Coarse-grained forms for equations describing
the microscopic motion of particles in a fluid

Shankar P. Das¹ and Akira Yoshimori²

¹ School of Physical Sciences, Jawaharlal Nehru University, New Delhi 110067, India.
² Department of Physics, Kyushu University, Fukuoka 812-8581, Japan

Abstract

Equations of motion for the microscopic number density \( \hat{\rho}(x, t) \) and the momentum density \( \hat{g}(x, t) \) of a fluid have been obtained in the past from the corresponding Langevin equations representing the dynamics of the fluid particles. In the present work we average these exact equations of microscopic dynamics over the local equilibrium distribution to obtain stochastic partial differential equations for the coarse grained densities with smooth spatial and temporal dependence. In particular, we consider Dean’s exact balance equation for the microscopic density of a system of interacting Brownian particles to obtain the basic equation of the dynamic density functional theory. In the thermally averaged equation for the coarse grained density \( \rho(x, t) \), the related dependence on the bare interaction potential in Dean’s equation is converted to that on the corresponding direct correlation functions of the density functional theory.
The equations of fluctuating nonlinear hydrodynamics (FNH) provide a very useful description of the time dependent behavior of various types of liquids, ranging from simple liquids, solutions, supercooled metastable liquids, mixtures, and complex liquids. These equations primarily represent the time evolution of the respective coarse grained densities for the system. Experimentally measurable correlation functions of these densities are obtained from the equations of FNH by averaging the corresponding quantities with respect to the noise. The simplest example of a collective density is the number density \( \rho(\mathbf{r}, t) \) of particles in a fluid. The latter is obtained as the average of the corresponding microscopic quantity, \( \hat{\rho}(\mathbf{x}, t) \equiv \rho(\mathbf{x}, t) \), where the hat over \( \rho \) indicates its dependence on the phase space coordinates of the fluid particles and the angular brackets denote an averaging over the appropriate ensemble. Dynamic equations for the microscopic densities (with a hat) are obtained exactly from the corresponding equations of motion of the individual fluid particles. For example, if the fluid particles follow reversible Newtonian dynamics (ND) the corresponding exact balance equations of hydrodynamics are known as Euler’s equations which are also time reversible. For fluid particles with dissipative dynamics, the corresponding equations for the microscopic densities are also time irreversible. Apart from being exact representations of the corresponding microscopic dynamics, these exact equations are formal and needs to be averaged over an appropriate ensemble for practical use. In this paper we obtain coarse grained forms of these exact equations by averaging them over a suitable local equilibrium distribution. The final equations are stochastic partial differential equations having smooth spatial and temporal dependence. The coefficients involved in the FNH equations are related to the thermodynamic properties of the fluid.

We begin by considering a \( N \) particle system in which the microscopic dynamics of the constituent particles is described by Smoluchowski equation involving only the particle coordinates. Let the \( \alpha \)-th particle (for \( \alpha = 1, \ldots, N \)) be of mass \( m \) and its position coordinates are given by \( \{ \mathbf{x}_\alpha \} \). A colloidal system with heavy particles in a solution is a typical example for such a system. In the over damped limit, the time dependence of the momentum of the particle is ignored and the stochastic equation of motion for the coordinate \( \mathbf{x}_\alpha \) of the \( \alpha \)-th particle is obtained as,

\[
\gamma_0 \frac{d\mathbf{x}_\alpha(t)}{dt} = - \sum_{\nu=1}^{N} \nabla_\alpha U(\mathbf{x}_\alpha - \mathbf{x}_\nu) + \xi_\alpha(t).
\]  

(1)
The quantity $\gamma_0$ which has dimension of $[MT^{-1}]$ is chosen to be unity in the following. $U(x_\alpha - x_\nu)$ is the interaction potential between particles $\alpha$ and $\nu$. The symbol $\nabla_\alpha$ with the Greek subscript $\alpha$ denotes the derivative operator with respect to the corresponding component of $x_\alpha$. The stochastic part or the noise $\xi_\alpha(t)$ in the equation of motion is white and gaussian. The noise correlation is given by,

$$\langle \xi_\alpha^i(t)\xi_\beta^j(t') \rangle = 2k_B T \delta_{\alpha\beta} \delta_{ij} \delta(t - t') .$$

