INVESTIGATION INTO THE COUPLING BETWEEN PLASMA-INDUCED CAPILLARY OSCILLATIONS AND STREAMER SELF-ORGANIZATION INSIDE BUBBLES IN WATER

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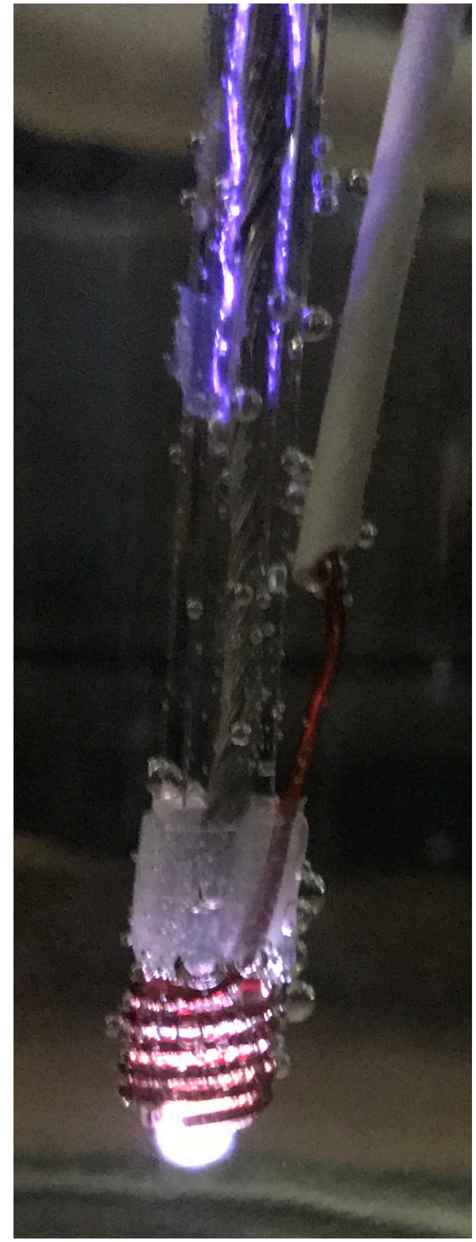
MOTIVATION

- **There is increased interest in applications of low temperature atmospheric pressure plasmas in liquids**
- **These technologies rely on the production and transport of plasma-derived reactive species into bulk liquid**
- **A common method of injecting plasma-derived reactive species into liquids is gas phase discharges in bubbles**

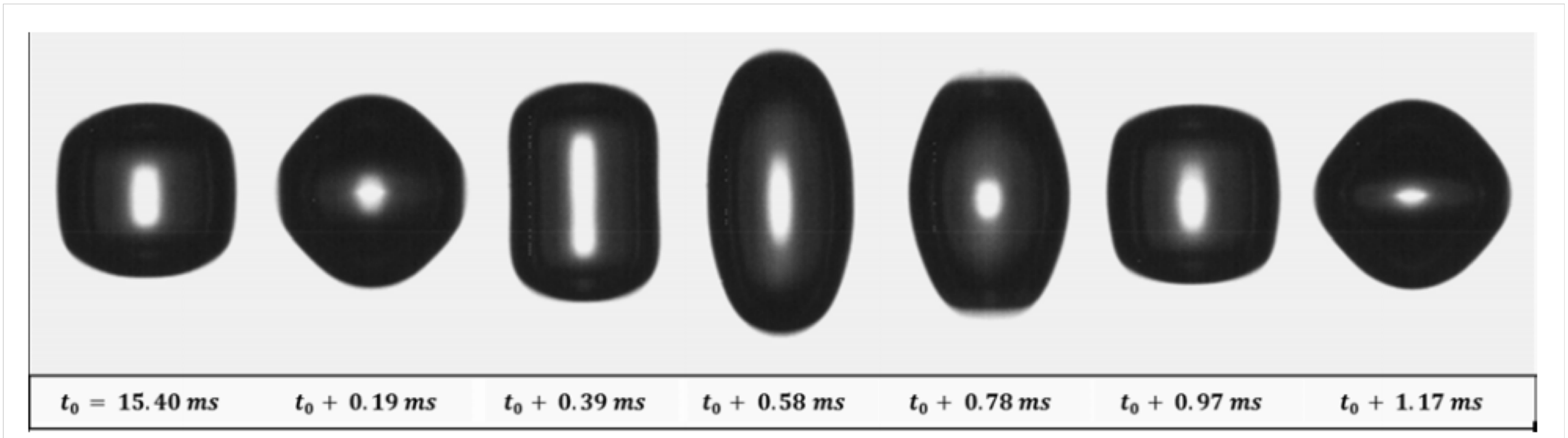
(left) Decomposition of methylene blue at various plasma treatment time

(right) Plasma jet in water



MOTIVATION

- The interaction between plasma and liquids can also initiate fluid dynamical effects at the interface, and possibly drive large-scale convection in solutions [1,2]
- While capillary waves on bubble surfaces have been observed [1], its effects on fluid flow and streamer formation are of interest



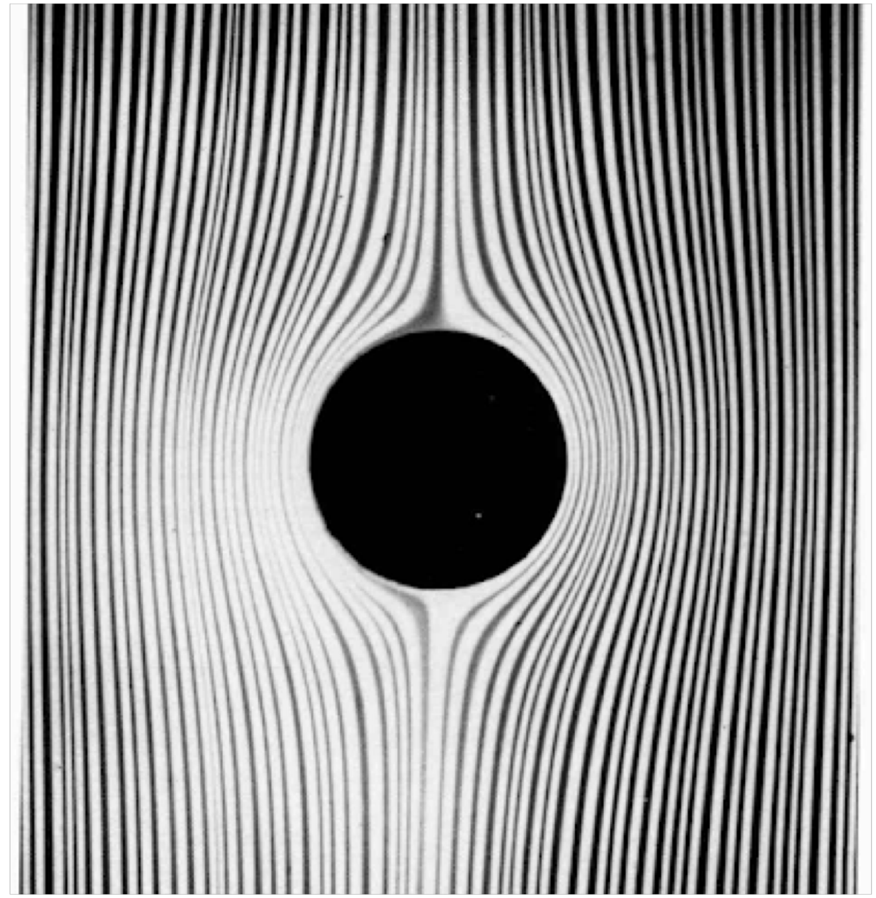
Spherical harmonic perturbation of spherical bubble [1]

