Vesicle dynamics in elongation flow: Wrinkling instability and bud formation.

Vasily Kantsler, Enrico Segre and Victor Steinberg
Department of Physics of Complex Systems,
Weizmann Institute of Science, Rehovot, 76100 Israel
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We present experimental results on the relaxation dynamics of vesicles subjected to a time-dependent elongation flow. We observed and characterized a new instability, which results in the formation of higher order modes of the vesicle shape (wrinkles), after a switch in the direction of the gradient of the velocity. This surprising generation of membrane wrinkles can be explained by the appearance of a negative surface tension during the vesicle deflation, due to compression in a sign-switching transient. Moreover, the formation of buds in the vesicle membrane has been observed in the vicinity of the dynamical transition point.

The stationary, stretched state is known to obey \( \Delta \dot{\varepsilon} = 0 \), with \( \varepsilon \) the elongation rate and \( \Delta \) the excess area per vesicle. This expression holds for the time scale of the experiment, with the excess area \( \Delta = S/R^2 - 4\pi \), where \( S \) is the total surface area of the vesicle and \( R \) its effective radius, defined via the volume \( V = \frac{4}{3} \pi R^3 \).

Measurements of the vesicle dynamics were conducted in the vicinity of the stagnation point (\( v_x = v_y = v_z = 0 \)) via epi-fluorescent or phase contrast microscopy. The flow was produced in a cross-slot micro-channel of 500 \( \mu \)m wide and 320 \( \mu \)m in height manufactured in elastomer (PDMS) by soft lithography [18]. The details of the design and of the arrangement will be published elsewhere.

Particle tracking velocimetry measurements of the flow field show that the deviation of the elongation rate \( (\Delta \dot{\varepsilon})_{x/y} / \dot{\varepsilon} \) across the size of the observation window is \(< 5\%\), deviations of \( \dot{\varepsilon} \) in the z-direction on the scale of the vesicle were \((\Delta \dot{\varepsilon})_z / \dot{\varepsilon} < 5\%\), and that the ratio of shear velocity gradient \( \gamma_{xy} \) to \( \dot{\varepsilon} \) on the size of the vesicle did not exceed \((\Delta \dot{\varepsilon})_z / \dot{\varepsilon} \). Experiments were performed in...
the range of velocity gradients $\dot{\varepsilon} = 0.05 \div 10$. We define the dimensionless strain $\chi = \dot{\varepsilon}\eta_{\text{out}} R^3/\kappa$, where $\kappa \approx 10^{-12}$ erg for DOPC [19]. The viscosity of the fluid inside the vesicle, $\eta_{\text{in}}$, can be different from the viscosity $\eta_{\text{out}}$ of the surrounding fluid, and their ratio $\lambda$ was varied across the experiments. The lipid solutions consisted of 85% DOPC (Sigma) and 15% NBD-PC (fluorescent lipid, Molecular Probes) dissolved in 9:1 v/v chloroform-methanol solvent (1.8 mg total lipids/ml), or DOPC in the solvent (1.5 mg/ml). The methods and conditions of the preparation of the vesicles for the experiments have been described previously [13, 14, 16]. The first set of the experiments was performed suddenly switching on the flow, starting after the vesicle had relaxed into an equilibrium shape. An initial growth of $\chi$ was followed by a dynamically excited relaxation to a stationary state. An example is shown in the lower inset of Fig[3]. The temporal evolution of the dynamics of the spectrum $u_k(t)$ is shown in Fig[2]. The temporal evolution of the power spectra is shown in Fig[2]. We found that the spectra show a $P_k \propto k^{-4}$ dependence for $\chi$ less than some critical value $\chi_c$, that is a well-known spectral decay due to thermal noise [1]. For larger $\chi$, the spectra become rather flat at smaller $k$, while the higher modes are still excited with the equilibrium spectral decay. From the rather sharp transition from the flat to the $k^{-4}$ spectrum around some $k = k_{thr}$, we can postulate that for $\chi > \chi_c$ modes with $k < k_{thr}$ are excited dynamically, while the higher modes are excited not by the flow but rather by thermal noise. We determine $\chi_c$ as the threshold above which the modes with $k \geq 3$ are excited ($k = 3$ is the first mode higher than elliptical). To this extent, we define $k^* = \sqrt{\sum_{k=3}^{19} k^2 P_k}$ and the restriction of $k^*$ on $\chi$, averaged over $\approx 200$ data points, is shown in Fig[3]. For $\chi < \chi_c$, $k^*$ remains constant, which means that the spectrum of all modes with $k \geq 3$ obeys the equilibrium distribution, $k^{-4}$. The growth of $k^*$ starts at $\chi = \chi_c$ with $\chi_c = 6.5 \pm 0.8$, which is identified as the onset of the wrinkling instability. The dependence of $k^*$ above the instability threshold can be fitted by $\sim \chi^{1/4}$ in a good agreement with numerical simulations in the same range of $\chi$ [17]. All the experiments were done for $\lambda = 1$, and no investigation on the influence of viscosity contrast on the wrinkling phenomena was done.

