Vacillating Breathing and Tumbling of Vesicles under Shear Flow

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(Received 27 July 2005; published 18 January 2006)

The dynamics of vesicles under a shear flow are analyzed analytically in the small deformation regime. We derive two coupled nonlinear equations which describe the vesicle orientation in the flow and its shape evolution. A new type of motion is found, namely, a "vacillating-breathing" mode: the vesicle orientation undergoes an oscillation around the flow direction, while the shape executes breathing dynamics. This solution coexists with tumbling. Moreover, we provide an explicit expression for the tumbling threshold. A rheological law for a dilute vesicle suspension is outlined.

DOI: 10.1103/PhysRevLett.96.028104 PACS numbers: 87.16.Dg, 83.50.Ha, 87.17.Jj, 87.19.Tt

Deformable entities under flow, like blood cells, or their biomimetics counterparts, represented by vesicles, reveal interesting dynamics, such as tank-treading [1] and tumbling [2,3]. In recent years nonequilibrium vesicle dynamics have received an increasing amount of interest both theoretically [1–4] and experimentally [5,6]. This interest is motivated by the fact that these systems constitute relatively simple models for the flow and viscoelastic behavior of real cells. Understanding the various intricate dynamics of individual vesicles under nonequilibrium conditions is an essential step in order to make progress in the study of their flow in various geometries, their collective behaviors, their rheological properties, and so on.

Biological and biomimetic flows in which we are interested here belong to the small Reynolds number limit where hydrodynamics are described by the Stokes equations. Despite the linearity of these equations, dynamics of vesicles belong to a class of highly nonlinear and nonlocal problems due to the free boundary character (the membrane shape is not known *a priori*).

By focusing on the relatively small deformation limit [7–9] of vesicles under shear flow, an analytical theory with an arbitrary viscosity contrast between the interior and the exterior of the vesicle is presented. We derive a nonlinear evolution equation (written for a complex variable) for the shape evolution as a function of relevant parameters. The evolution equation contains two dynamical variables: (i) the orientation angle of the vesicle in the flow, (ii) the amplitude of the shape deformation. This is an extension of the Keller-Skallak (KS)[10] model which assumed a shape-preserving solution. Relaxing this assumption we discover a new type of motion that we shall refer to as a vacillating-breathing mode: the vesicle orientation executes oscillations around the flow direction, whereas the long and short axes undergo a breathinglike motion. Moreover, we analyze tumbling and show marked differences with the KS analysis. Outcomes from the study of rheology are briefly outlined.

The vesicle is submitted to a linear shear flow $\mathbf{U}_0 = (\gamma y, 0, 0)$, where γ is the shear rate. The flow outside (and inside) the vesicle is described by the Stokes equations

$$\eta\nabla^2\mathbf{u} - \nabla p = 0, \qquad \nabla.\mathbf{u} = 0, \tag{1}$$

where $\mathbf{u}$ and p are the velocity and the pressure fields, respectively, and η designates the viscosity. The fields referring to the interior of the vesicle will be denoted with a bar (for example, $\bar{p}$, $\bar{\mathbf{u}}\ldots$). $\lambda = \bar{\eta}/\eta$ will designate the viscosity contrast.

The induced velocity fields outside and inside the vesicle are given by the classical Lamb solution [11]

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

and

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

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, which is represented by $p = \sum_n p_n$, where p_n are solid spherical harmonics. The various functions are given by [11] (i) in the exterior $\chi_{-n-1} = r^{-n-1}Q_n$, where the Q_n's depend on the angular variables only and are decomposed on an infinite series of surface spherical harmonics. Similar expressions hold for ϕ_{-n-1} and p_{-n-1} (the angular dependence for these two functions are denoted as S_n and T_n). (ii) In the interior the $r-$ dependence is r^n with the angular functions denoted with a bar (according to our convention). The coefficient of the expansion on spherical harmonics of the functions Q_n, S_n, and so on, are determined from boundary conditions: continuity of the velocity field and the forces at the membrane, $\mathbf{r} = \mathbf{r_m}$. The normal component of the force [12] is given by