[1] J. Phys. D: Appl Phys (2012) 45, 415203

[2] IEEE Trans. Plasma. Sci. (2016) 44, 1127

MOTIVATION

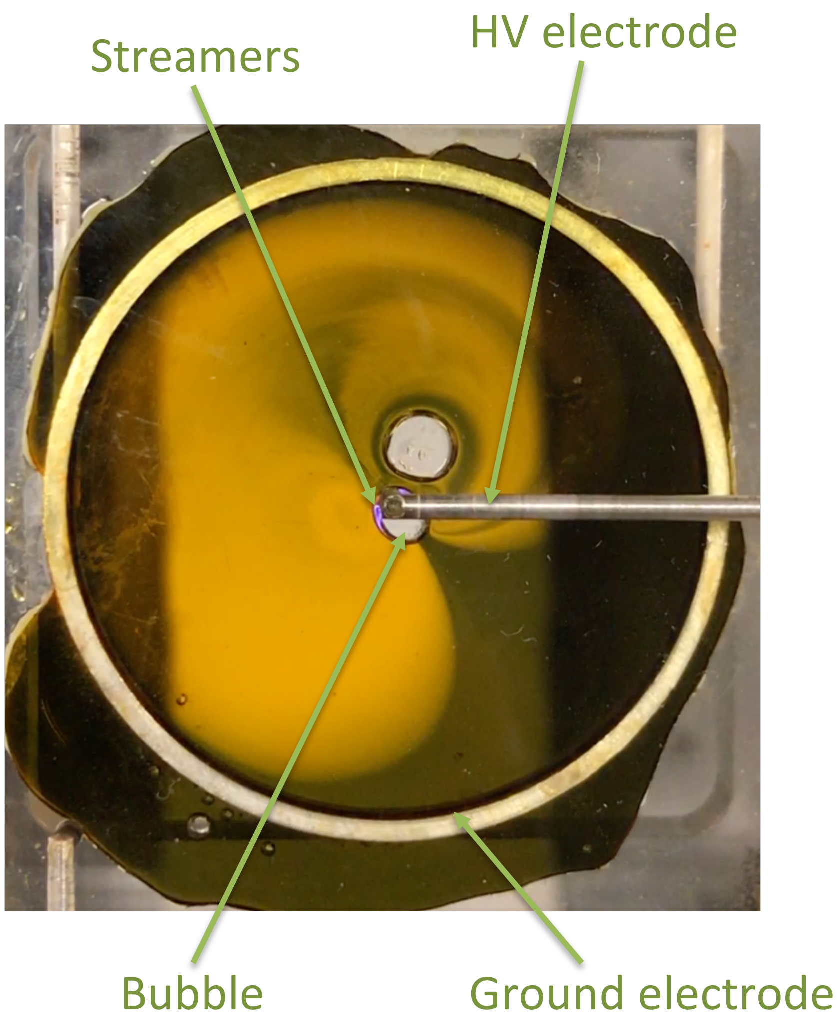
- A better understanding of forces at the interface gives insight to complex physics and chemistry in plasma-liquid interactions
- To render the interface available for optical interrogation, a 2-D Hele-Shaw-like cell was employed
  - A thin layer of liquid (~1 mm) is confined between two plates
  - 2-D bubble is produced in the center of fluid layer
  - Both plasma and liquid regions are accessible for optical measurements



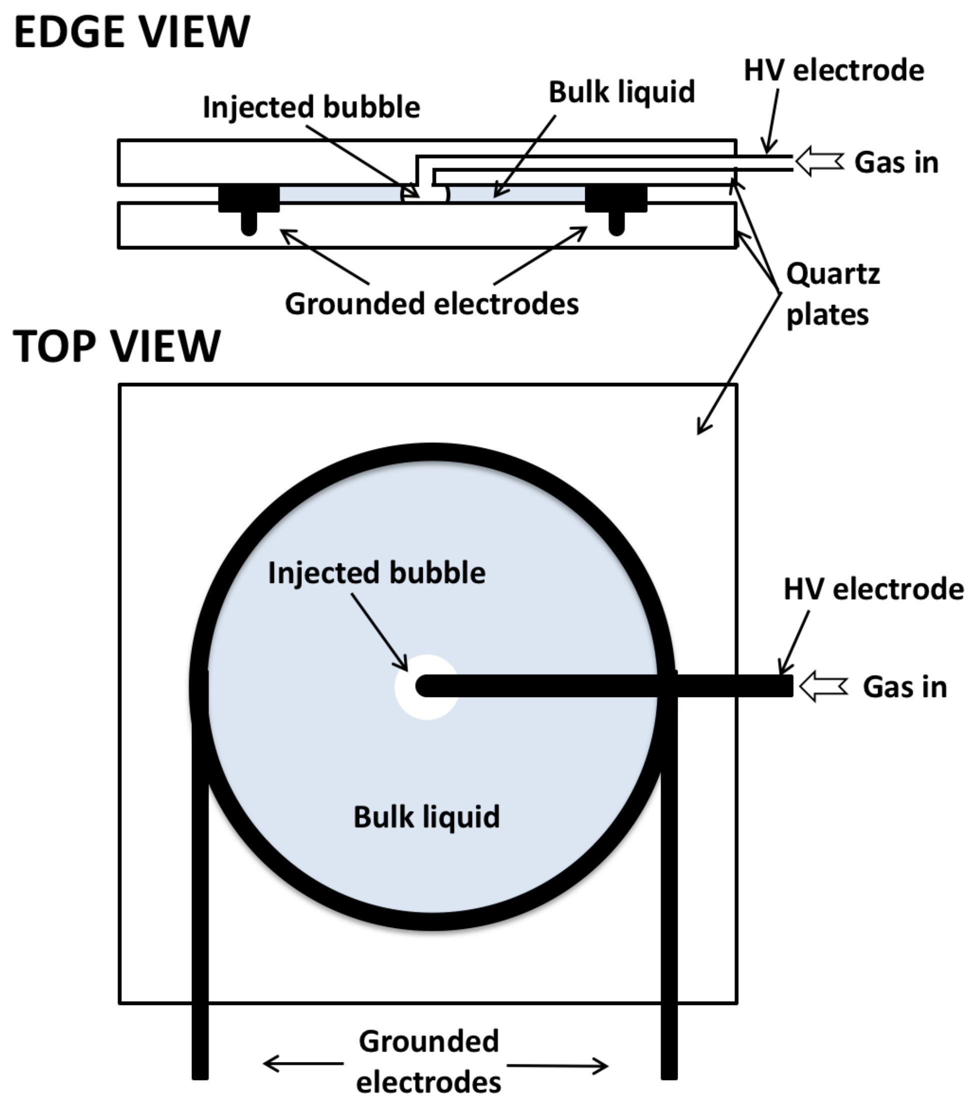
Hele-Shaw flow past a circle [1]

