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# Electrically mediated self-assembly and manipulation of drops at an interface†

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The fluid–fluid interface is a complex environment for a floating object where the statics and dynamics may be governed by capillarity, gravity, inertia, and other external body forces. Yet, the alignment of these forces in intricate ways may result in beautiful pattern formation and self-assembly of these objects, as in the case of crystalline order observed with bubble rafts or colloidal particles. While interfacial self-assembly has been explored widely, controlled manipulation of floating objects, e.g. drops, at the fluid–fluid interface still remains a challenge largely unexplored. In this work, we reveal the self-assembly and manipulation of water drops floating at an oil–air interface. We show that the assembly occurs due to electrostatic interactions between the drops and their environment. We highlight the role of the boundary surrounding the system by showing that even drops with a net zero electric charge can self-assemble under certain conditions. Using experiments and theory, we show that the depth of the oil bath plays an important role in setting the distance between the self-assembled drops. Furthermore, we demonstrate ways to manipulate the drops actively and passively at the interface.

## 1 Introduction

The ability to control and manipulate drops is crucial for many applications including ink-jet printing,<sup>1,2</sup> biological testing,<sup>3,4</sup> cell-based screening,<sup>5</sup> and chemical reactions.<sup>6,7</sup> Among the oldest methods is dielectrophoresis, the motion of uncharged dielectric objects under the application of a non-uniform electric field, which has been used in many contexts to manipulate particles and drops,<sup>8,9</sup> and has even been used to design microfluidic chips that allow for the programmable motion of drops on a surface.<sup>10,11</sup> Electrowetting<sup>12</sup> and electrowetting-on-dielectric<sup>13</sup> are two other strategies that are commonly employed in drop manipulation research and applications.<sup>14–18</sup> Most of the other existing methods focus on manipulating drops on solid substrates using external fields: electric,<sup>19–21</sup> magnetic,<sup>22–24</sup> ultrasonic,<sup>25,26</sup> and light.<sup>27,28</sup> The success of these techniques often relies on the surfaces being superhydrophobic.

While manipulation of drops on liquid surfaces rather than on solid ones offers an exciting alternative, it is challenging due to the dominating capillary forces and the so-called “Cheerios” interactions.<sup>29,30</sup> Previous works have, however, shown the self-assembly and pattern formation of drops at interfaces using vibration of a liquid bath<sup>31–34</sup> and drop evaporation.<sup>35,36</sup> Yet, manipulating the drops or even their self-assembled patterns at the interface remain intractable.

In this article, we present an experimental system with water drops floating at an oil–air interface, where the drops can be manually controlled and/or autonomously self-assembled in ways that depend on the boundary conditions. Furthermore, we show that the self-assembly is programmable, adding a layer of controllability. The two configurations that we present to manipulate drops at interfaces both rely on electrostatic interactions. In the first configuration, we show that the static charges on nearby surfaces can polarize neutrally charged drops and then be used to pattern or move the drops along the interface. The second configuration explores the assembly of triboelectrically charged drops at the oil–air interface. We identify the height of the oil bath as a key parameter that can be used to control the distance between the drops. We further show that the height of the bottom boundary can be patterned to direct the drops along desired trajectories at the interface.

Although the polarization of water drops in the presence of an external electric field<sup>37,38</sup> and the triboelectric charging of water drops<sup>39–43</sup> are well established, we harness these concepts to develop novel methods to manipulate drops specifically at a

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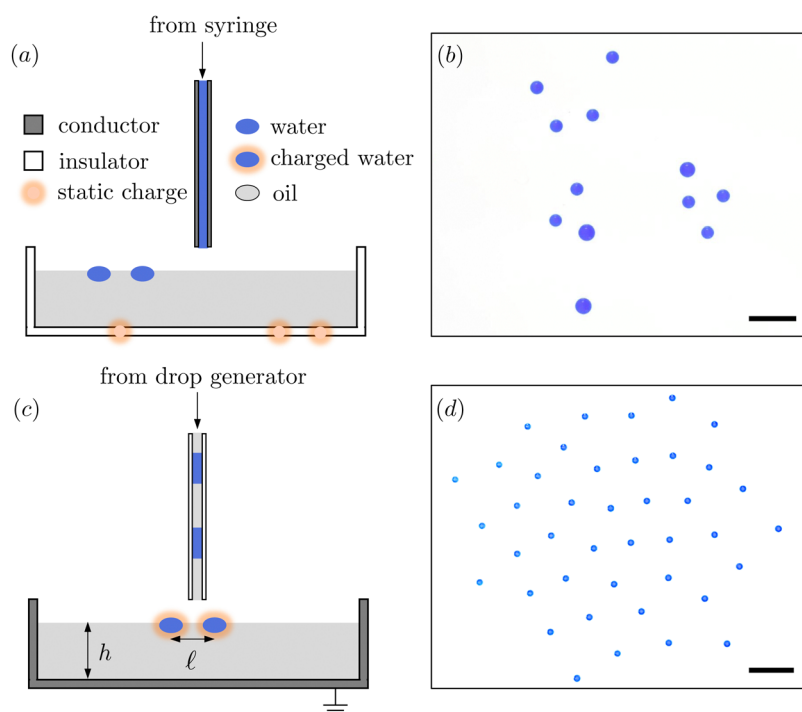
liquid–air interface. In so doing, we uncover and elucidate new behavior of floating drops such as self-assembly, trapping to a specific location at the interface, and spontaneous motion in response to the shape of the boundary, which have not been reported before to the best of our knowledge.

