

Bubbly flows in non-Newtonian and viscous Newtonian fluids

Experimental

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BROWN
School of Engineering

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CURRICULUM VITÆ

Mithun Ravisankar is a mechanical engineering graduate from Alagappa Chettiar Government College of Engineering and Technology, affiliated with Anna University, Tamil Nadu, India. Originally from Chennai, he developed an early fascination with how things work, particularly the behavior of machines and fluid systems, which grew into a passion for fluid mechanics, thermal sciences, and material behavior. His first exposure to fluid dynamics came during a research internship at Indian Institute of Technology Madras, where he studied how rail pressure affects droplet size in Common Rail Direct Injection (CRDI) diesel engine fuel atomization. This experience sparked a lasting appreciation for flow physics and the complexity of multiphase interactions.

He further explored the dynamics of smart materials during his undergraduate thesis on Shape Memory Alloys (SMAs), leading to a research assistantship at Indian Institute of Technology Indore. There, he simulated large deformations in the Friction Stir Welding of SMA plates by treating solids as incompressible fluids and applying the Carreau viscosity model to capture nonlinear behavior. He also addressed reviewer critiques by modeling asymmetric velocity fields using irrotational flow past a cylinder with circulation, while independently conducting experiments on laser-actuated SMA springs and authoring the resulting manuscript.

These experiences led him to pursue an Sc.M. in Fluids and Thermal Sciences at Brown University, where he worked under Prof. Roberto Zenit on bubble dynamics in viscoelastic fluids. His research focused on the clustering behavior of rising bubble pairs in viscoelastic media, particularly the velocity discontinuity that occurs at a critical bubble volume. Unlike the well-known Drafting-Kissing-Tumbling (DKT) interaction in Newtonian fluids, he discovered a unique phenomenon—Drafting-Kissing-Dancing (DKD)—in which bubble pairs exchange positions due to the dominance of elastic stresses. Mithun continued into the Ph.D. program at Brown, where he is currently researching bubble-induced turbulence and advancing the understanding of multiphase flow phenomena. His academic journey has combined hands-on experimentation, numerical modeling, and scientific writing, and he remains committed to exploring the physics of complex flows in pursuit of engineering solutions with real-world impact.

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1. **M. Ravisankar**, A. G. Correa, Y. Su, and R. Zenit. Hydrodynamic interaction of a bubble pair in viscoelastic shear-thinning fluids. *Journal of Non-Newtonian Fluid Mechanics*, 309:104912, 2022.
2. O. Atasi*, **M. Ravisankar***, D. Legendre, and R. Zenit. Presence of surfactants controls the stability of bubble chains in carbonated drinks. *Physical Review Fluids*, 8(5):053601, 2023.
3. **M. Ravisankar** and R. Zenit. Velocity fluctuations for bubbly flows at small Re. *Journal of Fluid Mechanics*, 1001:A34, 2024.
4. **M. Ravisankar**, M. E. Federle, M. Bahadori, A. Das, H. Kesari, and R. Zenit. The mechanics of the squash nick shot. *Proceedings of the National Academy of Sciences*, 122(20):e2505715122, 2025.
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PREFACE AND ACKNOWLEDGMENTS

This dissertation consists of original research by Mithun Ravisankar between August 2021 and October 2025 under the supervision of Professor Roberto Zenit at the Brown University School of Engineering. Contributions from others are an integral part of this work and are noted in the text where appropriate.

தெய்வத்தான் ஆகா தெனினும் முயற்சிதன்
மெய்வருத்தக் கூலி தரும்.

-திருவள்ளுவர்

Though fate-divine should make your labour vain;
Effort its labour's sure reward will gain.

-Thiruvalluvar

குறள்/Couplet: 619

பால்/Book: பொருட்பால்/Wealth

அதிகாரம்/Chapter: ஆள்வினையுடைமை/Manly Effort

I express my profound gratitude to my research advisor, *Prof. Roberto Zenit*, for your unwavering support, insightful guidance, and constant encouragement throughout my research and academic life at Brown University. It has been an absolute pleasure and honor to work with you. I am deeply thankful to you for introducing me to the fascinating world of bubbles and non-Newtonian fluids. Most of all, I am grateful for the example you set: showing me what it means to approach research with passion, energy, and more importantly, joy.

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1 INTRODUCTION

1.1 Turbulence: A Classical Perspective

Turbulence is one of the most complex and captivating phenomena in classical physics. From mixing cream in coffee and the smoke rising just after a candle is blown out, to crashing ocean waves and the banded clouds on Jupiter (see examples in Fig. 1.1), turbulence is all around us. Over 500 years ago, Leonardo da Vinci even sketched the chaotic patterns created by a jet of water plunging into a pool [25]. Our scientific understanding of turbulence began to take shape with Osborne Reynolds' experiments in 1883 [26]. He introduced the Reynolds number, $Re = UD/\nu$, a dimensionless measure of the ratio between inertial and viscous forces in a flow. Here, U is the velocity scale, D the characteristic length (like pipe diameter), and ν the kinematic viscosity. When $Re \gg 1$, inertial effects become dominate and when $Re > O(1000)$, the flow gradually transitions to turbuelence.

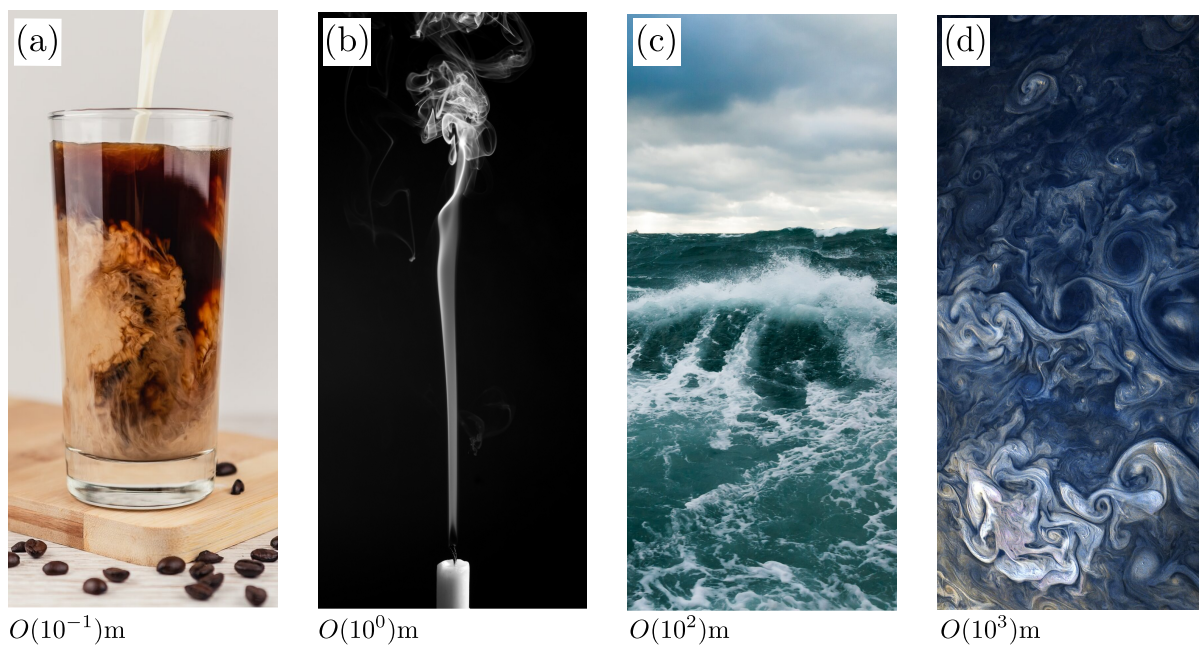


Figure 1.1: Examples of turbulent flows: (a) Mixing cream in coffee; (b) Smoke rising just after a candle is blown out; (c) Breaking wave in the ocean; (d) Banded clouds on Jupiter (Credits: NASA).

When describing turbulence, it is customary to highlight the characteristics that define its behavior. Turbulent flows are inherently irregular, unpredictable, and strongly chaotic. They span a wide range

of spatial and temporal scales, which allows them to efficiently transport momentum, heat, and other quantities throughout the fluid. Turbulence is also intrinsically dissipative: kinetic energy is continuously converted into heat through viscous effects, so sustaining turbulence requires a continuous input of energy. In the absence of forcing, turbulence naturally decays. This dissipative nature also makes turbulence statistically irreversible in time, meaning that the direction of time can be inferred from the evolution of a turbulent flow [27].

Turbulence, at its core, is described by the Navier–Stokes equations, but their complex, nonlinear nature makes them difficult to solve [28]. In order to overcome this complexity, a statistical description of turbulence was adopted. One of the most important contributions came from Andrei Kolmogorov, who showed that even chaotic flows follow simple, predictable patterns. Kolmogorov [29] proposed that in high Reynolds number flows, the smallest scales of turbulence behave in a statistically universal way, regardless of how the turbulence started. He imagined turbulence as a nested cascade of swirling eddies, where energy flows from large motions to progressively smaller ones, and eventually dissipated by viscosity at the smallest scales. This elegant picture led to the well-known $k^{-5/3}$ power law for the energy spectrum with respect to the wavenumber, k , a relationship that has been observed in a wide range of turbulent flows and remains central to our understanding of turbulence today.

While Kolmogorov’s theory provides a powerful framework for understanding turbulence, real world flows often show deviations from its predictions. Turbulence can be intermittent, with sudden bursts of intense motion, and these fluctuations lead to subtle changes in the expected scaling of the flow. Near walls, in rotating or stratified fluids, or in strongly sheared flows, the simple universality assumed by Kolmogorov may break down [30–32]. Modern experiments and high-resolution simulations have confirmed the classic $k^{-5/3}$ scaling over a wide range of conditions, but they have also revealed these more nuanced behaviors, prompting refinements to the theory. Grasping these complexities is key to accurately modeling turbulence in real-world applications, from engineering systems to atmospheric and astrophysical flows.

1.2 Bubbles and Bubble Induced Turbulence

Bubbles - It’s unbelievable to fathom that a simple pocket of voidness, referred to as a bubble has intrigued humans for almost two millennia. From gas bubbles escaping ocean seeps to the mesmerizing rise of

champagne bubbles in a glass [2], these ephemeral creations evoke a sense of wonder and playfulness. In the first recorded proverb, *Homo bulla*, the metaphor of man as a beautiful but exquisitely fragile and transient bubble, dates back to the Varro (116 BC – 27 BC) wrote the following in the first line of the first book of *De Re Rustica*, “for if, as they say, **man is a bubble**, all the more so is an old man”. Several other examples are meticulously summarized by Prosperetti [33, 34]. A long standing puzzle, aptly named as Leonardo’s paradox is that of a single bubble rising in a still liquid [33]. There is still an on going debate (see [35] and [36]) on what cause the zigzagging or spiraling paths of a rising bubble rather than the expected rectilinear vertical trajectory (see Fig. 1.2). A detailed review of gas bubble dynamics are discussed in [37].

For a single gas bubble rising in a quiescent liquid, the terminal velocity of the bubble, u_b , can be expressed as a function of the equivalent bubble diameter d_b , the liquid dynamic viscosity μ , the surface tension σ , the liquid density ρ , and the gravitational acceleration g . By applying the Buckingham– Π theorem, this relationship can be expressed in terms of dimensionless groups, most notably the Reynolds, Bond (or Eötvös), and Morton numbers:

$$\text{Re} = \frac{\rho u_b d_b}{\mu}, \quad \text{Bo} = \frac{\rho g d_b^2}{\sigma}, \quad \text{Mo} = \frac{g \mu^4}{\rho \sigma^3}. \quad (1.1)$$

Here, Re characterizes the ratio of inertial to viscous forces, Bo compares buoyancy to surface tension and Mo depends solely on the fluid properties and dictates the regime of bubble motion. As bubble volume increases (Eötvös number), its shape transitions from spherical, then ellipsoidal, and eventually adopting more complex form such as dimpled ellipsoidal caps. The Clift-Grace-Weber diagram [38] provides a framework for categorizing these behaviors based on dimensionless parameters. Additional non-dimensional groups are often introduced to describe specific dynamical balances:

$$\text{We} = \frac{\rho u_b^2 d_b}{\sigma}, \quad \text{Ca} = \frac{\mu u_b}{\sigma}, \quad \text{Oh} = \frac{\mu}{\sqrt{\rho \sigma d_b}}. \quad (1.2)$$

Here, the Weber number, We, compares inertial to surface tension forces; the Capillary number, Ca, compares viscous to surface tension forces; and the Ohnesorge number, Oh, compares viscous to inertial–capillary effects.

Bubble Induced Turbulence - In many practical applications, bubbles are dispersed within a continuous liquid phase, such as in oil pipelines, bubble column reactors for chemical processing, or vapor generators

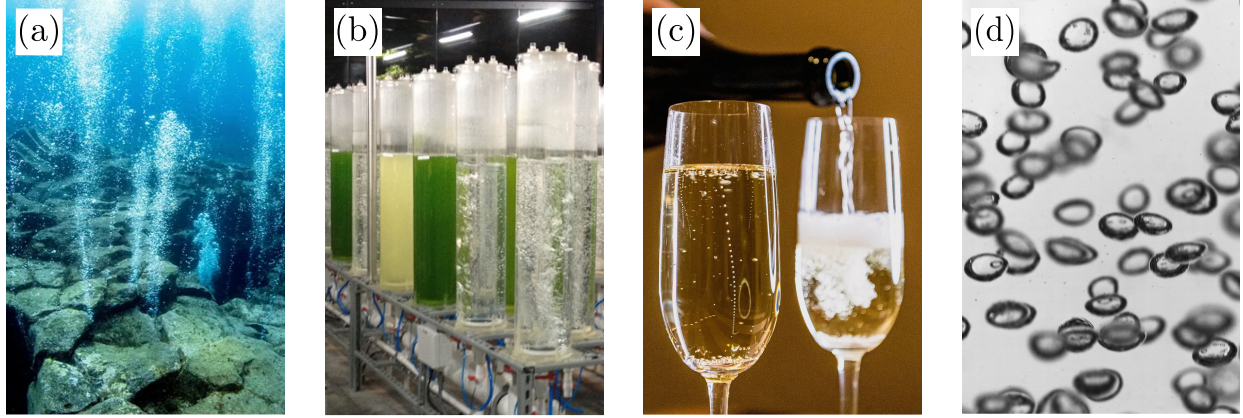


Figure 1.2: Examples of bubbly flows: (a) Gas bubbles from ocean seeps; (b) Bubbles in an algae bioreactor; (c) Bubbles rising in a glass of champagne (Credits: Colin Sullivan); (d) Bubbly flow in a Newtonian fluid.

in energy systems. Because of the large density difference between gas and liquid, bubbles do not travel at the same velocity as the surrounding fluid. Their motion induces velocity disturbances in the liquid, which collectively generate a complex form of agitation. In flows where the liquid is already turbulent, the resulting agitation arises from the two-way interaction between conventional shear-driven turbulence and bubble-induced disturbances. The overall flow behavior therefore depends on the structure and relative intensity of these two contributions, as well as the characteristic spatial and temporal scales associated with each. Understanding this interplay is crucial for accurately predicting mixing, mass transfer, and energy dissipation in multiphase flow systems.

Commonly found two-phase gas-liquid flows is the dispersion of air bubbles in a continuous liquid phase, also referred to as bubbly flows (see examples in Fig. 1.2). When the continuous liquid phase is turbulent, adding bubbles to such flows have proved to be beneficial in significant drag reduction [39]. Even in the case where a swarm of bubbles rising in an otherwise quiescent fluid, turbulent velocity fluctuations are imposed in the surrounding fluid entirely due to the motion of bubbles. Since the first pioneering work of Lance and Bataille [40], this turbulent-like nature due to the motion of bubbles, dubbed as the “pseudoturbulence”, has been the interest for a significant number of studies [41, 42]. The salient feature of the pseudoturbulence is that the fluctuating nature of the turbulence is observed due to the wake-wake interaction between the bubbles rising in a swarm. A paramount attribute of bubble-induced turbulence is its departure from the homogeneous isotropic Kolmogorov’s turbulence, see the recent review by Risso [43]. Specifically, the power spectra of liquid velocity fluctuations has been reported to decay as k^{-3} , where k is

the wavenumber. This occurs in the range where ordinary turbulence decays as $k^{-5/3}$, due to the addition of bubbles, the dissipation of turbulent kinetic energy is enhanced. This feature is one of the aspects that make bubbly flows to be attractive for some applications, particularly those which need high levels of agitation at small Reynolds numbers [44].

1.3 Viscoelasticity and Polymer Induced Turbulence

Viscoelasticity - When the Prophetess Deborah in the *Book of Judges* sang, “The mountains flowed before the Lord”, Reiner [45] realized she knew two things. First, that the mountains flow, which she said before the famous words of Heraclitus’, “Everything flows” (Greek: Πάντα ῥεῖ, *panta rhei*). Secondly, they flowed before the Lord and not before man, hinting the different time scales between God’s and his own. This property whether a material behaves like a solid or a fluid depending on the observation time (of a man), is referred to as viscoelasticity. Most biological fluids, like saliva, mucus, and blood, and pleural effusions are viscoelastic. The Deborah number is a dimensionless number that compares a material’s relaxation time to the time scale of an observation. A closely related number, the Weissenberg number, is also commonly used. In simple cases—like the lid-driven cavity flow of a fluid in a square duct—both numbers are the same. However, in non-square geometries, they differ and can be related through a geometric factor (see [46] for more details).

Elastic Turbulence - Viscoelastic fluids, like flexible polymer solutions, behave differently from Newtonian fluids due to their nonlinear properties. One striking example is that under certain shear conditions, these fluids can show turbulent-like motion even at very low Reynolds numbers, due to polymer stretching, an effect known as “elastic turbulence” [47]. Figure 1.3(a) shows an example of a swirling flow between two parallel disks [47]. In this case, the fluid velocity fluctuations are observed to decay at a power of $f^{-3.5}$, where f is the frequency. This is accompanied by significant stretching of the polymer molecules, resulting in an increase in the elastic stresses of up to two orders of magnitude. The discovery (or re-discovery) of elastic turbulence has attracted the attention of many research groups in the recent times [48].

Elasto-inertial Turbulence - When the Reynolds number is sufficiently high such that the inertial effects cannot be neglected, the modified turbulence due to the elasticity is referred to as “elasto-inertial turbulence” [49]. For certain applications like porous media flows [50], polymers are employed to achieve maximum

drag reduction [51, 52] and heat transfer enhancement [53]. A combination of experimental and numerical studies in the elastoinertial turbulence regime has revealed that a scaling exponent of -3 , in contrast to the classical Kolmogorov's $-5/3$ scaling observed in Newtonian fluids, is a distinctive signature of polymer induced turbulence [54–57]. Figure 1.3(b) shows the temporal power spectral density for concentration fluctuation for the Newtonian and polymer jet entering into a bath of Newtonian fluid at $Re = 800$.

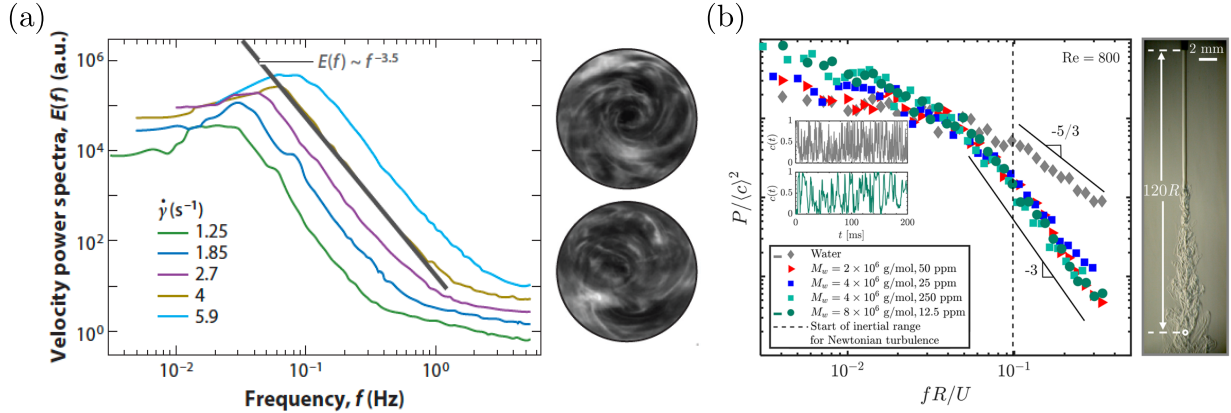


Figure 1.3: (a) Example of power spectra of velocity fluctuation and flow visualization of a swirling flow between two parallel disks at $Re = 0.7$; Adapted from [47]; (b) Temporal power spectral density for concentration fluctuations at $Re = 800$ for the Newtonian and polymer jet entering into a bath of Newtonian fluid; The inset compares the raw time signal of concentration fluctuation for the Newtonian jet and a polymer jet; Adapted from [57].

1.4 Motivations

Turbulent multiphase flows are common in both nature and engineering. Examples range from red blood cells moving through blood vessels and virus-laden droplets in air, to turbulent coolant flows in nuclear reactors, polymer-based drilling fluids in energy exploration, and air-lift reactors in chemical processing. Yet, much of our understanding of bubble dynamics still comes from studying them in simple Newtonian fluids like water.

The brief introduction above highlights the need to study and better quantify the modulation of turbulence due to both bubbles and viscoelasticity simultaneously. This problem is particularly relevant given the considerable range of applications in geophysics, biology, or industry. Figure 1.4(a) and (d) shows the cartoon of the shear-induced and bubble-induced turbulent flow composed of vortices of various length and time scales. Following the Kolmogorov's scaling, the energy spectra for a Newtonian shear-induced

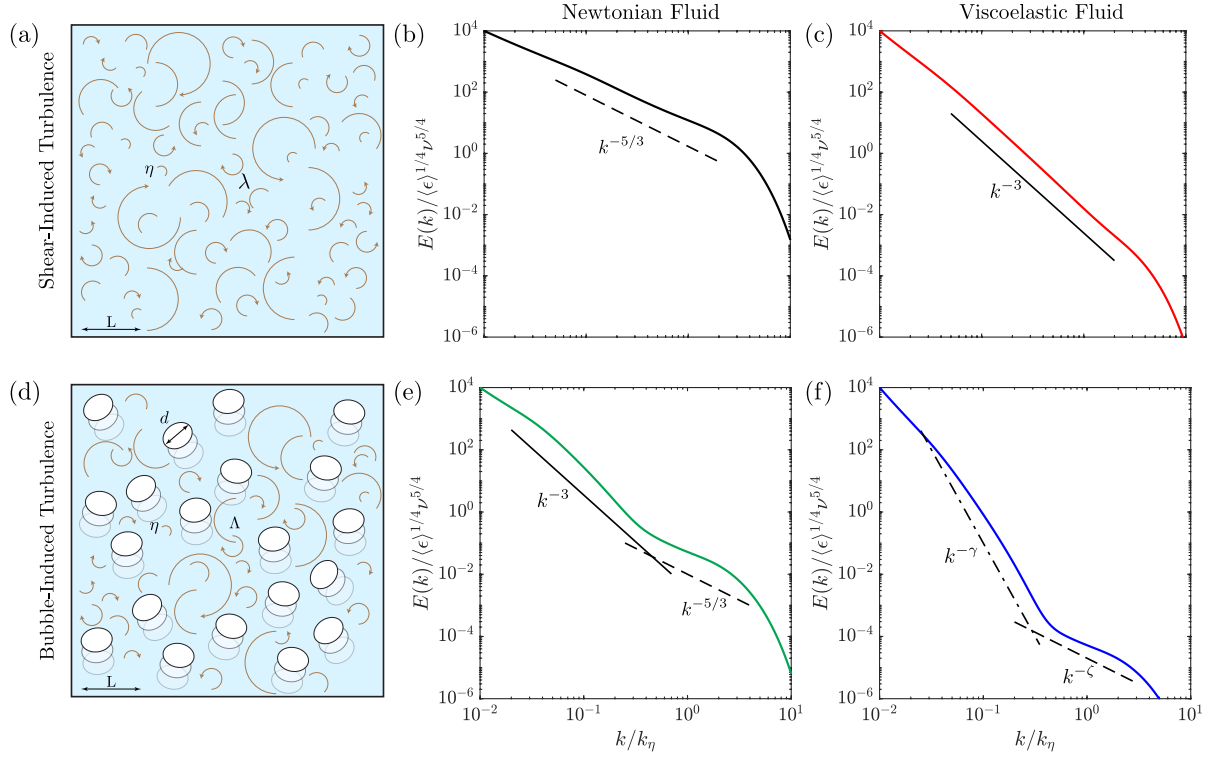


Figure 1.4: (a) Shear-induced turbulent flow composed of vortices (or eddies) of various length and time scales; Here, L is the integral scale, η is the dissipative scale, and λ is the Taylor microscale; (b) Normalized energy spectra for a Newtonian shear-induced turbulent flow; The classical Kolmogorov inertial scaling of $k^{-5/3}$ is shown as a dashed line; Here, ϵ is the dissipation rate and ν is the kinematic viscosity; (c) Normalized energy spectra for a viscoelastic shear-induced turbulent flow; The steeper scaling of k^{-3} due to the elasticity is shown by a solid line; (d) Bubble-induced turbulent flow composed of vortices (or eddies) of various length and times scales; Here, d is the bubble diameter and Λ is the Eulerian length scale; (e) Normalized energy spectra for a Newtonian bubble-induced turbulent flow; The $k^{-5/3}$ and k^{-3} power law is shown by a dashed line and a solid line, respectively; (f) Normalized energy spectra for a viscoelastic bubble-induced turbulent flow, where a different scaling behavior expected is shown as a dot-dashed line.

turbulent flow shows a $k^{-5/3}$ scaling, as shown in the Fig. 1.4(b). Whereas for a viscoelastic fluid, the energy spectra shows a k^{-3} scaling, as seen from Fig. 1.4(c). Because of the bubble wake-wake interactions, the energy spectra for a Newtonian bubble-induced turbulent flow shows a k^{-3} scaling for length scales larger than the bubble diameter, whereas for length scales smaller than the bubble diameter, the classical Kolmogorov's $k^{-5/3}$ scaling is recovered, as shown in the Fig. 1.4(e)

While the modulation of turbulence spectra by polymers [49, 57, 58] and by the bubbles [40, 44, 59] has interested the scientific community for decades, an intriguing open question remains: how do polymer solutions modify the bubble-induced turbulence? (as shown in Fig. 1.4(f)) From a fundamental

perspective, can polymers still trigger distinct changes in flow structures when turbulence is driven by rising bubbles—where energy input is discrete, anisotropic, and lacks organized shear? So far, this remains unexplored. An important follow-up question is whether inertia plays a role in how bubbles modulate turbulence in viscoelastic fluids.

1.5 Outline of the dissertation

In this thesis, we investigate how *elasticity* and *viscosity* influence the dynamics of bubble-induced turbulence. Specifically, we examine the wakes generated by a swarm of bubbles rising through a viscoelastic fluid. To quantify the resulting mixing and flow behavior, we employ various statistical tools, including energy spectra, spatial correlations, and other turbulence diagnostics.

Chapter 2 explores how adding polymers affects pseudoturbulence created by bubbles rising in a still fluid. We show that above a certain polymer concentration, the energy spectrum steepens beyond the usual k^{-3} scaling seen in Newtonian fluids, regardless of gas volume fraction in the inertial range. This change happens between the scales of bubble diameter and wake length. Using Lagrangian stretching, we also find that polymers increase flow coherence, highlighting how viscoelasticity can reshape turbulence even without background shear.

Chapter 3 investigates how bubble-induced turbulence changes with Reynolds number in a quiescent Newtonian fluid. Using PIV, we analyze the decaying turbulence behind a bubble swarm and find that at moderate $Re \sim O(100)$, the usual k^{-3} scaling becomes weaker and depends on gas volume fraction. As Re decreases further ($O(1)$ – $O(10)$), the spectrum shifts to a $k^{-5/3}$ scaling, suggesting that without strong bubble wakes, the turbulence becomes less universal and more sensitive to the number of bubbles—supporting earlier predictions by Mazzitelli *et al.* [60].

Chapter 4 examines how bubble-induced turbulence evolves with Reynolds number in quiescent viscoelastic fluids. By keeping the Weissenberg number constant and varying the Reynolds number, we observe that at moderate $Re \sim O(100)$, the steep spectral scaling typical of elastic pseudoturbulence begins to weaken and becomes sensitive to gas volume fraction. As Re drops further ($O(10)$ – $O(100)$), the spectrum shifts to a gas fraction dependent k^{-3} scaling, and eventually, as Re decreases to $O(1)$ – $O(10)$, the $k^{-5/3}$

scaling, characteristic of Newtonian fluids, is recovered. These results reveal how the viscous forces, even in viscoelastic fluids, can gradually override elastic effects.

Chapter 5 explores the dynamics of freely rising skirt bubbles across a range of Eötvös and Morton numbers. Using high-speed imaging and PIV, we revisit a problem largely untouched since the 1970s. We measure bubble shapes, velocities, and drag, and compare them with recent DNS results. Our experiments reveal a maximum stable skirt length and, for the first time, capture a secondary toroidal vortex forming within the skirt, visible in the bubble’s moving frame.

Finally, we conclude and summarize the work done in this thesis in Chapter 6.

The appendices present supplementary research undertaken alongside the PhD. Appendix A explores the stability of bubble chains in carbonated drinks. Appendix B examines the mechanics of the squash nick shot, while Appendix C showcases home-lab experiments designed to teach students about viscosity during COVID period.

2 ELASTIC PESUDOTURBULENCE IN POLYMER SOLUTIONS

Abstract We study the effects of polymer additives on pseudoturbulence induced by a swarm of bubbles rising in a quiescent fluid. We find that even in the absence of background shear, beyond a critical polymer concentration, the energy spectra of velocity fluctuations in bubble-induced turbulence decay more steeply with respect to the wavenumber k . This new scaling is significantly steeper than the classical k^{-3} scaling observed for bubbles in Newtonian fluids; it is independent of the gas volume fraction in the inertial limit and occurs within the length scales between the bubble wake length and the bubble diameter. Furthermore, we provide quantitative evidence that the presence of polymers enhances the coherence of the flow, as revealed by Lagrangian stretching statistics, highlighting the significant role of polymer additives in modifying the characteristics of pseudoturbulence. Our findings demonstrate that viscoelastic effects can modulate turbulence even in shear-free environments, offering a new perspective on the role of elastic stresses in complex multiphase flows.

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2.1 Introduction

Buoyancy-driven turbulent flows occur in many natural and industrial processes, such as in the upper ocean layer [61] and bubble column reactors [62] where bubbles play a pivotal role in inducing mixing. The addition of bubbles to turbulent flows has been studied for decades [40] owing to the significant drag reduction [39], heat transfer enhancement [63–65], among other effects [41, 42]. When a swarm of bubbles rise in an otherwise stagnant fluid, the disturbance caused by each bubble interacts with one another giving rise to velocity fluctuations in the continuous liquid phase. This fluctuating motion is referred to as bubble-induced turbulence, more commonly “pseudoturbulence” [66].

A paramount attribute of bubble-induced turbulence is its departure from the homogeneous isotropic Kolmogorov’s turbulence, as summarized by Risso [43]. Lance and Bataille [40] first observed that the addition of bubbles to a developed Newtonian turbulence leads to an energy spectra of velocity fluctuations, $E(k) \sim k^{-3}$, where k is the wavenumber, in the inertial subrange as opposed to the classical $k^{-5/3}$ decay observed in isotropic turbulence. This finding inspired numerous studies, both experimental [44, 59, 67, 68] and numerical [69, 70], which supported the idea that the k^{-3} scaling emerges in a homogeneous bubbly flow from both the spatial and temporal velocity disturbances. Specifically, the spatial contribution dominates at large scales, while the temporal contribution prevails at smaller scales [43].

It was previously argued that, in a statistically steady state, the k^{-3} scaling arises from a spectral balance between viscous dissipation and energy production by bubbles [40]. However, coarse-grained simulations of pseudoturbulence [71], using a spectral decomposition of the energy budget, refuted this argument, showing that no scales exist where energy production and dissipation are in equilibrium. Building on this complexity, an intriguing characteristic of pseudoturbulence is the emergence of the flow agitations in the continuous liquid phase independent of whether the dispersed phase is composed of solid spheres [72] or bubbles [59] for a wide range of Reynolds number and Weber number [70, 73]. This characteristic k^{-3} scaling arises from wake-wake interactions between rising bubbles, as confirmed by point particle simulations, which do not exhibit the k^{-3} scaling due to the absence of wakes behind the bubbles [74]. Recent experimental investigations further confirmed that reducing the bubble Reynolds number (as the wake behind the bubble vanishes) replaces the k^{-3} scaling observed in pseudoturbulence with a $k^{-5/3}$ scaling [3].

For certain applications like porous media flows [50], polymers are employed to achieve maximum drag reduction [51, 52] and heat transfer enhancement [53]. Because of the stretching property of the polymer molecules, efficient mixing can be achieved even in the absence of inertia (very small Reynolds number), referred to as “elastic turbulence” [58, 75]. When the Reynolds number is sufficiently high such that the inertial effects cannot be neglected, the modified turbulence due to the elasticity is referred to as elastoinertial turbulence [49]. A combination of experimental and numerical studies in the elastoinertial turbulence regime has revealed that a scaling exponent of -3 , in contrast to the classical Kolmogorov’s $-5/3$ scaling observed in Newtonian fluids, is a distinctive signature of polymer induced turbulence

[54–57]. Importantly, the -3 scaling observed in Newtonian pseudoturbulence and that in polymer-induced turbulence are coincidental and arise from entirely different mechanisms.

While the modulation of turbulence spectra by polymers [49, 57, 58] and by the bubbles [40, 44, 59] has interested the scientific community for decades, an intriguing open question remains: how do polymer solutions modify the bubble-induced turbulence? In this Letter, we report evidence of a significant modulation in the turbulent energy spectra by a swarm of bubbles rising in quiescent aqueous polymer solutions. We argue that the eddies generated by the wake interactions of bubbles in polymer solutions, which differ significantly from those in the Newtonian fluids [1, 76], play a critical role on the modulation of turbulence spectra. Consequently, there is an enhancement of large-scale coherent structures consistent with the recent experimental [77] and simulation results [53]. This new found insight opens new avenues in the elastoinertial turbulence and has direct relevance for polymer-based drilling slurries used in oil and natural gas explorations [78].

2.2 Experimental setup

In our experiments, mono-dispersed air bubbles are injected into a stagnant fluid at the bottom of the tank through a bank of identical capillary needles of inner diameter 0.6 mm arranged in a hexagonal array as shown in Fig. 2.1. The mean gas volume fraction, α is controlled by adjusting the gas flow rate using a flowmeter. Here, the mean gas volume fraction is measured from the increase in the liquid level after the injection of bubbles, $\alpha = (H_0/\Delta H + 1)^{-1}$, where H_0 is the initial liquid level and ΔH is the liquid level increase. Measuring liquid velocity fluctuations in two-phase gas-liquid flows is particularly challenging due to the dispersed nature of bubbly flows. Conventional hot-wire-based techniques are often used [44, 67, 79], but they are imperfect, as they require filtering out portions of the signal where bubbles interact with the sensor. To overcome these difficulties, the liquid velocity fluctuations was measured by abruptly stopping the bubble formation using a solenoid valve. Once the airflow ceased, the bubbly flow was sharply cut off, allowing the last bubbles to cross the measurement section and leaving the wake region free of bubbles. Then, the wake behind the bubble swarm was studied using high-speed particle image velocimetry (Photron FASTCAM SA5 at 500 frames per second). Since the measurement is carried out in the wake region (after the passage of the bubbles), there was no need to use the fluorescent particles. A green laser beam (532

nm) from an Nd:YAG laser system was used for illuminating the $55\ \mu\text{m}$ diameter tracer particles in the bubble column. The recorded images were then analyzed using PIVLab in MATLAB. The field of view is $38.5\ \text{mm} \times 38.5\ \text{mm}$ (1024×1024 pixels). For the PIV analysis, 32×32 pixels interrogation regions and 50% overlap is used in the first pass and 16×16 pixels interrogation regions with 50% overlap on the subsequent pass is used. Each snapshot is composed of 127×127 vectors. The spatial resolution (physical distance between two neighbouring vectors) is $0.3\ \text{mm}$. Spurious vectors are detected by median test and replaced by interpolating neighbor vectors.

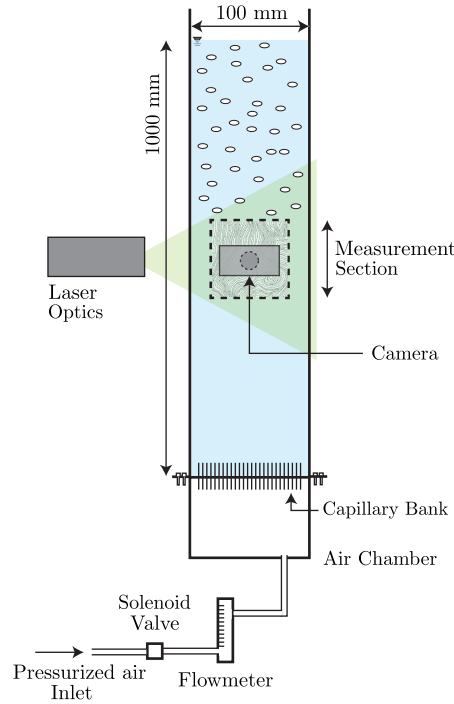


Figure 2.1: Schematic of the experimental setup used in the current study showing the abruptly stopped injection of air bubbles with a solenoid valve to record the flow fields behind the bubble swarm using PIV. The size of the measurement section is $38.5\ \text{mm} \times 38.5\ \text{mm}$, depicted by a dashed box.

2.3 Rheology of fluids

Aqueous solutions of non-ionic polyacrylamide polymer (PAAm) (Sigma Aldrich) with a molecular weight, M_w of $5 \times 10^6\ \text{g/mol}$ were prepared by diluting a stock solution, which was prepared by dissolving polymer in deionized water. The solutions were gently mixed using an overhead mixer for at least 24 hrs to achieve homogeneity. To minimize bubble-bubble coalescence, a small amount of magnesium sulfate salt

(0.025 mol/l) was added to all the solutions. The shear viscosity, density and surface tension of the fluids were measured using ARES-G2 Rheometer (TA Instruments), Density meter (Anton Paar) and Bubble pressure tensiometer (KRUS Scientific Instruments), respectively. The properties of all the fluids used in the experiments are listed in Table 2.1. The intrinsic viscosity, $[\eta]$, is obtained using the Mark-Houwink equation, $[\eta] = \kappa M_w^a$. For the aqueous polyacrylamide solutions used in the experiments, the intrinsic viscosity is determined to be $[\eta] = 10.57 \text{ dl/g}$, here the constants $\kappa = 10^{-4} \text{ dl/g}$ and $a = 0.75$ [80]. The solvent to solution viscosity ratio is given by $\eta_{sp} = \eta_s/\eta$, where $\eta_s = 1.1 \text{ mPa.s}$ is the solvent viscosity. The coil-overlap concentration ($c^* = 1000 \text{ ppm}$), was inferred from log-log plot of the zero-shear viscosity, η_0 , against a wide range of polymer concentration, c as shown in the Fig. 2.2(a). As the polymer concentration increases, the zero-shear viscosity changes the functional dependence from $\eta_0 \propto c^{0.6}$ to $\eta_0 \propto c^{3.3}$ clearly showing the non-linear polymer-polymer chain interactions [81]. To minimize the polymer-polymer chain interactions, we only consider fluids for which the polymer concentration, c , is well below the coil-overlap concentration, c^* . The shear-thinning observed in the dilute regime has a power index of $n \approx 0.95$ and hence ignored, following previous experimental studies [51, 57].