[1] http://luxsignifer.blogspot.com/2016/10/hele-shaw-flow-past-circle.html

EXPERIMENTAL SETUP

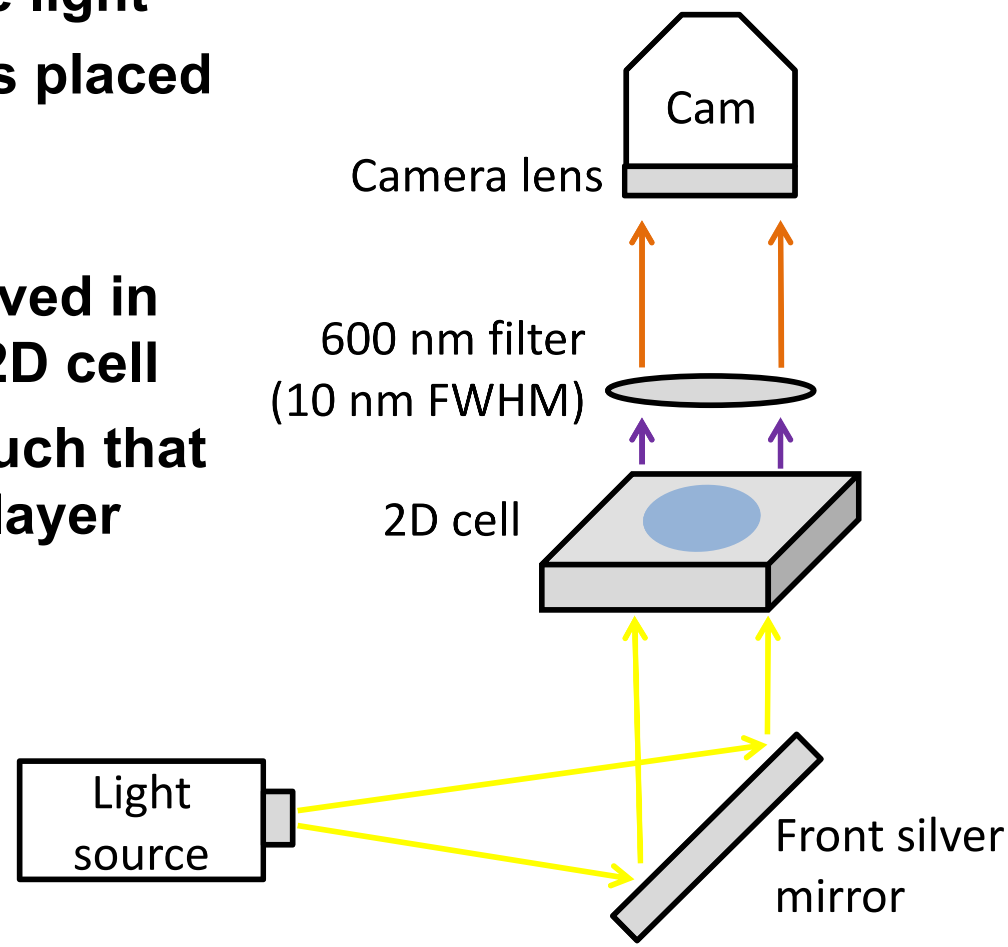


Plasma-induced chemical front in thymol blue solution



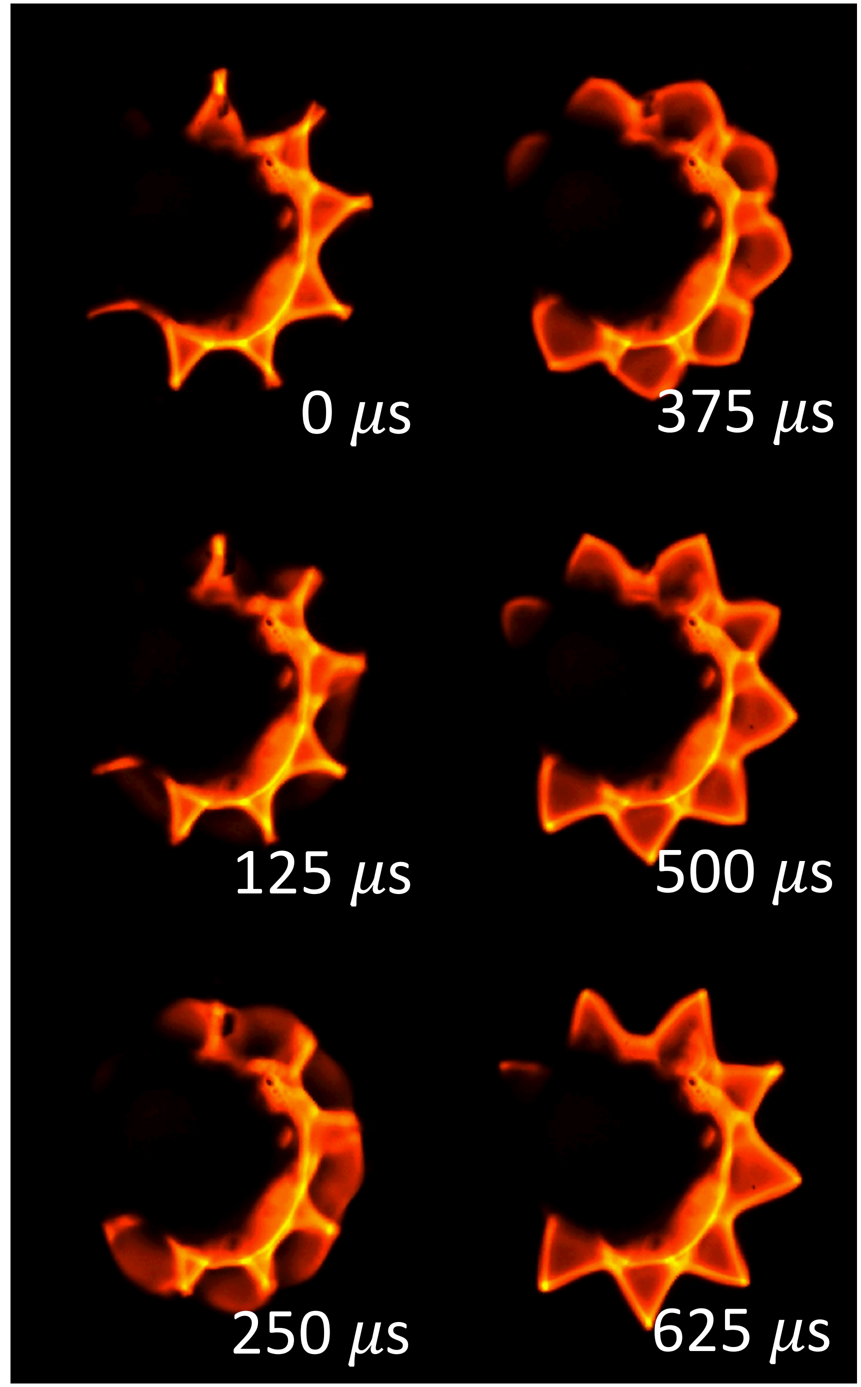
IMAGING OF AIR CAVITY OF BUBBLE USING INDIGO DYE

- **2-D cell was backlit with a white light**
- **A 600 nm (10 nm FWHM) filter is placed between cell and camera**
- **Indigo trisulfonate, a dye with absorbance at 600nm, is dissolved in deionized water and placed in 2D cell**
- **Backlight intensity is chosen such that bubble cavity is lit while liquid layer appears dark**
- **Imaging frequency is at least 4 times the excitation frequency of plasma**



