Dynamics and rheology of a dilute suspension of vesicles: higher order theory

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Abstract

Vesicles under shear flow exhibit various dynamics: tank-treading (tt), tumbling (tb) and vacillating-breathing (vb). The vb mode consists in a motion where the long axis of the vesicle oscillates about the flow direction, while the shape undergoes a breathing dynamics. We extend here the original theory [C. Misbah, Phys. Rev. Lett. **96**, 028104 (2006)] to the next order in a consistent manner. The consistent higher order theory reveals a direct bifurcation from tt to tb if $C_a \equiv \tau\dot{\gamma}$ is small enough—typically below 0.5 (τ= vesicle relaxation time towards equilibrium shape, $\dot{\gamma}$=shear rate). At larger C_a the tb is preceded by the vb mode. For $C_a \gg 1$ we recover the leading order original calculation, where the vb mode coexists with tb. The consistent calculation reveals several quantitative discrepancies with recent works, and points to new features. We briefly analyse rheology and find that the effective viscosity exhibits a minimum at $tt - tb$ and $tt - vb$ bifurcation points. At small C_a the minimum corresponds to a cusp singularity, while at high enough C_a the cusp is smeared out.

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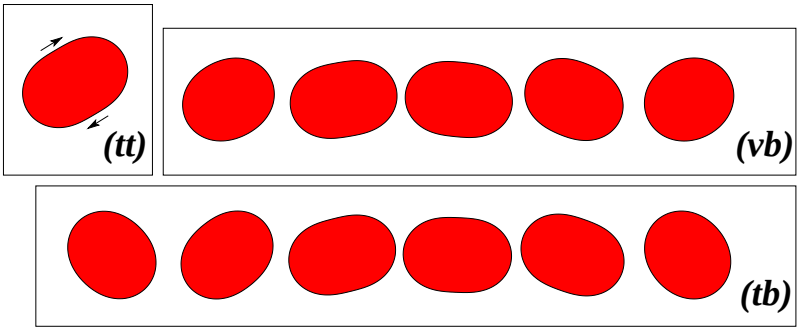

FIG. 1: The three types of motion are shown. The arrows for the tt regime refer to the tank-treading motion of the membrane. Note that for the tb the vesicle long axis makes a full rotation by an angle π, while in the vb the long axis executes an oscillation about the horizontal axis.

I. INTRODUCTION

Vesicles are closed membranes suspended in an aqueous medium. They constitute an interesting starting model for the study of dynamics of real cells, such as red blood cells. The study of their rheology should capture some essential features of blood rheology.

Under a linear shear flow, a vesicle (where the membrane is in its fluid state) is known to exhibit a tank-treading (tt) motion, while its long axis makes an angle, $\psi < \pi/4$, with the flow direction[1, 2]. In the presence of a viscosity contrast $\lambda = \eta_1/\eta_0$ (η_1 and η_0 are the internal and external viscosities, respectively), ψ decreases until it vanishes at a critical value of $\lambda = \lambda_c$. For a small enough $C_a \equiv \tau\dot{\gamma}$ (τ is the relaxation time towards the equilibrium shape in the absence of an imposed flow, $\dot{\gamma}$=shear rate) the tt exhibits a saddle-node bifurcation towards tumbling (tb)[3].

Recently, a new type of motion has been predicted[4], namely a vacillating breathing (vb) mode: the vesicle's long axis undergoes an oscillation (or vacillation) around the flow direction, while the shape executes a breathing motion. The three types of motion (tt, tb and vb) are shown on Fig. 1.

Shortly after this theoretical prediction, an experimental report on this type of mode has been presented[5] (the authors have used the term trembling in order to refer to the vb mode; actually trembling may evoke some kind of noisy dynamics, and seems to us an inappropriate denomination, since the vb mode is periodic in time) and in [6] a qualitatively similar motion called "transition motion" in the vicinity of the $tt - tb$ transition has been

observed. Nevertheless, a detailed experimental study of this vb mode would be interesting but has not been reported yet. Since then, works providing further understanding[7], or attempting[8, 9] to extend the original theory[4] to higher order deformation (with the aim to account for the experimental observation[5]) have been presented. Interesting features have emerged[8, 9] regarding the behavior of the vb mode as a function of C_a.

The first aim of this paper is to present the result of the consistent theory regarding the higher order calculation. We find significant differences with a recent work[8, 9] regarding the form of the evolution equation. This implies, in particular, that the location of the boundaries separating the various three regimes in parameter space is significantly affected. Furthermore, by accounting properly for higher order terms, it is shown that contrary to the belief in[8] the so-called self similarity (in that only two independent parameters survive in the final evolution equations) does not hold.

A second important report is to investigate how the effective viscosity derived recently in[4, 10] is affected by the higher order deformation. In the tt regime the effective viscosity derived in[4] is still a decreasing function of λ and is only slightly shifted by the higher order terms. It is found that for a small enough C_a the effective viscosity of the suspension (as a function of λ) still exhibits a cusp singularity at the $tt - tb$ bifurcation as reported in[10], while the cusp becomes a smooth minimum when C_a is high enough, namely when the $tt-vb$ bifurcation occurs.

The scheme of the paper is as follows. In Section II we present in details how a consistent calculation should be made, and develop the various necessary steps, including geometry, the calculation of the velocity field, and the treatment of the boundary conditions. Section III deals with the derivation of the leading order solution, whereas Section IV presents the outcome of the next order solution. The final evolution equation is presented in Section V, and a comparison with previous works is given in VI. Section VII is devoted to the main results of dynamics, and a brief discussion is devoted to rheology in Section VIII. Section IX is dedicated to a discussion and conclusion.

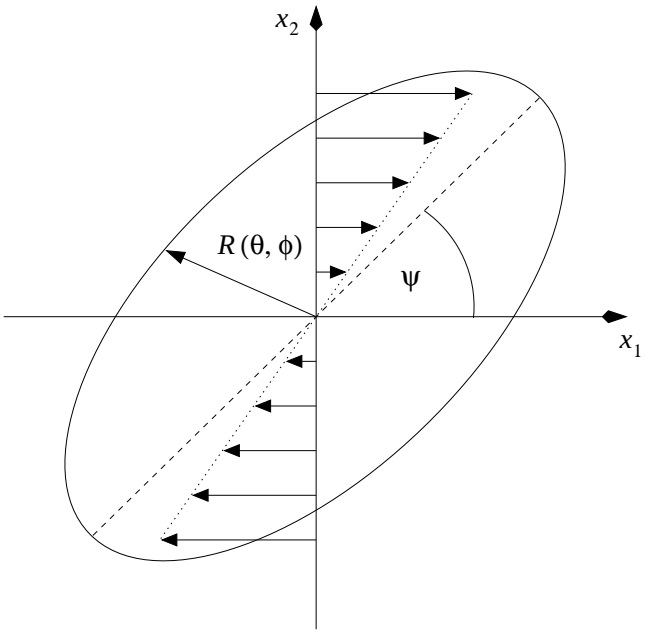

FIG. 2: Various geometrical quantities.

