

Exploding drops on lubricated surfaces

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(Received 3 May 2025; published 20 November 2025)

This paper is associated with a video winner of a 2024 American Physical Society's Division of Fluid Dynamics (DFD) Milton van Dyke Award for work presented at the DFD Gallery of Fluid Motion. The original video is available online at the Gallery of Fluid Motion, <https://doi.org/10.1103/APS.DFD.2024.GFM.V2634768>

DOI: [10.1103/dbc6-n7s6](https://doi.org/10.1103/dbc6-n7s6)

In 1882, Lord Rayleigh established the criteria for the stability of an electrically charged drop [1]. An evaporating drop carrying charge Q undergoes Coulomb explosion upon approaching a critical radius—now known as the Rayleigh limit—where electrostatic forces overcome surface tension. At this limit, the drop emits fine jets to expel excess charge, a phenomenon now associated with electrospray. For a long time, Rayleigh's work remained a theoretical curiosity: it was only in the 1960s that experimental confirmations of Rayleigh's predictions trickled in [2,3], and it was not until 2003 that jetting was captured on camera for a 50 μm -sized ethylene glycol drop levitated using a Paul trap [4]. By now, Rayleigh's predictions have been extensively validated by theory [5–7] and experiments [2–4,8–11]—but mostly for levitating *micro*drops not in contact with any surface [12–15].

Charged drops are more widespread than commonly acknowledged. For example, raindrops are naturally charged with a typical $Q \sim 1$ pC per drop (with either polarity) [16]; in typical laboratory settings, conventional pipetting produces positively charged drops with $Q \sim 50$ pC due to contact electrification [17–19]. Yet, surprisingly, there has been no reported instance of Coulomb explosions for evaporating drops on solid surfaces, even though the drops are likely to be charged. This paradox can be explained by the presence of contact-line pinning on surfaces which not only restricts the drop's free movement but also facilitates charge transfer between the evaporating drop and the underlying surface [20,21], effectively preventing Coulomb explosions.

Recently, we observed sustained, periodic Coulomb explosions (>60 cycles over 30 min) in sessile drops on a lubricated surface. Our experimental setup is strikingly simple: a conventional micropipette deposits a 1 μL deionized (DI) water drop onto a polymethylpentene (PMP) petri dish coated with a nanometric silicone oil film [Fig. 1(a)]. Through contact electrification with the plastic pipette tip, the drop acquires an initial charge of $Q = 100$ pC as measured using a Faraday cup [17–19]. The addition of a thin oil film eliminates contact-line pinning [22–24], enabling the drop to adopt a geometry dictated solely by surface tension and electrostatic forces.

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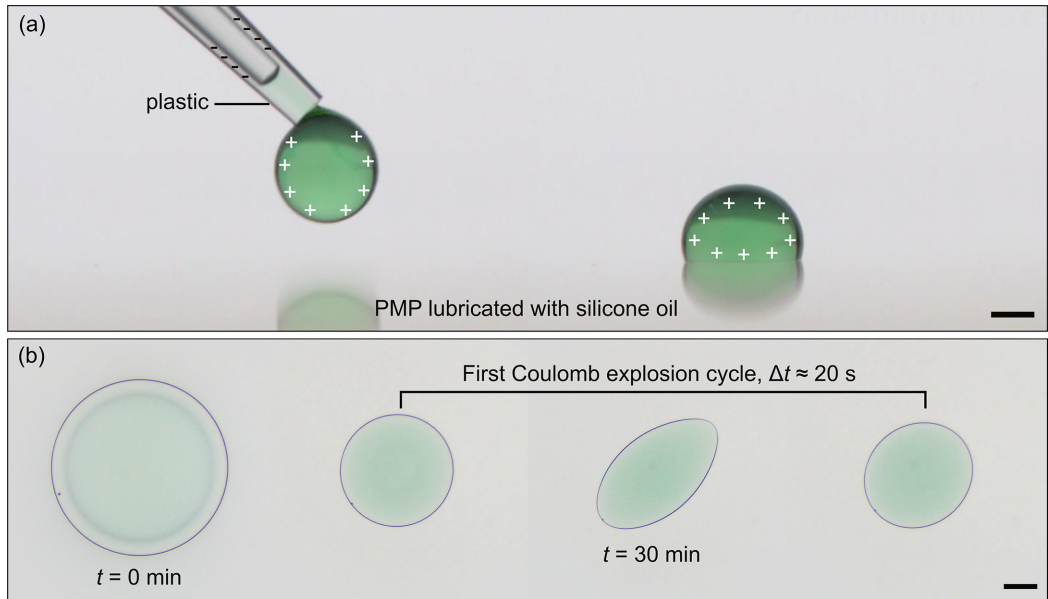


