

Covariant Hydrodynamics of Fluid Membranes

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We derive covariant, reparametrization invariant, hydrodynamical equations for a fluctuating fluid membrane, which describe both tangent-plane motion and shape-changing normal motion. We calculate the renormalization of parameters of a Rouse model in which there is a friction force proportional to the local membrane velocity. Our calculations include both the Fadeev-Popov determinant and a Liouville-like factor needed to ensure rotational invariance.

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Two-dimensional fluid membranes occur in a wide variety of physical and biological systems [1–3]. For example, when surfactant molecules consisting of a polar head group and a hydrocarbon tail are dissolved in water, they self-assemble into flexible bilayer membranes, which can organize into a variety [1,4] of structures, including lamellar phases in which nearly flat membranes form a periodic stack. The statistical and thermodynamic properties of fluid membranes [3] and the phases that they form are well described by a phenomenological model [5] in which membranes are viewed as 2D fluids with energies that depend only on shape, total area, and local 2D mass density if the membrane is compressible.

The dynamical properties of fluid membranes have received far less attention even though many experiments [6] provide dynamical as well as static information. In this paper, we outline the derivation of the equations governing the long-wavelength, low-frequency hydrodynamics of flexible fluid membranes. These equations are invariant under general time-dependent reparametrizations and reduce to those describing a two-dimensional fluid when the membrane is constrained to lie flat and not to change its shape. They also describe low-frequency modes associated with shape variations and couplings between shape modes and the tangent plane modes of a 2D fluid. Our treatment has much in common with that of Goldstein, Langer, and Jackson [7], who consider the dynamics of “fluid” interfaces without internal degrees of freedom.

Membranes can interact in different ways with the medium that surrounds them. Generally, they are imbedded in some 3D fluid, whose dynamics is governed by the 3D Navier-Stokes equations. Interactions between membranes and fluid are then determined by boundary conditions at the membrane-fluid interface. Though our formulation is sufficiently general to treat this situation, we focus our attention on the membrane generalization of the Rouse model [8] in which motion of the membrane relative to a rigid surrounding medium gives rise to a friction force proportional to the local membrane velocity. Applying field-theoretic techniques [9,10] used in the study of dynamic critical phenomena, we calculate

how long-wavelength static and dynamic coefficients of this model are renormalized under removal of high-wave-number degrees of freedom.

In addition to reproducing previous results [11–14] for the renormalization of the bending rigidity and surface tension, our calculations yield two new results of experimental importance. The first is that because of local crumpling [15,16], a nearly flat membrane will be compressible at long length scales even if it is incompressible at the shortest length scales. As a result, it will have a density mode that is *distinct* from the height mode. Both density and height modes are needed to reproduce the experimentally observed hydrodynamic modes [6,17] in a lamellar phases. The second result is that dynamic friction coefficients undergo independent length scale renormalization. Thus, the renormalization of the frequency of dynamical modes cannot be determined by static quantities (such as the bending rigidity) alone.

The field-theoretic description of fluid membranes presents two complications not found in more familiar theories. The first is that the Fadeev-Popov determinant [18] associated with fixing a gauge (or in this case a parametrization of surfaces) depends on membrane configuration. The second is that the number of degrees of freedom of membranes with different areas differ, and statistical sums over membranes of differing areas must be modified via the introduction of a term analogous to the Liouville factor [19,20] of string theories to reflect this difference. These two complications lead to an equilibrium probability distribution in a particular gauge that is not proportional simply to $e^{-\mathcal{H}/T}$ where \mathcal{H} is the Hamiltonian and T the temperature. Corrections to $e^{-\mathcal{H}/T}$ are present even in the purely static theory and will be discussed in detail in a separate publication [20].

