

Orientation and Dynamics of a Vesicle in Tank-Treading Motion in Shear Flow

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Experimental results on mean inclination angle and its fluctuation due to thermal noise in tank-treading motion of a vesicle in shear flow as a function of vesicle excess area, normalized shear rate, viscosity, and viscosity contrast between inner and outer fluids, ϵ , are presented. Good quantitative agreement with theory made for $\epsilon = 1$ was found. At $\epsilon > 1$ the dependence is altered significantly. Dependence of the vesicle shape on shear rate is consistent with theory. A tank-treading velocity of the vesicle membrane is found to be a periodic function close to that predicted by theory.

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Dynamics of deformable mesoscopic objects under hydrodynamic stresses determine rheology of many complex fluids, such as emulsions, suspensions of droplets or bubbles, solutions of vesicles, blood, biological fluids, etc. From a theoretical point of view this nonequilibrium problem is rather challenging due to the coupling between the object deformations and the flow that leads to a free-boundary hydrodynamic problem, where the object shape is not given *a priori* but determined by an interplay between flow, bending energy, and various physical constraints.

A vesicle, an example of such deformable objects, is a liquid droplet bounded by a closed phospholipid bilayer membrane (usually unilamellar) suspended in a fluid that can either be the same solvent as an inner one or different. Both the volume and the total surface area of the vesicle are conserved, and the latter means that the membrane dilatation is neglected. Vesicle surface undulations due to thermal fluctuations and an external flow were analyzed in Ref. [1].

In this Letter, we report experimental studies of average inclination angle and its fluctuations, membrane tank-treading velocity and shape of a single vesicle as a function of excess area, normalized shear rate, viscosity, and viscosity contrast, $\epsilon = \eta_{\text{in}}/\eta_{\text{out}}$. Here η_{in} and η_{out} are the dynamic viscosities of inner and outer fluids, respectively.

Dynamics of a vesicle subjected to a shear flow is governed by three main control parameters: the total excess area (dimensionless), $\Delta = S/R^2 - 4\pi$, the viscosity contrast, ϵ , and the dimensionless shear rate, $\chi = \dot{\gamma}\eta_{\text{out}}R^3/\kappa$, that characterizes the deformability of the vesicle under the shear rate, $\dot{\gamma}$. Here R is the effective vesicle radius related to its volume via $V = \frac{4}{3}\pi R^3$, S is the vesicle surface area that exceeds that of a sphere with the same volume by Δ , and κ is the bending rigidity.

As followed from theory [1] and confirmed by numerical simulations [2] at $\epsilon = 1$, a vesicle subjected to a shear flow acquires a stationary mean inclination angle at a given Δ with the membrane that undergoes a tank-treading motion. The stationary inclination angle results from the balance of

the following torques: torque associated with the rotational component of a shear velocity field, torque due to its elongation part, and torque resulting from the tank-treading motion of the membrane [3]. As the rotational torque component pushes the inclination angle away from $\pi/4$ towards lower values, the vesicle membrane acquires a tank-treading motion, through which the elongation component of the torque is introduced into the torque balance [4]. A theory based on a simple model of a fixed shape membrane [3] as well as a more intricate theory that takes into account the shape deformations due to flow [5], and numerical simulations [4,6], predict two types of a vesicle motion. For $\epsilon < \epsilon_c$, where the value of ϵ_c depends on the excess area [4,6], a vesicle achieves a fixed inclination angle with the membrane in a tank-treading motion. At $\epsilon > \epsilon_c$ a transition to a tumbling motion is predicted. The vesicle axis then rotates with respect to the flow direction. In this Letter, we will concentrate just on the first type of the vesicle dynamics.

In the case of $\epsilon = 1$, the detailed calculations that take into account both the vesicle shape deformations due to a shear flow and thermal fluctuations lead to the following expression for the mean inclination angle [1] in a quasi-spherical approximation at $\bar{\Delta} \ll 1$ [see Fig. 1(a) for definition of ϕ]:

$$\phi_0 = \frac{1}{2} \arctan\left(\frac{a - \bar{\Delta}}{\bar{\Delta}}\right)^{1/2} \approx \pi/4 - \frac{\bar{\Delta}^{1/2}}{2a^{1/2}}. \quad (1)$$

Here $a = 4(12/11)^2(2\pi/15) \simeq 1.994$; $\bar{\Delta}$ is the systematic part of the excess area that shows up due to the shear flow and is stored in the second harmonic of the shape deformations, where the last expression is valid at $\bar{\Delta}/a \ll 1$. We would like to point out that Eq. (1) is valid for all values of χ , and this expression is relevant to experimental measurements of the vesicle shape, since only $\bar{\Delta}$ can be measured.