II. ASYMPTOTIC EXPANSION

A. Vesicle geometry

The various geometrical quantities are shown on Fig.2. The time-dependent vesicle surface is described by $\mathbf{R} = R(\theta, \phi, t)\, \mathbf{e}_r$ where

$$R(\theta, \phi, t) = r_0 \left[1 + \epsilon f(\theta, \phi, t)\right]. \tag{1}$$

Here r_0 is the radius of the equivalent sphere of the vesicle and ϵ is a small parameter, which may be related to the excess area Δ via $\Delta = \epsilon^2$. This parameter serves formally as an expansion parameter ($\epsilon = 0$ is a sphere)[7]. For the following it is convenient to rescale spatial variables by r_0, so that we are left with

$$R(\theta, \phi, t) = 1 + \epsilon f(\theta, \phi, t). \tag{2}$$

Several notations and the spirit of some of the calculation used below are close to those developed for droplets and capsules in the small deformation theory[16, 21, 23]. The function f is expanded in powers of ϵ and decomposed on the basis of spherical harmonics. In principle

it can be decomposed on an infinite series of spherical harmonics as

$$f = \sum_{n=0}^{+\infty} f_n \tag{3}$$

with $f_n = \sum_{m=-n}^{+n} a_{nm}(t)\mathcal{Y}_{nm}(\theta,\phi)$, where $\mathcal{Y}_{nm}$ are the usual spherical harmonics, and a_{nm} are time-dependent amplitudes which are undetermined for the moment. We can alternatively write $f_n = F_{p_1 p_2 \ldots p_n}[\frac{\partial^n r^{-1}}{\partial x_{p_1}\partial x_{p_2}\ldots\partial x_{p_n}}]r^{n+1}|_{r=1}$ (where repeated indices are to be summed over). Most of our calculation is made with the second formulation. If only the leading order harmonics are retained, as adopted here, we can write up to order ϵ^2

$$f = f^{(0)} + \epsilon f^{(1)} = F^{(0)}_{ij} Y_{ij} + \epsilon \left(-\frac{6}{5} F^{(0)}_{ij} F^{(0)}_{ij} + F^{(1)}_{ij} Y_{ij} \right) \tag{4}$$

with the zeroth and first order amplitudes $F^{(0)}_{ij}$ and $F^{(1)}_{ij}$, respectively, and the abbreviation

$$Y_{ij} = \left(\frac{\partial^2 r^{-1}}{\partial x_i \partial x_j} \right)_{r=1}, \quad r = \left(x_1^2 + x_2^2 + x_3^2 \right)^{1/2}. \tag{5}$$

The $O(\epsilon)$ term $-6/5\, F^{(0)}_{ij} F^{(0)}_{ij}$ ensures constant volume.

As Y_{ij} is a second-order spherical harmonic, the tensors $F^{(0)}_{ij}$ and $F^{(1)}_{ij}$ have only five independent elements. We can thus demand that $F^{(0)}_{ij}$ and $F^{(1)}_{ij}$ are symmetric and traceless. In this case Eq. (4) can also be written as

$$f = \left[3F^{(0)}_{ij} x_i x_j + \epsilon \left(-\frac{6}{5} F^{(0)}_{ij} F^{(0)}_{ij} + 3F^{(1)}_{ij} x_i x_j \right) \right]_{r=1}. \tag{6}$$

In order to ensure that the excess area of the surface parameterized by (2) is ϵ^2, the five remaining coefficients $F^{(0)}_{ij}$ are not independent but coupled by the constraint

$$[F^{(0)}_{11}]^2 + [F^{(0)}_{22}]^2 + F^{(0)}_{11} F^{(0)}_{22} + [F^{(0)}_{12}]^2 + [F^{(0)}_{13}]^2 + [F^{(0)}_{23}]^2 \;=\; \frac{5}{96\pi}. \tag{7}$$

Eqs. (4,7) guarantee that the vesicle volume is $V = \frac{4}{3}\pi + O(\epsilon^3)$ and the vesicle surface area is $A = 4\pi + \Delta + O(\epsilon^3)$. We shall see later that including the $F^{(1)}_{ij}$ guarantees the constraint of excess area up to ϵ^4.

Our ansatz includes spherical harmonics of order 2 only. It is justified by the fact that the external flow just contains harmonics of order 2. At next-to-leading order modes of order 4 are excited, however, they do not couple back to the modes of second order. It is thus possible to derive a closed description that contains second-order spherical harmonics

only. It is only at higher orders (not included here) that higher order harmonics may affect dynamics.

Next we compute, up to $O(\epsilon^2)$, the two tangential vectors:

$$\mathbf{t}_\theta = \partial_\theta \mathbf{R}, \quad \mathbf{t}_\phi = \partial_\phi \mathbf{R} \tag{8}$$

and the unit normal vector

$$\mathbf{n} = \frac{\mathbf{t}_\theta \times \mathbf{t}_\phi}{|\mathbf{t}_\theta \times \mathbf{t}_\phi|}. \tag{9}$$

From the tangential vectors follows the metrics

$$g_{ij} = \mathbf{R}_i \cdot \mathbf{R}_j \tag{10}$$

with its inverse g^{ij}. Following Seifert [2], we evaluate the curvature tensor

$$h_{ij} = (\partial_i \partial_j \mathbf{R}) \cdot \mathbf{n}, \tag{11}$$

which yields the mean curvature

$$H = \frac{1}{2} g^{ik} h_{ki} \tag{12}$$

and the Gaussian curvature

$$K = \det(g^{ik} h_{kj}). \tag{13}$$

In terms of the amplitudes of the shape function we have, for example,

$$H \;=\; -1 - 2\epsilon\, F_{ij}^{(0)} Y_{ij} - \epsilon^2 \left[\frac{6}{5} F_{ij}^{(0)} F_{ij}^{(0)} + 2 F_{ij}^{(1)} Y_{ij} - 5 F_{ij}^{(0)} F_{lm}^{(0)}\, Y_{ij} Y_{lm} \right] + O(\epsilon^3). \tag{14}$$

B. Velocity field

In common experiments, the Reynolds number of the flow is much smaller than unity. The dynamics of the flow is thus adequately described by the Stokes equations for the velocity field $\mathbf{u}$ and the pressure p,

$$\eta_\alpha \nabla^2 \mathbf{v} = \nabla p, \tag{15}$$

where η_0 is the viscosity of the suspending fluid and η_1 is the viscosity of the fluid inside. We define as usually the ratio $\lambda \equiv \eta_1/\eta_0$. We can safely assume that the fluids are incompressible, and thus

$$\nabla \cdot \mathbf{v} = 0. \tag{16}$$

The total velocity field outside the vesicle can then be written as $\mathbf{v} = \mathbf{v}_0 + \mathbf{u}$, where $\mathbf{u}$ is the perturbation of the field due to the presence of the vesicle, and the imposed shear flow $\mathbf{v}_0$ is taken in the form $\mathbf{v}_0 = \dot{\gamma} x_2 \, \mathbf{e}_1$, where $\dot{\gamma}$ is the shear rate.

