

Experimental Notes: Surfactant-driven fracture of particle rafts.

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These are notes on easy ways to explore experiments on surfactant dynamics and how they influence particle rafts at air-water interfaces using inexpensive, homely items available in the kitchen. These notes are not aimed at the specialist, but are meant for a curious layperson or a student entering science. For ICTP students, please treat this as an introductory notes, there will be changes to the experimental protocol as we can do a better job than what one can manage in one's kitchen.

INTRODUCTION

Background

Liquids flow readily under application of some external force and yet retain stable shapes of the container they are held within under equilibrium conditions. A quiescent liquid in a container left open at the top to the ambient environment is bounded by the container walls on all sides except its top surface exposed to the air. This top surface of the liquid is generally flat for the most part, save in the region where the air-liquid surface comes in contact with the walls, where it forms a tiny lip we call a “*meniscus*”, we will discuss it later. What gives the liquid surface exposed to ambient environment a stable shape is a property we call *Surface Tension*. This property allows the air-liquid interface to behave very much like a stretched, elastic membrane that opposes distortion and takes on a shape that minimizes the surface area exposed to the ambient environment.

The macroscopic property of Surface Tension we measure at an interface has a microscopic origin like most properties in Condensed Matter & Statistical Physics. Consider Figure 1a which shows an approximately hemispherical drop of water resting on a solid surface and is exposed to air above. Within this droplet, we show a cartoon illustration of water molecules at a given instant in time. We notice that a given molecule residing in the bulk is attracted to all its neighbors due to the interaction posed by chemical bonds, since they act from all directions in the bulk they cancel each other out. However, molecules residing at the surface have only half the neighbors (ones within the bulk) which attract them inwards into the bulk. The same picture is depicted in a slightly blown up form in fig. 1b.

This microscopic interaction has two consequences: firstly, molecules at the surface are at a higher energy since they are left un-bonded with molecules in the air, and secondly, the molecules in the bulk apply an attractive force on the surface molecules. Both are two aspects of the same effect, which leads to adjustment of shape of the liquids to expose the smallest possible area, and the excess energy at the surface is what we macroscopically

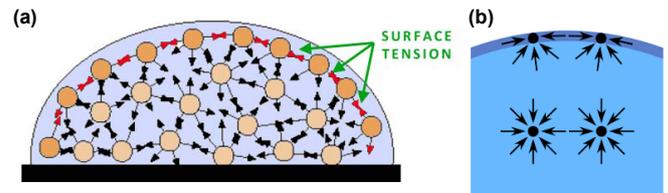


FIG. 1: A cartoon illustration of (a) a hemispherical drop of liquid resting on a solid surface with molecules depicted by circles and intermolecular attractive forces depicted by bi-directional arrows. (b) A blown up version of the picture depicted in (a). In both cases we notice molecules in the bulk experience intermolecular attractive forces from neighboring molecules along all directions, whereas molecules at the surface do not experience attractive forces from roughly half the volume that is now represented by air.

measure as the Surface Tension (γ) with units of Energy per unit area (mJ/m^2) or Force per unit length (mN/m).

I'd like to clarify two points: Firstly, Surface Tension is normally an equilibrium construct. I say “normally” because some caveats exist but they are not relevant to our experiment and we'll ignore them. Secondly, surface tension is the property of an interface, i.e. it arises only where two disparate media meet. So when we say the surface tension of water is $72 \text{ mN}/\text{m}$ for instance, we implicitly mean that $72 \text{ mN}/\text{m}$ is the surface tension of the air-water interface. Since most interfaces are normally exposed to air, we drop air and simply say surface tension of X or Y. If the medium is not air, then it becomes necessary to explicitly specify both media X and Y by quoting the surface tension of X-Y interface, such as water-oil interface for example. The definition of an interface is the surface where the density changes near-discontinuously (I say near because there is a short healing distance of a few molecule diameters, but in theory it may be ignored). The history of how the concept of Surface Tension came about is a fascinating story and should you be interested, a brief commentary is available in the introduction section of Ref. [1]. For a more lengthy and conceptual introduction to surface tension and all the phenomena associated with this property of interfaces, I recommend you to the beautiful book in Ref. [2].

