Simulation of droplet trains in microfluidic networks

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We show that in a microfluidic network with low Reynolds numbers, a system can be irreversible due to hysteresis effects. We simulated a network of pipes that was used in a recent experiment. The network consists of one loop connected to input and output pipes. A train of droplets enters the system at a uniform rate, but the droplets may leave the system in a periodic or even a chaotic pattern. The output pattern depends on the time interval between incoming droplets as well as the network geometry. For some parameters, the system is not reversible.

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Fluids behave differently at the micro scale, where factors such as surface tension, energy dissipation, and viscosity dominate. The Reynolds number (RN) is low and the flow stays laminar. This leads to nonintuitive behavior in a microfluidic Network (MN). MNs are networks of channels and pipes of 1 to 1000 μm [1,2]. They are applied in inkjet printers, lab-on-a-chip devices, fuel cells [3], and biochips [4,5]. Moreover, it is possible to make logical gates, signal encoders, and decoders [6,7].

Fuerstman *et al.* investigated coding and decoding signals using a simple network of microtubes. Their network is made of a single loop, with one input and one output [7] (Fig. 1). A constant flow of fluid enters the system from the input pipe and leaves from the output pipe. The fluid carries droplets of an immiscible liquid. The droplets are large enough to block the pipes but small enough to be stable along the path. At a T-junction, the droplets cannot split. Which path they take depends upon the fluxes in the pipes. The fluxes are determined by the droplets in the pipes so the process is history dependent. Droplets and carrier flow make a binary signal that carries information. This suggests that MNs can be represented by equivalent electrical circuits [8,9].

The flow rate in any branch is determined by the loop geometry and the presence of droplets in the branches. While the capillary force of a droplet slows down the current in one branch, the other branch become more favorable for the next incoming droplet. Considering droplets as 1 and no droplets as 0, we can define a binary signal. Such a signal can be encoded to a different signal by a MN as simple as a loop. The output pattern depends on the time intervals between input droplets and the geometry of the loop. This can be seen as a way to code a signal. It is possible to restore the original signal by a decoder. The reversible dynamics of fluids with low RNs suggest that the signal could be decoded if the output signal were fed to a system of pipes with the same geometry [7]. The possibility of making logical microfluidic devices has been examined by Prakash and Gershenfeldof based on the same concept [6] which has raised the interest of computer scientists [10].

To study a single loop encoder, we feed it with a uniform chain of droplets and see how the loop alters the intervals between droplets. In addition, we can track the droplets inside the system, providing information about the branch that a droplet selects. This information gives us another informative binary signal, which has been studied by Jousse *et al.* [11]. We ran a simulation based on the experiment by Fuerstman *et al.* [7]. Our approach is similar to the approach of Schindler and Ajdari [9]. The main difference is that we have taken in to account the velocity dependence effect of droplets. Our results show that the system can be chaotic, even though the RN is small. This is because of the nonlinear, history-dependent dynamics of the system. In addition, reversibility is not a general property of the system and it may break down even in the nonchaotic regime. We also investigated the relationship between patterns of output time intervals and the path selection of droplets.

In a pipe of length L, diameter D and at low RNs, the flux Q is related to the pressure difference of pipe ends, ΔP, via $\frac{\Delta P}{Q} \sim \frac{\mu L}{D^4}$, where μ is the viscosity. A droplet affects the flux by introducing a capillary pressure drop,

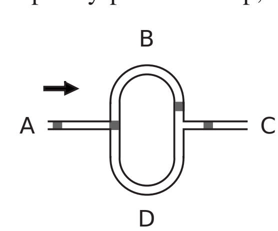

FIG. 1. *ABC* and *ADC* are identical except in their lengths, that is one of variable parameters. We call the system symmetric if the branches are equal and asymmetric otherwise.