A shear flow pulls out the hidden membrane area from the thermal fluctuations. The more excess area is available, the larger deviations of the mean inclination angle from the equilibrium one, $\pi/4$, obtained for small shear rates. It was reported in Ref. [7] that measurements of ϕ_0 as a function

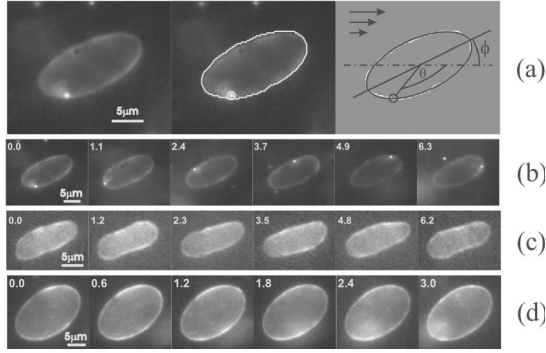


FIG. 1. (a) Image analysis of a vesicle and definitions of ϕ and θ . (b) Time series of snap shots of a vesicle with an attached fluorescent bead (at $\bar{\Delta} = 1$). (c) Time series of snap shots of a vesicle with low viscosity $\eta = 10^{-3}$ Pa \cdot s at $\bar{\Delta} = 1.4$ and $\zeta \approx 65$. (d) The same with high viscosity $\eta = 10^{-2}$ Pa \cdot s at $\bar{\Delta} = 0.4$ and $\zeta \approx 1300$; numbers in the left corners of each image are elapsing time in sec.

of $\bar{\Delta}$ are found to be in good agreement with Eq. (1) but the results were never presented. Reference [1] provides also the equation of motion for the complex amplitudes of the spherical harmonics that parametrize an instantaneous vesicle shape. From the phase of the complex amplitude of the second harmonics one can derive the rms of fluctuations of the vesicle inclination angle caused by the thermal fluctuations, $\langle (\phi - \phi_0)^2 \rangle^{1/2} \equiv \Delta\phi$, at $\chi \gg 1$:

$$\Delta\phi = \left(\frac{24}{55} \frac{1}{4a^{1/2}\zeta} \right)^{1/2} \approx 0.28\zeta^{-1/2}, \quad (2)$$

where $\zeta \equiv \chi\kappa\bar{\Delta}^{1/2}/k_B T$ is the dimensionless shear rate.

Measurements of the vesicle dynamics were conducted in a near-wall shear flow at a half height of a microchannel via epifluorescent microscopy. The rectangular channel with 250 μm height and 150 μm width was manufactured in an elastomer polydimethylsiloxane by a soft lithography [8]. An observation area $90 \times 67 \mu\text{m}^2$ was captured by Panasonic CCD camera and digitized via Ellips Rio frame grabber. A mechanical chopper, synchronized with CCD camera and installed in the Ar-ion laser beam path, reduced actual exposure time down to ~ 1 msec. It allowed to capture vesicles with velocity up to hundreds of $\mu\text{m}/\text{sec}$ without noticeable smearing of the vesicle image. The microchannel can be translated in the direction of the flow. Image processing and control of stepping motor drivers were performed under Matlab.

Vesicles were prepared by electroformation method [9]. Two different lipid solutions for the vesicle preparation were used. The first lipid solution consisted of 1.5 mg/ml of dioleoyl-phosphatidylcholine (DOPC, Sigma) with 0.3 mg/ml of fluorescent phosphatidylcholine (PC) lipid (Molecular Probes) dissolved in 9:1 v/v chloroform-methanol solvent. As an inner fluid, either a glucose-water solution ($\sim 5\%$ w/w) or a pure water, in which the

vesicles were swollen, were used [see Fig. 1(c) and 1(d)]. To increase the fluid viscosity dextran of 500 kDa molecular weight in various concentrations up to 6% w/w was added (variation in viscosity from 10^{-3} until 10^{-2} Pa \cdot s was achieved in such way). The viscosity contrast different from unity was achieved by centrifugation of a vesicle out of a glucose-water-dextran solution and replacing it by a sucrose-water solution of the same osmolarity ($\sim 9\%$ w/w) to compensate for a density difference. The viscosity measurements of every solution were carried out on a viscometer Vilastic-3.