Likewise, we write for the velocity field within the vesicle: $\bar{\mathbf{v}} = \boldsymbol{\omega} \times \mathbf{r}/2 + \bar{\mathbf{u}}$, where ω is the vorticity. Following Lamb [17], we write an ansatz for the unknown perturbation of the velocity field outside the vesicle in the form

$$\begin{aligned} \mathbf{u} = \quad & \sum_{n=0}^{\infty} \nabla \chi_{-n-1} \times \mathbf{r} + \nabla \phi_{-n-1} - \frac{n-2}{2n(2n-1)} r^2 \nabla p_{-n-1} \\ & + \frac{n+1}{n(2n-1)} \mathbf{r} p_{-n-1}, \end{aligned} \tag{17}$$

and inside the vesicle:

$$\begin{aligned} \bar{\mathbf{u}} = \quad & \sum_{n=0}^{\infty} \nabla \bar{\chi}_n \times \mathbf{r} + \nabla \bar{\phi}_n + \frac{n+3}{2(n+1)(2n+3)} r^2 \nabla \bar{p}_n \\ & - \frac{n}{(n+1)(2n+3)} \mathbf{r} \bar{p}_n. \end{aligned} \tag{18}$$

The first term expresses vortex motion in a uniform pressure field. The second term represents an irrotational motion which can exist in a field of uniform pressure. The last two terms are connected with the pressure distribution.

The functions $\bar{p}_n$, $\bar{\phi}_n$, and $\bar{\chi}_n$ in the Lamb solution are solid spherical harmonics of order n and p_{-n-1}, ϕ_{-n-1}, χ_{-n-1} are solid spherical harmonics of order $-n-1$ [16]. Splitting off their r-dependence, we write $\chi_{-n-1} = r^{-n-1} Q_n$, $\phi_{-n-1} = r^{-n-1} S_n$, and $p_{-n-1} = r^{-n-1} T_n$. Likewise for the quantities within the vesicle: $\bar{\chi}_n = r^n \bar{Q}_n$, $\bar{\phi}_n = r^n \bar{S}_n$, and $\bar{p}_n = r^n \bar{T}_n$. The precise values of the functions Q_n, S_n,... (which are surface spherical harmonics and thus depend only on the angles) are determined from the boundary conditions at the membrane, as will be seen later.

Since a shear flow induces a shape deformation from a sphere which involves only second order harmonics (i.e. $n = 2$), only $\mathcal{Y}_{2m}$ is active[2, 4]. The other modes are damped[7, 16]

to leading order. So we can write the Lamb solution for the full velocity field $\mathbf{v}$ as

$$\bar{\mathbf{v}} = \nabla\bar{\phi} + \frac{5}{42}r^2\nabla\bar{p} - \frac{2}{21}r\,\bar{p} + \dot{\gamma}/2\,(x_2\mathbf{e}_1 - x_1\mathbf{e}_2), \tag{19}$$

$$\mathbf{v} = \nabla\phi + \frac{1}{2}r\,p + \dot{\gamma}\,x_2\mathbf{e}_x. \tag{20}$$

where we explicitly write the external shear flow for the outer field and its rotational component for the inner field. Since there is only the second harmonic, we have dropped the subscripts (like in the terms p_{-n-1}, p_n, which would produce p_{-3} and p_2) in order to simplify the notations. The ansatz functions are expanded in powers of ϵ:

$$\bar{\phi} = \left[\bar{S}_{ij}^{(0)}Y_{ij} + \epsilon\,\bar{S}_{ij}^{(1)}Y_{ij}\right] r^2, \tag{21}$$

$$\phi = \left[S_{ij}^{(0)}Y_{ij} + \epsilon\,S_{ij}^{(1)}Y_{ij}\right] r^{-3} \tag{22}$$

and

$$\bar{p} = \left[\bar{T}_{ij}^{(0)}Y_{ij} + \epsilon\,\bar{T}_{ij}^{(1)}Y_{ij}\right] r^2 + \bar{p}_0, \tag{23}$$

$$p = \left[T_{ij}^{(0)}Y_{ij} + \epsilon\,T_{ij}^{(1)}Y_{ij}\right] r^{-3}. \tag{24}$$

C. Stress balance

We now formulate the stress balance at the membrane. To this end we have to evaluate the stresses exerted by the membrane as well as the hydrodynamical stresses that result from the fluids on both sides of the membrane.

The normal force exerted by the membrane is given by the Helfrich force[2]:

$$F_n = \kappa\left[2H(2H^2 - 2K) + 2\Delta_S H\right] - 2\,ZH. \tag{25}$$

H and K are the mean and the Gaussian curvature, respectively, and Δ_S is the Laplace-Beltrami operator. Here Z is a Lagrange multiplier which enforces local membrane area conservation. At zeroth order, the bracketed term in Eq. (25) vanishes since $H^2 = K^2 = 1$ and $\Delta_S H = 0$. It follows that, at leading order, the bending rigidity of the membrane is not involved into play. This is just a consequence of the formal expansion we have adopted. In order to arrive at a non-trivial solution at zeroth order, we formally require κ (and $\partial_t f$) to scale like ϵ^{-1}. In order to make all ϵ dependencies explicit, we introduce $\bar{\kappa} = \epsilon\kappa$ and write

$$F_n = \epsilon^{-1}\bar{\kappa}\left[2H(2H^2 - 2K) + 2\Delta_S H\right] - 2\,ZH. \tag{26}$$

By the same token, the isotropic part of Z must scale like ϵ^{-1}. The angular part, however, turns out to be $O(1)$. Hence we decompose the Lagrange multiplier Z in the following way:

$$Z = \epsilon^{-1}\left(Z_0^{(0)} + \epsilon\, Z_0^{(1)}\right) + Z_{ij}^{(0)} Y_{ij} + \epsilon\, Z_{ij}^{(1)} Y_{ij}. \tag{27}$$

The tangential force exerted by the membrane (due to its incompressibility) is

$$\mathbf{F}_t = \left(g^{11}\mathbf{t}_\theta + g^{21}\mathbf{t}_\phi\right)\partial_\theta Z + \left(g^{12}\mathbf{t}_\theta + g^{22}\mathbf{t}_\phi\right)\partial_\phi Z. \tag{28}$$

The fluid stresses are given by the hydrodynamical stress tensor:

$$\sigma_{ij} = -p\,\delta_{ij} + \eta_0\,(\partial_i v_j + \partial_j v_i), \tag{29}$$

$$\bar{\sigma}_{ij} = -\bar{p}\,\delta_{ij} + \eta_1\,(\partial_i \bar{v}_j + \partial_j \bar{v}_i). \tag{30}$$

The full stress balance at the membrane thus reads

$$\left[(\sigma_{ij} - \bar{\sigma}_{ij})n_j + F_n n_i\right]\mathbf{e}_i + \mathbf{F}_t = 0, \tag{31}$$

which has to be evaluated at $r = 1 + \epsilon f$.

D. Membrane incompressibility and kinematic condition

Membrane local incompressibility entails that the projected divergence of the velocity field must vanish on the membrane:

$$(\delta_{ij} - n_i n_j)\,\partial_i v_j = 0. \tag{32}$$

We have in addition to require continuity of the fluid velocities across the membrane,

$$v_i = \bar{v}_i \tag{33}$$

as well as equality with the velocity of the membrane (if we do not account for any permeation across the membrane). The latter condition reads

$$\epsilon\,\partial_t f = n_i v_i, \tag{34}$$

if we neglect $O(\epsilon^2)$ terms [21].