Non-equilibrium condition: Marangoni Stress

In the previous sub-section, I introduced surface tension (γ) as an equilibrium quantity, i.e. it is constant at every point along the interface. By “point”, I mean a small enough area relative to the entire surface area but large enough to contain enough molecules to obtain a reliable statistical average, this type of averaging comes under the topic of “intermediate asymptotics” in Applied Mathematics [3].

Let us now consider a situation we witness in our daily lives. Imagine a quiescent surface of water, its surface tension value (assuming the water is pure) is close to 72 mN/m – I say “close to” because surface tension values are always quoted for the reference temperature at which they were measured, we will take the value at room temperature and assume it a constant for simplicity. Now let us suppose you introduce a drop of soap or oil at this interface, generally such materials are called “*surfactants*” because they coat the surface. You notice it immediately spreads across the surface to coat a thin film of surfactant. This is because the surface tension of air-soap or air-oil interface is lower (usually anywhere in the range 30 - 60 mN/m depending on type of soap or oil) than water. Given the tendency to reduce the excess surface energy, the surface gets coated with the surfactant to arrive at a lower surface tension value.

I want to address two issues at this juncture. First, about the quantity of surfactant necessary to achieve a new equilibrium surface tension value, and second the time taken by the surfactant to spread across the surface. This second point is what our experiment is actually about for it relates to the non-equilibrium transition between two equilibrium surface tension values of the interface.

To help build your intuition, we will use a class of surfactants known as “*Amphiphiles*”. Molecules of amphiphilic surfactants possess a hydrophilic head which loves water and a hydrophobic tail which hates water. As a result, such molecules prefer to reside on air-water interface where the molecule’s head sticks into the bulk of water and the tail sticks out. All soaps and many oils are amphiphilic materials. When tiny quantities of amphiphilic surfactants are introduced on a clean surface of water exposed to air, the amphiphilic molecules start populating the surface and the surface tension of water starts to decrease. This continues with further introduction of surfactant until the amphiphiles completely crowd the surface. The surfactant concentration at which this point is reached is known as the Critical Micellar Concentration or CMC. The surface tension now stabilizes at a new lower value, and further introduction of surfactant does not decrease the surface tension any further. The excess surfactant molecules go into the bulk and form globules of amphiphilic molecules called Micelles with the

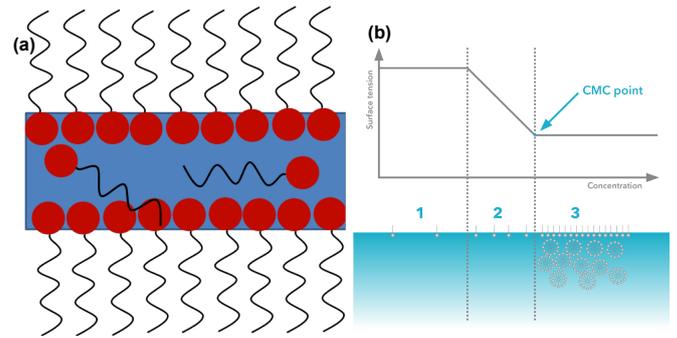


FIG. 2: (a) Cartoon illustration of amphiphilic molecules with a hydrophilic head (solid red circle) and a hydrophobic tail (wiggly black tail) populating the two sides of a soap film that provides it stability. (b) The high surface tension (stage 1) of water starts to decrease (stage 2) as amphiphilic surfactant is introduced until the surfactant concentration reaches a critical value called Critical Micellar Concentration (CMC) at which the surface tension value becomes constant (stage 3), i.e. attains new equilibrium value. Further introduction of surfactant cannot introduce more amphiphilic molecules on the surface as they have already crowded out the interface. The excess amphiphilic molecules form globular structures with tails sticking radially inwards and heads forming the surface of spherical globules known as micelles. These micelles then enter the bulk liquid.

hydrophobic tails pointing radially inwards into a sphere while the hydrophilic heads form the surface of the sphere of these micelles. These three stages are shown in fig. 2. This answers the first issue, now onto the second one.