(2)

The collective density $\hat{\rho}(x, t)$ is defined in terms of phase space variables $\{x_\alpha\} \equiv \Gamma_N(t)$ as:

$$\hat{\rho}(x, t) = \sum_{\alpha=1}^N \delta(x - x_\alpha(t)) .$$

(3)

Since the number $N$ of particles is constant $\hat{\rho}(x, t)$ is a microscopically conserved quantity. Dean\[14, 17\] obtained the following equation of motion for $\hat{\rho}$:

$$\frac{\partial}{\partial t} \hat{\rho}(x, t) = k_B T \nabla^2 \hat{\rho}(x, t) + \nabla \cdot \{ \hat{\rho}(x, t)\nabla \int dx' U(x - x') \hat{\rho}(x', t) \} + \hat{\zeta}(x, t) ,$$

(4)

where $\nabla$ denotes the derivative operator with respect to components of $x$. The correlation of the random force denoted by $\hat{\zeta}(x, t)$ is obtained as,

$$\hat{\zeta}(x, t) = -\sum_\alpha \nabla_\alpha \cdot \{ \delta(x - x_\alpha(t))\xi_\alpha(t) \} = \nabla \cdot \sum_\alpha \delta(x - x_\alpha(t))\xi_\alpha(t) .$$

(5)

$\hat{\zeta}(x, t)$ is dependent on the phase space coordinates through $x_\alpha$. Eqn. [11] is an exact representation of the stochastic dynamics described by eqns. [11]-[2]. Unlike the time reversible Euler equations, the Dean equation is dissipative since the corresponding microscopic dynamics described by eqn. [11] is dissipative. We average eqn. [11] to obtain a partial differential equation for the corresponding coarse grained density $\rho(x, t)$ which has smooth spatial and temporal dependence. This stochastic equation describes the time dependence of density fluctuations in a non-equilibrium state. The averaging should therefore be done over the corresponding non-equilibrium ensemble. We approximate the latter in terms of the local equilibrium ensemble. This is motivated from the fact that the hydrodynamic description of the non-equilibrium fluid corresponds to the time regime in which it has reached a state of local equilibrium. At this stage the local densities are sufficient to describe the state of the system. The average of the microscopic densities (dependent on phase space
coordinates) over the local equilibrium distribution function $f_{le}$ obtains the corresponding hydrodynamic field with smooth spatial and temporal dependence.

We note that the temperature $T$ enters the exact eqn. (4) only through the first term on the RHS. This is a consequence of using the chain rule of the stochastic differential equations of the Ito calculus in obtaining eqn. (4). Let a set of stochastic variables $x_i(t)$ ($i = 1, .., m$) satisfy the stochastic equation $\dot{x}_i = h_i + g_{ij} \xi_j$ with the correlation of the white noise $\xi_i$ being given by $<\xi_i(t)\xi_m(t')> = \delta_{im}\delta(t - t')$. Ito chain rule [18] obtains the stochastic differential equation for the variable $y(\{x_i\})$ in the form

$$\dot{y} = \sum_i \frac{\partial y}{\partial x_i} \dot{x}_i + \sum_{i,j,k} \frac{1}{2} \frac{\partial^2 y}{\partial x_i \partial x_j} g_{ik} g_{kj}.$$  

(6)

With eqn. (1) for dynamics of $x_\alpha$, the matrix $g_{ij} = \sqrt{2k_B T} \delta_{ij}$ is diagonal. The first term on the RHS of eqn. (4) follows from the $g_{ij}$’s in the second term on the RHS of eqn. (6). This holds even for a system of noninteracting particles. We define next the averaging procedure with respect to the local equilibrium distribution $f_{le}$. The latter is considered to have attained a stage when the system is at temperature $T$. This thermalization process is closely linked to the momentum distribution becoming the Maxwellian. So far the momentum current density has not been included in the formulation. The momentum current density $\hat{g}$ is the flux for number density $\hat{\rho}(x, t)$ in the corresponding balance equation representing number conservation.

