Comment on “Self-Running Droplet: Emergence of Regular Motion from Nonequilibrium Noise”

In a recent Letter [1] a spontaneous motion of an oil droplet in the surrounding aqueous media along a glass surface was reported. The authors suggested that the self-locomotion is driven by a difference in wettability ($\Delta \gamma_{ad}$) between the front and the rear of a droplet sustained by its translational motion. Whereas we do not question the presented experimental evidences, our comment concerns their interpretation and the underlying physical mechanism of the motion.

We have analyzed movies 2 and 3 (Ref.22 in [1]) in details using LabView software. For every frame of the de-fragmented movies both advanced and receding contact regions of the drop’s contour ($\sim 50$ points) were subtracted and approximated by polynomial functions. The drop contour was then extrapolated to the substrate and the contact angles and instantaneous velocities were determined.

For repetitive motion of a droplet on a narrow horizontal strip (movie 2, Fig.1a), the mean values of the advancing and receding contact angles are $\theta_a = (115.7 \pm 5.4)^\circ$, $\theta_r = (113.5 \pm 3.5)^\circ$, respectively, and $\Delta \theta = (2.2 \pm 6.4)^\circ \sim 0$ that substantially deviates from $11^\circ$ presented in [1].

The instantaneous velocity $U(t)$, exhibits sinusoidal behavior without any appreciable variation in $\Delta \theta$ in time, that is also inconsistent with [1].

The analysis of a droplet motion inside the vertical circle (movie 3, Fig.1b) shows a variation of $\Delta \theta$ vs. the angular position $\phi$ similar to that caused by a static interplay between gravity and capillarity in presence of the contact angle hysteresis [2]. If the motion was driven by $\Delta \gamma_{ad}$ between the front and the rear of the drop, one would expect $\cos \theta_a - \cos \theta_r$ to deviate noticeably from the prediction of [2] (the solid line). [5]

We have successfully reproduced most of the experiments of [1] with a similar system: WT/CTAB(0.45–1.0 mM) and benzonitrile/NaI(8.0–80 mM). We also performed a similar experiment with oil lenses deposited at the air-water interface [6]. The observed motion and the speed (up to $\sim 1$ cm/s) of the self-running lenses were similar to those measured for immersed drops in [1]. Thus, interaction with solid surface is not central for self-locomotion.

We argue that the mechanism of propulsion is that due to chemo-capillary effect. It has been predicted in [3] that Marangoni convection may result in a spontaneous motion of a reactive droplet suspended in a viscous fluid. Experimentally observed velocities of chemo-capillary drift cover a wide range from 0.1 mm/s [4] to 1 cm/s [5]. We found that drop interface consists of domains with low surface tension, $\gamma$. The transport across the w/o interface in the aria of those domains is accompanied by the formation of microemulsion phase inside the drop. [6]. It is well known that $\gamma$, between the microemulsion phase and the co-existing water-rich phase is ultralow, i.e., 3-4 orders of magnitude smaller than the bare oil-water value. Thus, the gradients of $\gamma$ in the vicinity of the domain’s fronts could be enormously large [5], yielding $\sim$cm/s speeds, while in [4] the motion is driven by weak near-equilibrium variation of $\gamma$ with surface concentration of surfactant.

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[1] Y. Sumino et al., Phys. Rev. Lett. 94, 068301 (2005).
[2] Y. I. Frenkel, J. Exptl. Theoret. Phys. 18, 659, (1948); (english translation: xxx.lanl.gov/abs/physics/0503051).
[3] A. A. Golovin and Yu. S. Ryazantsev, Fluid Dynamics 23, 370 (1990).
[4] S. R. Kovsintsev et al., Colloid Journal 63, 318 (2001).
[5] E. Chifu et al., J. Colloid Interface Sci. 93, 140 (1983); S. Bekki et al., J. Colloid Interface Sci. 152, 314 (1992).
[6] A. M. Bellocq et al., J. Colloid Interface Sci. 89, 427 (1982); A. M. Cazabat et al., Adv. Colloid Interface Sci. 16, 175 (1982).
[7] Replacing nitrobenzene (SG~1.2) used in [1] with benzonitrile (SG~1.01) allows deposition of lenses of 160 $\mu$m at the a/w interface.