The second lipid solution, which was mainly used for measurements of a membrane tank-treading velocity, consisted of 1.5 mg/ml of dimyristoyl-phosphatidylcholine (DMPC, Sigma), 0.3 mg/ml of fluorescent phosphoethanolamine (PE) lipid (Avanti Lipids, Inc.), and 0.05 mg/ml of Biotinyl PE (biotinated lipid used to attach avidin coated beads, Avanti Lipids, Inc.) dissolved in 9:1 v/v chloroform-methanol solvent. As a solvent 44% w/w glucose-water solution was used with 20 ppm of fluorescent, avidin coated beads (0.5 μm). The experiments were conducted at temperatures above 25 $^\circ\text{C}$ to keep the membrane in a liquid state. Such three-component lipid membrane bears three functions: to be a vesicle shell, to produce fluorescent image, and to provide means for attachment of fluorescent beads used for measurements of the membrane tank-treading velocity [see Fig. 1(b)].

A dilute solution of deflated vesicles was driven through a microchannel by a microsyringe pump at a constant flow rate. The profile of the horizontal velocity was measured by particle tracking velocimetry (PTV) with 1 μm fluorescent beads [see inset in Fig. 2(b)]. The measurements of vesicle dynamics were performed at a distance between about 5 and 45 μm from the wall. The deviation of the shear rate from a constant was at the most $\pm 8\%$ on the vesicle size. The lift force, that results in a vesicle drift comparable with those caused by various experimental factors, does not introduce any noticeable problem in the measurements.

We conducted two types of measurements of the vesicle inclination angles: in a laboratory frame collecting vesicle ensemble statistics for $\epsilon = 1$ at two viscosities $\eta = 10^{-3}$ and 10^{-2} Pa \cdot s, and in a frame moving with a velocity of a single captured vesicle to collect statistics in time for various viscosity contrasts. The latter measurements were performed in the following way. Image processing, first commenced at the grabbing rate 1.5 fps, until the program identified a proper vesicle to follow. Then the translation stage velocity was adjusted to move with the velocity of the captured vesicle, and the grabbing rate was switched to 25 fps. A vesicle can be usually traced for 10–20 sec that allowed to collect up to 500 measurements on a single vesicle. A series of the experiments were carried out on the membrane tank-treading velocities with a fluorescent bead attached to the vesicle membrane [Fig. 1(b)].

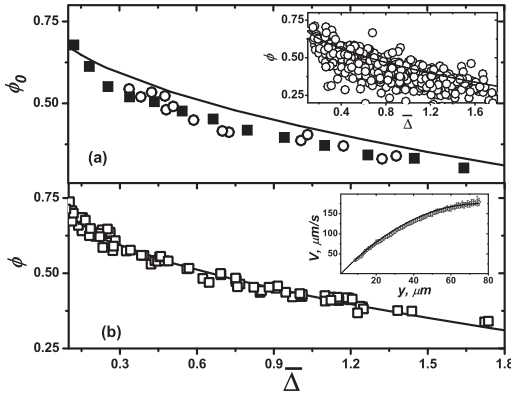


FIG. 2. (a) Average inclination angle, ϕ_0 , as a function of the excess area, $\bar{\Delta}$, at $\epsilon = 1$, $\zeta = 50$, and $\eta = 10^{-3}$ Pa·s; circles are time-averaged and squares are ensemble-averaged data. The inset: inclination angle, ϕ , as a function of $\bar{\Delta}$ at the same parameters. Solid lines on both plots are theoretical curves presented by Eq. (1). (b) ϕ as a function of $\bar{\Delta}$ at $\epsilon = 1$, $\zeta = 800$, and $\eta = 10^{-2}$ Pa·s. The inset: velocity profile measured for the flow discharge rate $Q = 3.3$ nl/sec at a middle height of a microchannel as a function of the distance from the wall in a horizontal plane, y . Solid line is the theoretical curve with the parameters of the experiment [13].

To define the inclination angle and the excess area of a vesicle moving at a given shear rate, image analysis in terms of shape recognition was done using Matlab IP toolbox that includes the following steps [see Fig. 1(a)]: (i) a binary area of a vesicle that provides the shape detection was defined; (ii) the elliptical approximation of the area and determination of main parameters, namely, long and short elliptic axes, $2L$ and $2B$, an inclination angle, ϕ , and the ellipse center coordinates, (x, y) , were carried out; (iii) determination of surface area and effective radius of the ellipse was done via the following expressions: $S \approx 4\pi\{[(L)^p(B)^p + Z^p(B)^p + (L)^p Z^p]/3\}^{1/p}$, and $R = (BLZ)^{1/3}$, where we used $Z = (L + B)/2$ [1] and $p = 1.607$ [10].

