Energy dissipation of fluid membranes

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We establish a dissipation functional for the in-plane viscosity of a fluid membrane. Guiding principles are a reparametrization and Galilean invariance and the requirement that the dissipation rate of a uniformly rotating membrane vanishes. From the dissipation functional we derive the equations of motion of the membrane. As an illustration, these equations are applied to study the dynamics of a nearly spherical vesicle.

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I. INTRODUCTION

Fluid membranes are flexible thin films built up by amphiphilic molecules [1]. Since the molecules can diffuse freely in the membrane, the latter may be seen as a two-dimensional fluid. Important macroscopic properties of membranes are bending elasticity, incompressibility, and viscosity. The basic static model for a continuum description of a membrane was given by Helfrich [2]. In his model the elastic energy of the membrane is determined by its local curvature alone. For the dynamical properties of membranes, however, hydrodynamic interactions are relevant. Two types of hydrodynamic interactions can be discerned: flow and dissipative processes inside the membrane, and the corresponding processes within the surrounding medium. So far, most discussions on the dynamics of membranes have considered the coupling of curvature forces of the membrane to hydrodynamic modes in the surrounding medium [3–5]. The internal viscosity of the membrane material has generally been neglected against the volume viscosity. For exceptions see [6,7], and [8] where the dissipation resulting from the friction between the layers of a bilayer membrane is considered. The inclusion of the membrane viscosity typically leads to modifications of dispersion relations.

In the present paper we concentrate on the internal viscoelastic dynamics of the fluid membrane. The degrees of freedom of the embedding medium are neglected completely. We will show in Sec. IV that there exists a dimensionless parameter β, which decides whether the in-plane or the bulk dissipation dominates. This parameter β is the ratio of the linear vesicle size R and a characteristic length $R_0=\nu/\eta$ determined by the in-plane and the bulk viscosities ν and η, respectively. For $R \ll R_0$ the bulk viscosity dominates and the theory of [3–5] is appropriate. In the opposite case $R \gg R_0$ our approach applies.

In order to describe the regime of dominating in-plane viscosity, we derive a dissipation functional. In the derivation, the covariance of the membrane with respect to reparametrizations is accounted for, together with Euclidean invariance and the invariance against uniform rotations. Covariance is essential to describe a fluid membrane in which there is no preferred coordinate system. Euclidean invariance is required in order to exclude dissipation in uniform translations of the membrane. The dissipation functional, together with the curvature energy, leads to equations of motion, which then are applied to the dynamics of almost spherical vesicles.

II. DISSIPATION OF ENERGY

The in-plane viscosity can be understood as resulting from the friction between two adjacent fluid lines of different flow velocities. Thus velocity gradients lead to dissipation of kinetic energy. The rate of dissipation can be described by a functional which should have the following properties: (i) $\partial_t E$ is negative semidefinite and covariant, reflecting fluidity [2]; (ii) $\partial_t E$ only depends on gradients of the tangential velocity field; (iii) a uniform translation or rotation does not lead to dissipation.

In order to embody these requirements, we first need to have a mathematical description of the membrane shape and its kinematics. This will be based on standard techniques of differential geometry [7]. The membrane is parametrized through the Euclidean vector

$$\vec{X}(\sigma;t)=\begin{bmatrix} X_1(\sigma;t) \\ X_2(\sigma;t) \\ X_3(\sigma;t) \end{bmatrix}, \tag{1}$$

where $(\sigma^1,\sigma^2)=:\sigma$ are the internal coordinates of the plane.

In coordinates moving with the fluid elements, the velocity field $\vec{U}=\partial_t\vec{X}(\sigma,t)$ can be decomposed in terms of the tangential vectors $\partial_i\vec{X}\equiv\partial\vec{X}/\partial\sigma^i$ and the normal vector $\vec{N}$ as $u_i=\partial_i\vec{X}\cdot\vec{U}$, and $v_\perp=\vec{N}\cdot\vec{U}$. The normal part $v_\perp$ is due to variations of the membrane shape. Analogous to conventional hydrodynamics [11] one may start with a dissipation functional that is bilinear in the covariant derivatives of the tangential flow, $A_{ij}:=D_iu_j$.

