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

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Vesicles under shear flow exhibit various dynamics: tank-treading (tt), tumbling (tb) and vacillating-breathing (vb). A consistent higher order theory reveals a direct bifurcation from tt to tb if $C_a \equiv \tau \gamma$ is small enough ($\tau=\text{vesicle relaxation time}$, $\gamma=\text{shear rate}$). At larger $C_a$ the tb is preceded by the vb mode. For $C_a \gg 1$ we recover the leading order calculation, where the vb mode coexists with tb. The consistent calculation reveals several quantitative discrepancies with recent works, and points to new features. We analyse rheology and find that the effective viscosity exhibits a minimum at $tt - tb$ and $tt - vb$ bifurcation points.

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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 $\lambda = \eta_m/\eta_a$ ($\eta_m$ and $\eta_a$ are the internal and external viscosities, respectively), $\psi$ decreases until it vanishes at a critical value of $\lambda = \lambda_c$. For a small enough $C_a \equiv \tau \gamma$ ($\tau=\text{relaxation time}$ towards the equilibrium shape in the absence of an imposed flow, $\gamma=\text{shear rate}$) the tt exhibits a saddle-node bifurcation towards tumbling (tb)$^2$.

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. Shortly after this theoretical prediction, an experimental report on this type of mode has been presented$^5$ (trembling denomination was used there) 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$^7,8$ 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 $C_a$ ($C_a$ scales out in the leading theory$^4$).

The first aim of this communication is to present the result of the consistent theory regarding the higher order calculation. We find significant differences with a recent work$^9$ regarding the form of the evolution equation. This implies, in particular, that the location of the boundaries separating the various three regimes in parameters space is significantly affected.

A second important report is to investigate how the effective viscosity derived recently in$^10$ is affected by the higher order deformation. For a small enough $C_a$ the effective viscosity of the suspension (as a function of $\lambda$) still exhibits a cusp singularity at the $tt - tb$ bifurcation$^10$, while the cusp becomes a smooth minimum when $C_a$ is high enough, namely when the $tt - vb$ bifurcation occurs.

The vesicle suspension is submitted to a linear shear flow $V_0 = (\gamma y, 0, 0)$. The fluid inside and outside the vesicle is described by the Stokes equations, with the following boundary conditions: (i) continuity of the (normal and tangential) velocity across the membrane, (ii) continuity of normal and tangential stress at the membrane, (iii) membrane incompressibility. The basic technical spirit can be found elsewhere$^2,4,11,12$. Here, we merely focus on the results. Lengths are reduced by the vesicle radius $r_0$ ($r_0$ designates the radius of a sphere having the same volume). The shape of the vesicle can be written in the general case as an infinite series on the basis of spherical harmonics $Y_{nm}$

$$r = 1 + \epsilon \sum_{n=0}^{\infty} \sum_{m=-n}^{n} F_{nm}(t) Y_{nm}(\theta, \phi)$$  \hspace{1cm} (1)$$

where $\epsilon$ is a small parameter expressing a small deviation from a sphere, $\theta$ and $\phi$ are the usual angles in spherical coordinates, and $F_{nm}(t)$ is a time dependent amplitude (to be determined) of the corresponding spherical harmonic. We shall set $\epsilon = \sqrt{\Delta}$, where $\Delta$ is the membrane excess area defined by $A = 4\pi + \Delta$, $A$ being the dimensionless area of the vesicle.

Using the expression of spherical harmonics in terms
of Cartesian coordinates, \( r_i \), we have
\[
\sum_{m=-2}^{2} F_{2m}(t) Y_{2m}(\theta, \phi) = \sum_{i,k=x,y,z} 3f_{ik}(t) r_i r_k,
\]
(2)

where \( f_{ik} \) are linear combinations of \( F_{2m} \). Since a shear flow induces a shape deformation from a sphere which involves only second order harmonics (i.e., \( n = 2 \)), only \( Y_{2m} \) enters the calculation [2, 3].

In recent works [8, 9], interesting attempts to deal with the higher order deformation (the next step beyond the leading order theory [4]) have been made. However, these authors take into account only the higher order contribution in the Helfrich bending force. We find that there are other contributions (especially stemming from the corresponding hydrodynamics response) which are at least of the same order. This is the first goal of this communication. The next goal is to analyze the far-reaching consequences of the present theory. Finally we examine some rheological implications.

The method follows the same strategy as in [4], and the technical details will be reported elsewhere, while we focus here on the main results. The extension of the leading order calculation [4] to higher order yields, for the amplitudes of the shape functions \( f_{ij} \), the following evolution equation (please compare with Eq. (3) in Ref. [10])
\[
\frac{Df_{ij}}{Dt} = \frac{20 e_{ij}}{23\lambda + 32} - \frac{24(Z_0 + 6\delta)}{23\lambda + 32} f_{ij} + \epsilon \left[ \frac{480}{7} \frac{\lambda - 2}{(23\lambda + 32)^2} Sd(\epsilon)_{ipj} \right]
+ \frac{288(4\lambda + 136)}{7(23\lambda + 32)^2} Z_0(432\lambda + 1008) \tilde{r} Sd(\epsilon)_{ipj},
\]
(3)

where \( e_{ij} = [\partial_i v_j + \partial_j v_i]/2 \) and \( Sd[b_{ij}] = \frac{1}{2}[b_{ij} + b_{ji} - \frac{\delta}{2}\partial_i \partial_j b_{il}] \). \( Z_0(t) \) is the isotropic component of the membrane tension and is determined by the condition of constant surface excess area. This constraint provides a relation between the \( f_{ij} \)'s which expresses the fact that the shape evolution equations comply with the available excess area. Making use of [3] fixes then \( Z_0 \) in terms of \( f_{ij} \) (and other parameters, like \( \Delta, C_a \)). This relation is lengthy and will be listed in an extended paper.