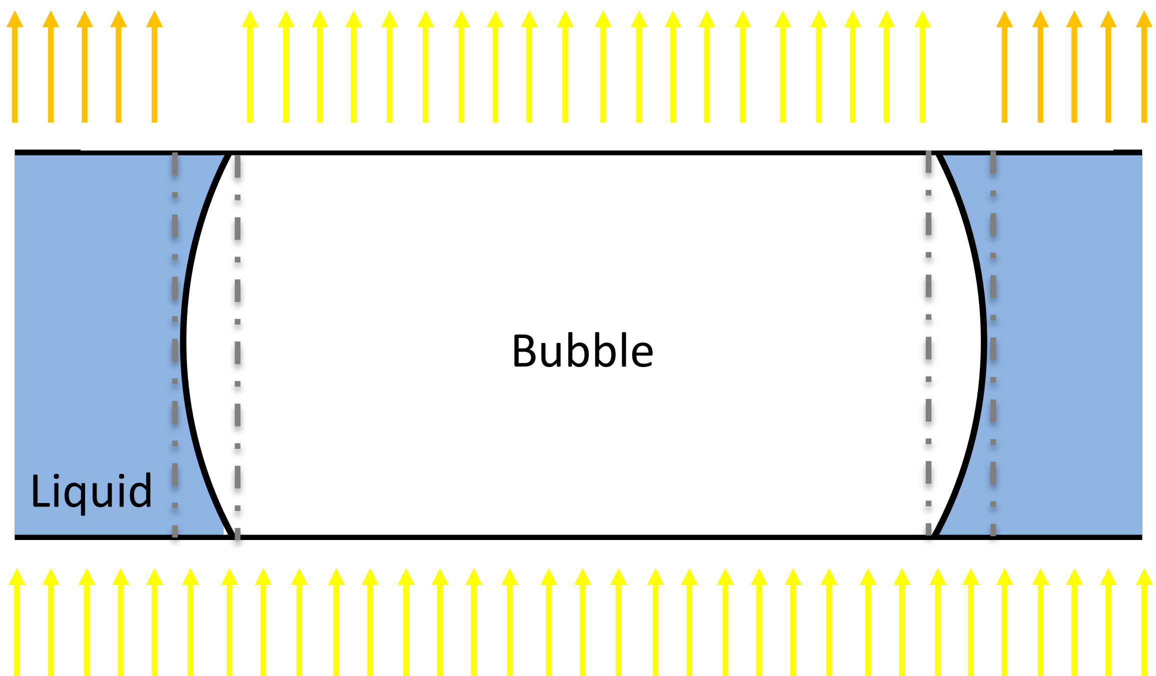
BUBBLE CAVITY DEFORMATION

- 20 kV is pulsed at 1 kHz
- Images are captured at 8000 fps (125 μs between frames)
- The orange lit region in the images represents the bubble cavity
- 9-pointed star polygon shape can be inferred
- Peaks oscillate at a frequency of 500 Hz, regaining their initial amplitudes in the duration of 2 excitation pulses



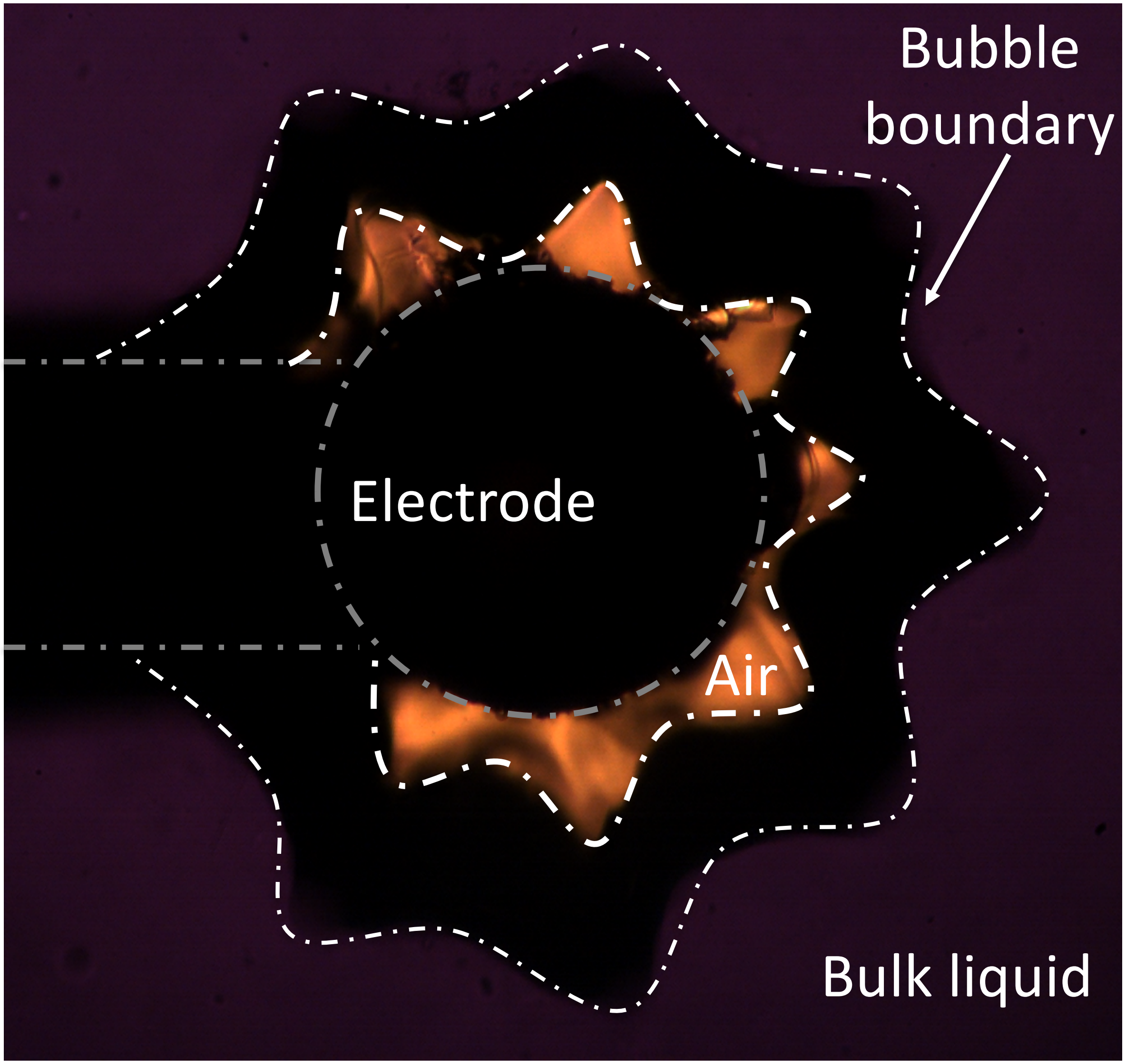
IMAGING OF CAPILLARY WAVE ON BUBBLE BOUNDARY

- 2D cell is backlit with white light
- Light traveling through bulk liquid and air bubble is unperturbed, appearing bright
- Slight curvature of the bubble boundary refracts light away, thus appears dark
- If a colored solution is used in the 2D cell, the bubble region might appear to be of a different color from the liquid region



Cross section of 2D cell, side view of bubble and surrounding liquid region

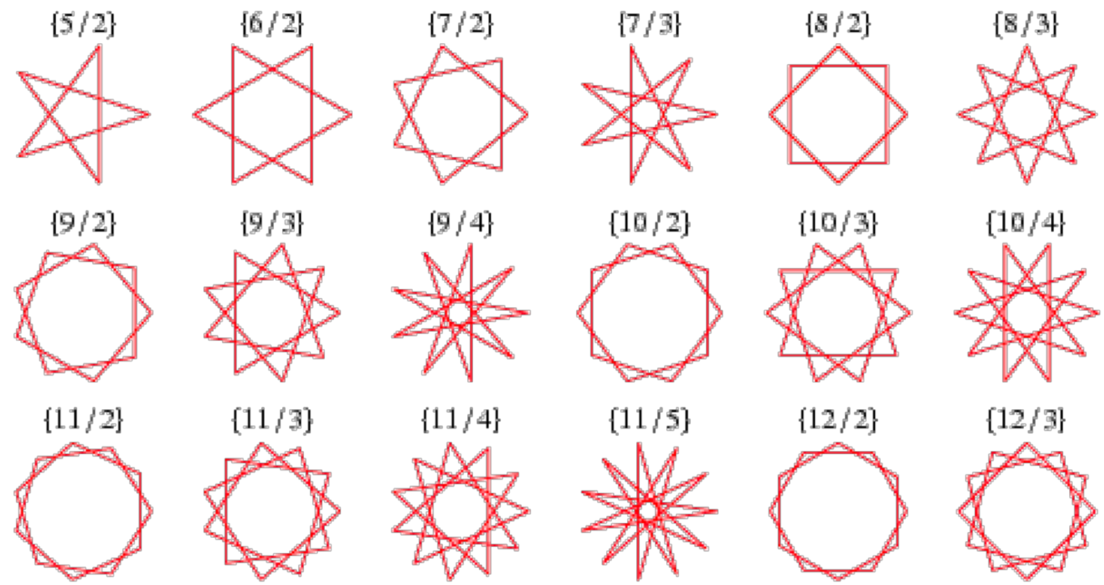
BUBBLE BOUNDARY PERTURBATION



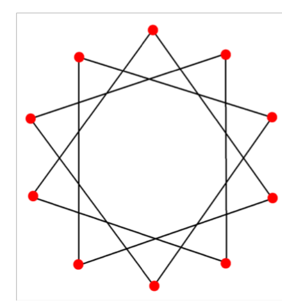
- **Bulk liquid appears purple as indigo solution is backlit**
- **Bubble cavity region appears orange**
- **The bubble boundary appears dark, but its perturbation matches the bubble cavity deformation**