Membrane coordinates in 3D Euclidean space are specified by a vector $\mathbf{R}(\tilde{u})$ as a function of a 2D parameter $\tilde{u} = (u^1, u^2)$. The energy of the membrane cannot depend on the way in which \mathbf{R} is parametrized and must, therefore, be invariant under reparametrization transformations of the form $\tilde{u} \rightarrow \tilde{u}' = \Theta(\tilde{u})$. If the membrane is compressible, energies will depend on the local mass density $\rho(\tilde{u})$. The simplest Hamiltonian [5,17] describing a free

compressible membrane is

$$\mathcal{H} = \frac{1}{2} \kappa \int d^2 u \sqrt{g} H^2 + \int d^2 u \sqrt{g} [f(\rho) - \mu \rho], \quad (1)$$

where $g = \det g_{ab}$ is the determinant of the metric tensor $g_{ab} = \partial_a \mathbf{R} \cdot \partial_b \mathbf{R}$ ($a, b = 1, 2$), $g^{ab} g_{bc} = \delta_c^b$, and $H = K_a^a$ is the mean curvature where $K_a^b = g^{bc} K_{ca}$ and K_{ab} is the curvature tensor. $f(\rho)$ is the local Helmholtz free energy density (which could have gradient terms in ρ), and μ is the chemical potential for particles in the membrane. In the Monge gauge, $\tilde{u} = (x, y)$ is a coordinate in the x - y plane, and $\mathbf{R} = (x, y, h(x, y))$ so that $H = \nabla^2 h$ to lowest order in h . The free energy density f can be expanded in powers of the deviation $\delta \rho = \rho - \rho_0$ from the mean-field equilibrium density ρ_0 determined by the equation of state, $\partial f / \partial \rho = \mu$. To harmonic order in $\delta \rho$, $f = f_0 + (1/2) \chi_0^{-1} (\delta \rho)^2 + \mu \delta \rho$ where χ_0 is the harmonic membrane compressibility.

To develop a hydrodynamical theory for any system, we need to identify all conservation laws and broken continuous symmetries. In a free membrane, energy, mass, and momentum are conserved. In addition, rigid translations of the membrane cost no energy. For simplicity, we will ignore energy conservation and assume all processes take place at constant temperature. Thus, the mass density ρ , tangent-plane momentum density j^a , and position \mathbf{R} are the membrane hydrodynamical variables whose characteristic frequencies go to zero at long wavelength. To discuss properties under reparametrization, it is useful to introduce explicit representations for these variables. Let \tilde{u}_0 be some time-independent parametrization, and let $\tilde{u}_{0,\alpha}(t)$ be the coordinate at time t in this space of the particle (molecule) of mass m in the membrane labeled α . Then Euclidean space coordinates of the membrane are given by $\mathbf{R}(\tilde{u}_0, t)$, and the position of particle α is $\mathbf{R}(\tilde{u}_{0,\alpha}(t), t)$. One can now introduce time-dependent parametrization via a time-dependent transformation $\tilde{u}(t) = \Theta_t(\tilde{u}_0)$ and $\tilde{u}_\alpha(t) = \Theta_t(\tilde{u}_{0,\alpha}(t))$, where $\Theta_t(\tilde{u})$ is a single-valued function of \tilde{u} at each t . The membrane mass density is then

$$\rho(\tilde{u}, t) = \frac{1}{\sqrt{g(t)}} \sum_\alpha m \delta(\tilde{u}(t) - \tilde{u}_\alpha(t)). \quad (2)$$

Here $g(t)$ is the determinant of the time-dependent metric tensor $g_{ab}(t)$. This density transforms as a scalar under arbitrary time-dependent reparametrizations; i.e., if $\tilde{u}' = Q_t(\tilde{u})$ for a single-valued function Q_t , then $\rho(\tilde{u}) = \rho'(\tilde{u}')$ where ρ' is the transformed density obtained by replacing \tilde{u}_α by $\tilde{u}'_\alpha = Q_t(\tilde{u}_\alpha)$ in Eq. (2). The tangent-plane momentum density $j^a(\tilde{u}, t)$ is the current of the conserved density ρ :

$$D_t \rho + D_a j^a \equiv \frac{1}{\sqrt{g}} \partial_t \sqrt{g} \rho + \frac{1}{\sqrt{g}} \partial_a \sqrt{g} j^a = 0, \quad (3)$$

where $\partial_t f(\tilde{u}, t) = \partial f(\tilde{u}, t) / \partial t|_{\tilde{u}}$ and

$$j^a(\tilde{u}, t) = \frac{1}{\sqrt{g(t)}} \sum_\alpha m \dot{\tilde{u}}_\alpha^a(t) \delta(\tilde{u}(t) - \tilde{u}_\alpha(t)). \quad (4)$$