Let us first discuss the evolution equation (3) and compare it to recent studies [4, 8, 9]. For that purpose, it is convenient to use another set of variables, namely the orientation angle \( \psi \) of the vesicle in the plane of the shear and the shape amplitude \( R \) by setting \( F_{22} = R e^{-2i\psi} [8] \). We may alternatively, instead of \( R \), use the definition \( R/2\sqrt{\Delta} = \cos(\Theta) \), as in [5]. We expand the full equation in powers of \( f_{ij} \) and retain terms up to the higher (fifth) order in a consistent manner. We then perform a straightforward conversion of variables in terms of \( \psi \) and \( \Theta \). We find for \( \Theta \) and \( \psi \) the following equations:
\[
T\partial_t \Theta = -S \sin \Theta \sin 2\Phi + \epsilon \Lambda_1 S \sin 2\Phi \times \cos 3\Theta + \epsilon \Lambda_2 S \sin 2\Phi \cos 2\Theta + \ldots \quad \text{(5)}
\]
\[
T\partial_t \psi = \frac{S}{2} \left[ \frac{\cos 2\psi}{\cos \Theta} [1 + \epsilon \Lambda_2 \sin \Theta] - \Lambda \right] + \ldots, \quad \text{(6)}
\]
where we define, as in [5],
\[
S = \frac{14\pi}{3\sqrt{3}} \frac{\gamma \eta r^3}{\kappa} \epsilon^{-2}, \quad \Lambda_1 = \frac{23\lambda + 32}{240} \sqrt{\frac{30}{\pi}} \epsilon, \quad \Lambda_2 = \frac{1}{25} \sqrt{\frac{10}{\pi}} \frac{49\lambda + 136}{23\lambda + 32}, \quad \Lambda_3 = \left( 10/7 \sqrt{10/\pi (\lambda - 2)/(23\lambda + 32)} \right). \quad \text{(7,8,9)}
\]

where \( \Lambda_1 \) and \( \Lambda_2 \) are the terms in the Helfrich bending force, and \( \Lambda_3 \) is the term proportional to \( \Lambda_1 \). 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 \( \Lambda_1 C_a/\epsilon \). If one has in mind a formal spirit (or a mathematical spirit, in that \( C_a \) is taken of order unity), then \( \epsilon \) 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 \( \Lambda_1 \); while \( T \) can be absorbed in a redefinition of time) does not hold. Indeed, we have three parameters, which are \( C_a, \lambda \) and \( \Delta \), the excess area (or equivalently \( \epsilon \)).

We present now the main outcome following from the study of Eq. (3). We first analyse the \( tt \) regime. Figure 1 presents the orientation angle as a function of \( \lambda \) and compare the results with previous studies. Instead of a square root singularity found for the leading order theory.
the present theory. The same order of discrepancy is found with Ref.\[8\], as shown in Fig.2. The results presented in Ref.\[4\] (in that the $\nu b$ mode coexists with $\nu b$ 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 of $\nu b$ preceding the $\nu b$ bifurcation, in qualitative agreement with Ref.\[3\], as shown Fig.2. The higher order calculation provided here shows significant differences with Ref.\[3\], as shown Fig.2. The results presented in Ref.\[3\] may be viewed as semi-qualitative given the disregard of other terms of the same order. Actually, a simplistic phenomenological model captures the main essential qualitative features of Fig.2[14].

In Fig.2 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 $C_a \rightarrow \infty$ we recover the results of Ref.\[4\] (in that the $\nu b$ mode coexists with $\nu b$ 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 of $\nu b$ preceding the $\nu b$ bifurcation, in qualitative agreement with Ref.\[3\]. The higher order calculation provided here shows significant differences with Ref.\[3\], as shown Fig.2. The results presented in Ref.\[3\] may be viewed as semi-qualitative given the disregard of other terms of the same order. Actually, a simplistic phenomenological model captures the main essential qualitative features of Fig.2[14].

In Fig.2 it was predicted that in the tumbling regime a $\nu b$ mode should take place. This was found to occur as an oscillator (like in a conservative system), since the frequency of 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 $\nu b$ mode becomes a limit cycle (in that all initial conditions in its domain of existence tend towards a closed trajectory in phase space, $(\psi, \alpha)$). As expected from the original theory[4] the $\nu b$ 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 basic understanding of the $\nu b$ 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 $\psi < 0$; see Figure 3) and a rotational part, tending to make a clockwise $\nu b$. Due to the membrane 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 $\nu b$, 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
itself on a time scale of the order of 1. \( \psi > \) to \( \tau_b \). Its actual elongation corresponds to the torque being less efficient. The vesicle feels, so to speak, that the flow compresses the vesicle. Due to this, the applied volume fraction.

For any \( C \) the leading order theory where a cusp singularity is observed in Ref.\[10\] (that \( \lambda \) exhibits a square root singularity, while at larger \( C \) the behavior is smoother.

We have checked that for high enough \( C \approx 100 \) (a quite accessible value in the experiments\[6\]) the full evolution equation produces a coexistence of the \( \psi \) mode and \( \tau_b \) solution, as predicted by the leading order theory (which is expected to be recovered formally at high enough \( C \)). We have checked that depending on initial conditions one mode prevails over the other (as described in\[4\]).

Finally, a systematic solution of Eq. (3) reveals new intriguing fixed points of the dynamics that do not correspond to those reported on so far, and they constitute presently an important line of inquiry. We hope to report along these lines in the future.

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