

## Smectic bubbles in strong external electric fields

Torsten Trittel <sup>1,2</sup> Christoph Klopp,<sup>2</sup> Caterina Tosarelli,<sup>3</sup>  
Emmanuelle Lacaze <sup>3</sup> and Ralf Stannarius <sup>1,2,\*</sup>

<sup>1</sup>*Fachbereich Technik, Technische Hochschule Brandenburg, Magdeburger Straße 50,  
14770 Brandenburg an der Havel, Germany*

<sup>2</sup>*Institut für Physik, Otto-von-Guericke-Universität Magdeburg, Universitätsplatz 2,  
39106 Magdeburg, Germany*

<sup>3</sup>*Institut des Nanosciences de Paris (INSP), Sorbonne Université,  
CNRS, 4 Place Jussieu, 75005 Paris, France*



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Smectic bubbles as fluid structures with a huge surface-to-volume ratio represent unique examples of quasi-two-dimensional (2D) fluids. We investigate the behavior of islands, local regions with larger film thickness, in the presence of strong inhomogeneous electric fields. The islands, which can be considered here as 2D analogs of liquid droplets, become charged and are repelled by a needle-shaped electrode in contact with the film. The mechanism has strong similarities to electrospraying and electrospinning, but because of the constraint of the thin film, the motion is confined to the film plane. Interestingly, the phenomenon depends on the polarity of the electric field. This indicates that the charging of the islands is related to the asymmetric ion mobilities in the film material. To prevent sedimentation of the islands, the experiments were performed under microgravity conditions on parabolic flights.

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### I. INTRODUCTION

Smectic freely suspended films [1] are classical examples of quasi-two-dimensional fluids. Films of only few nanometers thicknesses in the smectic A or C phases can freely span areas up to several square centimeters when they are supported by a lateral solid frame. Within the molecular layers, the film material flows like a liquid. Such films have been exploited in numerous experiments since their discovery [2–4]. A good overview of recent progress in freely suspended smectic films can be found in Refs. [5,6]. Smectic phases can also form curved films in the form of spherical bubbles [7,8], where the shape is stabilized by an inner Laplace pressure of few Pascal [9]. A peculiar variant are closed smectic bubbles, which can float freely like soap bubbles [10,11]. Here, our focus is on the effects of electric fields on smectic bubbles and on inclusions in their films.

Most of the classical experiments with smectics in electric fields are performed in sandwich cells, where the field is normal to the cell plane and of interest are primarily electro-optic effects in smectic C and C\* phases induced by switching orientation of the optic axis. As in cells, the optical axis in freely suspended films can be switched by means of electric fields [12–14]. With rotating electric

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\*Contact author: [ralf.stannarius@ovgu.de](mailto:ralf.stannarius@ovgu.de)

fields, spiral and target patterns of the c-director can be prepared [15,16]. In such experiments, two or more electrodes are usually in direct contact with the film material. Typical electric field strengths are of the order of  $10^3$  to  $10^4$  V/m.

In sandwich cells, flow is strongly inhibited by the presence of the glass plates. In comparison, effects of in-plane flow in freely suspended films can be much stronger. Macroscopic flow can be excited, for example, by mechanical shear [17,18], by the relaxation of elastic c-director distortions [19,20], by thermal gradients [21,22], or by electrically induced convection [23–26]. All these experiments were performed with flat horizontal films.

In an experiment with a capillary-supported sphere-shaped film (smectic bubble) that was placed between two planar electrodes without direct contact [8], it was shown qualitatively that thicker film regions on the bubble surface can be relocated by a uniform external electric field. This result is discussed in more detail below.

If one is interested in the motion of objects like solid or liquid inclusions or islands (regions with locally enhanced film thickness) on the surface of smectic bubbles without having to consider sedimentation in the gravitational field, microgravity ( $\mu g$ ) is needed. This was achieved earlier in parabolic flights, e.g., [10,11,27] and on board the ISS, e.g., [27–29]. Bubbles in these ISS experiments were supported by a thin capillary. The focus of these experiments was on the interactions of groups of droplets and islands in the films. An advantage of the bubble geometry over flat films in such experiments is the absence of large menisci, which otherwise interact with the inclusions and influence their behavior.

For the experiments presented here, the short microgravity phases during parabolic flights are sufficient and the comparably low  $\mu g$  quality is not critical. The typical parabolic flight scenario aboard an Airbus A310 airplane (at NOVESPACE, France) is a 22 second  $\mu g$  phase, preceded and followed by hypergravity ( $\approx 1.8 g$ ) phases of approximately 20 seconds each. The smectic films can be prepared in the preceding hypergravity phase and the bubbles are released to float freely after entry into  $\mu g$ , so that approximately 20 seconds are available for experiments. When the airplane enters the second hypergravity phase, freely floating bubbles disappear from the field of view, and they are lost. If bubbles are attached to a solid support, e.g., a capillary, one may re-use them in subsequent parabolas.

