Directional droplet transport on switchable ratchets by mechanowetting

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Abstract
Materials with a mechanical response to an external stimulus are promising for application in miniaturized cargo and fluid manipulation in microfluidic (lab-on-a-chip) systems and microsystems in general. One of the main challenges in droplet microfluidics is the precise control of the droplet motion, and existing technologies have drawbacks that can compromise the droplet contents. Here, we demonstrate how an on–off switchable ratchet topography combined with a simple actuation strategy can be exploited to accurately manipulate mm-sized droplets. Because of the mechanowetting principle, the three-phase line dynamically attaches to these deforming ratchets, affecting the droplet displacement in a controlled matter. We show that such topographies are capable of transporting droplets over a surface in a stepwise fashion. We calculate the forces generated by the surface using both a theoretical description of the three-phase line and fluid simulations, and we identify the window of applicability in terms of the droplet size relative to the sawtooth dimensions. Our results enable the design of microfluidic systems with deforming wall topographies for controlled droplet manipulation.

Keywords Droplets · Mechanowetting · Fluid dynamics

1 Introduction

Fluid manipulation at small length scales in microfluidic systems, such as a lab-on-a-chip biosensors, has enabled the decrease of the processing time of biochemical analyses by orders of magnitude, while reducing the processing costs and time compared to traditional processes (Whitesides 2006; Culbertson et al. 2014). One major challenge in the field of microfluidics is to transport fluids with high precision, which can be achieved by the downscaling of
conventional pumping systems (Laser and Santiago 2004), mimicking mucus transport out of the lungs using artificial cilia (Khaderi et al. 2011), manipulation of the surface-free energies in multi-fluid systems by means of electrowetting (Lin et al. 2012), structural design of topological liquid diodes (Li et al. 2017) and more (Zeng et al. 2002; Roy et al. 2009; Samiei et al. 2016; Malinowski et al. 2020). However, these techniques can have serious drawbacks, such as the non-uniform (parabolic) flow profile in conventional pumping and unwanted byproducts of electrochemical reactions in the case of electrowetting. Mechanical solutions to overcome this challenge have been presented, for example by tuning the droplet mobility by stretching microtextured substrates (Mazaltarim et al. 2021) and vibration of asymmetric microstructures (Tretjakov and Müller 2014). Here, we use mechanowetting, i.e., the control of three-phase line motion of two-fluid systems (e.g., drops, slugs) by mechanically deforming surfaces. In previous work, we have shown that mechanowetting can be used to transport fluid slugs in channels (de Jong et al. 2020; De Jong et al. 2021), as well as individual droplets on surfaces, by creating mechanical traveling surface waves (de Jong et al. 2019; De Jong et al. 2021).

The introduction of stimuli-responsive materials such as azobenzene-enhanced polymers (White and Broer 2015), magnetic artificial cilia (Khaderi et al. 2009; den Toonder and Onck 2013), responsive hydrogels (Kim et al. 2011) and nanoporous materials with very high surface-to-volume ratio (Detsi et al. 2011) has opened exciting new pathways to various applications, such as small cargo transport (Nistor et al. 2016), soft robotics (Huang et al. 2015; Li et al. 2016; Diller et al. 2014), magnetically driven fluid transport and swimmers (Khaderi et al. 2009; Namdeo 2014), high-precision electrochemical actuators (Hai-Jun and Jörg 2010) and haptic feedback for Braille-type displays (Camargo et al. 2012). However, it is very challenging to create traveling surface waves through coordinated action to break symmetry (Lv et al. 2016; Gelebart et al. 2017). Alternatively, surfaces with dedicated, individually addressable, (on–off) switchable features are significantly easier to make, because it would only require a locally applied external trigger, such as (laser) light. Switchable surfaces with good temporal and spatial control can also be achieved by combining static and dynamic structures into a hybrid system, e.g., by filling the voids between static ridges with a responsive compound (Lv et al. 2013).