## 2 Experimental setup

The two experimental configurations are summarized in Fig. 1: panels (a) and (b) correspond to the first configuration, and panels (c) and (d) correspond to the second. Both cases involve water drops floating at an oil–air interface. We use fluorinert FC-40 (Sigma-Aldrich) as the oil phase with a relative permittivity  $\epsilon_r = 1.9$ , surface tension  $\gamma_o = 17.3 \pm 0.3 \text{ mN m}^{-1}$ , viscosity  $\mu_o = 4.01 \text{ mPa s}$ , and density  $\rho_o = 1855 \text{ kg m}^{-3}$ . The permittivity and density are crucial properties for all observations presented here: a low relative permittivity allows for electrostatic interactions through the oil without screening and a high density allows for the water drops to float. Experiments with Novec 7500 (3M) as the oil phase,  $\epsilon_r = 5.8$ , did not show self-assembly. The aqueous phase is deionized water dyed with Erioglaucine disodium salt (0.4 wt%; Sigma-Aldrich) and the drop radius is  $O(1) \text{ mm}$ . Since the dye is a salt, a high concentration of the dye ensured that the drops were only modestly charged for the second configuration.<sup>40</sup> Note that the dye is not necessary for the qualitative observations described in this work.

In the first experimental configuration shown schematically in Fig. 1(a), the water drops are not charged and are dispensed onto an oil-filled Petri dish that is an insulator (dielectric). The drops are dispensed continuously from a syringe through a grounded metal needle, which ensured neutral charge of the drops. The neutral charge of the drops were checked using a Faraday cup and NanoCoulomb meter (Monroe Electronics; Model 284). We show that the uncharged drops can still be polarized by static charges on the surface of the container, or any nearby objects, and therefore can be assembled and controlled at the oil–air interface. Fig. 1(b) shows a top view of the drops self-assembled at the interface in configuration 1. The heterogeneity in size of the drops are due to the coalescence of the drops.

In the second configuration, the drops are charged and are dispensed onto an electrically grounded Petri dish covered with aluminum foil, as shown in Fig. 1(c). The drops are generated with a two-inlet one-outlet microfluidic T-junction device which creates a segmented flow of water drops in oil.<sup>44</sup> The flow rate of both phases was  $Q = 0.05 \text{ mL min}^{-1}$  as set using a syringe pump. At the outlet, the solution flowed through a Teflon tube (length 20 cm and inner diameter 0.38 mm), where the water drops picked up a net positive charge due to the charge separation that occurs in the electrical double layer.<sup>39,40,45,46</sup> Fig. 1(d) shows the self-assembly of the charged drops, which exhibit more order than the polarized case (Fig. 1(a) and (b)).