The derivatives $D_t = g^{-1/2} \partial_t g^{1/2}$ and D_a are, respectively, temporal and spatial covariant derivatives. The transformation rule for the momentum density under reparametrizations is $j^a(\tilde{u}) = (\partial u^a / \partial u'^b) [j'^b(\tilde{u}') - \partial_t \tilde{u}'^b \rho'(\tilde{u}')] / \rho'$, which reproduce the familiar result $j^a(\tilde{u}) = j'^a(\tilde{u}') + \tilde{v}^a \rho'(\tilde{u}')$ for Galilean transformations $\tilde{u}' = \tilde{u} - \tilde{v} t$ where \tilde{v} is a constant velocity. The conservation law, Eq. (3), is invariant under reparametrizations.

The mass density in 3D Euclidean space expressed in terms of $\rho(\tilde{u})$ is $\rho_3(\mathbf{x}) = \int d^2 u \sqrt{g} \rho(\tilde{u}) \delta(\mathbf{x} - \mathbf{R}(\tilde{u}, t))$. The 3D momentum density $J_3(\mathbf{x}, t)$ can be calculated using $\partial_t \rho_3 + \nabla \cdot \mathbf{J}_3 = 0$. The result is $\mathbf{J}_3(\mathbf{x}) = \int d^2 u \sqrt{g} \mathbf{J}(\tilde{u}) \delta(\mathbf{x} - \mathbf{R}(\tilde{u}, t))$, where

$$\mathbf{J}(\tilde{u}) = j^a \mathbf{e}_a + \rho \partial_t \mathbf{R} \quad (5)$$

is the membrane momentum density with $\mathbf{e}_a = \partial_a \mathbf{R}$ a covariant tangent plane vector. $\mathbf{J}(\tilde{u})$ has components both parallel and perpendicular to the local tangent plane. Components parallel to the tangent plane can arise either from the tangent plane momentum j^a or from $\partial_t \mathbf{R}$, but the component of \mathbf{J} normal to the membrane must come from the $\partial_t \mathbf{R}$. Different parametrizations will lead to different values of j^a and $\mathbf{e}^a \cdot \partial_t \mathbf{R}$, but leave \mathbf{J} unchanged [i.e., $\mathbf{J}(\tilde{u}) = \mathbf{J}'(\tilde{u}')$]. For example, translation of a flat membrane lying in the x - y plane with a constant velocity v in the x direction can be described by $j^a = 0$ and $\mathbf{R} = (x + vt, y, 0)$ or by $j^x = \rho v$, $j^y = 0$, and $\mathbf{R} = (x, y, 0)$.

The dynamical equations for j^a can now be obtained from the force equation [21],

$$\partial_t J_{3,i} + \nabla_j (v_j J_{3,i}) = f_i, \quad (6)$$

where $\mathbf{v}(\mathbf{x}) = \mathbf{J}_3 / \rho_3$ [or equivalently $\mathbf{v}(\tilde{u}) \equiv \mathbf{v}(\mathbf{R}(\tilde{u})) = \mathbf{J} / \rho$] is the membrane velocity, and f_i is the force density arising from membrane forces and dissipative couplings. Equation (6) is the complete nonlinear force equation. It reduces to Euler's equation for a three-dimensional isotropic fluid when $\mathbf{J}_3 = \rho_3 \mathbf{v}$ and $f_i = \nabla_j \sigma_{ij}$, where σ_{ij} is the stress tensor. The contribution to f_i from reversible membrane forces is

$$f_i = - \int d^2 u [\delta \mathcal{H} / \delta R_i(\tilde{u})] \delta(\mathbf{x} - \mathbf{R}(\tilde{u}, t)), \quad (7)$$

where the derivative is taken at constant $\sqrt{g} \rho$ (see [22]). The force equation for \mathbf{J}_3 can be reexpressed as an equation involving only membrane variables:

$$D_t \mathbf{J} + D_a [(j^a / \rho) \mathbf{J}] = - \frac{1}{\sqrt{g}} \frac{\delta \mathcal{H}}{\delta \mathbf{R}(\tilde{u})} + \mathbf{f}_{\text{dis}} + \boldsymbol{\zeta}, \quad (8)$$

where \mathbf{f}_{dis} is the dissipative force density and $\boldsymbol{\zeta}$ is a random Langevin force. This equation is invariant under reparametrizations. When \mathcal{H} is given by Eq. (1), then

$$\frac{1}{\sqrt{g}} \frac{\delta \mathcal{H}}{\delta R_i} = e_i^b \partial_b p + [\kappa Q - \sigma(\rho) H] \mathbf{N}, \quad (9)$$

where $p \equiv -\sigma(\rho) = -(f - \rho \partial f / \partial \rho) = -\sigma + \rho_0 \chi_0^{-1} \delta \rho$ is the 2D membrane pressure, \mathbf{N} is the unit normal

to the membrane, and $Q = H(K_b^a - \frac{1}{2}\delta_b^a H)(K_a^b - \frac{1}{2}\delta_a^b H) + D^2 H$, where D^2 is the covariant Laplacian, $g^{-1/2}\partial_a g^{1/2}g^{ab}\partial_b$. To linear order in the Monge gauge, $\kappa Q - \sigma H = -\sigma\nabla^2 h + \kappa\nabla^4 h$. If motion perpendicular to the membrane is prohibited, then all properties are intrinsic and Eqs. (3) and (8) reduce to equations such as those used in general-relativistic hydrodynamics [23]. They are also equivalent to the equations derived in Ref. [24] when $Q = 0$.

The dissipative force density \mathbf{f}_{dis} in general has contributions arising from interactions of the membrane with its surrounding medium and from intramembrane viscous forces. The latter are subdominant when the former are present, and we will ignore them. Interactions with the surrounding medium can be described by a phenomenological friction force proportional to the difference between the membrane velocity $\mathbf{v} = \mathbf{J}/\rho = v_n \mathbf{N} + v^a \mathbf{e}_a$ and the medium velocity $\mathbf{V} = V_n \mathbf{N} + V^a \mathbf{e}_a$ at the membrane:

$$\mathbf{f}_{\text{dis}} = -\gamma_n(v_n - V_n)\mathbf{N} - \gamma_t(v^a - V^a)\mathbf{e}_a, \quad (10)$$

with different friction coefficients γ_n and γ_t for flow perpendicular and parallel to the membrane. No slip boundary conditions are obtained by setting $\gamma_n = \gamma_t = \infty$, and the Rouse model by setting $\mathbf{V} = 0$.

We now restrict our attention to the Rouse model at low frequency where the inertial and nonlinear terms on the *left hand side* of Eq. (8) can be neglected. We also retain only linear terms in $\delta\rho$ in the force. In this case, the dynamical equations become

$$\begin{aligned} F_\rho &= D_t(\rho_0 + \delta\rho) - (\rho_0^2\chi_0^{-1}/\gamma_t)D^2\delta\rho \\ &\quad - \rho_0 D_a(\partial_t \mathbf{R} \cdot \mathbf{e}^a) + (\rho_0/\gamma_t)D_a\zeta^a = 0, \\ F_n &= \mathbf{N} \cdot \partial_t \mathbf{R} + (\kappa Q - \sigma H)/\gamma_n \\ &\quad + (\rho_0\chi_0^{-1}/\gamma_n)H\delta\rho - \zeta_n/\gamma_n = 0, \end{aligned} \quad (11)$$

where $\zeta_n = \boldsymbol{\zeta} \cdot \mathbf{N}$ and $\zeta^a = \boldsymbol{\zeta} \cdot \mathbf{e}^a$. Equations (11) define the functions F_n and F_ρ . In the Monge gauge, the equation for F_n can be written as

$$F_h = \sqrt{g}F_n = \partial_t h + \left. \frac{\sqrt{g}}{\gamma_n} \frac{\delta \mathcal{H}}{\delta h} \right|_\rho - \frac{\zeta_h}{\gamma_n} = 0, \quad (12)$$