$$F_n = \kappa[2H(2H^2 - 2K) + 2\Delta_B H] - 2\zeta H \tag{4}$$

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where κ is the membrane bending rigidity, H is the mean curvature, K the Gauss curvature, Δ_B is the Laplace-Beltrami operator, and $\zeta(\mathbf{r}_m, t)$ is a Lagrange multiplier which enforces the local membrane incompressibility. The tangential [7] part of the force is given by

$$F_\tau = -g^{ij}\mathbf{R_i}\partial_i\zeta \tag{5}$$

where g^{ij} are the elements of the inverse matrix of the metric $g_{ij} = \mathbf{R}_i.\mathbf{R}_j$ induced by the two tangential vectors $\mathbf{R}_i$. ζ is fixed from the projected zero divergence $(1 - n_i n_j)\partial_i u_j = 0$, where n_i is the ith component of the normal vector at the membrane.

From now on lengths will be reduced by the vesicle radius r_0 (r_0 designates the radius of a sphere having the same volume), and time by γ^{-1}. The shape of the vesicle will be parametrized as

$$r = 1 + \epsilon \sum f_n \tag{6}$$

where ϵ is a small parameter. Finally, we require that the membrane velocity coincide with that of the adjacent fluid on both sides of the membrane. The contribution of $\mathbf{U}_0$ in the balance equations contains spherical harmonics of order 2 only. This automatically implies that to leading order only Q_2, T_2,..., and f_2 survive. Each of these quantities are functions of the spherical harmonics. For example,

$$f_2 = \sum_{m=-2}^{2} F_{2m}\mathcal{Y}_{2m}(\theta, \phi) \tag{7}$$

where $\mathcal{Y}_{2m}$ are spherical harmonics. It follows that the vesicle shape evolution to leading order contains only the functions F_{22}, F_{21}, F_{20} (and their complex conjugates).

The algebra leading to the final results are technically involved, and will be reported on elsewhere. The evolution equation for F_{22} is found to be given by

$$-i\epsilon\partial_t F_{22} = F_{22} - h + 2h\Delta^{-1}(|F_{22}|^2 - F_{22}^2) \tag{8}$$

where Δ is the membrane excess area defined by $A = 4\pi + \Delta$, A being the dimensionless area of the vesicle, and $h = 60\sqrt{2\pi/15}/(32 + 23\lambda)$.

In this brief exposition we analyze only the motion in the plane of the shear. In that plane $\mathcal{Y}_{21} = 0$, and we are left only with F_{20} and F_{22}. F_{20} obeys $\epsilon\partial_t F_{20} = -ih\Delta^{-1}(F_{22} - F_{22}^*)$.

It is convenient to rewrite Eq. (8) in terms of a real and an imaginary part. For that purpose we set $F_{22} = Re^{-2i\psi}$. The phase is chosen as -2ψ since, for a shape-preserving motion, we wish (for the sake of comparison with previous works) that ψ coincide with the angle between the long axis and the shear direction. Extracting the real and imaginary parts from (8), we obtain

$$\epsilon\partial_t R = h\left[1 - 4\frac{R^2}{\Delta}\right]\sin(2\psi), \tag{9}$$

$$\epsilon\partial_t\psi = -\frac{1}{2} + \frac{h}{2R}\cos(2\psi). \tag{10}$$

These constitute the basic evolution equations in the small deformation theory. The second equation describes the overall orientation of the vesicle in the flow, while the first one governs the shape evolution. It is interesting to note that to leading order both for droplets [8,9], as well as for capsules [13], the evolution equations are linear. This markedly differs from the vesicle problem where, to leading order, the evolution equation (8) is nonlinear. This is traced back to the constraint of local area incompressibility. Similar types of equations have been suggested recently on the basis of heuristic arguments [3], and have reproduced successfully some numerical results. While the dependencies with ψ are identical, differences are found regarding the variable R, and a full discussion will be presented elsewhere. It is noteworthy that Eqs. (9) and (10), are free of κ (or, more precisely, free of $\chi = \eta\gamma r_0^3/\kappa$, an appropriate dimensionless parameter). Indeed, χ is fixed by Δ from the demand that the shape evolution must conform to the available excess area. The insensitivity to χ of the vesicle tilt angle in a shear flow was also reported numerically even for a large enough deformation [1,2].