The plots in Figs. 2(a) and 2(b) present results of the mean inclination angle measurements at $\epsilon = 1$ for two values of viscosity as a function of the excess area [Figs. 2(a) and 2(b)]. The inset in Fig. 2(a) demonstrates a rather large scatter of the inclination angle due to thermal fluctuations. For each curve with a specific value of viscosity up to 500 vesicles were investigated. Then the data were divided into 13 boxes corresponding to 13 values of $\bar{\Delta}$, and averaged over each box. In such a way, we obtained the dependence of ϕ_0 on $\bar{\Delta}$ averaged over an ensemble. In the case of the time-averaged data, up to 500 measurements in time were taken on one vesicle, and then conducted on 14 different vesicles with different $\bar{\Delta}$. As a result, the mean values, ϕ_0 , averaged either over ensemble or in time, show rather good agreement with the theoretical prediction, presented by the second right-hand side expres-

sion in Eq. (1) [Figs. 2(a) and 2(b)] [1,2]. Both expressions in Eq. (1) are valid at $\bar{\Delta}/a \ll 1$ but the second one surprisingly agrees better with the data up to $\bar{\Delta} = 1.8$. The data for a low viscosity fluid are slightly shifted down compared with the theoretical curve [see Fig. 2(a)]. This small discrepancy can be explained by the greater contribution of odd harmonics (first and third) into the mean inclination angle (through the phase) due to stronger fluctuations in a low viscosity fluid. The solid lines on all plots of ϕ_0 versus $\bar{\Delta}$ are not fits but theoretical predictions defined by Eq. (1) without any fitting parameter. It is also remarkable that for higher viscosity and higher shear rates the scatter reduces drastically [see Fig. 2(b)]. An increase in the viscosity contrast alters the dependence of ϕ_0 on $\bar{\Delta}$ that is seen from the data presented in Fig. 3 for values of $\epsilon = 1, 2.7$, and 5.6 . The solid line is the same theoretical curve defined by Eq. (1) as in Fig. 2, and the dotted and dashed curves are the fits. It is obvious from the plots that the larger the viscosity contrast, the stronger the decrease of ϕ_0 at the same $\bar{\Delta}$ [11]. Four points obtained by the numerical simulations at $\epsilon \approx 3.7$ in Ref. [4] were added for comparison. Similarity in the overall dependence is rather obvious.

The dependence of the rms of fluctuations of the inclination angle, $\Delta\phi$, on ζ is presented in Fig. 4 for three values of ϵ . For $\epsilon = 1$, the data are presented in linear-log coordinates together with the error bars. The latter results from two main sources of measurement errors: of the effective radius for smaller vesicles and of the inclination angle at higher values of χ . As clearly seen from the plots, the experimental data for all values of ϵ agree well with theoretical curves defined by Eq. (2) within the error bars.

We also study a vesicle deformation in a shear flow, described by the deformation parameter, $D = \frac{L-B}{L+B}$, as a function of the control parameter, $\zeta\bar{\Delta}^{-1/2} \equiv \chi\kappa/k_B T$, at different $\bar{\Delta}$. This issue has been already investigated ex-

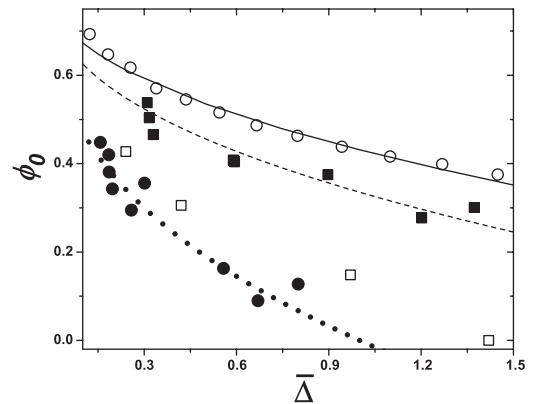


FIG. 3. ϕ_0 , as a function of $\bar{\Delta}$: circles— $\epsilon = 1$, $\eta = 10^{-2}$ Pa·s, ensemble-averaged data presented in Fig. 2(b); squares— $\epsilon = 2.7$; full circles— $\epsilon = 5.6$, open squares—numerical simulations from Ref. [4] at $\epsilon \approx 3.7$. Dotted curve is the fit $\phi_0 = \pi/4 - \alpha\bar{\Delta}^\beta$ with $\alpha = 0.785$, $\beta = 0.4$, and dashed curve with $\alpha = 0.45$, $\beta = 0.45$.