To understand the mechanics of surfactant spreading and the time it takes for the system to transition from a higher equilibrium surface tension value to a lower one, we introduce the concept of Marangoni Stress. When we introduce a drop of surfactant at the air-water interface, the surface tension is high everywhere along the air-water interface but low at the air-surfactant interface. If we measure the difference in surface tension in going from the air-surfactant interface a short distance to the air-water interface, and divide by the distance, we have in effect a surface tension gradient. If surface tension has units of force per unit length (mN/m), its gradient has units of a force per unit area (mN/m²) or a stress. This stress acts to spread the surfactant homogeneously across the surface until the gradient vanishes. This surface tension gradient is famously known as the Marangoni Stress for historical reasons.

The surface tension gradient was first observed and reported by James Thomson [4] through an effect now famous as “*The Tears of Wine.*” Incidentally, James Thomson was the brother of William Thomson who is better known to us as Lord Kelvin. However it was explained theoretically and put down in mathematical form by Carlo Marangoni in his 1865 Ph. D. dissertation [5]. Barring a few studies, the Marangoni effect caused

by surface tension gradients remained just a theoretical curiosity with few experimental studies. Whereas non-equilibrium phenomena caused by surface tension gradients are very easy to setup and observe, they are notoriously difficult to experimentally quantify. Part of the reason was also scientific culture, whereby the Marangoni effect remained the purview of surface science and was little explored within hydrodynamics. Starting in the 1980s both experiments and theoretical efforts to wed the surface science with hydrodynamics took place to understand the transient, non-equilibrium phenomenon of surfactant spreading over air-water interfaces. Further extensions occurred when studies undertook the effect of particles at air-water interfaces and how they were impacted by a spreading surfactant, the area within which the current experiment falls. I will share further details relevant to our experiment later in the document.

EXPERIMENT

Motivation

To understand the experiment you're about to conduct, let us start by asking what happens in the vicinity of a particle when introduced at the air water interface? Figure 3 shows a schematic of a pair of hydrophobic (fig. 3a) and hydrophilic (fig. 3b) particles at the air-water interface. Capillary effects act to deform the equilibrium air-water interface represented by the dashed line whereby the meniscus dips in case of hydrophobic particles because it wants to avoid the area exposed to the hydrophobic particle surface, but rises in case of hydrophilic particles and in fact coats a thin lubrication film over the particulate surface. Please note, I am intentionally representing particles with density less than water's, therefore you do not witness gravitational effects that might otherwise arise [6]. I do so because you will be using naturally buoyant particles in your experiments.

This difference in the menscii of hydrophilic and hydrophobic particles has a pronounced effect when the boundary is moved by some means to scoop up the particles into a smaller area to the point they come in contact with each other. When hydrophilic particles come in contact, they exploit the thin lubrication film resulting from the rising meniscus to slide over each other out of plane along the third dimension. However, if the same were done with hydrophobic particles, they do not possess the thin lubrication film to slide over each other. When in contact, they develop the ability to sustain stresses, i.e. they develop a non-zero yield stress and a non-zero shear modulus, two properties that define a solid. This system of hydrophobic particles, through the combination of their capillary interaction and the boundary condition now form a quasi two-dimensional solid. If interested in an elegant exposition of the theory underlying the me-