$$\frac{\partial}{\partial t} \hat{\rho}(x, t) = -\nabla \cdot \sum_\alpha p_\alpha \delta(x - r_\alpha(t)) = -\nabla \cdot \hat{g}(x, t)$$  

(7)

The probability function for the local equilibrium state is obtained in analogy with that of the equilibrium state [19]. We consider here the distribution function $f_{le}$ in terms of the corresponding local thermodynamic variables $\{\beta(x, t), \mu(x, t), v(x, t)\}$. The distribution $f_{le}(\Gamma'_N; t)$ in the local rest frame, moving with the local velocity $v(x, t)$, is obtained as

$$f_{le}(\Gamma'_N; t) = Q^{-1} \exp\left(-\int d\Gamma \beta(r, t) \left[\hat{e}'(r) - \mu(r, t) \hat{\rho}'(r)\right]\right).$$  

(8)

where $Q$ is the partition function $Q = \int d\Gamma f_{le}(\Gamma_N, t)$. The corresponding microscopically conserved densities of the energy and number of particles in the local frame are respectively denoted by $\hat{e}'$ and $\hat{\rho}'$. The fluid has attained a state with a fixed temperature $T$ ($\beta(r, t) \equiv \beta = 1/(k_B T)$) and the corresponding local equilibrium distribution is given by
The equal time correlation of the momentum density \( g'(x,t) \) in the local rest frame is now obtained as,

\[
\langle \hat{g}'_i(x_1,t)\hat{g}'_j(x_2,t) \rangle_{l.e} = \delta(x_1-x_2)\delta_{ij}\rho(x_1,t)k_BT
\]

(9)

where \( \rho(x,t) \equiv \langle \hat{\rho}(x,t) \rangle_{l.e} \) is the local density.

Having defined the distribution function we now average the microscopic eqn. (4) over the local equilibrium ensemble to obtain an equation for the coarse grained particle density in the following form:

\[
\frac{\partial \rho(x,t)}{\partial t} = k_BT\nabla^2 \rho(x,t) + \nabla \cdot \left[ \int dx' \{ \nabla U(x-x') \} \langle \hat{\rho}(x,t)\hat{\rho}(x',t) \rangle_{l.e} \right] + \zeta(x,t).
\]

(10)

The thermally-averaged equation (10) has stochastic and regular parts. The stochastic part or the noise \( \zeta(x,t) \) is obtained by coarse graining of the microscopic quantity \( \hat{\zeta}(x,t) \) defined in eqn.(5). Correlation of \( \zeta \) is understood as a combination of two steps. First, in \( \hat{\zeta} \) we average the noise \( \xi \) of the microscopic equations of motion over the different configurations of the Brownian particles so as to obtain the coarse grained noise \( \zeta \). This is indicated with a subscript \( \text{l.e} \) on the angular brackets. Second, we correlate the coarse grained noise \( \zeta \) at two different space time points while the equilibrium temperature of the bath is maintained at \( T \).

We denote the latter with a subscript \( B \) on the angular brackets to indicate the averaging over the bath variables. The correlation of the noise \( \zeta \) in the coarse grained equation is obtained by interchanging the order of the two operations stated above,

\[
\langle \zeta(x,t)\zeta(x',t') \rangle = \left\langle \left\langle \hat{\zeta}(x,t) \right\rangle_{l.e} \left\langle \hat{\zeta}(x',t') \right\rangle_{l.e} \right\rangle_B \\
\approx \left\langle \left\langle \hat{\zeta}(x,t)\hat{\zeta}(x',t') \right\rangle_{l.e} \right\rangle_B \\
= 2k_BT\nabla \rho(x,t)\nabla' \delta(x-x')\delta(t-t')
\]

(11)

\( \nabla' \) denotes the derivative operator with respect to the components of \( x' \). In the Markovian approximation of large separation between the characteristic time scales of the solute and that of the solvent variables, the noise correlation is independent of the coarse graining process.