POLYGRAM

- A regular polygram {n/k} is a polygon of n sides, where every vertex i is connected to vertex i+k
- When k = 1, the polygon is convex, where all interior angles are less than 180 degree

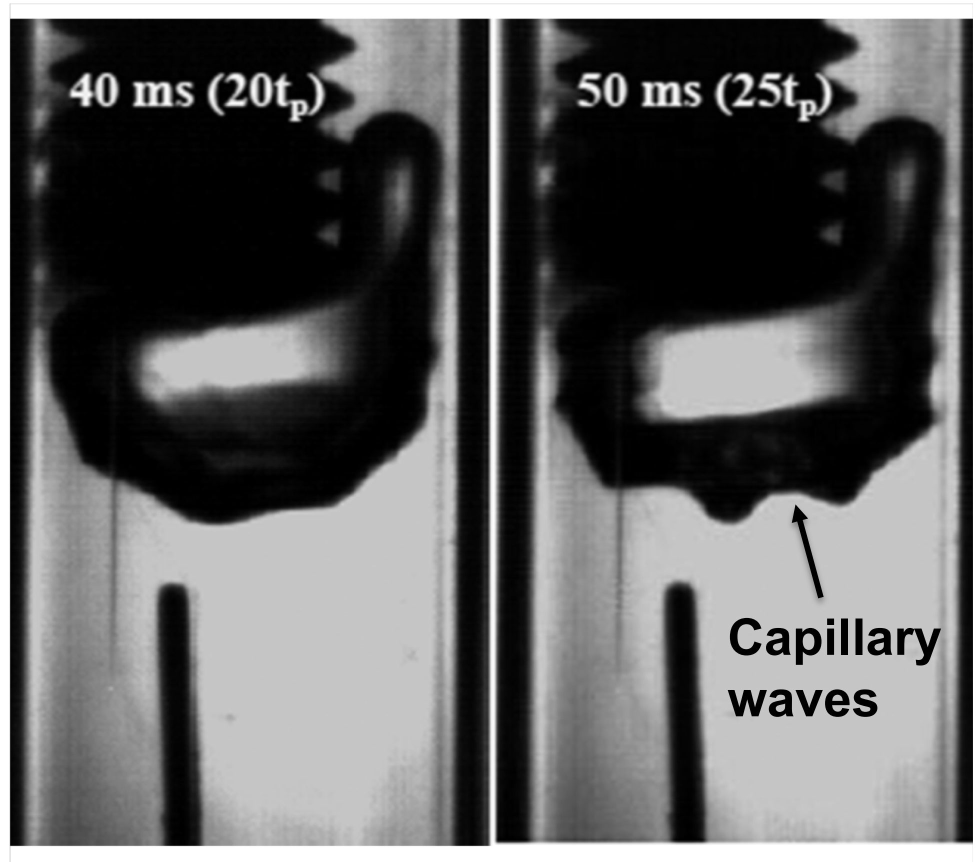


- For k > 1, the polygon is non-convex, where one of the interior angle is larger than 180 degree
- When intersecting lines are removed, simple concave isotoxal 2n-gons can be formed, represented as $\{n_\alpha\}$, where α = internal angle



PLASMA-INDUCED CAPILLARY WAVES IN ELECTRODE-ATTACHED BUBBLES

- Capillary waves were previously observed in bubbles in contact with biased threaded rod in water [1]
- Surface streamers propagating inside air bubbles excite capillary waves at the liquid-gas interface of the bubble
- Subsequent streamers were observed to closely follow the surface distortions
- Resonant behavior was observed as oscillation amplitude increased over several pulse periods

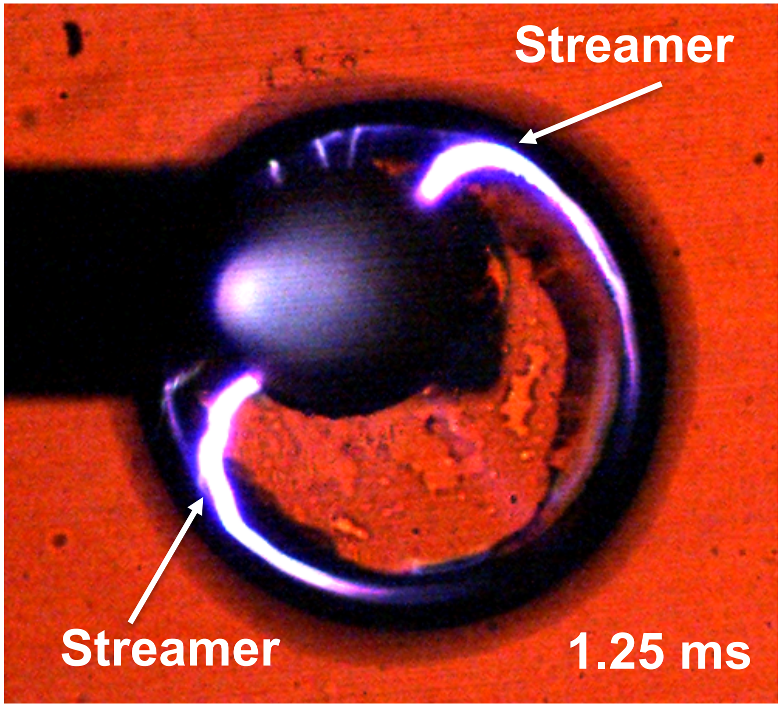


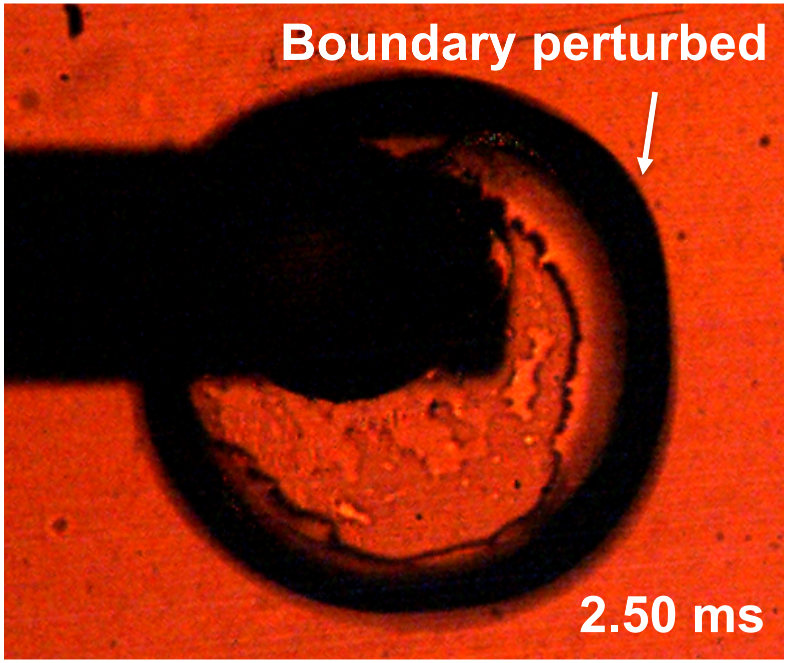
Bubble oscillations driven at 500 Hz [1]

[1] J. Phys. D: Appl Phys (2011) 44, 082001

FORMATION OF CAPILLARY WAVE

- Frequency of 60 ns 20 kV pulse: 200 Hz
- Imaged at 800 fps
- Salt solution with conductivity of 12.9 mS/cm
- First streamer excitation is shown on the right
- Streamers slightly perturbed bubble boundary at the contact point
- This is attributed to impulse of streamers