III. SOLUTION AT ZEROTH ORDER

In this section we shall deal with the leading order solution as originally derived by one of us[4]. By leading order we mean to keep only the terms having (0) in their superscript (e.g. $F_{ij}^{(0)}$). We shall present in the next section the solution to the next order. Following Frankel et al.[21], we determine the ansatz coefficients from the boundary conditions by performing surface integrals over the (spherical) vesicle. For example, the equation

$$\int \left(v_i^{(0)} - \bar{v}_i^{(0)}\right) x_j \, \mathrm{d}\Omega = 0, \tag{35}$$

which is an integral version of the continuity condition at zeroth order (actually a projection of the ith vector component on the subspace of first-order spherical harmonics), yields the five relations

$$\bar{T}_{ij}^{(0)} - T_{ij}^{(0)} + 10\bar{S}_{ij}^{(0)} = \frac{5}{3} e_{ij}. \tag{36}$$

Similarly, the integral

$$\int \left(v_p^{(0)} - \bar{v}_p^{(0)}\right) x_p x_i x_j \, \mathrm{d}\Omega \;=\; 0 \tag{37}$$

(which is just the projection of the radial velocity balance on the subspace of second-order spherical harmonics, but can also be understood as a projection of the ith component of the radial velocity on the subspace of first-order spherical harmonics) gives the equations

$$\frac{2}{7}\bar{T}_{ij}^{(0)} - T_{ij}^{(0)} + 4\bar{S}_{ij}^{(0)} + 6S_{ij}^{(0)} = \frac{2}{3} e_{ij}. \tag{38}$$

From the stress balance [Eq. (31)] we have, upon projection on the appropriate subspace,

$$\int \left[\left(\sigma_{iq}^{(0)} - \bar{\sigma}_{iq}^{(0)}\right) n_q^{(0)} + \left(F_n^{(0)} - 2Z_0^{(0)}\right) n_i^{(0)} + \mathbf{F}_t^{(0)} \cdot \mathbf{e}_i\right] x_j \, \mathrm{d}\Omega \;=\; 0, \tag{39}$$

which gives rise to

$$\lambda\bar{T}_{ij}^{(0)} + \frac{3}{2} T_{ij}^{(0)} + 10\lambda\bar{S}_{ij}^{(0)} = \frac{5}{3} e_{ij} + Z_{ij}^{(0)} - 4\left(Z_0^{(0)} + 6\bar{\kappa}\right) F_{ij}^{(0)} \tag{40}$$

and at the same time fixes

$$p_0 = \frac{2}{\epsilon}\left(Z_0^{(0)} + \epsilon Z_0^{(1)}\right). \tag{41}$$

Finally, evaluating the integral

$$\int \left[\left(\sigma_{pq}^{(0)} - \bar{\sigma}_{pq}^{(0)}\right) n_q^{(0)} + \left(F_n^{(0)} - 2Z_0^{(0)}\right) n_p^{(0)} + \mathbf{F}_t^{(0)} \cdot \mathbf{e}_p\right] x_p x_i x_j \, \mathrm{d}\Omega \;=\; 0, \tag{42}$$

we find

$$3T_{ij}^{(0)} - \frac{1}{7}\lambda\bar{T}_{ij}^{(0)} + 4\lambda\bar{S}_{ij}^{(0)} - 24S_{ij}^{(0)} = \frac{2}{3}e_{ij} - 2Z_{ij}^{(0)} - 4\left(Z_0^{(0)} + 6\bar{\kappa}\right)F_{ij}^{(0)}. \tag{43}$$

Eqs. (36,38,40,43) determine the values of $T_{ij}^{(0)}$, $\bar{T}_{ij}^{(0)}$, $S_{ij}^{(0)}$, and $\bar{S}_{ij}^{(0)}$ as a function of e_{ij}, $Z_0^{(0)}$, $Z_{ij}^{(0)}$, and $F_{ij}^{(0)}$.

Likewise, the angular components $Z_{ij}^{(0)}$ of the membrane tension are found from the surface integral over the projected divergence (Eq.(32))

$$\int \left(\delta_{pq} - n_p^{(0)}n_q^{(0)}\right)\,\partial_p v_q^{(0)}\,x_i x_j\,\mathrm{d}\Omega = 0, \tag{44}$$

which yields

$$\frac{3}{7}\bar{T}_{ij}^{(0)} + 2\bar{S}_{ij}^{(0)} = 0. \tag{45}$$

Having thus obtained the velocity field as a function of the shape amplitudes $F_{ij}^{(0)}$ (and e_{ij}, $Z_0^{(0)}$), we employ the same strategy to fulfil the kinematic condition (34):

$$\int \left(\epsilon\,\partial_t f^{(0)} - n_p^{(0)}v_p^{(0)}\right)x_i x_j\,\mathrm{d}\Omega = 0. \tag{46}$$

The result is a set of five equations that describe the dynamics of the amplitudes:

$$\epsilon\,\partial_t F_{ij}^{(0)} = \frac{20\,e_{ij}}{23\lambda + 32} - \frac{24\left(Z_0^{(0)} + 6\bar{\kappa}\right)}{23\lambda + 32}\,F_{ij}^{(0)}. \tag{47}$$

For the time being we leave the isotropic part $Z_0^{(0)}$ of the membrane tension undetermined, and it will be dealt with later in this paper.

IV. SOLUTION AT FIRST ORDER

The solution at first order is obtained in the same way as the solution at zeroth order, this is why we shall not dwell on this issue. However, we must now apply the boundary conditions at $r = R$ instead of $r = 1$ (in order to incorporate consistently the desired order in ϵ). To this end, we employ a Taylor series expansion around $r = 1$. For example, the velocity field becomes

$$v_i|_{r=1+\epsilon f} = \left[v_i^{(0)} + \epsilon f^{(0)}x_j\frac{\partial v_i^{(0)}}{\partial x_j} + \epsilon v_i^{(1)}\right]_{r=1} + O(\epsilon^2). \tag{48}$$

For the continuity condition we have to evaluate the integrals

$$\int \left(v_i^{(1)} - \bar{v}_i^{(1)} + f^{(0)} x_q \frac{\partial (v_i^{(0)} - \bar{v}_i^{(0)})}{\partial x_q} \right) x_i \, \mathrm{d}\Omega \;=\; 0, \tag{49}$$

$$\int \left(v_p^{(1)} - \bar{v}_p^{(1)} + f^{(0)} x_q \frac{\partial (v_p^{(0)} - \bar{v}_p^{(0)})}{\partial x_q} \right) x_p x_i x_j \, \mathrm{d}\Omega \;=\; 0. \tag{50}$$

Similar integrals are constructed for the first-order stress balance. The resulting equations allow for the determination of $T_{ij}^{(1)}$, $\bar{T}_{ij}^{(1)}$, $S_{ij}^{(1)}$, and $\bar{S}_{ij}^{(1)}$.

