

# Rechargeable self-assembled droplet microswimmers driven by surface phase transitions

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The design of artificial microswimmers is often inspired by the strategies of natural microorganisms. Many of these creatures exploit the fact that elasticity breaks the time-reversal symmetry of motion at low Reynolds numbers, but this principle has been notably absent from model systems of active, self-propelled microswimmers. Here we introduce a class of microswimmers that spontaneously self-assembles and swims without using external forces, driven instead by surface phase transitions induced by temperature variations. The swimmers are made from alkane droplets dispersed in an aqueous surfactant solution, which start to self-propel on cooling, pushed by rapidly growing thin elastic tails. When heated, the same droplets recharge by retracting their tails, swimming for up to tens of minutes in each cycle. Thermal oscillations of approximately 5 °C induce the swimmers to harness heat from the environment and recharge multiple times. We develop a detailed elasto-hydrodynamic model of these processes and highlight the molecular mechanisms involved. The system offers a convenient platform for examining symmetry breaking in the motion of swimmers exploiting flagellar elasticity. The mild conditions and biocompatible media render these microswimmers potential probes for studying biological propulsion and interactions between artificial and biological swimmers.

ue to their relative simplicity, natural microswimmers<sup>1,2</sup> and their artificial counterparts<sup>3-5</sup> are convenient systems for studying the complex behaviour of active matter. Indeed, intricate nanomachinery governs movement, from cell shaping and division to the propulsion of microorganisms in biology<sup>6</sup>. Developments in molecular machines<sup>3</sup> including nanocars7 foreshadow such life-like complexity in artificial swimmers, but fabrication and integration from molecular to nano- and microscales is non-trivial: 'a major challenge is finding robust ways to couple and integrate the energy-consuming building blocks to the mechanical structure'8. To this end, promising non-biological artificial muscles have already achieved programmable movement<sup>9</sup> and phase-transition-driven two-way elastic deformation<sup>10</sup>. Combinatorial approach methodology to multifunctionality<sup>11</sup> has resulted in self-sensing muscles<sup>12</sup>, and bottom-up synthesis techniques can be used to synthesize single molecules into polymer shapes<sup>13</sup>. A number of mechanisms for artificial swimmers have been used, including prominently chemical power<sup>14</sup>—catalytic particles creating bubbles, self-electrophoresis or releasing slightly dissolving compounds to drive Marangoni flows (for example, camphor boats)<sup>15</sup>. Others are driven by physical effects-thermophoresis<sup>16</sup> or external acoustic<sup>17</sup>, magnetic<sup>18</sup> and electric<sup>19</sup> fields, and they serve as a basis for the theoretical understanding of 'active matter'20.

Three main classes have emerged that are relatively easy to make in large quantities and are therefore accessible for the study of large ensembles of microparticle swarms. The first class—catalytic Janus microswimmers<sup>5</sup>—use catalysts on one side of their surface (for example, Pt or Pd) to decompose hydrogen peroxide in the surrounding solution, creating micro- or nanobubbles, and self-propel while the chemical fuel lasts.

The second major class of artificial microswimmers<sup>21</sup> uses light-absorbing Janus particles to induce local heating and asymmetric demixing in a binary lutidine/water mixture, thus generating spatial concentration gradients that induce self-diffusiophoretic motion of the particles. Both types of system have been extensively studied, but the need of toxic peroxide or lutidine prevents the study of such swimmers interacting with biological microswimmers in their native environment.

The third class of microswimmers are compatible with biological media and exploit external light or magnetic fields to create motion. Differential light absorption on relatively easy to fabricate Janus particles has been used for thermophoresis. Such swimmers have achieved ~50 µm s<sup>-1</sup> speeds with hollow particles, which allow loading with drugs for targeted delivery<sup>22</sup>. More recently, emulsion-based Pickering immobilization was employed to scale up and simplify the fabrication of such metal-containing Janus particles, capable of thermophoretic swimming<sup>23</sup>. Their potential beyond drug delivery also extends to therapies such as thrombosis ablation treatment<sup>24</sup>. Magnetic swimmers require more involved microfabrication of intricate screw-type structures<sup>25</sup>, which, however, can be controlled in deep tissues without restrictions for transparency or complications of light absorption. Therefore, they have excellent properties for the studies of mixed artificial and biological swimmers. Magnetic microrobots picked, encapsulated and delivered cells to