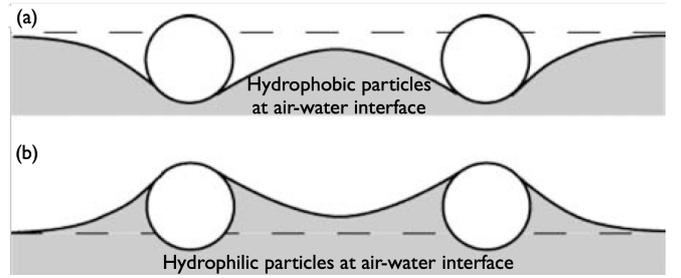


FIG. 3: Schematic representation of a pair of (a) hydrophobic and (b) hydrophilic particles at the air-water interface. The dashed line represents the equilibrium interface to impress how it deforms upon introduction of the particles. Whereas the meniscus resulting capillary effects dips to avoid exposure of water to (a) hydrophobic particles, it rises and coats a thin lubrication film across (b) hydrophilic particles.

chanics of these 2D solids complemented by simple experiments, the reader is encouraged to study Ref. [7]. For this reason tracer particles used in Particle Imaging Velocimetry (PIV) techniques in experimental hydrodynamics are hydrophilic, for if they are hydrophobic they will bias the experimental measurements. The specific gravity of the particles may however vary depending upon the experimental situation from naturally buoyant [8–10] to neutrally buoyant [11, 12].

Description of the Experimental Phenomenon

Consider a petri dish with water (dynamic viscosity $\mu = 10^{-3}$ N.s.m⁻²) or glycerol ($\mu = 10$ N.s.m⁻²). Sprinkle buoyant particles (hydrophilic or hydrophobic) of mean diameter $50 \mu\text{m}$ or so at its interface to form a 2D floating configuration. The dispersity of particles does not matter unless you want to study crystalline configurations, in which case you do need monodisperse particles. We do not focus on crystalline configurations here. So long as the mean particle diameter is of order 10s of μm , particle size has little bearing on the effect. Now dip a needle in a surfactant such as soap or oil and introduce the needle-tip at the air-liquid interface at the petri dish center. As the surfactant spreads to reduce the surface tension of the air-liquid interface, it pushes the particles radially outwards in a circle of growing radius. If the particles are hydrophilic (water-loving), this circle is smooth (see fig. 4a) because when hydrophilic particles come in contact with one another, they exploit the thin lubrication film coating them to slide over each other. But if the particles are hydrophobic (water-hating), the circle is initially smooth until the particles come in contact, but then they cannot slide over each other because the meniscus dips down and they do not have a thin lubrication film coating them. Therefore the hydrophobic particles can sustain stresses and form a quasi-2D solid, which then