In films of inhomogeneous thickness exposed to electric fields, one has to consider dielectric and charge effects. Dielectrically, a thicker film area or smectic island in an inhomogeneous electric field will experience a small force towards the higher field strength, but this effect is rather weak. The influence of electric fields on droplets (of the same material as the film) in uniformly thick films of a smectic bubble was observed in an ISS experiment [29]. Droplets of a few micrometer radius had formed a regular lattice in this experiment, with a lattice constant of approximately 25  $\mu m$ . Inhomogeneous electric fields were applied using a pair of needles that were not in contact but very close to the bubble surface [Fig. 1(a)]. The electric potential between the needles was 150 V, corresponding to an electric field strength of the order of several dozen kV/m near the needle. Droplets in the film near the needle were attracted by a positively charged electrode, but repelled from a negatively charged needle, and they moved on the film plane, distorting the lattice locally. A quantitative evaluation was not possible, but the experiment clearly demonstrated that the droplets carried a negative charge. As a possible explanation, it was suggested that the bubble is slightly negatively charged after inflation and that the charge per film area is larger in the droplets than in the surrounding film. A confirmation of this hypothesis is still pending.

As mentioned above, another observation, and probably the most relevant for the interpretation of the present experiment, was made in a sessile bubble under normal gravity [8]. The bubble had an initially inhomogeneous thickness, and the thicker section had settled to a spherical cap at the bottom, around the support capillary. When a uniform electric field of 6.8 kV/m was applied by two lateral planar electrodes, the island was dragged up towards the positive electrode [Fig. 1(b)]. A maximum angle of about  $50^\circ$  with respect to the horizontal was reached, but this state did not persist. Gradually, the island sank back to the bottom even when the electric field was kept constant.

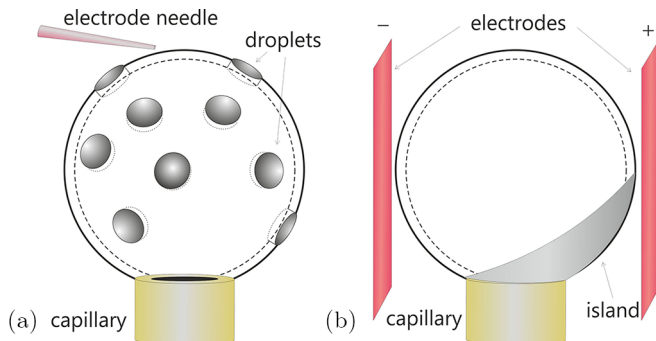


FIG. 1. Sketches of earlier experimental geometries of smectic bubbles in electric fields: (a) bubble with isotropic droplets on the ISS, the needle (shown here exaggerated with respect to the bubble size) acts as an electrode, the capillary as the counter-electrode [29]. (b) Bubble in the ground lab with two plate electrodes and a thicker smectic film region that is attracted temporarily toward the anode. Sketches are not to scale [8].

At still higher fields, islands that were originally disk-shaped were deformed to an elongated shape, stretching towards the positive electrode [8].

The purpose of the present study was to observe the effects of electric fields in the absence of gravitation effects. We hypothesized that islands would relocate on the surface of freely floating bubbles towards the side facing the anode. We also expected a possible drift of complete freely floating bubbles towards the anode.

## II. EXPERIMENTAL SETUP AND MATERIALS

Our original intention was the preparation of freely floating bubbles in homogeneous electric fields. A simple technique that works for soap bubbles is to inflate bubbles from a thin film across the tip of a straw or pipe and to release them by a sharp blow. This turned out to be unsuitable for smectic films. One can blow bubbles, but it is practically impossible to release them by blowing in air, as they are too sensitive to such quick shape changes and burst when they detach from the pipe end. An alternative is preparation by catenoid collapse [30], a technique that has been developed for reliable routine use [10,11]. It is used here.

The experiments were performed aboard an Airbus A310 at NOVESPACE, Bordeaux (France) during the 63rd CNES Parabolic Flight Campaign, September 26–October 7, 2022. The setup was a modified version of the apparatus used in earlier parabolic flights [31]. It is sketched in Fig. 2. The first sketch in (a) shows the two high-speed cameras C1 (Phantom V611) and C2 (VEO 710) and the catenoid drawing table T.