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**Fig. 1 | Emulsion droplets deform on cooling and eventually form dynamic swimmers with one or two fibre-extruding nozzles. a**, Schematic of the transformation of the initial oil drop into a swimmer with one or two tails, rapidly passing via a series of drop-shape shifts. The *T*<sup>1</sup> notation above the arrows represents the cooling of the sample. **b**, Images of swimmer formation observed experimentally on cooling a tetradecane drop. **c**, Microscopy image of tetradecane swimmers extruding one or two fibres with a diameter of  $-0.5 \,\mu$ m; extrusion rate  $U_{\rm f} \approx 6.5 \,\mu$ m s<sup>-1</sup> for drops extruding two fibres and  $U_{\rm f} \approx 12 \,\mu$ m s<sup>-1</sup> for drops extruding a single fibre. The swimmer speed is  $U_{\rm s} \approx 0.25 \,\mu$ m s<sup>-1</sup> for drops extruding two fibres at  $U_{\rm f} \approx 2.3 \pm 0.3 \,\mu$ m s<sup>-1</sup> and swimmer speed  $U_{\rm s} \approx 0.45 \pm 0.06 \,\mu$ m s<sup>-1</sup> (**d**) and swimmers extruding one fibre with  $U_{\rm f} \approx 0.85 \pm 0.10 \,\mu$ m s<sup>-1</sup> and swimmer speed  $U_{\rm s} \approx 0.28 \pm 0.05 \,\mu$ m s<sup>-1</sup> (**e**). In all experiments, the alkane drops are dispersed in 1.5 wt% Brij 58 surfactant solution. Scale bars, 20  $\mu$ m. The quoted values of  $U_{\rm s}$  and  $U_{\rm f}$  are for the specific drops shown in these images. The relation between  $U_{\rm s}$  and  $U_{\rm f}$  is expressed by equation (3). The statistically averaged values of parameter *c* in equation (3) are presented and discussed in the main text.

locations while protecting them from shear forces<sup>26</sup>, while others were used to capture non-motile sperm cells, propel them and fertilize an egg<sup>27</sup>. Since external magnetic fields synchronize all the swimmers' movements and orientation, this class possesses some inherent limitations for studying complexity in active matter.

Nonetheless, a minimal non-living model swimmer, which is easy to generate (for example, by self-assembly) and operates in biologically compatible media, remains an outstanding experimental challenge. This challenge is considerably more difficult if one wishes to assemble a swimmer that uses elasticity for hydrodynamic propulsion, can internally store energy and can be recharged.

Here we present a new class of active microswimmers grown via bottom-up molecular self-assembly using only three simple components: alkane oil drops and water containing a dissolved surfactant. The operating temperature window is tuneable by the choice of oil and surfactant, and small (~5 °C) thermal oscillations in the environment are sufficient to drive and recharge the swimmers, requiring no additional fuel. The experiment requires only an optical microscope with a thermally controlled sample holder. The compatibility of our system with bacteria and higher organisms<sup>28,29</sup> provides an opportunity to study interactions between artificial and biological swimmers, also in populated swarms.

On cooling, the alkane droplets spontaneously eject thin elastic filaments, which—due to viscous friction with the surrounding fluid—push the droplets and induce swimming. On subsequent heating of the environment, the filaments retract completely, thus returning the droplets to their initial state and recharging the system. The internal liquid-to-plastic phase transition that occurs on the surface of the oil drops and drives these phenomena is reversible, while the inherent elasticity of the filaments breaks the time-reversal symmetry of the droplet motion (although the latter occurs at low Reynolds numbers), thus generating partially irreversible swimming motion. Developing a detailed elasto-hydrodynamic model of the filament dynamics, we provide a quantitative insight into the swimming behaviour of the droplets and highlight some similarities with the beating patterns of flagellated swimmers<sup>30</sup>.