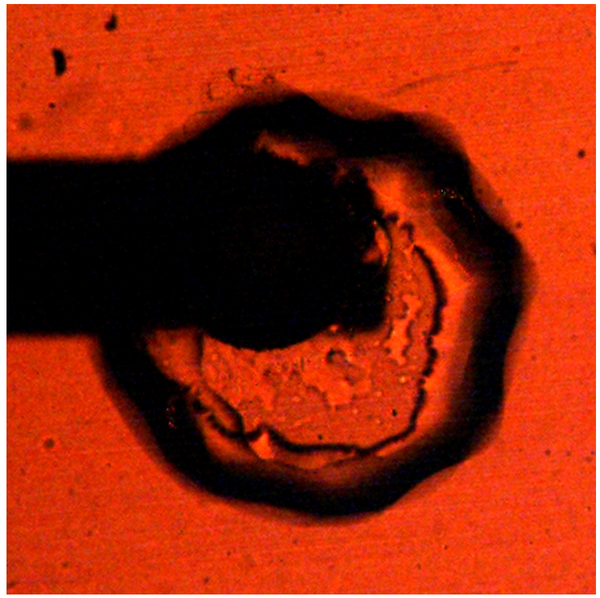




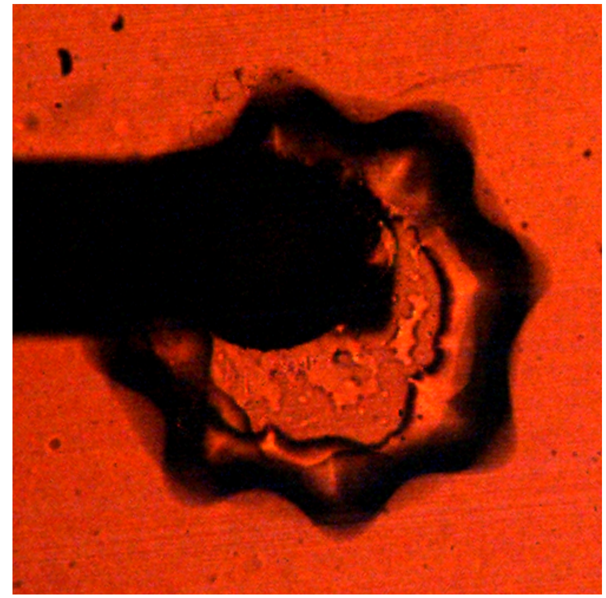
DEVELOPMENT OF CAPILLARY WAVE

- Each pulse progressively perturbs the bubble boundary, increasing the amplitude of the oscillation

Initial
t = 0 ms



1.25 ms after 10th pulse
t = 47.5 ms

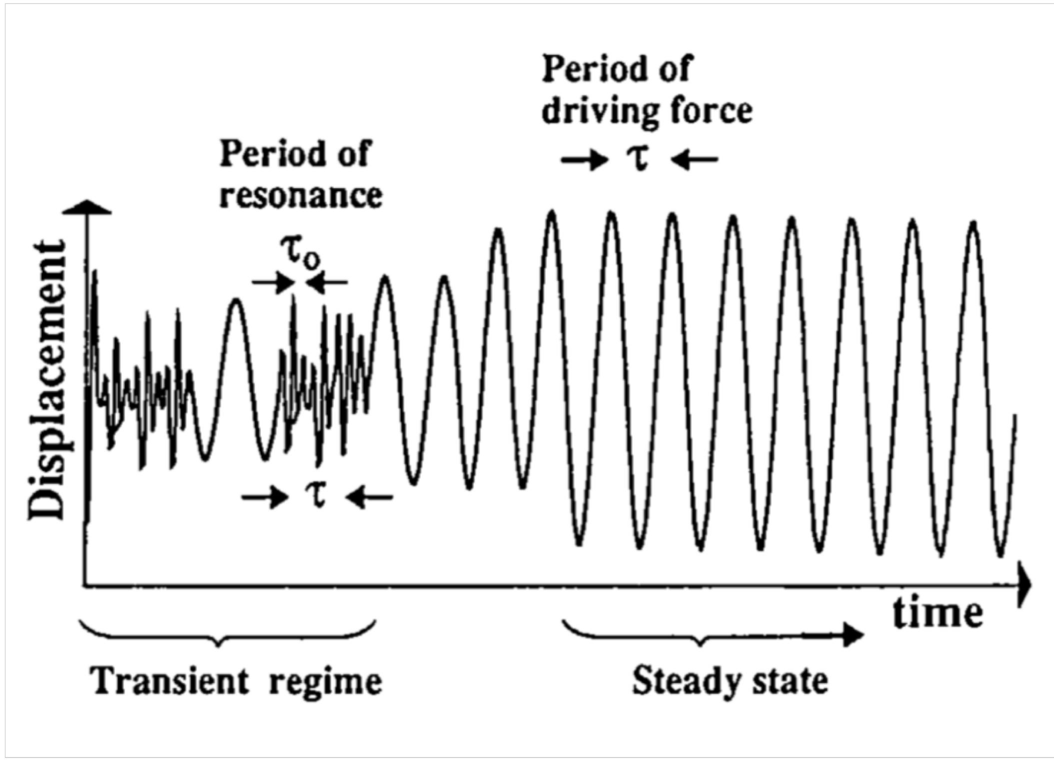


1.25 ms after 11th pulse
t = 52.5 ms

- Local perturbations slowly initiate capillary waves that travel along on the bubble surface, developing into a n = 10 capillary wave mode after approximately 11 pulses

FORCED DAMPED OSCILLATOR

- Small amplitude pulsation of bubble can be thought of as a forced damped linear oscillator
- In this case,
  - Driving force: Streamer impulse
  - Damping force: viscosity, surface tension
  - Oscillator: bubble boundary
- Qualitatively, the oscillator initially responds according to its resonance frequency (ω_0) and the driving frequency (ω), eventually reaching its steady state (driven at ω)



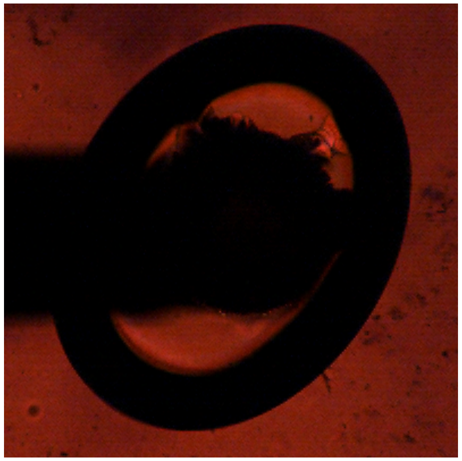
Schematic of transient and steady-state response of oscillator (Acoustic bubble, 1994)

EFFECTS OF FREQUENCY ON CAPILLARY WAVE MODES

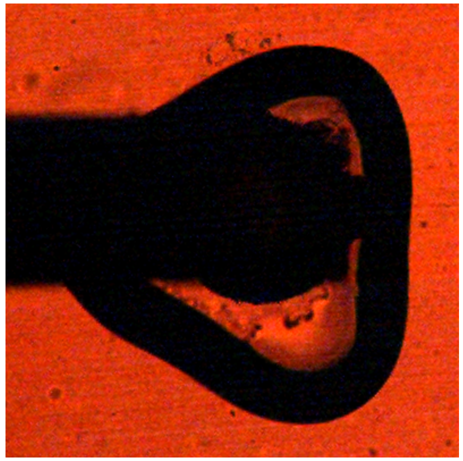
- Excitation frequency ranges from 100 Hz to 1 kHz
- Bubble boundary was perturbed, with observed modes ranging from n = 2 to n = 12
- At frequency under 100 Hz, bubble remains unperturbed and thus circular
- There is loose correlation between excitation frequency and observed steady-state capillary wave modes