Sketch (b) shows the details of the drawing table: a fixed catenoid holder ring R1, a movable ring R2 and the stepper motor SM that is used to control the distances between the two coaxial rings. Both rings have the same radius  $R = 25$  mm. Initially, the rings R1 and R2 are in contact and the liquid crystal is spread at their edges so that it wets the two rings. When the rings are slowly moved apart by the stepper motor SM, a cylindrical and later catenoid-shaped smectic film is formed that spans the gap between R1 and R2. This preparation is done in the hypergravity phase. The ring R2 is stopped just before the critical distance for catenoid collapse is reached. When the airplane enters the microgravity phase, the rings are further separated, exceeding the critical distance of  $1.32548 R$  for catenoid collapse. The result of this collapse, which is relevant here, is the satellite bubble [30], which is then observed by the two synchronized fast cameras at a frame rate of 5000 fps. The essential modification with respect to the earlier setup are two hollow electrode needles (outer diameter  $2R_{\text{needle}} = 1.05$  mm) that are placed near the central axes of R1 and R2, but slightly offset (to avoid contact when the rings touch at the start of the catenoid drawing process). The needles positioned in the rings are shown in (c). In addition, we also tested a geometry with plate electrodes instead of the needles. The plate electrodes were placed behind the catenoid

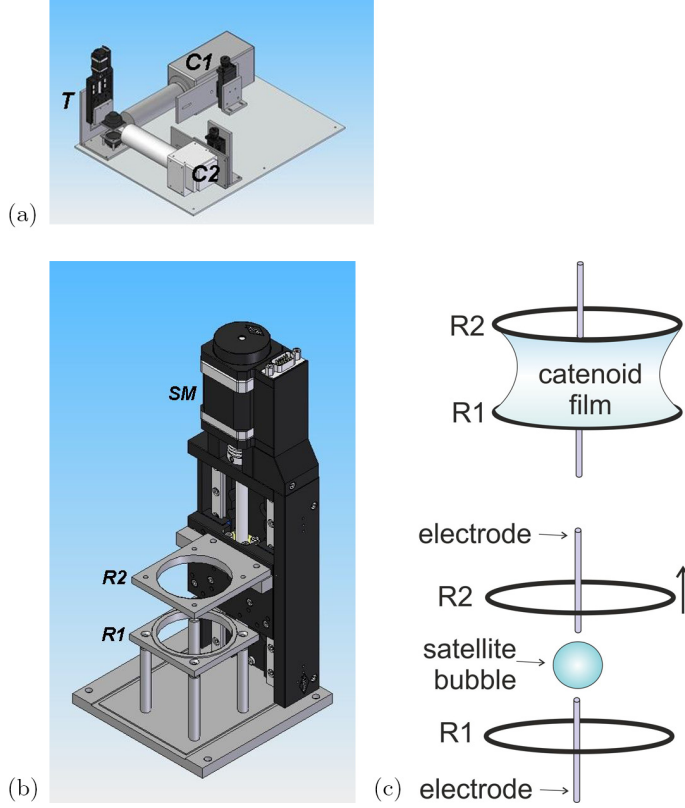


FIG. 2. Catenoid film preparation setup (modified from [31]) used in the present study. (a) View of the positions of the high-speed cameras C1 (V611) and C2 (VEO710) and the drawing table; (b) details of the drawing table  $T$  with two support rings and the stepper motor (SM) to move R2; (c) sketch of the smectic film during catenoid drawing (top) and after catenoid collapse (bottom) with the freely floating smectic bubble. The two thin cylinders represent the electrodes. The electrodes are connected to the rings, their tips being located approximately 15 mm above/below rings.

holder rings. High-voltage electric fields were created with an XP Power QH10-5 DC-DC Converter, 1 kV dc/1.25 mA.

The liquid crystal material used was a 50%/50% mixture of 5-n-octyl-2-[4-(n-hexyloxy) phenyl]pyrimidine and 5-n-decyl-2-[4-(n-octyloxy) phenyl]pyrimidine, which is smectic C in a broad temperature range around room temperature. In order to obtain samples with a controlled concentration of ions, we added 0.1 wt.% of tetrabutylammonium bromide. It dissolves in the liquid crystal without changing the smectic C phase, and forms a small  $Br^-$  anion and a bulky cation. This asymmetry turns out to be responsible for several important effects found in our study.

After the catenoid had collapsed, we observed the resulting smectic bubbles with and without applied electric voltages by means of the two high-speed cameras. The recording was stopped manually by a post-trigger and the images recorded in the relevant time period before the trigger event were stored for evaluation.

### III. EXPERIMENTAL OBSERVATIONS

We performed different sets of experiments. In some experiments, the bubble was floating freely when the electric field was applied. In other experiments, we took advantage of the fact that some

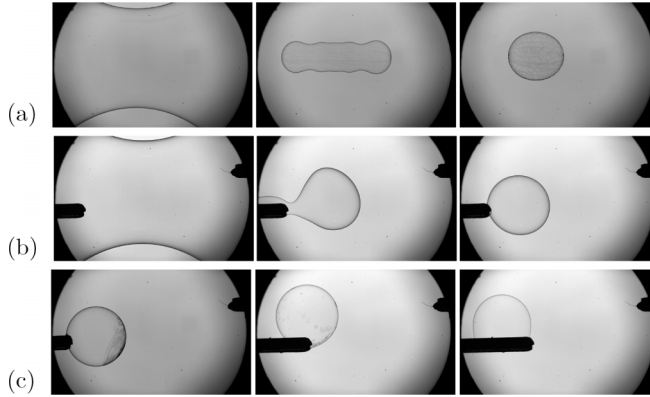


FIG. 3. Catenoid collapse in the catenoid preparation used in the present study, seen with the VEO710 in transmitted light. (a) Snapshots with planar electrodes (outside the field of view), taken in 5 ms steps, a freely floating satellite bubble forms; (b) snapshots with needle electrodes taken in 8.44 ms steps, the bubble is captured by the left electrode and survives. (c) Three different stationary attachment geometries: frontal at the tip (left), sideward at the tip (middle), and lateral (right). Image sizes are  $14.1 \times 8.8 \text{ mm}^2$ .