As an illustration, we present results obtained with oily drops of alkane (pentadecane or tetradecane), placed in ~1.5 wt% aqueous surfactant solution (Brij 58) that is cooled at a rate of 0.1–  $1.0 \,^{\circ}$ C min<sup>-1</sup>, down to ~8  $^{\circ}$ C for pentadecane (C<sub>15</sub>) or ~2  $^{\circ}$ C for tetradecane (C<sub>14</sub>). The cooling results in initial changes in the drop shape, which quickly reach a steady-state spheroidal shape with four (or five) 'spikes' arranged in the positions of the corners of a tetrahedron (or of a pentagonal pyramid with a tetragonal base) at the particle surface (Fig. 1). One or two of the spikes transform into nozzles that quickly extrude material from the sphere into long filaments with a uniform diameter (Fig. 1 and Supplementary Videos 1–3). We observed that swimmers with a single tail are preferably



**Fig. 2 | Main parameters describing the swimmers shape and motion. a**, Detailed diagram of the spiked ball swimmer, where the effective swimmer radius is *a*; swimmer velocity,  $U_{si}$  fibre extrusion velocity,  $U_{Fi}$  and radius of curvature of the first filament bend,  $R_c$ . The filament radius is *r* and plastic shell thickness,  $\delta$ . **b**, Schematic cross-section of the fibre. In the centre, the fibres are filled with liquid oil, whereas ordered layers of plastic rotator phase are formed on their surface. This rotator phase ensures the fibres' elasticity. **c,d**, Microscopy images of one-tailed swimmers overlaid with several different quantities measured from the experiment. The swimmer shown in **c** is made from tetradecane oil and that in **d** from pentadecane oil. Scale bars, 20 µm.

formed at lower cooling rates (for example,  $0.1 \,^{\circ}\text{C min}^{-1}$ ), whereas the main fraction of swimmers had two tails at a higher cooling rate (~0.5  $\,^{\circ}\text{C min}^{-1}$ ), while drops with one and two tails were observed to coexist in the same sample in the transition range of cooling rates, namely, ~0.2–0.3  $\,^{\circ}\text{C min}^{-1}$ . No notable dependence of the number of extruded tails on the drop size was observed, thus excluding a strong influence of the local interfacial curvature for spikes transformation into fibre-extruding nozzles.

 $C_{15}$  and  $C_{14}$  droplets extruded fibres with notably different diameters, namely,  $d \approx 2.0 \pm 0.2 \,\mu$ m for  $C_{15}$  and  $d \approx 0.5 \pm 0.1 \,\mu$ m for  $C_{14}$ , independently of whether one or two fibres were extruded from a given drop (Fig. 1b,c and Fig. 1d,e show illustrative examples, respectively). For convenience, hereafter, we term the fibres for  $C_{15}$ drops as 'thick' and for  $C_{14}$  drops as 'thin'.

The time for which swimming can be observed in a given system mostly depends on the cooling rate. Depending on the specific oil–surfactant combination, the swimmers are observed in a specific temperature range, for example, 3-4 °C wide for C<sub>15</sub> drops. Performing experiments with different cooling rates, we change the duration of the period in which the emulsion temperature falls within this range. For example, at a cooling rate of 0.50 °C min<sup>-1</sup>, about 5–10 min is available for swimming, whereas this time is ~20 min at a lower cooling rate of 0.15 °C min<sup>-1</sup>.

In our previous studies, quasi-static shapes were obtained on the slow cooling of emulsion droplets<sup>31</sup>. The experiments showed that these shape transformations (artificial morphogenesis) were driven by the formation of a two-dimensional plastic rotator phase at the drop surface, with somewhat different stability in microconfinement than bulk rotator phases<sup>32</sup>, and with thickness between several and dozens of nanometres, depending on the system<sup>31,33</sup>. This plastic rotator phase is formed at the edges of the deformed drops (that is, at the edges of polyhedral shapes, at the periphery of flattened platelets and on the surface of cylindrical protrusions), thus forming a frame of plastic rods that gradually extends with time by incorporating new alkane molecules from the liquid interior of the drops. The molecules in the rotator phase have some rotational freedom and

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occupy a larger volume compared with the truly crystalline phase of the alkanes-this freedom leads to higher molecular mobility and inherent plasticity of the rotator phases. The energy gain on the formation of this plastic rotator phase on the surface of the deforming drops is greater than the energy penalty from the expanding interfacial area, where the interfacial energy was measured to be in the range of 3 to 10 mJ m<sup>-2</sup> (ref. <sup>34</sup>). Molecular dynamics tools to model rotator phase transitions were only recently established<sup>35</sup>, but the main stages of the observed drop-shape evolution sequence were explained with a theoretical model that analysed the energy dependence on the droplet shape, which included both drop surface energy and energy of the forming plastic frame<sup>36,37</sup>. The observed shapes were also interpreted and discussed<sup>38</sup> as a new type of 'tensegrity' (tensorial integrity) structure<sup>39</sup>, which acquire mechanical stability by balancing the compression stress of the interfacial tension with rigidity of the plastic frame forming at the drop surface.