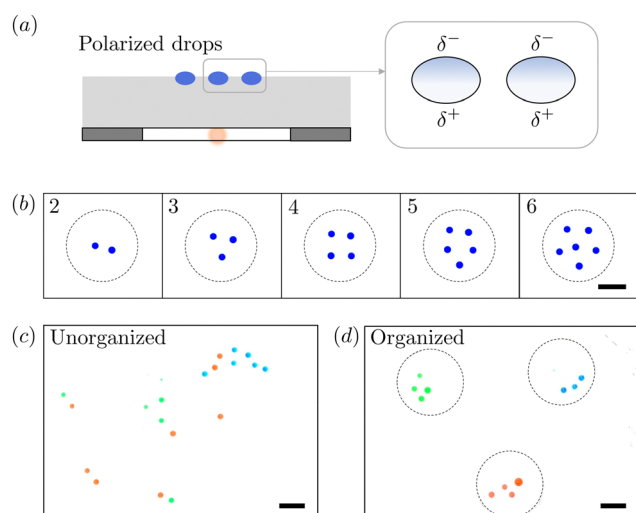


**Fig. 1** The experimental setup for the two configurations. (a) Schematic of the first configuration where the drops are dispensed from a syringe through a metal needle. The drops, which have a neutral charge, float at the oil–air interface and the container is an insulator (dielectric). The yellow spots on the container schematically show localized static charge that may be present on the dielectric. (b) Top view experimental image showing the preferential self-assembly of the drops in configuration 1. (c) Schematic of the second configuration where the drops are produced by a microfluidic drop generator and are dispensed onto the container through an insulating tube. The drops in this configuration have a net positive electric charge and the container is a conductor that is grounded. (d) Top view experimental image showing the self-assembly of the charged drops. Scale bars in (b) and (d) represent 1 cm.

The amount of charge that the water drops pick up is a function of the length of the tube, diameter of the tube, flow rate, concentration of dye in the drop, size of the drop, and the humidity of the environment. For simplicity, we kept all these parameters constant in the experiments presented here. The relative humidity was approximately 20% and the ambient temperature was about 21 °C. We note that the experimental system is very sensitive to the external fields present in the environment in the sense that the position and the movement of the drops at the interface can be influenced by fields created by static charges on nearby surfaces including the hand of the experimentalist.

### 3 Polarized drops

While we rely on electrostatic interactions to manipulate and control the drops at the interface, we show that it is not necessary for the drops to have a net electric charge. Fig. 2(a) shows a schematic of the oil–water system in a Petri dish covered with aluminum foil with a small circular window cut out in the middle. The window exposes the plastic underneath, which is a dielectric surface prone to static charges. In fact, we can place charges on dielectric surfaces by bringing two dielectric surfaces



**Fig. 2** The preferential assembly of polarized but uncharged drops (configuration 1). (a) Schematic showing the experimental setup where the container is covered with aluminum foil (dark gray) with only a small window in the middle cut out to expose the dielectric material. The inset shows a schematic of the drops polarized, in this case, by a negative charge on the surface below. (b) Experimental images showing drops assembled near the static charge that are present on the dielectric surface. Note that the interior of the dotted circle represents the area that is exposed to the dielectric material underneath. The assembled drops from  $N = 2$ – $6$  show 2D crystalline order. (c) Experimental image showing an “unorganized” state where the entire dielectric surface of the container is exposed. The drops dyed with different colors are randomly dispensed over the surface and get attracted by nearby charges. (d) Experimental image showing an “organized” state, where three windows expose the dielectric surface of the container. Drops are dispensed inside each window based on their color such that each window contains a set of different colored drops. Scale bars in (b–d) represent 1 cm.

into contact, causing a charge separation that leaves one surface with an excess negative, and the other with an excess positive charge.<sup>45</sup> In our experiments, we do this by touching the exposed surface of the Petri dish with a gloved finger after neutralizing the surface with a Zerostat pistol (Sigma-Aldrich).

Water is a strong dielectric ( $\epsilon_r = 78.5$ ) and can be polarized easily in the presence of an electric field. When the uncharged water drops encounter the field produced by the static charges and become polarized, as shown in the inset in Fig. 2(a), there are two consequences: the drops are attracted toward the charge causing the drops to move to the location on the interface above the charge, which then leads to dipole–dipole repulsion between the drops. The combination of these two effects results in self-assembly of the drops at the interface above the location of the static charges. Fig. 2(b) shows the patterns formed by  $N$  number of drops as they assemble above the exposed window on the Petri dish; the dotted circles in the images represent the outline of the exposed window. The reason for covering most of the Petri dish with aluminum foil and only exposing a small window is to localize the effect and to eliminate unwanted field interactions. As shown in the figure, the drops show 2D crystalline order from  $N = 2$  to  $N = 6$ . Adding more drops resulted in a breakdown of this order and coalescence between some of the drops. This breakdown could be because the attractive force bringing the drops toward the charge is stronger than the dipole–dipole repulsion between the drops. The capillary length of the system is  $l_c = \sqrt{\gamma_o/(\rho_o g)} \approx 1$  mm, which means that an additional attractive force contribution from the Cheerios effect might also be present and dominant for separation distances close to  $l_c$ . Similar crystalline patterns have also been observed with solid particles at interfaces due to electric or magnetic forces.<sup>47–50</sup>