The dissipation functional then is given by the most general bilinear form

$$\partial_t E=-\frac{\nu}{2}\int d^D\sigma\sqrt{g}\,A_{ij}A^{ij} -\frac{\tilde{\nu}}{2}\int d^D\sigma\sqrt{g}\,A_{ij}A^{ji}\,, \tag{2}$$

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where $\sqrt{g}\,d^D\sigma$ is the invariant area element with $D=2$, and $g=\det(g_{ij})$ is the determinant of the metric tensor $g_{ij}=\partial_i\vec{X}\cdot\partial_j\vec{X}$. A term $(A_i^i)^2$ is not included since it does not describe shear flow but a compression. For an incompressible membrane, $A_i^i=2Hv_\perp$ (H is the mean curvature) [12]. The expression (2) is covariant and positive semidefinite for $\nu\geq\tilde{\nu}$, but not yet invariant under a uniform translation with a constant velocity $\vec{V}$. Namely, the velocity field $\vec{U}$ and its components transform like

$$\vec{U}'(\sigma,t)=\vec{U}(\sigma,t)+\vec{V}\ ,$$
$$u_j'=u_j+\partial_j\vec{X}\cdot\vec{V}\ ,$$
$$v_\perp'=v_\perp+\vec{N}\cdot\vec{V}\ . \tag{3}$$

Hence in terms of the extrinsic curvature-tensor $K_{ij}=-\partial_i\vec{X}\cdot\partial_j\vec{N}$ we obtain

$$\begin{aligned}\partial_t E'=&-\frac{\nu}{2}\int d^D\sigma\sqrt{g}\,(D_iu_j')(D^iu'^j)\\&-\frac{\tilde{\nu}}{2}\int d^D\sigma\sqrt{g}\,(D_iu_j')(D^ju'^i)\\=&\,\partial_tE\\&-\frac{\nu}{2}\int d^D\sigma\sqrt{g}\,[2u_jD_i(K^{ij}\vec{N}\cdot\vec{V})+K_{ij}K^{ij}(\vec{N}\cdot\vec{V})^2]\\&-\frac{\tilde{\nu}}{2}\int d^D\sigma\sqrt{g}\,[2u_jD_i(K^{ji}\vec{N}\cdot\vec{V})+K_{ij}K^{ji}(\vec{N}\cdot\vec{V})^2]\ .\end{aligned} \tag{4}$$

Now, the additional terms in Eq. (4) can be absorbed into a redefinition of A_{ij}. From (3), one has $D_iu_j'=D_iu_j+K_{ij}(v_\perp-v_\perp')$, which suggests to replace A_{ij} in (2) by the covariant expression

$$A_{ij}:=D_iu_j-K_{ij}v_\perp\equiv\partial_i\vec{U}\cdot\partial_j\vec{X}\ . \tag{5}$$

This ensures the Galilean invariance of (2).

However, the dissipation functional (2) in general still does not vanish for a uniform rotation where $\vec{U}=\vec{\omega}\times\vec{X}$, i.e.,

$$A_{ij}=\partial_j\vec{X}\cdot\partial_i\vec{U}=\vec{\omega}\cdot(\partial_i\vec{X}\times\partial_j\vec{X})=-A_{ji}\ . \tag{6}$$

As a consequence, a uniform rotation is dissipation free only if $\nu=\tilde{\nu}$, implying

$$\begin{aligned}\partial_tE=&-\frac{\nu}{2}\int d^D\sigma\sqrt{g}\,(A_{ij}A^{ij}+A_{ij}A^{ji})\\\equiv&-\nu\int d^D\sigma\sqrt{g}\,A_{(ij)}A^{(ij)}\ ,\end{aligned} \tag{7}$$

with $A_{(ij)}=\frac{1}{2}(A_{ij}+A_{ji})$. In coordinates moving with the fluid elements, the tensor $A_{(ij)}$ is given by the time derivative of the metric tensor g_{ij}:

$$\partial_tg_{ij}=\partial_i(\partial_t\vec{X})\cdot\partial_j\vec{X}+\partial_i\vec{X}\cdot\partial_j(\partial_t\vec{X})=A_{ij}+A_{ji}\ . \tag{8}$$

Hence in these coordinates, the dissipation functional (7) reads

$$\partial_tE=-\frac{\nu}{4}\int d^D\sigma\sqrt{g}\,\partial_tg^{ij}\partial_tg_{ij}\ . \tag{9}$$

The dissipation functional (7) is the central result of our paper. As explained it is negative semidefinite, covariant, and invariant under uniform translations and rotations.