Fluids (in water)	c/c^*	ρ (kg/m ³)	σ (mN/m)	η (mPa.s)	λ_e (ms)	d (mm)	U_b (mm/s)	Re	We	Wi	El
10% Glycerin	-	1003.0	73.23	1.30	≈ 0	2.8 ± 0.2	290	626	3.2	0	0
50 ppm PAAm	0.05	1000.7	74.52	1.13	8.15	3.1 ± 0.2	224	614	2.1	0.6	0.001
100 ppm PAAm	0.1	1001.4	74.35	1.27	29.1	3.4 ± 0.1	222	595	2.3	1.9	0.003
150 ppm PAAm	0.15	1001.5	74.44	1.48	40.6	3.5 ± 0.1	218	502	2.2	2.5	0.005
200 ppm PAAm	0.2	1001.8	74.50	1.65	62.7	3.5 ± 0.2	212	451	2.2	3.8	0.008
300 ppm PAAm	0.3	1002.6	74.41	2.12	86.8	3.5 ± 0.2	206	340	2.0	5.0	0.015

Table 2.1: Physical properties of the fluids: c - polymer concentration, c^* - coil-overlap concentration; ρ - density; σ - surface tension; η - viscosity; λ_e - relaxation time; d - bubble diameter; U_b - bubble velocity; Re - Reynolds number; We - Weber number; Wi - Weissenberg number; El - Elasticity number.

The relaxation time, λ_e , is determined using a dripping-onto-substrate method, following Dinic *et al.* [82]. For this, a pendant drop of fluid is brought into contact with a sessile drop on a glass substrate. This causes an unstable liquid bridge to form between the nozzle (of diameter $2h_0$) and the sessile drop, undergoing a capillary-driven thinning. By visualizing the collapse at 5000 frames per second, the liquid bridge undergoing inertio-capillary (observed before $t - t_c = 0$) and elasto-capillary thinning dynamics (linear regime observed after $t - t_c = 0$) can be determined from the subpixel image analysis using MATLAB

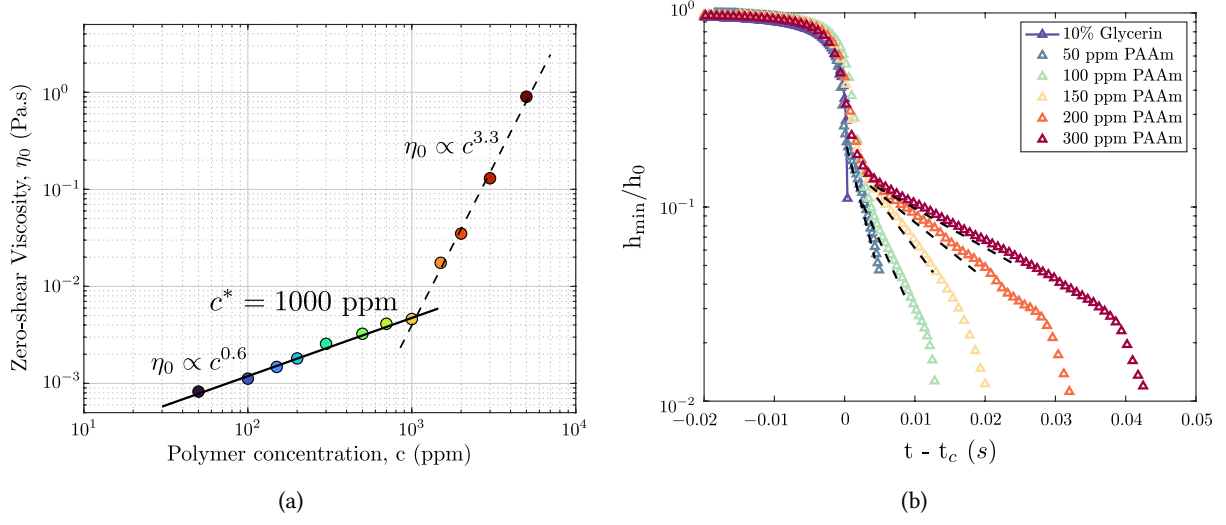


Figure 2.2: (a) Zero-shear viscosity versus a wide range of polymer concentration for aqueous solutions of PAAm. The intersection of solid and dashed lines determine the coil-overlap concentration for PAAm solutions; (b) Evolution of minimum neck radius, $h_{\min}$, normalized by the nozzle radius, h_0 , with the time axis shifted by the transition points, t_c , of different polymer concentration fluids. The relaxation time, λ_e , is determined from the slope of the dashed lines for each polymer concentration.

as shown in the Fig. 2.2(b). Here, t_c marks the transition from the inertio-capillary to elasto-capillary regime [82]. The relaxation time for varying polymer concentration is obtained from elastocapillary thinning dynamics described using

$$\frac{h_{\min}(t)}{h_0} \approx \left(\frac{Gh_0}{2\sigma} \right)^{1/3} \exp[-(t - t_c)/3\lambda_e], \quad (2.1)$$

where G is the elastic modulus.

2.4 Results and discussions

2.4.1 Spatial correlation and second-order structure function

Figure 2.3(a,c) show the snapshots of bubbly flow and Fig. 2.3(b,d) show the dimensionless vorticity fields, $\omega(d/U_b)$, obtained from the bubble swarm wake for the Newtonian fluid and viscoelastic fluid, respectively. We note that the differences in the flow field are not caused due to the difference in the Reynolds number considered. From the instantaneous streamlines overlaid on the normalized vorticity fields it is evident that the flow structures in the wake of the bubble swarm in the polymeric fluid is significantly different from that

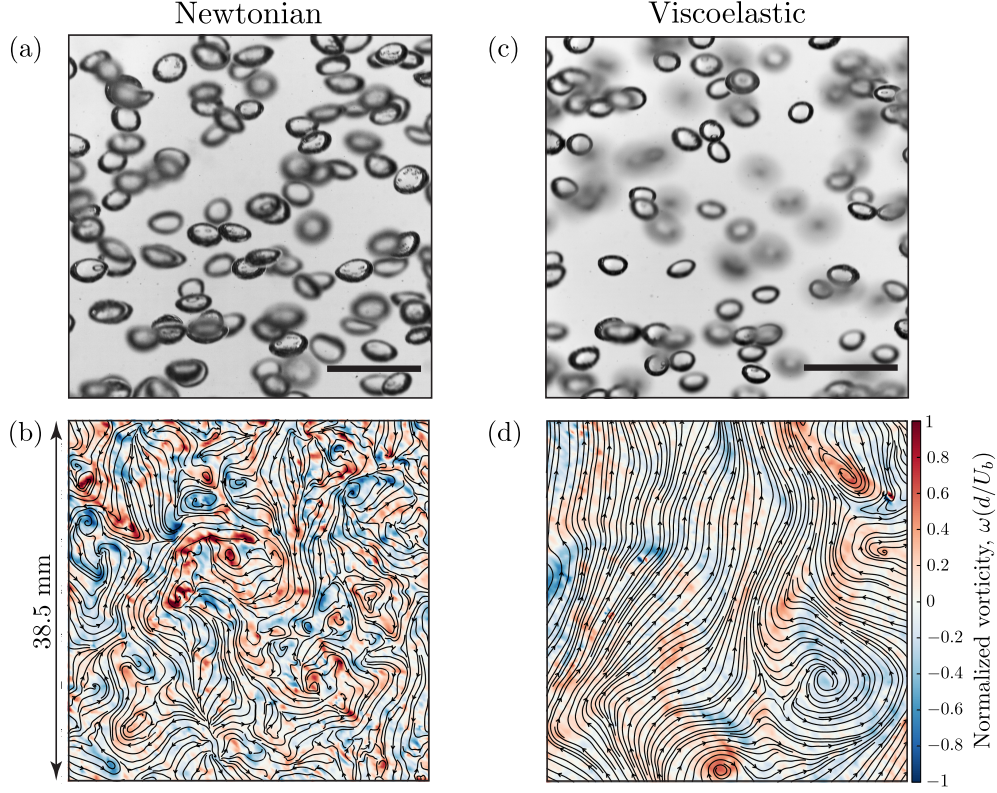


Figure 2.3: Sample snapshots of bubbly flow in (a) Newtonian fluid ($Re = 626$, $Wi = 0$ and $El = 0$) and (c) viscoelastic fluid $Re = 451$, $Wi = 3.8$ and $El = 0.008$) with a gas volume fraction of $\alpha \approx 0.025$. The scale bar is 10 mm. Sample snapshots of the vorticity, normalized by the flow time, d/U_b , obtained from the PIV measurement section behind the bubble swarm at a constant gas volume fraction of $\alpha \approx 0.025$ is shown for the (b) Newtonian ($Re = 626$, $Wi = 0$ and $El = 0$) and (d) viscoelastic fluids ($Re = 451$, $Wi = 3.8$ and $El = 0.008$). Solid lines are the instantaneous streamlines in the wake.

of the Newtonian fluid. We can quantify this observation considering the spatial correlation across a length scale r of the horizontal velocity, u_x , and vorticity magnitude, ω , defined as, $\bar{R}_{xx} = \langle u_x(\mathbf{x}) \cdot u_x(\mathbf{x} + \mathbf{r}) \rangle / \langle u_x^2 \rangle$ and $\bar{R}_\omega = \langle (\omega(\mathbf{x}) \cdot \omega(\mathbf{x} + \mathbf{r})) \rangle / \langle \omega^2 \rangle$, respectively. Here, r lies solely in the horizontal plane and the $\langle \rangle$ represent the average in space. Figures 2.4(a) and 2.4(b) show that the correlation of horizontal velocity and vorticity magnitude increases over longer distances as the polymer concentration increases, at a constant gas volume fraction of $\alpha \approx 0.025$, respectively. The average vortex size can be then estimated by the integral length scale of the vorticity, $L_\omega = \int_0^\infty \bar{R}_\omega dr$. Inset in the Fig. 2.4(b) shows that the average vortex size increases with the Weissenberg number. We note that the spatial correlations over the normalized distance does not change significantly with the gas volume fraction. Overall, this indicates that polymer stresses enhance the prominence of large scale flow structures, in agreement with previous experimental [83] and numerical

simulations [53]. Here, the horizontal velocity is preferred over the velocity magnitude, because the probability density functions (PDFs) of the horizontal velocity fluctuations are symmetric, whereas the vertical velocity fluctuations are positively skewed. In other words, there is an anisotropy in the vertical direction due to the mean motion of bubbles.

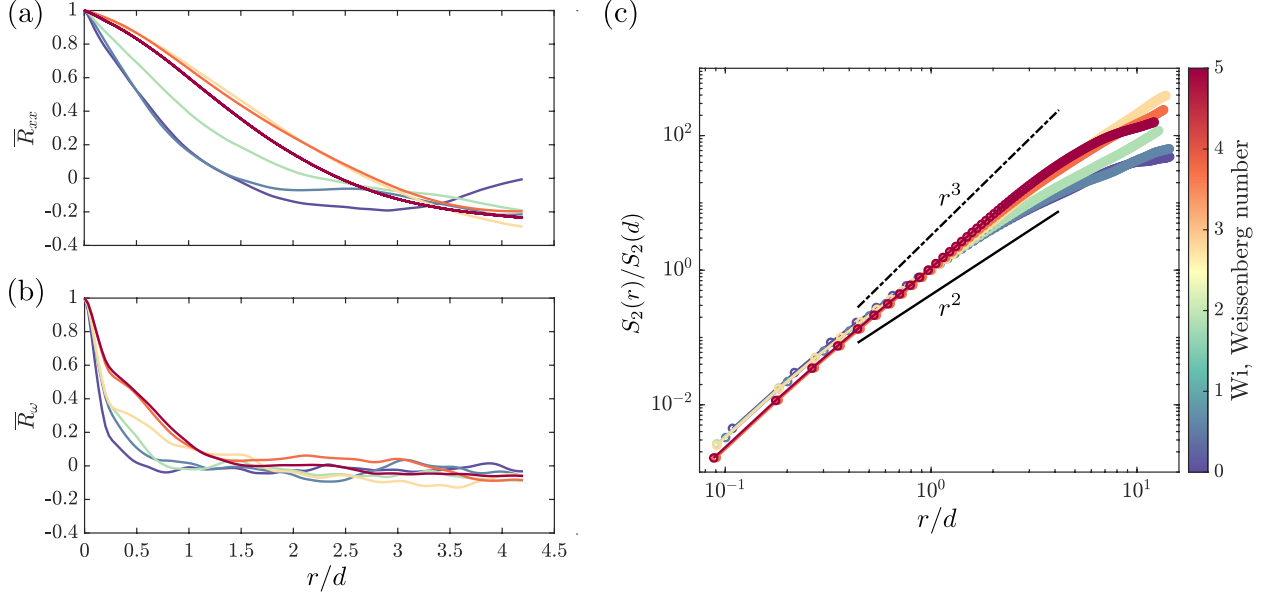


Figure 2.4: Spatial correlation of the (a) horizontal velocity and (b) vorticity magnitude observed in the wake of the bubble swarm for a range of Weissenberg numbers at a constant gas volume fraction of $\alpha \approx 0.025$. When the Weissenberg number is increased, the flow behind the bubbly swarm gets increasingly correlated over longer distance; (c) The normalized second-order structure function, $S_2(r)$ as a function of normalized length scale r for a range of Weissenberg numbers at a constant gas volume fraction of $\alpha \approx 0.025$. The solid and dot-dashed lines correspond to the r^2 and r^3 scaling, respectively. The color of each line in figures (a), (b), and (c) represents the value of the Weissenberg number, as indicated by the color bar on the right in figure (c).

Now, we consider the second-order longitudinal velocity structure function defined as,

$S_2(r) = \langle [u_x(\mathbf{x} + \mathbf{r}) - u_x(\mathbf{x})] \cdot (\mathbf{r}/r)^2 \rangle$ to explore on how the energy spectra will decay [84]. Here the $\langle \rangle$ denotes the spatial average. From the theory of homogeneous and isotropic turbulence for Newtonian incompressible fluids [28], we know that when the second-order velocity structure function scales $S_2(r) \sim r^\beta$, then the energy spectra should scale as $E(k) \sim k^{-(\beta+1)}$. Hence, for the classical Kolmogorov turbulence, the second-order velocity structure function behaves as $S_2(r) \sim r^{2/3}$ in the inertial range and $S_2(r) \sim r^2$ in the dissipative range. Figure 2.4(c) shows the second-order velocity structure function observed in the wake of the bubble swarm as a function of normalized length scale r for a range of Weissenberg number at a

constant gas volume fraction of $\alpha \approx 0.025$. In Newtonian pseudoturbulence, however, because of the bubble wake-wake interactions in the intermediate length scales, instead of the $S_2(r) \sim r^{2/3}$ scaling in the inertial range the second-order structure function scales as $S_2(r) \sim r^2$ [73, 85]. Note that the $S_2(r) \sim r^2$ scaling for the Newtonian pseudoturbulence in the intermediate range is identical to that of the scaling behaviour in the dissipative range for the classical Newtonian Kolmogorov turbulence. As the polymer concentration increases the second-order structure function $S_2(r)$ for pseudoturbulence shows a steepness close to r^3 . However, this can be deceptive as any energy spectra $E(k) \sim k^{-\gamma}$ where γ is greater than 3 will also show r^2 dependence in the second-order structure function, analogous to the two dimensional turbulence [86] and in some instances can only be identified by the higher order structure functions [87]. Thus for the viscoelastic pseudoturbulence we can conjecture that the energy spectra should decay as $E(k) \sim k^{-\gamma}$, where γ is greater than 4.

Probability density function of the velocity fluctuations We now show the statistics of the velocity fluctuations and how they were altered in the presence of polymers. From previous studies [59, 68, 88], it has been shown that the PDFs of the horizontal velocity fluctuations are symmetric and non-Gaussian, whereas for the vertical component, the PDFs are strongly positively skewed (i.e. enhanced probability of upward fluctuations). This is because the positive skewness is the result of the velocity fluctuations in the vicinity of the bubble, which move vertically [59, 88]. Therefore, to capture these effects accurately, it is essential to analyze the velocity field within the bubble swarm rather than in the wake of the bubble swarm.

For that purpose, similar to [73, 89], fluorescent particles within the bubble swarm were added to capture the velocity fluctuations produced in the bubble vicinity. By nature, this particular visualization technique within the bubble swarm is restricted to low gas volume fractions. Figure 2.5 shows the PDF of the horizontal and vertical velocity fluctuations within the bubble swarm normalized by the standard deviation for gas volume fraction of $\alpha = 0.010$ for a Newtonian fluid. The multiple colors correspond to the consecutive instances (blue corresponds to $t = 0$ and red corresponds to $t = 2$ ms) in the decaying agitations left behind the bubble swarm. Both the horizontal and vertical velocity fluctuations PDFs are in good quantitative agreement with the results of Riboux *et al.* [59] who showed that PDFs corresponding to the flow in the vicinity of the bubbles was positively skewed whereas the PDFs corresponding to the wake of the bubble swarm was Gaussian. This was further corroborated by Alm  ras *et al.* [67] from the conditioned

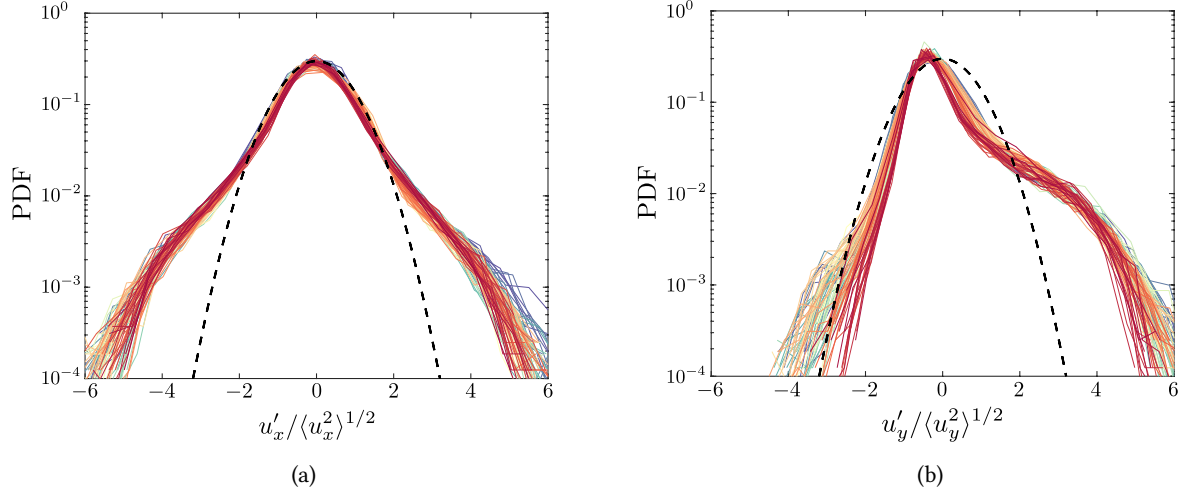


Figure 2.5: PDF of the liquid velocity fluctuations (a) in the horizontal and (b) vertical direction within the bubbly flows in Newtonian fluid ($Wi = 0$) normalized by the standard deviation for $\alpha = 0.010$. The dashed line corresponds to the Gaussian profile. Here, the color gradients, ranging from blue to red, correspond to 2 ms of measurement within the bubble swarm

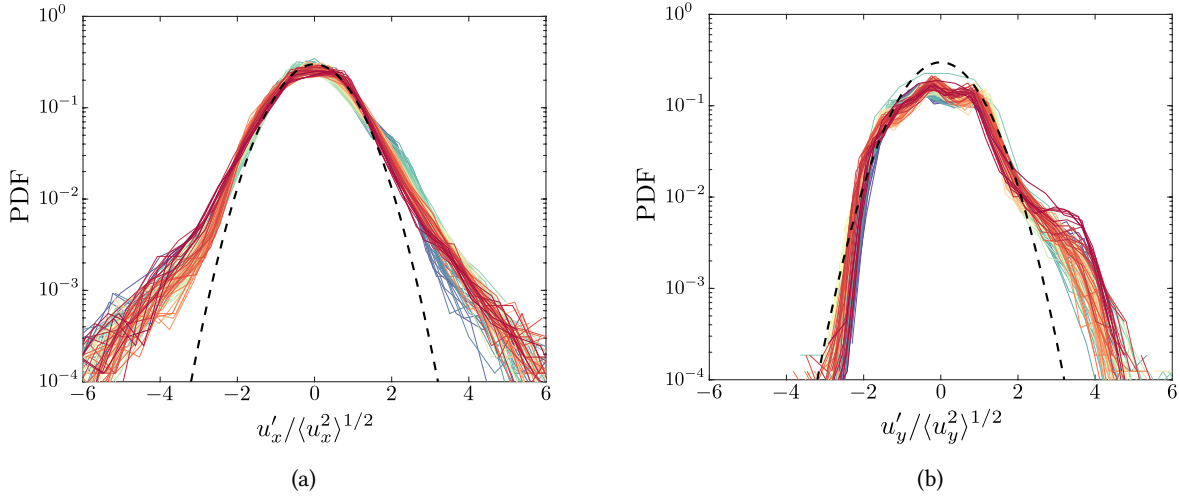


Figure 2.6: PDF of the liquid velocity fluctuations (a) in the horizontal and (b) vertical direction within the bubbly flows in viscoelastic fluid ($Wi = 3.8$) normalized by the standard deviation for $\alpha = 0.010$. The dashed line corresponds to the Gaussian profile. Here, the color gradients, ranging from blue to red, correspond to 2 ms of measurement within the bubble swarm

PDFs of a single bubble in a turbulent flow showing strong skewness in the primary and secondary wake whereas the conditioned PDFs in the far field are Gaussian (See Fig. 14(d) in [67]). Thus, the exponential tail is mainly due to the agitations by the wakes from the rising bubbles and their interactions, leading to a larger probability of upward fluctuations. For the case of viscoelastic fluids, as shown in the Fig. 2.6,

the PDFs of the horizontal and vertical velocity fluctuations are similar in trend to that of the Newtonian fluids. The main difference is that there is a shrinkage in the exponential tail as observed in the case of Newtonian fluid. This can be interpreted as the polymer additives suppressing the exponential tails of velocity distributions by decreasing the occurrence of large scale velocity fluctuations [83].

2.4.2 Variance of the velocity fluctuations

Figure 2.7(a) shows the variance of the liquid velocity fluctuations as a function of gas volume fraction for a range of Weissenberg numbers. We first focus on the case of Newtonian fluids ($Wi = 0$). From their experiments, Alm  ras *et al.* [67] showed that the evolution of liquid agitation in bubbly turbulent flows depends on the so-called critical bubblance parameter, b_c . Here, the bubble parameter, which compares the energy of fluctuations produced by the bubble swarm to the energy of fluctuations produced by the incident turbulence, is given by,

$$b = \frac{V_r^2 \alpha}{u_0'^2}, \quad (2.2)$$

where, u_0' is the incident turbulent fluctuations produced in the absence of bubbles (single-phase flows) and V_r is the relative rising velocity of the bubble. They showed that regardless of the incident turbulent fluctuations (u_0'), the standard deviations of the liquid velocity fluctuations (u_{rms}) showed a non-monotonic evolution with the bubblance parameter, b . The conclusions from their work is that the normalized velocity fluctuations, u_{rms}/u_0' evolves as $b^{0.4}$ when $b < b_c$, where $b_c \approx 0.7$ in their experiments. Whereas for $b > b_c$, the normalized velocity fluctuations increases much faster as $b^{1.3}$.

Note that $b = 0$ corresponds to single-phase flow, whereas $b = \infty$ corresponds to a bubble swarm rising in a quiescent liquid (pseudo-turbulence). In the current experiments, since the bubbles are rising in a quiescent liquid ($b = \infty$), we choose to compare the results using the gas volume fraction, α . Figure 2.7(a) shows the variance of the velocity fluctuations as a function of gas volume fraction for a family of Weissenberg numbers. The velocity fluctuations are determined by $\mathbf{u}' = \mathbf{u} - \langle \mathbf{u} \rangle$. Here $\langle \rangle$ represent the average in space. For brevity, only the vertical velocity fluctuations are shown. It is immediately evident that the variance of the velocity fluctuations for a family of Weissenberg numbers are of the same order. Following the work of Cartellier and Rivi  re [90], the variance of the velocity fluctuations is

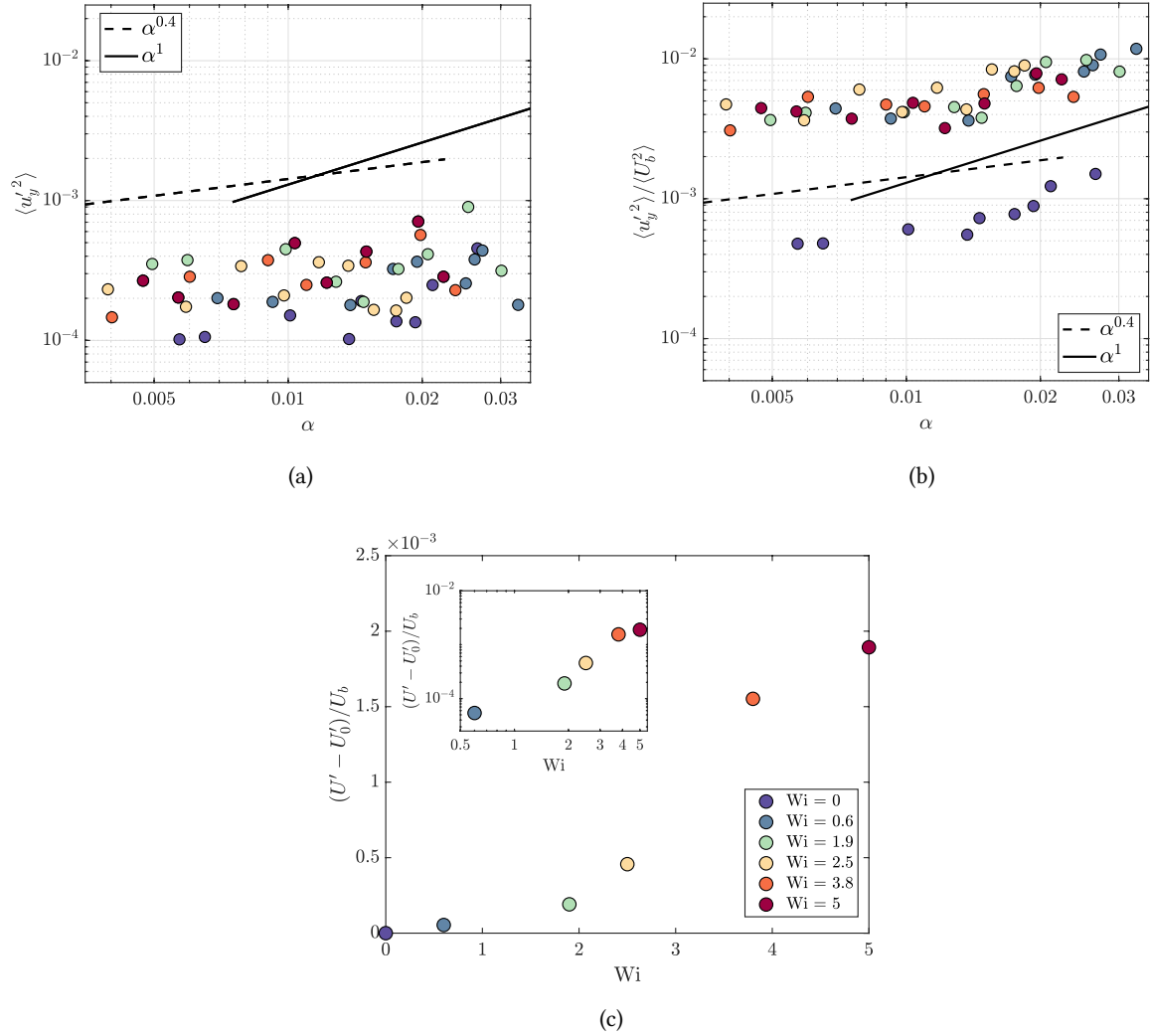


Figure 2.7: (a) Variance of the velocity fluctuations, $\langle u_y'^2 \rangle$, in the vertical direction as a function of the gas volume fraction, α , for a family of Weissenberg numbers; (b) Variance of the velocity fluctuations, $\langle u_y'^2 \rangle$, in the vertical direction normalized by the square of the mean bubble velocity, $\langle U_b^2 \rangle$, as a function of the gas volume fraction, α , for a family of Weissenberg numbers. The dashed and solid lines show functional dependencies of $\langle u_y'^2 \rangle \propto \alpha^{0.4}$ and $\langle u_y'^2 \rangle \propto \alpha^1$, respectively; (c) Difference in the normalized RMS velocity fluctuations relative to the Newtonian case as a function of the Weissenberg number at a constant gas volume fraction of $\alpha \approx 0.02$; Inset shows the same plot in the log-log axis.

normalized by the bubble velocity squared (see Fig. 2.7(b)). Similar to the previous works ([44, 67, 90]), for the Newtonian fluid ($Wi = 0$) two regimes can be observed, separated by a critical gas volume fraction of $\alpha_c \approx 0.015$. For $\alpha < \alpha_c$, the normalized velocity fluctuations increase monotonically as $\langle u_y'^2 \rangle / \langle U_b^2 \rangle \propto \alpha^{0.4}$ (dashed line). Whereas for $\alpha > \alpha_c$, the normalized velocity fluctuations increase much faster as $\langle u_y'^2 \rangle / \langle U_b^2 \rangle \propto \alpha^1$ (solid line). A similar trend is observed for the case of viscoelastic fluids on increasing the Weissenberg

number. However, the normalized velocity fluctuations are an order of magnitude higher compared to that of the Newtonian case. This shows that the turbulence intensity is larger in the presence of elastic effects. This is further quantified using the root-mean-square (RMS) of the velocity fluctuations, given by $U' = \sqrt{(2\langle u_x^2 \rangle + \langle u_y^2 \rangle)/3}$. As shown in Fig. 2.7(c), the difference in the normalized RMS velocity fluctuations relative to the Newtonian case, U'_0 , increases with the Weissenberg number at a constant gas volume fraction $\alpha \approx 0.02$, consistent with the previous studies [91, 92]. A clear transition from the Newtonian case occurs at a critical value of Weissenberg number, $Wi_{cr} \approx 2.5$.

2.4.3 Energy spectra of the velocity fluctuations in Newtonian fluids

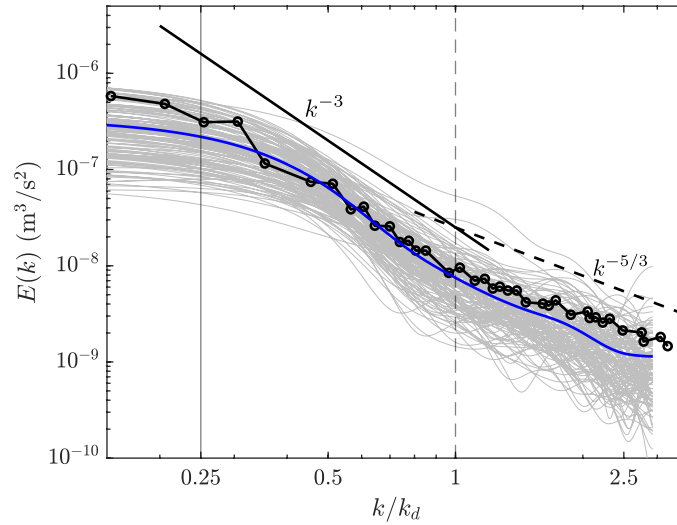


Figure 2.8: Spatial spectrum of the horizontal velocity for each horizontal row of the PIV velocity field for the Newtonian fluid at $Re = 626$ and gas volume fraction, $\alpha \approx 0.0025$ is shown by the gray lines. The solid blue line corresponds to the average energy spectra of the horizontal velocity fluctuations. The abscissa is normalized by the wavenumber corresponding to the bubble diameter, k_d . We overlay the spectrum obtained from experiments by Riboux *et al.* [59] (solid black line with symbols) for $Re = 670$ and gas volume fraction, $\alpha \approx 0.0046$ and find it to be in agreement with our experiments. The solid and the dashed gray line in the abscissa denote the upper and lower boundaries of the k^{-3} spectra observed by [59].

In the current experiments, the energy spectra of horizontal velocity fluctuations is calculated using the Welch method for each horizontal row of horizontal velocity fluctuations obtained from the particle image velocimetry (PIV) of the wake behind the bubble swarm, as detailed in [59]. The average energy spectra of horizontal velocity fluctuation is then obtained by taking the mean of energy spectra of all the rows, as shown in Fig. 2.8.

The average energy spectra of vertical velocity fluctuations in the vertical direction is obtained in the same manner as mentioned above. As established by Riboux *et al.* [59, 69], we have replicated the k^{-3} scaling is observed between the Eulerian integral length scale ($\Lambda = d/C_d$) and the bubble diameter (d) in our benchmark Newtonian fluid bubbly flow experiments, where $C_d = \frac{16}{\text{Re}}\kappa(\text{Re})$ is the drag coefficient of a single rising bubble obtained from [93]. Here $\kappa(\text{Re}) = \frac{16+3.315\text{Re}^{1/2}+3\text{Re}}{16+3.315\text{Re}^{1/2}+\text{Re}}$. We note that the k^{-3} scaling, observed less than a decade in the wavenumber space, is a defining feature of pseudoturbulence. As discussed in the main manuscript, pseudoturbulence originates from the wake interactions between rising bubbles. As a result, its characteristics are limited to the spatial range between the bubble wake length and the bubble diameter. In wavenumber space, k^{-3} scaling is observed when $k \leq k_d$. Note that, at scales smaller than the bubble diameter (wavenumber $k > k_d$), the classical Kolmogorov's power-law decay, $E(k) \sim k^{-5/3}$ is recovered, again, in agreement with previous studies [43, 59, 73].

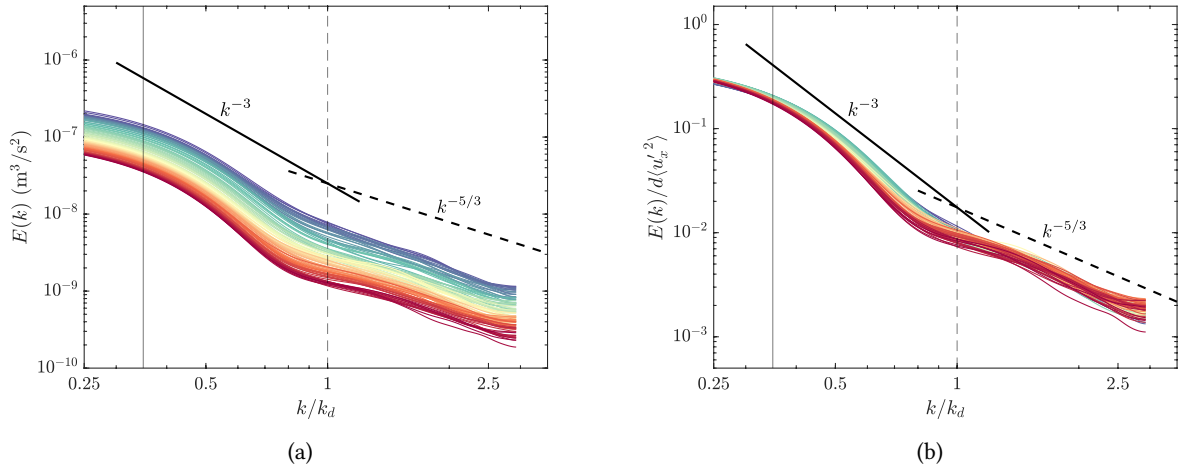


Figure 2.9: (a) Energy spectra and (b) normalized energy spectra of the horizontal liquid velocity fluctuations with $\text{Re} = 626$ and gas volume fraction, $\alpha \approx 0.025$. The multiple colors correspond to the consecutive instances in the decaying agitations left behind the bubble swarm (blue corresponds to $t = 0$ and red corresponds to $t = 20$ ms). The k^{-3} scaling in the intermediate scales (solid black line) and $k^{-5/3}$ scaling in the small scales (dashed black line) in accordance to [71].

Figure 2.9(a) shows the energy spectra of the decaying agitations measured after the bubbles have been switched off. For brevity only the horizontal energy spectra of the liquid velocity fluctuations are presented. It is evident that as the time increases (from blue to red), the agitation decays, hence the energy associated

decreases; however, the shape of the curve remains unchanged. In other words, if the spectra is normalized by the corresponding variance, the spectra collapse into a single curve as shown in the Fig. 2.9(b). Therefore, to be consistent across the different experimental conditions, the flow is measured immediately after the bubble swarm has left the measurement section, where the energy associated is greater.

2.4.4 Energy spectra of the velocity fluctuations in viscoelastic fluids

Time dependence on the spectra of the velocity fluctuations Figure 2.10(a) and (b) show the energy spectra of horizontal and vertical velocity fluctuations normalized by their respective variance and bubble diameter for a range of Weissenberg numbers at a constant gas volume fraction of $\alpha \approx 0.025$. Here the abscissa is normalised by the wavenumber corresponding to the bubble diameter, $k_d = 2\pi/d$. As is well established from decades of research [40, 43, 44, 59, 72, 88], for a bubbly flow in Newtonian fluid ($Wi = 0$), the energy spectra of the velocity fluctuations, $E(k)$, decays as k^{-3} , which is dubbed as the signature of pseudoturbulence for wavenumbers $k \leq k_d$. For different gas volume fractions, normalizing the energy spectra by the respective variance of the velocity fluctuations results in a single curve. This indicates that the velocity fluctuations increase with the gas volume fraction.

A striking difference in the energy spectral decay is observed when the polymer concentration increases ($Wi > 0$). The k^{-3} scaling of the energy spectra (for wavenumber $k \leq k_d$) is gradually replaced by a much steeper scaling of $E(k) \sim k^{-20/3}$ beyond a critical Weissenberg number, $Wi_{cr} \approx 2.5$. Such a steep exponent (approximately -20/3) has not been reported in the context of turbulence modulation by the elasticity. Similar to the Newtonian case, there is no change in the $E(k) \sim k^{-20/3}$ scaling in polymer solutions for a range of gas volume fractions. This is in agreement with the previous works which showed that the energy spectra does not depend on the gas volume fraction or the bubble diameter [44, 59]. Note that, at scales smaller than the bubble diameter (for wavenumber $k \geq k_d$), the classical Kolmogorov's power-law decay is recovered for all cases, irrespective of the Weissenberg number. The compensated energy spectra shows this information in a slightly different manner.