The polarization effect of the water drops can be further leveraged to sort and organize drops at the interface. Fig. 2(c) shows an “unorganized” state of the drops, dyed red, blue, or green, when the Petri dish below is fully exposed. The randomly distributed static charges on the boundary leads to a random distribution of the drops along the interface. To achieve a more “organized” pattern, where the location of the drops can be dictated in advance, we covered the Petri dish with aluminum foil only exposing three circular areas as shown in Fig. 2(d). Each of the colored drops was released above its corresponding window. Thus, we were able to collect and retain the three different colored drops in three different regions. Patterning the surface of the container with static charges is therefore a simple way to dictate the location of the uncharged floating drops and to create local self-assembled colonies.

### 4 Charged drops

The focus so far has been on understanding the collective behavior of uncharged, polarized drops at the oil–air interface in an insulating container. Here, we switch our attention to the second configuration (Fig. 1(c) and (d)): charged drops in a conductive container.

In the experiments, we dispense electrically charged drops onto the center of the container with the oil bath at height  $h$ .

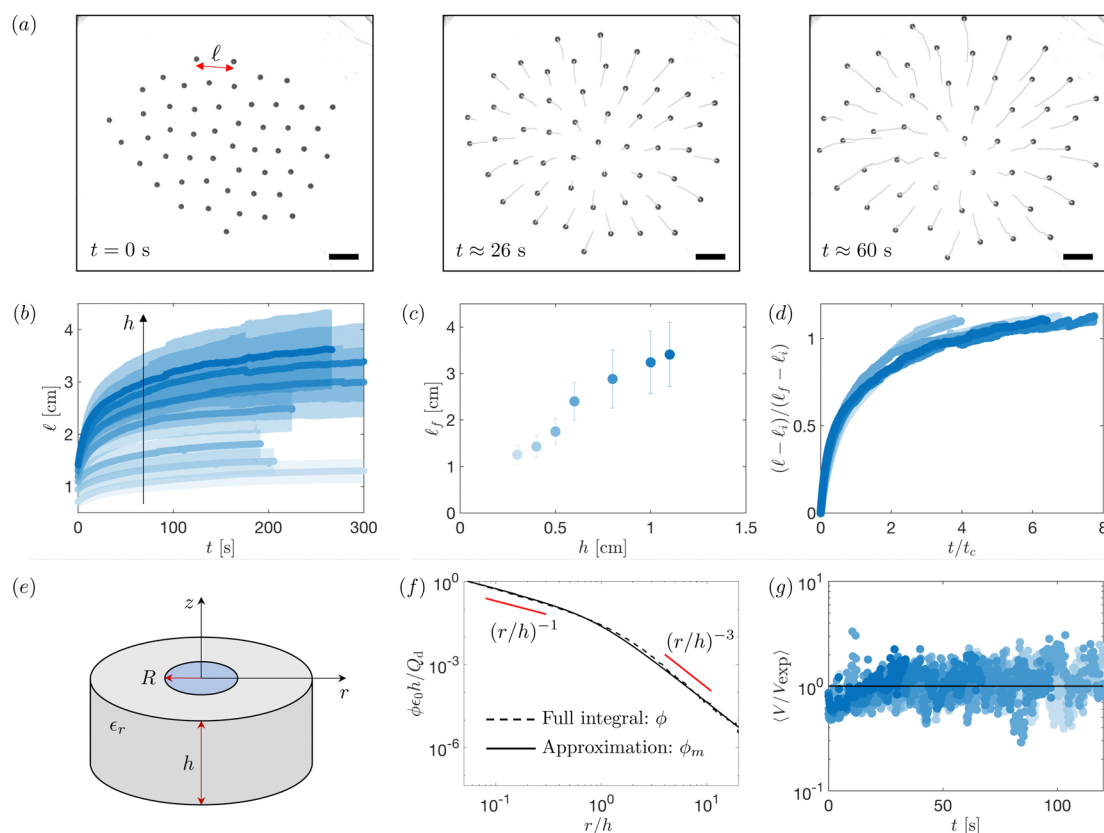
The drops have a projected radius of about  $R = 1.5$  mm, as imaged from above, and have a charge of approximately  $Q_d = 0.07 \pm 0.01$  nC as measured using a Faraday cup and Nano-Coulomb meter (see ESI,† Fig. S1). As subsequent drops are added, the previous drops get pushed away from the center towards the edges due to electrostatic repulsion and due to the flow of oil in the bath. Therefore, in order to ensure that all experiments start with a similar initial condition and that the patterns we observe are independent of the dynamics that occur during the introduction of the drops, upon stopping the drop dispensation we bring the drops closer together at the interface and constrict them using a dielectric stylus, before allowing them to spread apart and relax.