where $\sqrt{g} = [1 + (\nabla h)^2]^{1/2}$ and $\zeta_h = \sqrt{g}\zeta_n$ and where we used the fact that $n_3 = g^{-1/2}$. When changes in density are suppressed, this equation reduces to the nonlinear rotationally invariant Kardar-Parisi-Zhang equation [25] for interface growth in the limit of zero velocity. In the harmonic limit and in the absence of noise, the equations for ρ and h reduce to

$$\partial_t \rho = \frac{\rho_0^2}{\chi_0 \gamma_t} \nabla^2 \delta\rho, \quad \partial_t h = -\frac{1}{\gamma_n} (-\sigma \nabla^2 + \kappa \nabla^4) h, \quad (13)$$

and predict a density mode with dispersion $\omega = -i(\rho_0^2\chi_0^{-1}/\gamma_t)q^2$ and a height mode with dispersion $\omega = -i(\sigma q^2 + \kappa q^4)/\gamma_n$. The density mode is identical to that for thin films on a solid substrate [26].

The Langevin noise $\zeta(\bar{u}, t)$ is a stochastic force arising from high-wave-number degrees of freedom. Its statistical properties should be chosen so that the time-dependent probability distribution for observable decays to the equilibrium distribution at long times. In flat-space systems without a gauge symmetry, the thermal equilibrium probability distribution P is proportional to $e^{-\mathcal{H}/T}$, and this condition is met by choosing the Langevin noise to have a zero mean and a white noise spectrum. In the present case, the equilibrium distribution in a particular gauge is proportional to $\Delta_{\text{FP}}\Delta_L e^{-\mathcal{H}/T}$ where Δ_F is the Fadeev-Popov determinant [18] and Δ_L is the Liouville factor [19,20] relating the measure of a general fluctuating surface to a reference surface with a grid in which each area element in parameter space maps onto equal area elements of the physical surface. In the Monge gauge [14,20] $\Delta_{\text{FP}} = \prod_{\mathbf{x}} g^{-1/2}$, and $\Delta_L = \exp[-\mu_1 \int d^2x (\nabla h)^2]$ to lowest order in h , where μ_1 is a temperature-dependent potential. To lowest order in the temperature, $\mu_1 = \partial\delta\tau/\partial\Lambda_x$, where $\delta\tau = T \int^\Lambda [d^2q/(2\pi)^2] \ln[(\sigma q^2 + \kappa q^4)\lambda^2/T]$ is the one-loop correction to the surface tension with Λ the wave-number cutoff, Λ_x the cutoff along the direction x , and λ the thermal wavelength. The factors Δ_{FP} and Δ_L lead to the probability distribution $P \sim \exp[-(\mathcal{H} + \mathcal{H}_\Delta)T]$ where to lowest order in $(\nabla h)^2$, $\mathcal{H}_\Delta = \mu_\Delta \int d^2x (\nabla h)^2$, where $\mu_\Delta = T \int^\Lambda d^2q/(2\pi)^2 + \mu_1$. If we choose $\langle \zeta_h \rangle = -\sqrt{g}(\delta\mathcal{H}_\Delta/\delta h)$ and

$$\langle \delta\zeta_i(\bar{u}, t) \delta\zeta_j(\bar{u}', t') \rangle = 2T\gamma_{ij}g^{-1/2}\delta(\bar{u} - \bar{u}')\delta(t - t'), \quad (14)$$

where $\gamma_{ij} = \gamma_n n_i n_j + \gamma_t e_i^a e_{aj}$, then the dynamic probability distribution will decay to $e^{-(\mathcal{H} + \mathcal{H}_\Delta)/T}$ at long time.