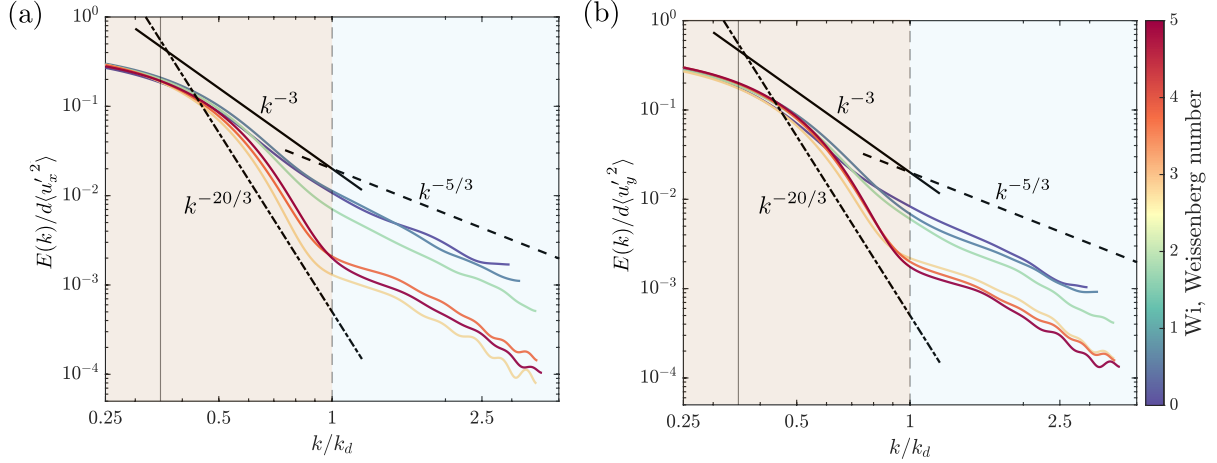


Figure 2.10: (a) Horizontal, and (b) vertical spectra of the liquid velocity fluctuations normalized by the variance and bubble diameter for a range of Weissenberg numbers at a constant gas volume fraction of $\alpha \approx 0.025$. The abscissa is normalized by the wavenumber corresponding to the bubble diameter, k_d . The solid and the dashed gray line in the abscissa denote the upper and lower boundaries of the k^{-3} spectra in the Newtonian fluid. The solid and dot-dashed black lines correspond to the pseudoturbulence scaling for Newtonian (k^{-3}) and viscoelastic ($k^{-20/3}$) fluids at length scales relevant to the bubble wake length and the bubble diameter, respectively. The dashed black line corresponds to Kolmogorov scaling ($k^{-5/3}$) at length scales smaller than the bubble diameter for both Newtonian and viscoelastic cases. The region dominated by the wake interactions (with wavenumber $k \leq k_d$) and by the viscous dissipation effects (with wavenumber $k \geq k_d$) are shaded with light brown and light blue color, respectively.

To understand the scaling observed in the pseudoturbulence of polymeric fluids, we consider the spectral decomposition of the energy balance,

$$\frac{d}{dt}E(k, t) = T(k, t) - T^{[p]}(k, t) - D(k, t) + P(k, t), \quad (2.3)$$

where D represents the kinetic energy dissipation at a wavenumber k , T is the kinetic energy transfer across wavenumber k within the solvent, $T^{[p]}$ is the kinetic energy transfer across wavenumber k between the solvent and polymer molecules, and P is the energy production due to the rising bubbles, respectively. At statistically steady state, the left-hand side of the Eq. 2.3 becomes zero, thus,

$$T(k) - T^{[p]}(k) = D(k) - P(k). \quad (2.4)$$

We first focus on the case of Newtonian fluids ($Wi = 0$), meaning $T^{[p]}(k) = 0$. Using a scale-by-scale energy budget Zamansky *et al.* [71] showed that at large scales ($k/k_d < 0.4$), the rate of energy injected

by the bubbles, $P(k)$, dominates over the viscous dissipation, $D(k) = 2\nu k^2 E(k)$, thus $P(k) \approx -T(k)$, leading to $E(k) \sim k^{-1}$. Whereas, at small scales ($k/k_d > 1$), viscous dissipation becomes dominant, thus $D(k) \approx T(k)$, resulting in the classical Kolmogorov scaling $E(k) \sim k^{-5/3}$. However, in the intermediate scales, contributions from all the terms are non-negligible, thus, $T(k) = D(k) - P(k)$. From numerous experiments and simulations [43, 59, 67, 69, 71, 72, 94], we know that the contribution from all these terms gives rise to $E(k) \sim k^{-3}$ in the intermediate scales, driven by the wake-wake interactions.

In the case of polymeric fluids ($Wi > 0$), the kinetic energy transfer across the wavenumber k between the solvent and polymer molecules, $T^{[p]}(k) \neq 0$. From Fig. 2.10, it is evident that even in the polymeric fluids, the energy spectra at large ($k/k_d \leq 0.4$) and at small scales ($k/k_d \geq 1$) behaves similar to that of the Newtonian case, leading to believe that the $T^{[p]}$ is important only in the intermediate scale. Note that the term $T^{[p]}$ is strictly positive in statistically steady state, as it is equal to the elastic energy dissipated by the polymers [92]. Because of this dissipative nature of the polymers, the net effect of $T^{[p]}$ over all scales is to remove kinetic energy from the solvent and thus the negative sign in the Eq. 2.4 [95]. Thus, the contribution from the $T^{[p]}$ causes the energy spectra to decay more steeply than k^{-3} , leading to a scaling of $E(k) \sim k^{-\gamma}$, where γ is greater than 4. This further confirms the scaling observed from the second-order structure function. From experiments we show that the $\gamma \approx 20/3$ for the elastic pseudoturbulence.

Dependence of gas volume fraction on the spectra of velocity fluctuations For a Newtonian fluid, the average energy spectra, when normalized by the variance of the velocity fluctuations and bubble diameter, collapses into a single curve for different gas volume fractions as shown in the Fig. 2.11. It is to be noted that, unlike the reported temporal power spectral density (refer to [67, 68]), the energy spectra obtained from the PIV images show a k^{-3} scaling at large scales for less than a decade in the horizontal axis. Since the pseudoturbulence k^{-3} scaling is a result of wake-wake interaction, the spatial energy spectra is expected to be observed between the bubble wake length and the bubble diameter ($k \leq k_d$). We report that in the viscoelastic fluids, similar to the Newtonian counter part, the average energy spectra when normalized by the variance of the velocity fluctuations and bubble diameter collapses into a single curve for a family of gas volume fractions with a steeper $k^{-20/3}$ scaling instead of the k^{-3} scaling as shown in the Fig. 2.12.

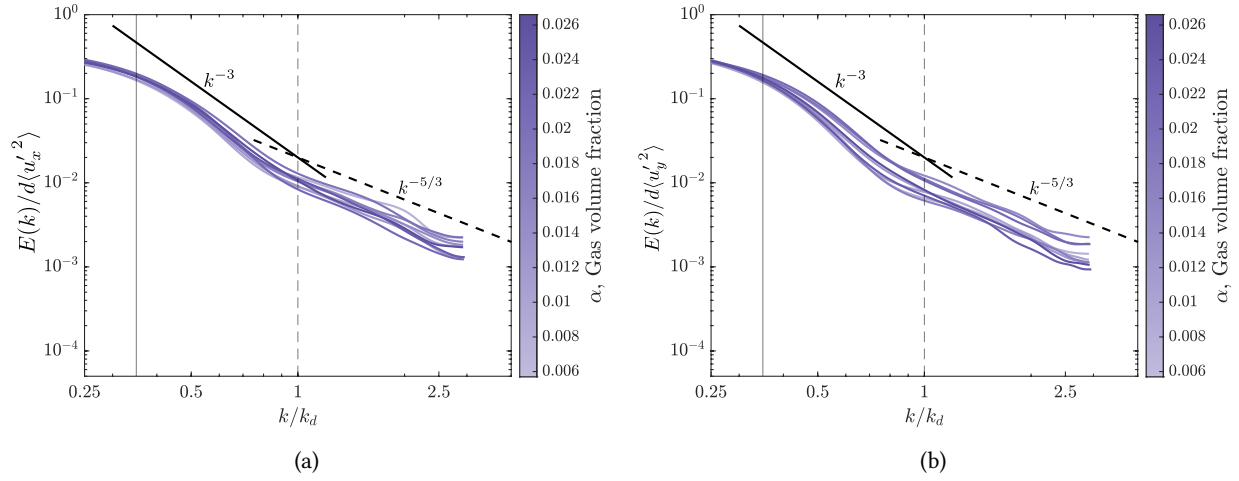


Figure 2.11: (a) Horizontal, and (b) vertical spectra of the liquid velocity fluctuations normalized by the variance and bubble diameter for a family of gas volume fraction in a Newtonian fluid. The abscissa is normalized by the wavenumber corresponding to the bubble diameter, k_d . The solid and the dashed grey line in the abscissa denote the upper and lower boundaries of the k^{-3} power spectral decay.

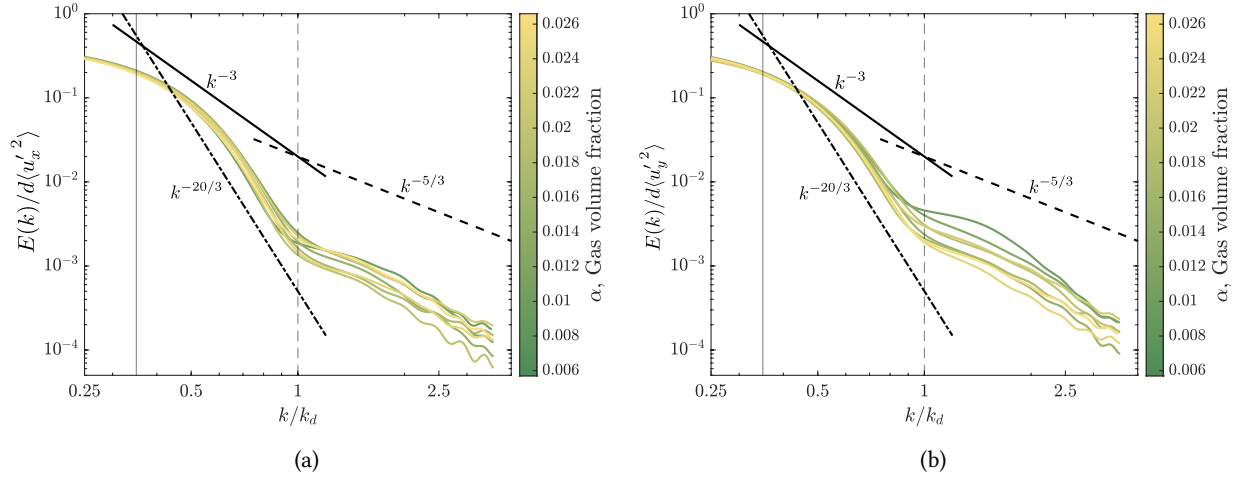


Figure 2.12: (a) Horizontal, and (b) vertical spectra of the liquid velocity fluctuations normalized by the variance and bubble diameter for a family of gas volume fraction in viscoelastic fluid with a Weissenberg number of 3.8. The abscissa is normalized by the wavenumber corresponding to the bubble diameter, k_d . The solid and the dashed grey line in the abscissa denote the upper and lower boundaries of the k^{-3} power spectral decay in the Newtonian fluid.

Figure 2.13 shows the compensated energy spectra of the (a) horizontal and (b) vertical liquid velocity fluctuations showing the emergence of k^{-3} scaling for a range of Weissenberg numbers at a constant gas volume fraction $\alpha \approx 0.025$, respectively. The abscissa is normalized by the wavenumber corresponding to the bubble diameter, k_d . We note that for $Wi < Wi_{cr}$, the k^{-3} scaling is observed for wavenumbers, $k < k_d$.

Furthermore, it is evident from the Fig. 2.13(c) and (d) that for $Wi \geq Wi_{cr}$, the $k^{-20/3}$ scaling is observed for wavenumbers, $k < k_d$.

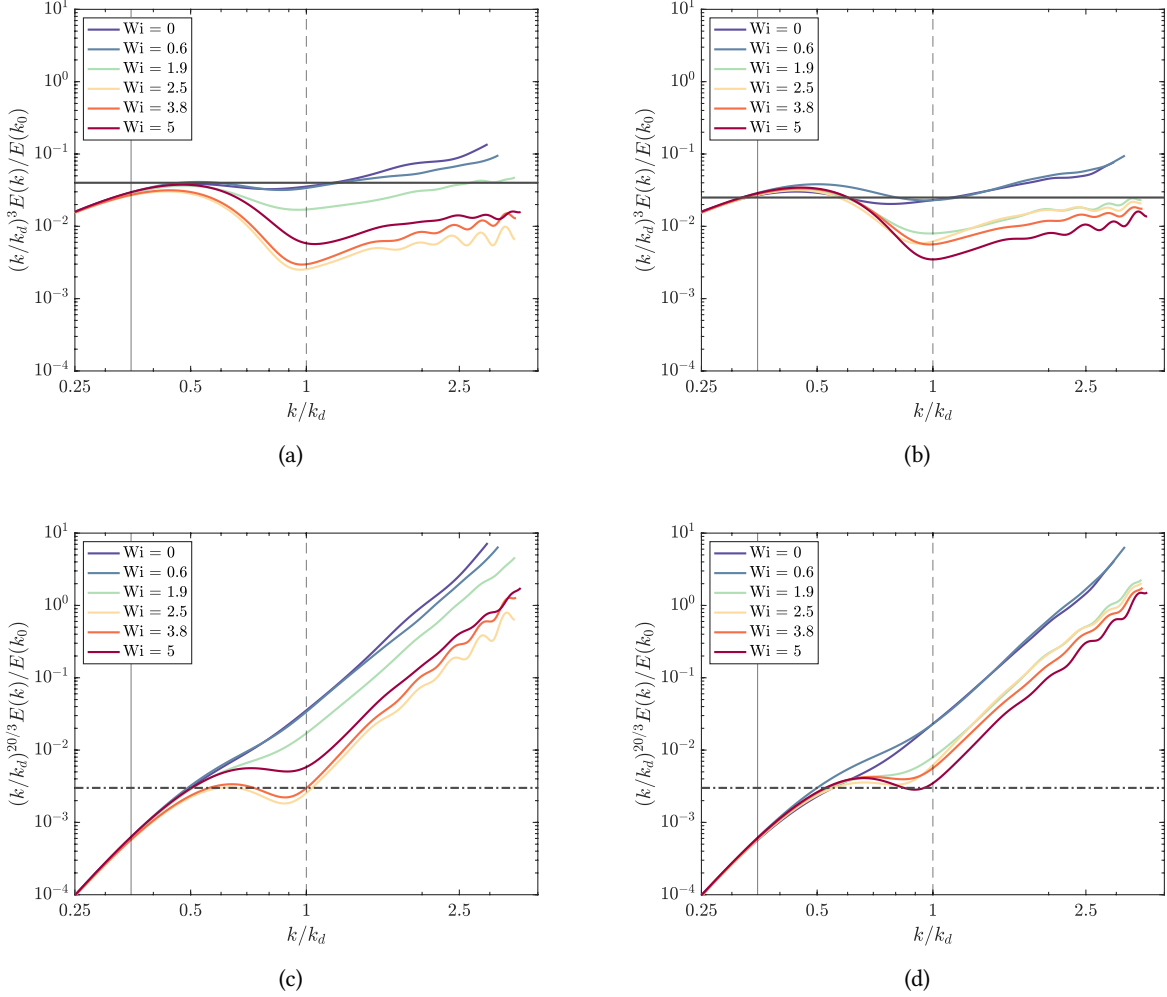


Figure 2.13: Compensated (a) horizontal, and (b) vertical spectra of the liquid velocity fluctuations showing the emergence of k^{-3} scaling for a range of Weissenberg numbers at a constant gas volume fraction $\alpha \approx 0.025$. Compensated (c) horizontal, and (d) vertical spectra of the liquid velocity fluctuations showing the emergence of $k^{-20/3}$ scaling for a range of Weissenberg numbers at a constant gas volume fraction $\alpha \approx 0.025$. The abscissa is normalized by the wavenumber corresponding to the bubble diameter, k_d . The solid line and the dot-dashed line denote the k^{-3} and $k^{-20/3}$ scaling, respectively.

2.4.5 Effect of Weissenberg number on the dissipation rate

The turbulent dissipation rate is defined as,

$$\epsilon = 2\nu\langle s_{ij}s_{ij}\rangle, \quad (2.5)$$

where ν is the kinematic viscosity and s_{ij} is the fluctuating rate of strain. To estimate the dissipation rate, ϵ , using two velocity components from the PIV data, we use the local isotropic assumption [96],

$$\epsilon = \nu \left\langle 4 \left(\frac{\partial u'_x}{\partial x} \right)^2 + 4 \left(\frac{\partial u'_y}{\partial y} \right)^2 + 3 \left(\frac{\partial u'_x}{\partial y} \right)^2 + 3 \left(\frac{\partial u'_y}{\partial x} \right)^2 + 4 \left(\frac{\partial u'_x}{\partial x} \frac{\partial u'_y}{\partial y} \right) + 6 \left(\frac{\partial u'_x}{\partial y} \frac{\partial u'_y}{\partial x} \right) \right\rangle. \quad (2.6)$$

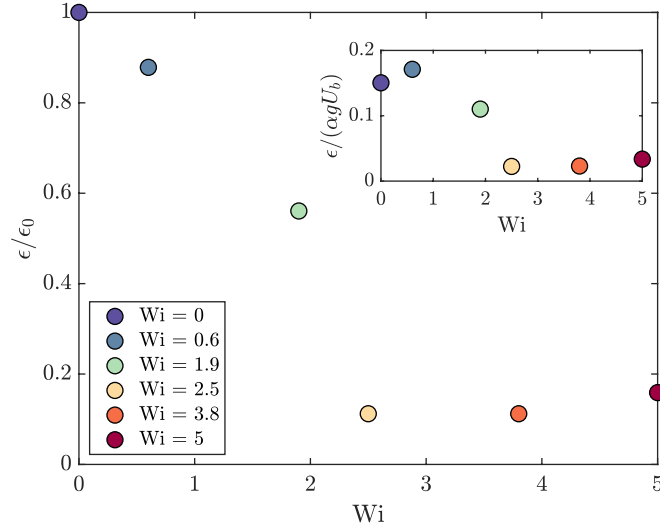


Figure 2.14: Dissipation rate, normalized by its Newtonian counterpart, as a function of the Weissenberg number at a constant gas volume fraction of $\alpha \approx 0.025$; Inset shows the dissipation rate normalized by the energy injected through the buoyancy force.

Figure 2.14 illustrates the energy dissipation rate, normalized by its Newtonian counterpart (ϵ_0), as a function of the Weissenberg number (Wi) at a constant gas volume fraction $\alpha \approx 0.025$. For values below the critical Weissenberg number ($Wi_{cr} \approx 2.5$), the normalized dissipation rate decreases rapidly. Beyond this threshold, the dissipation rate saturates to 15% of the Newtonian case. This highlights the dissipative characteristics of the polymers, consistent with the previous studies [97–100]. The total average energy injected by the bubbles can be approximated as the work done by the buoyancy force, $P_{total} \sim \alpha g U_b$. However, only a fraction of this total energy is effectively transferred into the wakes [43, 71]. The inset in

Fig. 2.14 shows the dissipation rate normalized by the total power injected into the system by the bubbles, further emphasizing the role of polymers in the dissipation process.

2.4.6 Finite Time Lyapunov Exponent

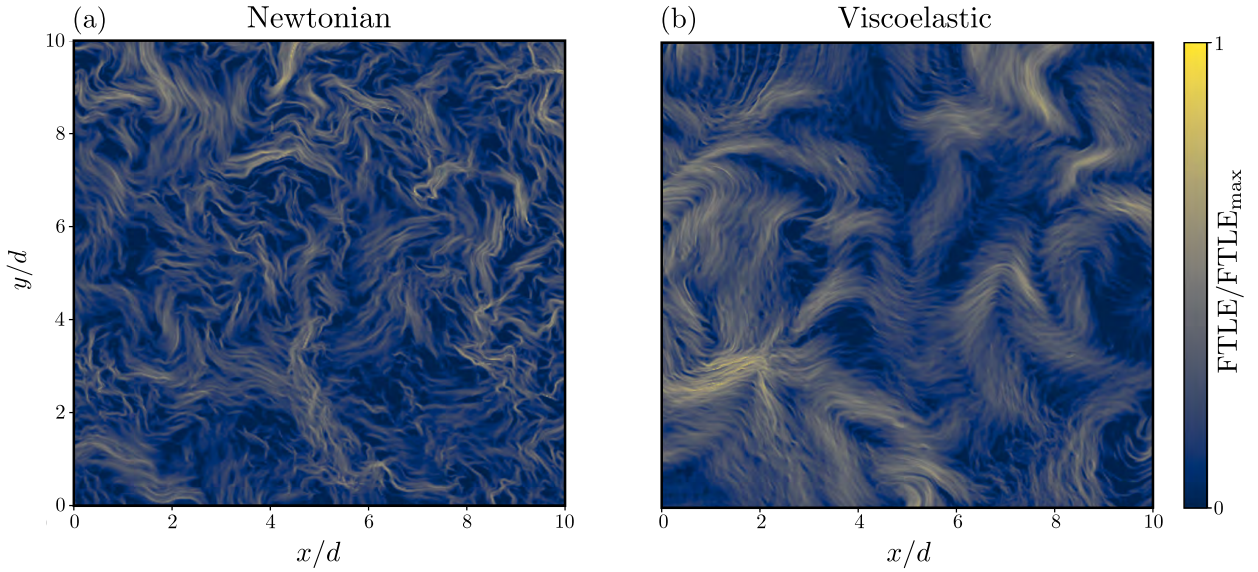


Figure 2.15: Normalized FTLE field at the same dimensionless flow time interval, $t^* = 10$ right after the bubbles have left the measurement section in (a) Newtonian ($Wi = 0$) and (b) viscoelastic fluid ($Wi = 3.8$). In the contour, 0 corresponds to the trenches and 1 corresponds to the ridges. Notice the suppression of small scale structures and enhancement of large scale structures in the presence of polymer.

Compensated energy spectra of velocity fluctuations Motivated by recent work in relating the polymeric stress to the Lagrangian stretching fields [101], we calculate the Lagrangian coherent structures (LCS) [102, 103] to understand the effect of elasticity on the flow structures. In the present study, to further demonstrate the enhancement in the large scale turbulence over the small scale turbulence, the coherent structures are determined using finite time Lyapunov exponent (FTLE). In other words, when two states (or particles dispersed in a flow) separated by an infinitesimally small distance, δ_0 , the distance between these two states evolves as $\delta_0 e^{\lambda_L t}$, where λ_L is the Lyapunov exponent. Thus, the FTLE field measures particle separation rate in the flow. This is helpful in understanding chaotic mixing of fluid elements over extended times [104]. Since FTLE is sufficient to locate the repelling and attracting LCS [105], the forward FTLE fields is determined using the TBarrier tool [106]. As noted by Peng and Dabiri [107], dynamical systems approach to identify coherent structures can be extended to multiphase flows provided the particle trajectories can

be calculated from the equations of motion or directly measured from experiments. Calculating FTLE of the flow fields in the bubble swarm wake provides insights on how a scalar field or a Stokesian seeding particles would evolve. Thus, with the velocity data known, the particle trajectories over time, $\mathbf{x}(t; t_0, \mathbf{x}_0)$, are determined from their initial position, $\mathbf{x}_0$ and initial time, t_0 . The flow map, $\mathbf{F}_{t_0, t_1}(\mathbf{x}_0) = \mathbf{x}(t; t_0, \mathbf{x}_0)$ is defined from these particle trajectories. The FTLE field over the time interval t_0 to t_1 is defined as

$$\text{FTLE}_{t_0, t_1}(\mathbf{x}_0) = \frac{1}{2|t_1 - t_0|} \log \lambda_{\max}(\mathbf{C}_{t_0, t_1}(\mathbf{x}_0)) \quad (2.7)$$

where $\lambda_{\max}$ is the largest eigenvalue of the right Cauchy-Green strain tensor, $\mathbf{C}_{t_0, t_1} = [\nabla \mathbf{F}_{t_0, t_1}]^T \mathbf{F}_{t_0, t_1}$. The time interval to determine the FTLE field is chosen to be the same dimensionless flow time $t^* = tU_b/d$ for all the fluids considered in the experiments. Figure 2.15(a) and (b) shows the normalized FTLE fields in the wake region behind the swarm of bubbles in Newtonian and viscoelastic fluid, respectively. The contours of the normalized FTLE fields, a value of 0 corresponds to trenches while 1 corresponds to ridges. Ridges in the forward FTLE are associated with the regions of high shear i.e., the regions of intense mixing and stretching in the flow [104]. Compared to that of the Newtonian FTLE field, it is immediately clear that the presence of polymers significantly changes the regions of high shear. In other words, the small scale fine structures are smoothened in the presence of polymers. This confirms that the spatio-temporal evolution of the material regions in the viscoelastic fluid becomes more coherent at large scales [108].

Probability distribution of the Lyapunov exponent Figure 2.16(a) illustrates the skewness in the probability distribution of the Lyapunov exponent, normalized by the corresponding mean, further emphasizing quantitatively that the large-scale flow structures become more coherent. Additionally, the mean Lyapunov exponent, $\langle \lambda_L \rangle$ is related to the polymer stretching rate [101]. The polymeric response becomes significant when the local stretching rate exceeds the inverse of the relaxation time, $\langle \lambda_L \rangle > 1/\lambda_e$. This is the coil-stretch transition [109]. In our experiments, we stayed way below coil-overlap concentration, c^* , to avoid non-linear polymer-polymer chain interaction. Thus, the normalized Lagrangian lyapunov exponent, $\langle \lambda_L \rangle \lambda_e < 1$. Figure 2.16(b) shows the Lyapunov exponent rescaled with polymer relaxation time as a function of the Weissenberg number. Inset shows that as the polymer concentration increases, the mean stretching field, $\langle S \rangle = e^{\langle \lambda_L \rangle \lambda_e}$ increases. Since the Lagrangian stretching field is intrinsically related to

the polymer stress, the dissipative nature of the polymer results in the steeper decay in the energy spectra observed in the current study.

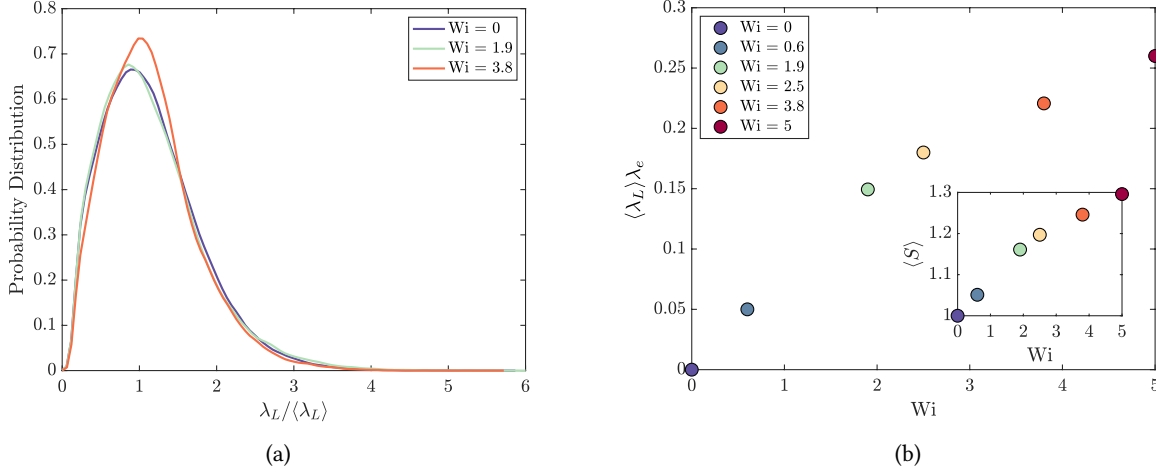


Figure 2.16: (a) Probability distribution of the normalized Lyapunov exponent; (b) Mean Lyapunov exponent rescaled with polymer relaxation time as a function of the Weissenberg number. Inset: Mean stretching field as a function of the Weissenberg number.

2.5 Conclusions

In this Letter, we have demonstrated for the first time that the addition of polymer additives significantly alters the bubble-induced turbulence, opening new avenues for investigating elasto-inertial turbulence [49]. Our findings suggest that the dissipative nature of the polymers results in a more rapid decay of the energy spectrum, exceeding the typical k^{-3} scaling expected for the pseudoturbulence. As the polymer concentration is increased, the flow structure is correlated over longer distances and the energy spectra steepens from the classical $E(k) \sim k^{-3}$ and saturates at $E(k) \sim k^{-20/3}$ indicating an enhancement of energy transfer at intermediate length scales. Notably, the $-20/3$ exponent has not been observed for the turbulence of viscoelastic fluids to date; however, such a large energy decay for elastic pseudoturbulence lacks theoretical insights. Evidence from our experiments suggests that the $-20/3$ scaling exponent results from the synergistic contribution due to the presence of both bubbles and polymers. Given that bubbles in Newtonian fluids enhance heat transfer by nearly 20 times [64] and polymers alone increase heat transfer by 50% [53], the evidence presented here suggests that combining bubbles and polymer solutions could result in a dramatic enhancement of heat transfer efficiency.

3 VELOCITY FLUCTUATIONS FOR BUBBLY FLOWS AT SMALL RE IN NEWTONIAN FLUIDS

Abstract We experimentally investigate the effect of Reynolds number (Re) on the turbulence induced by the motion of bubbles in a quiescent Newtonian fluid at small Re. The energy spectra, $E(k)$, is determined from the decaying turbulence behind the bubble swarm obtained using particle image velocimetry (PIV). We show that when $Re \sim O(100)$, the slope of the normalized energy spectra is no longer independent on the gas volume fraction and the k^{-3} subrange is significantly narrower, where k is the wavenumber. This is further corroborated using second-order longitudinal velocity structure function and spatial correlation of the velocity vector behind the bubble swarm. On further decreasing the bubble Reynolds number ($O(1) < Re < O(10)$), the signature k^{-3} of the energy spectra for the bubble induced turbulence is replaced by $k^{-5/3}$ scaling. Thus, we provide experimental evidence to the claim by Mazzitelli *et al.* [60] that at low Reynolds numbers the normalized energy spectra of the bubble induced turbulence will no longer show the k^{-3} scaling because of the absence of bubble wake and that the energy spectra will depend on the number of bubbles, thus non-universal.

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3.1 Introduction

When a swarm of bubbles rises in an otherwise stagnant fluid due to the motion of bubbles disturbances are created in the surrounding fluid giving rise to velocity fluctuations. At a moderate to high Reynolds number, Re, these velocity fluctuations in the wake behind the bubbles interacts with one another giving rise to the emergence of k^{-3} scaling of the energy spectra, $E(k)$, of velocity fluctuations. This signature k^{-3} scaling of the energy spectra instead of the classical $k^{-5/3}$ Kolmogorov's scaling is regarded as the bubble induced turbulence, also referred to as pseudoturbulence [43].

Lance and Bataille [40] were the first to observe the emergence of k^{-3} scaling of energy spectra, $E(k)$, with respect to the wavenumber, k for the bubble induced turbulence in Newtonian fluids. They argued that in a spectral space, the balance between the energy produced by the motion of bubbles and the viscous dissipation in a statistically steady state gives rise to the emergence of k^{-3} scaling. Following this pioneering work, a number of numerical [41, 110, 111] and experimental [44, 67–69, 112] works supported the emergence of k^{-3} scaling for both the spatial and temporal velocity fluctuations generated by the bubble motion [43]. Amoura *et al.* [72] showed experimentally that irrespective of the dispersed phase, be it bubbles or random fixed solid spheres, the velocity fluctuations generated in the continuous phase gives rise to the k^{-3} scaling. Pandey *et al.* [113] identified the coexistence of Kolmogorov’s turbulence with the bubble induced turbulence for a wide range of Reynolds number and Galilei number (ratio of buoyancy to viscous forces). Recently, Zamansky *et al.* [71] proposed that the k^{-3} subrange of the energy spectra results from the mean shear rate imposed by the bubbles, ruling out the speculation by [40] that the balance between the spectral production and the spectral dissipation.

Experimentally determining the liquid velocity fluctuations in two-phase gas-liquid flows is challenging. Due to the dispersed nature of the bubbly flow, laser-based techniques within the bubble swarm can only be used for very dilute flows. On the other hand, the hot-wire-based techniques [44, 67, 79], are far from perfect as their use implies elaborate signal processing. To overcome these difficulties, Riboux *et al.* [59] measured the liquid velocity fluctuations by abruptly stopping the bubble formation using a solenoid valve thus leaving the wake behind the bubble swarm free of bubbles to be analyzed using particle image velocimetry (PIV). They showed that the k^{-3} scaling, independent of the gas volume fraction and bubble diameter, is observed between the Eulerian length scale ($\Lambda = D/C_d$) and the bubble diameter (D), where C_d is the drag coefficient of a single rising bubble. Later Risso [43] modified the Eulerian length scale, $\Lambda = D/\sqrt{C_d \text{Re}}$ to include the Reynolds number.

An intriguing question is to explore what happens to the energy spectra of the bubbly flows at lower Reynolds number [111, 114]. When $\text{Re} < 20$, the bubbles will not have a significant wakes behind them [115, 116]. A clue on what to expect for smaller Reynolds numbers was summarized by Mazzitelli *et al.* [60], who conducted numerical simulations for bubbly Newtonian fluids considering bubbles as point particles. Specifically, they did not observe the k^{-3} scaling. Thus concluding the essentiality of the wakes. They further argued that the energy spectrum slope for bubbly flows with little to no wakes behind them will

depend on the number of bubbles and will therefore be non-universal [60, 74]. Most surprisingly, to our knowledge, there are only a few experimental studies in this regime [44, 79, 90]. Cartellier and Rivière [90] studied bubble induced agitations at $Re \sim O(1)$; however, to delay the onset of large scale instabilities (transition into heterogeneous bubbly regime) an inner tube within the bubble column was used to create the liquid flow rate due to the gas lift. Further, they did not address the nature of energy spectra of liquid velocity fluctuations.

In the present study we will focus only on the homogeneous bubbly regime to address the effect of Reynolds number on the bubble induced turbulence. Using the particle image velocimetry technique proposed by Riboux *et al.* [59], the liquid velocity fluctuations are determined to calculate the energy spectra. We find that as the Reynolds number decreases the slope of the energy spectra emerges as $k^{-5/3}$ instead of the k^{-3} scaling observed for the bubble induced turbulence. Further, we show that this slope of the energy spectra depends on the number of bubbles in the column. Understanding the spectral structure fluid velocity fluctuations in two-phase flows at low Re could be justified by their relevance in modern applications such as microfluidics [117] and hydrogen production [118].

3.2 Experimental setup and methods

3.2.1 Experimental setup

Figure 3.1(a) shows the schematic of the experimental setup used in the present study. It consists of a transparent acrylic tank of height 1000 mm with a cross-section of 100 mm $\times$ 50 mm. Monodispersed air bubbles are injected at the bottom of the tank through a removable capillary bank. The capillary bank is custom made with identical capillaries of inner diameter 0.6 mm in tandem with a secondary capillary of inner diameter 0.25 mm, arranged in a hexagonal array to increase the number of identical capillaries per unit area [44]. The tandem arrangement provides the sufficient hydraulic resistance through the capillaries such that individual bubbles are formed in a quasi-steady manner, thereby avoiding the generation of gas jets [119]. The gas flow rate is adjusted using a needle valve.

The velocity fluctuations in the liquid phase is measured from the wake behind the bubble swarm, following the method used by Riboux *et al.* [59]. The airflow is abruptly stopped by using a rapid solenoid valve. Then, the region behind the bubble swarm is studied using high-speed particle image velocimetry

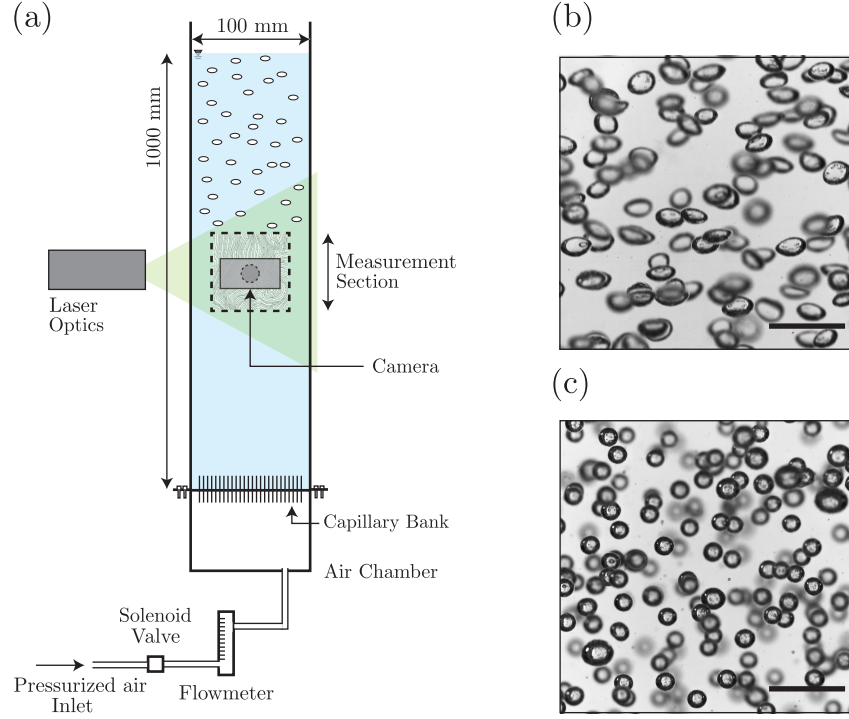


Figure 3.1: (a) Schematic of the experimental setup used in the current study. Using a solenoid valve, the formation of bubbles are abruptly stopped and the wake behind the bubble swarm is visualized using PIV in the measurement section. The size of the measurement section is 40 mm $\times$ 40 mm. A sample snapshot of a bubbly flow with gas volume fraction of $\alpha \approx 0.025$ in Newtonian fluid with viscosity (b) $Re = 626$ and $We = 3.2$, and (c) $Re = 6$ and $We = 0.5$, respectively. The scale bar is 10 mm.

(Photron FASTCAM SA5 at 500 frames per second). The recorded images were then analyzed using PIVLab in MATLAB. For the PIV analysis, 32×32 pixels interrogation regions in the first pass and 16×16 pixels interrogation regions in the second pass with 50% overlap on the subsequent pass is used. The physical distance between two neighbouring vectors is 0.3 mm. Spurious vectors are detected by median test and replaced by interpolating neighbor vectors. From the 2D PIV data, the liquid velocity fluctuations in the bubble swarm wake are obtained as follows $\mathbf{u}' = \mathbf{u} - \langle \mathbf{u} \rangle$. Here $\langle \rangle$ represent the average in space. The energy spectra, $E(k)$, of the velocity fluctuations are determined from the decaying turbulence behind the bubble swarm following [59]. The energy spectra of horizontal and vertical velocity fluctuations are then obtained using the Welch method by averaging the energy spectra of each rows and columns, respectively. To determine the velocity fields within the bubbly swarm at low gas volume fraction, fluorescent particles with an orange filter in the camera was used. The details of the fluorescent particle imaging technique can be found from our earlier study [1].