For a case with  $h = 0.4$  cm, Fig. 3(a) shows an image sequence at the moment the array was allowed to relax, which we label  $t = 0$  s, and later at  $t \approx 26$  s and  $t \approx 60$  s. The gray tails map the trajectory of each drop from its initial position. Note that the length of the tails is indicative of the speed of the

drops, which for reasons of symmetry is larger for the drops along the periphery. In what follows, we will show that this system is dominated by electrostatic repulsion. The drops are always moving away from each other although they will reach a quasi-equilibrium configuration within the observation time of the experiments.

#### 4.1 Effect of the height of the oil bath

We analyzed how the distance between the drops evolves as each of the assemblies with varying  $h$  reach their quasi-equilibrium pattern. The distance  $\hat{\ell}$  for a given drop is defined as the average distance to its three closest neighbors. We chose to consider three neighbors since the drops on the outer edge of the pattern might only have three neighbors – two on each side and one towards the interior. Fig. 3(b) shows the average distance  $\ell$ , representing the average of  $\hat{\ell}$  for all the drops on the surface, as a function of time  $t$ , with  $t = 0$  being defined as the



**Fig. 3** The distance between the drops as a function of the height  $h$  of the oil bath. (a) Image sequence showing the self-assembly of drops for a case with  $h = 0.4$  cm at the moment the assembly was allowed to relax,  $t = 0$  s, and later at  $t \approx 26$  s and  $t \approx 60$  s. The gray tails map the trajectory of each drop and the scale bars represent 1 cm. (b) The time evolution of the average distance  $\ell$  between the drops for several  $h$  values, as the drop assembly relaxes and reaches a quasi-equilibrium configuration with typical spacing  $\ell_t$ . From the lightest to the darkest, the marker colors correspond to  $h = 0.3, 0.4, 0.5, 0.6, 0.8, 1.0$ , and  $1.1$  cm. The shaded error bars are standard deviations from averaging the distance for all the drops in the frame. (c) The quasi-equilibrium distance  $\ell_f$  as a function of  $h$ . (d) Rescaling the  $x$  and  $y$  axis, all curves can be collapsed. Here,  $t_c = (\ell_f - \ell_i)/u_0$  and  $u_0$  is the average rate of expansion of the drops. (e) Schematic showing the geometry considered in the theoretical model. The drop is represented as a flat circle of radius  $R$ , which is a distance  $h$  from the bottom conductive boundary. (f) The theoretical result showing the exact electrostatic potential  $\phi$  (dashed line) and the approximate solution  $\phi_m$  (eqn (1); solid line) at the interface,  $z = 0$ , in the point charge limit ( $R \rightarrow \infty$ ). (g) The ratio of the magnitude of the theoretical velocity of a drop to the magnitude of the experimentally measured velocity averaged over all drops,  $\langle V/V_{\text{exp}} \rangle$ , is plotted as a function of time. The solid black line represents  $\langle V/V_{\text{exp}} \rangle = 1$ .

time when the assemblies were allowed to relax from the initial state of confinement. We observe that  $\ell$  increases over time, although very slowly at long time, and seem to approach different values as a function of  $h$ . To characterize this behavior, we define the quasi-equilibrium distance  $\ell_f$  as the average of the distance when the local fractional change in  $\ell$  is below a threshold:  $\Delta\ell/\ell < 10^{-4}$ . Since  $\ell(t)$  is always increasing,  $\ell_f$  is not simply the largest value but some representation of  $\ell(t)$  as  $t \rightarrow \infty$ . In Fig. 3(c), we plot  $\ell_f$  as a function of  $h$ . As  $h$  is increased,  $\ell_f$  also increases. But for large  $h$ ,  $\ell_f$  is only weakly dependent on  $h$ , which is expected since the effect of the bottom boundary will decrease as the distance from the boundary is increased. In Fig. 3(d), we rescale the time evolution of  $\ell$  by only considering the fractional change in distance  $(\ell - \ell_i)/(\ell_f - \ell_i)$  over time, where  $\ell_i = \ell|_{t=0}$ . We rescale time with an empirical time scale  $t_c = (\ell_f - \ell_i)/u_0$ , where  $u_0 = \frac{d\ell_i}{dt}$  is the average rate of expansion of the drops as taken as the slope of the experimental data in Fig. 3(b) near  $t = 0$ . The collapse of the data indicates that the initial spacing  $\ell_i$  of the drops and height  $h$  of the bath set the time scale of the dynamics.