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$$\Delta P_{\mathrm{Ca}} \sim \frac{\sigma}{R}\mathrm{Ca}^{2/3}, \tag{1}$$

where σ is the surface tension between the gas and the liquid and $\mathrm{Ca}=\frac{\mu v}{\sigma}$, where Ca is the capillary number [12–14]. Here, v indicates the droplet velocity, which is $v=\frac{Q}{\pi R^2}$; therefore, Eq. (1) is true for gas bubbles in a liquid. Nevertheless, it can be used for small droplets when capillary forces dominate viscosity [11].

Flux conservation on the network nodes results in a system of cubic equations, which is solved numerically. The flux rates of the pipes are preserved as long as the number of droplets in all of the pipes is unchanged. Therefore, we calculate the position of the droplets over time until a new droplet enters a pipe or a moving droplet meets the end of a pipe. We call these new events. When a droplet reaches the bifurcation, it selects the branch that has a stronger flow. For the case of equal flux, we suppose that one of the branches is always more favorable. Then we recalculate the ΔPs and the Qs after any new event and continue in this way.

We studied the effect of the length difference (ΔL) between the branches on the output signal. We also investigated the impact of the time interval of input droplets (ΔT) on the output signal while all other parameters were fixed. We performed the simulation for two different settings. In one setting, ($\Delta L=0$) with a ratio $\frac{L}{D}=40$ (symmetric). In the other, there is a 10% difference in the length of the branches, but the diameters are the same (asymmetric). For both configurations, we studied the time coding of the output signal as a function of the time intervals between entering droplets.

To study the reversibility, we consider two different ways of signal decoding. In both methods, the sequence of the time intervals between the output droplets is saved and used as an input signal in the reversed process. In the first method, which we call in-place decoding, we stopped the coding process and reversed the direction of flow. In the second method, we supposed that we had only the coded signal and a decoding device, identical to the coding device. From the application viewpoint, this method may be more interesting. In contrast with the first method, we call it out-of-place decoding.

In the simulation, droplets entered at a uniform flat rate. Therefore, an input signal is specified by a single parameter, ΔT_i, the time interval between entering droplets. However, in the output, the time interval between the droplets might be different, so the output signal is specified by the sequence, $\Delta T_n=T_n-T_{n-1}$, where n is the number of droplets that have exited.

For large enough ΔT_is, the output pattern is identical to the input pattern. However, by decreasing ΔT_i, we start to get different patterns at the output. This happens if a droplet reaches the entering T-junction while the former droplet has not yet left the loop. Therefore, the droplet at the junction will select the branch that is empty and has a higher flow rate. At this point, we start to get a periodic output pattern (periodic sequence of ΔT_ns) that is different from the uniform input signal. The period length increases as we decrease ΔT_i.

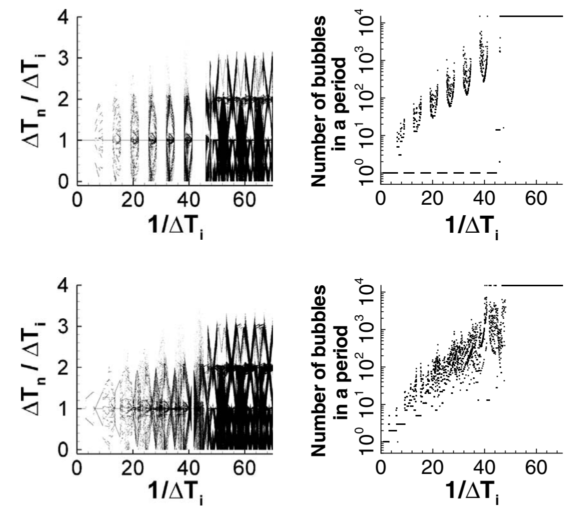

FIG. 2. Output time intervals versus input time interval (top) for symmetric setup (left) and asymmetric setup (right). The period of the output signal as a function of the input time interval is shown below each case.

By decreasing ΔT_i even more, we get disordered patterns with no visible order or periodicity (Fig. 2). In this case, the period length equals the number of droplets involved in the system, which means the system either is aperiodic (chaotic) or has a period that is longer than the simulation time. The same behavior is demonstrated in the work of Fuerstman *et al.* by Poincare maps [7].