As we pointed out in the introduction, the generation of higher order modes in the vesicle shape, i.e. wrinkles, observed during the relaxation dynamics should lead to a tremendous increase in the elastic energy, which is unlikely, for a vesicle with positive surface tension. On the other hand, a recent theory predicts that a sudden switch...
FIG. 2: Wrinkling instability. Snapshots of vesicle dynamics in time-dependent elongation flow at $\lambda = 1$, $\Delta \approx 1$ and: (a) $\chi = 8.1$, (b) $\chi = 81$, (c) $\chi = 323.5$. The scale bar is 20 $\mu m$, numbers are $t \dot{\epsilon}$. Plots below the images are the data analysis for each of the cases above: (d) - amplitudes $A(\theta, t)$ of higher harmonics versus $\theta$ and $t \dot{\epsilon}$ (values in color), (e) $|u_k|^2(t)$ are the instantaneous Fourier spectra of the amplitudes $A(\theta, t)$ at various $t \dot{\epsilon}$ (values in color), (f) $D(t)$ versus $t \dot{\epsilon}$. Columns ex_a, ex_b, ex_c correspond to the data presented in row (a), (b), (c).

In direction of the velocity gradient, $\dot{\epsilon}(t) = \text{sign}(t) \dot{\epsilon}_0$, is effectively equivalent to a negative surface tension and leads to the excitation of modes with $k^2 \leq \beta \chi / \sqrt{\Delta}$, where $\beta$ is the numerical factor [17]. Then the theory gives $\chi_c \simeq 1.2$ for $k = 3$ and $\Delta \simeq 0.6$, which corresponds to our average $\langle \Delta_i \rangle$ over the data set in the transient region. The theoretical value of $\chi_c$ is of the same order as the experimental one, and the difference can be attributed, first, to uncertainty in the value of $\kappa$ taken and to rather rough estimates based on an isotropic surface tension [17].

Another interesting phenomenon observed is the formation of buds. These could be seen intermittently in the experiments: sometimes the vesicle surface folds to
FIG. 3: $k^*$ versus $\chi$. The arrow defines $\chi_c$, the onset of excitation of the mode $k = 3$. Dashed line is a fit $\sim \chi^{1/4}$ above the instability threshold. The upper inset illustrates the image analysis. Lower inset: averaged power spectrum for various $\chi$: squares – 2.6; open circles – 24; circles – 116; dashed lines show a $\propto k^{-4}$ dependence.

FIG. 4: Formation of buds. (a) $\chi = 6.8, \Delta = 0.9$; (b) $\chi = 179, \Delta = 0.4$. The scale bar is 10 $\mu$m, numbers are $\dot{t}$.

To summarize, we presented new experimental results about the relaxation dynamics of vesicles in elongation flows suddenly switched on or reversed. When the vesicle relaxes from its equilibrium shape towards a new stationary, stretched shape in the elongation flow, the scaling of the dynamics with the elongation rate and the linear dependence of the relaxation velocity on the viscosity contrast were found to be in agreement with the theoretical predictions [3, 4, 17]. We also observed and characterized a new instability, that results in the excitation of higher order modes, i.e. wrinkles, in the membrane, during the vesicle relaxation following the reversal of the velocity gradient. This unexpected generation of higher order modes suggests that only the appearance of a negative surface tension during the vesicle deflation due to compression in the transient can explain the effect. A recent theory [17] used this physical picture to derive a criterion for the onset of the instability and the power law dependence of the average wave number of the higher order modes as a function of $\chi$, which is in reasonable agreement with our experiment. Finally we observed and report here, albeit without a quantitative investigation, the phenomenon of bud formation during the transient dynamics, particularly close to the wrinkling instability threshold.

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