The first and second terms in the RHS of eqn. (10) are in terms of slowly varying quantities. These are conveniently expressed in terms of the Liouville operator \( \mathcal{L} \). For the
$N$ particle system $\mathcal{L}$ is defined as

$$
\mathcal{L} = \sum_{\alpha=1}^{N} \left[ \frac{\partial H}{\partial p_{\alpha}} \frac{\partial}{\partial r_{\alpha}} - \frac{\partial H}{\partial r_{\alpha}} \frac{\partial}{\partial p_{\alpha}} \right],
$$

(12)

where the Hamiltonian $H = K + V$ is a sum of kinetic and potential parts. The momentum dependent or the kinetic energy part is $K = \sum_{\alpha=1}^{N} \frac{p_{\alpha}^2}{2m}$ and the interaction part $V$ is a function of the particle coordinates only. The time rate of change of $\hat{\rho}(x, t)$ is expressed in terms of the operator $\mathcal{L}$ to obtain $i\mathcal{L} \hat{\rho} = -\nabla \cdot \hat{\mathbf{g}} = \dot{\hat{\rho}}(x, t)$. Note however that if $\mathcal{L}$ acts on the momentum current density $\hat{\mathbf{g}}$, it does not obtain the corresponding time derivative.

Using the definitions (12) and (7) respectively of the Liouville operator $\mathcal{L}$ and the current $\mathbf{g}'$ in the local rest frame, we obtain

$$
i\mathcal{L} \hat{g}'_i(x) = -\sum_j \nabla_j \sum_{\alpha=1}^{N} p'^i_{\alpha} p'^j_{\alpha} \delta(x - x_{\alpha}) + \int d\mathbf{x}' \{ \nabla_i U(x - x') \} \dot{\hat{\rho}}(x, t) \hat{\rho}(x', t) .
$$

(13)

Next taking an average over the local equilibrium ensemble in the co-moving frame, we obtain

$$
\langle -i\mathcal{L} \hat{g}'_i(x, t) \rangle_{1e} = k_B T \nabla_i \hat{\rho}(x, t) + \int d\mathbf{x}' \{ \nabla_i U(x - x') \} \langle \dot{\hat{\rho}}(x, t) \hat{\rho}(x', t) \rangle_{1e} .
$$

(14)

Since $\mathcal{L}$ involves derivative operators, integrating by parts and using the property $i\mathcal{L}H = 0$, we obtain

$$
\langle -i\mathcal{L} \hat{g}'_i(x) \rangle_{1e} = \int d\Gamma \hat{g}'_i(x) \frac{1}{Q} i\mathcal{L} \exp \left[ -\beta \{ H - \int d\mathbf{x}' \mu(x') \hat{\rho}(x') \} \right]

= \beta \int d\mathbf{x}' \mu(x') \langle \hat{g}'_i(x) i\mathcal{L} \hat{\rho}(x') \rangle_{1e}

= -\beta \int d\mathbf{x}' \mu(x') \sum_j \langle \hat{g}'_i(x) \nabla_j \hat{g}'_j(x') \rangle_{1e} = \rho(x) \nabla_i \mu(x) .
$$

(15)

In reaching the last equality we have used the key relation (9) for momentum correlation in the local equilibrium distribution which has thermalized at temperature $T$.

The quantity on the RHS of eqn. (15) is a thermodynamic property. We link this to the Helmholtz free energy $F$ expressed as a functional of the inhomogeneous density $\rho(x)$. Using the equilibrium relation $F - G = \Omega \equiv -PV$, where $\Omega$ is the thermodynamic potential, we have in the density functional formalism [20, 21].
\[ F[\rho(x)] \equiv \Omega[\rho(x)] + \int d\mathbf{x} \rho(\mathbf{x}) \mu(\mathbf{x}) . \]

\( \Omega[\rho] \) is a functional of the density obtained from the equivalent result of grand canonical ensemble partition function \( \Omega[\rho(x)] \equiv -k_BT \ln \Xi \). The density functional theory identifies the equilibrium density by minimizing the grand potential \( \delta \Omega/\delta \rho(x) = 0 \). Using the above relations it then follows that the corresponding Helmholtz free energy functional satisfies \( \delta F[\rho]/\delta \rho(x) = \mu(\mathbf{x}) \). Eqn. (10) then reduces with the help of eqns. (14) and (15) to the form

\[ \frac{\partial \rho}{\partial t} = D_0 \nabla \cdot \left[ \rho \nabla \frac{\delta F}{\delta \rho} \right] + \zeta . \]