Membrane incompressibility (32) provides us with the expression of $Z_{ij}^{(1)}$, and from the kinematic condition (34) we find at first order the following evolution equations for the amplitudes:

$$\begin{aligned}
\epsilon \, \partial_t F_{ij}^{(1)} = & -\frac{\omega_s}{2} \left(\epsilon_{psi} F_{pj}^{(0)} + \epsilon_{psj} F_{pi}^{(0)} \right) - \frac{24}{23\lambda + 32} \left[Z_0^{(1)} F_{ij}^{(0)} + \left(Z_0^{(0)} + 6\bar{\kappa} \right) F_{ij}^{(1)} \right] \\
& + \frac{4800}{7} \frac{\lambda - 2}{(23\lambda + 32)^2} \, \mathcal{S}d \left[F_{ip}^{(0)} e_{pj} \right] \\
& + \frac{288}{7} \frac{1}{(23\lambda + 32)^2} \, \mathcal{S}d \left[F_{ip}^{(0)} F_{pj}^{(0)} \right] \left[(138\lambda + 192) \, \bar{\kappa} + (49\lambda + 136) \left(Z_0^{(0)} + 6\bar{\kappa} \right) \right],
\end{aligned} \tag{51}$$

where we have introduced the notation $\mathcal{S}d[b_{ij}] = \frac{1}{2}[b_{ij} + b_{ji} - \frac{2}{3}\delta_{ij} b_{ll}]$.

Note that this equation still contains the undetermined functions $Z_0^{(0)}(t)$ and $Z_0^{(1)}(t)$, which must be chosen such that the dynamics of the amplitudes comply with the available excess area relative to the sphere.

V. COMBINING ZEROTH AND FIRST ORDER SOLUTIONS

We now proceed by casting the solutions at zeroth order, Eq. (47), and at first order, Eq. (51), into a single equation. To this end, we set $F_{ij} = F_{ij}^{(0)} + \epsilon F_{ij}^{(1)}$ and $Z_0 = Z_0^{(0)} + \epsilon Z_0^{(1)}$. Thus we obtain a single evolution equation for the amplitudes F_{ij}:

$$\begin{aligned}
\epsilon \, \frac{\mathcal{D} F_{ij}}{\mathcal{D} \bar{t}} \;=\; & \frac{20 \, \bar{e}_{ij}}{23\lambda + 32} - \frac{24 \, (Z_0 + 6\epsilon C_a^{-1})}{23\lambda + 32} \, F_{ij} \\
& + \epsilon \left[\frac{4800}{7} \frac{\lambda - 2}{(23\lambda + 32)^2} \, \mathcal{S}d[F_{ip} \bar{e}_{pj}] + \frac{288}{7} \frac{(49\lambda + 136) \, Z_0 + (432\lambda + 1008) \, \epsilon C_a^{-1}}{(23\lambda + 32)^2} \, \mathcal{S}d[F_{ip} F_{pj}] \right].
\end{aligned} \tag{52}$$

The quantity

$$\frac{\mathcal{D} F_{ij}}{\mathcal{D} \bar{t}} \equiv \partial_{\bar{t}} F_{ij} + \frac{1}{2} \left(\epsilon_{psi} F_{pj} + \epsilon_{psj} F_{pi} \right), \tag{53}$$

entering this equation, is the Jaumann derivative (note that we consider all the calculation in the advection frame of the vesicle, so that we have partial derivative instead of material derivative as is usually written in the Jaumann derivative). ϵ_{psj} is the Levi-Civita tensor. In Eq.(52) time is dimensionalized by $\dot{\gamma}^{-1}$, and $\bar{e}_{ij}$ by $\dot{\gamma}$. The capillary number C_a is defined as

$$C_a = \frac{\eta_0 \dot{\gamma} r_0^3}{\kappa} \equiv \tau \dot{\gamma} \tag{54}$$

where τ is a typical time scale for the relaxation of the vesicle towards its equilibrium when the flow is set to zero.

We shall now determine the Lagrange multiplier Z_0 by imposing that the shape functions F_{ij} must comply with the available excess area. In terms of F_{ij}, the surface of the vesicle is given by

$$A = 4\pi + \Delta + O(\epsilon^4) = 4\pi + \epsilon^2 \frac{96\pi}{5} \left(F_{11}^2 + F_{22}^2 + F_{11}F_{22} + F_{12}^2 + F_{13}^2 + F_{23}^2 \right) + O(\epsilon^4). \tag{55}$$

Note that after recasting the zeroth- and first-order amplitudes into one amplitude, the surface area is observed up to $O(\epsilon^4)$. This is a very important point, since problems with constraints always trigger higher order nonlinearities than those initially present in the physical problem[26].

Evaluating $\partial_t A = 0$ and substituting Eq. (52) for $\partial_t F_{ij}$, we find the expression for Z_0, which reads:

$$Z_0 = \frac{(8\pi\, F_{12} - 6C_a^{-1}\epsilon) + \epsilon\, A_0}{1 + \epsilon\, B_0} \tag{56}$$

with the abbreviations

$$\begin{aligned} A_0 &= \frac{8\pi \left[1200\,(\lambda - 2)\; C_0 - 31104\,(3\lambda + 7)\,\epsilon C_a^{-1}\, D_0 \right]}{35\,(23\lambda + 32)}, \\ B_0 &= \frac{1728\pi\,(49\lambda + 136)\, D_0}{35\,(23\lambda + 32)}, \end{aligned}$$

which contain the following combinations of the amplitudes:

$$\begin{aligned} C_0 &= F_{11}F_{12} + F_{22}F_{12} + F_{13}F_{23}, \\ D_0 &= F_{11}F_{22}^2 - F_{11}F_{12}^2 + F_{22}F_{13}^2 + F_{11}F_{23}^2 + F_{11}^2F_{22} - 2F_{23}F_{12}F_{13} - F_{12}^2F_{22}. \end{aligned}$$

VI. GENERAL COMMENTS AND COMPARISON WITH OTHER WORKS

Let us make some general comments. If we set formally $\epsilon = 0$ on the right hand side of Eq.(52) and in Eq.(56), we obtain the following equation

$$\epsilon \frac{\mathcal{D}F_{ij}}{\mathcal{D}\bar{t}} = \frac{20\bar{e}_{ij}}{23\lambda + 32} - \frac{192\pi}{23\lambda + 32}\frac{F_{12}F_{ij}}{\Delta}. \tag{57}$$

This is the evolution equation derived in [4] (where time is rescaled by $\dot{\gamma}^{-1}$), which we call "leading order theory". Note that even to this leading order the evolution equation is nonlinear. This nonlinearity is triggered by local membrane incompressibility. Note that this markedly differs from droplet[16, 21] and capsule[23] theories where the leading order equations are linear. The nonlinearities induce bifurcations and lead to the three dynamical modes: *tt*, *tb* and *vb*.

In the leading order theory the membrane rigidity (or C_a) scales out from the evolution equation. Following Ref.[4], two groups [8, 9] attempted very recently to include higher order contributions beyond Eq.(57). The calculations presented by Lebedev et al.[8] and Nogushi and Gompper[8, 9] do not seem to conform to our theory. Lebedev et al.[8] add in the Helfrich force the next order term, but they ignored the corresponding hydrodynamical response. As shown below the ignored terms are higher than those retained. This does not only induce quantitative variations, but also it is shown below that the so-called self-similarity[8] (in that the equations contain only two independent parameters) does not hold.

Nogushi and Gompper[9] retain the full Helfrich force (without truncation), but as Lebedev et al.[8] they did not take into account the corresponding hydrodynamic response. In addition, the authors combine, without justification, various ingredients: (i) leading order theory[4] for the amplitude of vesicle deformation in order to compute the hydrodynamical response, (ii) the full Helfrich force (without including the corresponding velocity field), (iii) the semi-phenomenological Keller-Skalak[15] theory for the orientation angle of the vesicle; this last point will become more clear in the next section. At present we do not understand why the authors consider worthwhile to use a leading order theory for the amplitude of deformation, but a semi-phenomenological theory for the orientation angle.