Frequency (Hz)	Steady-state wave modes
100	2
200	3,4
500	6
800	8
1000	8,9,10,12

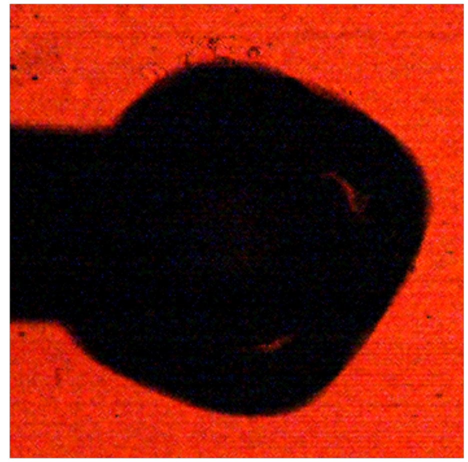
CAPILLARY WAVE MODES



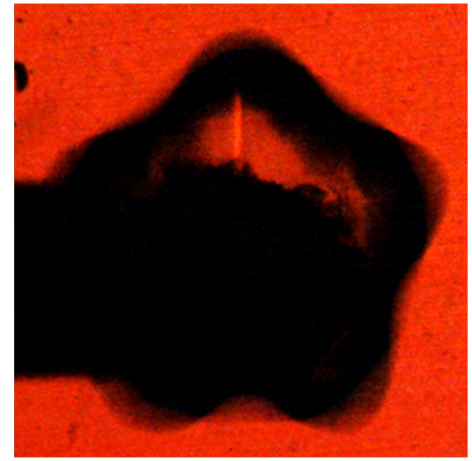
n = 2, f = 100Hz



n = 3, f = 200Hz

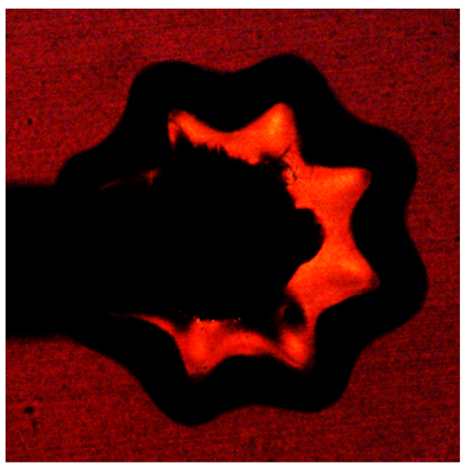


n = 4, f = 200Hz

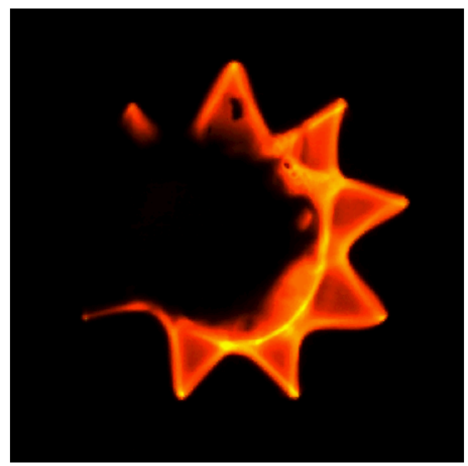


n = 5, f = 200 Hz

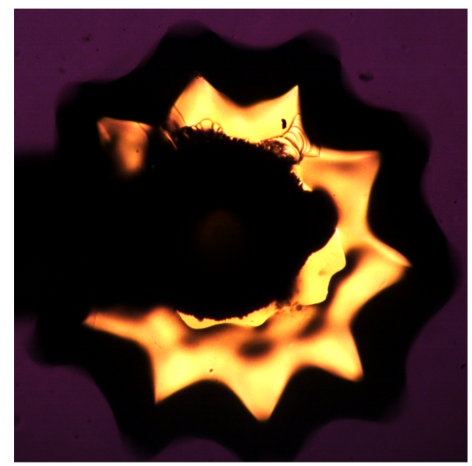
n = 6, f = 500Hz



n = 8, f = 800Hz



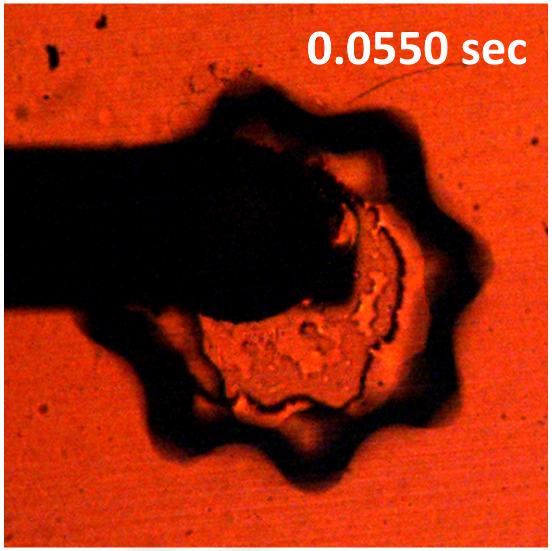
n = 9, f = 1kHz

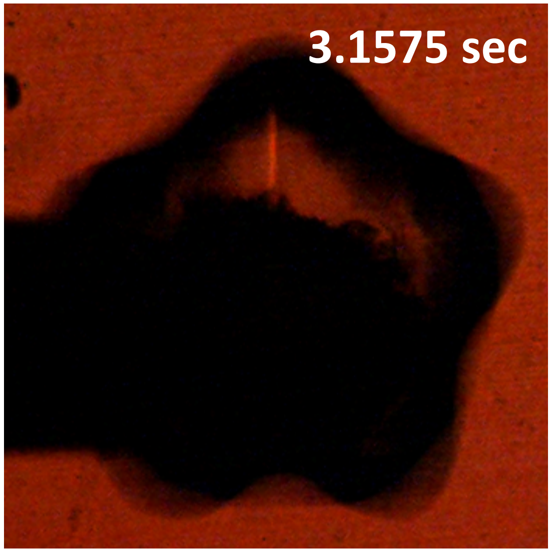


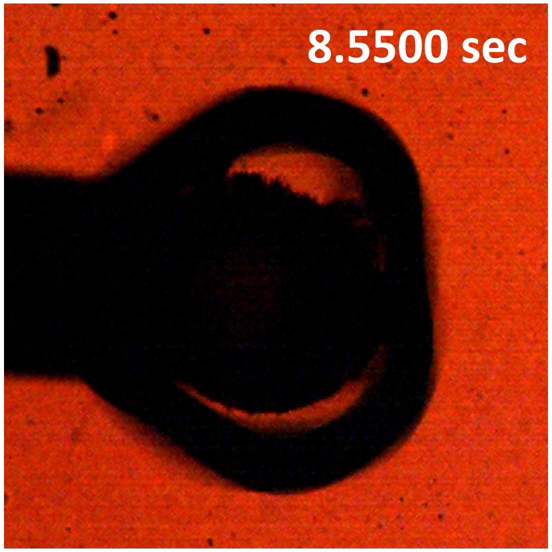
n = 10, f = 1kHz

TRANSITION BETWEEN TRANSIENT AND STEADY-STATE RESPONSE

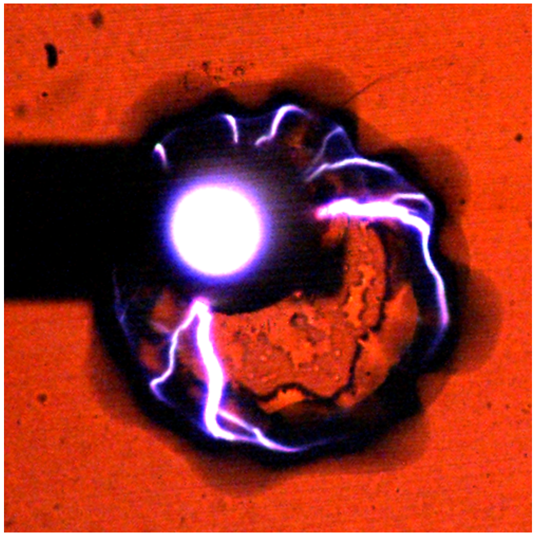
- Given the driving frequency of 200 Hz, we observed the steady-state mode n = 3 to be dominant
- Initial formation of capillary waves are often resonant modes of higher n (n = 8 here)
- During the transient regime, the bubble can transition through multiple modes (n = 4, 5, 6)
- Eventually it relaxes into its steady state mode (n = 3)