In this paper, we present a method of directional droplet manipulation by exploiting the local, selective actuation of a simple sawtooth surface structure. By adapting the spatial design to the typical droplet size, we demonstrate how a conceptually simple surface structure with a localized on–off switching mechanism is able to generate capillary forces in the desired transporting direction. We propose several sawtooth (ratcheting) actuation sequences and identify the different transporting regimes by calculating the forces corresponding to each step in the process. To this end, we performed theoretical calculations based on the three-phase line interaction with the switching surface, as well as a series of three-dimensional, two-phase computational fluid dynamics (CFD) simulations with deforming boundary conditions to accurately study the capillary droplet interaction with the switchable solid topography.

## 2 Dimensional analysis

To characterize the switchable ratchet flow, we normalize the parameters of the system as follows. The (mean) droplet velocity $U$ is normalized with $\lambda f$ in the Strouhal number, i.e., $St = \lambda f / U$, where $\lambda$ is the width of one sawtooth and $f$ is the on–off switching frequency (i.e., in the time period $\Delta t = 1/f$, one sawtooth has been fully actuated and retracted). The relative velocities are defined through $St^{-1}$ as the number of sawtooth lengths traveled by the droplet in one sawtooth actuation cycle. Three-phase line motion is quantified by the Capillary number, $Ca = U \mu / \gamma_v$, and the Reynolds number $Re = \rho Ud / \mu$ determines the inertial flow regime, where $\rho$ is the liquid density, $d$ the droplet size and $\mu$ the dynamic viscosity. The Young equation (Young 1805), i.e., $\cos \theta_v = (\gamma_{sl} - \gamma_{sv}) / \gamma_v$, where $\theta_v$ is the Young contact angle, non-dimensionalizes the solid–liquid, solid–vapor, and liquid–vapor surface free energies $\gamma_{sl}$, $\gamma_{sv}$ and $\gamma_v$, respectively. The relative time is incorporated by the phase parameter $\varphi = ft$. In addition to these well-known dimensionless parameters, we distinguish the size ratios $d / \lambda$ and $A / \lambda$, where diameter $d$ is the droplet base length, and the amplitude $A$ is the maximum sawtooth height. In addition to the Strouhal number, we introduce the relative center-of-mass displacement $x_{cm} / \lambda$, where $x_{cm}$ is the center-of-mass location of the drop. The performance of the switchable ratchet will be expressed in terms of the dimensionless force, i.e., $F / \gamma_v$, where $F$ is (the $x$ component of) the capillary force working on the droplet, which is generated by the interaction of the droplet with the switchable sawtooth.

## 3 Theoretical model

To study the fundamentals of the switchable ratchet mechanism, the theoretical model from de Jong et al. (2019) was adopted to account for the capillary forces generated by the switching sawtooth topography. We consider a substrate with a periodic distribution of equal-sized sawtooths of width $\lambda$. The height function of a one-dimensional, piecewise-linear sawtooth located between $x = 0$ and $x = \lambda$ can be described by
Here, $0 < \lambda_r < 1$ is the relative sawtooth size that defines the location of the peak of the sawtooth relative to the full sawtooth size (i.e., $h(\lambda \lambda_r) = A$). The relative sawtooth size is set to $\lambda_r = 0.75$, unless stated otherwise. The sawtooth profile in Eq. 1 is plotted in Fig. 1a. For the theoretical model, the deformation of the surface is modeled by instantaneously deforming the surface to its final shape. Starting from a flat surface, the sawtooth of amplitude $A$ is actuated, which, at every point of the three-phase line, induces a distortion of the contact angle, i.e., $\theta_Y \rightarrow \theta_Y + \Delta \theta$ (see the Supplementary Information), with

$$h(x) = \begin{cases} 
0 & \frac{x}{\lambda} < 0 \\
\frac{\lambda}{\lambda_r} \left(1 - \frac{x}{\lambda} \right) & 0 \leq \frac{x}{\lambda} \leq \lambda_r \\
\frac{A}{1-\lambda_r} \left(1 - \frac{x}{\lambda} \right) & \lambda_r \leq \frac{x}{\lambda} \leq 1 \\
\frac{x}{\lambda} & \frac{x}{\lambda} > 1 
\end{cases}$$

(1)