It should be stressed that a consistent theory [Eq. (52)], as presented here, induces higher and higher nonlinearities due the constraint of a given available excess area. In section VII we shall present the main results which follow from the full equation Eq.(52).

A. Post-expansion of the full evolution equation

In order to compare more precisely with previous analyses, it is convenient to expand the evolution equation Eq.(52) in powers of F_{ij}. For that purpose, we admit that F_{ij} is small enough, albeit it is formally of order unity. This is a priori justified by the fact that due to the available excess area constraint (7) the sum of the amplitudes is $5/96\pi \ll 1$. The idea is to express F_{ij} in terms of the orientation angle and the amplitude of deformation. For that purpose we make use of the following identity

$$\sum_{i,k=1,2,3} 3x_i x_k F_{ik}(t) = \sum_{m=-2}^{2} a_{2m}(t)\mathcal{Y}_{2m}(\theta,\phi) \tag{58}$$

(note that a_{2m} was called F_{2m} in [4]).

Then using as in[4] $a_{22} = \mathcal{R}e^{-2i\psi}$, ψ coincides with the orientation angle of the vesicles (Fig.2), and $\mathcal{R}$ is the amplitude of deformation of the vesicle. Instead of using $\mathcal{R}$, and for the sake of comparison with[8] we use the variable Θ defined by $\mathcal{R}/2\epsilon = \cos(\Theta)$. We expand the full equation [Eq.(52)] in powers of F_{ij} and retain terms up to the higher (fifth) order in F_{ij} in a consistent manner. We then perform a straightforward conversion of variables in terms of ψ and Θ. We find for Θ and ψ the following equations (where now we use physical time instead of $\bar{t}$):

$$\begin{aligned} T\partial_t\Theta &= -S\sin\Theta\sin 2\psi + \cos 3\Theta + \epsilon\,\Lambda_1\, S\sin 2\psi\,(\cos 4\Theta + \cos 2\Theta) && (59)\\ &\quad + \epsilon\,\Lambda_2\, S\sin 2\psi\cos 2\Theta + ... && (60)\\ T\partial_t\psi &= \frac{S}{2}\left\{\frac{\cos 2\psi}{\cos\Theta}\left[1+\epsilon\Lambda_2\sin\Theta\right] - \Lambda\right\} + ..., && (61) \end{aligned}$$

where we define,

$$\begin{aligned} S &= \frac{7C_a}{9}\frac{\sqrt{3}\pi}{\epsilon^2}, && (62)\\ T &= \frac{7C_a}{720}\sqrt{10\pi}\,\frac{23\lambda+32}{\dot{\gamma}}, && (63)\\ \Lambda &= \frac{1}{240}\sqrt{\frac{30}{\pi}}\,(23\lambda+32)\epsilon, && (64)\\ \Lambda_1 &= \frac{1}{28}\sqrt{\frac{10}{\pi}}\,\frac{49\lambda+136}{23\lambda+32}, && (65)\\ \Lambda_2 &= \frac{10}{7}\sqrt{\frac{10}{\pi}}\,\frac{\lambda-2}{23\lambda+32}, && (66) \end{aligned}$$

and '...' stands for other terms of the series. The first term on the right hand side of Eq. (59) corresponds to the leading order theory presented in [4]. The first and second term correspond to the situation treated in [8] where only the higher order contribution in the membrane bending force is included[8, 9]. Taking the corresponding hydrodynamical response to the same order into account (as done here) induces significant changes. A new term, for example, is the third term on the right hand side of Eq. (59) (proportional to Λ_1). This term is at least of the same order as $\cos 3\Theta$. Indeed, the term proportional to $\Lambda_1 S$ is of the order of $\Lambda_1 C_a/\epsilon$. Even if we consider ϵ not too small (the situation is worse otherwise!), we have a term proportional to C_a. If one has in mind a physical situation, then it is known that most experimental observations operate at C_a significantly larger than one[5, 6], and that therefore the neglected terms are higher than those retained. If one has in mind a formal spirit (or a mathematical spirit, in that C_a is taken of order unity), then ϵ should be regarded as small. In that case the neglected terms are of order $1/\epsilon$, and are much higher than the retained term in the Helfrich energy, namely $\cos 3\Theta$ (which is of order one). As a natural consequence of this, the so-called similarity equations (put forward in[8], in that the evolution equations contain only 2 independent parameters, S and Λ; while T can be absorbed in a redefinition of time) does not hold. Indeed, we have three parameters, which are C_a, λ and Δ, the excess area (or equivalently ϵ); we could of course use S, Λ and Δ as well, but the community is used rather to the quantities C_a, λ and Δ, and there is no need to contribute to proliferation of symbols.

VII. RESULTS

Equation (52) constitutes our basic result that we shall analyze now. We first analyze the *tt* regime. Figure 3 presents the orientation angle as a function of λ and compares the results with previous studies. Instead of a square root singularity found for the leading order theory (and in the Keller-Skalak regime[15]), the angle crosses zero quasi-linearly. A point which is worth of mention is that the *tt* angle becomes negative before the solution ceases to exist (signature of the *tb* regime). Before the solution ceases to exist the *vb* mode takes place, as discussed below.

In [4] it was predicted that in the tumbling regime a *vb* mode should take place. This was found to occur as an oscillator (like in a conservative system), since the frequency of

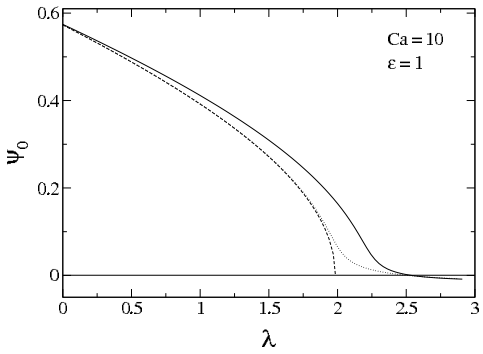

FIG. 3: The angle ψ_0 in the tt regime. Dashed line: the leading order theory[4], dotted line: the theory of Ref.[8], the full line: the present theory.

oscillation about the fixed point $\psi = 0$ was found to be purely imaginary. By including higher order terms the frequency acquires a non zero real part[8], and the vb mode becomes a limit cycle [in that all initial conditions in its domain of existence tend towards a closed trajectory in phase space, (ψ, Θ)]. As expected from the original theory[4] the vb mode still occurs in the vicinity of the tumbling threshold. This happens provided that the shape dynamics evolve with time (breathing of the shape). C_a is a direct measure for the comparison between the shape evolution time scale and the shearing time. The original theory[4] corresponds formally to $C_a \to \infty$, as can be seen from Eq.(59) and the definition of T and S. Including higher order terms leads to the appearance of C_a in the equation.

