Arnold Tongues in a Microfluidic Drop Emitter

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The Letter reports an experimental study of microfluidic droplets produced in T junctions and subjected to a local periodic forcing. Synchronized and quasiperiodic regimes—organized into Arnold tongues and devil staircases—are reported for the first time for a system dedicated to drop emission. The nature of the dynamical regime controls the droplet characteristics. These phenomena are mostly controlled by the characteristics of the forcing and the flow conditions.

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Emulsions are dispersions of micrometric droplets in liquids. Emulsions are widely used in a number of industrial domains, such as pharmaceutical [1,2], cosmetic [3], and food industries [4,5]. Over the last 50 years, emulsion science made considerable progress at fundamental and engineering levels. Recently, microfluidic technology offered new prospectives for this domain. In microfluidic technology, droplets of micrometer sizes are produced [6–12] and individually manipulated. This defines a ‘‘bottom up’’ approach to emulsion science, by opposition to the ‘‘top down’’ traditional techniques that typically handle large populations of droplets. In this context, tasks out of reach by traditional approaches, such as controlling the droplet size without changing the conditions of production, may be envisioned. To achieve such a task, a natural idea is to operate with a physical perturbation imposed close to the region where the droplets are formed and keep the flow rates, in the average, constant. Nonetheless, the drop formation process is a nonlinear instability, and its coupling with an actuator may give rise to complex behavior. The purpose of this Letter is to unravel the phenomena occurring in such a situation, and examine the consequences on the characteristics of the emitted droplets. By working with mechanical integrated actuators in T junction drop emitters, we obtained Arnold tongues and devil staircases, with characteristics mostly controlled by the flow conditions. Arnold tongues were observed in physical and chemical systems [13,14], but to the best of our knowledge, their observation in a drop emitter, miniaturized or not, along which their impact on the drop size characteristics have never been reported so far.

The experimental system is sketched in Fig. 1. It consists in a main channel that forms a T junction with a channel perpendicular to it (‘‘side channel’’) equipped with a local actuator. The actuator is made using multisoft layer lithography [15]. Briefly, this technology consists of microfabricating two polydimethylsiloxane (PDMS) channels, placed on top of each other, and separated by a thin membrane. Taking advantage of the elastic properties of PDMS, one may easily deflect the membrane by applying pressure on the membrane. In the experiments we are carrying out, the actuating channels are 20 μm deep and 100 μm wide. The dimensions of the working channels are either 20×200 or 40×200 μm^2. The membrane that separates the two channels (working and actuating) is 20 μm thick. In our device, the actuating channel stands on the lower level, the working channel on the upper one.

Tetradecane with surfactant (SPAN 80 at concentrations between 1% and 3% in mass) and deionized water mixed with fluorescein are the working fluids. They are driven, respectively, through the main and side channels. They meet at the T junction, forming droplets. We used hydrostatic pressure or syringe pumps to drive the fluids, spanning a range of flow rates lying between 0.1 and 20 μl/ min. The actuation channel is filled with water. Electromagnetic valves driven by waveform generators force the pressure above the membrane to vary periodically in time. The membrane thus periodically deflects at controlled amplitudes and frequencies. The observations of the oil-water structures are made using fluorescence microscopy. In order to characterize the dynamics of the system, we measure the local fluorescence intensity at a few hun-

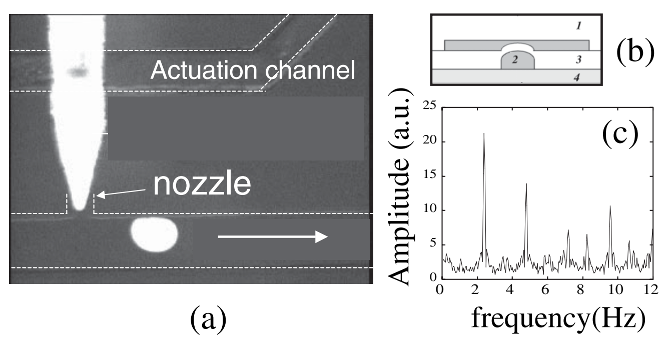

FIG. 1. (a) Top view of the experiment. Tetradecane (black) and water labeled with fluorescein (white) meet in the T junction, emitting droplets. (b) Sketch of the actuation system (1) working channel; (2) actuation channel; textured regions such as (3) PDMS; (4) glass substrate. The working and the actuation channels are separated by a membrane. (c) Typical Fourier power spectrum for a T junction producing droplets without actuation.