In the pure tank-treading regime where the shape is fixed, we have

$$R_0 = \frac{\sqrt{\Delta}}{2},$$
$$\psi_0 = \pm\frac{1}{2}\cos^{-1}\left(\frac{\sqrt{\Delta}}{2h}\right) = \pm\frac{1}{2}\cos^{-1}\left[\frac{(23\lambda + 32)}{120}\sqrt{\frac{15\Delta}{2\pi}}\right]. \tag{11}$$

For $\lambda = 1$ we obtain Seifert's result [7]. The above solution is subject to the condition $\sqrt{\Delta}/2h < 1$, or, equivalently,

$$\lambda < \lambda_c \equiv -\frac{32}{23} + \frac{120}{23}\sqrt{\frac{2\pi}{15\Delta}}. \tag{12}$$

Above λ_c steady solutions cease to exist, and tumbling takes place. We define the reduced volume as $\tau = [V/(4\pi/3)]/(A/4\pi)^{3/2}$ (it follows that $\Delta = 4\pi[\tau^{-2/3} - 1]$, V is the enclosed volume). For $\tau \simeq 1$, λ_c diverges as $1/\sqrt{\tau - 1}$. For a given Δ one finds from (11), by using the expression of λ_c, that $\psi_0 \sim \pm\sqrt{\lambda_c - \lambda}$ (the "+" solution is stable and the "−" one is unstable, which is a signature of a saddle-node bifurcation). Figure 1 shows the boundary between a tank-treading regime (lower part) and the tumbling one. The results following from the KS theory are also shown (they reproduce well the full numerical results [2]). Also shown are the results obtained from an expansion $\psi_0 \simeq \pi/4 - (23\lambda + 32)/140\sqrt{15\Delta/2\pi}$ (valid for a small argument) from which we determine the tumbling threshold by setting $\psi_0 = 0$. Surprisingly, the expanded (or extrapolated) result provides a significantly better agree-

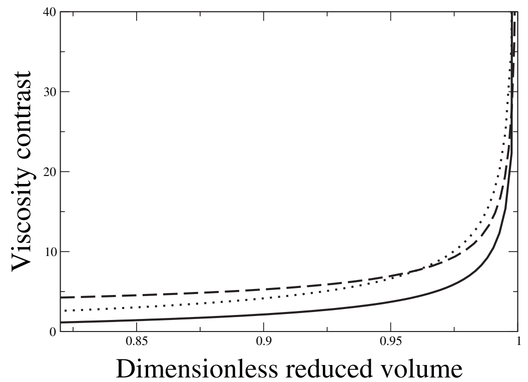

FIG. 1. The tumbling boundary. Solid line: present calculation. Dashed line: the KS theory. Dotted line: the present theory obtained by expansion of ψ_0.

ment with the KS theory even for $\tau \simeq 0.9$ (corresponding to $\Delta = 1$).

Tumbling [10] may be qualitatively understood under a shape-preserving assumption ($R = R_0$). We then obtain from Eq. (8) the Jeffery [10] form

$$\partial_t \psi = -\frac{1}{2} + B\cos(2\psi), \qquad B = \sqrt{\frac{2\pi}{15\Delta}}\frac{60}{23\lambda + 32}. \tag{13}$$

For $B < 1/2$ ($\lambda > \lambda_c$) the tank-treading regime ceases to exist in favor of tumbling via a saddle-node bifurcation.