Here, $n$ is the outward-pointing normal vector of the three-phase line in the horizontal plane. When this perturbation is applied to the Young Equation, a restoring tension $f$ (force per unit length) is generated, which can be written as

$$f = [\gamma_Y \sin \theta_Y \Delta \theta] n.$$

(3)

When integrated over the three-phase line of the drop, we obtain an expression for the capillary force that works on the droplet, i.e.,

$$F = \int_{TPL} f \, ds,$$

(4)

where $ds$ is an infinitesimal part of the three-phase line. Equation 4 provides a net force measure that quantifies the performance of the switchable sawtooth geometries. For small deformations, the droplet can be approximated as a spherical cap, so that the three-phase line can be approximated as a circle, which allows us to rewrite Eq. 4 as

$$\frac{F}{\gamma_Y} = \frac{\sin \theta_Y}{2} \int_0^{2\pi} n_c \arctan[n_c \cdot \nabla h] \, d\phi.$$

(5)

where $n_c = [\cos \phi, \sin \phi]$ is the outward-pointing normal of the circular three-phase line and $\phi$ is the angular part in the cylindrical coordinate system. Note that the surface tension enters in the description of the force directly through Eq. 5 and indirectly through the dependence of $\theta_Y$ on $\gamma_Y$. For surfaces with several simultaneously active sawtooths, the theory can trivially be extended by summing multiple, phase-shifted instances of Eq. 1.

### 4 Numerical model

We performed a series of CFD simulations using a two-phase CFD model, which was developed within the OpenFOAM framework Weller et al. (1998). Here, we solve the Navier–Stokes equations for momentum and the continuity equation, complemented by the smoothed continuous-surface-force formulation of the volume-of-fluid method (SCSF-VOF). For the fluids labeled 1 and 2 with their respective densities $\rho_1, \rho_2$ and kinematic viscosities $\nu_1, \nu_2$, the fluid velocity $u$ and pressure $p$ are calculated by solving the coupled momentum, continuity and phase evolution equations, i.e. Hirt and Nichols (1981), Brackbill et al. (1992), Raeini et al. (2012).
\[
\frac{\partial \rho u}{\partial t} + \nabla \cdot (\rho u u) = -\nabla p + \rho g \\
+ \nabla \cdot [v(\nabla u + (\nabla u)^T)] \\
+ \rho \gamma h \kappa \nabla \alpha, \\
\]

where \( g \) is the body force vector (accounting for the gravity forces), \( \gamma_h \), the liquid–vapor surface free energy, \( \kappa = \nabla \cdot (\nabla a / |\nabla a|) \) the interface curvature, \( 0 \leq \alpha \leq 1 \) the indicator function, which is 1 for fluid 1 and 0 for fluid 2, in this paper set to water and air, respectively. The corrective indicator function, which is 1 for fluid 1 and 0 for fluid 2, in this paper set to water and air, respectively. The corrective

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho u) = 0,
\]

\[
\frac{\partial \alpha}{\partial t} + \nabla \cdot (u \alpha) = -\nabla \cdot (u_0 \alpha(1 - \alpha)),
\]

where \( a \) is the switchable nature of the sawtooth is captured within the definition of the amplitude in Eq. 1, i.e., \( A \rightarrow A(t) \), which is set to \( A(t) = A(1 - \cos(2\pi \phi)) / 2 \), where \( 0 \leq \phi \leq 1 \). The different cells are actuated in an alternating fashion to facilitate the droplet movement and the time dependence is shown in Fig. 1a,b. Here, the actuation sequence notation \( \{+n, \pm m\} \) denotes the actuation strategy consisting of two steps that are repeated. In the first step the sawtooth that is located \( n \) unit cells (each unit cell consists of a single sawtooth) next to the previous cell is actuated (e.g., the +2 step in Fig. 1a). Then, in the next step a different sawtooth is actuated, shifted by \( \pm m \) unit cells (e.g., the -1 step in Fig. 1a), completing the \( \{+2, -1\} \) sequence.