bubbles from the previous parabola had survived the hypergravity phases and stuck to one of the electrodes. Such bubbles could be observed during several following parabolas, so that multiple experiments with one and the same bubble were possible.

#### A. Freely floating bubbles

After catenoid collapse, we observe two different principal scenarios, depending on the electrode geometry. Typical snapshots of collapsing catenoids in  $\mu g$  are seen in Fig. 3. The support rings (located to the left and right of each image) are at least 33 mm apart when the collapse occurs, thus they are not within the field of view. In (a), an example for the collapse with plate electrode geometry is shown. Since the plates are behind the rings, i.e., have a larger distance to each other than the two support rings, they do not affect the collapse. Like in the earlier experiments without electrodes (e.g. [10,31]), a single freely floating bubble is the most probable outcome. In a few cases, the films burst during collapse, and in about 20–30% of the experiments, more than one bubble was formed. These multiple satellite bubbles are usually aligned along the central axis, and they tend to coalesce within a few hundred milliseconds.

The experiments with plate electrodes behind the support rings turned out to be unsuccessful in the sense that no effects of electric fields were found at all. The electric field strength was on the order of 20 kV/m. Possibly, effects might be found on longer time scales, which were not accessible in the microgravity time of the parabolas. Therefore, the setup was modified and the majority of experiments were performed with needle-shaped electrodes as sketched in Fig. 2(c).

#### B. Pinned bubbles

The needle-shaped electrodes were placed near the ring centers, their tips reaching further than the ring planes [see Fig. 2(c)]. Therefore, they are slightly displaced off-axis so that they do not touch each other when the rings are brought together. Figure 3(b) shows a typical experiment with the needle electrodes. The main observation is that most of the collapses did not create freely floating bubbles, but satellite bubbles were trapped at one of the electrodes. The simple reason is that the collapsing catenoid film gets in contact with the needles before pinch-off. Then, it sticks to the metal. In about half of the cases, the trapped bubbles burst shortly after contacting the electrode tips, but when they survived the collapse, they formed stable closed bubbles that remained attached

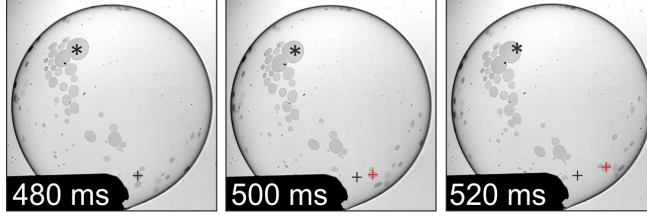


FIG. 4. Islands floating away from the electrode tip, snapshots are taken in 20 ms steps (times given with respect to the field-on switch). The black crosses are fixed at the positions of a marked island in the first frame, red crosses in the following images show the displacement of that island from the first position. The asterisk at the center of the original position of a large island in the group at the top shows that this group is also involved in the global convective motion. Background film thickness 25 nm, island thicknesses  $\approx 50$  nm. The time stamps are plotted on the needle electrode (black bar at the bottom). Images are strongly contrast enhanced. Image sizes  $6 \times 6 \text{ mm}^2$ .

there. These were robust enough to survive even during subsequent hypergravity phases so that they could be investigated in several parabolas. In principle, three attachment geometries can be distinguished, with examples shown in Fig. 3(c). In the left image, a bubble is positioned directly at the tip, almost aligned with the needle axis. In the right image, the bubble is fixed alongside the needle, and the tip is free. The most interesting geometry for our experiments was that where the bubble was positioned at the side of the tip (middle image), so that the film immediately in front of the needle tip experiences a strong electric field component in the film plane.

Islands are formed spontaneously during the catenoid collapse in most of the experiments. The reason is that when the film area is reduced in the collapse, excess smectic material must be stacked somewhere on the film. As a consequence of the smectic layer structure, the excess material forms surplus layers, which are circular in equilibrium because of the line tension at their edges (caused by dislocations in the smectic layer structure). The background film thicknesses in our experiment are between 20 and 40 nm. They cannot be controlled, but depend on the film that initially forms when the support rings are separated. The islands possess between one and a few dozen surplus layers (the single layer thickness is around 2.5 nm). The darker they appear in the images, the thicker they are. Optically, they are well below the first destructive interference order.