We have now put the bare diffusion constant \( D_0 = \gamma_0/(\beta \kappa) \) in the RHS above to keep the dimensional factor explicit \[22, 23\]. The free energy \( F[\rho] \) of the density functional theory is expressed (in units of \( k_B T \)) as a sum of two parts \( F[\rho] = F_{\text{id}} + F_{\text{ex}} \), respectively denoting the non-interacting or ideal gas part \( F_{\text{id}}[\rho] \) and the interaction part \( F_{\text{ex}}[\rho] \). The ideal gas part is obtained as

\[ F_{\text{id}}[\rho] = k_B T \int d\mathbf{x} \rho(\mathbf{x}) \left[ \ln \left( \Lambda_0^3 \rho(\mathbf{x}) \right) - 1 \right] . \]

where \( \Lambda_0 = h/\sqrt{2\pi \kappa m k_B T} \) is the thermal wave length. The interaction part or the so called excess part \( F_{\text{ex}}[\rho] \) is generally expressed in a functional Taylor expansion in terms of direct correlation functions \( c^{(i)}(x_1, ..., x_i) \) at a density \( \rho_0 \).

\[ c^{(i)}(x_1, ..., x_i; \rho_0) = \left[ \frac{\delta^i F_{\text{ex}}[\rho]}{\delta \rho(x_1) ... \delta \rho(x_i)} \right]_{\rho=\rho_0} \]

The two point function for \( i = 2 \) is the Ornstein-Zernike direct correlation function \( c^{(2)} \).

Going beyond the over damped limit considered above, the equations of motion of the fluid particles are obtained in terms of both the respective momentum and position coordinates. This is termed as the Fokker-Planck dynamics (FPD). For the \( N \) particle system the momentum \( p_\alpha \) of the \( \alpha \)-th particle is taken as \( \dot{\mathbf{x}}_\alpha(t) \) (mass taken as unity) and its time rate of change is

\[ \frac{d\mathbf{p}_\alpha(t)}{dt} = -\sum_{\beta=1}^{N} \nabla_\alpha U(\mathbf{x}_\alpha(t) - \mathbf{x}_\beta(t)) - \gamma_0 \mathbf{p}_\alpha + \mathbf{\xi}_\alpha(t), \]

\[ \frac{\partial \rho}{\partial t} = D_0 \nabla \cdot \left[ \rho \nabla \frac{\delta F}{\delta \rho} \right] + \zeta . \]

\[ \frac{\partial \rho}{\partial t} = D_0 \nabla \cdot \left[ \rho \nabla \frac{\delta F}{\delta \rho} \right] + \zeta . \]
where \( \gamma_0 \) is a dissipative coefficient. Note that this model does not conserve momentum microscopically unless the dissipative coefficient \( \gamma_0 \) has a \( \nabla^2 \) operator associated with it. Balance equations for the collective densities \( \{ \hat{\rho}(x,t), \hat{g}(x,t) \} \) have been obtained by Nakamura and Yoshimori\[15\]. By averaging these microscopic balance equations, we obtain the corresponding equations of motion for the coarse grained variables \( \{ \rho(x,t), g(x,t) \} \).

\[
\frac{\partial}{\partial t} \rho(x,t) + \nabla \cdot g(x,t) = 0 \tag{21}
\]

\[
\frac{\partial}{\partial t} g_i(x,t) + \gamma_0 g_i(x,t) + \nabla^j \left\langle \left\{ \sum_{\alpha} \frac{p_{\alpha i} p_{\alpha j}}{m} \delta(x - x_{\alpha}(t)) \right\} \right\rangle_{le} + \int dx' \left\{ \nabla_i U(x - x') \right\} \left\langle \hat{\rho}(x,t) \hat{\rho}(x', t) \right\rangle_{le} = \theta_i(x,t) \tag{22}
\]