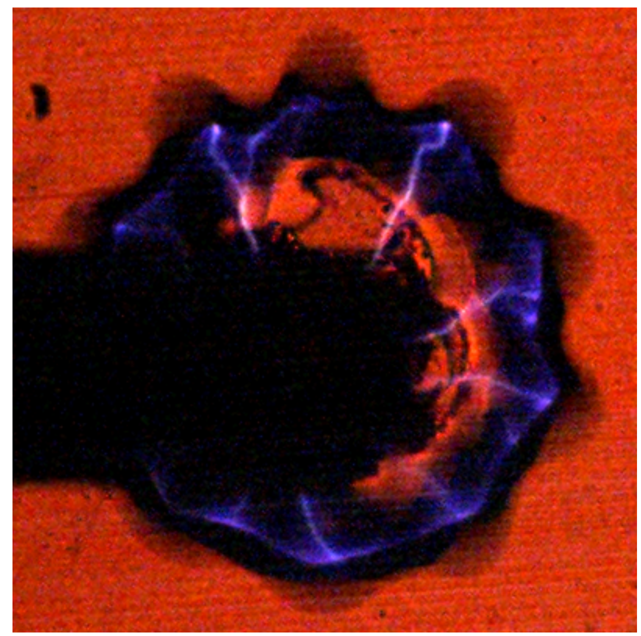


SELF-ORGANIZATION OF STREAMERS

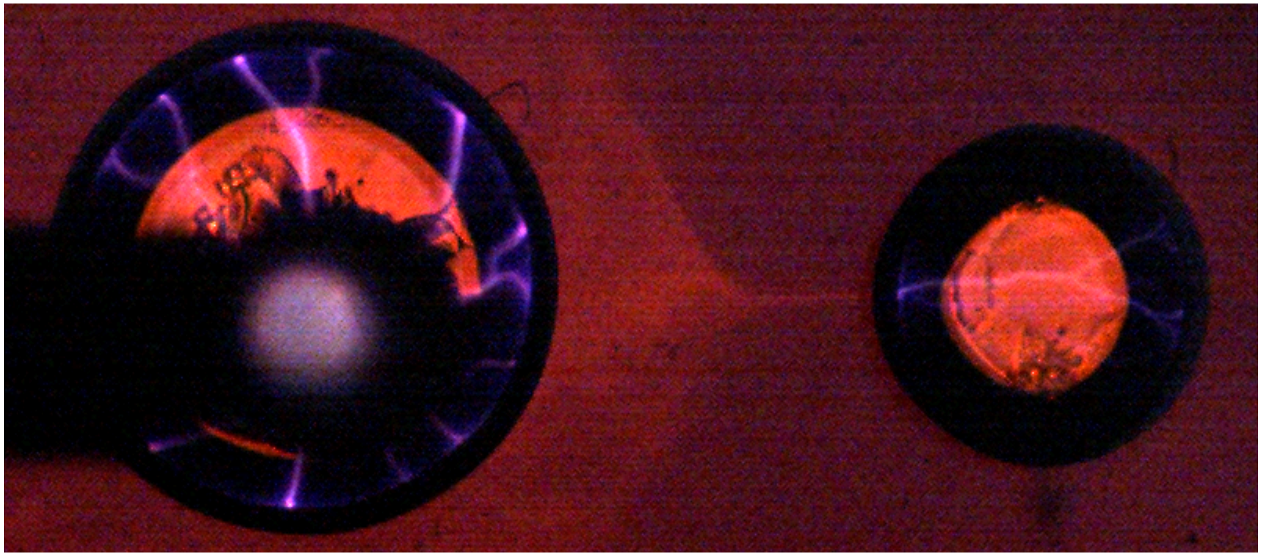


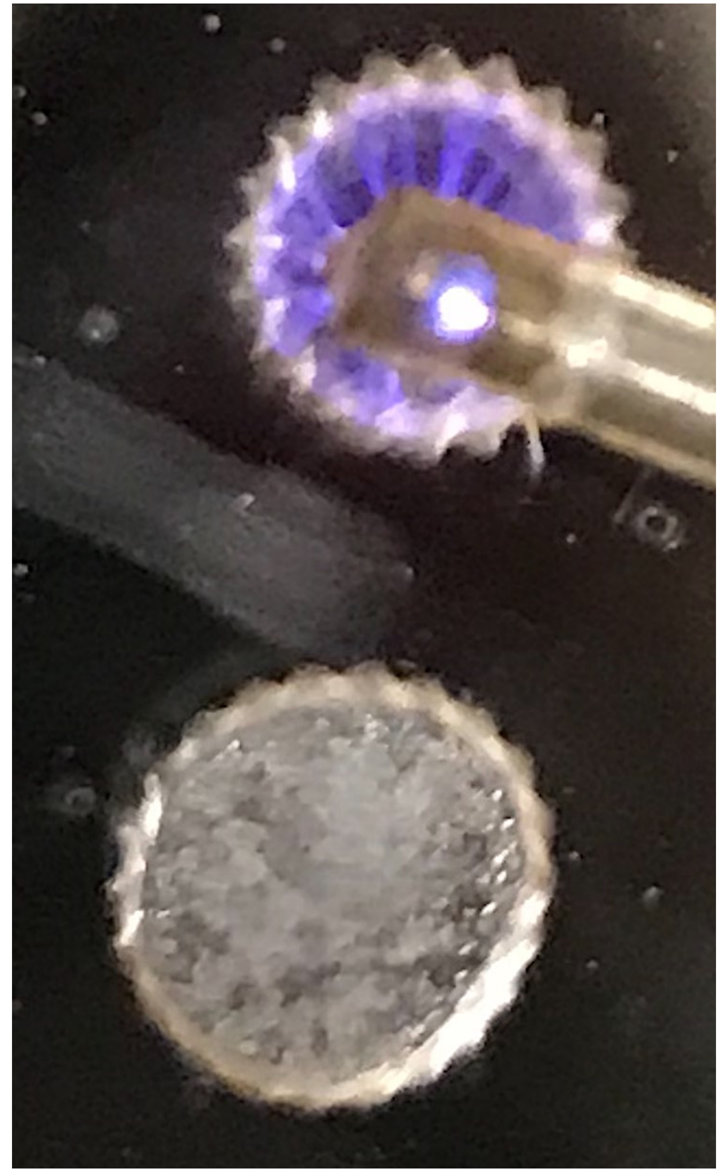
- For an asymmetrical bubble, where high voltage electrode is not centered, streamers reaching for surface initiate surface ionization waves on interface
- These ionization waves travel along the perturbed interface

- For a symmetrical bubble, where high voltage electrode is centered, streamers touch the interface at the points
- Possibly due to these points representing locations of shorter gaps before pulse
- The self-organization of streamers provides positive feedback for continuous capillary wave propagation



EFFECTS ON SECONDARY BUBBLE





- **Streamers and capillary wave pulsation can develop in an adjacent secondary bubble**
- **In the absence of discharges in secondary bubble, capillary waves can still develop, indicating pressure field propagating through bulk liquid**

CONCLUSION

- Capillary waves were formed on bubble surface when streamers were excited inside a 2-D bubble
- A positive correlation between excitation frequency and induced capillary wave modes was observed
- Capillary wave development can be attributed to momentum transfer from streamer impact
- There is a transient regime where resonant modes are excited before steady-state is achieved
- Capillary waves can be excited in secondary bubbles in the bulk liquid
- Self-organization of streamers are observed inside bubble, possibly due to positive feedback from capillary wave excitation
- Characterization of capillary wave excitation in bubbles can give insight into the optimization of reactive species production at the plasma-liquid interface