In the new class of oil-surfactant systems described here, the previously observed transformations occur very quickly and the drops rapidly transmute into swimmers (Fig. 1 and Supplementary Videos 1-3). As all the other shape changes are driven by a few layers of plastic phase formed on the surface of the self-shaping drops, we postulate that the elastic filaments have a similar structure. Therefore, the swimmers' actuation is caused by self-assembled thin elastic flagella-like tails (Fig. 2a) with a shell of thickness  $\delta$  composed of alkane molecules ordered in a plastic rotator phase and an interior of liquid alkane (Fig. 2b). Our experiments showed that two main conditions should be simultaneously satisfied to observe swimmers of this type: (1) the surfactant should be with a longer tail than alkane molecules so that the adsorption layers of the surfactant freeze before the drop interior; (2) the cooling rate should be very low, around  $\leq 1$  °C, thus allowing the drops to pass through all the preceding stages to form a swimmer before their complete freezing.

We quantify the relationships between all key parameters describing the behaviour of the observed swimmers. The swimming,  $U_{s}$ , and extrusion,  $U_{\rm P}$  speeds were measured to increase approximately linearly with the cooling rate, which is consistent with the faster formation of the plastic rotator phase on the drop surface (Fig. 3a). Both  $U_{\rm s}$  and  $U_{\rm F}$  are notably higher for single-tailed swimmers compared with two-tailed swimmers at a given cooling rate. This result is also expected, because the rate of plastic phase formation on the drop surface is expected to be the same for a given system and cooling rate, independent of the number of extruded fibres, while this same phase is distributed into one or two fibres in the two types of swimmer. Under equivalent conditions, droplets with a smaller initial diameter extrude fibres at a lower rate compared with bigger droplets, again because bigger drops have a higher surface area from which the rotator phase of the fibres originates. All these experimental trends are quantitatively captured in the model explained below (equation (3)).

To clarify the origin of this new type of microswimmer propulsion, we balance the hydrodynamic Stokes drag on the rapidly extruding cylindrical filaments with the drag created by the friction of the propelled mother drop (approximated as a sphere) with the surrounding fluid. The resulting elasto-hydrodynamic model of the swimmers advances the understanding of elastic fibre extrusion dynamics<sup>40-42</sup> and is described in detail in Supplementary Section II. Briefly, because of the filaments friction with the viscous medium, we estimate the fibre elastic stiffness from the measured periodicity of fibre buckling with characteristic wavelength l (Fig. 2). While past calculations only focussed on hydrodynamic friction and bending of fibres with a fixed basis<sup>40</sup>, our theoretical approach builds on a different numerical scheme<sup>41</sup> and fully characterizes free particle swimming using the extrusion of elastic filament (Fig. 2). The relationship that we derive between the swimming speed of a droplet extruding one or two filaments, U<sub>s</sub>, and the extrusion speed of that filament,  $U_{\rm P}$  is



**Fig. 3** | **Droplet swimming speed. a**, Dependence of swimming speed,  $U_s$ , on cooling rate (filled symbols) and heating rate (empty symbols) for  $C_{15}$  swimmers in 1.5 wt% Brij 58 solution, extruding one fibre (red circles) or two fibres (green triangles). **b**, Relation between swimming speed,  $U_s$ , fibre extrusion speed,  $U_r$ , and drop radius, *a*, for the same experimental data (equation (3)). Note that equation (3) used to construct the plot in **b** is derived only for the extruding fibres—the data with retraction are shown for comparison only. The error bars represent the standard deviations calculated from our data points.

$$U_{\rm S} = c \, U_{\rm F} \, l/a, \tag{1}$$

where *a* is the radius of the main body of the swimmer and *c* is a dimensionless constant. We treat the filaments as a uniform in length material with characteristic (elasto-hydrodynamic) buckling length *l*, which depends on the bending stiffness *A*, extrusion speed and resistance to the flow of filament in the direction parallel to its axis per unit length  $\xi_{\parallel}$  as

$$l = \left( A / \xi_{||} U_{\rm F} \right)^{1/3}.$$
 (2)