In Fig. 4 we report on the phase diagram and compare it to previous theories[4, 8]. For small C_a we find a direct (saddle-node) bifurcation from tt to tb, in agreement with Ref.[3]. At $C_a \to \infty$ we recover the results of Ref.[4] (in that the vb mode coexists with tb and whether one prevails over the other depends on initial conditions; this is not shown on the figure 2). At intermediate values of C_a we find a belt (or a domain) of vb mode preceding the tb bifurcation, in qualitative agreement with [8, 9], as shown on Fig. 4. The bifurcation from the tt to vb is of Hopf type (the imaginary part of the stability eigenvalue of the tt mode is imaginary). At larger C_a, the vb mode ceases to exist, while the tb mode takes place. The tb mode occurs, at small C_a, as a saddle node bifurcation from tt. It merges at larger C_a as a consequence of cessation of existence of the vb mode. Actually this coincides with the cessation of existence of the tt branch (which becomes unstable– instability leading to vb–, but it continues to exist until it hits the tb boundary).

The higher order calculation provided here shows significant differences with[8, 9], as

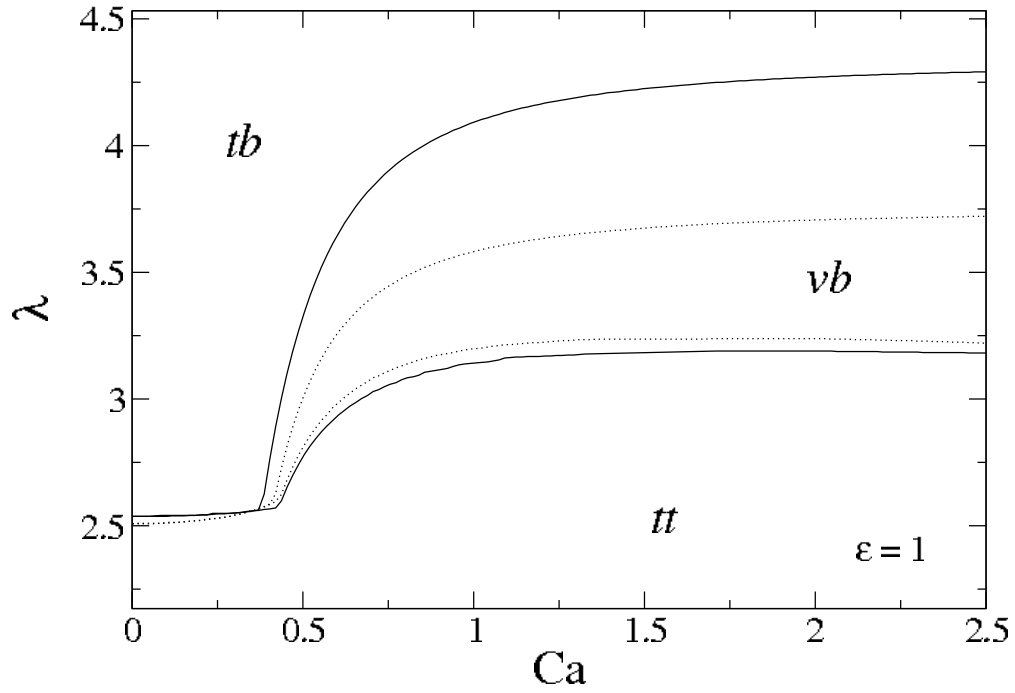

FIG. 4: The dotted line: theory in Ref.[8], the full line: the present theory. The same order of discrepancy is found with Ref.[9].

shown in Fig. 4. The results presented in [8, 9] may be viewed as semi-qualitative given the disregard of other terms of the same order, as commented in the previous section. Actually, a simplistic phenomenological model [11] captures the main essential qualitative features of Fig. 4.

Figure 5 shows a snapshot of the *vb* mode. Note that a pure swinging (a terminology usually used for oscillation of rigid objects, and adopted in [9]) would be impossible within the Stokes limit, since this is forbidden by the symmetry of the Stokes equation upon time reversal. The breathing is a necessary condition for the present mode. In the upper half plane (i.e. when $\psi > 0$) the shape in the *vb* regime (dashed line in the figure) is different from the one in the lower plane (dotted line, rounded shape). This asymmetry makes this dynamics possible owing to the fact that the two shapes (i.e. for $\psi > 0$ and $\psi < 0$) can not be deduced from each other by a simple mirror symmetry with respect to the horizontal axis.

The basic understanding of the *vb* mode is as follows. First we recall that a shear flow is a sum of a straining part along $\pm\pi/4$ (which elongates the vesicle for $\psi > 0$ and compresses it for $\psi < 0$) and a rotational part, tending to make a clockwise *tb*. Due to the membrane

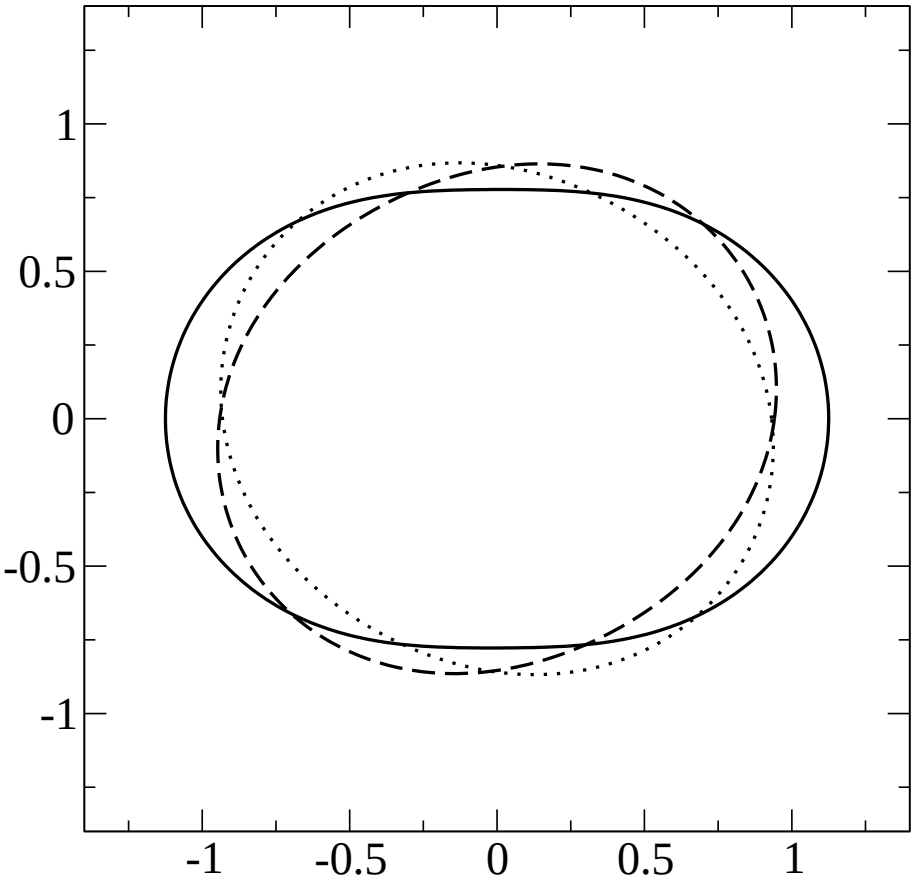

FIG. 5: A snapshot of the vb mode. Solid line: $\psi = 0$. Dotted line: $\psi \simeq -0.5$. Dashed line $\psi \simeq 0.5$

fluidity the torque associated with the shear is partially transferred to tt of the membrane, so that (due to torque balance) the equilibrium angle for tt is $0 < \psi_0 < \pi/4$. Further, an elongated vesicle tumbles more easily than a compressed one[3]. Suppose we are in the tt regime ($\psi_0 > 0$), but in the vicinity of tb, so $\psi_0 \simeq 0$. For small C_a the vesicle's response is fast as compared to shear, so that its shape is adiabatically slaved to shear (a quasi shape-preserving dynamics): a direct bifurcation from tt to tb occurs[3]. When $C_a \simeq 1$, the shape does not anymore follow adiabatically the shear. When tumbling starts to occur ψ becomes slightly negative. There the flow compresses the vesicle. Due to this, the applied torque is less efficient. The vesicle feels, so to speak, that its actual elongation corresponds to the tt regime and not to tb. The vesicle returns back to its tt position, where $\psi > 0$, and it feels now an elongation (which manifests itself on a time scale of the order of $1/\dot{\gamma}$). Due to elongation in this position, tumbling becomes again favorable, and the vesicle returns to $\psi < 0$, and so on. We may say that the vesicle *hesitates* or *vacillates* between tb and tt. The compromise is the vb mode.