A 1 kV potential across a  $\approx 1$  cm electrode gap seems not too large at first glance, yet one has to keep in mind that the largest electric field strengths occur in the vicinity of the needle tips. Assuming a spherical charge with the same radius as the needles and the same electric potential, a rough estimate yields around 1 MV/m at the surface of the electrode tip. The field strength drops approximately with  $1/r^2$  in the vicinity of the electrode tip, where  $r$  is the distance from the center of a hypothetical sphere forming the needle tip. The counter-electrode is sufficiently far from that tip so that its exact position and influence can be neglected in the region of the trapped bubbles. Its contribution to the electric field formed by a superposition of the fields of the two electrodes is more than one order of magnitude smaller than that of the electrode that holds the bubble.

*a. Laterally attached bubbles.* We will first consider the case of the laterally attached bubbles, Fig. 3(c), right. The electric field in this region is comparably low, as the charges in the needle will be concentrated near its tip. The electric field is in good approximation radial with respect to the needle axis. When the field is applied, the islands on the bubble start to move, yet in an irregular fashion. There is no clear pattern in their motion. The bubbles in this geometry were thus not further evaluated.

*b. Bubbles attached sideward at the tip.* The second geometry, that of the middle image in Fig. 3(c) is much more interesting. When an electric field is switched on, the islands at the tip are pushed away from the needle toward the opposite electrode. This process sets in immediately, and it is found for both polarities of the field, yet with quantitatively different strengths. Figure 4

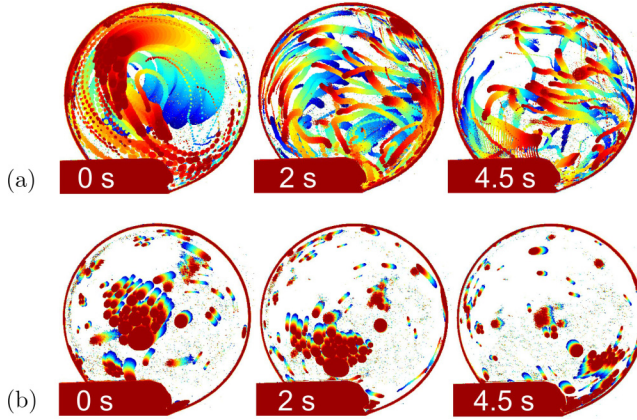


FIG. 5. Island motion driven by a positive (a) and negative (b) potential of 1 kV at the supporting electrode (red area at the bottom). The images are overlays of earlier, blue frames at the times indicated (after field-on) and subsequent images taken during the following 0.5 s, color-coded from green over yellow, orange, and red to brownish. The bubble diameter is 5.5 mm.

shows an example of islands on the film that are driven away by an electric potential of +1 kV. The crosses mark a selected island that may be used to determine the speed. An average speed of 15 mm/s is found in this bubble, 0.5 seconds after the voltage was switched on. The correlated motion of the island pattern suggests that it is not the interaction of the electric field with individual islands, but the complete film flows away from the tip. The flow pattern has the form of a convective roll that includes the entire bubble. The background film thickness is 25 nm, while the islands are approximately 50 nm thick.

In order to visualize these flow patterns qualitatively, we use superimposed contrast-enhanced color-coded images of bubbles in electric fields as shown in Fig. 5. The color coding is as follows. Earliest images are coded in blue, then we proceed via green, yellow, orange, and red to brownish. The result is best seen in Fig. 5(a) in the left image, where a group of large islands starts right of the center of the bubble image and moves counterclockwise around the center ending left of the central point. Each color scale covers a 0.5 second time interval from blue to red. The three images were composed starting at times 0 s, 2 s, and 4.5 s after the field was switched on. The two rows (a) and (b) refer to the polarities + and - of the supporting electrode, respectively. We cannot distinguish in these transmission images between islands at the front and rear sides of the bubble, but this can be done with the help of the second, vertical camera view. It reveals that most of the islands in this particular experiment are on the rear hemisphere of this bubble. The flow in both hemispheres is practically equivalent. It is obvious that not only are individual islands displaced in the film, but the complete film is moving. It is also evident that the effect gradually slows with time. Most interestingly, the polarity plays an essential role in these experiments. This is seen when Figs. 5(a) and 5(b) are compared. Evidently, the effect is much larger when the needle serves as anode, as seen in Fig. 5(a). The flow is much weaker when the needle is used as a cathode. It is important to note that both sets were recorded with exactly the same bubble during different parabolas. This means that influences from background film thicknesses or other bubble features on the flow can be excluded. The cathode induces much less flow than the anode. This is qualitatively in agreement with the observation of islands in combined electric and gravitation fields [8], although the detailed mechanisms cannot be the same, as will be discussed later. During the 31 parabolas available in this flight, we found five bubbles in the geometry of Figs. 4 and 5; in two of them with the needle as anode, the average velocities measured near the tip were initially  $\approx 30$  mm/s. After 0.4 s, the flow reduced to  $\approx 15$  mm/s (Fig. 4), and after 1.2 s to 9 mm/s. When the needle serves as cathode, the observed motion is much slower. The velocities were found to be in the range between 0.5 mm/s