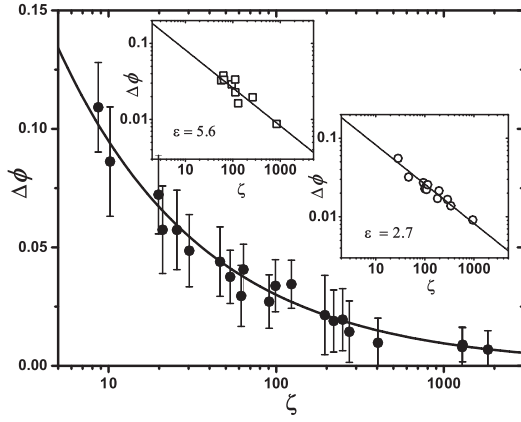


FIG. 4. rms of fluctuations of the inclination angle, $\Delta\phi$, vs the normalized shear rate, ζ : full circles with error bars— $\epsilon = 1$ in linear-log presentation; open circles— $\epsilon = 2.7$, and squares— $\epsilon = 5.6$ in the insets in log-log presentation. Solid lines are theoretical curves presented by Eq. (2).

perimentally in Ref. [12], and a comparison with theory was made [1]. However, quantitative comparison with the data was absent. In Fig. 5 we present the data on the deformation parameter, D versus $\zeta\bar{\Delta}^{-1/2}$, for several values of the final excess area. All three data sets present the crossover behavior and show a tendency to a saturation in the deformation parameter at sufficiently large shear rates. The latter is qualitatively consistent with the theory [1] though not quantitatively due to large values of $\bar{\Delta}$ found in the experiment, whereas the theory is developed for small deviations from the spherical shape.

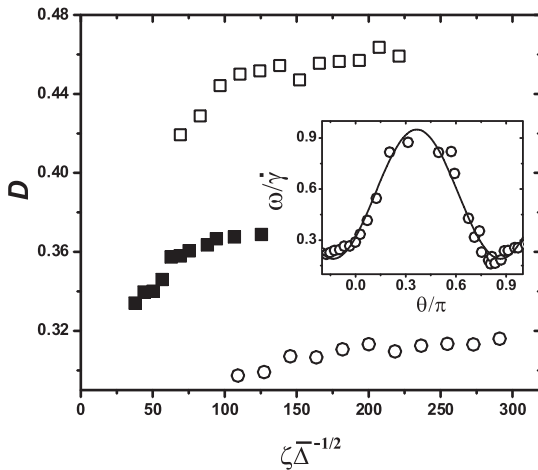


FIG. 5. Dependence of the deformation parameter, D , as a function of $\zeta\bar{\Delta}^{-1/2}$ for several values of $\bar{\Delta}$: open squares—1.1, full squares—0.8, open circles—0.7. The inset: the reduced local rotation velocity of a vesicle membrane in a tank-treading motion, $\omega/\dot{\gamma}$, as a function of the angle along the membrane, θ , at $\bar{\Delta} = 1$. Solid line is the theoretical fit.

And finally, in the inset in Fig. 5 we show the result of the measurements of the local rotation velocity of a vesicle membrane, $\omega \equiv \frac{d\theta}{dt}$, normalized by the shear rate, $\dot{\gamma}$, as a function of angle θ along the membrane defined in Fig. 1(a). As predicted by theory [3,4], the tank-treading rotation velocity (or local frequency) is periodic and is described by the dependence $\frac{\omega}{\dot{\gamma}} = C + D \cos(2\theta)$ with $C = 0.57$ and $D = 0.38$, found from the fit.

To conclude, the experimental results on the mean inclination angle and its rms of fluctuations show surprising agreement with the theoretical predictions [1], in spite of the fact that the theory is made for quasispherical vesicles, whereas vesicles with rather strong deviations from sphericity (prolate) are mostly observed in the experiment. At the viscosity contrast larger than unity the dependence of ϕ_0 on $\bar{\Delta}$ differs significantly from that found at $\epsilon = 1$. The larger the viscosity contrast, the smaller ϕ_0 at the same $\bar{\Delta}$. The dependence of the vesicle shape on the shear rate is also consistent with the theory. The tank-treading rotation velocity of a vesicle membrane is found to be a periodic function.

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