The dependence of the collective behavior of the drops on the bath height  $h$  is captured by the representative electrostatic potential  $\phi$  of the system and can be explained with a simplified model. Consider the geometry shown in Fig. 3(e), where the drop is assumed to be a flat circle of radius  $R$ , located at the origin, and having a uniform charge distribution. We consider a domain that is infinite in the radial direction  $r$  and semi-infinite in the axial direction  $z$ , where there is a finite height  $h$  of oil below the drop with a relative permittivity  $\epsilon_r$  and an infinite layer of air above the drop. An exact analytical solution does not exist for  $\phi$  in closed form. But, the approximate solution, labelled  $\phi_m$ , representing the electrostatic potential at the interface generated by one drop can be written as (see Section SII, ESI†)

$$\phi_m(r, z=0) = \frac{Q_d h^2}{2\pi\epsilon_0 r[(\epsilon_r + 1)h^2 + \epsilon_r^2 r^2]}, \quad (1)$$

where  $Q_d$  is the total charge of the drop,  $\epsilon_0 = 8.854 \times 10^{-12} \text{ F m}^{-1}$  is the permittivity of free space, and  $r$  is the distance from the center of the drop. Our model captures the dielectric discontinuity at the oil–air interface and the conductive boundary condition at the bottom. Fig. 3(f) shows  $\phi_m$  plotted along with the exact numerical solution  $\phi$  in the point charge limit ( $R \rightarrow 0$ ), evaluated at  $z = 0$ , in dimensionless form. Note that for shallow bath heights, corresponding to large  $r/h$ , the dimensionless potential scales as  $(r/h)^{-3}$  which means that the potential has a quadratic dependence with the height, as also seen from eqn (1). A detailed calculation can be found in the ESI† (see Fig. S2).

It is important to note that the patterns we observe in Fig. 3(a) and the separation distances  $\ell_f$  that we measure are mesoscale, with  $\ell_f \gg l_c$ , which clearly indicates that the electrostatic interactions are much stronger than the Cheerios effect (see ESI†, Section SIII and Fig. S3). We analyze the velocity of drops spreading as a function of time and compare with the theoretical prediction using the calculated electrostatic potential

in order to validate that the system is dominated by electrostatic repulsion. Assuming that inertia is negligible, the evolution of the speed of a single drop for a purely repulsive system can be derived by balancing the viscous drag force with the electrostatic force. In the experiments, the Reynolds number  $\text{Re} = \rho_o V R / \mu_o$ , where  $V$  is the speed of the drops, ranges from 0.007 to 0.7, with the maximum corresponding to the early time dynamics of the experiments with the largest bath height. Taking the viscous drag to be that of a sphere in a bulk fluid modified by a fitting parameter  $\beta$  (see ESI†, Section SIV and Fig. S4), which accounts for any excess drag due to the presence of the interface and the bottom surface of the container in shallow cases, the velocity is  $\mathbf{V} = \frac{1}{6\beta\pi\mu_o R} \frac{dU_e}{dr} \hat{\mathbf{e}}_r$ . Here  $U_e = Q_d \phi_m(r, z=0)$  is the electrostatic energy and  $\hat{\mathbf{e}}_r$  is the unit vector from the drop towards its neighbor. The theoretical velocity of a two-body system separated by a distance  $r$  is then (see ESI†)

$$\mathbf{V} = -\frac{Q_d^2 h^2 [(\epsilon_r + 1)h^2 + 3\epsilon_r^2 r^2]}{12\beta\pi^2\mu_o\epsilon_0 R r^2 [(\epsilon_r + 1)h^2 + \epsilon_r^2 r^2]^2} \hat{\mathbf{e}}_r. \quad (2)$$

We calculate the theoretical velocity of each of the drops in the container at every time step by taking the distances and unit vectors as input from the experimental data and summing the two-body interactions between a drop and six of its closest neighbors to calculate the resultant velocity vector. Considering more than six neighbors had negligible effect on the resultant velocity vector. The magnitude  $V$  of this theoretical velocity in the absence of any fitting, *i.e.* taking the fitting parameter  $\beta$  to be 1, can be compared to the magnitude of the experimentally measured velocity  $V_{\text{exp}}$  for each drop in the container. The ratio of the two velocities averaged over all the drops for a given time step,  $\langle V|_{\beta=1}/V_{\text{exp}} \rangle$ , is nearly time-independent as shown in Fig. S4(a) in the ESI† indicating that our model captures the spatiotemporal variations in the drop velocities well. Using the fitting parameter,  $\beta$ , we can further collapse  $\langle V/V_{\text{exp}} \rangle$  around unity, as shown in Fig. 3(g), suggesting that the model with one fitting parameter  $\beta$  captures the major experimental features. The scatter in the data may be attributed to the point charge approximation of the drop in the model and the nonlinear many-body interactions not considered in the model. Nevertheless, the reasonable agreement between the experimental data and the model prediction suggests that the dynamics are dominated by electrostatic repulsion.