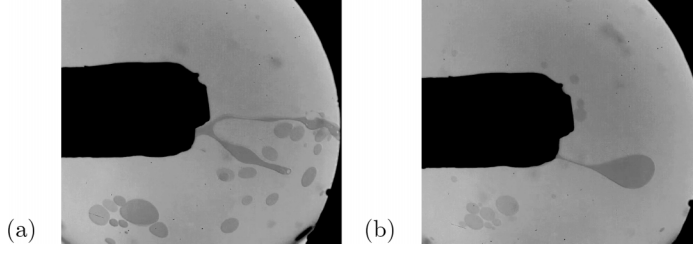


FIG. 6. Island deformation near the tip driven by a positive (a) and negative (b) potential of 1 kV at the supporting electrode. Image sizes  $3.2 \times 2.8 \text{ mm}^2$ .

and 1.5 mm/s. The acceleration of the flow can also be seen from the second camera perspective. The effects of acceleration are shown in Fig. 6. The film is accelerated at the tip, which leads to a transient deformation of islands. As seen in the two images, the effect is much stronger at the anode than at the cathode.

When a positive electric potential is switched off again, a remarkable effect is seen: The flow field reverses. This is visualized quantitatively in Fig. 7 with the same color-coding as in Fig. 5. The left images show the situation before the field is switched off. The times refer to the beginning of the color coding respective to the moment the field is switched off, each color scale covers 0.5 s from blue to red. It is seen clearly, in particular one second after the field is down, that the colors run clockwise from early (blue) to late (red). Again, the displacement of the global pattern indicates that it is not the islands relocating in the film but the complete film is flowing. After four seconds, this backward flow has ceased completely. With the negative potential, backward flow is practically absent.

*c. Symmetrically front-attached bubbles.* Finally, we describe the observations in frontally attached bubbles [Fig. 3(c), left image]. In that case, the system is axially symmetric, and this changes the effects and the structure of the flow qualitatively. The smectic film is now at rest, because flow away from the electrode tip is inhibited. The smectic film keeps its thickness and therefore the flow field has to be divergence-free. If the material would be driven away from the electrode in an axially symmetric way, the smectic film would have to dilute and create dislocations, which costs more energy than the electric forces provide. In the OASIS experiment [27], this symmetry was broken

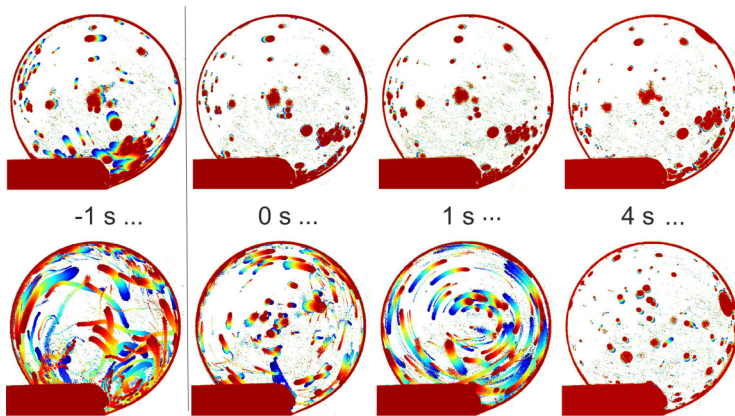


FIG. 7. Motion after a negative (top) and positive (bottom) potential of 1 kV is switched off at time  $t = 0$ . The brownish bar is the electrode needle. Times refer to both top and bottom sequences. The bubble diameter is 5.5 mm, color codes as in Fig. 5.