The noise \( \theta_i(x,t) \) in the generalized Langevin equation is obtained by coarse graining of the noise \( \hat{\theta}_i(x,t) \) which is defined in terms of the noise \( \xi_\alpha \) in the micro-dynamic equations.

\[
\hat{\theta}_i(x,t) = \sum_{\alpha=1}^N \delta(x - x_{\alpha}(t)) \xi^i_\alpha(t) \tag{23}
\]

Using the same approximations as discussed with eqn. \[11\], the correlation of the multiplicative noise \( \theta_i(x,t) \) is obtained as,

\[
\langle \theta_i(x,t) \theta_j(x', t') \rangle = 2k_B T \gamma_0 \rho(x,t) \delta(x - x') \delta(t - t') \tag{24}
\]

We evaluate the third term on the LHS of eqn. \[22\] by making a change of variables \( p_\alpha = p'_\alpha + m v(r'_\alpha) \) and \( r_\alpha = r'_\alpha \) in the locally moving frame with velocity \( v(r'_\alpha) \). Using the symmetry of the distribution function \[8\] for the locally moving frame under the \( p' \rightarrow - p' \) transformation this term reduces to

\[
k_B T \nabla_i \rho(x,t) + \nabla_j \left[ \frac{g_i(x,t) g_j(x,t)}{\rho(x,t)} \right] \tag{25}
\]

Substituting the above result in eqn. \[22\], and using the relations \[14\]-\[15\], which also hold for the Fokker-Planck dynamics, we obtain the following stochastic equation for the coarse grained momentum density

\[
\frac{\partial g_i}{\partial t} + \nabla_j \left[ \frac{g_i g_j}{\rho} \right] + \rho \nabla_i \frac{\delta F}{\delta \rho} + \gamma_0 g_i = \theta_i \tag{26}
\]

Eqn. \[26\] is very similar in form to the corresponding equations of fluctuating nonlinear hydrodynamics for the particles which follow reversible ND\[8\],\[24\]. There is however one
important difference. Unlike in the case of the FPD, for the ND the dissipative term involves a $1/\rho$ nonlinearity \[8\]. On the other hand, in the FPD the noise correlation given by eqn. \[21\] involves the density and hence the noise is multiplicative. The presence of the $1/\rho$ nonlinearity in the ND case is important with respect to the ergodicity-nonergodicity (ENE) transition of the self-consistent mode coupling theory \[25, 26\] of dense liquids. The latter has been extensively used for studying glassy relaxations \[27, 28\]. The ENE transition being referred to is a consequence of feedback effects from slowly decaying density fluctuations in a dense liquid. It has been established \[8\] that the $1/\rho$ nonlinearity in the generalized Langevin equation is essential in smoothing off this transition. For Newtonian dynamics of particles, the single equation \[17\] for $\rho$ has also been obtained \[24\] by eliminating the current $g$ from a field theoretic formulation of the problem. This however requires in the ND case applying the so called adiabatic approximation in which momentum fluctuations decay much faster than the density fluctuations.

In summary, the present work fills up an important gap between a) a nonlinear diffusion equation for the coarse grained density field in terms of thermodynamic direct correlation functions and b) the exact representation of the Brownian dynamics of a system of particles (e.g. Dean’s equation). Thus the single equation \[17\] for $\rho(x, t)$ is the coarse grained form of Dean’s exact equation for the microscopic density $\hat{\rho}(x, t)$. The first equation has been used in a large number of works on fluids and is well known to the community as an ingredient of the dynamic density functional theory. The driving free energy for this coarse grained equation is obtained here in the standard form of density functional theory. The role of the bare interaction potential $\beta U(r)$ in Dean’s equation is replaced by the corresponding direct correlation function $c(r)$. The averaged equation which forms the basis for field theoretic models for the dynamics of fluids, has often been considered (without proof) to be exact for interacting Brownian particles. The effects of nonlinearities in the coarse grained equation are studied with various analytic techniques which are available for calculating renormalized time correlation functions in terms of appropriate mode coupling contributions \[27, 28\]. These models often form the basis for studying dynamics heterogeneities and growing dynamic correlation lengths in supercooled liquids.
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