### 5 Results and discussion

By applying the theory to the switching sawtooth topography, we can distinguish between two main modes of transportation, a caterpillar-type mode of actuation (i) and the more straightforward, sequential sawtooth actuation (ii). The caterpillar mode consists of two steps, where we actuate the sawtooth near the advancing and receding line in an alternating fashion, which comes down to repeating phase shifts of \( \pm n \) (Fig. 2a, I → II) and \( -(n - 1) \) (Fig. 2a, II → III), where \( n \geq 2 \) is an integer number. This is denoted by the actuation sequence \( \{n, -(n - 1)\} \). The first panels of Fig. 2a, b show the initial equilibrium position. The sawtooth between \( x / \lambda = -1 \) and 0 retracts and the sawtooth between 1 and 2 is actuated (i.e., the \( +n \) step, with \( n = 2 \)), which is indicated by the variation in color intensity as used in Fig. 1a. This generates a force in the positive \( x \) direction (indicated by the red arrow in Fig. 2b), leading to the droplet being pulled to reach the equilibrium state in panel III (in this configuration there are three-phase line tensions, but they all cancel out leading to an integrated zero net force). Now, when the previous sawtooth has been retracted and the sawtooth between 0 and 1 is actuated (i.e., the \( -(n - 1) \) step), again a force in the positive \( x \) direction is generated (see panel IV). This pushing force causes the droplet to move over the sawtooth and relocate to the state similar to panel I, but shifted by \( \lambda \). This means that, after two sawtooth actuations, we have transported the droplet by the amount \( \lambda \) by sequentially pulling and pushing, effectively generating a relative velocity of \( U / \lambda f = 0.5 \).

Conceptually, the sequential mode is simpler than the caterpillar mode, as it only consists of a single unit step. In Fig. 2c, we demonstrate one cycle for \( d / \lambda = 3.0 \). Here, the actuation sequence is \( \{+m, +m\} \), with \( m = 1 \), which means that this mode of transport solely relies on pushing of the droplet over the corrugations. Starting from the equilibrium position where the receding part of the droplet is connected to the sawtooth (panel I), we deactivate the original sawtooth and actuate the next one, i.e., the +1 step in panel II. The newly formed corrugation generates a force in the +\( x \) direction, which will push the droplet to a state similar to panel I, shifted by the amount \( \lambda \), corresponding to the relative velocity \( U / \lambda f = 1.0 \). The same arguments hold for Fig. 2d, where the actuation sequence \( \{+2, +2\} \) is visualized for \( d / \lambda = 4.5 \). Here, the total droplet displacement is \( 2\lambda \), i.e., \( U / \lambda f = 2.0 \).

The relative velocities (i.e., the number of sawtooth lengths traveled per sawtooth actuation cycle) of the caterpillar and sequential modes are shown in Fig. 3a. The caterpillar \( \{+2, -1\} \) mode shows a relative velocity of
The sequential $\{+1,+1\}$ mode in Fig. 3a shows a larger relative velocity of $U/\lambda f = 1$. It is found that the sequential mode is constraint to the droplet size compared to the sawtooth width, $d/\lambda$. In order for the droplet to be displaced, its geometric center needs to be positioned such that it is in front of the actuated sawtooth, suggesting that the droplet size needs to be at least twice the size of the sawtooth. The caterpillar mode generates a relative velocity lower than that of the sequential mode, as indicated by $U/\lambda f$. On the other hand, the caterpillar $\{+2,-1\}$ mode also works at smaller $d/\lambda$, allowing for smaller droplet sizes to be transported (for a given sawtooth size) or for larger sawtooths (for a given droplet size).

In Fig. 3b, the forces acting on the droplet of the different modes are plotted. In order to reach the maximum acceleration, the largest possible force needs to be generated. This force depends on the location of the droplet with respect to the sawtooth. So, rather than the maximum forces, we analyze the performance by plotting the force at the moment that the sawtooth is actuated. This is done because here, the most important aspect is the onset of the drop motion. Once the motion is initiated, the droplet will move in the direction of the force to the closest equilibrium point. From Fig. 3b,
it can be seen that the initial forces for the caterpillar mode (red graphs) are much larger than for the sequential mode (blue graph). This difference in forces is also apparent from the size of the red arrows in Fig. 2b–IV (caterpillar) and c–II (sequential). This highlights that the caterpillar mode is more robust compared to the sequential mode.