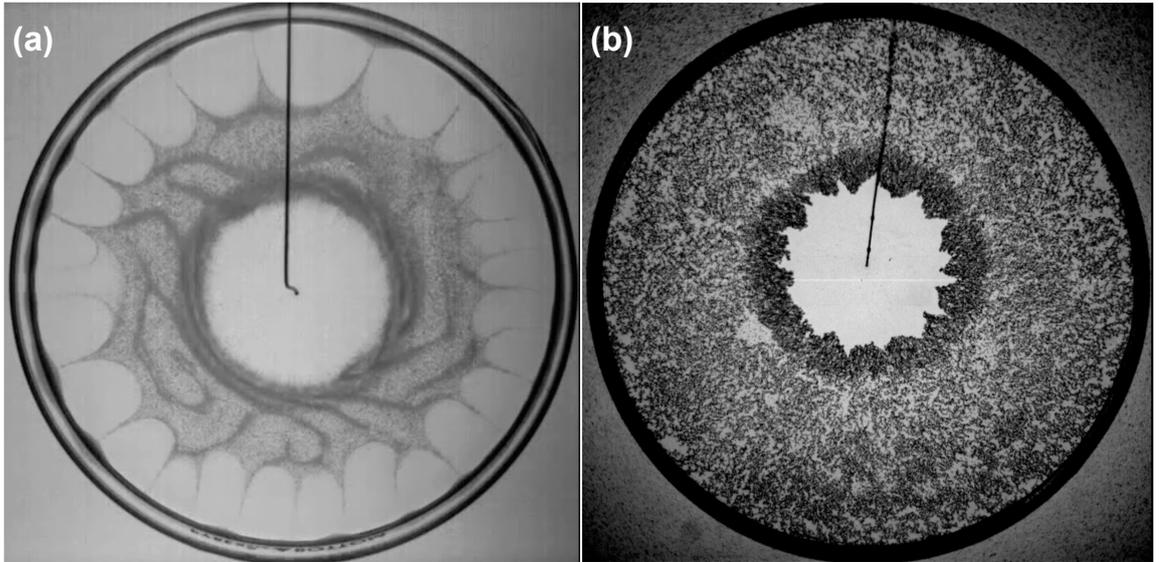


FIG. 4: The spreading surfactant front visualized using (a) hydrophilic and (b) hydrophobic particles leads to starkly different particle patterns. The particulate band (dark annulus) appears as a smooth circle for (a) hydrophilic particles as opposed to periodic fracture patterns for (b) hydrophobic particles.

fractures (see fig. 4b) as periodic cracks [13, 14].

The purpose of the experiment will be to collect a video recording with your smartphone of the surfactant spreading and subsequent fracture of the 2D solid and perform image analysis to extract the radius of the growing surfactant-particle front $R_S(t)$ as a function of time t . Towards the end of the introductory section, I mentioned investigations into surface tension gradient problems took off only around 1980s. Since we will use some of these results, let me point you to some of the literature. References [15–18] serve as good introductions for the “Tears of Wine” problem as well as a generally good introduction, but historical and conceptual.

The class of studies we focus on however concern the transient spreading of a surfactant across an air-water interface. When a surfactant is introduced from a point source (such as a needle as we do) at air-water interface of a deep fluid layer, the radial location of the spreading surfactant front ($R_S(t)$) from the source is given by the expression:

$$R_S(t) = K \left(\frac{\Delta\gamma^2}{\mu\rho} \right)^{1/4} t^{3/4} \quad (1)$$

where $\Delta\gamma = \gamma_{(water-air)} - \gamma_{(surfactant-water)} - \gamma_{(surfactant-air)}$ is called the Harkins spreading coefficient at the line of three phase contact, i.e. the surfactant front, μ is the dynamic viscosity of water and ρ is its density, and K is a numerical coefficient in the range $0.665 \leq K \leq 1.52$.

To the best of my knowledge, Eq. 1 was first introduced in Ref. [19] and was experimentally demonstrated in Ref. [20]. A more convincing experimental proof and

a physically intuitive explanation for the $R_S(t) \sim t^{3/4}$ scaling is provided in Ref. [21], but a theoretically rigorous proof subject to certain requirements that takes into account the spreading surfactant’s shearing of the underlying water and the nucleation of a shear boundary layer (commonly known in Hydrodynamics as the Blasius boundary layer) was provided by Olivier Jensen in Ref. [22].

The goal of our experiment is to test the scaling in Eq. 1. Ideally, this scaling would be easier to verify with hydrophilic particles as they result in a smoother ring of the surfactant front, but hydrophilic particles are not easy to find in daily situations. So we will use hydrophobic particles, but a nice aspect of that is we get to explore the fracture of particle rafts.

EXPERIMENT

Ingredients

This is a low-tech experiment and the phenomenon is robust enough to be observed with very minimal effort. Things needed for this experiment are:

- 1) A Petri dish or a glass bowl.
- 2) Water – tap water or mineral water suffices.
- 3) Pepper flakes or any type of flour used in the kitchen.
- 4) A safety pin.
- 5) Some liquid soap.
- 6) A sheet of white or black paper – white if you use pepper flakes and black if you use flour.
- 7) Glycerine or glycerol, if you have access.

