

Supplementary Materials

All of the supplementary movies represent the time evolution (t in simulation units) of a two-dimensional active gel droplet, illustrating different types of movement. The entire system (a square box of side 120 lattice units, with periodic boundary conditions) is represented. The colours refer to the concentration of active material: red is the concentration $\phi = 2$ inside the droplet, and white represents concentration $\phi = 0$ outside; the transition values have been coloured in blue to facilitate the visualisation of the interface. The arrows (or rods – in movies 7 and 8 for the nematic case) represent the average particle orientation inside the droplet.

Supplementary Movie 1: *Rotating Extensile Droplet (normal anchoring)*

For high enough activity ($\zeta = 0.007$ in the system represented) extensile active droplets with imposed normal anchoring spontaneously rotate. First, the droplet elongates in the direction of the polarisation, while remaining static. Then, the droplet deforms to assume a “bean-shape” and starts to rotate (anti-clockwise in the movie). The rotation is powered by two internal bend deformations. These distortions lead to active forces that push the droplet in different directions and cause it to rotate.

Supplementary Movie 2: *Oscillating Extensile Droplet (normal anchoring)*

For intermediate values of the activity ($\zeta = 0.005$ in the system represented) active droplets with imposed normal anchoring exhibit oscillatory motion. After the initial elongated, static state, the droplet starts to rotate intermittently, alternating between clockwise and counter-clockwise movements. When switching between the two senses of rotation, the droplet is maximally elongated and minimally bent.

Supplementary Movie 3: *Stop-and-go Rotating Extensile Droplet (normal anchoring)*

Close to the onset of steady rotation ($\zeta = 0.0054$ in the system represented) droplets rotate always in the same sense but with a stop-and-go type of movement. The shape of the droplet varies, being more elongated and aligned when the rotation slows down and more deformed and bent when the droplet rotates.

Supplementary Movie 4: *Rotating Contractile Droplet (planar anchoring)*

High enough contractile activity ($\zeta = -0.0065$ in the system represented) causes active droplets to spontaneously rotate for an imposed planar anchoring. The droplet starts by elongating in the direction perpendicular to the polarisation, remaining static. Then, the droplet deforms into a “S-shape” and starts to rotate (clockwise in the movie). The rotation is, this time, powered by a pair of internal splay deformations. Just like in the extensile case, the distortions lead to active forces that push the droplet in different directions, causing it to rotate.

Supplementary Movie 5: *Rotating Asymmetric Contractile Droplet (planar anchoring)*

For significantly high values of the contractile activity ($\zeta = -0.0080$ in the system represented) rotating active droplets with imposed planar anchoring exhibit an asymmetric shape and rotate around a point that does not coincide with the center of mass. This is most likely a centrifugal effect caused by the increasing rotational velocity.

Supplementary Movie 6: *“Walking” Extensile Droplet (normal anchoring)*

In the regime of high activity and intermediate anchoring strength ($\zeta = -0.0080$ and $W = -0.03$ in the system represented) extensile droplets with normal imposed anchoring exhibit a hybrid type of movement. This exotic dynamics arises from a combination between translation and rotation. As the droplet alternates motile spells and partial rotations, it gives the impression of moving in small “steps”.

Simulations of nematic systems (used to generate movies 7 and 8) have been run using a slightly different set of parameters from the polar active gel simulations (stated in the main text). For the nematic case, we have used: $a = 0.16$, $k = 0.08$, $\alpha = 0.5$, $\kappa = 0.01$, $\xi = 1.1$, and $W = 0.1, -0.1$ for planar and normal anchoring, respectively.

Supplementary Movie 7: *Rotating Nematic Extensile Droplet (normal anchoring)*

Nematic extensile droplets (the activity parameter value in the system represented is $\zeta = 0.0025$) rotate for an imposed normal anchoring, in an analogous way to polar extensile droplets with rotation powered by a pair of elastic deformations.

Supplementary Movie 8 *Rotating Nematic Contractile Droplet (Planar anchoring)*

The nematic analogue of the polar contractile rotating state is found in nematic contractile droplets ($\zeta = -0.0035$ in the system represented)for an imposed planar anchoring as well.