The results of our simulations show that the buckling length can be extracted from the radius of curvature  $R_c$  of the first buckle, which is measured with high precision from the experimental observations, as  $l \approx 1.2R_c$  (Fig. 2). Combining equations (1) and (2) with the theoretical estimates<sup>42</sup> for  $\xi_{\parallel}$ , we are able to describe the dynamics of the swimmer using a,  $l_{i}^{T}U_{F}$  and  $U_{S}$  as experimentally accessible quantities, while c and A were determined from equations (1) and (2), respectively. We analysed more than 50 swimmers in total, finding the swimming speed  $U_{\rm s}$  to be rather high, around  $1 \,\mu\text{m}\,\text{s}^{-1}$ , and appears as 10–50% of  $U_{\rm F}$  of thick fibres and 3–5% of  $U_{\rm F}$  of thin fibres. From these experiments, we determined bending stiffness  $A \approx 210 \pm 60 \text{ Nm}^2$  with  $l \approx 25 \pm 5 \mu \text{m}$  for thick fibres and  $A \approx 25 \pm 8 \,\mathrm{Nm^2}$  with  $l \approx 7 \pm 2 \,\mu\mathrm{m}$  for thin fibres. Both one-tailed and two-tailed swimmers are described by this model with the same value of the only material parameter A, a clear indication of the self-consistency of the theoretical approach. This interpretation also showed that the constant c is fairly independent of the materials used and the drop size. Statistically averaging the values of *c* determined for the individual droplets, we obtained  $c \approx 0.142 \pm 0.035$  for swimmers with one filament, which is ~50% higher than  $c \approx 0.093 \pm 0.031$ determined for swimmers extruding two filaments in the same system, probably because the propulsion forces exerted by the two fibres in the two-tailed swimmers do not act in the same direction.

The combination of equations (1) and (2) yields the following relationship, which can be experimentally verified:

$$U_{\rm S} = \frac{c}{a} \left(\frac{A}{\xi_{\parallel}}\right)^{1/3} U_{\rm F}^{2/3}.$$
 (3)

Indeed, the plot of  $U_{\rm S}$  versus  $U_{\rm F}^{2/3}/a$  for swimmers of the same composition, which presumably have the same values of A and  $\xi_{\parallel}$ , gives a straight line (Fig. 3b). Note that the scaling prediction in equation (3) reflects the intricate coupling between the hydrodynamic

propulsion force created by the extruding fibre and its elastic properties, which lead to fibre buckling and thus modulating the propulsion force (Supplementary Section IV.G provides further details).

With our model, we also quantitatively describe the undulations of the orientation angle of the swimmers with respect to their direction of motion (Fig. 4). Using the experimentally available values of a, l and  $U_{\rm p}$  we predict the period and amplitude of the angle oscillations, which agrees very well with the measured data for the last two quantities (Supplementary Table 2 and Supplementary Sections IV.F and IV.I).

The retraction of fibres—on subsequent emulsion heating—is also of high interest, because it defines the reversibility of the process. The retraction is driven by the positive interfacial tension at the oil-water interface, which draws the fibre inwards to minimize the liquid interfacial area. This is unlike fibre extrusion where buckling instability is coupled with elasticity-induced undulations; during retraction, both fibre and droplet are pulled towards a single point on the nozzle, resulting in reduced undulating movement of the droplet-filament system. The quantitative comparison of the data for fibre extrusion and contraction shows that the swimmer speeds during extrusion are higher than those on retraction at the same magnitudes of cooling and heating rates (Fig. 3a). The slope of the relation between  $U_{\rm S}$  and  $U_{\rm F}^{2/3}/a$  is also somewhat lower for retraction in the case of two-tailed swimmers (Fig. 3b). This difference in the swimming speeds on cooling and heating-enabled by fibre elasticity—shows that the swimming is partially reversible. A similar difference is also observed in simulations (Supplementary Videos 4 and 5). Further experiments are needed to more precisely quantify the irreversibility of the observed processes.