Like many other chaotic systems, we observe that the system re-emerges into the chaotic regime at different points for the symmetric and the asymmetric models. There are some intermediate intervals where the coder loop is invisible and the output and input are identical. As shown in Fig. 2, similar behavior has been observed in both symmetric and asymmetric networks, though the figures are different. This indicates that even at low RNs, the system shows chaotic behavior as a result of the history-dependent dynamics of the model. The same has been observed in ecological models [15].

We studied the reversibility (decoding) of the process in both the in-place and the out-of-place methods. In the chaotic regimes, the process is not decodable with either method, the input signal information is lost and it cannot be retrieved. It is possible, however, to restore the original signal when we have periodic outputs. However, it is not always true that all of the processes are reversible when they are not chaotic. This is more surprising for the case of in-place decoding.

The behavior of the system for different input time intervals are in Fig. 3 for the asymmetric setup. In the figure, the patterns of time intervals are shown for before and after the reversion of the flow. First, the flow goes through the network and the output is shown for some droplets (the left part of each graph), then we look at the time interval between these droplets when the flow is reversed (the right side of each graph). The graphs differ only in the time intervals of the input signals. Therefore, as shown in the figure, even for periodic output signals, the initial signal is sometimes recon-

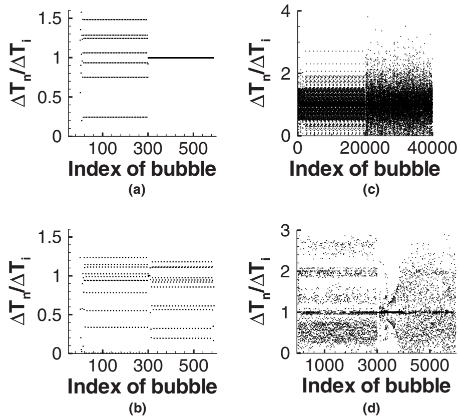

FIG. 3. In-place decoding for the asymmetric model. The ΔTs are shown for before and after the reversion of flow for 300 (a and b), 20 000 (c) and 3000 (d) droplets, in the left and the right side of each graph (see text). For $\Delta T_i=0.1$ (a), there is a regular and periodic coded signal which is decoded perfectly. For $\Delta T_i=\frac{1}{11.45}$ (b), there is a periodic coded output, but the decoded pattern is a different periodic signal. For $\Delta T_i=\frac{1}{42.60}$ (c), the coded signal is periodic, but in reversion, it is chaotic. For $\Delta T_i=\frac{1}{54.00}$ (d), the output pattern is aperiodic, with the values concentrated on some strips. Reversion produces a similar pattern with more fluctuations.

structable. It is also possible that a periodic encoded signal shows chaotic behavior when decoded [Fig. 3(c)]. Similar behavior is observed for the symmetric setup. Here, as we are at low RNs, time reversibility is expected. Again, the history dependence of the process plays its role. A droplet chooses a branch that has a faster flow, but when it leaves the branch, the flows might have been changed. As a result, when we reverse flow, the droplet that has just left a branch might go into the other branch.

Looking at Fig. 3, one can ask whether the decoded signals are a stationary solution. These figures have been selected for demonstration because the solution is converged upon quickly. Thus, we do in-place decoding for the whole range of ΔT_i with appropriate wait time for both the encoding and decoding processes. The map of encoded signals for both symmetric and asymmetric models has been previously shown in Fig. 2. The corresponding decoded signals are in Fig. 4. For both symmetric and asymmetric models, there are some areas in which the signals are decodeable.