Fluids (in water)	ρ (kg/m ³)	σ (mN/m)	μ (Pa.s)	D (mm)	U (mm/s)	η (mm)	Re	We
● 10% Glycerin	1003.0	73.23	0.001	2.8 ± 0.2	290	0.10	626	3.2
● 50% Glycerin	1140.6	72.38	0.005	2.6 ± 0.1	243	0.43	144	2.1
● 60% Glycerin	1142.1	71.97	0.007	2.6 ± 0.1	206	0.47	87	1.8
● 75% Glycerin	1195.3	70.07	0.029	2.5 ± 0.2	114	1.15	11	0.6
● 85% Glycerin	1224.0	70.04	0.048	2.3 ± 0.1	109	1.45	6	0.5

Table 3.1: Physical properties of the fluids: ρ - density; σ - surface tension; μ - viscosity; D - average bubble diameter; U - average bubble velocity; η - Kolmogorov's length scale; Re - Reynolds number; We - Weber number.

3.2.2 Test fluids

To prepare viscous Newtonian fluids, water-glycerin mixtures were used. The properties of the fluids used in the study, shear viscosity, density and surface tension were measured using ARES-G2 Rheometer (TA Instruments), Density meter (Anton Paar) and Bubble pressure tensiometer (KRUS Scientific Instruments), are listed in Table 3.1. To reduce bubble coalescence, a small amount of magnesium sulfate salt (0.05 mol/l) was added to all the fluids used in the study, following Lessard and Zieminski [120]. The relevant dimensionless numbers used in the current study are (i) Reynolds number, $Re = \rho U D / \mu$, where U is the average bubble velocity, D is the average bubble diameter, ρ is the fluid density, and μ is the fluid viscosity, and (ii) Weber number, $We = \rho U^2 D / \sigma$, where σ is the surface tension. The average bubble diameter and velocity were obtained from the probability distribution obtained from the image processing. The Kolmogorov's length scale was calculated as $\eta = (v^3 / \epsilon)^{1/4}$, where $v = \mu / \rho$ is the kinematic viscosity and ϵ is the dissipation rate. In turn, ϵ determined from the fluctuating rate of strain tensor using local isotropic assumption following Xu and Chen [96]. The mean gas volume fraction, $\alpha = (H_0 / \Delta H + 1)^{-1}$, is measured from the increase in the liquid level after the injection of bubbles, where H_0 is the initial liquid level and ΔH is the liquid level increase.

3.3 Results and discussion

3.3.1 Probability density functions of the velocity fluctuations

Figure 3.2 shows the probability density functions (PDFs) of the horizontal and vertical velocity fluctuations within the bubble swarm normalized by the corresponding standard deviations for a bubble Reynolds number, $Re = 6$ at a constant gas volume fraction of $\alpha \approx 0.010$. The PDFs of the vertical velocity fluctuations are positively skewed whereas the PDFs of the horizontal velocity fluctuations are symmetric and non-Gaussian, similar to that of the bubbly flows at high Reynolds number [59, 67, 88]. Here, determining the PDFs within the bubble swarm is essential as the velocity fields in the wake of the bubble swarm will be symmetric and Gaussian [73, 89]. Though there is little to no wake for low Reynolds number bubbles, here, the exponential tail and the positive skewness in the vertical velocity fluctuations are attributed to the large fluctuations in the vicinity of the bubbles [59, 67].

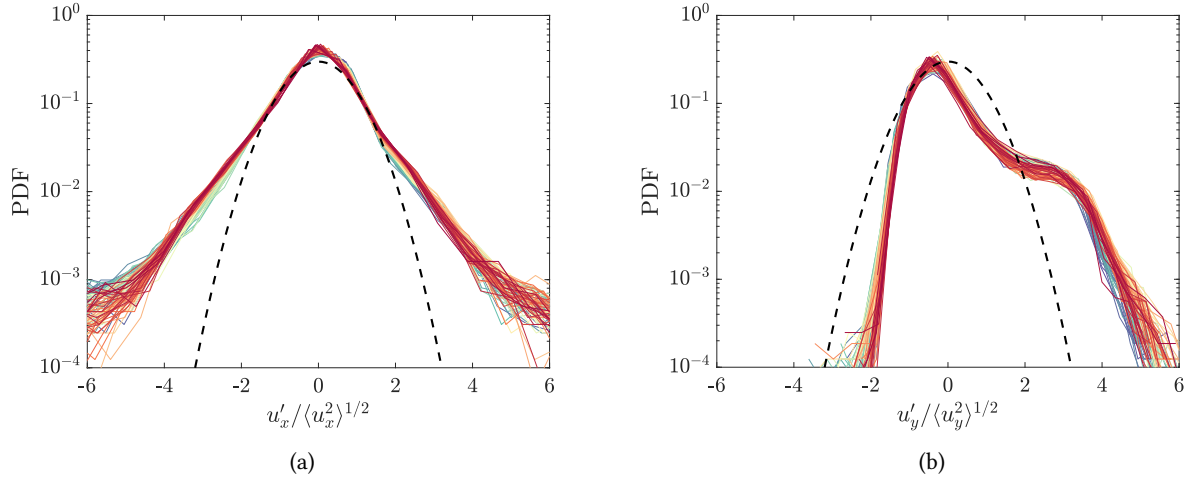


Figure 3.2: PDF of the liquid velocity fluctuations (a) in the horizontal and (b) vertical direction within the bubbly flows in viscous Newtonian fluid ($Re = 6$) normalized by the standard deviation for gas volume fraction of $\alpha = 0.010$. The dashed line corresponds to the Gaussian profile. Here, the color gradients, ranging from blue to red, correspond to 2 ms of measurement within the bubble swarm

3.3.2 Energy spectra of the velocity fluctuations

We investigate the energy spectra of velocity fluctuations for different bubble Reynolds number at a fixed gas volume fraction. Figure 4.1 shows the horizontal and vertical energy spectra of velocity fluctuations

normalized by the corresponding variance and bubble diameter for a family of bubble Reynolds number at a constant gas volume fraction of $\alpha \approx 0.025$. Here, the abscissa is normalized by the wavenumber corresponding to the bubble diameter, $k_d = 2\pi/D$. As seen in the Fig. 4.1, at large scales (small wavenumbers) the energy spectra of the liquid velocity fluctuations scales as k^{-1} in agreement with Zamansky *et al.* [71]. At the intermediate scales, for Reynolds numbers, $Re \gg O(10)$, the signature k^{-3} scaling of the energy spectra is obtained similar to [44, 59], which also serves as a validation of our experimental method. When the bubble Reynolds number $Re \sim O(100)$, the k^{-3} subrange is significantly narrower compared to that when $Re = 626$. On further decreasing the Reynolds number, for $Re = 11$ and 6, the signature k^{-3} scaling for the pseudoturbulence does not emerge. The numerical results obtained for the bubble induced turbulence by Bunner and Tryggvason [111] for Re between 12 and 30 found energy spectra decays with a k^{-3} scaling, which agrees with the present findings. To our knowledge no other data for $Re < O(10)$ exists.

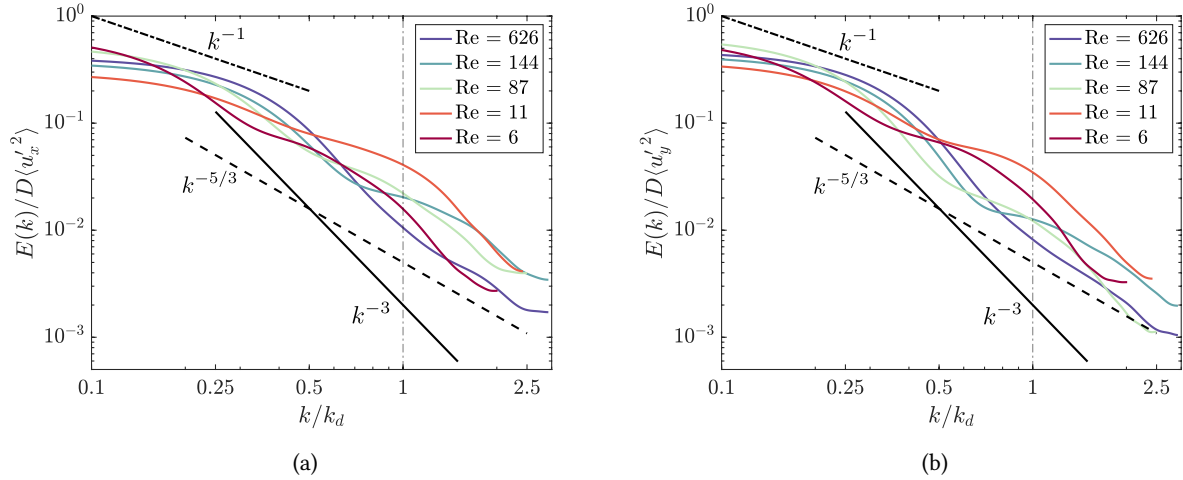


Figure 3.3: (a) Horizontal, and (b) vertical spectra of the liquid velocity fluctuations normalized by the bubble diameter and the variances for a family of Reynolds number at a constant gas volume fraction of $\alpha \approx 0.025$. The abscissa is normalized by the wavenumber corresponding to the bubble diameter, k_d . The solid, dashed and dot-dashed black lines correspond to the k^{-3} , $k^{-5/3}$, and k^{-1} scaling, respectively.

It has been argued that the observed k^{-3} scaling is the result of the contribution from the bubble wakes [43]. In the current study, however, for $O(1) < Re < O(10)$, no significant wake behind the bubbles are expected to appear [115]. Thus, instead of the signature k^{-3} scaling for the pseudoturbulence, the energy spectra of velocity fluctuations induced by the low Reynolds number bubbles in our experiments scales as $k^{-5/3}$. We argue that $k^{-5/3}$ scaling emerges from the small scale disturbances generated by the

bubble motion, which are strong enough to generate a large scale flow with the same characteristics as the Kolmogorov's turbulence [74]. Thus, we provide the first experimental evidence that at very low Reynolds numbers bubbly flows, the energy spectra of the liquid velocity fluctuations scales as $k^{-5/3}$. Further, we note that at small scales (large wavenumbers) the energy spectra of the liquid velocity fluctuations for $O(1) < \text{Re} < O(10)$ follows an exponential decay due to the dominance of viscosity beyond the Kolmogorov's microscale, η , as listed in the Table 3.1.

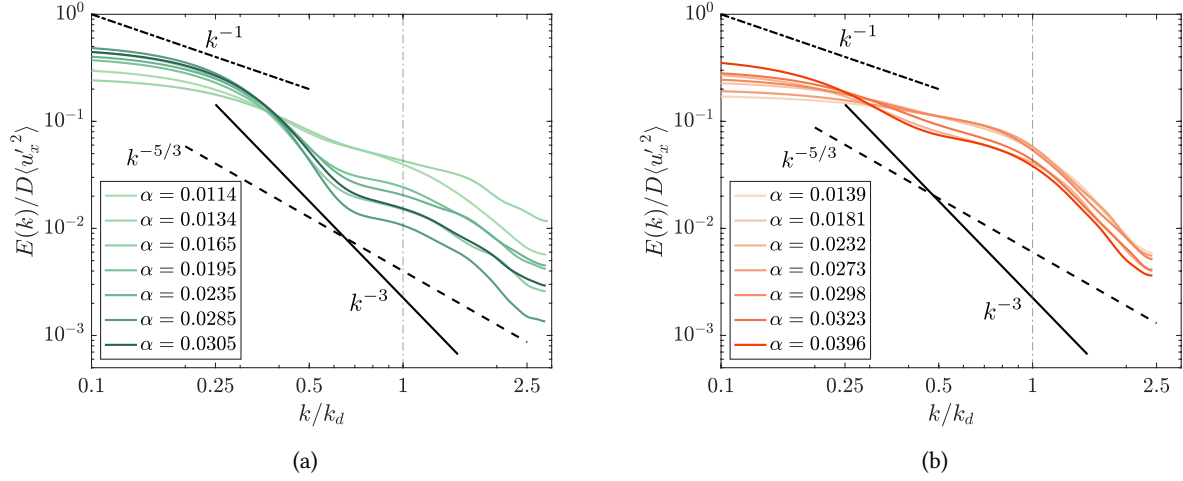


Figure 3.4: Horizontal spectra of the liquid velocity fluctuations normalized by the bubble diameter and the variances for a family of gas volume fraction at (a) $\text{Re} = 114$ and (b) $\text{Re} = 11$. The abscissa is normalized by the wavenumber corresponding to the bubble diameter, k_d . The solid, dashed and dot-dashed black lines correspond to the k^{-3} , $k^{-5/3}$, and k^{-1} scaling, respectively.

Further, Riboux *et al.* [59] suggested that the k^{-3} scaling observed in the pseudoturbulence does not depend on the bubble diameter and gas volume fraction for a wide range of Reynolds numbers. We show that this is not the case at small Reynolds number. Figure 4.2(a) shows the horizontal spectra of liquid velocity fluctuations normalized by the bubble diameter and the variance for a family of gas volume fraction at $\text{Re} = 114$. It is to be noted that at very low gas volume fractions, though the bubbles have significant wake, the energy spectra does not show the k^{-3} scaling. However, as the gas volume fraction increases, the k^{-3} scaling emerges. Note that this k^{-3} subrange is significantly narrower. This is because as the number of bubbles (i.e the gas volume fraction) increases the average inter distance between the bubbles decreases, thus leading to wake interactions. This is further corroborated using spatial correlation in the next section. Whereas, for $\text{Re} = 11$ as seen in the Fig. 4.2(b), the k^{-3} scaling is not observed, instead the $k^{-5/3}$ scaling

is obtained. This is in agreement with the results from Mazzitelli *et al.* [60] that the slope of the energy spectra no longer show the signature k^{-3} scaling for the pseudoturbulence and that the slope of the energy spectra depend on the number of bubbles and therefore is non-universal.

3.3.3 Structure functions

To demonstrate that at low Reynolds numbers, the energy spectra of the bubbly flows exhibit a $k^{-5/3}$ scaling, we consider the second-order longitudinal velocity structure function across a length scale r defined as,

$$S_2(r) = \langle [u_x(\mathbf{x} + \mathbf{r}) - u_x(\mathbf{x})] \cdot (\mathbf{r}/r)^2 \rangle. \quad (3.1)$$

Here the $\langle \rangle$ denotes the spatial average. For a homogeneous and isotropic turbulence in a Newtonian incompressible fluids, it is known that if the second-order velocity structure function scales as $S_2(r) \sim r^\beta$, then the corresponding energy spectra should scale as $E(k) \sim k^{-(\beta+1)}$ [28]. For classical Kolmogorov turbulence, the second-order velocity structure function scales as $S_2(r) \sim r^{2/3}$ in the inertial range and as $S_2(r) \sim r^2$ in the dissipative range. Figure 3.5 shows the normalized second-order velocity structure function observed in the wake of the bubble swarm as a function of normalized length scale r , across a range of Reynolds numbers for a constant gas volume fraction of $\alpha \approx 0.025$.

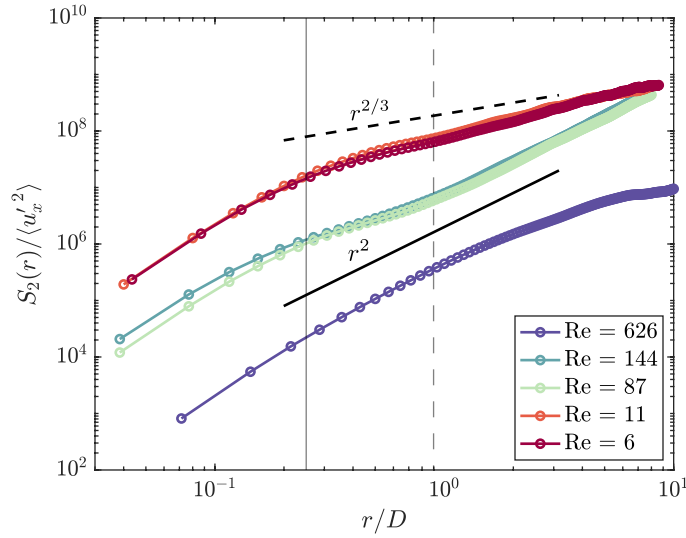


Figure 3.5: The normalized second-order structure function, $S_2(r)$ as a function of normalized length scale r for a family of Reynolds numbers at a constant gas volume fraction of $\alpha \approx 0.025$. The solid and dashed lines correspond to r^2 and $r^{2/3}$ scaling, respectively.

For all experiments, when the normalized length scale $r/D < 0.25$ (dissipative range), the structure function scales as $S_2(r) \sim r^2$. In the inertial range, because of the bubble wake-wake interactions, instead of the $S_2(r) \sim r^{2/3}$ scaling, the second-order structure function scales as $S_2(r) \sim r^2$ for $Re \gg O(100)$ [73]. At Reynolds numbers around $Re \sim O(100)$, for length scales smaller than the bubble diameter ($r/D < 1$), the structure function scales as $S_2(r) \sim r^{2/3}$. Whereas, for length scales larger than the bubble diameter ($r/D > 1$), the structure function scales as $S_2(r) \sim r^2$. This highlights that the k^{-3} scaling in the energy spectra can be recovered for bubbles with sufficiently large wakes (i.e. $Re = 144$ and $Re = 87$). On further decreasing the Reynolds number $Re \sim O(10)$, the structure function follows $S_2(r) \sim r^{2/3}$ for length scales larger than that of the dissipative range and thus the corresponding energy spectra of the velocity fluctuations at low Reynolds number bubbly flows scales as $E(k) \sim k^{-5/3}$, as shown in Fig. 4.1.

3.3.4 Spatial correlation

To further understand why the slope of the energy spectra depends on the gas volume fraction for low to moderate Reynolds number, the spatial correlation of the horizontal velocity fields are considered. The spatial correlation of the horizontal velocity, $\mathbf{u}_x$ defined as,

$$\bar{R}_{xx} = \frac{\langle u_x(\mathbf{x}) \cdot u_x(\mathbf{x} + \mathbf{r}) \rangle}{\langle u_x^2 \rangle}. \quad (3.2)$$

Here $\langle \rangle$ represent the average in space. Figure 4.3(a) shows the spatial correlation of the horizontal velocity behind the bubble swarm at a constant gas volume fraction of $\alpha \approx 0.025$. It is evident that as the Re decreases from 626 to 144 to 87, we observe a higher correlation over longer distances. This may be because the size of the bubble decreases as fluid viscosity increases (see Table 3.1). Therefore, more number of bubbles are needed to reach a fixed gas volume fraction (say $\alpha \approx 0.025$). This longer correlation with the decrease in the Reynolds number is in agreement with the numerical results from Esmaeeli and Tryggvason [110], who showed that there is an emergence of flow structures many times larger than the bubble size, which is also observed in particle suspensions at finite Reynolds numbers [121].

However, on further decreasing the Reynolds number ($Re = 11$ and 6), the velocity is decorrelated quickly as seen in the Fig. 4.3(a). This agrees with the previous literature [40, 43, 59, 60] that the presence of wake is essential for the signature k^{-3} scaling to emerge as seen in the Fig. 4.1. Figure 4.3(b) shows the

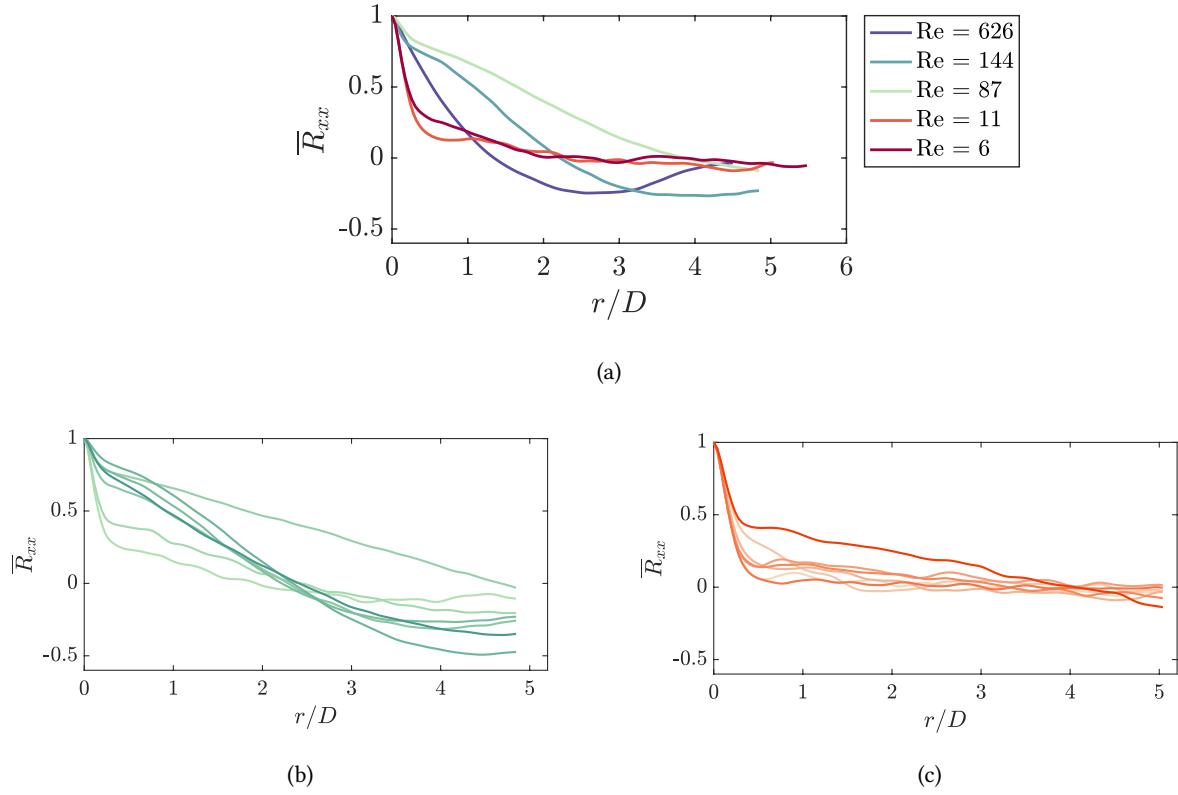


Figure 3.6: Spatial correlation of the horizontal velocity observed in the wake of the bubble swarm (a) at a constant gas volume fraction of $\alpha \approx 0.025$ for a family of Reynolds number. Spatial correlation of the horizontal velocity for a family of gas volume fractions (mentioned in Fig. 4.2) at (b) $Re = 114$ and (c) $Re = 11$.

spatial correlation of the horizontal velocity observed behind the bubble swarm for a family of gas volume fractions at $Re = 114$. It is immediately evident that as the gas volume fraction increases, the horizontal velocity is correlated over a longer distance. Thus, agreeing with our results for the energy spectra, as seen in Fig. 4.2(a), dependence on the number of bubbles. However, as seen in the Fig. 4.3(c) when the $Re = 11$, even with higher gas volume fraction, the horizontal velocity is quickly decorrelated beyond one bubble radius.

3.4 Conclusions

The bubbly flow properties in viscous Newtonian fluids have been studied experimentally by varying the concentration of glycerin in water mixture. Particle image velocimetry (PIV) technique was used to visualize the wake behind the bubble swarm to determine the velocity fluctuations in the decaying agitations. We

demonstrated experimentally that the signature k^{-3} scaling of the pseudoturbulence is replaced by $k^{-5/3}$ scaling for the energy spectra of velocity fluctuations induced by low Reynolds number bubbles. We showed that as the Reynolds number decreases to $Re \sim O(100)$, the k^{-3} subrange becomes significantly narrower. Further, for low Reynolds bubbly flows the slope of the energy spectra depends on the number of bubbles in the flow.

These experimental results agree with the numerical results by Mazzitelli *et al.* [60] that the energy spectra is non-universal for bubbles with $O(1) < Re < O(10)$. To understand why the slope of the energy spectra depends on the gas volume fraction, the spatial correlation of the velocity field was considered. For a constant gas volume fraction, as the $O(10) < Re < O(100)$, a higher correlation over longer distances was observed in agreement with the results from Esmaeeli and Tryggvason [110]. As the bubble size decreases with Reynolds number, more number of bubbles (i.e. higher gas volume fraction) are required to maintain the same gas volume fraction. Thus, as the inter-bubble distance among bubbles decreases the wake interactions are more pronounced. Whereas for $O(1) < Re < O(10)$ the velocity quickly decorrelates beyond the bubble radius, thus the signature k^{-3} scaling for the bubble induced turbulence does not emerge.

4 VELOCITY FLUCTUATIONS FOR BUBBLY FLOWS AT SMALL RE IN VISCOELASTIC FLUIDS

Abstract We experimentally investigate the effect of Reynolds number (Re) on turbulence induced by rising bubbles in a quiescent viscoelastic fluid at low Re. Recent work by **M. Ravisankar** and Zenit [5] showed that bubble-induced turbulence in viscoelastic fluids, termed as ‘elastic pseudoturbulence’, exhibits a steeper decay in the energy spectra of liquid velocity fluctuations, arising from the synergistic interaction between bubbles and fluid elasticity. In this study, we explore how this elastic pseudoturbulence evolves with Re while keeping the Weissenberg number constant. At moderate Reynolds numbers, $Re \sim O(100)$, the steep spectral decay typical of elastic pseudoturbulence begins to weaken and becomes sensitive to the gas volume fraction. As Re decreases further to $O(10)$ – $O(100)$, the spectrum transitions to a gas volume fraction dependent k^{-3} scaling, characteristic of Newtonian pseudoturbulence. This transition is further supported by measurements of the spatial correlation of velocity vectors behind the bubble swarm. These observations suggest that viscous forces can increasingly dominate over elastic effects as Re decreases, even in viscoelastic fluids.

Information This work is to be submitted to *Physical Review Fluids Letter*[7].

4.1 Introduction

When a swarm of bubbles rises in an otherwise stagnant fluid, their motion creates velocity fluctuations in the surrounding fluid. These velocity fluctuations in the wake behind the bubbles interact with one another, giving rise to the emergence of a k^{-3} scaling in the energy spectra, $E(k)$, of velocity fluctuations. This characteristic k^{-3} scaling, instead of the classical $k^{-5/3}$ Kolmogorov scaling, is regarded as bubble-induced turbulence, also referred to as pseudoturbulence [43].

The first observation of this k^{-3} scaling in Newtonian fluids was made by Lance and Bataille [40], who attributed it to a balance between energy input from rising bubbles and viscous dissipation in a statistically steady state. Since then, both numerical simulations [41, 110, 111] and experimental studies [44, 67–69, 112] have confirmed this spectral behavior in bubble swarms. Notably, Amoura *et al.* [72] demonstrated that

such scaling can arise in the continuous phase even when bubbles are replaced by randomly distributed solid spheres. Furthermore, Pandey *et al.* [113] showed that pseudoturbulence and classical turbulence can coexist across a broad range of Reynolds and Galilei numbers. More recently, Zamansky *et al.* [71] proposed that the k^{-3} subrange is driven by a mean shear rate induced by the bubbles, challenging the earlier hypothesis by Lance and Bataille [40] that spectral balance was responsible.

Recent work by **M. Ravisankar** and Zenit [5] revealed that in viscoelastic fluids, bubble-induced turbulence leads to a steeper decay in the energy spectrum of liquid velocity fluctuations. This behavior stems from the combined effects of bubble motion and fluid elasticity. Their analysis showed that polymers enhance the coherence of the flow, as evidenced by Lagrangian stretching statistics, highlighting that even a small amounts of elasticity can reshape pseudoturbulence. Notably, they found that elastic stresses, closely related to flow stretching [101], increase with polymer concentration. As a result, polymers dampen small-scale chaotic motions and promote larger, more organized flow structures, driving the steeper spectral decay.

Previous experimental work by **M. Ravisankar** and Zenit [3] examined bubble dynamics in Newtonian fluids at low Reynolds numbers and found that as the bubble Reynolds number decreases, the wakes behind the bubbles diminish. This leads to a shift in the energy spectrum toward a $k^{-5/3}$ scaling, rather than the typical k^{-3} seen in classical pseudoturbulence. Interestingly, in viscoelastic fluids, bubble interactions behave quite differently. **M. Ravisankar** *et al.* [1] showed that pairwise interactions at low Reynolds numbers are strongly influenced by fluid elasticity, suggesting that the turbulence characteristics could also change in this regime. To explore this, we adopt the particle image velocimetry (PIV) technique developed by Riboux *et al.* [59] to measure liquid velocity fluctuations in the wake of a bubble swarm. Our results show that at constant Weissenberg number as the Reynolds number decreases, the energy spectra follow a k^{-3} scaling—unlike the steeper $k^{-20/3}$ decay typically observed in viscoelastic bubbly flows at higher Re. Moreover, we find that this spectral slope is not fixed but depends on the number of bubbles in the system, pointing to a strong interplay between bubble concentration, fluid elasticity, and turbulent structure.

4.2 Experimental details

The experimental setup consists of a transparent acrylic tank of 1000 mm tall with a cross-section of 100 mm $\times$ 50 mm. Monodisperse air bubbles are introduced at the base of the tank through a removable capillary bank. The gas flow rate was controlled by a needle valve. Here, the mean gas volume fraction is measured from the increase in the liquid level after the injection of bubbles, $\alpha = (H_0/\Delta H + 1)^{-1}$, where H_0 is the initial liquid level and ΔH is the liquid level increase. Airflow is abruptly stopped using a solenoid valve, and the wake region, free of bubbles, is then captured using high-speed particle image velocimetry (Photron FASTCAM SA5 at 500 fps). The resulting image sequences are analyzed in MATLAB using PIVLab. From the 2D PIV data, velocity fluctuations in the bubble swarm wake are computed as $\mathbf{u}' = \mathbf{u} - \langle \mathbf{u} \rangle$, where $\langle \cdot \rangle$ denotes spatial averaging. The energy spectra, $E(k)$, associated with these fluctuations are extracted from the decaying turbulence behind the swarm, following [59]. Horizontal and vertical energy spectra are obtained using the Welch method, by averaging across rows and columns, respectively. Further methodological details are available in our previous works [3, 5].

Fluids (in water)	c/c^*	ρ (kg/m ³)	σ (mN/m)	η (mPa.s)	λ_e (ms)	d (mm)	U_b (mm/s)	Re	We	Wi	El
0% Glycerin	0.3	1002.6	74.41	2.12	86.8	3.5 ± 0.2	206	340	2	5	0.015
25% Glycerin	0.3	1117.9	72.06	5.21	75.4	3.1 ± 0.1	194	129	2	4.7	0.036
50% Glycerin	0.3	1131.1	71.11	7.63	92.3	2.9 ± 0.1	157	67	1	4.9	0.073
50% Glycerin	0.4	1133.2	70.95	9.22	105.1	2.8 ± 0.2	143	49	1	5.3	0.108

Table 4.1: Physical properties of the fluids: c - polymer concentration, c^* - coil-overlap concentration; ρ - density; σ - surface tension; η - viscosity; λ_e - relaxation time; d - bubble diameter; U_b - bubble velocity; Re - Reynolds number; We - Weber number; Wi - Weissenberg number; El - Elasticity number.

To prepare viscous viscoelastic fluids, we used water–glycerin mixtures combined with aqueous solutions of non-ionic polyacrylamide (PAAm, $M_w = 5 \times 10^6$ g/mol, Sigma Aldrich). Stock solutions were made by dissolving PAAm in deionized water and gently mixed using an overhead stirrer for at least 24 hours. To reduce bubble coalescence, 0.05 mol/L magnesium sulfate was added to all solutions. Fluid properties—shear viscosity, density, and surface tension—were measured using an ARES-G2 Rheometer (TA Instruments), a density meter (Anton Paar), and a bubble pressure tensiometer (KRÜSS Scientific Instruments), respectively. These are listed in Table 4.1. The coil-overlap concentration ($c^* = 1000$ ppm) was estimated from a log–log plot of zero-shear viscosity η_0 versus polymer concentration. All tests were done in the dilute regime

($c \ll c^*$) to avoid polymer–polymer interactions. The weak shear-thinning behavior observed ($n \approx 0.95$) was neglected, consistent with previous studies [51, 57]. Relevant dimensionless numbers include (i) Reynolds number, $Re = \rho U_b d / \eta$, where U_b is the bubble velocity, d is the bubble diameter, ρ is the fluid density, and η is the fluid viscosity, (ii) Weber number, $We = \rho U_b^2 d / \sigma$, where σ is the surface tension, (iii) Weissenberg number, which measures the elasticity in the polymer solutions, is defined as, $Wi = \lambda_e U_b / d$, where λ_e is the relaxation time, and (iv) Elasticity number, which compares the elastic stresses to the inertial stresses, is defined as, $El = Wi / Re$. See [5] for the details on Rheological characterization.

4.3 Results and discussion

4.3.1 Energy spectra of the velocity fluctuations

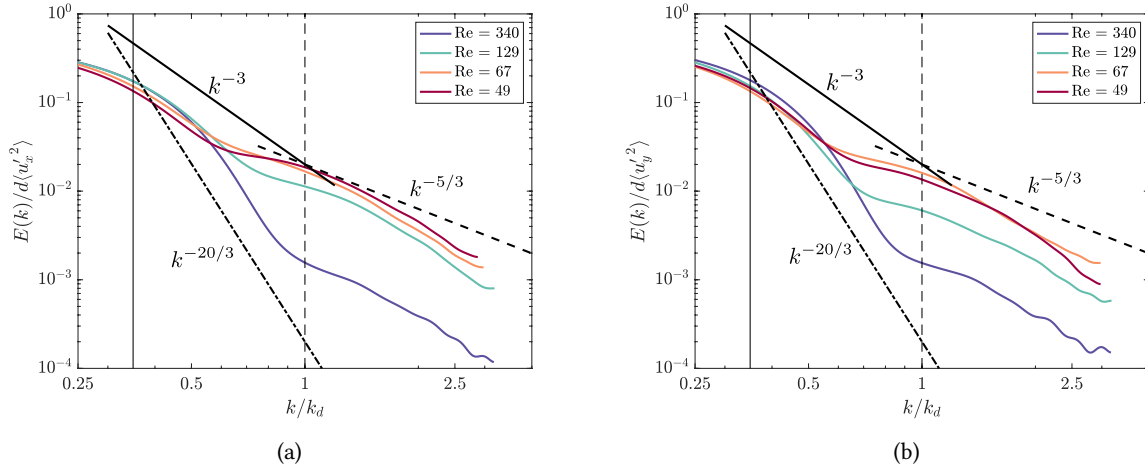


Figure 4.1: (a) Horizontal, and (b) vertical spectra of the liquid velocity fluctuations normalized by the bubble diameter and the variances for a family of Reynolds number at a constant gas volume fraction of $\alpha \approx 0.025$ and constant Weissenberg number of $Wi \approx 5$. The abscissa is normalized by the wavenumber corresponding to the bubble diameter, k_d . The solid, dashed and dot-dashed black lines correspond to the k^{-3} , $k^{-5/3}$, and $k^{-20/3}$ scaling, respectively.

Figure 4.1 shows the horizontal and vertical energy spectra of velocity fluctuations, normalized by the corresponding variance and bubble diameter, for a range of bubble Reynolds numbers at a constant gas volume fraction of $\alpha \approx 0.025$ and a fixed Weissenberg number. The abscissa is normalized by the wavenumber corresponding to the bubble diameter, $k_d = 2\pi/d$. At intermediate scales, for high Reynolds

numbers ($Re \gg O(100)$), we observe the characteristic $k^{-20/3}$ scaling of the energy spectra in the viscoelastic fluids, consistent with **M. Ravisankar** and Zenit [5], thus validating our experimental approach.

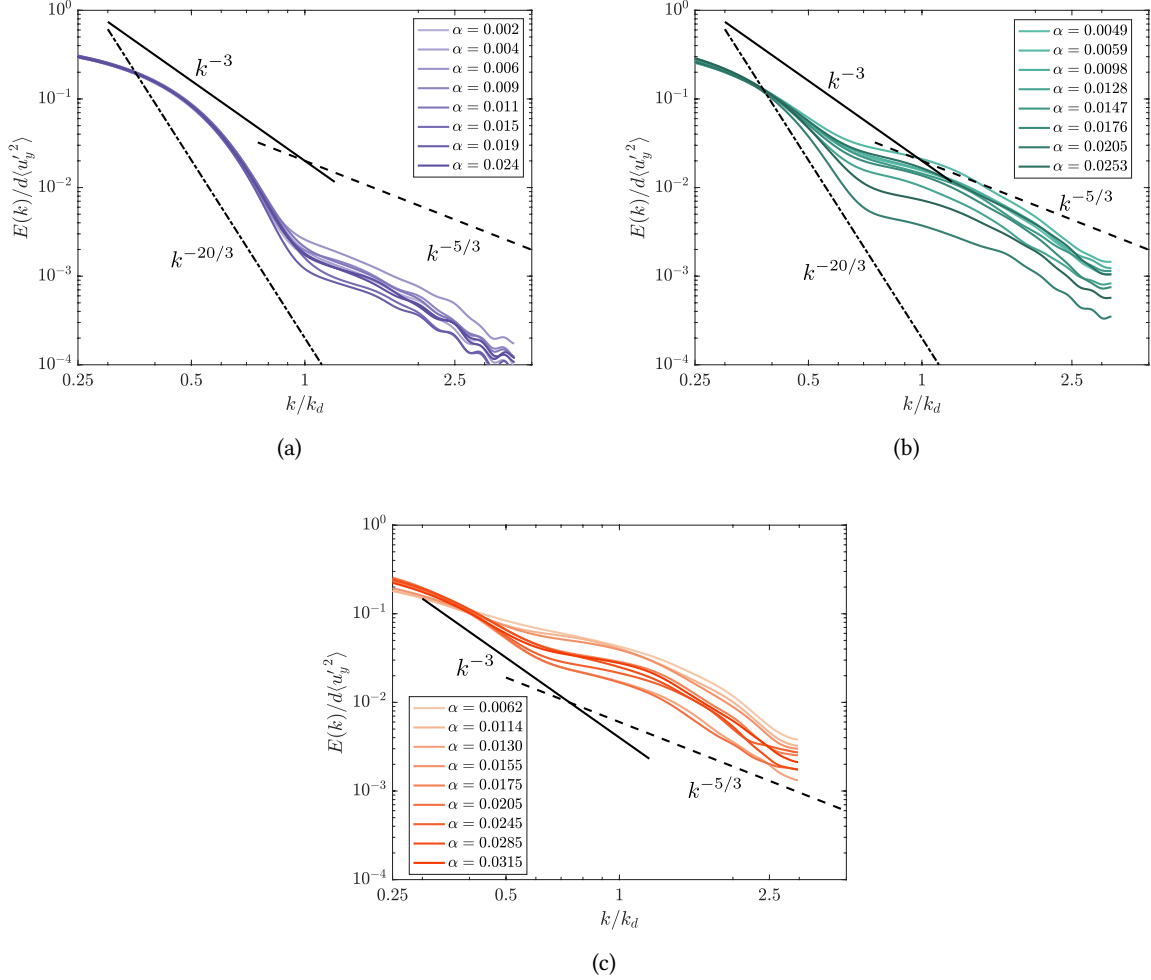


Figure 4.2: Vertical spectra of the liquid velocity fluctuations in viscoelastic fluid ($Wi = 5$) normalized by the variance and bubble diameter for a family of gas volume fraction at a Reynolds number of (a) $Re = 340$, (b) $Re = 129$ and (c) $Re = 67$. The abscissa is normalized by the wavenumber corresponding to the bubble diameter, k_d . The solid, dashed and dot-dashed black lines correspond to the k^{-3} , $k^{-5/3}$, and $k^{-20/3}$ scaling, respectively.