The analysis of two-tailed droplets suggests an analogy with biological swimmers. The undulating motion of our swimmers resembles-to some extent-that of several eukaryotic microorganisms that swim by waving their flexible flagella. In particular, the swimmers that extrude two fibres show similarities to the motion exhibited by biflagellate algae, akin to Chlamydomonas reinhardtii (Supplementary Videos 2 and 3). One question for these biological swimmers concerns the mechanism by which their two flagella synchronize. It had long been thought that the synchrony of beating flagella is achieved through hydrodynamic interaction<sup>30,43</sup>, as seen in artificially driven colloidal systems<sup>44</sup>. The flow fields around two single-flagellated algae have also been shown to lead to concerted motion provided the distance of flow-mediated interactions was sufficiently small<sup>45</sup>. This view has been questioned with increasing evidence that intracellular coupling must play a mediatory role in the coordination of beating<sup>46,47</sup>. To test



**Fig. 4 | Kinematics of swimming. a,b**, Microscopy images of pentadecane swimmers dispersed in 1.5 wt% Brij 58 solution, extruding two fibres. Note that the thickness of the extruded fibres decreases with time due to the molecular rearrangement of the alkane molecules in the fibres and the length of the fibres increases remarkably. The fibres are either extruded out-of-phase (Supplementary Video 3) (a) or extruded in-phase (Supplementary Video 2) (b). **c,d**, Dependence of angle  $\Phi$  with time. This angle is defined as the angle formed between the extruding tip, the white dot in the centre of the extruding drop and the vertical axis, as shown in the inset in **d**. For out-of-phase extrusion, the angle oscillates with time (**c**), whereas for in-phase extrusion, it remains almost constant (**d**). Scale bars, 20 µm.

these possibilities with our model system, we studied droplets that were extruding two fibres. By analysing the video records, we observed that the extrusion speeds of the two fibres extruded by a given drop were equal. However, for the various droplets, these pairs of fibres were in different relative phases, which resulted in different ranges of undulation angles  $\Phi$  for the main droplets (Fig. 4 and Supplementary Videos 2 and 3). Unlike the beating patterns observed in Chlamydomonas that exhibit changes in the relative phase between the two flagella, our analysis of 30 droplets swimming by extruding two filaments in 12 videos show that for a given droplet, the phase difference between the extruded filaments remains constant without any sign of synchronization. These results suggest that at distances comparable to the sizes of our droplets, the hydrodynamic interactions and coupling through the body of the droplet are too weak to induce a notable change in the phase difference and fibre synchronization. One can expect that the internal coupling inside the organism most likely plays a key role for similarly sized biological swimmers.

Another analogy with living systems is the emergent ability to harvest energy from changes in the environment (for example, in day/night cycles). When we warm rotator phase fibres (often several millimetres long and only a micrometre wide), they fully retract all the way back to the mother drop (Supplementary Videos 6 and 7). Supplying gentle temperature oscillations of less than 5°C, which do not lead to oil drop freezing, provides enough energy to completely recharge the swimmers in every cooling/heating cycle and let them swim for multiple cycles. Supplementary Video 8 shows a droplet in three consecutive extrusion/retraction cycles. The enthalpy of freezing of hexadecane is  $\Delta H^{\circ} \approx 235 \, \text{kJ kg}^{-1}$  (ref. <sup>48</sup>), and since approximately 75% of this enthalpy (that is, 175 kJ kg<sup>-1</sup>) is due to the liquid-rotator phase transition<sup>48</sup>, the stored potential energy in the particles that could be used for swimming exceeds the maximum energy density of a lead-acid battery, namely, ~140 kJ kg<sup>-1</sup> (40 Wh kg<sup>-1</sup>) (ref. <sup>49</sup>).

In conclusion, we present a new class of active, elastic microswimmers produced by simply cooling a three-component system-oil droplets in an aqueous surfactant solution. The swimmers in this class are not restricted to the specific examples presented and discussed in this Article. We typically observed such active swimmers when surfactants of different types (ionic or non-ionic) have saturated hydrophobic tails that are one to three carbon atoms longer than the alkane molecules and the emulsions are slowly cooled at ~0.1-0.5 °C min<sup>-1</sup>. The temperature interval of the swimming behaviour can be tuned by selecting alkanes with appropriate melting temperature in the drops. Our theoretical model has identified the key parameters governing the motion and may inspire new discoveries in active matter. By coupling buckling instability with filament extrusion, we quantitatively reveal the origin of partial time irreversibility of this mode of swimming at low Reynolds numbers<sup>28</sup> and provide some insights into the motion of living microswimmers. Note that the non-ionic surfactants used in our study are biocompatible and have been applied in various biosystems<sup>50-54</sup>, although the biocompatibility of our systems with real microorganisms has to be investigated in subsequent studies. We highlight the potential for hydrodynamic studies in the area of active matter by referring to mixed systems of artificial and biological microswimmers that can be explored in diluted or in dense populations to reveal collective effects.

#### Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/ s41567-021-01291-3.

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### Methods

**Preparation of samples and observations.** For the preparation of microswimmers, we used emulsions prepared with tetradecane or pentadecane oil dispersed in 1.5 wt% Brij 58 aqueous surfactant solution prepared with deionized water (purified by an Elix 3 module (Millipore)). All the chemical substances were obtained from Sigma-Aldrich. The surfactant was used as received and the alkanes (purity, 99%) were purified from surface-active contamination by passing through a glass column filled with Florisil adsorbent.

Emulsions were prepared using laboratory microkit membrane emulsification module from Shirasu Porous Glass Technology, working with tubular glass membranes with an outer diameter of 10 mm and a working area of approximately  $3 \text{ cm}^2$ . Membranes with mean pore size of 5 and  $10 \,\mu\text{m}$  were used.

For optical observations, a sample of the prepared emulsion was placed in a glass capillary with rectangular cross-sections (width, 1 or 2 mm; height, 100 µm; length, 50 mm) and the capillary was placed into a custom-made cooling chamber connected to a cryo-thermostat (Julabo CF30), allowing the precise control of temperature. To ensure the correct measurement of temperature, a calibrated thermocouple probe was inserted in the next orifice and the temperatures were recorded during the experiments. All the observations were made in transmitted cross-polarized white light. Long-focus objectives (×10, ×20 and ×50) were used to observe the drops on sample cooling. An additional  $\lambda$  plate (compensator plate) was placed between the polarizer and analyser, the latter two being oriented at 90° with respect to each other. The  $\lambda$  plate was oriented at 45° with respect to both analyser and polarizer. Under these conditions, the liquid background and fluid objects have a magenta colour. Observations were performed with an Axio Imager. M2m microscope (Zeiss). The cooling rates applied were varied between 0.05 and 1.00 °C min<sup>-1</sup> and the heating rate, between 0.1 and 3.0 °C min<sup>-1</sup>.

**Procedure for video analysis.** The obtained microscopy images were analysed using ImageJ software to extract data for R,  $R_{\odot}$ ,  $U_{\rm S}$  and  $U_{\rm P}$ . For measurements of  $U_{\rm P}$  the built-in segmented-line command was used and the fibres were manually outlined. The fibre extrusion speed was calculated as the slope of the newly extruded fibre length per unit time (Supplementary Fig. 1). For the measurement of  $U_{\rm S}$ , the MTrackJ plugin was used, which allows an easy tracking of the data versus time (Supplementary Video 9). Due to the complexity of the system, all the measurements were manually performed.

**Reporting Summary.** Further information on research design is available in the Nature Research Reporting Summary linked to this article.

#### Data availability

Source data are provided with this paper. The data that support the findings of this study are available from the corresponding authors upon reasonable request.

#### Code availability

The code used in this study is available from the corresponding authors upon reasonable request.

#### Acknowledgements

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#### Author contributions

D.C. discovered the phenomenon and clarified the experimental conditions under which this new type of swimmer is obtained and can be controlled. D.C., S.T. and N.D. suggested studying the process in more detail. D.C. and S.T. designed the experimental part of the study. S.K.S. designed the part of the study about filament retraction. D.C. performed most of the experiments with respect to fibre extrusion, summarized the obtained results and analysed them (with inputs from S.T., N.D. and S.K.S.), while E.E.L., D.C. and J.C. performed most of experiments for fibre retraction (with input from S.K.S.). E.E.L. clarified the experimental conditions for controlled retraction of the tails. S.K.S. made the first analytical model for swimming by using the estimates of sphere and cylinder drag forces. M.L. and E.L. developed the theoretical description for the extrusion of fibre and motion of droplets. S.K.S., D.C. and M.L. analysed movies and developed insights into relating the dynamic features to the material properties of fibres. M.L. and G.D.C. developed the computer code used in the numerical simulations. S.K.S. and N.D. prepared the initial manuscript draft. D.C. edited the manuscript and prepared the figures and movies. M.L. prepared the theoretical part of the Supporting Information. M.L. and E.L. edited the manuscript. All the authors critically read the manuscript and approved it.