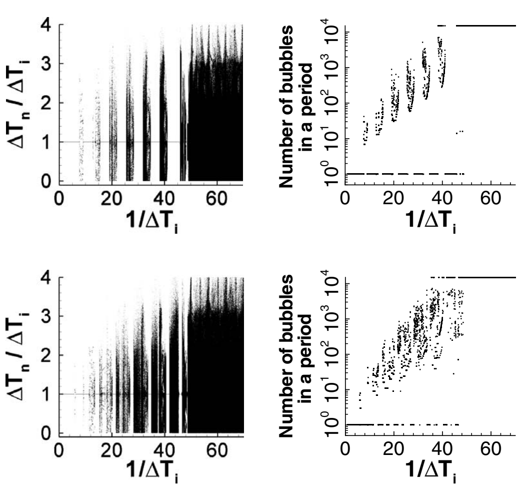

FIG. 4. Time intervals between the droplets of in-placed decoded signals as a function of uniform time intervals between the droplets in the original signal (top) and the period of the output signal as a function of the input ΔT (bottom). For both symmetric (left) and asymmetric (right) setups, there are some regions where the signal is decoded successfully. The corresponding encoded signals are in Fig. 2.

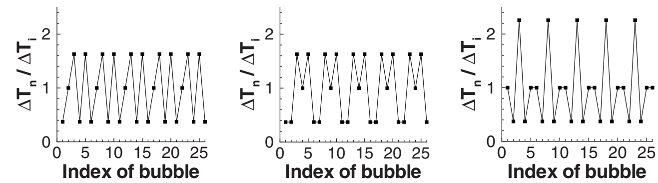

FIG. 5. Out-of-place decoding: a periodic output signal (left) from the asymmetric setup for $\Delta T_i=\frac{1}{15}$ has a period of 5. The output depends on the phase of the signal in the decoding process if we start from droplets 1 (middle) or 4 (bottom). If we start with the others (the numbers 2, 3, or 5), the original uniform signal is reconstructed.

In the out-of-place decoding, the signal can again be decoded for some periodic signals. In periodic signals, there is an additional degree of freedom: the phase, which is dependent upon which droplet should go first when we feed a periodic signal into an empty device. We observed that, for the periodic signals, the result is sensitive to this phase. In a periodic pattern, if we start decoding from different phases, we might get different signals. The resulting signals are again periodic and the original signal may be among them (Fig. 5). Figure 5(a) shows a periodic signal that is encoded by an asymmetric setup with a flow of droplets with $\Delta T_i=\frac{1}{15}$. The numbers in the figure indicate the phase. As the signal has a period of 5, we can feed this sequence into an empty decoding device starting with any of these droplets. The figure shows that different patterns are found if we start the decoding process with droplets numbered 1 and 4, while the original sequence is constructed if we start from the other three droplets. Similar features are observed for symmetric model; however, the results of only the asymmetric model are presented here.

We can go further by looking at the intermediate pathway and studying the patterns of pipe sections. We can follow the train of the droplets and see which branch they are taking. We call this up ($\uparrow$) or down ($\downarrow$). This gives us another way of decoding a signal. This was introduced by Jousse *et al.* [11]. We look at the signal in our asymmetric model. We suppose that the shorter branch is labeled $\uparrow$. If we count the number of droplets passing through "up" and "down" branches call them by $n_\uparrow$ and $n_\downarrow$, respectively, then $p=\frac{n_\uparrow}{n_\uparrow+n_\downarrow}$ and $q=\frac{n_\downarrow}{n_\uparrow+n_\downarrow}$ are the probabilities of a bubble passing through "up" or "down" branches, respectively. We expect, in the long run, that more droplets pass through the shorter branch ($\uparrow$) than the longer one ($\downarrow$). So, $1\geq r=p-q\geq 0$. Here r not only is related to the pipe lengths, but also varies with ΔT_i. For a large enough ΔT_i, when any droplet reaches an empty loop, all droplets pass through the shorter branch and $r=1$. After decreasing ΔT_i the droplets alternate between the "up"

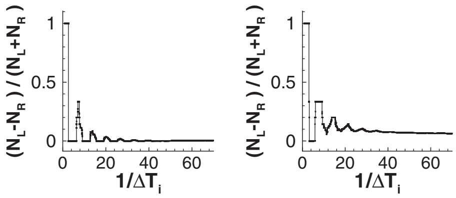

FIG. 6. The ratio of the difference between the number of droplets passing through pipes versus the input time interval, for the symmetric (left) and the asymmetric (right) setups.