The CFD simulations are different from the theoretical approach since the sawtooths are switched at frequency \( f \), in contrast with the quasi-static approach used in the theory. Additionally, the theory does not take into account droplet deformation caused by surface deformation at (non-zero) amplitude \( A \). In Fig. 4a–c, two simulations are detailed, demonstrating transport for the \{+2, −1\} and \{+1, +1\} modes. The simulation snapshots (top view) in Fig. 4b, c are very close to those theoretically predicted in Fig. 2b, c. We simulated a large range of \( d/\lambda \) values and plotted the results alongside the theory in Fig. 3a. It is shown that for the caterpillar mode \{+2, −1\} transport for \( 1.43 \leq d/\lambda \leq 2.12 \) is confirmed by the CFD model, see Fig. 3a (red squares).

For the \{+1, +1\} mode, the droplet is picked up earlier in the CFD simulations than the theory predicts. This observation can be explained by two phenomena. First, the three-phase line (and consequently the droplet) deforms when subjected to a topography change, which means that it will deviate from a spherical-cap shape. Second, when the sawtooth is actuated, fluid in the bulk of the droplet is displaced.

This means that effectively, the droplet footprint increases in size during the process of actuation, and contracts when de-actuating the surface. Both phenomena give rise to an equilibrium position that is slightly different from the position predicted by the theory. Where the CFD simulations already show droplet movement at \( d/\lambda > 1.88 \), the theory predicts motion only at \( d/\lambda > 2 \) which can be seen in Fig. 3a. In the \{+2, −1\} mode, the droplet is not transported for \( d/\lambda > 2.12 \) in the CFD model, while the theory predicts transport up to \( d/\lambda = 2.38 \).

The CFD model allows us to study dynamic effects by increasing the frequency to \( 4 \leq f \leq 100 \) Hz. To this end, we perform a series of simulations in a domain with periodic boundary conditions, such that multiple instances of the \{+2, −1\} mode are actuated, defining different cells. These cells are separated by (at least) two sawtooth widths \( \lambda \) each. This shift is necessary to ensure that the droplet of interest is not affected by an actuated sawtooth in a neighboring...
cell. In Fig. 5a, the relative transporting speed response for $d/\lambda = 1.65$ is plotted as a function of the frequency for the caterpillar mode. Near $f = 33$ Hz, it is observed that the traveling direction is reversed which is due to the fact that the actuation frequency is equal to a resonance frequency. While the sawtooth is retracting, the horizontal velocity of the droplet becomes negative as a consequence of the impulse that was given to the drop during actuation, i.e., the observed reversal is the resonance of the droplet at the actuation frequency. The net effect is that the droplet is propelled in the negative direction, which, moments later, causes it to attach to a sawtooth in a different cell location upstream. At frequencies higher than 85 Hz, the transport mechanism breaks down, which is caused by the relatively slow settling time in the pulling step. Here, the driving force is not strong enough to overcome the viscous forces. When the actuation cycle of the pulling sawtooth is finished, the droplet has not settled in the right location (i.e., the position shown in Fig. 2b-III) to picked up by the next sawtooth.

To determine the effect of the surface and fluid parameters, simplifying assumptions can be made for Eq. 4 and Eq. 1. For sufficiently small values of $A/\lambda$ ($< 0.1$) and considering that the horizontal component gradient of the (piecewise linear) height function $\partial h/\partial x \propto A/\lambda$, the force is proportional to the amplitude De Jong et al. (2021), i.e.,

$$\frac{F_x}{\gamma_d d} \propto \frac{A}{\lambda}. \quad (9)$$

For larger values of $A/\lambda$ the deformation under the droplet becomes so large, that the direct fluid displacement due to the activation of the sawtooth (i.e., $\sim 0.5 \lambda d$) becomes significant, so that the actuation force no longer scales linearly with the amplitude. Additionally, the dependence of the forces on the contact angle (and the surface tension) follows directly from Eq. 4, i.e.,

$$\frac{F_x}{\gamma_d d} \propto \sin \theta_y. \quad (10)$$

This means that, when keeping $d$ constant, the dimensionless force is strongest at $\theta_y = 90^\circ$. This dependence is similar to that in our previous works de Jong et al. (2020), De Jong et al. (2021), de Jong et al. (2019).