We further calculate the time that it would take for two drops starting at the center of the container to repel and reach the side walls of the container. The distance from the center to the side walls of the container is approximately  $l = 7.5 \text{ cm}$  for the smallest container that we used. The time  $t_f$  that it would take the drops to traverse that distance can be calculated by  $t_f = \int_0^l (1/V) dr$ , which is about 3 hours and 40 hours, respectively, for  $h = 1.1 \text{ cm}$  and  $h = 0.3 \text{ cm}$ . These large durations are a result of the quickly decaying strength of the electrostatic potential, which scales as  $r^{-3}$ , and is also the reason why the distance between the drops appears to approach a quasi-equilibrium value.



## 5 Manipulation of the drops

With the understanding of the collective behavior of the drops in both the polarized and the charged cases, we now offer two ways to manipulate the drops at the oil–air interface.

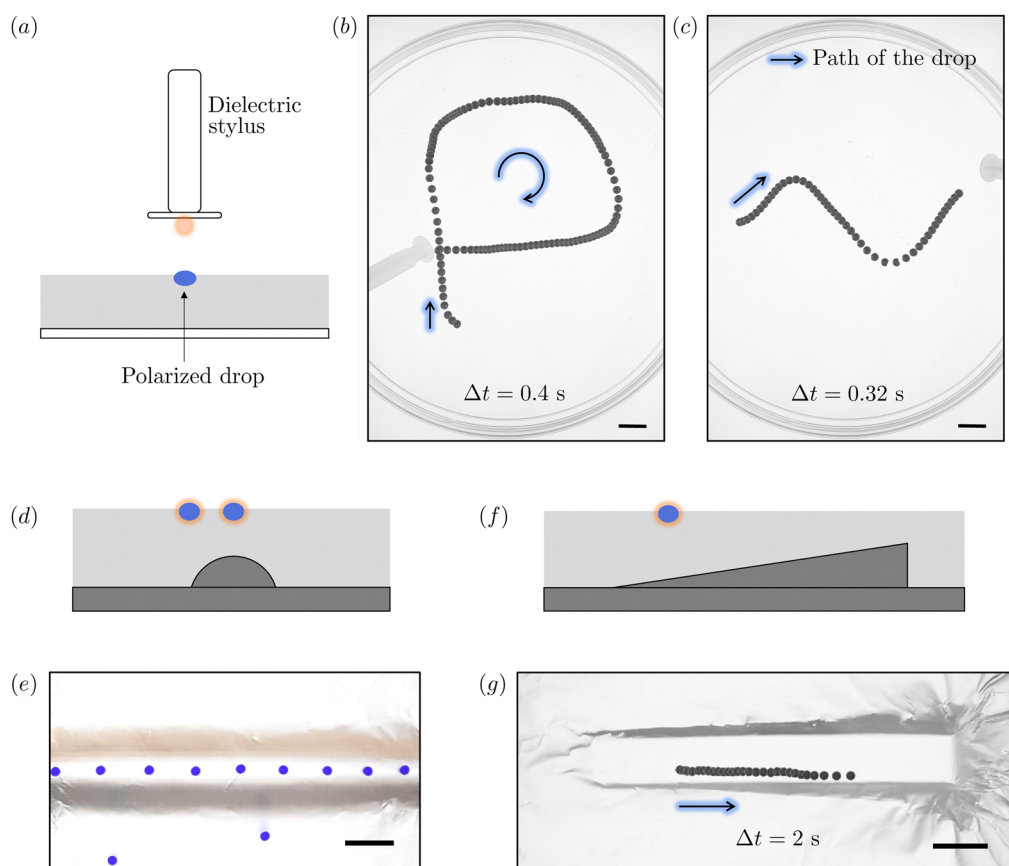
### 5.1 Active control with a stylus

We showed that static charges present on the surface of the container, which is underneath the drops, can affect the position of the drops at the oil–air interface. We now present a more dynamic, yet simple, approach using a dielectric stylus, as shown schematically in Fig. 4(a). We use the plunger of a syringe as the dielectric stylus and, after neutralizing the surface with a Zerostat pistol, place charges on the flat bottom surface of the stylus using contact electrification. The stylus can now be used to move the drop as desired on the interface. In Fig. 4(b) and (c), we provide images showing (b) the spatiotemporal evolution of a single drop as the stylus is used to draw the letter “P” and (c) an approximate sine wave traced by a single drop. Since the electrostatic