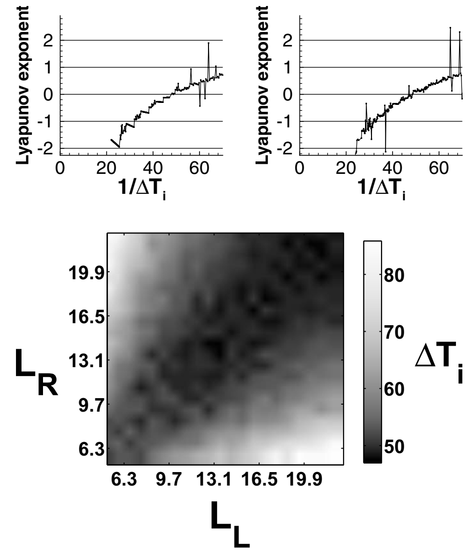

FIG. 7. top-λ versus input time interval for symmetric (left) and asymmetric (right) setups. down-X and Y axis are the lengths of the branches. The color represents the input time interval where λ changes sign and the system becomes chaotic.

and "down" branches. At this point, $r=0$. It should be noted that, the up-down signal is alternating but the output interval is still uniform and identical to the input signal. Thus, if the up-down signal is periodic, the sequence of time intervals is also periodic, but they do not need to have the same period. As we have considered only a very small difference between the pipe lengths, for any periodic up-down signal and in any period, the difference between the numbers of ↑ and ↓ is 0 or 1. This means that for any signal with period P, r is either 0 or $\frac{1}{P}$, for even and odd P numbers, respectively (Fig. 6).

To quantify the chaotic behavior, we calculated the Lyapunov exponent λ. We changed the distance between an incoming droplet and the next droplet by a factor of 10^{-3} and then watched the output ΔTs. Now, we can compare the perturbed system with the original system and determine λ: $\delta_n = \Delta T_n - \Delta \acute{T}_n$ Here $\Delta \acute{T}_n$ is the original output time interval at step n and $\Delta \acute{T}_n$ is the perturbed output time intervals. The difference δ_n at step n depends on time, which is denoted by n here. As time passes, δ_n can diverge or converge, which will tell whether the system is chaotic or not. The time dependence of δ_n should read as $\delta_n = e^{(t_n - t_0)\lambda}\delta_0$, where δ_0 is the perturbation. In Fig. 7, one can see that λ depends upon the input time interval, $\lambda > 0$ means that δ_n diverges, so the system is chaotic. The phase diagram (Fig. 7) shows the input time interval at which λ changes its sign, depending on the size of the branches.

We used a microfluidic network and a set of deterministic equations to simulate the passage of droplets in the network. The fluid had a constant flow, which carried the droplets. The time interval between successive droplets was constant at the input. We found that patterns of droplets at the output depend upon the time interval between the input droplets. We observed that, in general, we have both periodic and chaotic patterns of output, depending on the rate of entering droplets. This observation agrees with experiments by Fuerstman *et al.* [7].

We found that, although deterministic, the system is not always reversible. We reversed the time in simulations and found that in some situations, the process is irreversible and droplets choose the other branch when returning. This also depends upon the input time interval of the droplets. We observed that there is a relationship between irreversibility and chaos in our system. In general, when the output signal is periodic, it may be decoded to the original input signal just by reversing the flow direction, similar to the experiments of Fuerstman *et al.* [7]. When it is chaotic, however, it is not possible to generate the input signal (decode). Finally, by calculating the Lyapunov exponent, we generated the phase diagram of both periodic and chaotic systems (Fig. 7). This phase diagram is useful if we were to design a device that could encode a signal, like the work of [7]. Given the signal properties, this diagram helps to set the system dimensions in a way that guarantees reversibility.

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