Methods

Step 1: First wash your dish and wipe it dry with a paper napkin or clean cloth. This is a strict no when the experiment is conducted in lab conditions but its ok for a kitchen experiment.

Step 2: Pour some water into the dish. Tap water usually has organic impurities that affect the surface tension of water. If possible, please use mineral water. In lab conditions, we used de-ionized water.

Step 3: Place a white/black sheet of paper on a table and place the dish on it. We do this so you have a background contrast when you image the experiment with your smartphone. Normally in lab conditions, we place the dish on a light tablet and image the transmitted light through the dish, water, and particles. This allows you to image the particulate shock. But for our kitchen experiment, we don't have access to a light tablet.

Step 4: Place a few books or blocks on both sides of the dish to act as a support on which to rest your smartphone for imaging. Place the smartphone with the display facing you (away from the dish). Open the camera app in video mode to adjust the smartphone such that it captures the entire dish surface. Play with the height of the supports and the camera magnification until the entire dish covers your camera's field of view.

Step 5: Make sure you turn on the flash mode and turn off lights in your room and close window curtains. Since we do not have the luxury to provide controlled illumination as done under lab conditions, we make use of the camera flash to obtain a homogeneous illumination with no shadows at a reliable intensity.

Step 6: Scoop up some pepper flakes or flour with a spoon and very gently tap the spoon to sprinkle them onto the water surface so they coat it uniformly. Alternatively, if you have a sprinkler for salt and pepper, the pepper sprinkler works very well. In reality, pepper flakes as well as flour have natural oils within them. When you introduce them on the water surface, this has the effect of dispersing all particles more or less uniformly across the surface. You want to introduce enough pepper flakes to uniformly coat the surface. Fewer than that and you may not see the effect. Whereas sprinkling a few excess flakes is not a problem, introducing too many introduces inertia and you may not see the effect, so there is a sweet spot where the optimal experimental conditions are attained.

Step 7: Now take a long needle or safety pin. If using a safety pin, open up its bends so you can stretch

it out into a straight, long needle. Dip it in soap.

Step 8: Now start recording the video on your camera in slow motion (Slo Mo) mode. Bring the needle tip previously dipped in soap to the petri dish center and poke the surface. Try not to occlude the camera's field of vision with your hand or the camera will fail to record the surfactant dynamics.

Step 9: You will notice the configuration of pepper flakes are pushed radially away from the point when you dipped the needle and the configuration of pepper flakes have cracked.

Step 10: Stop the video recording. Your experiment is over.

Note1: If you do the above experiment with glycerine/glycerol instead of water, the viscosity of glycerine is 1300 times that of water, so the dynamics slows down considerably. This is due to the presence of the μ term in Eq. 1. In such case, record the video in normal mode, not in slow motion. However glycerine is also hygroscopic, i.e. it adsorbs vapor from the ambient air causing the viscosity in the surface layer of glycerine to decrease. To avoid this, you should perform the experiment within a few minutes of pouring the glycerine into the dish.

Note2: Normally, we perform these experiments with naturally buoyant hollow glass spheres (commercially called glass bubbles), which are available in both hydrophilic and hydrophobic form. We treat the petri dish and these glass bubbles to make sure they are all clean and will not contaminate the water surface during the experiment. In our case, when we introduce the pepper flakes or flour on water, the leaching of natural oils from pepper flakes or flour already contaminates the surface and reduces the surface tension. However, the surface tension of soap is lower than that of oil. That is the reason we dip the needle in soap to perform this experiment. If we were to dip the needle in oil, nothing will happen because there's no surface tension difference, hence no gradient.