III. THE EQUATION OF MOTION

The discussion of hydrodynamics of the membrane will be restricted to a Stokes-like regime. We accordingly balance the friction forces with the curvature and constraint forces, neglecting inertial terms. The resulting equation of motion reads

$$-\frac{1}{2}\frac{\delta_c(\partial_tE)}{\delta_c\vec{U}}=-\frac{\delta_c\mathcal{H}}{\delta_c\vec{X}}+g^{ij}\partial_i\phi\partial_j\vec{X}+\phi\Delta_{\rm cov}\vec{X}\ , \tag{10}$$

and is similar to the model of Ref. [12] but with a more realistic friction force. In (10) $\mathcal{H}$ is the Helfrich Hamiltonian

$$\mathcal{H}=2\kappa\int d^D\sigma\sqrt{g}\,H^2 \tag{11}$$

with the mean curvature $H=\frac{1}{2}K_i^i$ and bending rigidity κ, and ϕ means a Lagrange multiplier related to the constraint of incompressibility described by local conservation of the area

$$2v_\perp H-D_iu^i=A_i^i=0\ . \tag{12}$$

From (7) we obtain

$$\begin{aligned}\frac{\delta_c(\partial_tE)}{\delta_c\vec{U}}&=2\nu D_i(\partial_j\vec{X}A^{(ij)})\\&=2\nu\vec{N}K_{ij}A^{ij}+2\nu(D_iA^{(ij)})\partial_j\vec{X}\ ,\end{aligned} \tag{13}$$

which after insertion into (10) and decomposition into normal and tangential parts yields

$$-\nu K_{ij}A^{ij}=-\vec{N}\cdot\frac{\delta_c\mathcal{H}}{\delta_c\vec{X}}+2H\phi\ , \tag{14}$$

$$-\nu g_{jk}D_iA^{(ij)}=\partial_k\phi\ . \tag{15}$$

Equations (14) and (15) constitute another important result of our paper. Together with Eqs. (11) and (12) they represent a closed description of an incompressible viscous membrane. Problems arise if the system has modes with a vanishing friction force, i.e., modes that leave the metric tensor invariant: $\delta g_{ij}=0$. In this case our equation of motion is meaningless and external friction forces should be included.

IV. THE DEFORMED SPHERE

Now we apply the above results to an almost spherical vesicle. Consider a sphere with radius R as a reference configuration $\vec{X}_0$ where $H_0=-1/R$ and $K_{0,ij}=-(1/R)g_{0,ij}$. For small deviations $\xi(\varphi,\vartheta,t)$ (with spherical coordinate φ,ϑ) from the unit sphere we have $\vec{X}(\varphi,\vartheta,t)=\vec{X}_0+\xi(\varphi,\vartheta,t)\vec{N}_0$, which, to linear order in ξ, implies $v_\perp=\partial_t\xi$. We furthermore decompose the tangential flow into a gradient and a rotational part

$$u_i = \partial_i \psi + \epsilon_{0,i}^{j} \partial_j \rho \ ,$$

with $\epsilon_{0,ij} \equiv \vec{N}_0 \cdot [(\partial_i \vec{X}_0) \times \partial_j \vec{X}_0]$. Equations (14), (15), and (12), linearized in ψ, ρ, and ξ then read

$$\kappa \left[\Delta_0^2 \xi + \frac{2}{R^2} \Delta_0 \xi \right] = -\frac{2}{R} \phi \ , \tag{16}$$

$$\partial_j \left[\Delta_0 \psi + \frac{2}{R^2} \psi \right] + \epsilon_{0,j}^{l} \partial_l \left[\Delta_0 \rho + \frac{2}{R^2} \rho \right] = -\frac{2}{\nu} \partial_j \phi \ , \tag{17}$$

$$-\frac{2}{R} \partial_t \xi = \Delta_0 \psi \ . \tag{18}$$

Integration of the tangential part (17) yields

$$\Delta_0 \psi + \frac{2}{R^2} \psi + \frac{2}{\nu} \phi = \mathrm{const}_1 \ , \tag{19}$$

$$\Delta_0 \rho + \frac{2}{R^2} \rho = \mathrm{const}_2 \ . \tag{20}$$

Since ρ and ψ are assumed to be small, we find $\mathrm{const}_1 = \mathrm{const}_2 = 0$.