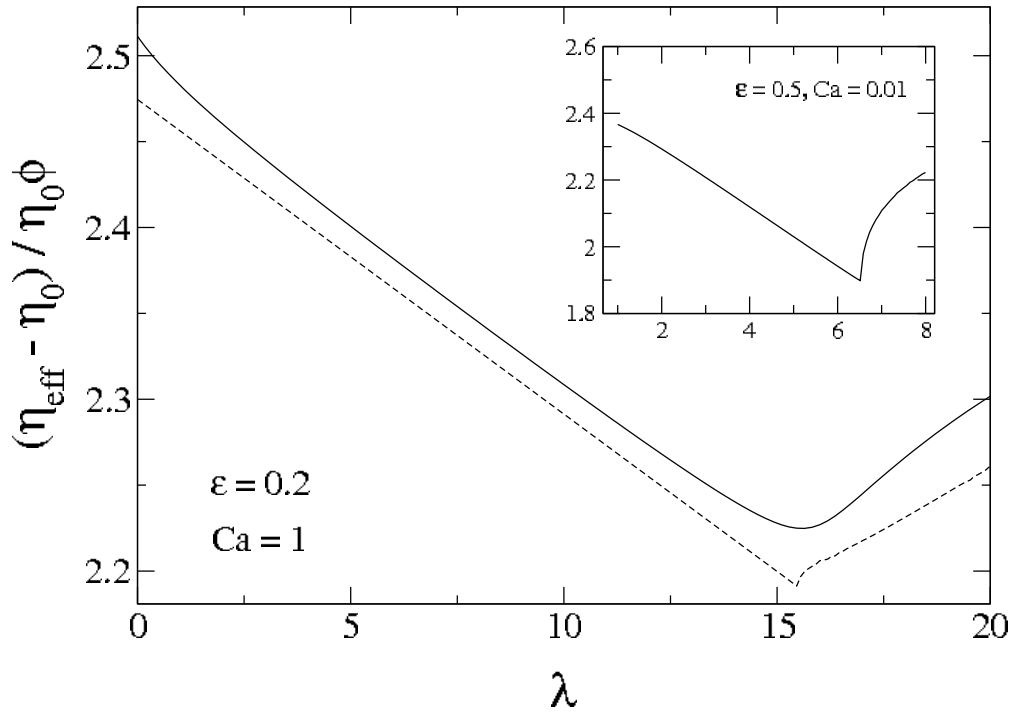

FIG. 6: The dashed line in the outer graph corresponds to the leading order theory where a cusp singularity is observed for any C_a. ϕ is the label of the vertical axis is the suspension volume fraction.

VIII. A BRIEF DISCUSSION OF RHEOLOGY

We discuss briefly the implication of the higher order theory on rheology. A complete discussion on this topic, and the comparison between droplets and capsules theory will be presented in the future.

Recently a link between the different modes and rheology has been presented[10]. It is thus natural to ask how higher order terms would modify the reported picture. Eq. (52) constitutes a basis for the derivation of the constitutive law, as in [10]. Here we focus only on the effective viscosity as a function of λ. In the *tb* and *vb* regimes we make an average of the effective viscosity over a period of oscillation. The results are reported on Fig.6.

We see that at small enough C_a, the cusp singularity (inset of Fig.6)[10] persists as in the leading order theory, while at larger C_a the cusp is smeared out by the fact that the transition towards the *vb* mode does not show a singularity as does a saddle-node bifurcation.

IX. DISCUSSION AND CONCLUSION

We have extended in a consistent manner the original theory[4] to higher order, and have analyzed the far reaching consequences. We have found that three parameters (i.e. the full set of dimensionless parameters that we can construct from the original model) survive to the next consistent leading order, thus ruling out some suspicions on self-similarity solutions, as announced in[8]. We have then analyzed the phase diagram of the evolution equation, and briefly discussed the behavior of the effective viscosity in the dilute regime. A particular point is that the next order terms wash out the cusp singularity of the effective viscosity[10] at the bifurcation point, provided that C_a is large enough. At low enough C_a the cusp singularity persists.

We have checked that for high enough $C_a \simeq 100$ (a quite accessible value in the experiments[6]) the full evolution equation produces a pseudo-coexistence of the vb and tb solution. By pseudo we mean the following: if we start with an adequate initial condition (say ψ small), but the physical parameters are such that the tb mode should be expected from the phase diagram (Fig.4), then the system can spend a long time in the vb regime (say about 50 cycles, or more; typically the number of cycles is of order $1/C_a$), before it falls onto the attracting mode, namely the tb one. If, on the contrary, the vb mode is expected from the phase diagram, but the initial condition is such that ψ is large enough to enforce tb, then the system spends a long period of time in the tb regime before it exhibits vb. Since, to date, no systematic experimental study of the vb mode has been reported (the only reports show about two temporal periods[5, 6]), our analysis shows that it may prove very difficult to locate experimentally the boundaries of the various modes.

Finally, this model has focused on vesicles. In order to make a step forward towards red blood cells, it is necessary to include the cytoskeleton structure, which is known to exhibit nonlinear viscoelasticity. We hope to tackle this question in the future.

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[26] Suppose that we have a quantity, say a vector $\mathbf{m}$, whose norm must be constant in the course of time–like a director in liquid crystals–. In the harmonic approximation the vector $\mathbf{m}$ obeys a linear equation, say $\partial_t \mathbf{m} = \mathbf{L}(\mathbf{m})$ where L is a certain linear operator. This equation does not conserve the norm of $\mathbf{m}$. In order to fulfill the constraint, we must add a term $\mu\mathbf{m}$ and write $\partial_t \mathbf{m} = \mathbf{L}(\mathbf{m}) + \mu\mathbf{m}$, where μ is a Lagrange multiplier enforcing constant norm of $\mathbf{m}$. Taking the scalar product with $\mathbf{m}$ and setting $\partial_t \mathbf{m^2} = \mathbf{0}$, determines μ and the final evolution equation becomes $\partial_t \mathbf{m} = \mathbf{L}(\mathbf{m}) - \mathbf{m}\mathbf{L}(\mathbf{m})\mathbf{m}$. So the constrained equation has acquired a third nonlinearity, or in terms of energy, it has a quartic term, while the original non constrained energy is harmonic. This problem is somewhat akin to ours where the evolution equation (51) acquires highly nonlinear terms hidden in Z_0.