Analysis

To perform the analysis, I would like you to read the paper [13] as we will closely follow a section of the steps in image and data analysis explained therein. We will focus on testing Eq. 1 for our experiment. For that, we need not calculate the azimuthally averaged radial packing fraction at each time instant $\phi_\theta(r, t)$. Be that as it may, we will go through part of those image analysis steps in the following sense. We will merely focus on image intensity values to distinguish particle-laden from

particle-free regions, but we will not perform the calibrations necessary to estimate the packing fraction itself. For your benefit packing fraction is defined in a 2D system such as ours as the ratio of the area occupied by the particles to the total surface area. What I mean will become clear shortly as you read the image analysis steps I outline below. Part of a scientist's research activity involves searching and identifying the tools necessary to perform the analysis in question. So I will identify the steps and tools but will not spoon-feed you. I will expect you to take my pointers and perform your own search and proceed with image analysis.

Image Analysis

Step 1: Take the movie recording from your smartphone and transfer it to a laptop. Then convert the movie into a series of consecutive images. I use Apple's Quicktime professional version, but there are several open source programs out there such as ImageJ that allow you to do this. I will let you search online for a suitable toolkit that allows you to perform this task on your laptop.

Step 2: Having converted the movie to images, let me digress a bit on what we normally do. As you know, there are several image standards, JPEG, GIF, PNG, etc. When performing image analysis a good standard to follow is to choose an image standard that does not compress your images because compression always leads to loss of information – this is a direct consequence of Claude Shannon's Information Theory. Loss of information during compression can result in degradation of image quality for image analysis to extract quantitative data, this qualifies as introduction of systematic error into your experiment. The best image standard is TIFF so that there is no information loss from image compression. Within TIFF standard, if you store the movie as a series of TIFF images instead of multipart TIFF with multiple images within the same file, it causes less headache because it keeps your code and algorithms simple.

For the purposes of your current experiment, it does not matter what image standard you pick, but if you have the freedom of choice, I would advise you select TIFF or PNG because PNG is a lossless compression standard. If its not possible, do by all means go with any other image standard.

Step 3: Now you have your experimental images and you are ready to commence your analysis. The first task is to write a program in a programming language of your choice (e.g. Matlab, C, C++, Java, Python etc.) to open an image file and convert it to grayscale image.

Step 4: With the grayscale image, identify the X and Y pixel coordinates of the point where the needle touched the interface. From there count the number of pixels to the petri dish edge, this is the radius of your petri dish – approximately since you cannot possibly identify the exact petri dish center and impinge the needle there. Now extract the intensity at each pixel along the line connecting the center to the petri dish edge. This gives you the pixel intensity $I(r, t)$ at each pixel point along the radius r and at the given time instant t .

To learn how to draw a line, I suggest you look up the Bresenham line algorithm. You repeat the above process of computing $I(r, t)$ at each azimuthal angle. In other words, you are sweeping a circle with a line that runs the length of a circle of radius r . Take the average over all the lines along all azimuthal angles θ , the more lines you scan, the better your average and you now have $I_\theta(r, t)$, i.e. the azimuthally averaged Intensity as a function of radial distance r and time t . To achieve this, an elegant way is to combine Bresenham line algorithm with the Bresenham circle algorithm.

Step 5: Doing the above for all images you can now construct a graph of the kind shown in figure 2a of Ref. [13].

Step 6: You can see a shock front proceed from left to right along x-axis for radial distance (r) as a function of time t whereby the Intensity $I_\theta(r, t)$ on the y-axis discontinuously varies at a particular radial distance. This shock front is basically the location of the surfactant front in your experiment. Taking its derivative, you can now obtain the location of the surfactant front as a function of radial distance, i.e. $R_S(t)$. Plotting this quantity as I have done in figure 2b of Ref. [13] in a log-log plot should give you a straight line of slope $3/4$, implying the surfactant front proceeds with time t as $R_S \sim t^{3/4}$.

Step 7: If you repeat your experiment with glycerol you will notice this scaling shifts down, i.e., the y-intercept of the log-log plot decreases. That is because you have increased dynamical viscosity μ by a factor of 10^4 and since it is in the denominator, the plot shifts down.

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