As the Reynolds number decreases to $Re \sim O(100)$, the $k^{-20/3}$ subrange becomes noticeably narrower compared to the case of $Re = 340$. On further decreasing the $Re = 67$ and $Re = 49$, while keeping the Weissenberg number constant, the energy spectra restores to the signature k^{-3} scaling of the Newtonian pseudoturbulence. While previous studies have linked the k^{-3} scaling in viscoelastic flows to elastic turbulence, we note that though the k^{-3} scaling is coincidental for the pseudoturbulence and elastic

turbulence, in the case of bubble-induced flow, there is no background shear flow, moreover the disturbances created by the bubbles are intermittent and anisotropic in nature. Therefore, we note that the k^{-3} scaling observed on decreasing the Reynolds number is entirely due to the bubble-induced turbulence and not the elastic effect.

Figure 4.2(a) shows the vertical spectra at $Re = 340$, the $k^{-20/3}$ scaling of the energy spectra is independent of the gas volume fraction. However, as in our previous study [3] for Newtonian fluids, at lower Re , the energy spectra begin to depend on gas volume fraction in viscoelastic fluids. As seen in Fig. 4.2(b) at $Re = 129$, the $k^{-20/3}$ scaling becomes sensitive to the gas volume fraction. This narrowing is due to reduced bubble wake length, which will be discussed in detail using spatial correlation analysis in the next section. For $Re = 67$ and 49 (Figs. 4.2(b) and (c)), we do not observe $k^{-20/3}$ scaling. Instead, the spectra follow a gas volume fraction dependent k^{-3} scaling, similar to observations in Newtonian fluids at low Re [3]. These findings confirm that the energy spectra slope departs from the canonical $k^{-20/3}$ behavior of elastic pseudoturbulence at low Reynolds numbers. Instead, the slope becomes dependent on bubble concentration, indicating that it is non-universal, similar to the Newtonian case [60, 74].

4.3.2 Spatial correlation

To better understand why the slope of the energy spectra depends on gas volume fraction at low to moderate Reynolds numbers, we examine the spatial correlation of the horizontal velocity field. The spatial correlation of the horizontal velocity component, $\mathbf{u}_x$, is defined as:

$$\bar{R}_{xx} = \frac{\langle \mathbf{u}_x(\mathbf{x}) \cdot \mathbf{u}_x(\mathbf{x} + \mathbf{r}) \rangle}{\langle u_x^2 \rangle}, \quad (4.1)$$

where $\langle \cdot \rangle$ denotes spatial averaging. Figure 4.3(a) shows the spatial correlation of the horizontal velocity behind the bubble swarm at a constant gas volume fraction of $\alpha \approx 0.025$. As Re decreases from 340 to 49, we observe a clear reduction in velocity correlation over longer distances. This is likely due to the decrease in bubble size with increasing fluid viscosity (see Table 4.1), which means more bubbles are needed to maintain the same gas volume fraction. However, at lower Reynolds numbers ($Re = 67$ and 49), the velocity field decorrelates rapidly, as evident in Fig. 4.3(a). This observation aligns with previous studies

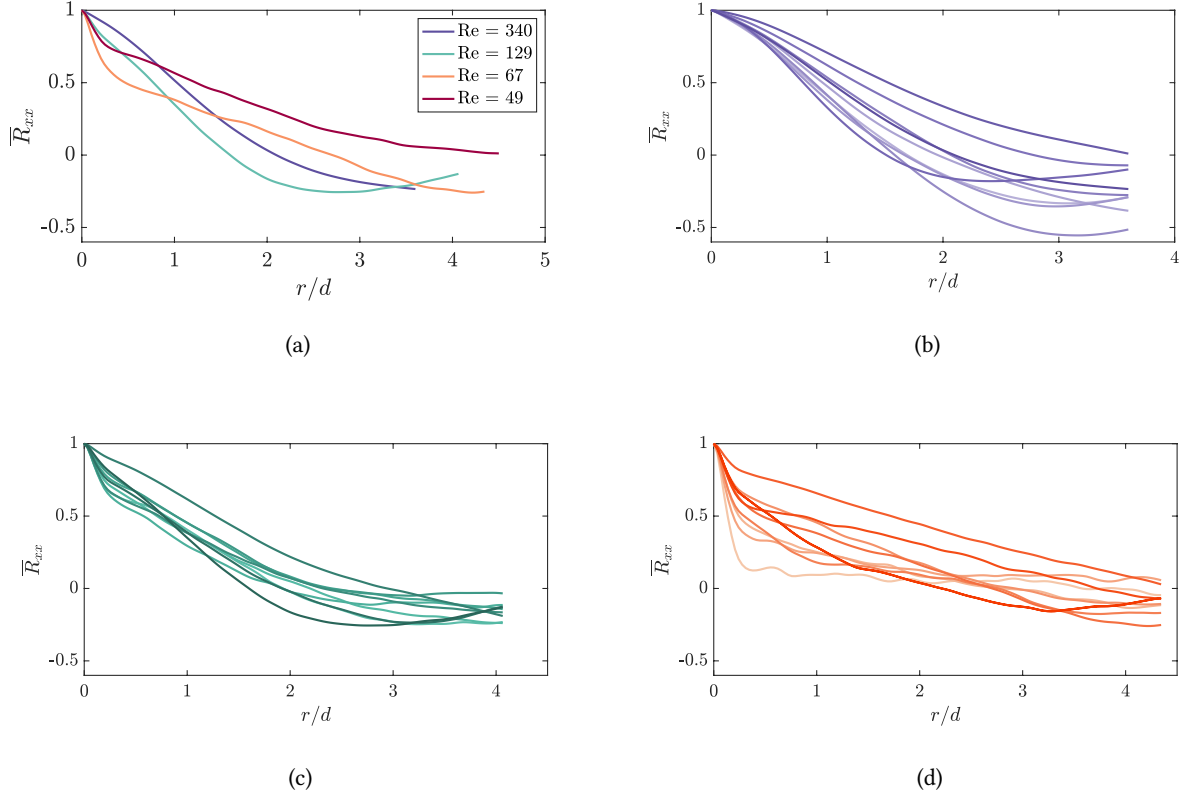


Figure 4.3: Spatial correlation of the horizontal velocity observed in the wake of the bubble swarm (a) at a constant gas volume fraction of $\alpha \approx 0.025$ and a constant Weissenberg number of $Wi \approx 5$ for a family of Reynolds number. Spatial correlation of the horizontal velocity for a family of gas volume fractions (mentioned in Fig. 4.2) at constant Weissenberg number of $Wi \approx 5$ and Reynolds number (b) $Re = 340$, (c) $Re = 129$ and (d) $Re = 67$.

[40, 43, 59, 60], which emphasize that the presence of wakes is crucial for the emergence of the characteristic pseudoturbulence scaling seen in Fig. 4.1.

Figure 4.3(c) presents the spatial correlation of horizontal velocity for varying gas volume fractions at $Re = 129$. It is immediately apparent that increasing the gas volume fraction enhances velocity correlation over longer distances. This supports our findings in the energy spectra (Fig. 4.2(b)), reinforcing the dependence on the number of bubbles. However, as shown in Fig. 4.3(c), when $Re = 49$, even at higher gas volume fractions, the horizontal velocity rapidly decorrelates beyond one bubble radius.

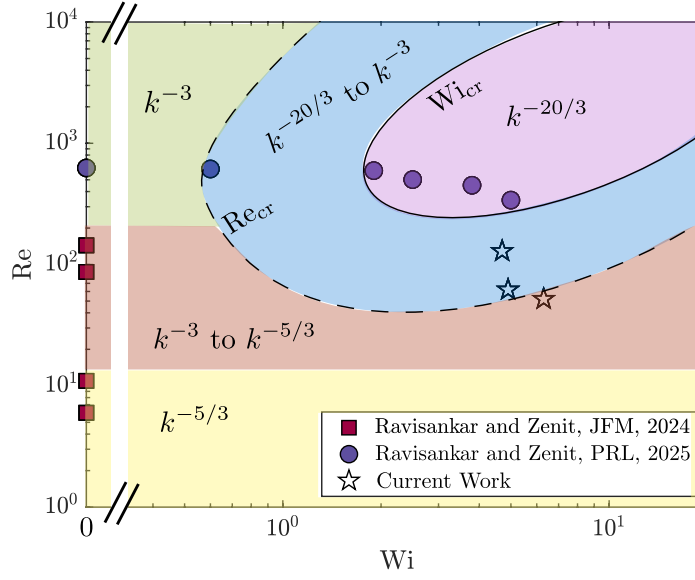


Figure 4.4: Regime map of the bubble-induced turbulence.

4.4 Conclusions

We experimentally investigated bubbly flows in viscoelastic fluids at low Reynolds numbers while keeping the Weissenberg number constant. By fixing the Weissenberg number and varying the Reynolds number, we found that at moderate Reynolds numbers ($Re \sim O(100)$), the steep spectral scaling characteristic of elastic pseudoturbulence, $k^{-20/3}$, began to weaken and became sensitive to the gas volume fraction. As Re decreased further (from $O(100)$ to $O(10)$), the energy spectrum transitioned to a k^{-3} scaling that depended on the gas volume fraction. Eventually, at $Re \sim O(10)$, the k^{-3} scaling typical of Newtonian pseudoturbulence reappeared. These observations show how viscous forces can gradually dominate and suppress elastic effects in viscoelastic fluids at low Reynolds numbers. Our findings are consistent with the recent work by **M. Ravisankar** and Zenit [3], which showed that the energy spectra in Newtonian bubbly flows become non-universal in the low Reynolds number regime ($O(1) < Re < O(10)$). To explore why the spectral slope depends on the gas volume fraction, we analyzed the spatial correlation of the velocity field. At a constant gas volume fraction, decreasing Re led to reduced velocity correlation over longer distances. In the low- Re range ($O(1) < Re < O(10)$), the velocity rapidly decorrelated beyond the bubble radius, preventing the emergence of the characteristic $k^{-20/3}$ scaling seen in elastic pseudoturbulence. Instead, a gas-volume-dependent k^{-3} scaling emerged. These results are summarized in a regime map (Fig.

4.4) that illustrates how the energy spectral scaling depends on the relative dominance of elastic, inertial, and viscous forces. When both elastic and inertial effects are strong, the spectrum follows a $k^{-20/3}$ scaling. If only inertial forces dominate, the scaling transitions to k^{-3} . When viscous forces become dominant, the energy spectrum exhibits a $k^{-5/3}$ decay.

5 EXPERIMENTAL EVIDENCE OF A DOUBLE RECIRCULATION WITHIN THE SKIRT OF A SKIRT BUBBLE

Abstract We investigate the dynamics of freely rising bubbles for a range of Eötvös and Morton numbers where a distinct skirt appears. This regime has not been studied in detail through experiments since the late 1970s. Using modern flow visualization techniques, such as high-speed photography and particle image velocimetry (PIV), we gain new insights by analyzing bubble shapes, terminal velocities, and drag forces, and compare our results with recent Direct Numerical Simulations (DNS) of skirt bubble behavior. Our findings confirm the existence of a maximum skirt length, beyond which the skirt becomes unstable. Notably, for the first time in experiments, we provide evidence of a secondary toroidal vortex inside the bubble skirt, observed in a moving frame of reference in agreement with the predictions from DNS.

Information This work is submitted to *International Journal of Multiphase Flow* [6].

5.1 Introduction

Bubbles rising through a quiescent fluid may seem like a simple everyday event, but their motion is governed by surprisingly complex physics. The behavior of gas bubbles rising through liquids has been a topic of interest in fluid dynamics for over a century, owing to its relevance in industrial processes, such as chemical reactors, wastewater treatment, and oil recovery [37, 43, 62, 122, 123], as well as for interpreting geophysical natural phenomena like underwater volcanic activity and gas seepage in oceanic environments [33, 61, 124–126].

The dynamics of a rising bubble (its speed, shape, and trajectory), are determined by the balance of buoyancy, viscous, surface tension, and inertial forces [37]. These competing effects are commonly characterized using dimensionless numbers such as the Reynolds (Re), Eötvös (Eo), Weber (We), and Morton (Mo) numbers. Over the years, studies have revealed that bubbles exhibit a variety of ascent patterns, from smooth straight to zigzagging or wobbling motions as they deform [36, 116, 127]. Moreover, surface contaminants in the surrounding fluid also influence the bubble trajectory [2]. The Clift-Grace-Weber diagram [38] provides a framework for categorizing these behaviors based on dimensionless parameters.

As bubble volume increases (Eötvös number), its shape transitions from spherical, then ellipsoidal, and eventually adopting more complex form such as dimpled ellipsoidal caps. For a recent review of gas bubble dynamics, see Legendre and Zenit [37].

Among the various complex bubble dynamics, one particularly intriguing phenomenon is the formation of a thin layer of air at the rear of large bubbles rising in a very viscous fluid, commonly referred to as a “skirt”. Once formed, skirts can remain steady, oscillate, or destabilize, sometimes shedding smaller fragments or large volumes of air known as ‘exfoliation’ [128, 129]. This skirt formation is governed by the balance between viscous stresses and surface tension and arises in a specific regime of intermediate Morton numbers ($10^{-1} < Mo < 10^4$), high Eötvös numbers ($Eo > 200$), and moderate Reynolds numbers ($10 < Re < 100$) [130, 131]. Notably, this skirt formation was first identified in drops made of fatty acid falling into a paraffin oil, as early as the 19th century by Thomson and Newall [132].

Several studies have deepened our understanding of how skirts form and evolve around rising bubbles. A simplified model to estimate skirt thickness was proposed by Guthrie and Bradshaw [129] assuming a fully developed vertical flow inside the skirt. This idea was later extended to falling drops by Bhaga and Weber [131]. Ohta and Sussman [130] showed that while skirt thickness increases with higher density and viscosity ratios between the discrete and continuous phases, it has little effect on the bubble’s terminal velocity, indicating that skirt formation is driven more by internal and surface flow dynamics than by overall bubble motion. Recent direct numerical simulations by Legendre [133] proposed a unified relation for the terminal velocity and drag force across various bubble shapes (dimpled, skirted, and spherical cap). Their work also highlighted the emergence of a near-parallel flow within the skirt film, suggesting the formation of a Poiseuille-like profile and associated vorticity patterns. More importantly, they identified a secondary toroidal vortex within the skirt, previously undetected in experiments. Despite these insights, the validation of such flow structures and skirt dynamics remains largely dependent on decades-old experimental data [131].

This gap in experimental verification motivates the present study, which aims to revisit the skirt bubble regime using modern flow visualization techniques, such as high-speed photography and particle image velocimetry (PIV). Specifically, we conduct a systematic investigation of the conditions under which skirts form, evolve, and destabilize in viscous fluids. We examine how changes in Eötvös and Morton numbers influence bubble shape and dynamics, quantify terminal velocities and drag forces. More importantly, we

use PIV measurements to resolve the internal flow field within the skirt to compare with recent numerical predictions [133].

5.2 Experimental details

The schematic of the experimental setup to form and study the skirt bubble is shown in the Fig. 5.1(a). It consists of a 20 cm × 20 cm × 2 m tank with a hemispherical “dumping cup”. Bubbles are formed by injected desired volume of air into an inverted dumping cup, the bubble is then released by rotating the cup. Corn syrup and water mixtures were used as the working fluids. Using a diffused back light setup using an LED light source, the bubble radius and the terminal velocity were measured using a high-speed camera (Photron FASTCAM SA3) with a frame rate of 500 frames/s. The equivalent bubble diameter, d , was determined as,

$$d = 2 \left(\frac{V_{cup}}{\frac{4}{3}\pi} \right)^{1/3} \quad (5.1)$$

where $V_{cup} = \frac{1}{6}\pi h(3r^2 + h^2)$ is the volume of air in the inverted dumping cup. Here, r and h are the radius of the base and height of the cup, respectively. The physical properties of the fluids are listed in Table 5.1. The shear viscosity, density and surface tension were measured using ARES-G2 Rheometer (TA Instruments), Density meter (Anton Paar) and Bubble pressure tensiometer (KRUS Scientific Instruments), respectively. The experimental results are presented in terms of the dimensionless numbers defined as follows: (i) Eötvös number, $Eo = \rho g d^2 / \sigma$, (ii) Morton number, $Mo = g \mu^4 / \rho \sigma^3$, (iii) Weber number, $We = \rho U_B^2 d / \sigma$, and (iv) Reynolds number, $Re = \rho U_B d / \mu$, where ρ is the surrounding fluid density, σ is the surface tension, μ is the fluid viscosity, g is the acceleration due to gravity, d is the equivalent spherical bubble diameter, and U_B is the bubble velocity. In addition to these, two geometric parameters are considered: a represents the frontal curvature, defined as the inverse of the radius of curvature at the leading edge of the bubble, and L denotes the skirt length, measured along the bubble surface from the rear point to the tip of the skirt, as shown in the Fig. 5.1(b). The bubble shapes observed in the current study are compared with those reported by Clift *et al.* [134] in the Fig. 5.1(c).

The flow fields around and within the skirt bubbles were obtained using particle image velocimetry (PIV). A Laser beam (532 nm) from a Nd:YAG laser system, generated into a thin sheet using a cylindrical lens, was used for illuminating the bubble column. To avoid the Mie-scattering from the bubble interface,

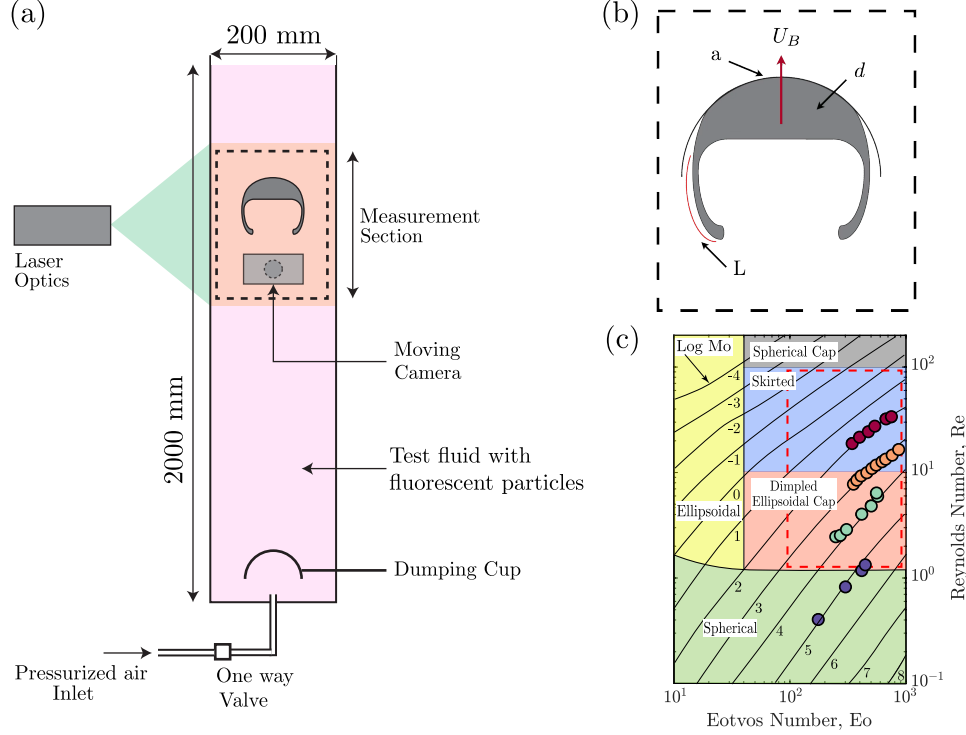


Figure 5.1: (a) Schematic of the experimental setup used in the current study showing the dumping cup installed at the bottom of the tank to form large bubbles; (b) Nomenclature used in the skirt bubble description. Here, a is the front curvature, d is the equivalent bubble diameter, L is the skirt length and U_B is the terminal velocity of the bubble; (c) The current study overlaid on the bubble shapes reported by Clift *et al.* [134].

the $55 \mu\text{m}$ diameter tracer particles were dyed with Rhodamine B. The tracer particles were suspended in an Ethanol Rhodamine B solution. To ensure that the dye infiltrated homogeneously along the particle surface, the suspension was kept at 30°C for 24 hrs. The dyed particles were then washed thoroughly in water. When illuminated by a green laser sheet, the dyed tracer particles emit orange color fluorescence. Using an orange filter in the camera, only the fluorescence light from the tracer particles was recorded to obtain the flow fields around and within the skirt bubbles. The recorded images were then analyzed using PIVLab in MATLAB.

Table 5.1: Physical properties of the fluids: ρ - density; σ - surface tension; μ - viscosity; d - equivalent bubble diameter; U_B - terminal bubble velocity; Re - Reynolds number, We - Weber number; Mo - Morton number; Eo - Eötvös number.

Fluids (in water)	ρ (kg/m ³)	σ (mN/m)	μ (Pa.s)	d (mm)	U_B (mm/s)	Re	We	Mo	Eo
80% Corn Syrup	1316	50.4	1	36 - 54	398 - 483	20 - 34	155 - 322	58	251 - 766
82% Corn Syrup	1320	50.2	2	37 - 57	322 - 436	7 - 16	101 - 289	923	356 - 1027
85% Corn Syrup	1337	50.2	3.3	31 - 46	191 - 341	2.5 - 6.5	31 - 144	6890	255 - 574
90% Corn Syrup	1350	50.1	8	26 - 41	94 - 190	0.4 - 1.3	6 - 40	238000	175 - 442

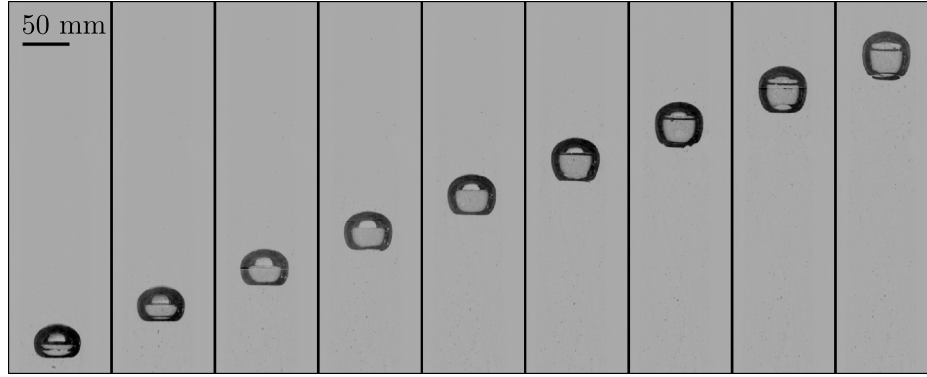


Figure 5.2: Series of snapshots showing the evolution of the skirt in an 80% corn syrup and water mixture (●) with a fixed Mo = 58 and Eo $\approx$ 550. Time interval between the snapshots is 0.16 s.

5.3 Results and discussion

5.3.1 Development of the skirt, transient process

We first examine the time evolution of the bubble velocity and the skirt length as the bubble rises. Figure 5.2 shows how the bubble shape evolves over time in a fluid with a fixed Mo = 58 and Eo = 550. The bubble maintains a nearly straight vertical trajectory. In Figure 5.3(a), the transient bubble velocity, U , normalized by its terminal value, U_B , is plotted against normalized time tU_B/d , for different Eötvös numbers. The results show that the bubble accelerates quickly and reaches its terminal velocity early in its rise. In contrast, Figure 5.3(b) shows the evolution of normalized skirt length over time. Unlike the velocity, the skirt length continues to grow even after the terminal velocity is reached, eventually stabilizing around the normalized time, $tU_B/d \approx 15$. This clear difference in timescales highlights that the bubble reaches its terminal velocity much faster than the time it takes for the skirt to fully form. These observations align with the findings of Legendre [133], reinforcing that the viscous relaxation time for a bubble to reach its terminal speed is

significantly shorter than the characteristic time required for skirt development. These plots also show that despite its relatively long length, the skirt does not add significant drag force to the bubble.

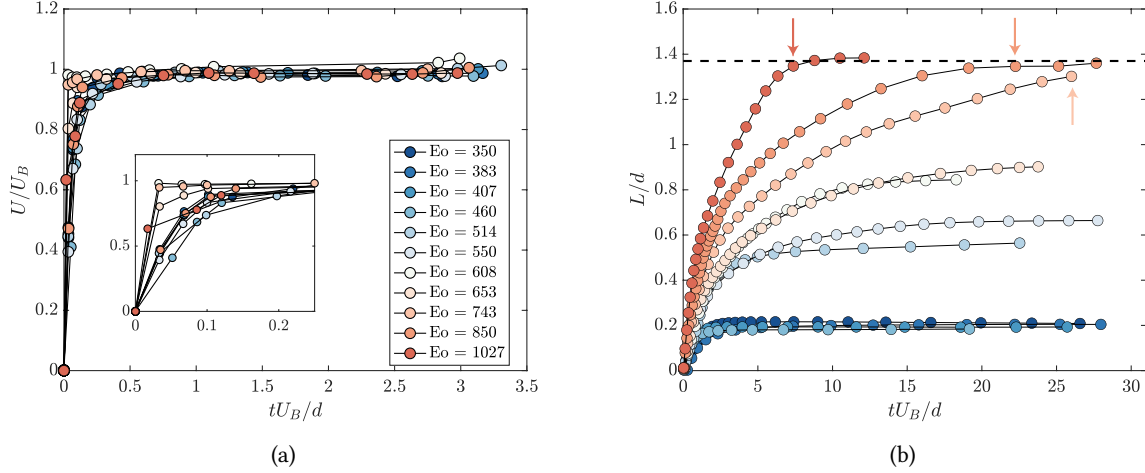


Figure 5.3: (a) Transient bubble velocity, U , normalized by the terminal velocity for a range of Eötvös number in a fluid with Morton number, $Mo = 923$. The inset highlights the early-time evolution; (b) the transient skirt length normalized by the bubble equivalent diameter as a function of normalized time, tU_B/d , for a range of Eötvös number in a fluid with Morton number, $Mo = 923$. The dashed line corresponds to the skirt length limit obtained by Legendre [133], $L \approx 1.37d$. Arrows indicate the normalized skirt length at which the skirt becomes unstable and begins to fragment.

5.3.2 Terminal properties: shape, tail length, velocity and drag coefficient

Figure 5.4 shows the terminal bubble shapes at various Eötvös numbers for a fixed Morton number of $Mo = 923$. In all cases, the Eötvös number is varied by changing the bubble volume. At $Eo \approx 250$, the rear of the bubble begins to deform and its curvature inverts, resulting in a shape known as a dimpled ellipsoidal cap (Fig. 5.4(a)). When Eo increases to around 500, the dimple opens up and the bubble forms a short skirt (Fig. 5.4(b,c)). As the Eötvös number increases further, in the range $500 \leq Eo \leq 700$, the skirt becomes slowly grows in length with time (Fig. 5.2) but eventually reaches a terminal length 5.4(c,d). For $Eo \geq 700$, the skirt becomes unstable (Fig. 5.4(f)).

For large Eötvös numbers, the skirt becomes unstable. Specifically, when the skirt length approaches $L \approx 1.37d$, it reaches a threshold beyond which the skirt becomes unstable, in agreement with Legendre [133]. Because of this instability, the skirt length no longer increases in time, indicated by the arrows in Figure 5.3(b). Figure 5.5(a) shows the normalized skirt length as a function of corrected Reynolds number,

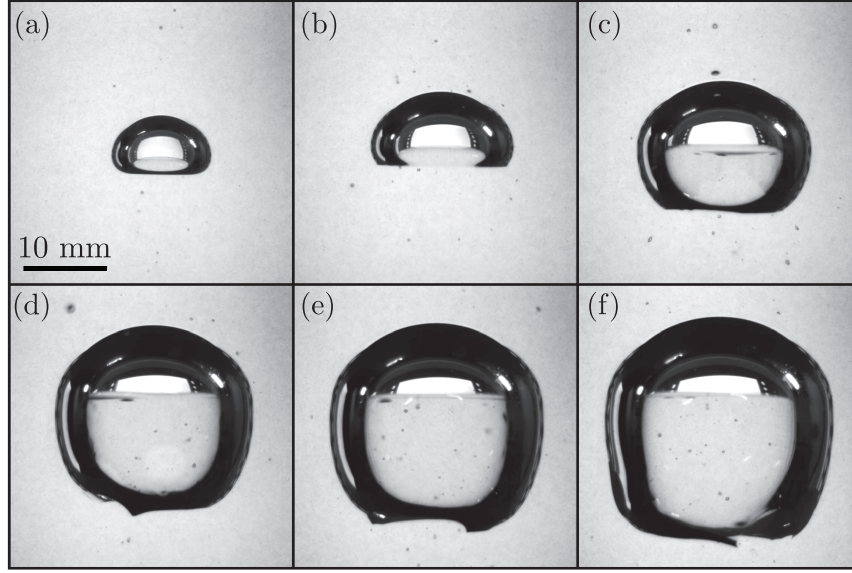


Figure 5.4: Snapshots of bubble shapes in an 82% corn syrup and water mixture (●) with a fixed Morton number of $Mo = 923$, are shown across different Eötvös numbers: (a) $Eo \approx 250$; (b) $Eo \approx 350$; (c) $Eo \approx 450$; (d) $Eo \approx 550$; (e) $Eo \approx 650$; and (f) $Eo \approx 750$.

$Re - Re_c$. Here, $Re_c \approx 150/Ca$, is the Reynolds number corresponding to the onset of the skirt, where the capillary number, $Ca = \mu U_b / \sigma$. For values of $Re - Re_c$ smaller than 10, the skirt length evolves roughly linearly, as predicted by Legendre [133], described by

$$\frac{L}{d} = 0.15(Re - Re_c). \quad (5.2)$$

For $Re - Re_c \geq 10$, a plateau is observed to a constant value, $L \approx 1.37d$. However, experimentally this plateau is not reached because the bubble becomes unstable before the skirt reaches this threshold. The inset shows the map of Re vs We , where the bubble shape transitions from spherical to dimpled shape to skirted behavior based on the We .

Figure 5.5(b) shows the evolution of the normalized front curvature as a function of the Reynolds number. The normalized curvature a/d follows a unique evolution from the spherical shape, $a = d/2$, to the spherical cap regime, where $a = 1.14d$ [135]. Legendre [133] showed that the ratio between the cap radius a to the bubble diameter d can be fitted to

$$\frac{a}{d} = \frac{12 + 1.14Re^{1.25}}{24 + Re^{1.25}}. \quad (5.3)$$

Our experimental data align remarkably well with this prediction, validating the robustness of the proposed correlation across the range of Reynolds numbers studied.

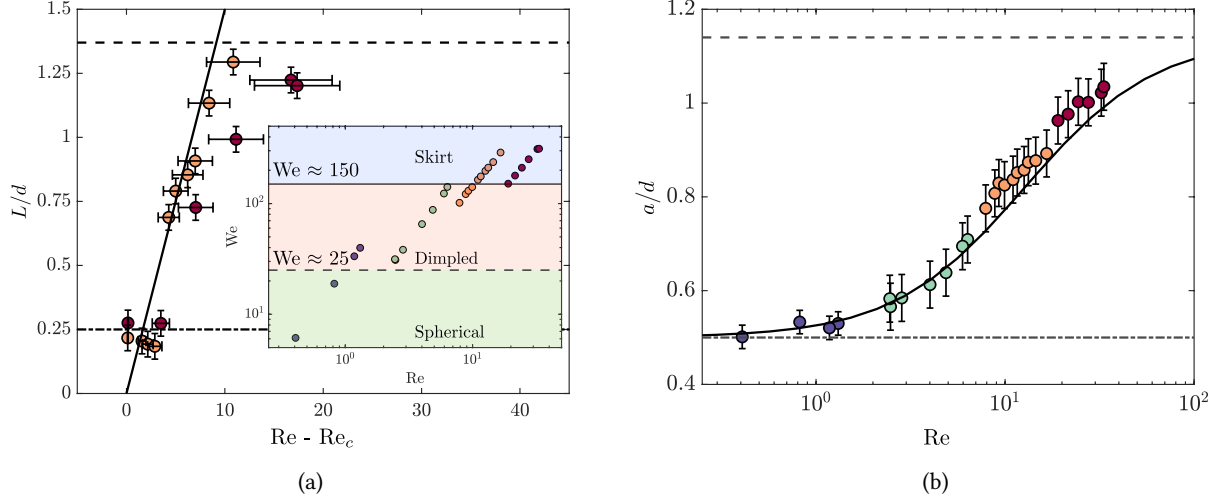


Figure 5.5: (a) Normalized skirt length as a function of modified Reynolds number. The dashed line and dot-dashed line correspond to $L = 0.25d$ and $L = 1.37d$, respectively; The inset shows the map of Re vs We in the current study. For $We \approx 25$, the bubble transitions from spherical shape to a dimpled shape and for $We \approx 150$, the dimpled bubble transitions to the skirted behavior; (b) Front curvature of the bubble normalized by the equivalent bubble diameter as a function of the Reynolds number, Re . Here, the solid line corresponds to the relation from [133], as denoted in the Equation 5.3. The dot-dashed line and the dashed line correspond to the Hadamard-Rybczynski and spherical cap relations, respectively.

We now consider the terminal velocity and the drag coefficient of the bubbles (dimpled and skirt bubbles). For the limit $Re \rightarrow 0$, the balance between drag [136, 137] and buoyancy, leads to a terminal velocity, $U_B = \rho d^2 g / 12\mu$, which can be written in terms of Reynolds number as,

$$U_B = \left(\frac{Re}{12} \right)^{1/2} \sqrt{gd}. \quad (5.4)$$

On the other hand, for a spherical-cap bubble, Davies and Taylor [135] estimated the terminal velocity by balancing the dynamic pressure from potential flow around a sphere with the hydrostatic pressure variation at the bubble nose, yielding,

$$U_B = 0.707 \sqrt{gd}. \quad (5.5)$$

Legendre [133] extended this expression for the rising velocity applicable to bubbles of any shape,

$$\frac{U_B}{\sqrt{gd}} = \left(2 + \frac{12}{\text{Re}}\right)^{-1/2}. \quad (5.6)$$

Similarly, the evolution of the drag coefficient for all the bubble shapes is satisfactorily described using,

$$C_D = \frac{8}{3} + \frac{16}{\text{Re}}. \quad (5.7)$$

Figures 5.6(a) and (b) show the terminal velocity and drag coefficient as a function of Reynolds number from the present experiments. The data closely follow the solid lines, which corresponds to the predictions of Legendre [133], confirming that the proposed relations reliably capture the dynamics across different bubble shapes.

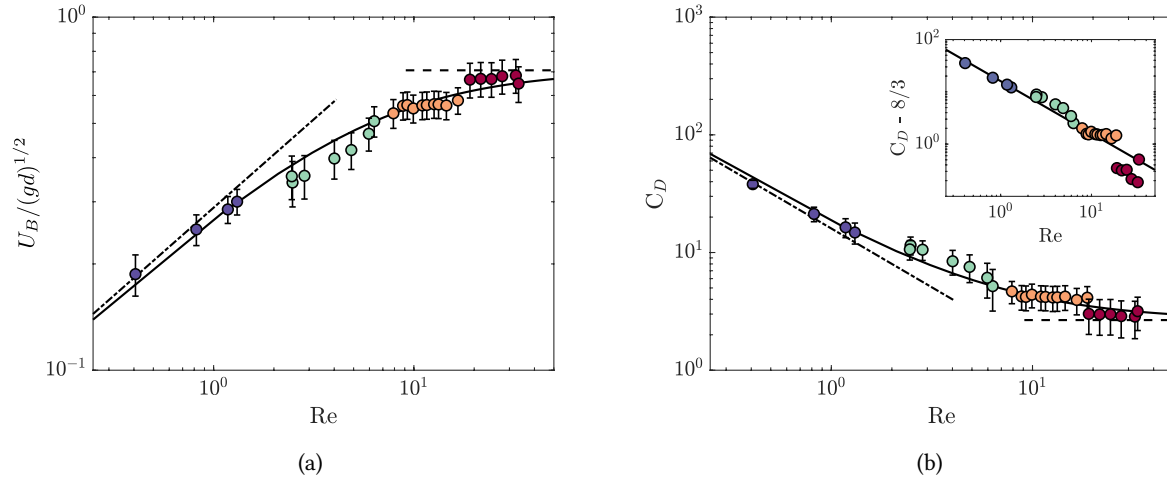


Figure 5.6: (a) Bubble terminal velocity U_B normalized by $(gd)^{1/2}$ as a function of Re ; (b) Drag coefficient C_D as a function of Re . The solid line in (a) and (b) corresponds to the relation from [133]. The dot-dashed line and the dashed line in (a) and (b) correspond to the Hadamard-Rybczynski and spherical cap relations, respectively.

5.4 Flow fields around the skirt bubble

Early experiments on skirt bubbles [38, 131] identified only a single vortex in the wake, whereas more recent numerical simulations [128, 130, 133, 138] have revealed the presence of a secondary vortex inside the skirt. We now investigate the flow fields within freely rising skirt bubbles using particle image velocimetry (PIV).

Figure 5.7 shows the average flow fields around the skirt bubbles in the moving frame of reference with a fixed Morton number of $Mo = 58$ across different Eötvös numbers (a) $Eo \approx 550$, (b) $Eo \approx 653$ and, (c) $Eo \approx 743$, respectively. Analyzing the flow in the moving frame, where the bubble is stationary and the fluid moves past it, provides a clearer view of the flow structures around and inside the bubble, making it easier to compare with numerical simulations.

In Figure 5.7(a) ($Eo \approx 550$), no secondary vortex is observed, as the skirt is too short to sustain it. When the Eötvös number increases to $Eo \approx 653$ (Fig. 5.7(b)), the skirt elongates and the remnants of a secondary vortex begin to appear. However, flow near the bubble surface is partially obscured by the cylindrical shape of the bubble, which prevents direct visualization of the vortex core. At a higher $Eo \approx 743$ (Fig. 5.7(c)), the stagnation point (denoted by the green dot) between the primary and secondary vortices shifts downward, indicating that the secondary vortex is becoming stronger and larger. This elongation also makes the skirt dynamically unstable, as evidenced by the stagnation point moving off-center in the averaged flow field, showing how skirt instability disrupts flow symmetry. Thus, we provide the first experimental confirmation of a secondary vortex inside a skirt bubble, in agreement with the previous numerical simulations [133, 138].