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dred microns from the T junction downstream. This signal is controlled by the fluorescence emitted by the droplets driven along the main channel. When equal size droplets are emitted at a constant frequency, the signal that we obtain is periodic in time. When the emission is aperiodic and the droplets have irregular sizes, the observed signal is aperiodic. This measurement thus allows one to characterize the dynamical state of the system. The fluorescence intensity is recorded and analyzed by various methods (Fourier transforms, statistical distributions, etc.). We also systematically measured the droplets' areas by using standard image processing techniques.

In the absence of an external forcing, and within the range of flow rates we considered, one produces water droplets in oil with sizes comparable to the microchannel's width. The drop formation process proceeds in several steps: water penetrates into the main channel, forms a blob, developing a neck; the neck elongates and becomes thinner as the water blob is advected downstream; it eventually breaks up, giving rise to the emission of a droplet. This process is periodic in time, as confirmed by the Fourier spectra that include a single fundamental peak and its harmonics [see Fig. 1(c)]. We have documented in detail the dependence of the fundamental frequency f_0 with the flow-rate conditions.

The mechanical actuators affect the drop formation process in a rather subtle way: the actuators modulate the water flow rate, which in turn modulates the natural frequency f_0. We deal here with a parametric resonance process interacting with a nonlinear instability. From the knowledge of similar dynamical situations [16], one may expect that synchronized and quasiperiodic regimes develop in our system. This is what we observed: an example of a synchronized regime, obtained with oil and water being driven by gravitational forces, is shown in Fig. 2(a). Here, the system is forced at a frequency f_f equal to 1.013 times the natural frequency f_0, and we observe that the natural oscillator synchronizes with the external frequency. Synchronized states are obtained by forcing the system at a frequency close to a multiple (or some rational number) times the natural frequency. An example is shown in Fig. 2(b), where the system is forced at 3.16 times the natural frequency f_0 and the droplet emission synchronizes at a frequency equal to one-third of the forcing frequency. Similar synchronized states were obtained, in which droplets are emitted at $1/2$, $2/3$ [see Fig. 2(c)], $3/5$, $1/4$, or $1/5$ the forcing frequency. In the synchronized states, the system delivers regular drops at regular intervals of time. The other regimes we observed are quasiperiodic (QP). An example is shown in Fig. 2(d), where the system was forced at a ratio frequency $f_0/f_f = 0.53$. In this case, the spectrum displays a series of distinct peaks given by a linear combination of two independent frequencies (with integer coefficients)—the natural one and the forcing one. This is characteristic of a

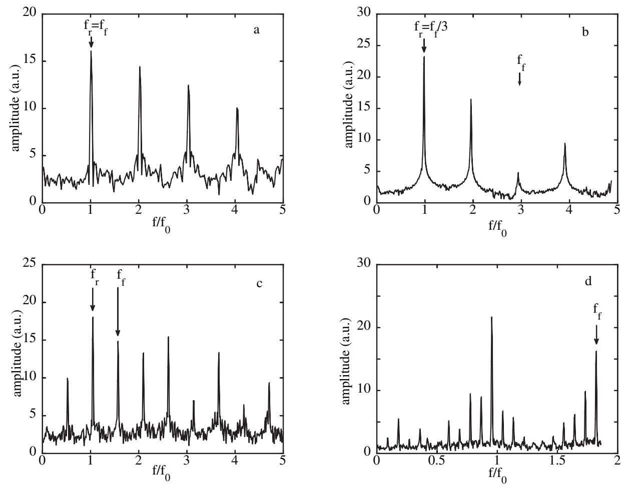

FIG. 2. Fourier power spectra obtained at different forcing frequencies, with pressure driven flows, in a 40 μm deep microchannel; the natural frequency f_0 is 2.37 Hz in cases (a), (b), and (c). (a) $f_f = 2.4$ Hz ($1/1$ resonance). (b) $f_f = 7.5$ Hz ($1/3$ resonance). (c) $f_f = 3.76$ Hz ($2/3$ resonance). (d) (Here, the natural frequency is 6.7 Hz.) $f_f = 12.8$ Hz (quasiperiodic regime).

quasiperiodic regime [16]. In the quasiperiodic regimes, the droplets are emitted irregularly and they have irregular sizes.

By varying the forcing frequency f_f and spanning a range of actuation pressures P (which controls the forcing amplitude), one can map out the phase-locked and quasiperiodic states on a single diagram (see Fig. 3). In this diagram, the gray regions represent conditions for which the droplet emission is synchronized at the forcing frequency times some rational number. One obtains tongues—Arnold tongues—with widths growing with the forcing amplitude. Outside the tongues, the regimes are quasiperiodic.