FIG. 1. Spontaneous Coulomb explosion of an evaporating drop on a lubricated surface. (a) A $1\ \mu\text{L}$ water drop, charged via conventional pipetting, is deposited on a lubricated surface. Food dye is added for visualization. Scale bar is 1 mm. (b) Top view of the same water drop undergoing Coulomb explosion. As the drop evaporates and shrinks to a critical size ($t = 30$ min), the first explosion cycle is triggered. Scale bar is $200\ \mu\text{m}$. The video associated with this image is available at <https://doi.org/10.1103/APS.DFD.2024.GFM.V2634768>

The initial phase of evaporation ($t < 30$ min) is relatively uneventful, with the drop shrinking isotropically [Fig. 1(b)]. As the drop evaporates, the charges become more concentrated, and the increasing charge density drives the system toward the Rayleigh limit. At $t = 30$ min, the drop reaches its first critical size, triggering the Coulomb explosion cycle [Figs. 1(b) and 2(a)]. The drop first elongates to an ellipsoid, reaching maximum elongation within ~ 10 s [Fig. 1(b)]. At this point, electrospay occurs, releasing excess charges, enabling the mother drop to spring back to a circle [Fig. 1(b); see video for full sequence]. Remarkably, this cycle repeats with high periodicity, occurring more than 60 times during the subsequent 30 min of observation.

Electrospray is observed each time the charged drop reaches maximum elongation, with a sharp conical tip (Taylor cone) forms at one pole of the ellipsoid, as captured by high-speed imaging [Figs. 2(a)–2(c)]. One conspicuous feature of the phenomenon described here is the great diversity of length scales (microns to millimeters) and especially of timescales (microseconds to minutes).

The entire evaporation process takes place for half an hour or more, with each explosion cycle lasting for tens of seconds (see video for full sequence). At the microscopic level, jets are formed within a millisecond [Fig. 2(b)], which then disintegrate into microdroplets within microseconds [Fig. 2(c)]. From the smallest timescale to the largest is a span of some nine orders of magnitude. These charged microdroplets mutually repel upon exiting the oil meniscus, initially moving at $\sim 1\ \text{mm s}^{-1}$ before slowing to $\sim 0.1\ \text{mm s}^{-1}$, traveling distances far exceeding their own size before fully evaporating [Fig. 2(a)].

To demonstrate that the presence of charge is critical, we deposited a water drop onto the lubricated surface using an earthed metal needle, thereby ensuring zero initial charge [Fig. 3(a)]. The uncharged drop evaporated without pinning, maintaining a perfect spherical geometry [Fig. 3(b); see video for full sequence]. Notably, even as the drop shrank to minimal sizes over a 60-minute period, no Coulomb explosions were observed—confirming that the presence of electrical charge is a necessary condition for this behavior.