To quantify vortex strength, we compute the circulation, defined as

$$\Gamma = \iint_A \omega dA, \quad (5.8)$$

where $\omega = \partial v / \partial x - \partial u / \partial y$ is the out-of-plane vorticity and A is the area enclosing the vortex. The normalized circulation ratio of the secondary to the primary vortex increases from about 10^{-2} at $Eo \approx 653$ to roughly 10^{-1} at $Eo \approx 743$, confirming the growing influence of the secondary vortex as the skirt lengthens. Although optical scattering near the bubble surface limits direct visualization of the secondary vortex core, its presence is inferred from the stagnation point position and the evolving flow patterns. This is quantified by the distance from the bubble surface to the stagnation point, denoted as y_s . Figure 5.7(d) presents the normalized stagnation distance as a function of the modified Reynolds number. Because the formation of the secondary vortex is directly linked to the skirt length, the normalized stagnation distance exhibits a similar trend to that of the skirt length with respect to the modified Reynolds number.

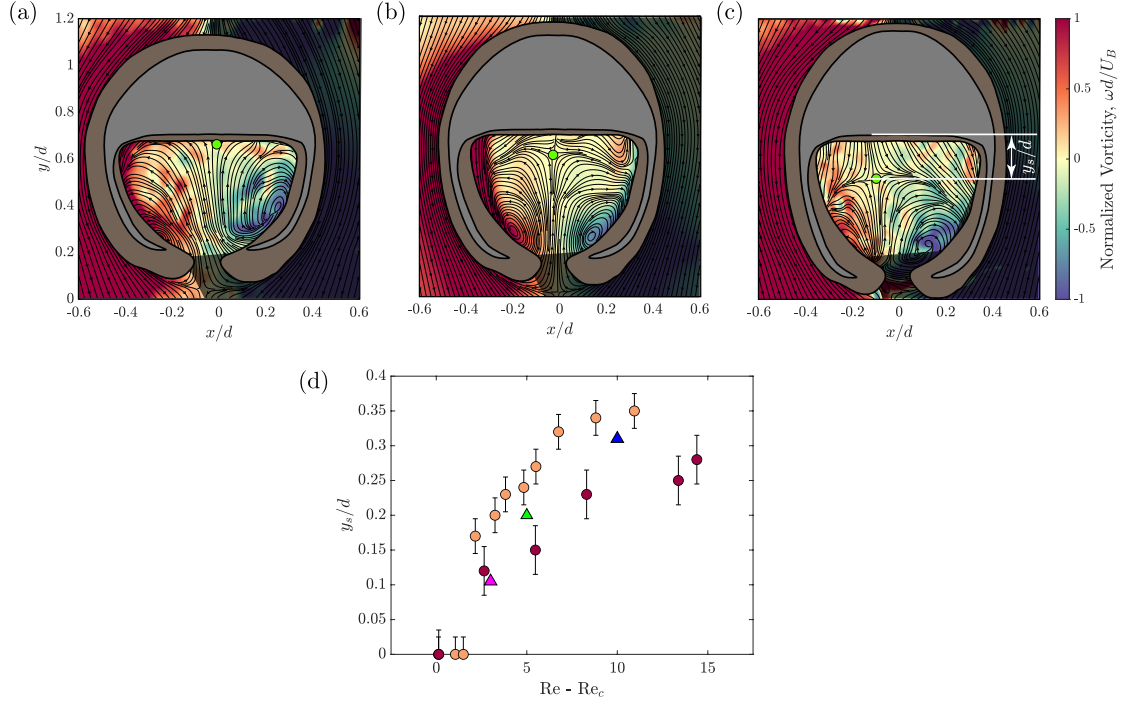


Figure 5.7: Snapshots of average vorticity field, normalized by the flow time, d/U_B , around the skirt bubbles in the moving frame of reference with a fixed Morton number of $Mo = 58$ across different Eötvös numbers: (a) $Eo \approx 550$, (b) $Eo \approx 653$ and, (c) $Eo \approx 743$, respectively. Solid lines indicate averaged streamlines in the wake. Green dot corresponds to the stagnation point in the wake within the skirt bubble. (d) Normalized stagnation distance as a function of modified Reynolds number. Numerical results from Legendre [133]: $\blacktriangle$ - $Mo = 500$; $Eo = 330$; $\blacktriangleup$ - $Mo = 10$; $Eo = 200$; $\blacktriangledown$ - $Mo = 100$; $Eo = 330$.

5.5 Conclusions

In this study, we experimentally investigated the dynamics of skirt bubbles rising freely in quiescent liquid, across a range of Eötvös and Morton numbers. We confirmed the existence of a maximum skirt length beyond which the skirt becomes unstable. Most notably, we provided the first experimental evidence of a secondary toroidal vortex forming within the bubble skirt in a moving frame of reference. Although the core of the secondary vortex could not be directly visualized due to optical scattering near the bubble surface, its presence was indirectly identified through the location of the stagnation point formed between the primary and secondary vortices. Our results show that the position and strength of this secondary vortex evolve with increasing bubble size. These findings not only support recent DNS results but also contribute new experimental insights into the complex internal flow structures of skirt bubbles. Our study serves as the base for future experiments aimed at precisely measuring the skirt thickness and understanding its impact on flow stability.

6 CONCLUSIONS

Pseudoturbulence refers to the turbulent-like motion observed in a quiescent fluid when a swarm of bubbles rises through it, driven by interactions between the wakes of individual bubbles. In this thesis, we investigate the dynamics of bubbly flows across various flow environments using particle image velocimetry (PIV) experiments. The key findings are summarized in Figure 6.1, which highlights the different regimes of bubble-induced turbulence based on the properties of the continuous phase and the bubble Reynolds number.

6.1 Summary

In Chapter 2, we explored what happens when polymer additives are introduced into bubbly flows, and what we found was both surprising and exciting. We discovered that polymers fundamentally reshape the turbulence generated by rising bubbles. As polymer concentration increased, the energy in the flow decayed much faster than expected, steepening well beyond the classical k^{-3} scaling of pseudoturbulence to a striking $k^{-20/3}$. This level of spectral decay had never been observed before in viscoelastic turbulence. At a constant gas volume fraction of $\alpha \approx 0.025$, the correlation between horizontal velocity and vorticity magnitude increased over longer distances as the polymer concentration increased. The average vortex size, estimated using the integral length scale of vorticity, $L_\omega = \int_0^\infty \bar{R}_\omega dr$, also increased with the Weissenberg number. These findings indicated that polymer stresses enhanced the prominence of large-scale flow structures, consistent with previous experimental [83] and numerical [53] studies. To understand why this happens, we examined how fluid elements stretch and deform over time by calculating Lagrangian coherent structures using FTLE analysis. This is important, as the stretching field is related to the polymeric stresses within the flow. These measurements revealed that polymers increased the stretching in the flow and smoothed out small-scale chaotic motions, creating more organized, large-scale structures. In simple terms, since the Lagrangian stretching field is intrinsically related to the polymer stress [101], the dissipative nature of the polymer resulted in the steeper decay in the energy spectra observed. Thus, the $-20/3$ scaling, previously unreported in viscoelastic turbulence, likely emerges from the coupled dynamics of bubbles and polymers.

In Chapter 3, we experimentally studied bubbly flows in viscous Newtonian fluids by varying the glycerin concentration in water. Our results showed that the classical k^{-3} scaling of pseudoturbulence was replaced by a $k^{-5/3}$ scaling as the Reynolds number decreased, particularly around $\text{Re} \sim O(100)$. As Re decreased, the k^{-3} subrange narrowed significantly, and the energy spectra slope depended on the number of bubbles in the flow. These findings agreed with numerical results [60] showing non-universal energy spectra for Reynolds numbers between $O(1)$ and $O(10)$. To explain this, we examined spatial velocity correlations. At moderate Reynolds numbers ($O(10) < \text{Re} < O(100)$), longer-range correlations were observed, consistent with [110]. However, when Re dropped further ($\text{Re} \sim O(10)$), bubbles did not produce significant wakes behind them [115, 116], causing velocity to decorrelate quickly beyond the bubble radius and preventing the emergence of the k^{-3} scaling despite higher bubble densities. This strongly confirms the essential role of wakes in the development of the k^{-3} scaling.

In Chapter 4, we experimentally studied bubbly flows in viscoelastic fluids at low Reynolds numbers while keeping the Weissenberg number constant. By maintaining a fixed Weissenberg number and varying the Reynolds number, we observed that at moderate $\text{Re} \sim O(100)$, the steep spectral scaling typical of elastic pseudoturbulence, $k^{-20/3}$, began to weaken and became sensitive to the gas volume fraction. As Re decreased further (from $O(10)$ to $O(100)$), the spectrum shifted to a gas volume fraction-dependent k^{-3} scaling, and eventually, as Re dropped to $O(1)$ – $O(10)$, the $k^{-5/3}$ scaling characteristic of Newtonian fluids was recovered. These results revealed how viscous forces, even in viscoelastic fluids, gradually overrode elastic effects at small Reynolds number.

In Chapter 5, we investigated the dynamics of skirt bubbles rising freely in quiescent liquid, across a range of Eötvös and Morton numbers. We confirmed the existence of a maximum skirt length beyond which the skirt becomes unstable. Most notably, we provided the first experimental evidence of a secondary toroidal vortex forming within the bubble skirt in a moving frame of reference. Although the core of the secondary vortex could not be directly visualized due to optical scattering near the bubble surface, its presence was indirectly identified through the location of the stagnation point formed between the primary and secondary vortices. Our results show that the position and strength of this secondary vortex evolve with increasing bubble size. These findings not only support recent DNS results but also contribute new experimental insights into the complex internal flow structures of skirt bubbles.

6.2 Future directions

In Chapter 2, we explored how pseudoturbulence behaves in a still, quiescent viscoelastic fluid. A natural next step would be to ask: what changes when the background is already turbulent? For example, in turbulent pipe flows, adding polymers is known to reduce drag [49]. Similarly, bubbles can also affect turbulence—but how they do so depends on their size [39]. So an exciting question arises: what happens when you have both, a turbulent viscoelastic fluid and bubbles rising through it? And even further, how do these combined effects influence heat transfer? A simple setup like a Hele-Shaw cell could be a good starting point to explore these interactions.

In Chapters 3 and 4, we looked at pseudoturbulence at low Reynolds numbers in both Newtonian and viscoelastic fluids. Our focus was on the wake left behind by a bubble swarm. But what actually happens within the swarm? That remains a fascinating open question. One way to investigate this, inspired by Amoura *et al.* [72], could be to randomly distribute solid spheres in a tank and investigate the flow. This could help us better understand how consistent the flow behavior really is across different conditions at low Reynolds numbers.

There is also an ongoing debate in the bubbly flow community around the observed $k^{-5/3}$ scaling at scales smaller than the bubble diameter [139]. Part of the confusion comes from how we collect the data. Some experiments, like ours, look at the flow behind the bubble swarm, capturing the decaying wake [3, 5, 59]. Others try to measure the flow within the swarm but often need to interpolate data where bubbles obscure the velocity field [73, 140]. Both methods have their limitations. Interestingly, experiments with a turbulent background—especially those using hot-wire anemometry—tend not to see the $k^{-5/3}$ scaling at all [68, 72]. Clearly, there is a need for new experiments that can measure the full 3D velocity field around and within the swarm, without relying on interpolation.

Finally, after several decades, in Chapter 5, we experimentally revisited the skirt bubbles using advanced imaging techniques. As noted by Legendre and Zenit [37], there’s now a real opportunity to build a comprehensive “bubble map” for viscoelastic fluids, taking into account the Eötvös number, Morton number, Reynolds number, and Weissenberg number. It would be fascinating to see how skirt bubbles behave in such complex fluids, and what new physics might emerge.

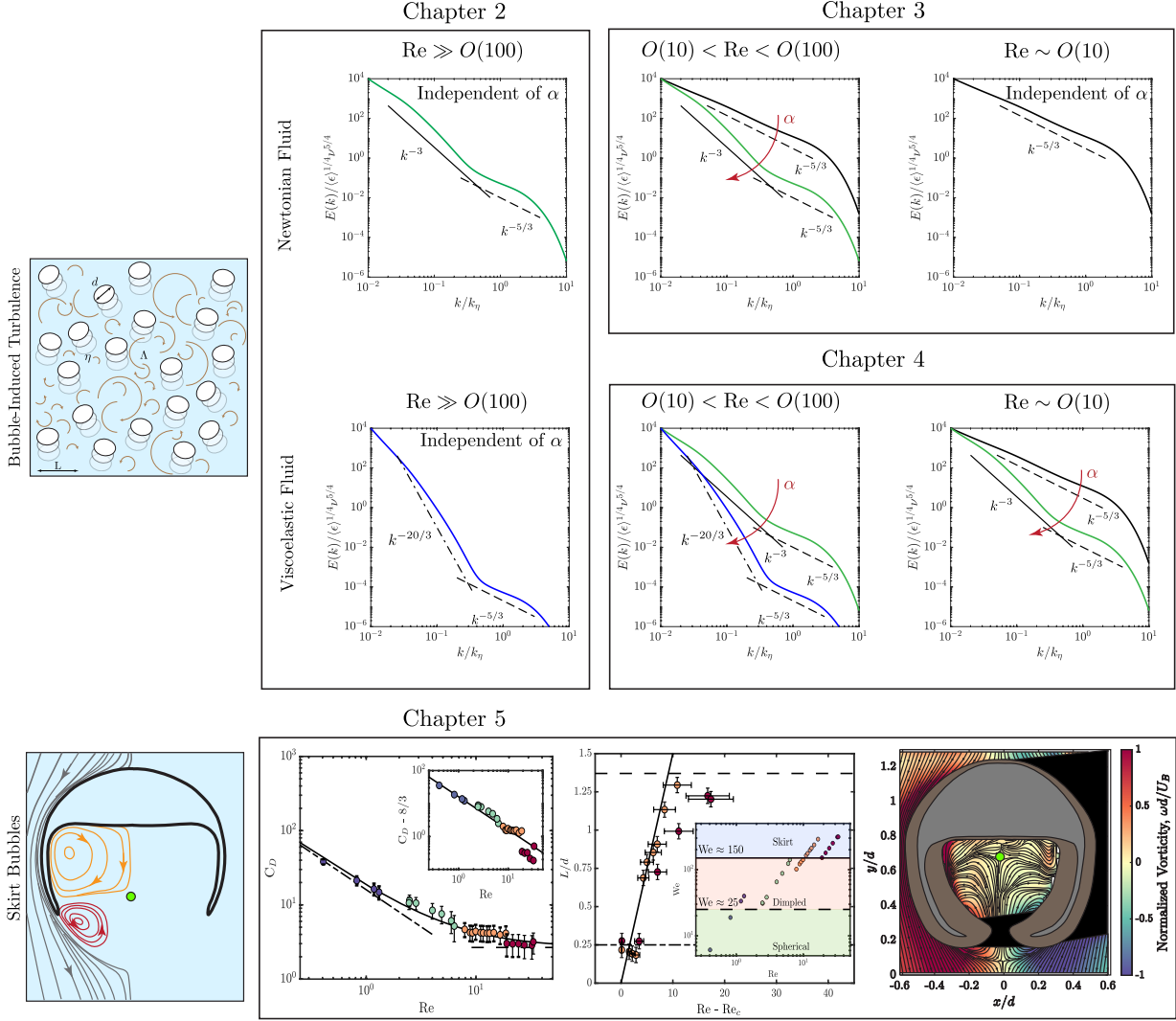


Figure 6.1: This thesis primarily explored bubble-induced turbulence, as detailed in Chapters 2–4. In Chapter 2, we showed for the first time how polymer additives altered bubble-induced turbulence. Even without background shear, once the polymer concentration exceeded a critical threshold, the energy spectrum of velocity fluctuations steepened from the classical k^{-3} to a much steeper $k^{-20/3}$ decay. This new scaling appeared between the bubble wake length and the bubble diameter and remained independent of the gas volume fraction in the inertial range. In Chapter 3, we focused on how bubble inertia affected pseudoturbulence in Newtonian fluids. We found that at lower Reynolds numbers ($O(10) < Re < O(100)$), the slope of the energy spectra depended on the gas volume fraction, α , between the length scales relevant to the bubble wake length and bubble diameter. These results aligned with numerical findings by Mazzitelli *et al.* [60], showing that energy spectra were non-universal. On further decreasing the Reynolds number ($Re \sim O(10)$), the k^{-3} scaling was not observed; instead, the $k^{-5/3}$ scaling was obtained. In Chapter 4, we extended this analysis to polymeric fluids. As with Newtonian fluids, reducing the Reynolds number caused a shift from k^{-3} to $k^{-20/3}$ scaling, depending on the gas volume fraction. When $Re \sim O(10)$, the energy spectrum transitioned from $k^{-5/3}$ to k^{-3} , again depending on the gas volume fraction. Finally, in Chapter 5, we revisited skirt bubble dynamics. Through experiments across a range of Eötvös and Morton numbers, we confirmed the existence of a maximum skirt length beyond which instability set in. Most notably, we presented the first experimental evidence of a secondary toroidal vortex forming within the bubble skirt in a moving reference frame.

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A PRESENCE OF SURFACTANTS CONTROLS THE STABILITY OF BUBBLE CHAINS IN CARBONATED DRINKS

Abstract Bubbles appear when a carbonated drink is poured in a glass. Very stable bubble chains are clearly observed in champagne, showing an almost straight line from microscopic nucleation sites from which they are continuously formed. In some other drinks such as soda, such chains are not straight (not stable). Considering pair interactions for spherical clean bubbles, bubble chains should not be stable which contradicts these observations. The aim of this work is to explain the conditions for bubble chain stability. For this purpose, experiments and direct numerical simulation are conducted. The bubble size as well as the level of interface contamination are varied, to match the range of parameters in typical drinks. Both factors are shown to affect the bubble chain stability. The transition from stable to unstable behavior results from the reversal of the lift force, which is induced by the bubble wake. A criteria based on the production of vorticity at the bubble surface is proposed to identify the conditions of transition from stable to unstable bubble chains. Beyond carbonated drinks, understanding bubble clustering has impact in many two-phase problems of current importance.

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A.1 Introduction

Flavour and smell are not the only sensorial traits of drinking champagne. The bubble chains that appear in a tall glass complete the trifecta: the beauty and visual satisfaction of observing the consecutive and ordered formation of bubbles are an essential part of the experience. Bubbles in carbonated drinks, not just champagne, form as excess Carbon Dioxide exsolves from the supersaturated liquid. The supersaturation condition is easily achieved since the bottling pressure is higher to atmospheric, hence more gas can be dissolved in the liquid. The chains form as bubbles are continuously formed at a certain nucleation site; once the bubble reaches a certain critical size for which buoyancy overcomes the surface tension force

that keeps the bubble attached, the bubble begins to ascend freely. Immediately after the bubble has been dislodged, a new one being to form. This process continues for several minutes until most of the dissolved gas comes out of solution [141]. The speed at which bubble rise in the liquid depends on their size and the liquid properties [142–144]. Once the bubbles reach the liquid surface they briefly remain at it while the liquid film drains [145–149]. When the liquid film becomes sufficiently thin, it spontaneously bursts with a characteristic sound [150] and forming aerosol droplets [149, 151, 152]. All these processes make a glass of carbonated liquid a complex laboratory of two-phase flows with intricate physico-chemical processes [141, 151–154].

The present investigation focuses on the stability of bubble chains: as the bubbles form consecutively from the same nucleation site, should they remain in-line or not? Stable in-line motion is commonly observed in Champagne and other carbonated wines, but is this what one should expect by considering the hydrodynamic interactions among bubbles? The flow disturbance created by the motion of a single bubble ascending will exert certain influence on a subsequent bubble moving behind. Beyond the curiosity of bubble chains in drinks, these results are important in clarifying the key mechanisms that lead to bubble distributions in bubbly flows in general. The nature of hydrodynamic interactions among bubbles determines whether or not cluster are formed [155] which, in turn, greatly influence the mixing and agitation levels of bubbly flows[43].

Let us first consider the case of a two spherical bubbles, with clean interfaces, rising in-line. If the Reynolds number (defined in the Methods section), is higher than 10, the in-line motion is unstable because bubbles are ejected from the wake of any previous bubble. The process is explained by the wake of the leading bubble. Its interaction with the trailing bubble results in the appearance of a lift force [156, 157], F_L :

$$F_L = C_L \rho_l \frac{4\pi R^3}{3} (\mathbf{V}_L - \mathbf{V}_B) \times (\nabla \times \mathbf{V}_L) \quad (\text{A.1})$$

where $\mathbf{V}_L$ is the velocity field of the leading bubble wake, $\mathbf{V}_B$ is the velocity field of the trailing bubble and C_L is a lift coefficient (positive) for spherical and clean bubbles. Such unstable in-line bubble interaction has been confirmed by experiments [158] and simulations [157]. Recent simulations have also revealed that the interaction between two bubbles, in the inertial regime, is full of subtleties [159, 160].

For the case of bubbles in champagne, and other aqueous carbonated drinks, the bubble Reynolds number increases monotonically after the bubble formation as the bubble continues to grow. However, and most importantly, it reaches a value of $Re > 10$ a few millimeters after formation [161]. Hence, one would expect that stable bubble chains would not be observed in a glass champagne. In this investigation we argue that a stable bubble chain is possible only if the lift force acting on the trailing bubble changes sign. In such a case, the bubbles would remain in-line regardless of small disturbances.

Previous investigations have addressed the lift reversal process. Two mechanisms are known to cause lift reversal, bubble deformation [143, 162, 163] and surface contamination [164]. Either one, or both, can lead to a reversal of the lift force. Harper [165] envisioned that surfactants were in fact responsible for the stabilization of bubble chains observed on common beverages, but did not propose a physical mechanism to explain the stabilization.

The main objective of this work is to clarify the origin of the in-line stabilisation of bubble chain, inspired by the particular case of bubble chains in carbonated drinks. Experiments and numerical simulations are conducted to vary the bubble size and surface contamination independently. To simplify the system, we consider non-growing bubbles. In the case of Carbon Dioxide bubbles rising in supersaturated liquids, the bubbles grow as they ascend. Since the grow rate, $\dot{R}$, is smaller than the bubble velocity $\dot{R}/U_B < 1$, the added mass effect can be neglected [161].

A.2 Materials and Methods

A.2.1 Relevant dimensionless groups

The problem of a bubble of radius R rising steadily in a fluid of viscosity μ and density ρ under the action of gravity g , with a gas-liquid interfacial tension σ_0 , is completely characterized by two non dimensional numbers. We consider here the Bond number and Archimedes numbers:

$$Bo = \frac{\rho g R^2}{\sigma_0}, \quad Ar = \frac{\rho^2 g R^3}{\mu^2}. \quad (\text{A.2})$$

Note that the Archimedes number is in fact a Reynolds number

$$Re = \frac{\rho R U_B}{\mu} \quad (\text{A.3})$$

where characteristic velocity $U_B = \rho g R^2 / \mu$ is obtained from the balance of the bubble viscous drag with buoyancy. The bubble deformation can be quantified considering the bubble aspect ratio defined as $\chi = D_L / D_S$ where D_L and D_S are the long and short diameters of an ellipsoidal bubble. It is well known that χ is a function of Bo [166].

The transfer of surfactants is characterized by the bulk and the interface Péclet numbers, respectively defined as:

$$Pe = \frac{\rho g R^3}{\mu D}, \quad Pe_s = \frac{\rho g R^3}{\mu D_s}, \quad (\text{A.4})$$

where D and D_s are the surfactant diffusivity coefficient in the bulk and the bubble surface, respectively. An estimation of Pe_s for an air bubble of millimetric radius inside a liquid containing fatty acids or other long chain surfactants ($D_s \sim O(10^{-10})$) gives $Pe_s \approx 10^8$, indicating negligible surface diffusion. Therefore, D_s has been set to zero in the simulations.

Furthermore, the surfactant presence at the interface introduces three additional non dimensional numbers

$$\alpha = \frac{k_a C_0 \mu}{\rho g R}, \quad La = \frac{k_a C_0}{k_d} \quad \text{and} \quad Ma = \frac{\mathcal{R} T \Gamma_\infty}{\sigma_0}, \quad (\text{A.5})$$

namely the solubility parameter α , the Langmuir number La and the Marangoni number Ma . C_0 is the surfactant concentration in the liquid bulk, Γ_∞ is the maximum concentration of surfactants at the interface and k_a and k_d are the adsorption and desorption kinetic constants, respectively. $\mathcal{R}$ is the ideal gas constant and T is the absolute temperature. The Langmuir number La compares the adsorption and desorption source terms at the interface. Clean interfaces are observed for $La \rightarrow 0$ while saturated interfaces for $La \rightarrow \infty$.

The level of rigidity of the interface due to the Marangoni effect can be quantified by comparing the two terms in the right hand side of Eq. A.15, namely the surface tension gradient $\nabla_I \sigma \sim La Ma \sigma_0 / R$ and the liquid viscous shear $\tau \sim \mu_L U_B / R$ experienced by a non contaminated bubble. Therefore the ‘rigidity’

parameter Λ [148, 167] is

$$\Lambda = \frac{Ma}{Bo}. \quad (\text{A.6})$$

The value of $La\Lambda$ increases with interface contamination by surfactants, which in turn result in the increase of the interfacial vorticity as shown in Eq. A.16.

A.2.2 Range of parameters for bubbly drinks

The density, surface tension and viscosities for water (Pellegrino sparkling water), a lager beer (Tecate, 4.5% ethanol), sparkling wine (Segura Viudas Brut, 12.5% ethanol) and champagne (Charles de Cazanove, 12% ethanol) were measured using a pycnometer (25ml), a tensiometer (BPT Mobile, KRUSS Scientific Instruments) and rheometer (ARES-G2 Rheometer, TA instruments, concentric cylinders), respectively. The values of these properties are shown in Table A.1. These test beverages were degassed before testing, by keeping them in a light vacuum environment (1 mbar) for one hour.

Table A.1: Values of physical properties water (carbonated water, Pellegrino), beer (lager, Tecate) and champagne (sparkling wine, Brut Segura Viudas). The values of the dimensionless numbers were obtained considering bubbles sizes ranging $0.25 < R < 1$ mm.

Fluid	ρ (kg/m ³)	μ (mPa s)	σ (mN/m)	Bo	Ar	La	Ma
Water	998	1.00	72.0	0.008 - 0.13	152 - 9726	0 - 0.1	0.1
Beer	1124	1.36	55.0	0.013 - 0.20	105 - 6700	15 - 38	0.0014
Sparkling wine	1100	1.47	48.0	0.014 - 0.23	86 - 5493	0.2 - 0.7	0.3
Champagne	992	1.44	58.0	0.010 - 0.17	73 - 4653	0.2 - 0.7	0.3

Considering bubble radii ranging from 0.25 to 1 mm, the expected values of Ar , Bo and La for beer, champagne and sparkling water, are summarized in Table A.1.

To estimate the values of La and Ma for these drinks we considered the type of surfactants and their expected concentrations [168, 169]. The equilibrium adsorption parameters for the Langmuir isotherm were determined for various surfactants from equilibrium surface tension measurements.

For champagne, the surfactants are mainly fatty acids that contribute to the organoleptic sensations [151]. For fatty acids, the Langmuir adsorption isotherm was fitted to the equilibrium surface tension measurements, which resulted in $0.02 \text{ m}^3/\text{mol} < k_a/k_d < 0.07 \text{ m}^3/\text{mol}$ and $\Gamma_\infty = 10^{-5} \text{ mol/m}^2$ [168]. Assuming that $C_0 = 10 \text{ mol/m}^3$, La ranges from 0.2 to 0.7 and $Ma \approx 0.3$. Therefore, we estimated that for

bubble diameters ranging from 0.25 to 1 mm moving in champagne, the product of the ‘rigidity parameter’ with the Langmuir number is in the range $0.2 < La\Lambda < 14$. The reader is referred to [170, 171] for a more detailed account on the sorption isotherms of small molecules.

For beer most surfactants are proteins of molecular weight ranging from 5 to 100 kDa [172, 173]. The adsorption of proteins to air/liquid interfaces is significantly different than the adsorption of small molecules. Most importantly, this adsorption cannot be described by the Langmuir isotherm. Indeed, proteins adsorption requires higher activation energy than fatty acids, once adsorbed protein unfold from their three dimensional structure and create multilayers of adsorbed proteins conferring elastic and viscous properties to the surface. Also, the adsorption of proteins can be considered as irreversible [169, 174, 175]. Nevertheless, attempts to fit adsorption isotherms of proteins with the Langmuir isotherm exist. Surface coverage measurements for albumine (molecular weight of 67 kDa) is one example. These measurements gave $k_a/k_d = 1.3 \times 10^4 \text{ m}^3/\text{mol}$ and $\Gamma_\infty = 4 \times 10^{-8} \text{ mol/m}^2$ [176, 177]. Assuming a concentration of surface active proteins ranging from 0.08 to 0.2 g/l [178] and an average molecular weight of 67 kDa we estimate $0.0012 < C_0 < 0.003 \text{ mol/m}^3$. Therefore, we obtain $Ma \approx 0.0014$ and $15 < La < 40$. For bubble from 0.25 to 1 mm in radius, we infer $La\Lambda = Ma La/Bo \approx 0.1 - 1.6$. Note that other adsorption isotherms have been derived to describe the adsorption of proteins on air/liquid or liquid/solid interfaces [179, 180] ; however, the use and implementation of these isotherms is beyond the scope of this study.

Lastly, for water, we arbitrarily assigned small values of Ma and La to denote that a small amount of surfactants are present due to ordinary laboratory conditions.

A.2.3 Experimental setup and materials

Bubble chain experiments were conducted in a rectangular plexiglass tank of $50 \times 50 \times 400 \text{ mm}^3$. At the bottom of the tank, a needle was inserted through a self-healing rubber stopper. Blunt edge needles of three different inner diameters, D_c , were used to produce bubbles of different sizes (0.16 mm, 0.26 mm and 1.55mm). Air was introduced to the needle to produce bubbles using a syringe pump (Harvard Apparatus PHD 2000 series), considering flow rates ranging from 1 to 10 ml/min. The different flow rates resulted in experiments with different bubble formation frequencies which, in turn, resulted in changes in the inter-bubble spacing.

To match the Archimedes and Bond numbers for bubbles ascending in typical beverages, a mixture of 35-65 % (by weight) of water-glycerin was used. The resulting fluid had a viscosity of 0.01 Pa s, which was measured using a rheometer (ARES-G2 Rheometer, TA instruments, concentric cylinders). The properties of the bubbles produced for each capillary are shown in Table A.2.

Table A.2: Experimental conditions. We used four capillaries with inner diameter D_c to produce bubbles of different radius R .

D_c (mm)	R (mm)	Bo	Ar	La
0.16, ◐, •	1.1 - 1.5	0.18 - 0.39	155 - 476	0.0
	0.8 - 1.0	0.12 - 0.19	57 - 115	0.5
	0.8 - 1.0	0.14 - 0.20	69 - 113	1.5
0.26, ◐, •	1.3 - 1.5	0.26 - 0.39	262 - 474	0.0
	1.2 - 1.3	0.30 - 0.35	237 - 292	0.5
	1.3 - 1.4	0.38 - 0.44	293 - 359	1.5
1.55, ◐, •	2.0 - 2.1	0.63 - 0.74	993 - 1243	0.0
	1.8 - 2.2	0.69 - 0.94	815 - 1310	0.5
	1.6 - 1.9	0.56 - 0.82	517 - 908	1.5
0.05, Spark. Water, ★	0.25	0.008	152	0.1
0.05, Spark. Water, ★	0.64	0.056	2610	0.1
0.06, Spark. Water, ★	0.76	0.078	4333	0.1
0.16, Spark. Water, ★	0.98	0.128	9042	0.1
0.05, Beer, ★	0.21	0.009	62	27.5
0.05, Beer, ★	0.38	0.028	354	27.5
0.06, Beer, ★	0.67	0.089	2015	27.5
0.16, Beer, ★	0.84	0.143	4042	27.5
0.05, Spark. Wine, ★	0.19	0.008	37	0.45
0.06, Spark. Wine, ★	0.59	0.078	1128	0.45
0.16, Spark. Wine, ★	0.77	0.133	2507	0.45
0.05, Champagne, ★	0.39	0.026	276	0.45
0.05, Champagne, ★	0.52	0.045	654	0.45
0.06, Champagne, ★	0.86	0.125	2985	0.45
0.16, Champagne, ★	0.91	0.139	3506	0.45

To calculate the value of La , we considered that for SDS aqueous solutions, $k_a/k_d = 0.11 \text{ m}^3/\text{mol}$, according to [168]. The critical micellar concentration (CMC) is 10 mM, approximately; for higher concentrations of SDS, we can consider that the surface concentration no longer increases. Therefore, we assume that $C_\infty = 10 \text{ mM}$. Therefore, different amount of SDS were added to the fluid to reach values of La relevant to those expected for bubbly drinks.

A series of experiments were conducted with actual sparkling water, beer, sparkling wine and real champagne, considering bubble sizes similar to those observed in carbonated liquids. We used a glass

capillary that was heated and stretched to have a very small inner diameter where the bubbles were formed. The properties of these liquids appear in Table A.1 and the test conditions are in Table A.2.

A.2.4 Numerical simulations

Direct numerical simulations were conducted with the in house JADIM code by solving the Navier-Stokes equations coupled with the Level-Set method. We refer the reader to [148, 181] for a detailed description of the method and its validation. Briefly, Navier-Stokes equations are solved for two Newtonian and incompressible fluids using the finite volume method (second-order accurate in time and space). Continuity is ensured through a projection method, and the capillary contribution is considered through the classical Continuum Surface Force method [182]. The interface position is tracked using the Level-Set method where the transport of the signed distance to the interface is controlled through the re-distancing method. The important aspect of the considered numerical approach used here is its ability to account for the interface contamination by the presence of surfactant in the liquid bulk. The concentration C of surfactant in the liquid bulk is given by the classical advection-diffusion equation

$$\frac{\partial C}{\partial t} + \mathbf{v}_c \cdot \nabla C = \nabla \cdot (D_c \nabla C), \quad (\text{A.7})$$

while the surfactant concentration Γ at the interface is given by [183, 184]

$$\frac{\partial \Gamma}{\partial t} + \nabla_s \cdot (\mathbf{v}_s \Gamma) = D_s \nabla_s^2 \Gamma + S_\Gamma, \quad (\text{A.8})$$

D_c and D_s are the diffusion coefficients of the surfactants in the liquid phase and along the interface, respectively, $\mathbf{v}_c$ is the velocity field in the liquid phase, $\mathbf{v}_s$ is the projection of $\mathbf{v}_c$ on the tangent to the interface, $\nabla_s = (\mathbf{I} - (\mathbf{n} \otimes \mathbf{n})) \cdot \nabla$ is the surface gradient operator and S_Γ is the adsorption/desorption source term of surfactants at the interface [184, 185],

$$S_\Gamma = k_a C_I (\Gamma_\infty - \Gamma) - k_d \Gamma, \quad (\text{A.9})$$

where k_a and k_d are adsorption and desorption kinetic constants, respectively, Γ_∞ is the maximum concentration of surfactants on the interfaces, and C_I is the surfactant concentration at the vicinity of the gas-liquid

interfaces. Considering Eqs. A.8-A.9, in a steady state ($S_\Gamma = 0$), the bubble interface is covered by the uniform concentration

$$\Gamma_0 = \Gamma_\infty \frac{La}{1 + La} \quad (\text{A.10})$$

This concentration is the relevant concentration for the interface concentration normalization (see Eq. A.16).

The variation of the surface tension σ with Γ is described using the Langmuir adsorption isotherm [186]:

$$\sigma = \max \left[\sigma_\infty, \sigma_0 \left(1 + \frac{\mathcal{R}T\Gamma_\infty}{\sigma_0} \ln \left(1 - \frac{\Gamma}{\Gamma_\infty} \right) \right) \right] \quad (\text{A.11})$$

$\mathcal{R}$ is the ideal gas constant and T is the absolute temperature. σ_∞ is a threshold surface tension, set to $0.05\sigma_0$ for stability purposes. It has been verified that the exact value of this threshold does not affect the bubbles velocity or trajectory and that the surface tension never reaches this threshold for the range of parameter investigated in this study.

A.3 Results

A.3.1 Experiments

To characterize the system we consider millimetric-sized Nitrogen bubbles forming in a capillary tube and ascending in different viscous liquids. We vary the Bond, Bo , the Archimedes, Ar and the Langmuir, La , numbers, defined in the Methods Section.

Fig. A.1 shows images of bubble chains (superposing many different instants) using a capillary tube and a clean water-glycerin mixture, considering different gas flow rates. From similitude arguments, the bubble size, liquid properties and gas flow rate are equivalent to bubbles moving in champagne [187] and other carbonated beverages[188]. In other words, the Archimedes and Bond numbers are matched as closely as possible for this experiments to those of carbonated drinks. The lateral dispersion of bubbles from the point of formation indicates that these chains are not stable. As the gas flow rate increases, the bubble frequency increases and, in turn, the separation distance in between bubbles decreases. The side-ways motion of the bubbles appears sooner as the bubble separation decreases. This is expected, since the hydrodynamic interaction is stronger for shorter distances.

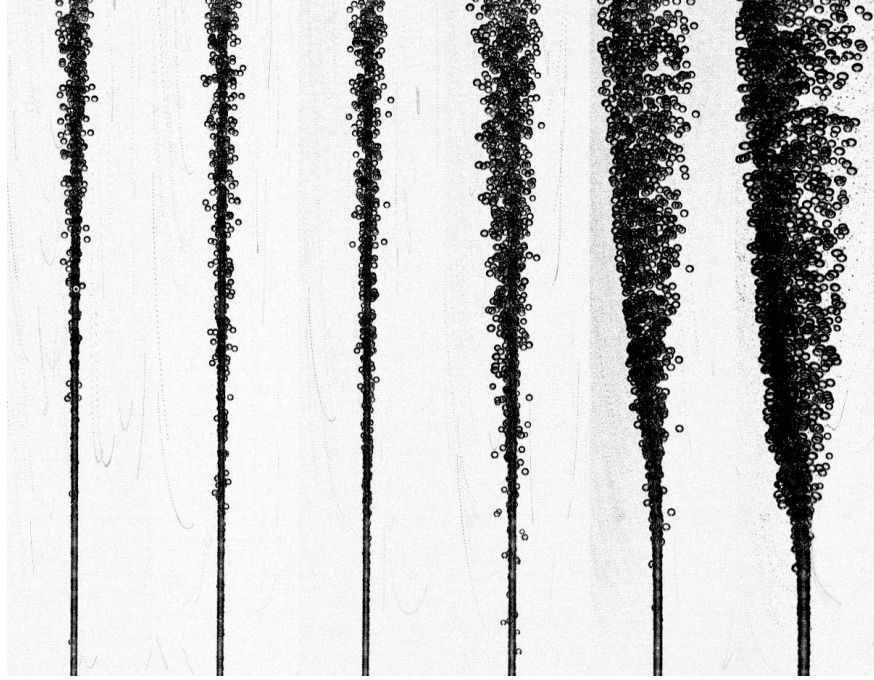


Figure A.1: Bubble chains forming from a capillary in a clean fluid. The flow rate through the capillary increases from 2 to 10 ml/min, from left to right. The bubble frequency ranges from 6 to 18 bubbles/s. $Ar \approx 150$, $Bo \approx 0.2$ and $La = 0$.