We examined horizontal cuts of such a diagram, by keeping P fixed and varying the forcing frequency f_f. A typical plot characterizing such a cut is shown in Fig. 3(b). Here, the "winding number" W is determined by counting the number of drops N_d emitted during a large number N_f of forcing periods, and computing the ratio N_d/N_f. The evolution of W with f_f has the form of a staircase that evokes the "devil staircase" obtained close to the chaotic line in the circle map model [16]. The staircase of Fig. 3(b) reveals three plateaus (there are others located at rational numbers such as $2/3$, but they are hard to distinguish). Outside the plateaus, one has quasiperiodic regimes, with winding numbers varying with the forcing frequency.

We found that the width of the synchronized regions and thus the sizes of the steps in the devil staircases mostly depend, for a fixed geometry, on the flow rates and—as shown above—on the actuating pressure amplitude P. The other parameters (SPAN 80 concentrations in the range 1%–3%, detailed waveform of the forcing) play a minor

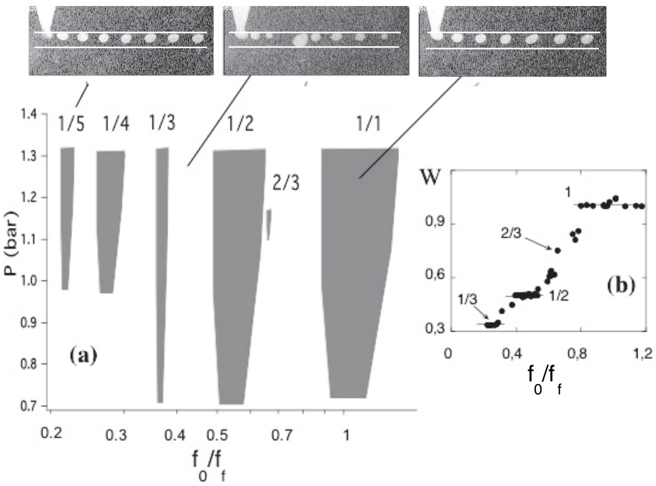

FIG. 3. (a) Synchronized (gray zones) and quasiperiodic regimes (white zones) shown on a diagram where the ordinate is P and the abscissa is the ratio of the natural frequency f_0 over the forced frequency f_f. Syringe pumps are used, imposing oil and water flow rates of, respectively, 4 and 0.3 μl/ min . The microchannel depth is 20 μm. Droplets are shown in some cases. (b) Evolution of the winding numbers W with the ratio of the natural frequency over the forcing frequency. The microchannel depth is 40 μm in this case, and the oil and water flow rates are 2 and 0.40 μl/ min , respectively. The plateaus, which correspond to specific rational numbers, reveal synchronized states; outside the plateaus, the regimes are quasiperiodic and the winding number varies with the forcing frequency. This plot is often called "devil staircase."

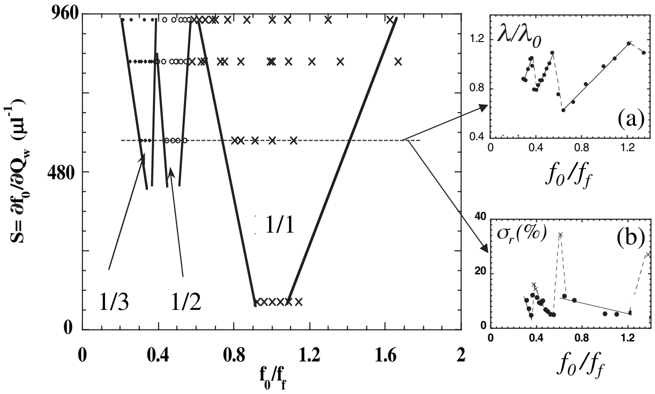

FIG. 4. Regions of synchronization and quasiperiodicity obtained for an oil flow rate of 6.7 μl/ min and an actuation pressure $P = 2$ bar. The range of variation of the water flow rate extends from 0.3 to 4 μl/ min . The susceptibility S is determined, without forcing, from the experimental curve $f_0 = f(Q_w)$. The symbols appearing on the plot signal the various synchronized regimes obtained at 1 (crosses), 1/2 (circles), or 1/3 (diamonds) of the forcing frequency. (a) Evolution with f_0/f_f of the ratio of the droplet size λ (as compared to λ_0, obtained without forcing. (b) Evolution with f_0/f_f the relative dispersion σ_r of the droplet sizes. On this plot, the crosses correspond to QP regimes and the disks to synchronized states. Plots (a) and (b) are obtained for $S = 550$ μl^{-1}.