Equations (11) and (12) are nonlinear and lead to couplings between the density and height modes. To study the effects of these couplings perturbatively, it is convenient to introduce a generating function similar to those used in the study of stochastic equations and dynamic critical phenomena [9,10], and we define $Z = \langle \int dF_\rho dF_h \delta(F_\rho) \delta(F_h) \rangle$ where the brackets $\langle \rangle$ signify an average over the random noise and F_ρ and F_h are the equations of motion defined in Eqs. (11) and (12). We then introduce fields $\hat{\rho}$ and \hat{h} to provide an integral representation of the delta functions. The generating function is then $\hat{Z} = \int \mathcal{D}[\hat{\rho}] \mathcal{D}[\hat{h}] \mathcal{D}[\delta\rho] \mathcal{D}[h] e^{-L}$ where $L = \int dt d^2x \mathcal{L}$ and

$$\begin{aligned} \mathcal{L} &= \hat{h} \left\{ \partial_t h + \sqrt{g}\gamma_n^{-1} [\kappa Q - \sigma(\rho)H + \delta\mathcal{H}_\Delta/\delta h] \right. \\ &\quad + \sqrt{g}\hat{\rho} [D_t(\rho_0 + \delta\rho) - (\rho_0^2\chi_0^{-1}/\gamma_t)D^2\delta\rho \\ &\quad \quad \quad \left. - \rho_0 D_a(\partial_t \mathbf{R} \cdot \mathbf{e}^a)] + \sqrt{g}(\rho_0^2 T/\gamma_t) D_a \hat{\rho} D^a \hat{\rho} \right. \\ &\quad \left. - \ln J + T\sqrt{g}\gamma_n^{-1} \hat{h}^2 \right\}. \end{aligned} \quad (15)$$

Here J is related to the Jacobian of the transformation from F_ρ and F_h to $\delta\rho$ and h . It plays no role in the one-loop calculations we present here.

We can now calculate how parameters in \mathcal{L} renormalize under the removal of high-wave-number degrees of freedom. Using standard renormalization procedures, we remove height degrees of freedom with wave numbers in the shell $\Lambda/e' < q < \Lambda$ and rescale wave number according to $q \rightarrow e^{-l}q$ without rescaling fields. Our results are

$$\frac{d\sigma}{dl} = \frac{T\Lambda^2}{4\pi} \ln[(\kappa\Lambda^2 + \sigma)\lambda^2/T], \quad (16)$$

$$\frac{d\kappa}{dl} = -\frac{3T}{4\pi} \frac{\kappa\Lambda^2}{\kappa\Lambda^2 + \sigma}, \quad (17)$$

$$\frac{d\chi}{dl} = \frac{\chi T}{4\pi} \frac{\Lambda^2}{\kappa\Lambda^2 + \sigma} + \frac{\rho_0^2 T}{4\pi} \frac{\Lambda^2}{(\kappa\Lambda^2 + \sigma)^2}, \quad (18)$$

$$\frac{d\gamma_t}{dl} = \frac{T}{4\pi} \gamma_t \left(2 - \frac{\gamma_t}{\gamma_n}\right) \frac{\Lambda^2}{\kappa\Lambda^2 + \sigma}, \quad (19)$$

$$\frac{d\gamma_n}{dl} = -\frac{T}{4\pi} \gamma_n \frac{\Lambda^2}{\kappa\Lambda^2 + \sigma}. \quad (20)$$

There are several observations to make about these results. First, the equations governing the renormalization of κ and σ are identical to those obtained previously [11–13] from static equilibrium calculations. Second, Eq. (18) for equilibrium compressibility is a new result showing that χ at length scale Λ'^{-1} is greater than χ at the shorter length scale Λ^{-1} . The physical origin of this effect is membrane crumpling. It is easier to compress a crumpled than a flat membrane. At the molecular length scale, there is no crumpling, but at larger scales there is. Even if the membrane is incompressible [$\chi_0 = \chi(\Lambda) = 0$] at the molecular length scale, it is necessarily compressible at longer length scales. Third, we find nontrivial rescaling of the friction coefficients γ_n and γ_t implying that the renormalization of dynamical modes cannot be determined by renormalization of static coefficients alone. Finally, we emphasize the necessity of including both Fadeev-Popov and Liouville factors to obtain the proper rotationally invariant renormalization of the surface tension σ from a correlation function rather than from a free energy. In static calculations, one can always use the free energy to calculate σ , but the same Fadeev-Popov and Liouville corrections are required to obtain σ from a height-height correlation function. Details of the derivation of the Liouville factor Δ_L appear in a separate paper [20]. Free energies cannot be calculated in a dynamical calculation, and σ must be obtained from a correlation function.

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