The quantities ψ and ϕ can now be eliminated from Eqs. (16), (18), and (19), with the result

$$\left[\Delta_0 + \frac{2}{R^2} \right] \partial_t \xi = -\frac{\kappa R^2}{2\nu} \Delta_0^2 \left[\Delta_0 + \frac{2}{R^2} \right] \xi \ . \tag{21}$$

For all modes with $l \neq 2$ this reduces to the remarkably simple result

$$\partial_t \xi = -\frac{\kappa R^2}{2\nu} \Delta_0^2 \xi \ . \tag{22}$$

This, e.g., shows that the condition of incompressibility in the present case does not lead to long-ranged interactions. Equation (22) has solutions for $l \geq 2$

$$\xi_{lm}(\varphi, \vartheta, t) = c_{lm} e^{-\alpha_l t} Y_{l,m}(\varphi, \vartheta) \tag{23}$$

with spherical harmonics $Y_{l,m}$ and the damping constants

$$\alpha_l = \frac{\kappa}{2\nu R^2} l^2 (l+1)^2 \ . \tag{24}$$

For these modes, the tangential flow is a pure potential flow since $\rho = 0$.

The $l = 1$ modes describe a uniform translation (ψ) or rotation (ρ). The $l = 0$ mode is constant, corresponding to conservation of total area.

We will now compare our results to the model given by Milner and Safran in [4]. They calculate damping constants ω_l for the modes of a spherical vesicle taking into account the viscosity of the surrounding medium but neglecting in-plane dissipation. They result in

$$\omega_l = \frac{\kappa}{\eta R^3} \frac{[l(l+1) - \gamma_{\mathrm{eff}}] l(l+1)(l+2)(l-1)}{(2l+1)(2l^2+2l-1)} \ ,$$

where γ_{eff} is a dimensionless effective surface tension running from 0 to 8 and η is the volume viscosity of water. For $\gamma_{\mathrm{eff}} = 0$ the ratio of our damping constant (24) to the one of Milner and Safran is

$$\frac{\omega_l}{\alpha_l} = \frac{1}{\beta} \frac{2(l+2)(l-1)}{(2l+1)(2l^2+2l-1)} \ , \tag{25}$$

where β is the dimensionless ratio $\beta := R/R_0$ with $R_0 := \nu/\eta$. Typical values for the in-plane viscosity are of the order of 10^{-7} N s/m [9,10] while the volume viscosity of water is about 10^{-3} N s/m^2. Assuming a droplet of typical size (10 μm) we obtain $\beta = 10$, e.g., a ratio $\omega_{l=2}/\alpha_{l=2} = 1.45$, indicating that both dissipative mechanisms are of the same order. For vesicles with a linear size $R \gg R_0$ ($\beta \gg 1$) the in-plane damping will dominate.

In order to obtain experimental results for the damping constants ω_l, one may measure optically the time correlation of fluctuating vesicles, as in the experiments of [3]. In our region the time correlation function for the deviation from the sphere is given by

$$\langle \xi_{l,m}(0) \xi_{l,m}(t) \rangle = \frac{kTR^2}{\kappa [2l - l(l+1)]^2} \exp(-\alpha_l t) \ , \tag{26}$$

where the fluctuation-dissipation theorem is used. α_l is our damping constant and the prefactor of the exponential comes from the static Helfrich energy as in [3].

V. SUMMARY

We have considered a regime of membrane dynamics, where the internal viscosity of the membrane dominates other dissipative processes and also inertial terms. To start with, we wrote down a functional, which describes the dissipation of energy, deriving from gradients of the tangential velocities in the membrane. This functional is covariant, negative semidefinite, and is not affected by uniform translations or rotations. From this functional, we derived equations of motion for a membrane corresponding to the Stokes regime in ordinary hydrodynamics, but living on a two-dimensional curved manifold.

These equations have been applied to a nearly spherical vesicle. The resulting linearized equation for radial deviations from the sphere is remarkably simple and does not show long-ranged interactions that one would expect in case of an incompressible membrane.

Comparing our result to another model taking into account volume dissipation in the surrounding medium, we demonstrate the relevance of in-plane dissipation and give the range of validity.

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