To investigate the reason behind the observed unstable behavior of bubble chains we conducted two types of experiments. First, the bubble size was increased considering the same clean fluid by changing the size of the capillary through which the bubbles are produced. These results are shown in Fig. A.2 (a). In this case, the bubble chain is unstable for small sizes (left and middle images) but transitions to a stable in-line chain behavior for large bubbles (right image). Note that, the Bond and Archimedes numbers for these large bubbles are larger than that expected for bubble-chains on champagne and other beverages (see methods section).

In the second experiment, we kept the bubble size relatively fixed by using the same capillary tube; hence, Bo and Ar did not change significantly. But in this case, we gradually added a surfactant (Sodium Dodecyl Sulfate) to increase the surface contamination. Fig. A.2 (b) shows that as the amount of surfactants increases, the bubble chains transitions from being unstable to stable. Experiments for only one flow rate are shown for both cases (increasing size and increasing surfactant); experiments for other flow rates (not shown) display the same behavior.

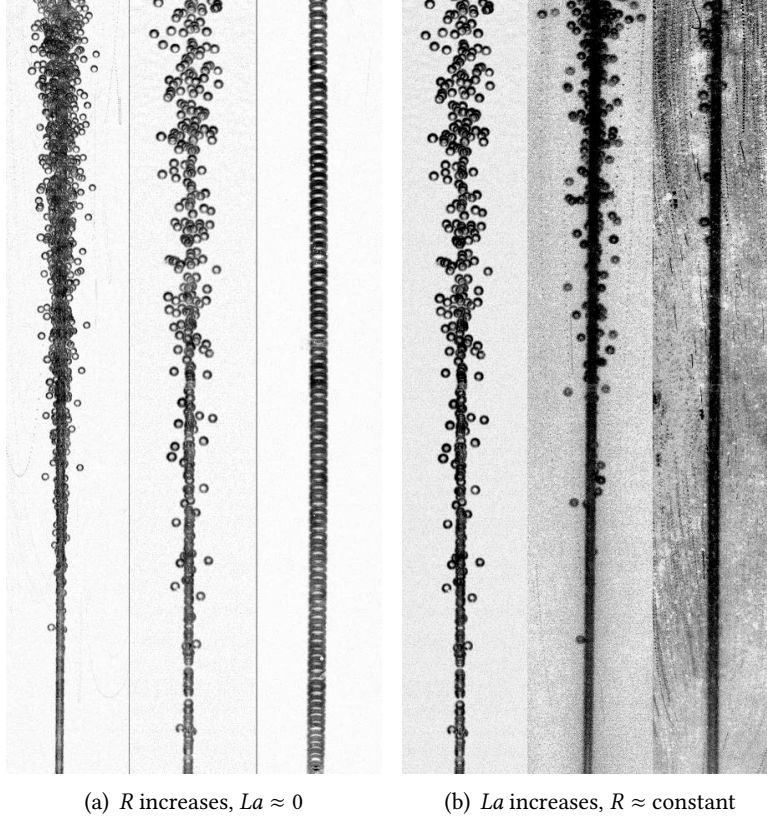


Figure A.2: The effect of bubble size and surface contamination on bubble chain. (a) The bubble size increases (from left to right), for a liquid without surfactants $La = 0$; $Ar=(150, 240, 1050)$ and $Bo=(0.18, 0.25, 0.66)$ for (left, center, right). (b) The bubble size is approximately the same, but the amount of surfactant increases (from left to right). $La=(0, 0.1, 0.5)$, $Ar=(240, 150, 225)$ and $Bo=(0.24, 0.21, 0.32)$ for (left, center, right). R is the bubble radius.

These two experiments clearly indicate that there are two possibilities to stabilize a bubble chain. However, the physical mechanisms that leads to stabilization remains unexplained. To help understand this issue, we conducted numerical simulations.

A.3.2 Numerical simulations

The details of the numerical scheme can be found in Section II.D and in the references cited therein. Most importantly, these simulations are capable of modeling the motion of deformable bubbles considering the presence of surfactants.

The simulations considered a pair of gas bubbles, initially at rest, rising due to buoyancy contained in stagnant fluid domain. To simulate the conditions found in a bubble chain, periodic boundary conditions

were imposed on the top and bottom walls of the domain. Many simulations were conducted considering different bubble sizes and contamination levels. The bubble pair is initially placed slightly off the in-line configuration (less the $0.2 R$).

Fig. A.3 shows typical results for two characteristic cases. In Fig. A.3 (a) and (b) the results of a nearly spherical bubble pair ($Ar = 400$) without surfactants ($La = 0$) are shown. The bubbles start close to an in-line configuration at the start of the simulation; after some time, the bubbles no longer stay vertically aligned, moving away from each other as shown in Fig. A.3 (a). During this initial transit period, the wake behind the bubbles develops. To visualize the wake, we show isovorticity surfaces of the stream-wise vorticity, as shown in Fig. A.3 (b). The interaction of the bubble with the wake in front of it leads to the appearance of a lift force as indicated by Eqn. A.1. For the case of spherical bubbles and the absence of surfactants, the wake has two strands of counter-rotating vorticity, in agreement with [189]. This wake structure induces a lift force in opposite directions for each subsequent bubble. Therefore, the bubbles are pushed sideways, leading to an increasing separation and deviation from the in-line configuration. These chains are unstable.

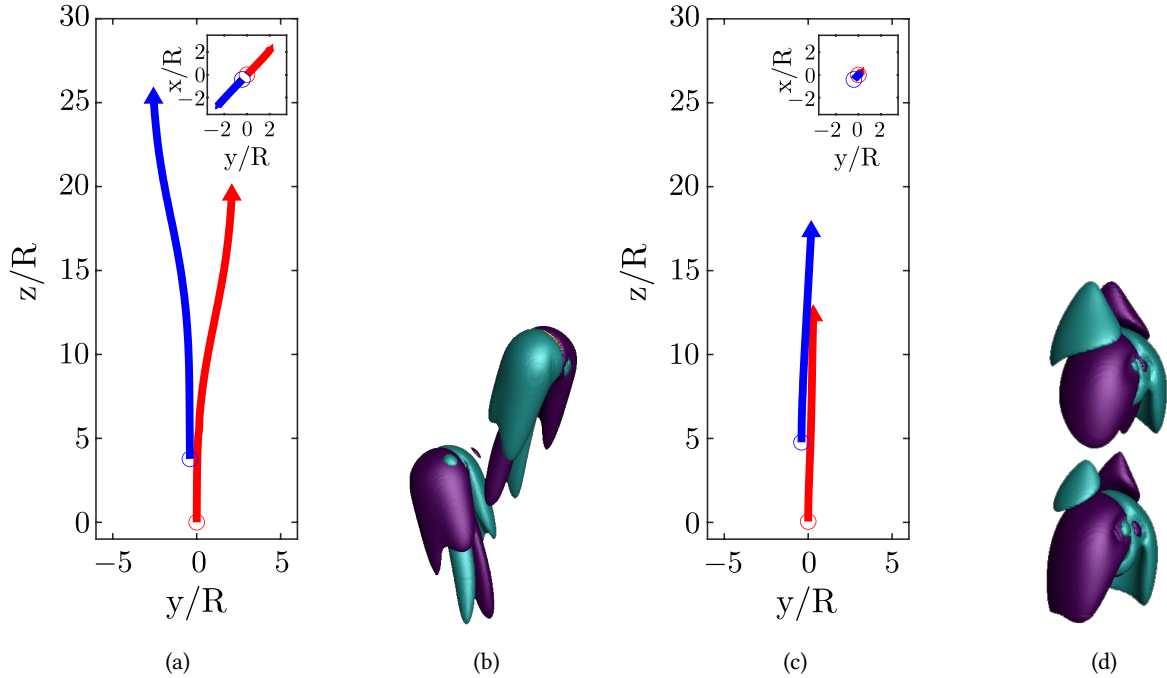


Figure A.3: Simulation of two interacting bubbles ascending in a fluid. (a) and (c) show the trajectories of the two bubbles, distances are in bubble radius units; (b) and (d) show streamwise iso-vorticity surfaces around the bubbles, $\omega_x U_B / R = \pm 0.05$. For both cases $Bo = 0.05$, $Ar = 400$; for (a-b) $La = 0$ and for (b-c) $La = 1.0$.

In contrast, by considering also spherical bubbles ($Ar = 400$) but including the effect of surfactants such that $La = 1$ the behaviour changes significantly. Fig. A.3 (c) shows the trajectory for the two bubbles. Clearly, the initial in-line configuration remains unchanged even after the initial transient period. The structure of the wake around the bubbles in this case, shown in Fig. A.3 (d), is significantly different from the case without surfactants. In particular, the vortex pair developing behind the trailing and leading bubble have the same rotation direction. This configuration induces a lift force of same direction for both bubbles; therefore, the lift force does not destabilize the in-line configuration.

As in the case of experiments, simulations were conducted for varying Ar , Bo and La to identify the conditions that lead to stability or not. In agreement with the experiments, stable chains were observed for larger bubbles (more deformed) or when surfactants were present. More importantly, since the entire flow field around the bubble pair is known, the vorticity field could be readily obtained. As discussed below, the reversal of the lift force can be attributed to the amount of vorticity created at the bubble surface. Therefore, a criteria for stability can be proposed based on the maximum surface vorticity for each case.

For larger bubbles (for higher Reynolds and Weber numbers), it is well known that the trajectories transitions to a zigzagging path [116]. Although the range of sizes for which such transition occurs is beyond those occurring in carbonated drinks, a few simulations were also conducted for such large bubbles (not shown here). It was found that the presence of surfactants, the transition to a not-straight path occurs at a smaller value of Reynolds and Weber numbers, in agreement with the experimental observations of [190].

A.4 Discussion

For the range of Bond and Archimedes numbers relevant to bubbly drinks, the in-line clean bubble configuration is unstable. In other words, stable bubble chains cannot be observed because hydrodynamic interactions lead to dispersion. The dispersion is the result of the wake-induced lift force. Increasing the bubble injection frequency increases the lateral dispersion, as shown in Fig. A.1. An increase in the bubble frequency results in a smaller separation distance between successive bubbles, for which the wake-induced lift force is larger. Hallez and Legendre [157] showed that the magnitude of the wake-induced lift is proportional to the rotational part of the wake velocity field; the lift magnitude evolves as $1/(r \sin(\theta))$

where r is the distance between the two bubbles centers and θ is the angle between the center line and the vertical.

To elaborate further on this idea, we measured the bubble chain dispersion angle, α for different unstable chains. This is done by superposing the position of bubbles at different instants onto the same frame, as done in Fig. A.1 but considering many more of bubbles. Figure A.4 show two examples for small and high bubble frequencies, (a) and (b), respectively. The angle is calculated by fitting a line from the point at which the chain first becomes unstable to the edge of the bubble cloud on the top of the image, as indicated by the yellow lines in the image. The angle is then plotted as a function of frequency for all experiments, in Fig. A.4 (c). The plot below shows only the case of small bubbles for different levels of surfactants. The dispersion angle of the unstable bubble chain increases linearly with bubble frequency. This implies that the bubble-bubble distance also influences the instability. Adding surfactants reduces the slope of the trend (the angle increases at a smaller rate with frequency), but the linearity is preserved.

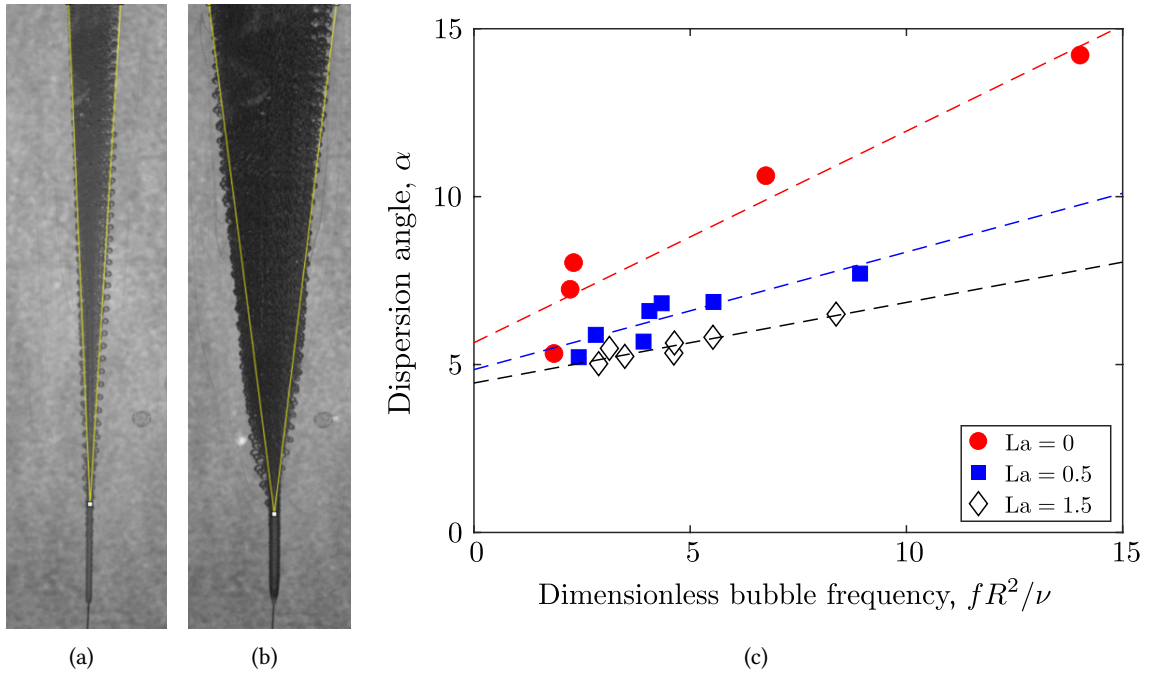


Figure A.4: Dispersion angle of the bubble chain. (a) and (b) show images of superposed bubbles over time for the 1 ml/min and 10 ml/min, respectively shown in Fig. A.1. In (c), the dispersion angle is shown as a function of the dimensionless bubble frequency, fR^2/ν , for all experiments conducted using the capillary with diameter $D_C = 0.16$ mm.

To rationalize this idea we conduct an order of magnitude analysis of the effect of bubble spacing on the induced lift force. From Eqn. A.1, we can infer that the lift force, F_L , felt by a trailing bubble evolves as

$$F_L \sim C_L R^3 \frac{U_B^2}{L} \left(1 - \frac{R}{L}\right) \quad (\text{A.12})$$

where L is the separation distance between bubbles, $L = U_B/f$, where f is the bubble frequency. When C_L has a positive value, the trailing bubble will move sideways at a speed W_B . That motion induces a viscous drag given by $F_D \sim \mu W_B R$. If we balance lift and drag in the horizontal direction (for large values of R/L), we can obtain

$$\alpha \sim \frac{W_B}{U_B} \sim C_L f \frac{R^2}{\nu} \quad (\text{A.13})$$

where α is the half-angle of the bubble dispersion of an unstable chain (assuming small α) and ν is the fluid kinematic viscosity. This expression indicates that the angle increases linearly with bubble frequency, in agreement with experimental observations shown in Fig. A.4. Furthermore, as surfactants are added C_L is expected to decrease (to eventually reverse, as discussed below), leading to smaller spreading angle. This fact is also in agreement with the experimental measurements.

Interestingly, the dependence of the dispersion angle of bubble chains with bubble frequency resembles behaviour of the so-called blooming jets [191]. For such pulsed-jets the angle at which they spread also depends linearly on the pulsating frequency.

The experiments also showed that increasing the bubbles size, without surfactants, a stable bubble chain configuration could be observed, as shown in Fig. A.2 (a). The bubble chain becomes stable because the lift force changes sign. The origin of the lift reversal for deformed bubbles in a shear flow at large bubble Reynolds number was explained by Adoua et al. [189]. The positive lift effect ($C_L > 0$ in Eq. A.1) results of the tilting and stretching mechanisms of the vorticity of the upstream flow in the bubble wake [156, 192]. The vorticity produced at the bubble surface is also tilted and stretched in the wake; however, its contribution gives rise to a streamwise vorticity of opposite sign and thus an induced lift contribution of opposite direction. When the surface vorticity exceeds a certain amount, its contribution becomes dominant and therefore the induced lift changes direction ($C_L < 0$). As the bubble size increases, the bubble shape evolves becoming more oblate which, in turn, increases the production of surface vorticity. Hence, it is

expected to observe a lift reversal as bubbles become more deformed. Such a change on the lift direction been observed experimentally for air bubbles in different fluids including water [143, 163].

For the case of bubbles in champagne and other carbonated drinks the bubble size is too small to explain the deformation-induced lift reversal. Typically, for a bubble in water, the lift reversal is observed for a bubble Reynolds number $Re \approx 1000$ corresponding to a bubble aspect ratio $\chi \approx 2$ [163]. The current experiments and simulations showed that a stable chains could be observed for small spherical bubbles if the surface was contaminated with surfactants. Therefore, we can argue that surfactants can also induce a reversal of the lift force. Such a reversal has been reported in previous experiments [164]. When a sufficient amount of surfactants is present, the bubble surface becomes immobile. In other words, the surface of the bubble can no longer sustain slip. Such non-slip condition leads to an increase of the production of vorticity at the surface which is transported to the bubble wake and is sufficient to change the wake structure. In this case, again, the lift force changes sign which implies that the trailing bubble in the wake is pushed to remain in-line forming a stable chain.

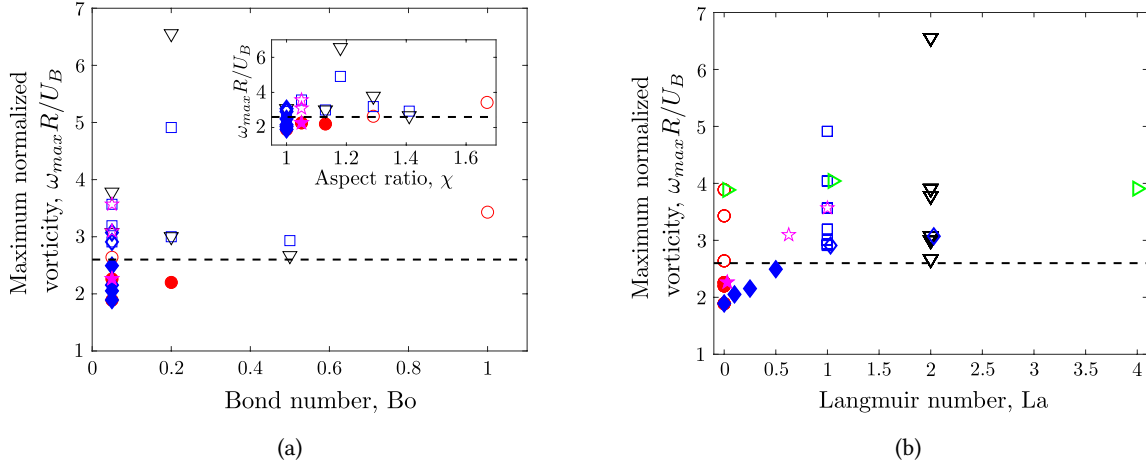


Figure A.5: Maximum normalized azimuthal vorticity on bubble surface as function of Bond number (a) and Langmuir number (b). Filled and empty symbols show unstable and stable chains, respectively. The dashed line shows the threshold value of $\omega_{\max} R/U_B = 2.65$. The red circles, blue squares and black triangles shown simulations for different values of Bo but $La = 0$, $La = 1$ or $La = 2$, respectively. The blue diamonds, magenta stars and green side-ways triangles show simulations for which both Bo and La were varied. Note that the Ar was also varied in the simulations ($400 < Ar < 11000$).

Since we have access to all flow properties from the numerical simulations we can calculate the amount to vorticity created at the bubble surface. As discussed above, we expect the surface vorticity to exceed a certain value for increasing bubble deformation, contamination or both. The magnitude of the vorticity at

the bubble surface is calculated for each time step of the simulation. To quantify the vorticity production we consider two quantities: the maximum value or a surface average value. Each of these quantities increases rapidly when the simulations starts to quickly reach a steady state value after the initial transient. The time evolution of these two quantities is very similar, varying only in magnitude. We use the maximum value following previous studies [143, 193].

The deformation of an air bubble in water can be described in terms of the Bond number, Bo , or the bubble aspect ratio, χ [163, 166]. In Fig. A.5 (a) the maximum value of the surface vorticity for each simulation is shown as a function of the Bond number, Bo , and the bubble aspect ratio, χ . For instance, for a certain bubble size, the Bond number is fixed. By progressively adding surfactants the maximum amount of surface vorticity increases. It is observed that for all cases the bubble pair becomes stable once maximum surface vorticity exceeds a threshold value of $\omega_{\max} R/U_B = 2.65$. This value can be surpassed by adding surfactants or by increasing Bo or χ . The plot also shows that either the aspect ratio or the bubble Bond number can be used to quantify the bubble deformation. We found that the transition could be readily detected by either considering the maximum or mean value of the surface vorticity. The only difference was the value of threshold.

Considering now the effect of surfactant at the interface, in Fig. A.5 (b) the maximum vorticity is shown as a function of the Langmuir number for different values of the Bond number. As in the previous case, it is clear that the bubbles remain in an in-line configuration once the surface vorticity exceeds the threshold value. Note that some additional simulations are shown here where the value of La gradually increased for a fixed Bo (blue diamonds). The results are in good agreement with the rest of the simulations.

Additionally, to further sustain the argument above we projected the value of the surface vorticity onto the bubble surface for a few selected cases, shown in Appendix A. The images show that the maximum vorticity is located at the line of maximum curvature for uncontaminated bubbles ($La=0$) and at the location where the surfactant gradient is highest for contaminated bubbles ($La>0$).

As shown above, the threshold value for $\omega_{\max} \sim 2.65 U_B/R$ is observed at the transition from unstable to stable bubble line. The same vorticity amount is required to reverse the lift force when either bubble deformation or bubble surface contamination are increased. This confirms that the transition from unstable to stable is related to the reversal of the lift experienced in the wake of bubble.

To understand why this is the case, let us consider the stress balance at the bubble interface. At the bubble surface, the viscous shear stress, Σ , balances the gradient of surface tension, $\nabla_I \sigma$:

$$\mathbf{n}_b \cdot \Sigma \cdot (\mathbf{I} - \mathbf{n}_b \mathbf{n}_b) = -\nabla_I \sigma \quad (\text{A.14})$$

where $\mathbf{n}_b$ is the normal vector to the bubble surface and $\nabla_I = (\mathbf{I} - \mathbf{n}_b \mathbf{n}_b) \cdot \nabla$ is the surface gradient operator. The gradient of surface tension can be expressed as a function of the surfactant concentration using the Langmuir adsorption isotherm [186]. The tangential viscous shear along the flow direction is $\mu(\partial u_I / \partial r - u_I / R_c)$ and the interfacial azimuthal vorticity writes $\omega_I = \partial u_I / \partial r + u_I / R_c$ with u_I the local bubble interfacial velocity and R_c the radius of curvature. The interfacial vorticity can then be directly expressed as a function of the two above mentioned mechanisms, namely deformation and surface contamination, as

$$\omega_I = 2 \frac{u_I}{R_c} + \frac{\mathcal{R} T \Gamma_\infty}{\mu(\Gamma_\infty - \Gamma)} \nabla_I \Gamma \quad (\text{A.15})$$

This relation shows that both the surface deformation and the surface contamination can increase the magnitude of the interfacial vorticity ω_I . Indeed, deformation reduces at the bubble equator the radius of curvature R_c and the surface contamination results in an accumulation of surfactants at the rear of the bubble. This expression can be normalized by considering the bubble equivalent radius R , the characteristic rising velocity of a bubble $\rho g R^2 / \mu$ and the concentration of surfactant at equilibrium $\Gamma_0 = \Gamma_\infty La / (1 + La)$ (see section Material and Methods). The normalized vorticity ω_I^* (denoting all normalized variables with $*$) at the interface can be written as:

$$\omega_I^* = 2 \frac{u_I^*}{R_c^*} + \frac{La \Lambda}{1 + La(1 - \Gamma^*)} \nabla_I^* \Gamma^*. \quad (\text{A.16})$$

With these ideas, we can now propose a stability map that considers both the effect of bubble deformation and surface mobility (first and second terms in the right hand side of Eq. A.16, respectively) as the main factors determining the stability of a bubble chain. The bubble deformation can be readily quantified by the Bo number. While La quantifies the amount of surfactants that can be transferred from the liquid to the bubble surface, a better measure of the effect of surfactants on the interface behavior is given by the parameter $La \Lambda$, as demonstrated by Eq. A.16.

Fig. A.6 shows both experiments and simulation results in a map of Bo and $La\Lambda$. The filled and empty markers show unstable and stable chains, respectively. The combination of bubble deformation and surface rigidity clearly determine the stability conditions of a bubble chain. Furthermore, the transition results from the amount of vorticity created at the bubble surface as indicated by the threshold value (dashed line). If the contamination level and the bubble size are known, the map presented here can be used to determine the stability of a bubble chain. Again, small bubbles, as those typically observed in carbonated drinks are not expected to be stable in clean liquids. However, the presence of surfactants leads to surface immobilization to result in stable chains.

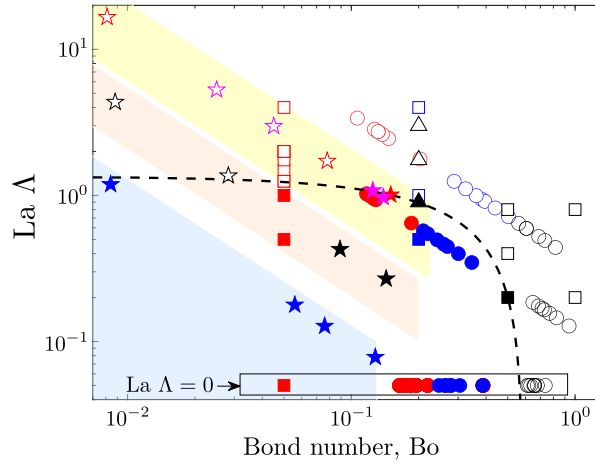


Figure A.6: Stability map showing the stable and unstable chain behaviour in terms of the Bond number and the product of the rigidity parameter with the Langmuir number. Filled and empty symbols show unstable and stable chains, respectively. Circles and stars show experimental data, while squares and triangles show numerical results. The data enclosed in the rectangle corresponds to cases for $La\Lambda = 0$ (clean fluids). The dashed line presents an line for which $\omega_{\max}R/U_B \approx 2.65$ which marks the transition. The stars (blue, black, red and magenta) show the measurements conducted with carbonated water, beer, sparkling wine and champagne, respectively. The yellow, orange and blue shaded areas represent the expected range of values for champagne, beer and water, respectively, according to Table A.1.

We can also note that when bubbles are very small, for $Ar < 1$, a wake will not develop [38] and therefore, regardless of contamination, a lift force does not appear. In aqueous liquids, an $Ar < 1$ corresponds to $Bo < 10^{-3}$. Bubble chains to the left of this value in the map shown in Fig. A.6 will be stable.

In this study, we neglect the effect of bubble growth. Since the growth rate of the bubbles is smaller than the bubbles velocity, the flow field around the bubbles is decoupled from the bubble growth, i.e, at every moment, the flow field is at equilibrium with the bubble size and shape. Therefore, Fig. A.6 can be used to predict the evolution of chain stability as the bubbles grow. Indeed, in that situation, Bo and Ar increase

as the bubble rise; however, $La\Lambda$ decreases as the bubble grows. Therefore, the chain may transition from stable to unstable and vice versa, as the bubbles grow.

To end and to sustain that these claims are valid for bubbles in typical carbonated drinks, including champagne, we attempt to quantify the Bond number, the rigidity parameters and the Langmuir number for three types of carbonated beverages. We also conduct experiments with actual beverages. The calculation of Bo is straight forward, since the values of density, size and surface tension are known. For the calculation of Λ and La , on the other hand, some assumptions must be made of the surface absorption and nature of the typical surfactants in these beverages (see Methods section for details). Considering bubbles ranging in radius from 0.25 to 1 mm, carbonated water is not expected to produce stable chains since a small amount of surfactants is present. The blue shaded area in Fig. A.6 show the range of parameters valid for water. For champagne, the type of surfactants and the quantities are well studied and known. Hence, it is possible to confidently assign the range of expected values of Λ and Bo for the same range of sizes. The yellow shaded region, shown in Fig. A.6, clearly indicates that for this beverage stability of bubble chains is expected for small bubbles. This is in agreement with what is typically observed. For the case of beer, on the other hand, we had to make assumptions to infer Λ , since the typical surfactants are very different. We predict that for the same range of bubbles sizes, denoted by the orange-shaded area in the figure, bubble chains can either be stable or not. This observation can be seen in some beer glasses, but more carefully conducted experiments are needed to assess bubble chain stability and to understand how proteins immobilize the surface of bubbles in beer.

The experiments conducted with very small bubbles in water, beer, sparkling wine and champagne are also shown in Fig. A.6, depicted by the stars. Special glass-pulled capillaries were fabricated to produce bubbles in the size range of actual beverages. The sizes and values of the relevant parameters are shown in Table A.2. The chains were stable for sparkling wine and champagne, considering small bubbles. Note that in one case, we were able to produce a weakly unstable bubble chain for sparkling wine ($Bo = 0.13$). For beer, the smallest bubble size produced stable chains, while the other two sizes showed unstable chains. For water, experiments with these small bubble sizes were all unstable. These observation are in good agreement with our experiments with mock fluids and numerical simulations.

A.5 Conclusions

In summary, we have demonstrated that the stable bubble chains typically observed in champagne occur due to the presence of surfactants. For certain bubble sizes, the bubbles can only move in-line if enough vorticity is produced on their surface. If bubbles are small, the deformation is not enough to produce sufficient vorticity. Hence, surface immobilization (resulting from the presence of surfactants) provides a second source of vorticity that leads to a stable chain. The current results explain why one observes stable bubble chains in champagne, but also provide an interesting case study of sources of vorticity in bubbly two-phase flows. Additionally, since these effects are now identified, the stability of a bubble chain could be used to assess the level of surfactant contamination in any liquid. This idea could be of practical use.

B THE MECHANICS OF THE SQUASH NICK SHOT

Abstract Squash is a widely popular racket sport, practiced by millions of people worldwide, played inside a walled court. When played well, players can last for several minutes before the ball bounces twice on the floor. There is, however, an unanswerable shot. When the ball hits the nick between a vertical wall and the floor, under certain conditions, it rolls without any vertical bounce. We study this process experimentally. We determined that the ball must hit the vertical wall first at a narrow range of heights above the floor, but most importantly, it must touch the floor before finishing its rolling time on the vertical wall. When the rolling time is shorter than the contact time, the vertical momentum is canceled due to a mechanical frustration condition. This behavior is explained considering a contact model, which agrees with the experimental observations. In addition to its relevance to other walled ball sports, this understanding could help design rolling shock dampers with possible practical applications.

Information This work is published in *Proceedings of the National Academy of Sciences* [4]. We thank Mark Johnson and Alex Zaslavsky from the Rhode Island Squash Association and Stefano Crema from Squash Busters Providence for animated discussions. We are also grateful to Coach Arthur Gaskin, from Brown University, for his wisdom in all things squash.

B.1 Introduction

Squash is a racket sport played in a four-walled court between two players. Players take turns hitting the ball in allowable areas on the walls such that the opponent is unable to return the ball back before bouncing twice on the floor. According to the Professional Squash Association [194], squash is played by 3 million people in over 150 countries. The sport was recently approved to be included in the 2028 Summer Olympics games in Los Angeles.

The pace, speed and court size of racket sports is determined by the mechanics of the ball [195, 196]. What distinguishes squash from other racket sports is the ball and its interaction with the walls. The ball is a soft-deformable hollow rubber shell that ‘squashes’ significantly when it impacts the wall (hence the name); it is characterized by a low coefficient of restitution [197, 198]. Professional players become incredibly

skilled at retrieving the ball from all parts of the court making rallies long and physically demanding. One shot in squash is highly sought-after because it is unanswerable, the nick shot. When the ball hits the nick (the corner between a vertical wall and the floor), in some conditions, it rolls on the floor instead of bouncing [199]. This shot occurs roughly 8.7 % in professional games (see Supplemental Information, SI); hence, it plays a significant role in the outcome of a match. Similar shots are also observed in racquetball, padel, Gaelic handball, fronton (American handball) and Jai Alai (in this sport, the nick shot is called “chula”). From our observations, we determined that nick shots occur when the ball first hits one of the vertical walls (side or back) moving approximately perpendicular to it (see Fig. B.1(a)), descending at angles of incidence, θ_0 , ranging from 13 to 40 degrees, and at a range of different speeds. To our knowledge, no mechanical interpretation of this peculiar behavior exists.

Experiments were conducted to identify the set of conditions needed to observe a rolling nick shot. We rationalize our measurements by placing the results on a map of the ratio of the characteristic times (contact to rolling) and the dimensionless elasticity of the ball (the Cauchy number) for collisions near a nick. We conclude by offering an explanation of the behavior sustained on a mechanical model that predicts the conditions at which the ball ceases to bounce vertically after the interaction with the nick. Understanding the mechanics of the nick shot could be relevant beyond squash and sports mechanics. This knowledge could be useful in designing rotational dampers in sport injury prevention [200] and manufacturing control [201].

B.2 Experimental Setup

The experiments were conducted in the controlled environment of our lab rather than on the squash courts. To simulate the player hitting the squash ball, a pressurized air cannon was used to launch a squash ball towards an acrylic wall. The air cannon, constructed of PVC piping with an inner diameter close to that of the diameter of the balls, a plastic chamber, a manometer, and a valve. Once pressurized to a certain value, the valve was quickly opened to release the squash ball at a certain speed U_0 , aimed at the corner at an incident angle θ_0 . The pressure inside the air cannon was varied from 15 psi to 40 psi to produce a range of impact speeds, as seen in Table 1 in the main manuscript. The acrylic wall represents the nick, where the vertical wall and the floor meet. The corner, constructed from two pieces of 7.5 cm thick acrylic plate, was

mounted using aluminum profiles about 1 meter away from the air cannon. The angle at which the acrylic corner was mounted was then varied from 20 to 40 degrees to produce different incident angles. The impact location H was varied by displacing the acrylic corner back and forth, while keeping the position of the cannon fixed.

Three different squash balls were used, a single dot (for intermediate), a double dot (for professional players), and a hard ball (used in doubles), all manufactured by Dunlop. The diameter of the balls were $D = 40$ mm, and their masses were $m = 24$ grams.

The ball's Young's modulus was determined experimentally following the procedure described below. For the collision experiments and the compression tests, the balls were tested at room temperature, 22°C, and at 40°C, which we determined to be a typical temperature for a squash ball during a hard game. The ball was heated using a heating pad. The temperature was measured using a hand-held infrared heat gun, with an accuracy of $\pm 2^\circ\text{C}$.

The interaction between the squash ball and the wall was recorded using a high-speed camera (*Photron FASTCAM SA5*) filming at 5000 fps, with a shutter speed of 1/20000. Examples of typical experiments can be seen in movies below. The squash ball was marked up with white stripes to help image tracking.

The images collected were analyzed using image processing techniques on Matlab. Using the diameter of the squash ball for calibration, the location of the ball was tracked in time, as shown in Fig. 1(c) of the main manuscript. Hence, both vertical and horizontal velocities of the ball geometric center were determined during the interaction with the wall. The ball speed was determined in two directions, considering a central difference scheme. The occurrence of a nick shot was determined from these measurements. A collision was considered a nick if the vertical velocity after the interaction was smaller than $0.1U_0 \sin \theta_0$.

B.2.1 Measurement of the ball's Young's Modulus

To determine the Young's Modulus of the squash balls, compressive tests were conducted using an Instron machine. The machine was set to compress each ball at a controlled rate of 2 mm/s up to a predetermined deformation limit of 25 mm, well below the permanent deformation limit. The applied force as a function of deformation showed a non-linear behavior. We, therefore, used the two-parameter hyperelastic Gent model [202, 203], to determine the effective Young's modulus of the balls. The stress, σ was calculated as the ratio of the compression force over the cross sectional area of the ball. The strain, ϵ , was calculated as

the ratio of the deformation and the ball diameter. The data was fitted to

$$\sigma = G_m \left(\lambda - \frac{1}{\lambda^2} \right), \quad (\text{B.1})$$

where, $\lambda = 1 + \epsilon$ is the stretch ratio and G_m is normalized shear modulus defined as

$$G_m = G J_m / (J_m - I_1 + 3)$$

where G is the shear modulus, J_m is the locking parameter and I_1 is the first invariant of the left Cauchy-Green deformation gradient tensor. The fitting of the data to this model yielded the material constants, G and J_m . The Young's modulus was determined by

$$E = 2G(1 + \nu), \quad (\text{B.2})$$

where $\nu = 0.5$ is the Poisson ration for rubber. The values of E are shown in Table 1 of the main document.

B.2.2 Materials and Methods

Controlled collisions were produced with an air gun. It deployed squash balls (Dunlop, Professional 2-Dot, one yellow dot and elite doubles) at a range of speeds, close to those use in regular squash. The incident angle and the initial contact with the vertical wall was varied, by moving the target and its inclination. The collisions were recorded with a high speed camera. The position was determined using image processing software; the velocity was calculated using a central-difference scheme. The ball was directed to hit the nick between a vertical and a horizontal wall at different locations, H , speeds, U_0 , and incidence angles, θ_0 , see Fig. B.1(a). Experiments were conducted for three different balls, two different ball temperatures and two fixed incident angles. All experiments consider no initial rotational speed, $\omega_0 = 0$. Table B.1 summarizes all the experimental conditions considered.

We rationalize the interaction by considering the following phases: (i) contact with the wall, $t = t_1$; (ii) compression and rolling, $t_1 < t < t_2$; (iii) contact with the floor $t = t_2$; and (iv) bounce or nick process, $t > t_2$. Figures B.1(a) and B.1(b) show nomenclature and snapshots of a typical collision at $t = t_1$ and at $t = t_2$, respectively. Figures B.1(c) and B.1(d) show the trajectory and the speed time evolution, respectively,

Table B.1: Range of conditions and ball properties for all experiments. For all cases, $0.5 < H^* < 1.0$.

Case	Ball type	E (kPa)	U_o (m/s)	Ca
1	Single dot (○, ●)	99	33 to 58	0.05 to 0.12
2	Double dot (□, ■)	105	18 to 58	0.04 to 0.13
3	Doubles (△, ▲)	471	17 to 58	0.16 to 2.26
4	Single dot, at 40° C (○, ●)	96	30 to 52	0.04 to 0.15
5	Double dot, at 40° C (□, ■)	90	33 to 56	0.04 to 0.11
6	Doubles, at 40° C (△, ▲)	450	30 to 55	0.21 to 0.62

of three typical collisions (cases C1, C2 and C3, see movies in the SI). In particular, the case C1 shows the nick behavior, depicted by the red symbols in the figure. The ball first touches the vertical wall at a point A , at distance $H^* = H/D > 0.62$, at $t = t_1$. Immediately after, the ball slows down as it exchanges momentum with the wall. As the ball compresses, it also rolls without sliding at the contact point A [204] (conditions discussed in the SI). Hence, the ball's center of mass continues to move downwards (see vertical speed of the ball, U_{cm} , for $t_1 < t < t_2$), even though the point A is fixed. As the interaction evolves, the ball eventually touches the floor on point B at $t = t_2$, Fig. B.1(b). Soon after this happens, the vertical velocity of the ball becomes zero. The most impressive part of the nick behavior is displayed after this moment. As the ball decompresses, its horizontal velocity gradually increases until losing contact with the wall but, most importantly, its vertical velocity remains very close to zero. In other words, the ball leaves the corner interaction with a nearly horizontal trajectory, as seen in Fig. B.1(c). Two other cases are shown in the figure, for $H^* = 0.51$ and $H^* = 0.81$. Even though the ball collides at approximately the same speed and incident angle, the nick behavior is not observed (detailed description in SI).