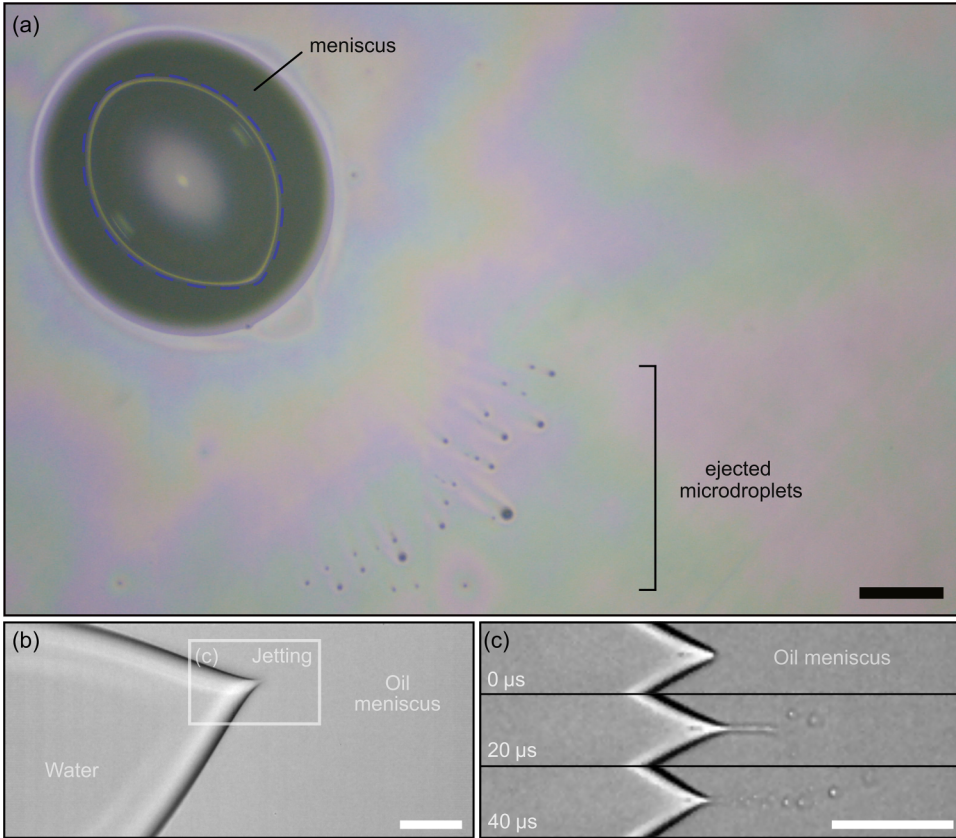


FIG. 2. Electro spray event during Coulomb explosion in a charged drop. (a) The charged microdroplets exit the oil meniscus, repel one another, and fan out reminiscent of electrosprays. They also travel distances a few times the drop size. The image was captured with a digital camera under diffused white light, revealing colorful thin-film interference patterns. These patterns arise from submicrometric variations in the lubricant's thickness [25]. Scale bar is 200 μm . (b) Microscopic view of jet formation from a Taylor cone, (c) which disintegrates into microdroplets within a few microseconds, and forms the spray in (a). Scale bars in (b) and (c) are 50 μm . The video associated with this image is available at <https://doi.org/10.1103/APS.DFD.2024.GFM.V2634768>

In this work, we have shown how electrospray can be spontaneously generated on charged sessile drops on a surface using only commercially available materials (PMP petri dishes and silicone oil), remarkably without requiring the ~ 10 kV potentials needed in conventional electrospray systems. By simply varying drop composition, the platform can potentially be repurposed for micro-/nanopatterning, portable ion sources, and on-demand nanoparticle synthesis.

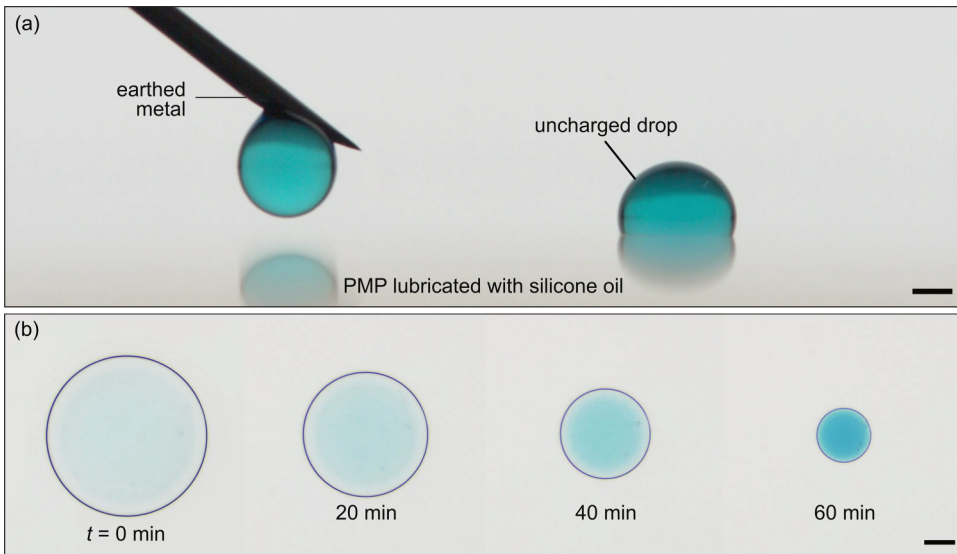


FIG. 3. Evaporation of an uncharged water drop on a lubricated surface. (a) Initial deposition of a $1\ \mu\text{L}$ water drop using an earthed metal needle, ensuring no initial charge. Blue food dye is added here for visualization. Scale bar is 1 mm. (b) Top-view time-lapse showing evaporation of the same drop over 60 min in the absence of contact line pinning and maintained spherical geometry. Scale bar is $200\ \mu\text{m}$. The video associated with this image is available at <https://doi.org/10.1103/APS.DFD.2024.GFM.V2634768>

This work was supported by KAUST start-up fund BAS/1/1416-01-01.

Data availability. The data are not publicly available. The data are available from the authors upon reasonable request.

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