### **Competing interests**

The authors declare no competing interests.

#### Additional information

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**Correspondence and requests for materials** should be addressed to M.L., S.K.S., E.L. or N.D.

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# nature research

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<b>C</b>	<b>.</b>	

## Software and code

olicy information about availability of computer code		
Data collection	The obtained microscopy pictures were analysed using ImageJ software to extract data for R, Rc, US and UF. For measurement of the swimming speed, US, MTrackJ plugin was used	
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# Life sciences study design

All studies must dis	sclose on these points even when the disclosure is negative.	
Sample size	We anaylize more than 50 swimmers	
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Randomization	We allocate them by the size of the drops and number of extruded fibers	
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Describe the research sample (e.g. a group of tagged Passer domesticus, all Stenocereus thurberi within Organ Pipe Cactus National

Research sample	Monument), and provide a rationale for the sample choice. When relevant, describe the organism taxa, source, sex, age range and any manipulations. State what population the sample is meant to represent when applicable. For studies involving existing datasets, describe the data and its source.	
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Imaging type(s)	Specify: functional, structural, diffusion, perfusion.	
Field strength	Specify in Tesla	
Sequence & imaging parameters	Specify the pulse sequence type (gradient echo, spin echo, etc.), imaging type (EPI, spiral, etc.), field of view, matrix size, slice thickness, orientation and TE/TR/flip angle.	
Area of acquisition	State whether a whole brain scan was used OR define the area of acquisition, describing how the region was determined.	
Diffusion MRI Used	Not used	
Parameters Specify # of directions, b-values, whether single shell or multi-shell, and if cardiac gating was used.		
Preprocessing		
Preprocessing software	Provide detail on software version and revision number and on specific parameters (model/functions, brain extraction, segmentation, smoothing kernel size, etc.).	
Normalization	If data were normalized/standardized, describe the approach(es): specify linear or non-linear and define image types used for transformation OR indicate that data were not normalized and explain rationale for lack of normalization.	
Normalization template	Describe the template used for normalization/transformation, specifying subject space or group standardized space (e.g. original Talairach, MNI305, ICBM152) OR indicate that the data were not normalized.	
Noise and artifact removal	Describe your procedure(s) for artifact and structured noise removal, specifying motion parameters, tissue signals and	

Define your software and/or method and criteria for volume censoring, and state the extent of such censoring.

physiological signals (heart rate, respiration).

Volume censoring

## Statistical modeling & inference

Model type and settings Specify type second level	e (mass univariate, multivariate, RSA, predictive, etc.) and describe essential details of the model at the first and els (e.g. fixed, random or mixed effects; drift or auto-correlation).
Effect(s) tested	ise effect in terms of the task or stimulus conditions instead of psychological concepts and indicate whether factorial designs were used.
Specify type of analysis: Whole brain	ROI-based Both
Anatomical loca	tion(s) Describe how anatomical locations were determined (e.g. specify whether automated labeling algorithms or probabilistic atlases were used).
Statistic type for inference (See <u>Eklund et al. 2016</u> )	el-wise or cluster-wise and report all relevant parameters for cluster-wise methods.
Correction Describe th	e type of correction and how it is obtained for multiple comparisons (e.g. FWE, FDR, permutation or Monte Carlo).
Models & analysis         n/a       Involved in the study          Functional and/or effective connectivit          Graph analysis          Multivariate modeling or predictive and	y alysis
Functional and/or effective connectivity	Report the measures of dependence used and the model details (e.g. Pearson correlation, partial correlation, mutual information).
Graph analysis	Report the dependent variable and connectivity measure, specifying weighted graph or binarized graph, subject- or group-level, and the global and/or node summaries used (e.g. clustering coefficient, efficiency, etc.).
Multivariate modeling and predictive analy	sis Specify independent variables, features extraction and dimension reduction, model, training and evaluation metrics.

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