{aligned} f &= \frac{1}{6\pi a} \cdot \frac{1}{1 + 4.41 a / L} \approx \frac{1}{6\pi a} \quad \text{for Quasi2D, and} \\ f &= \frac{1}{4\pi} \ln\left(\frac{L}{3.71 a} \right) \quad \text{for True2D,} \\ f &\approx \frac{1}{4\pi} \left(- \ln\left(-k_c a / 2 \right) - \gamma \right) \quad \text{for (2+1)D (}\textbf{rough estimate}\text{),} \end{aligned} \tag{9}$$

and L is the system size.

2 Linearized FHD

The linearized FHD equations can be given a precise meaning. With these equations we also have a freedom to include equilibrium (background) fluctuations or only focus on non-equilibrium ones, since fluctuations are additive. Let us denote

$$\left(\frac{k_B T}{\eta}\right) = \epsilon$$

and also introduce a coefficient α that is either zero (no background fluctuations) or 1.

2.1 Density Gradient

Now we consider a system out of equilibrium, in the presence of gradients. First, we consider a constant macroscopic gradient $\boldsymbol{\nabla} c = \boldsymbol{g}$ of the total number density, without color. We imagine here that the gradient is very weak so the system is essentially uniform, and that the gradient is imposed and maintained externally (say by boundary conditions). This allows us to perform a standard linearized FHD calculation to study the spectrum of the nonequilibrium long-ranged (giant) fluctuations [4]. In the presence of the weak gradient the linearized FHD equation has an extra term $\boldsymbol{w} \cdot \boldsymbol{g}$, where $\delta c = c - c_0$ is the fluctuation around the background value,

$$\begin{aligned}\partial_t \delta c(\boldsymbol{r}, t) &= \chi \boldsymbol{\nabla}^2 \delta c(\boldsymbol{r}, t) + \sqrt{2\alpha c_0 \chi}\, \boldsymbol{\nabla} \cdot \boldsymbol{\mathcal{W}} \\ &+ \epsilon \boldsymbol{\nabla} \cdot \left(c_0 \int \boldsymbol{\mathcal{R}}(\boldsymbol{r} - \boldsymbol{r}') \boldsymbol{\nabla}' \delta c(\boldsymbol{r}', t)\, d\boldsymbol{r}' \right) - \alpha c_0 \left(\boldsymbol{\nabla} \cdot \boldsymbol{w} \right) - \boldsymbol{w} \cdot \boldsymbol{g}.\end{aligned} \tag{10}$$

Let us assume a gradient in the z direction, $\boldsymbol{\nabla} c_0 = g\hat{\boldsymbol{y}}$, and that the wavenumber is perpendicular to the gradient, $\boldsymbol{k} \perp \boldsymbol{y}$. In Fourier space (10) becomes

$$\begin{aligned}\partial_t \left(\widehat{\delta c}\right) &= -\left(\chi k^2 + \epsilon c_0 k c_1(ak)\right) \widehat{\delta c} \\ &+ \sqrt{2\alpha c_0 \chi}\, \left(ik \mathcal{Z}_{\boldsymbol{k}}\right) - i\alpha c_0 \sqrt{2\epsilon k c_1\left(ka\right)}\, \mathcal{Z}_{\boldsymbol{k}}^{(1)} \\ &- g\sqrt{\frac{2\epsilon c_2\left(ka\right)}{k}}\, \mathcal{Z}_{\boldsymbol{k}}^{(2)}.\end{aligned} \tag{11}$$

From this equation we obtain the static structure factor

$$S(\boldsymbol{k}) = \left\langle \left| \widehat{(\delta c)} \right|^2 \right\rangle = S_0 + \Delta S = \alpha c_0 + \epsilon g^2 \frac{c_2\left(ka\right)}{k\left(\chi k^2 + c_0 \epsilon k c_1(ak)\right)}. \tag{12}$$

The second term above is the nonequilibrium fluctuations on top of the equilibrium $S_0 = c_0$.

2.2 Giant Color Fluctuations

Now we consider fluctuations of color when we have a **gradient in color but without a gradient in density**. To write the equations we linearize (3) and then add fluctuations in order to obey fluctuation-dissipation balance at equilibrium,

$$\boldsymbol{S}\left(\boldsymbol{k}\right) = \left[\begin{array}{cc} \left\langle \left(\widehat{\delta c}_R\right) \left(\widehat{\delta c}_R\right)^{\star} \right\rangle & \left\langle \left(\widehat{\delta c}_R\right) \left(\widehat{\delta c}_G\right)^{\star} \right\rangle \\ \left\langle \left(\widehat{\delta c}_G\right) \left(\widehat{\delta c}_R\right)^{\star} \right\rangle & \left\langle \left(\widehat{\delta c}_G\right) \left(\widehat{\delta c}_G\right)^{\star} \right\rangle \end{array} \right] = \boldsymbol{S}_0 = \left[\begin{array}{cc} c_R & 0 \\ 0 & c_G \end{array} \right] \quad \text{at equilibrium,}$$