role. The dependence in P and in the flow conditions can be accounted for by formalizing a preceding remark about parametric resonance. In our system, the natural frequency f_0 depends on the water flow rate Q_w that is modulated by the actuator at an amplitude ε and a forcing frequency f_f, for small ε, and one may write the following expression:

$$f_0(Q_w) = f_0(\bar{Q}_w + \varepsilon \sin 2\pi f_f t) \approx f_0(\bar{Q}_w) + \varepsilon \frac{\partial f_0}{\partial Q_w} \sin 2\pi f_f t$$

in which $\bar{Q}_W$ is the time-averaged water flow rate, f_f the forcing frequency, t the time, and $\frac{\partial f_0}{\partial Q_W}$ the derivative of f_0 with respect to the water flow rate $\bar{Q}_W$, for a fixed oil flow rate, without forcing. The formula shows that the "susceptibility" $S = \frac{\partial f_0}{\partial Q_W}$ controls, in addition to the actuation pressure P (incorporated in the parameter ε), the effective amplitude of the forcing, and thereby the sizes of the domains of existence of the synchronized regimes. This feature is shown in Fig. 4 in which the oil flow rate along with pressure P is kept constant, but different water flow rates are imposed. We obtain Arnold tongues similar to Fig. 3(a), but now depending on the susceptibility S instead of the actuation pressure P. We plotted the evolution of the droplet sizes with the forcing frequency, for a particular value of S [see Fig. 4(a)]. The curve has the form of a saw, each tooth corresponding to a synchronized region. Along each tooth, the droplet volume varies as the inverse of the actual emission frequency, a law that can be accounted for by invoking volume conservation. Figure 4(b) is reminiscent of the devil staircase. We also measured in the same conditions the evolution of the dispersion in size of the emulsion (i.e., the standard type deviation of the droplet size distribution divided by their mean size) with the forcing frequency [see Fig. 4(b)]. We found that in quasiperiodic regimes, the dispersion is several times larger than in synchronized states. This reveals a spectacular impact of the nature of the dynamical regime on the droplet characteristics.

Synchronized regimes are interesting from the viewpoint of droplet size control and production of monodispersed emulsions. According to our study, broad domains of synchronization are obtained at large P and large S. One remarkable example is shown in Fig. 5: here, we imposed S on the order of 2000 μl^{-1}—and we worked at a constant maximum actuation pressure in a 20 μm high microchannel. In this case, the devil staircase shrinks to a single step located at a winding number equal to 1. Here, synchronization holds throughout the 1 order of magnitude range of variations of the forcing frequency. On Fig. 5, we have reported the evolution of the drop volume in a function of the actuation frequency. The graph is related to that of Fig. 4(a), but with a single tooth. The present flow conditions appear as particularly suitable for droplet control.

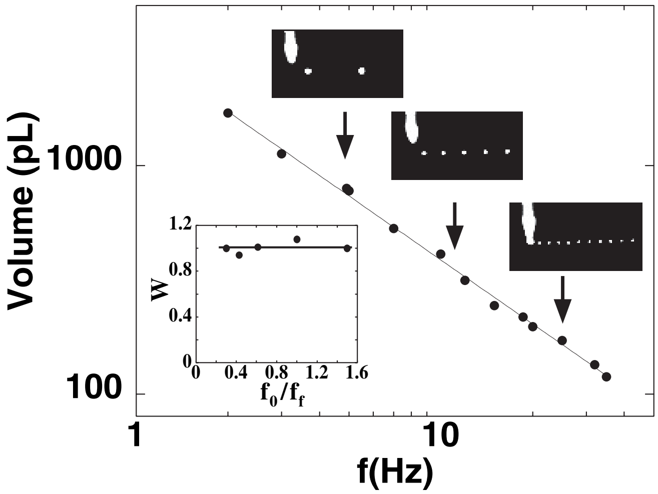

FIG. 5. Evolution of the droplet volumes in a function of the actuation frequency, in a 20 μm deep microchannel; oil and water flow rates are, respectively, 4 and 0.1 μl/min, and the actuation pressure P is 1.7 bar. The pictures represent droplets at various frequencies. The inset shows the evolution of the winding number with f_0/f_f, and indicates that the devil staircase includes one single step.

To conclude, we observed complex behavior in a droplet emission system dedicated to produce controlled emulsions in a microfluidic system. By working with appropriate flow conditions, one may either favor synchronized regimes giving rise to monodisperse emulsions of controlled sizes or promote quasiperiodic regimes giving rise to polydisperse emulsions. The complex behavior we observed here can be qualitatively accounted for by referring to a parametric resonance mechanism coupled to a nonlinear instability. Arnold tongues and devil staircases along with their spectacular impact on the droplet characteristics have never been observed so far in a hydrodynamic system, miniaturized or not, dedicated to droplet production.

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