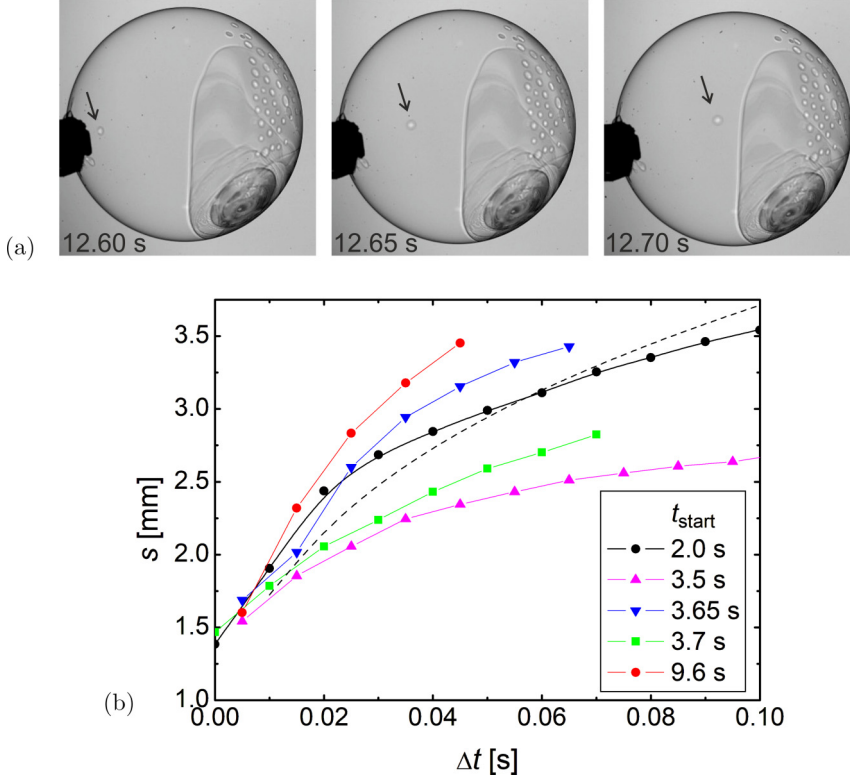


FIG. 8. (a) Individual droplet (marked by the arrow) ejected from the positively charged electrode at 1 kV in a frontally attached bubble. It detached approximately 12.59 s after the field was switched on. The droplets form a lattice when they reach the cap opposite to the electrode. Image sizes  $4.6 \times 4.6 \text{ mm}^2$ . (b) Measured positions of droplets versus time  $\Delta t$  after detachment at time  $t_{\text{start}}$ , where  $s$  is the path length on the bubble surface. Solid lines only guide the eye.

because of the different electrode arrangements. Thus, the field caused displacement of objects on the bubble.

Still, there are notable electric-field effects, at least when the bubble sticks to the positively charged electrode. The meniscus at the electrode tip becomes unstable and small droplets are formed. They detach from the meniscus and are repelled by the anode to move in the resting film. These droplets are flat lenses [9], the difference to islands is easily recognized optically. The droplets have no sharp borders, and they are brighter than the background film. The optical images indicate diameters of  $\approx 100 \text{ }\mu\text{m}$  and heights of the order of 250 nm, with only little dispersity.

We assume that these droplets move respective to the surrounding immobile film, and their velocities are governed by the balance of electric and viscous forces. Even though one cannot expect that all droplets carry the same charges, we find that all velocities are comparable. Figure 8(a) shows a typical droplet moving from the electrode tip toward the opposite side of the bubble through a uniform film region. At the right side, opposite to the electrode, an inhomogeneous, thicker film region is found. This thicker film region is repelled by the electric field and remains thus fixed as long as the bubble is in microgravity. Inside this region, the droplets are collected to form a rather regular hexagonal lattice. This side-effect is discussed below.

We note two important differences to the motion of islands in the previous geometry. The first one is that the film itself is not moving; only the objects on the film move. This is confirmed by the observations of the positions and arrangements of islands and film thickness inhomogeneities. Even

more important, the effect is time-independent; we see no trend in the speed of droplets ejected one or twelve seconds after the field was switched on. In contrast, the flow of the films in Figs. 4 and 5 ceases after a few seconds. There is still another difference when the field is switched off: in the frontally attached bubbles, the droplets do not flow back. After the end of  $\mu g$  and re-entry into the hypergravity phase of the parabolic flight, the islands and droplets sediment in the gravitational field at the bottom of the bubble.

#### IV. DISCUSSION AND SUMMARY

Before giving an explanation of the observed effects, we summarize the observations made in the previous section. We start with the bubbles that are attached symmetrically at the electrode tip. These bubbles show only marginal flow in the background film. However, droplets are ejected from the meniscus when the electrode is positively charged. The droplets move towards the opposite side of the bubble and are collected there in a quite-regular lattice. The ejection speed does not depend upon the time the field is on; there is no 'aging' effect. When the supporting needle served as cathode, the effect was not observed.

Our explanation is as follows. The effect is related to the well-known electrospraying [32,33] and electrospinning techniques [34,35]. The meniscus of the smectic film is strongly charged at the electrode tip. Then, the repelling electric forces overcome the surface tension that keeps the meniscus stable. The meniscus develops elongated extrusions, which detach from the meniscus to form the small droplets that are seen in Fig. 8(a). The repelling electric forces are counteracted by viscous forces that are roughly proportional to the velocity of the droplets [36,37]. In a simple model that disregards the angle between the local electric field and the film area, one would obtain  $v \propto s^{-2}$  and a solution  $s \propto \Delta t^{1/3}$ . Actually, the path of the droplets on the surface of the bubble is curved and the field is oblique to the local film plane, but still, the dashed curve in Fig. 8(b) qualitatively reflects the motion of the droplets. This process is active as long as there is material in the meniscus, which is much longer than the  $\mu g$  time set by the duration of one parabola. When the droplets reach the opposite side of the bubble, they are collected there. Being charged with the same sign, they repel each other and form a nearly hexagonal lattice that maximizes their mutual distances. A similar observation was made in experiments with smectic bubbles at the ISS [29]. An interesting detail of the present observation is that the curvature of the film surface is not large compared to the lattice constant as in Ref. [29]. This interesting geometry has been studied in colloidal crystals by several groups, e.g., Refs. [38–40]. Because of the limited number of droplets in our system, however, a quantitative analysis of the detailed arrangement and defect structures was not possible, it requires more systematic experiments.