We impose the gradient $\boldsymbol{\nabla} c_R = -\boldsymbol{\nabla} c_G = \boldsymbol{g}$ and perform the same computation as in the previous section. The linearized equations are

$$\begin{aligned}\partial_t \delta c_R(\boldsymbol{r},t) &= \chi \boldsymbol{\nabla}^2 \delta c_R(\boldsymbol{r},t) + \sqrt{2\alpha c_R \chi}\, \boldsymbol{\nabla}\cdot\boldsymbol{\mathcal{W}} - \alpha c_R \left(\boldsymbol{\nabla}\cdot\boldsymbol{w}\right) - \boldsymbol{w}\cdot\boldsymbol{g} \\ &+ \epsilon \boldsymbol{\nabla}\cdot\left(c_R \int \boldsymbol{\mathcal{R}}(\boldsymbol{r}-\boldsymbol{r}')\boldsymbol{\nabla}'\left(\delta c_R(\boldsymbol{r}',t) + \delta c_G(\boldsymbol{r}',t)\right)\, d\boldsymbol{r}'\right). \end{aligned} \tag{13}$$

$$\begin{aligned}\partial_t \delta c_G(\boldsymbol{r},t) &= \chi \boldsymbol{\nabla}^2 \delta c_G(\boldsymbol{r},t) + \sqrt{2\alpha c_G \chi}\, \boldsymbol{\nabla}\cdot\boldsymbol{\mathcal{W}} - \alpha c_G \left(\boldsymbol{\nabla}\cdot\boldsymbol{w}\right) + \boldsymbol{w}\cdot\boldsymbol{g} \\ &+ \epsilon \boldsymbol{\nabla}\cdot\left(c_G \int \boldsymbol{\mathcal{R}}(\boldsymbol{r}-\boldsymbol{r}')\boldsymbol{\nabla}'\left(\delta c_R(\boldsymbol{r}',t) + \delta c_G(\boldsymbol{r}',t)\right)\, d\boldsymbol{r}'\right). \end{aligned} \tag{14}$$

In Fourier space the linearized equations are

$$\begin{aligned}\partial_t \begin{bmatrix} \widehat{\delta c_R} \\ \widehat{\delta c_G} \end{bmatrix} &= -\chi k^2 \begin{bmatrix} \widehat{\delta c_R} \\ \widehat{\delta c_G} \end{bmatrix} - \epsilon k c_1(ak) \begin{bmatrix} c_R & c_R \\ c_G & c_G \end{bmatrix} \begin{bmatrix} \widehat{\delta c_R} \\ \widehat{\delta c_G} \end{bmatrix} \\ &+ \begin{bmatrix} ik\sqrt{2\alpha\chi c_R} \\ 0 \end{bmatrix} \mathcal{Z}_k^{(R)} + \begin{bmatrix} 0 \\ ik\sqrt{2\alpha\chi c_G} \end{bmatrix} \mathcal{Z}_k^{(G)} \\ &- i\sqrt{2\epsilon\alpha k c_1(ka)} \begin{bmatrix} c_R \\ c_G \end{bmatrix} \mathcal{Z}_{\boldsymbol{k}}^{(1)} - \sqrt{\frac{2\epsilon c_2(ka)}{k}} \begin{bmatrix} g \\ -g \end{bmatrix} \mathcal{Z}_{\boldsymbol{k}}^{(2)}. \end{aligned} \tag{15}$$

Solving this linear system of SODEs (Ornstein-Uhlenbeck process) we obtain

$$\boldsymbol{S}(\boldsymbol{k}) = \boldsymbol{S}_0 + \Delta\boldsymbol{S} = \alpha \begin{bmatrix} c_R & 0 \\ 0 & c_G \end{bmatrix} + \epsilon g^2 \frac{c_2(ka)}{\chi k^3} \begin{bmatrix} 1 & -1 \\ -1 & 1 \end{bmatrix}, \tag{16}$$

where we now see that the giant fluctuations have a spectrum $\sim 1/k^3$ and therefore have the potential to be sufficiently strong to be measurable, although they are still not as large as in True2D. Note that if we look at the total density $\delta c = \delta c_R + \delta c_G$ its spectrum is flat, since the system is in equilibrium aside from color gradients,

$$\left\langle \left(\widehat{\delta c}\right)\left(\widehat{\delta c}\right)^{\star} \right\rangle = S_{RR} + S_{GG} + S_{RG} + S_{GR} = \alpha\left(c_R + c_G\right) = \alpha c.$$

References

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