Interesting enough dynamics are revealed when the assumption of a shape-preserving motion is relaxed [in which case Eqs. (9) and (10) are solved numerically]. The tumbling regime is accompanied with an oscillation of the long and short axes (qualitatively similar to Fig. 4). Comparison of the KS theory to the present analysis reveals significant differences. $d\psi/dt$ is plotted as a function of $\cos(2\psi)$ (Fig. 2). This leads, for the KS theory, to a straight line [as can be seen from Eq. (13)]. Taking into account the deformability of the vesicle, a marked difference is found as shown on Fig. 2 (full line). A linear fit, dictated by the KS theory, produces the dashed-dotted line in Fig. 2. This fit conveys the impression that the effective rotation frequency (represented by A) is smaller (in absolute value) than the KS one. The same holds for B (representing the slope).

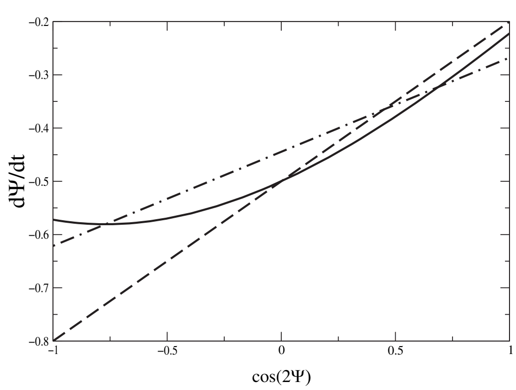

FIG. 2. $\dot{\psi}$ as a function of $\cos(2\psi)$. Solid line: present calculation. Dashed line: the KS theory. Dashed-dotted line: a linear fit from the full calculation. Parameters are $\Delta = 1$ and $h = 0.3$.

A systematic analysis of (9) and (10) reveals the existence of a new kind of motion (Fig. 3): the vesicle aligns along the flow, by executing asymmetric oscillations, whereas the long and short axes show a breathing motion (Fig. 4). This mode is referred to as a *vacillating-breathing* (VB) mode. It coexists with the tumbling one, each having its own basin of attraction. Tumbling occurs if the initial conditions (for R and ψ) are large enough (say of order 1), while the VB mode prevails for small initial values (around 0.1). The coexistence of the two solutions may be inferred from the following reasoning.

The set (9) and (10) admit another fixed point $\psi_0 = 0$, $R_0 = h$. The linear stability of this fixed point (with perturbations $\sim e^{\omega t}$) yields

$$\omega = \pm i\sqrt{\frac{2}{h}\left(1 - \frac{4h^2}{\Delta}\right)}. \tag{14}$$

ω is purely imaginary provided that $4h^2/\Delta < 1$. This condition is nothing but the one fixing the tumbling domain. In other words, ω is purely imaginary in the tumbling regime. This entails that the system behaves about $\psi = 0$ as an oscillator with a period of order $2\pi/\omega$. We have checked that this provides a good agreement with the full nonlinear solution. Note that even a perturbation theory can correctly capture coexistence, as is documented in bifurcations theory [14].

Following Einstein [15], we evaluate the volume average of the stress tensor (actually Einstein evaluated the dissipation) which contains a contribution stemming from the applied shear field, and another due to the presence of the vesicle. This leads us to the following effective viscosity (for a dilute enough suspension):

$$\eta_{\rm eff} = \eta\left[1 + \frac{5}{2}\phi\frac{23\lambda - 16}{23\lambda + 32} + \phi\sqrt{\frac{15}{8\pi}}\frac{(4h^2 - \Delta)}{h}\right] \tag{15}$$

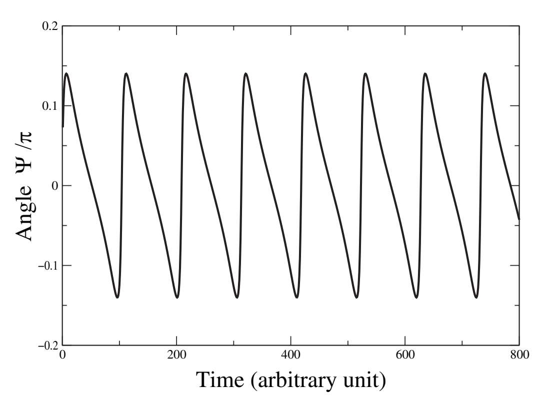

FIG. 3. The behavior of the angle as a function of time for the VB mode. Parameters are $\Delta = 1$ and $h = 0.4$.