interactions are instantaneous and the friction is low due to the low viscosity of the oil subphase, the drops can be manipulated rather quickly. We believe that this method has the potential for future applications due to its simplicity and effectiveness.

### 5.2 Passive control with a prescribed shape of the container

We established that the height of the oil bath  $h$  affects the electrostatic potential at the interface. Therefore, patterning the height profile of the bottom boundary creates a non-uniform potential that allows for preferential assembly and spontaneous motion of the drops. Fig. 4(d) shows a schematic of the experimental system, where we use a section of a cylinder to create an uneven height profile. Next, Fig. 4(e) shows the experimental image at a quasi-equilibrium state, where the charged drops have moved and arranged themselves along the shallow ridge over the elevated surface. The underlying physics that drives the drops to the shallow region can be explained by the classic problem in electrostatics of a point charge next to a conducting sphere. If a point charge is confined to a horizontal plane of



**Fig. 4** Manipulation of the drops at the interface. (a) Schematic showing configuration 1 with polarized but uncharged drops where the polarization comes from static charges on the surface of a dielectric stylus. (b) An image showing the spatiotemporal evolution of a drop as a stylus is used to write out the letter “P” with the drop. The time elapsed was 45 seconds and the arrows with the blue halo show the path of the drop. (c) An image showing another spatiotemporal evolution of a drop as a stylus is used to make a sine wave. The time elapsed was 15 seconds. (d) Schematic showing a cross-section of the hemispherical cylinder placed on the bottom to create a non-uniform surface. (e) Image showing the drops preferentially assembled and aligned over the ridge. The horizontal feature on the image corresponds to the elevated bottom surface. (f) A schematic of the geometry considered in an experiment, where an inclined plane with an angle of about  $2^\circ$  is placed below the drop. (g) A spatiotemporal image showing the trajectory of the spontaneous motion of a charged drop over the inclined plane. The time elapsed was 48 seconds. All scale bars in the figure represent 1 cm.

height  $h$  above a conducting sphere, the location at which the force along the plane vanishes will be directly above the center of the sphere. Although the ideas remain the same, we consider a hemispherical boss shape to theoretically explain this phenomenon in the ESI† (see Section SV and Fig. S5).

The feature that the charged drop will move along the interface to be close the conducting bottom surface can be further leveraged to create unmediated motion of the drops along a desired path. In experiments, we placed a charged drop above an inclined plane, as schematically shown in Fig. 4(f), with an inclination angle of about  $2^\circ$ . The drop spontaneously moved toward the shallow end. A spatiotemporal plot showing the trajectory of the drop is displayed in Fig. 4(g), where the time between each frame shown is 2 s. Although the dynamics of the motion will depend on the details of the geometry, as shown in the model in the ESI,† Section V, this is a simple way to produce unmediated and spontaneous motion at the interface.

## 6 Conclusions

In conclusion, a novel experimental system was introduced to control and manipulate water drops floating at an oil–air interface. We showed that uncharged drops can be trapped at desired locations on the interface, or effortlessly moved around in determined trajectories along the interface through polarization *via* static surface charges. On the other hand, a collection of charged drops self-assembled at the interface, dominated by the electrostatic repulsive forces that was found to be a function of the height  $h$  of the oil bath. Exploiting the dependence on  $h$ , we showed that patterning the height profile of the bottom boundary allowed for preferential assembly and directed motion of the drops. With this work, we extend the research area of drop manipulation to the liquid–air interface and also offer our experimental setup as a potential platform to study many other problems in physics including 2D solids, colloidal crystals, and elastic waves.

## Author contributions

PRK, AAP, and HAS conceived the idea. PRK performed the experiments and collected the data. PRK, Jpds, GT, AAP and HAS discussed and interpreted the results, and wrote the paper.

## Data availability

Data for this article are available on github at [https://github.com/paulkaneelil/drop\\_assembly\\_project](https://github.com/paulkaneelil/drop_assembly_project).

## Conflicts of interest

A provisional patent is being filed for the methods described in this work.

## Acknowledgements

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