Note that the droplets generated by electrospraying are in the smectic phase, in contrast to the lattices observed earlier [29] at the ISS, where the film was heated to the melting point and the droplets contain molten isotropic mesogen material.

The proposed explanation of the polarity effect is as follows: When the electrode acts as an anode, the bulky, relatively immobile cations remain in the smectic material and charge it positively, while the very mobile small  $Br^-$  anions may leave the film material and gather at the electrode. This adds to the charging of the meniscus material that causes the ejection of droplets. However, when the needle serves as the cathode, mobile anions can quickly move away from the electrode tip without taking along liquid crystal material. This reduces the electro-spraying effect. Finally, since no background film material is transported, a backflow effect is not observed when the field is switched off.

Ion mobilities are expected to be of the order of  $10^{-9} \dots 10^{-8} \text{ m}^2/(\text{Vs})$  [41,42]. In fields of more than  $10^5 \text{ V/m}$  near the electrode tip, ions can travel several millimeters during the parabolic flight experiments and thus charge the films locally if anion and cation mobilities differ substantially. This is expected here since the ion mobilities are related to diffusivity, which depends strongly on the size and 'bulkiness' of the charge carriers.

The situation is different when the bubble is attached asymmetrically at the tip, as shown in Fig. 4. Here, the charging of the film material leads to a similar electro-spraying phenomenon, but now it involves the complete smectic film in front of the tip, not only small droplets that detach from the meniscus. When the charged film material is blown away from the needle tip, it is replaced by other charged material. The entire film is thus set in convective motion.

Here, we also find an asymmetry of the two polarities of the needle. The induced flow is much stronger at an anode, and it is approximately one order of magnitude weaker at a cathode. The explanation is the same as above for the frontally attached bubbles. It is related to the different mobilities of cations and anions of the salt.

This field-induced convective motion gradually ceases over time. The flowing film gradually becomes charged. The limited  $\mu g$  time did not allow to follow the process over long enough time. After re-entry into hypergravity at the end of the parabola, the film shows irregular flow patterns caused by gravitation forces on the islands.

The asymmetrically attached bubbles exhibit an unexpected backflow effect after the field is switched off. We assume that it occurs because the unidirectional flow of film material causes a distortion of the film meniscus and extracts some material that was sucked into the needle by capillary forces. When the electric field ends abruptly, the meniscus recovers its equilibrium shape and the material is sucked back into the needle, causing a transient reversal of the flow direction.

It is also reasonable to assume that the flowing film gradually becomes charged, and the electric forces will be balanced when the complete smectic film is sufficiently charged, then the flow stops. This occurs on a time scale of few seconds. The reason for the polar asymmetry lies, again, in the different ion mobilities. When the bubble is attached to the anode, the film directly at the electrode tip will quickly be charged positively, and is thus repelled in the direction of the field. When the needle is negatively charged, the negative ions are repelled but carry much less film material because they are more mobile in the smectic fluid.

## V. SUMMARY AND OUTLOOK

The experiments performed in this study are unique and they open a new perspective on electric field effects in charged fluids. The distinguishing feature in our experiment is the quasi-two-dimensional local geometry of the fluid films, with lateral dimensions of several millimeters but thicknesses in the range of dozens of nanometers, almost six orders of magnitude smaller. It would be desirable to collect more data in new microgravity experiments, but parabolic flights are not only expensive, they also involve extremely long lead times. In potential future experiments, it would be possible to obtain quantitative data of the influence of viscosities by varying temperatures, film thicknesses, and the smectic film material. An investigation of the influences of electric field strengths and bubble sizes would also be useful. However, access to reflight opportunities is not at the discretion of the authors of this study.

Also, the quantitative investigation of the effect of salt concentration and the modification of the salt dopants could be very interesting. For instance, the interpretation presented here could be tested by adding salts that form large, bulky anions and small, mobile cations. It would be desirable for a quantitative interpretation to measure the charge transport properties in independent experiments. With our large salt concentrations, we can safely assume that electronic charge contributions which were measured in liquid crystals with time-of-flight experiments [43,44] can be neglected with respect to those of ionic charges [41,42,45–47]. However, quantitative data of ion mobilities for the salt used in our experiments are not available so far.

One could probably perform a modification of the experiment under normal gravity in horizontal flat films. The convective flow structures found in the sideward attached bubbles will of course be absent, they are inhibited by the film boundaries, but it might be possible to eject droplets with high-voltage electrodes inserted in flat films.

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## DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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