The presented transporting method does not require sawtooth structures with $\lambda_r = 0.75$, or even asymmetrically shaped corrugations in general. It can also be done with a symmetric bump (e.g., Eq. 1 with $\lambda_r = 0.5$). Figure 5b–d show that the asymmetry of the used corrugation, combined with the appropriate choice of actuation sequence (see also Fig. 3a), enhances the range of relative droplet sizes at which crosses are CFD results where no transport was measured and the dots are the CFD results where transport at $U/\lambda f = 1$ was observed. c Pushing force for $\{+2, -1\}$, calculated using the theoretical model (i.e., $f \to 0$), $\theta_y = 90^\circ$. Same as (b), except for the dots, which represent CFD results where $U/\lambda f = 0.5$. The dashed triangle represents an area that can be used for optimization (see the main text). d The same as (c), for the pulling force in the $\{+2, -1\}$ sequence.

Fig. 5 Various dependencies of the sawtooth droplet transporting efficiency. a Frequency dependence of the relative transport velocity for the $\{+2, -1\}$ mode, showing direction reversal near $f = 33$ Hz and transport breakdown for $f > 85$ Hz. Here, $d/\lambda = 1.65$ and $\theta_y = 90^\circ$. b Pushing force for the $\{+1, +1\}$ sequence, $\theta_y = 90^\circ$. Here, the background color represents the theoretical forces generated by the sawtooth topographies corresponding to the legend, the red line is the theoretical transporting limit, i.e., outside of this line, $F = 0$, the red
droplets can be transported. For example, values of $\lambda_r < 0.5$ are more suitable for transport in the $-x$ direction, when combined with a different actuation sequence (e.g., $\{-2, +1\}$ or $\{-1, -1\}$). Moreover, when $\lambda_r < 0.5$, transport for both types of actuation sequences can also be realized, albeit with smaller net forces, which is also shown in Fig. 5b for the $\{+1, +1\}$ sequence and in Fig. 5c, d for $\{+2, -1\}$. Note that in Fig. 5b–d, the shown range deliberately excludes $\lambda_r = 1$ and 0, since those values represent singularities in $\nabla h$ when Eq. 1 is used as height function.

The sawtooth surfaces that were analyzed in this paper can be generated in an experimental setting in multiple ways. For example, by using switchable materials such as (micro-)structures embedded in) hydrogels (Sidorenko et al. 2008; Kim et al. 2011) or azobenzene-embedded liquid crystal polymers (White and Broer 2015; Liu et al. 2015; Liu and Broer 2015), one can create topographical features capable of switching between an “on” and “off” state, by applying the appropriate stimulus. For example, the azobenzene-embedded liquid crystal polymers can be made responsive to polarized UV light by controlling the orientation of the embedded liquid crystal polymers (Liu et al. 2015; Liu and Kim et al. 2011) or azobenzene-embedded liquid crystal structures embedded in) hydrogels (Sidorenko et al. 2008; White and Broer 2015). However, these advantages come at the cost of tighter limitations in droplet sizes.

6 Conclusions

In this paper, the effects of a switchable sawtooth topography were investigated using both theoretical and numerical approaches. By adapting the sawtooth size $\lambda$ to the droplet size $d$, we identified the various transport regimes for two actuation sequences, i.e., the caterpillar ($\{+n, -(n-1)\}$) and sequential ($\{+m, +m\}$) modes. We found that the conceptually simpler sequential mode is able to generate higher relative velocities and is able to propel a larger range of relative droplet sizes, while the caterpillar mode is able to generate higher forces and offers therefore a more robust way of droplet transport. We anticipate that our findings and results will open new avenues towards designing digital microfluidic systems, in which the capillary interaction between droplets and deforming surfaces can be exploited.

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Author Contributions EDJ designed and performed the simulations, and PRO wrote the paper. All authors discussed the results.

Declarations

Conflict of interest The authors declare that they have no conflict of interest.

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