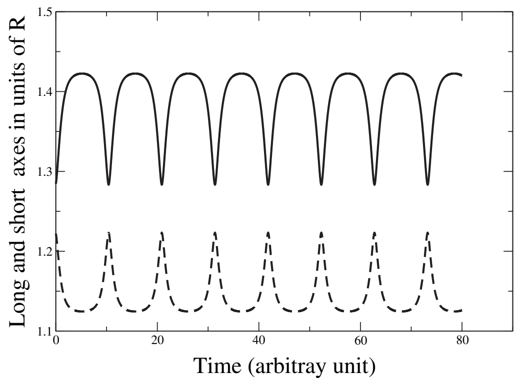

FIG. 4. The behavior of the long and short axes as functions of time for the VB mode, with same parameters as in Fig. 3. We never observed full interchange of short and long axes.

where ϕ is the volume fraction of the vesicles. Interesting limits are recovered. In the spherical case ($\Delta = 0$) and when $\lambda \to \infty$ (rigid spheres) we obtain $\eta_{\rm eff} = \eta[1 + \frac{5}{2}\phi]$, which is the famous Einstein result [15]. This result was initially developed (to our best knowledge) for spherical rigid particles. Actually, this result is more general, since it remains valid for $\Delta = 0$ even for an arbitrary viscosity contrast. This is a consequence of the fact that for a sphere (be it fluid inside or not) the enclosed fluid executes a global solidlike motion (this is a trivial solution and it is unique, owing to the Stokes linearity). An alternative expression for $\eta_{\rm eff}$ is $\eta_{\rm eff} = \eta[1 + 5\phi/2 - \phi\Delta(23\lambda + 32)/16\pi]$. Thus, with increasing the excess area, the effective viscosity should decrease according to this law. This is not devoid of experimental testability. For $\Delta = 0.5$ (corresponding to only 4% in relative excess area where a perturbative scheme is expected to make a sense), we find for $\lambda = 1$ and 2, $\eta_{\rm eff} \simeq \eta[1 + 2\phi]$ and $\eta_{\rm eff} = \eta[1 + 1.5\phi]$, respectively. These are significant enough shifts. Note that we prefer, to some extent, the form (15) since it tells us some important information: the last term vanishes exactly at the tumbling threshold (for $4h^2 = \Delta$). This raises naturally the question about what happens beyond. For example, what is the behavior of effective viscosity in the tumbling regime, the VB one, and so on? In short, we should connect the underlying dynamics to the rheological properties. We hope to investigate this matter further in a future paper.

Several issues deserve future consideration, however. Firstly, thermal fluctuations may play a role [7], and especially for low enough shear rates. Recent experiments by Kanstler and Steinberg [6] reported on this aspect in the tank-treading regime. Their average orientation angle fits remarkably well with deterministic equations [7]. In this respect, the fluctuation-free theory is expected to deliver the essential features regarding the average. Notable effects can, however, be detected due to fluctuations. A precise study of the interplay between shear and fluctuations, both for individual dynamics and rheology, should be addressed in the future. Secondly, while the role of χ is unessential for the fluctuation-free tank treading, this seems not clear yet for other regimes. Finally, higher order expansions are necessary in order to have access to a wider domain of applicability of the theory.

Fruitful discussions with, and a critical reading of the Letter by, T. Podgorski are acknowledged. I am grateful to M. Mader and V. Vitkova for stimulating discussions, and to CNES and CNRS (ACI "modélisation de la cellule et du myocarde") for financial support.

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