B.3 Condition for a nick shot

We hypothesize that the nick will occur if the ball makes contact with the floor before it has ended its rolling motion. In other words, the contact time has to be longer than the rolling time. We can estimate the characteristic time for the compression, t_c , considering the collision of an elastic ball against a rigid wall [205]. The rolling time, t_r , can be calculated by considering the displacement of the ball's center of mass during contact and the rotational inertia of the ball (details of calculations appear in the SI).

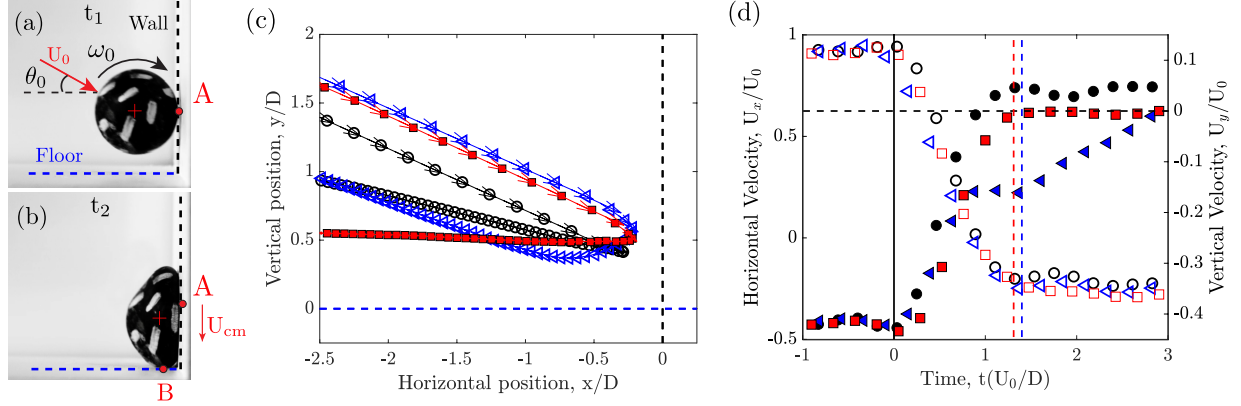


Figure B.1: (a) Nomenclature, ball first contact with vertical wall, $t = t_1$. (b) Ball first contact with floor, $t = t_2$. (c) Trajectory of the ball for three cases: Case C1, filled red squares, nick shot, $Ca = 0.25$, $H^* = 0.62$, $\theta_0 = 24.43^\circ$ and $\tau = 0.53$; Case C2, empty black circles, ball hits wall and floor simultaneously, $Ca = 0.24$, $H^* = 0.51$, $\theta_0 = 24.34^\circ$ and $\tau = 0.76$; Case C3, empty blue triangles, ball hits vertical wall at higher H , $Ca = 0.25$, $H^* = 0.81$; $\theta = 24.31^\circ$ and $\tau = 1.10$. (d) Normalized normalized ball velocity (U_x/U_0 and U_y/U_0) as function of normalized time, tU_0/D . Empty and filled symbols show horizontal and vertical velocities, respectively (left and right vertical axes, respectively). Case C1, red symbols; C2, black symbols; C3 blue symbols; vertical solid black line corresponds to time $t_1 = 0$, vertical red dashed line shows t_2 for case C1; vertical dashed blue line shows t_2 for case C3.

If we define $\tau = t_r/t_c$, the condition for a nick shot is

$$\tau < 1. \quad (\text{B.3})$$

Considering explicit expressions for t_c and t_r , we obtain

$$\tau = \beta H^* Ca^{2/5} \frac{(\cos \theta_0)^{1/5}}{\sin \theta_0 + 2\kappa\omega^*} \quad (\text{B.4})$$

where $\beta = 0.6058$ is a numerical constant, $\omega^* = \omega_0 D/U_0$, $H^* = H/D$ and Ca is the Cauchy number defined as

$$Ca = \frac{E}{\rho_{ball} U_0^2}$$

where E is the Young's modulus and $\rho_{ball} = 6m/(\pi D^3)$, is the density of the ball. Using Eqn. B.10, a map of conditions for the nick to occur can be obtained, considering the collision kinematic conditions (U_0 , ω_0 , θ_0), the location of first contact with the wall (H^*), and the ball elasticity parameter (Ca).

B.3.1 Characteristic times for wall collision

To identified the conditions for the nick shot to occur, two characteristic times are needed: the compression time and the rolling time. Here, we detail how those times are determined.

Contact time The contact time for the impact of an elastic sphere is determined by the balance of the impact force and the elastic deformation, neglecting friction and energy dissipation [205]. The impact time is give by

$$t_c = \alpha \left(\frac{m^2}{DE^2 U_0 \cos \theta_0} \right)^{1/5} \quad (\text{B.5})$$

where $\alpha = 3.29$, m is the mass of the ball, U_0 is the ball speed at contact, θ_0 is the angle at contact and E is the Young's modulus.

Rolling time Assuming that there is no slip between the ball and the wall, a condition met for contact with friction and at small incident angles [204], the ball will execute a rolling motion on the wall with in contact with it. The time that the wall will take to reach the floor, while rolling, is

$$t_r = \frac{H}{U_{cm}}$$

where H is the distance above the floor where the ball first makes contact with the wall. U_{cm} is the velocity of ball's center of mass at time t_1 (see Fig. 1(a) in the main text), which can be defined as

$$U_{cm} = \frac{D(U_0 \sin \theta_0 - D\omega_0/2)}{D_J + D} \quad (\text{B.6})$$

where $D_J = 2J/(mD)$, J being the ball's polar moment of inertia and ω_0 is the ball's angular velocity at the moment of contact. This calculation the classic rolling theory [205]. Therefore, we can rewrite t_r as:

$$t_r = \frac{H(2D_J + D)}{D(U_0 \sin \theta_0 + D_J \omega_0)} \quad (\text{B.7})$$

where $D_J = 2J/(mD)$. A squash ball is a thick spherical shell of thickness T , for which the polar moment of inertial is

$$J = \frac{1}{10} m D^2 \frac{1 - (1 - 2T/D)^5}{1 - (1 - 2T/D)^3}. \quad (\text{B.8})$$

Considering that $T/D = 0.113$ for a squash ball, $J = \kappa m D^2$ where $\kappa = 0.1346$.

Therefore, the rolling time simplifies to:

$$t_r = \frac{H(4\kappa + 1)}{U_0 \sin \theta_0 + 2\kappa D \omega_0} \quad (\text{B.9})$$

The time ratio The ratio of rolling to contact time is defined as

$$\tau = \frac{t_r}{t_c}.$$

Therefore, from Eqns. B.5 and B.9, we can write

$$\tau = \frac{1}{\alpha} \left(\frac{DE^2 U_0 \cos \theta_0}{m^2} \right)^{1/5} \frac{H(4\kappa + 1)}{U_0 \sin \theta_0 + 2\kappa D \omega_0}$$

After simplification, we have

$$\tau = \beta H^* \text{Ca}^{2/5} \frac{(\cos \theta_0)^{1/5}}{\sin \theta_0 + 2\kappa \omega^*} \quad (\text{B.10})$$

where $\beta = 0.6058$ is a numerical constant, $\omega^* = \omega_0 D / U_0$, $H^* = H / D$ and

$$\text{Ca} = \frac{E}{\rho_{ball} U_0^2}$$

where Ca is the Cauchy number and $\rho_{ball} = 6m / (\pi D^3)$.

B.4 Results and discussion

A vast experimental campaign was conducted (Table B.1) to test the conditions set by Eqn. B.10. In all experiments, no initial rotation was considered ($\omega^* = 0$) and only two representative incident angles, θ_0 , were tested. Figure B.2 shows all the experimental results. In these maps, we plot the value of τ for a range of values H^* and Ca. On the plots, the filled symbols indicate the cases nick was observed. A shot was considered to be a nick if the rebound vertical speed was smaller than $0.1 U_0 \sin \theta_0$. Clearly, two conditions can be identified: $\tau < 1$ and $0.6 < H^* < 0.75$.

Even without any fitting parameters, Eqn. B.10 predicts the conditions very well.

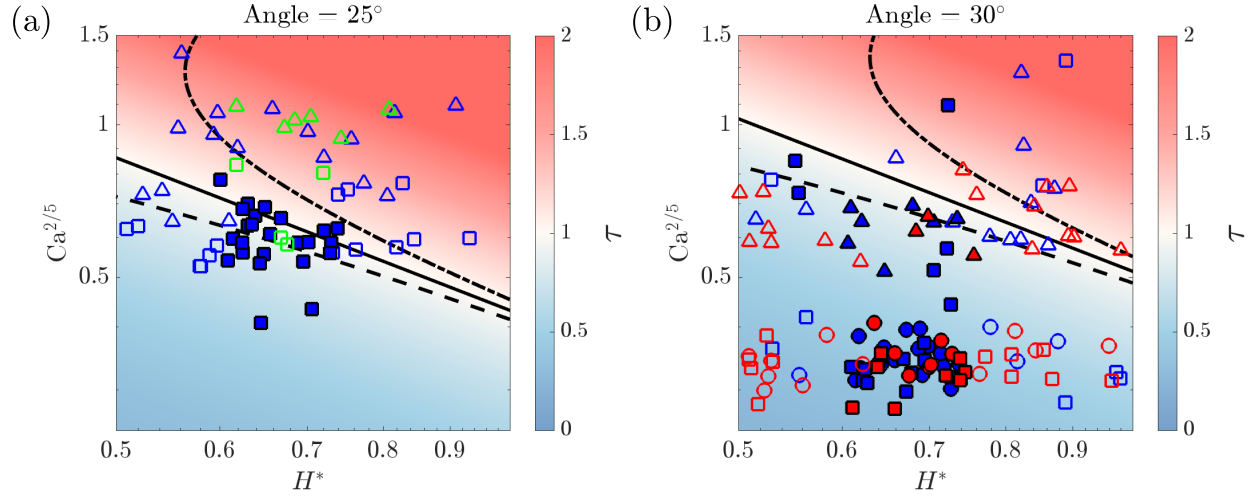


Figure B.2: Map of conditions for nick occurrence: value of the time ratio τ (indicated by the color) as a function of $Ca^{2/5}$ and H^* for a fixed incidence angle θ_0 and no initial rotation $\omega_0 = 0$. (a) $\theta_0 = 25^\circ$; (b) $\theta_0 = 30^\circ$. Symbols according to Table B.1. Filled symbols (resp. empty) show the cases when the nick shot was observed (resp. was not observed). Green symbols on (a) show experiments when the wall was lubricated with a viscous fluid. The dot-dashed and dashed lines consider $\omega_0 = \pm 100 \text{ s}^{-1}$, topspin and backspin, respectively.

Several other conditions were also tested, including ball temperature, ball type, ball speeds, and wall lubrication, all of which are shown in Fig. B.2. A detailed account appears in the SI, including a discussion on the effect of initial ball spin. The conditions for the occurrence of the nick shot hold for all these tests.

Considering a mechanical model (described in the SI), we can explain why the ball does not bounce vertically when the nick shot conditions are satisfied. The rolling process that occurs when the ball is in contact with the vertical wall cannot proceed when a second contact point, with the floor, is imposed. This second contact point induces a torque in the opposite direction of the rolling motion, canceling it, resulting in a mechanical equilibrium condition for which both the linear and rotational momentum of the ball become null. In other words, the ball gets stuck on the corner when the nick conditions are met. Because of the energy stored during the compression phase with the vertical wall, the ball can still rebound horizontally. This is why the ball moves away from the wall after the collision with no vertical momentum.

Understanding the nick mechanics can be used to refine squash technique. Mastering the nick shot requires precision and low τ , achieved with higher speed (lower Ca), steeper angles, or softer balls (lower E). The ideal shot comes from a fully extended arm, hit with force, and when the ball is warm, in accordance to what is observed in professional play.

The theory developed for the mechanism of the nick shot could have practical use beyond squash. Abrupt changes in angular momentum, typically associated with head impacts [206–208], could be reduced. Our model predicts that increasing mass prolongs momentum transfer ($\propto m^{2/5}$), while larger size ($\propto D^{-1/5}$) and greater stiffness ($\propto E^{-2/5}$) shorten it. Thus, a heavier, larger, and more compliant damper that slides on impact would be more effective in reducing large rotational rates.

Statistics of nick shot occurrence To determine how often nick shots are played, one of us (RZ) watched all the matches (men and women, 32 matches) played at the Tournament of Champions of 2023, transmitted by SquashTV. This prestigious tournament, part of the professional tour, is played every January in New York City. We counted the number of times a player won a point with a nick shot. It occurred 8.7 % times. Note that not all of these shots were rolling nicks. Those occurred only 1.2% of all winning shots.

B.5 The mechanical model of the ball contact

B.5.1 Stage 1: Contact with the wall, $t = t_1$

Before making contact with the vertical wall, the ball travels at a velocity $\mathbf{U}_0$, approaching the wall at an angle θ_0 , as depicted in Fig. 1(a) of the main text. When the ball reaches the wall, its horizontal location is $x/D = 0.5$ while its vertical location, $H^* = y/D$, depends on how close to the floor it reaches the vertical wall. The ball makes contact with the wall at a material point on its surface, denoted as A . After making contact, the ball experiences an impulse which changes its center of mass velocity, as well as its angular velocity. From the balance of linear and angular momentum, the change of the ball's center of mass velocity is given by

$$\Delta U_{cm} = D_J \Delta \omega$$

where $\Delta \omega$ is the change of angular velocity before and after the impact and $D_J = 2J/(mD)$, with J being the polar moment of inertia (see Eqn. 5 above).

Assuming that the velocity of a material point of the ball at location A (no slip) is zero, we can conclude that

$$U_0 \sin \theta_0 + \Delta U_{cm} - \omega D/2 = 0$$

and that

$$\Delta\omega = \frac{\frac{1}{2}U_0 \sin \theta_0 - \omega_0 D}{D_J + D}.$$

Therefore, the speed of the center of mass immediately after contact is

$$U_{cm} = \frac{D(U_0 \sin \theta_0 - D\omega_0/2)}{D_J + D}. \quad (\text{B.11})$$

B.5.2 Stage 2: contact with the floor, while already in contact with the wall, $t = t_2$

As described in the main text, when the ball has not yet concluded its rolling motion on the vertical wall but makes contact with the floor, the nick behavior is manifested. In this case the ball is observed to roll on the floor with no vertical velocity.

When the ball makes simultaneous contact with the vertical wall, at point A with the vertical wall and at point B with the floor, it experiences impulses from these two points that will change the ball's velocity. Let the change of the ball's center of mass at $t = t_2$ be

$$\Delta U_{cm}(t_2) = P_B/m + P_A/m$$

where P_B and P_A are the impulses at B and A , respectively.

At point A , considering that at $t = t_2$ the material point is still fixed (non slip), we will have

$$U_{cm}(t_2) = D\omega/2. \quad (\text{B.12})$$

Now, we can also assume that at the material point B the the velocity is null. Therefore, following a similar argument as before, we can conclude that

$$U_{cm}(t_2) = -D\omega/2. \quad (\text{B.13})$$

The only possibility for Eqns. B.12 and B.13 to be both satisfied is for $\omega = 0$. From it, it follows that $U_{cm} = 0$.

B.5.3 Stage 3: restitution

As shown above, if the ball makes simultaneous contact with the vertical wall and floor, while rolling on the vertical wall, its velocity and rotation cancels out. However, due to the impact, the ball still has energy stored in elastic deformation. Since the energy is mostly from deformation in the horizontal direction, the ball will regain kinetic energy in this direction. Due to the contact at point B , the ball will roll as the elastic energy is restored into kinetic energy in the horizontal direction.

B.6 Detailed description of experiments

Experiments when the nick shot is not observed In this section we provide a detailed description of two experiments shown in Fig. 1(c) and 1(d) (case C2 and C3) that do not exhibit the nick behavior.

Our observations indicate that the nick behavior occurs when the balls touches the floor before it has completed the rolling phase with the vertical wall. To illustrate that this is indeed the nick condition, we show two other cases in Fig.1 of the main document, with very similar conditions but reaching the corner at slightly different H^* values. Case C2, shown in Figs. 1(c) and 1(d) by the black markers and the movie below, displays a collision for which $H^* = 0.51$; in this case, the ball makes contact with both vertical wall and floor at approximately the same time ($t_1 \approx t_2$). The compression-decompression cycle occurs with both walls at the same time and rolling does not occur. As explained above, rolling is not possible while having two no-sling contact points. The ball bounces off with at a slightly lower angle that the incoming one, see Fig. 1(d). Case C3, displayed by the blue symbols, also in Figs. 1(c) and 1(d) and movie below, shows an interaction for which $H^* = 0.81$. The collision occurs at a higher location than C1 (nick case). The evolution of the ball position and velocity is very different. The evolution of position and speed, for times soon after $t = t_1$ occurs in a manner similar to Case C1: the ball compresses and rolls on the vertical wall. Since the vertical location is higher, the ball is capable of completing its rolling motion with respect to point A by the time it touches the floor at time t_2 . Hence, the ball is free to continue its vertical motion downwards, starting a compression phase with the floor. During this compression phase with the floor, the ball is not longer in contact with the wall on point A . Therefore, the motion of the center of mass of the ball has a negative velocity even for times longer than t_2 even as the ball is already moving away from the wall in the

horizontal direction, as seen on Fig. 1(d). Subsequently, the ball bounces off from the floor with a certain angle, with both horizontal and vertical momentum.

Testing several collision conditions A vast experimental campaign was conducted, including three balls, two temperatures, two incident angles, for a range of impact speeds and initial contact locations. Furthermore, for one test we added a small amount of oil to the wall to lubricate the contact to induce sliding on the wall upon contact. The nick conditions were identified to occur for $0.6 < H^* < 0.75$ and $\tau < 1$. All experiments conducted in this investigation satisfied these conditions, as shown, as shown in Fig. 2 of the main document.

High speed collisions An interesting group of data is that for $\tau < 1$ but $H^* > 0.75$ (data on lower right hand side of Fig. 2(b)). One would expect the condition of $\tau < 1$ to be sufficient since it quantifies the ratio of rolling to contact time.

It is not possible for a ball to reach the horizontal wall for $H^* > 0.78$, if one considers that the maximum possible stretching on the wall upon impact (see above). However, the calculation from Eqn. 2, from the main text, results in values with $\tau < 1$. We note that these experiments have small values of the Cauchy number ($Ca < 0.16$) which correspond for high impact speeds ($U_0 \approx 45$ m/s). For such a range of speeds, the deformation of the ball is significant. We do not expect of the calculation of the contact and rolling times to be valid in such a case.

Experiments at different temperatures For the experiments at $\theta_0 = 30^\circ$, tests were conducted with the same three balls but at a higher temperature (40°C), to replicate the conditions observed in a real squash game. At higher temperatures the elastic properties of the ball change (see Table 1 of the main document), leading to an increase in the coefficient of restitution [209]. The material becomes softer at higher temperatures (the value of E decreases). The results for these tests are shown in Fig. 2(b) of the main text by the red symbols. These data also satisfy the nick occurrence conditions. Since, in general, E decreases with temperature, the attainable values of Ca are smaller.

Collisions over lubricated walls An additional test was conducted for the experiments at $\theta_0 = 25^\circ$ to further test our rolling hypothesis. For a set of experiments involving the double-dot and doubles balls, at

room temperature, the vertical wall was covered by a thin film of viscous fluid (glycerol) with the intention to lower the surface friction. It is well known that when the wall is lubricated by a viscous fluid, sliding (instead of rolling) occurs upon contact[210]. The results from these tests are shown by the green symbols in Fig. 2(a) of the main text. The nick shot was prevented from occurring when the collision corresponded to a value of $\tau < 1$. Upon closer inspection of the high speed videos, for these collisions the ball slid upon contact; hence, the rolling frustration mechanisms did not occur.

B.7 Effect of initial spin

Experiments were not conducted with initial rotation. However, this effect is often present while playing squash. Our estimation, obtained by watching videos from the Professional Squash Association, is that the ball rotation never exceeds 100 s^{-1} during normal playing.

Tangential recoil A ball with backspin could bounce with an angle different from the incident one. Considering both normal and tangential coefficients of restitution and the conservation of angular momentum during an oblique collision with spin, according to Cross [211]:

$$\frac{U_y^{\text{after}}}{U_y^{\text{before}}} = \frac{1 - 4\kappa e_t}{1 + 4\kappa} + \frac{2\kappa(1 + e_t)}{1 + 4\kappa} \left(\frac{D\omega_0}{U_0 \sin \theta_0} \right) \quad (\text{B.14})$$

where U_y^{after} and U_y^{before} are the vertical speeds after and before the collision, $\kappa = 0.1346$ (see calculation above) and e_t is the tangential coefficient of restitution, defined as $e_t = (U_y^{\text{after}} - \omega^{\text{after}} D/2) / (U_y^{\text{before}} - \omega^{\text{before}} D/2)$. From Eqn. (12), the condition required for a rebound with zero vertical speed is:

$$\frac{\omega_0 D}{U_0} = \frac{(4\kappa e_t - 1) \sin \theta_0}{2\kappa(1 + e_t)}. \quad (\text{B.15})$$

Considering, for instance $\theta_0 = 25^\circ$ (as in Fig. 2(a) of the main text), and $e_t = 0.8$ (typical value), leads to

$$\omega^* = \frac{\omega_0 D}{U_0} = -0.49.$$

For ball speeds in a squash game, this condition corresponds to a rotation rate of around $-500 \text{ s}^{-1} = -30000 \text{ RPM}$ (backspin). This value of backspin is not attainable in regular play. Therefore, a collision with backspin cannot explain the occurrence of the nick shot.

Model predictions with initial spin Our model does account for ball spin. The term $2\kappa\omega^*$ in Eqn. (2) of the main text can be varied to analyze the effect of spin. Our observations (from PSA videos), indicate that ω_0 is never larger than 100 s^{-1} , with either top or backspin. Fig. 2 (main text) shows two lines that include the effect of top (dashed-dotted) and backspin (dashed) for $\omega_0 = 100 \text{ s}^{-1}$. When spin is considered, the rotation time is either longer or shorter (top or backspin, respectively); therefore, the condition for $\tau = 1$ is displaced to higher or smaller values of Ca for top and backspin, respectively. In other words, the nick shot is possible but the range of speeds and ball elasticities are slightly different.

B.8 Collision videos

Videos for typical ball collisions have been placed on a data repository. All videos were recorded at 5000 fps, with a shuttle speed of 1/20000, using a Photron FASTCAM SA5 camera.

Movie S1. Case C1: Nick collision. [HERE](#)

Conditions: $\text{Ca} = 0.25$, $H^* = 0.62$, $\theta_0 = 24.43^\circ$ and $\tau = 0.87$.

Movie S2. Case C2: No nick collision. [HERE](#)

Conditions: $\text{Ca} = 0.24$, $H^* = 0.51$, $\theta_0 = 24.34^\circ$, and $\tau = 0.76$.

Movie S3. Case C3: No nick collision. [HERE](#)

Conditions: $\text{Ca} = 0.25$, $H^* = 0.81$, $\theta_0 = 24.31^\circ$, and $\tau = 1.10$.

C MAKE A PANCAKE: LEARN ABOUT VISCOSITY

Abstract One of the most important fluid-mechanical concepts in an introductory class is the fluid viscosity. While other properties are more intuitive, such as density or surface tension, dynamic viscosity is hard to assimilate: its units do not offer an immediate physical measure of its meaning and its formal definition is more involved. In an effort to help teaching this concept, we propose a new method. Instead of the classical sedimenting-sphere experiment, we ask the students to “make pancakes”: pour a known volume of a viscous fluid onto a horizontal surface to observe how it spreads. Using a phone to video-record the experiment, the time rate of change of the circular fluid blob radius can be determined with standard image processing techniques. By fitting the data to a viscous gravity current model, the value of dynamic viscosity can be inferred in viscous dominated flows, neglecting surface tension effects. We show examples using pure glycerin and mixtures of corn syrup and water to measure the fluid viscosity with notable accuracy and repeatability. This simple flow, surprisingly, has not been widely used as a teaching tool to date.

Information This work is to be submitted to *Physical Review Physics Education Research* [8]. The authors acknowledge the undergraduate students, Ajay Harishankar Kumar, Advay Mansingka, Amick Sollenberger, and Portia Tieze of the Fall 2020 course ENGN 0810 at the School of Engineering, Brown University who actively participated in designing the laboratory work (for the COVID period) which became the basis for this paper. Special thanks to Daniel M. Harris for helpful discussions.

C.1 Introduction

When teaching undergraduate physics and engineering students about fluid mechanics, the concept of fluid viscosity is usually taught using a falling ball experiment. First conceived by Fritz H  ppler [212], the falling ball viscometer determines the viscosity of a Newtonian fluid from the terminal velocity of the sedimenting sphere. When dropped in the fluid, the sphere initially accelerates to reach a steady state after a short time. At a steady state, the viscous drag and the buoyancy force of the sphere are balanced by the gravitational

force. With the density, ρ_s , and diameter, d_s , of the sphere known, the dynamic viscosity of the fluid, μ , can be determined from Stokes' law,

$$\mu = \frac{d_s^2(\rho_s - \rho)g}{18 u_s}, \quad (\text{C.1})$$

where ρ is the fluid density, u_s is the terminal velocity of the sphere and g is the acceleration due to gravity. This falling sphere determination of dynamic viscosity is accurate only for the viscous-dominated flows for which Stokes' law applies. Since then, there have been improvements done to the falling ball experiments to obtain more accurate measurements directed specifically towards the undergraduate students [213]. For example, using electrical circuits [214] and video recordings [215] to determine the precise velocity of the falling ball in these experiments. Further, it is well known that these falling ball experiments can also be used in determining the viscosity of non-Newtonian fluids [216]. Recently, miniature ball drop experiments are designed to measure viscosity of non-Newtonian fluids at moderate shear rates [217]. However, in all these experiments the students do not see the fluid moving around the sphere.

Students often confuse dynamic viscosity with density and erroneously refer to a fluid as dense, when it is actually viscous. When introducing the subject, students are often told that the dynamic viscosity is the constant of proportionality in between the shear stress and velocity gradient for a fluid placed in between a moving plate and a fixed plate as shown in the Fig. C.1.

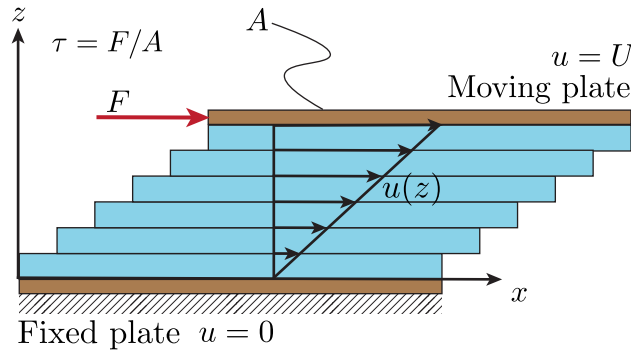


Figure C.1: Fluid in shear between two parallel plates

This relation, referred to as Newton's law of viscosity, is given by,

$$\tau = \mu \frac{\partial u}{\partial z}, \quad (\text{C.2})$$

where τ is the shear stress, which is force, F over the area, A , and $\frac{\partial u}{\partial z}$ is the velocity gradient along the z direction. Hence, the units of viscosity are:

$$[\mu] = \left[\frac{M}{LT} \right] = \frac{kg}{m.s} = \frac{N.s}{m^2} = Pa.s, \quad (C.3)$$

which, again, do not offer much physical insight and nor it gives an indication that the viscosity is a measure of resistance to flow. On the other hand the kinematic viscosity, defined as

$$\nu = \frac{\mu}{\rho}, \quad (C.4)$$

where ρ is the density, can be readily interpreted physically as a momentum diffusivity, analogous to thermal diffusivity. This paper is focused solely on dynamic viscosity, μ .

There are other methods that can be used for teaching purposes. A commonly used viscometer, an instrument which is used to measure the viscosity of a fluid, in undergraduate laboratory is an Ostwald viscometer. It consists of a U shaped glass tube to reservoir bulbs separated by a capillary tube. The unknown viscosity of the fluid with density, ρ_1 , is obtained by measuring the time, t_1 , it takes for the fluid to flow between two marked points on the viscometer. Repeating the same measurement, now with a reference fluid with known viscosity, μ_0 and density, ρ_0 , at the same temperature, the unknown viscosity of the fluid, μ_1 , can be determined as

$$\mu_1 = \mu_0 \frac{\rho_1 t_1}{\rho_0 t_0}. \quad (C.5)$$

One of the drawbacks of this measurement is that this method works only for relatively low-viscosity fluids. More importantly, viscosity and density of a reference fluid should be known to begin with. Commercially available viscometer overcomes these drawbacks by determining the dynamic viscosity of the fluid by measuring the torque resulting from the viscous drag of the fluid flow between two plates or a bob and cone arrangement. However, this sophistication comes with a big price. And again, the measurement is obtained “blindly”: the students do not see viscosity opposing the flow. Other methods consider a weight-controlled capillary viscometer which determines the viscosity of a fluid draining through the capillary tube with the help of a force sensor [218] and a capillary based paper sensor for measuring the viscosity of blood

plasma using a smartphone app [219]. Delvert et al. [220] reported a simple yet elegant experimental setup to measure viscosity of air and other gases at different temperature or relative humidity.

This paper proposes an alternative method to measure dynamic viscosity by observing a flow. Further, the experiments were designed to provide a hands-on experience for the students. The experiments are based on the original study of lava extrusion spreading on the crater floor of the Soufrière of St. Vincent (West Indies) in 1979. The lava dome spread horizontally for five months and reached a ‘pancake’ shape with a height of 130 m and a mean diameter of 870 m. With the help of the recorded data and theoretical analysis, Huppert et al. [221] determined the effective viscosity of the lava to be 2×10^{11} Pa.s.

Section C.2 describes the theoretical analysis of the spreading of viscous drops on a horizontal surface. In Section C.3, the procedure to make ‘pancakes’ and record them as they radially spread are detailed. In Section C.4, we show that the dynamic viscosity of fluids determined using this theory matches excellently with those measured from the rheometer. We finish with a few concluding remarks in Section C.5.

C.2 Theory

When a blob of a viscous fluid is placed or poured on a flat surface, the fluid spreads radially under the action of its own weight. This is referred to as a viscous gravity current, which propagates from the balance of the weight of the fluid and the viscous force. Huppert [222] solved the problem analytically considering a viscous dominated flow (small Reynolds number, $Re \ll 1$), neglecting surface tension (large Bond number, $Bo \gg 1$) and wettability effects. The Reynolds number, which compares the inertial to viscous forces, is defined as

$$Re = \frac{\rho U H}{\mu}, \quad (C.6)$$

where U and H are the characteristic velocity and height, respectively. The Bond number, which compares the gravitational to surface tension forces, is defined as

$$Bo = \frac{\rho g L^2}{\sigma}, \quad (C.7)$$

where L is the horizontal extent of the gravity current and σ is the surface tension. Wettability of the surface on which the fluid spreads can affect the surface tension; thereby affecting the measurements. For the theory to accurately predict the viscosity, a wetting surface is necessary.

By solving the mass and momentum conservation equations, as described in the Appendix, the radius of the viscous blob, $R(t)$, can be obtained as

$$R(t) = 0.779 \left(\frac{g\rho V^3 t}{\mu} \right)^{1/8}, \quad (\text{C.8})$$

where t is the time and V is the fluid volume. If V is known and R is measured in time, the fluid viscosity can be inferred from the Equation C.8, for given values of g and ρ .

We note that this theory of determining the fluid viscosity through observing the spreading drop is not limited to viscous Newtonian fluids and can be extended to non-Newtonian fluids as reported in [223, 224].

C.2.1 Details of the derivation

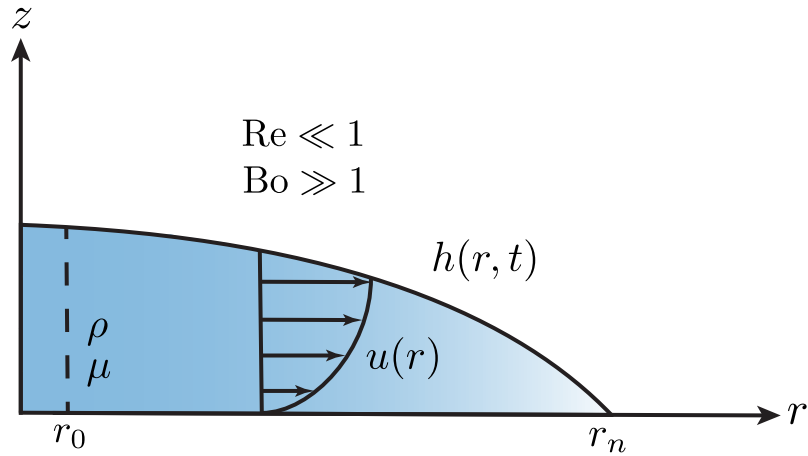


Figure C.2: Sketch of the propagating gravity current with a uniform velocity profile for $\text{Re} \ll 1$ and $\text{Bo} \gg 1$.

Consider a blob of fluid with density, ρ , spreading on a flat surface with the coordinate system as shown in the Fig. C.2. Since the vertical velocity is negligible ($\text{Re} \ll 1$), the hydrostatic pressure, P , in the fluid is

$$P = P_0 + \rho g(h - z), \quad (\text{C.9})$$

where P_0 is the atmospheric pressure at the surface, h is the thickness of the current. The equation of motion of the fluid is,

$$\frac{1}{\rho} \frac{\partial P}{\partial r} = g \frac{\partial h}{\partial r} = \frac{\mu}{\rho} \frac{\partial^2 u}{\partial z^2}, \quad (\text{C.10})$$

where $u(r, z, t)$ is the radial velocity and r is the radius of the fluid blob. The derivative of u with respect to the radial direction is neglected assuming that the thickness of the fluid blob is much less than the radius of the fluid blob (lubrication theory). Solving the Equation C.10 using the no-slip boundary condition at the floor and a shear stress free top surface of the fluid blob, the velocity profile can be obtained as,

$$u(r, t) = \frac{1}{2} \frac{\rho g}{\mu} \frac{\partial h}{\partial r} z(2h - z). \quad (\text{C.11})$$

Note, since the $\text{Re} \ll 1$, the velocity profile of the current is parabolic as shown in the Fig. C.2. The governing equation of motion for the free surface of a viscous gravity current spreading radially is given by the kinematic condition,

$$\frac{\partial h}{\partial t} + \frac{1}{r} \frac{\partial}{\partial r} \left(r \int_0^h u dz \right) = 0. \quad (\text{C.12})$$

Substituting the velocity profile from Equation C.11, the governing equation becomes,

$$\frac{\partial h}{\partial t} - \frac{\rho g}{3\mu r} \frac{\partial}{\partial r} \left(r h^3 \frac{\partial h}{\partial r} \right) = 0. \quad (\text{C.13})$$

From mass conservation,

$$\int_0^{R(t)} 2\pi r h(r, t) dr = V t^\alpha, \quad (\text{C.14})$$

where V is either a fixed fluid volume (for which $\alpha = 0$) or a constant flow rate (for which $\alpha = 1$) [225].

The solution to the Huppert's model will be presented here for a fixed volume case. Writing the Equation C.13 in a non-dimensional form, we get

$$\frac{\partial \tilde{h}}{\partial \tilde{t}} - \frac{\rho g h^{*2}}{3\mu u} \frac{1}{\tilde{r}} \frac{\partial}{\partial \tilde{r}} \left(\tilde{r} \tilde{h}^3 \frac{\partial \tilde{h}}{\partial \tilde{r}} \right) = 0, \quad (\text{C.15})$$

where $\tilde{h} = h/h^*$, $\tilde{r} = \epsilon r/h^*$ and $\tilde{t} = t\epsilon u/h^*$. Here, h^* is the initial height of the fluid blob at time, $t = 0$. ϵ is the ratio of radial spread to height of the fluid blob. With the volume and the radius known, the height can be determined by approximating the fluid blob to be a cylinder.

For the Equation C.15 to be non-dimensional, the ratio of radial spread to height of the fluid blob should be

$$\epsilon = \frac{3\mu u}{\rho g h^{*2}}. \quad (\text{C.16})$$

Thus the non-dimensional equation is given by

$$\frac{\partial \tilde{h}}{\partial \tilde{t}} - \frac{1}{\tilde{r}} \frac{\partial}{\partial \tilde{r}} \left(\tilde{r} \tilde{h}^3 \frac{\partial \tilde{h}}{\partial \tilde{r}} \right) = 0. \quad (\text{C.17})$$

Now, we can introduce a similarity solution such that, $\tilde{h} = \tilde{t}^a f(\xi)$, where $\xi = \tilde{r}/\tilde{t}^b$. Plugging the similarity solution in Equation C.17, we get

$$\tilde{t}^{a-1} (af - b\xi f') = \tilde{t}^{4a-2b} \frac{1}{\xi} (f' f^3 \xi)'. \quad (\text{C.18})$$

Here, the ' corresponds to the partial derivative with respect to ξ . Equating the powers of t on both sides for dimensional homogeneity leads to

$$3a = 2b - 1. \quad (\text{C.19})$$

In non-dimensional form, Equation C.14 for the constant volume case becomes

$$V = 2\pi \int_0^{\epsilon R/h^*} \frac{h^{*3}}{\epsilon^2} \tilde{r} \tilde{h} d\tilde{r}. \quad (\text{C.20})$$

Plugging the similarity solution, $\tilde{h} = \tilde{t}^a f(\xi)$, we get

$$V = 2\pi \int_0^c \tilde{t}^{a+2b} f \xi d\xi. \quad (\text{C.21})$$

Equating the powers of t on both sides for dimensional homogeneity, we have

$$a + 2b = 0. \quad (\text{C.22})$$

Solving for a and b from Equation C.19 and C.22, we get, $a = \frac{-1}{4}$ and $b = \frac{1}{8}$. Plugging this back into the Equation C.18,

$$\frac{f}{4} + \frac{\xi}{8}f' + \frac{1}{\xi}(f'f^3\xi)' = 0. \quad (\text{C.23})$$

Integrating Equation C.23, we have

$$f^3 = \frac{-3\xi^2}{16} + C, \quad (\text{C.24})$$

where C is some constant. The similarity solution,

$$\tilde{h} = \tilde{t}^{-1/4} \left(C - \frac{3}{16} \frac{\tilde{r}^2}{\tilde{t}^{1/4}} \right)^{1/3}. \quad (\text{C.25})$$

Note, $h = h^*\tilde{h} = 0$, when $r = R$, thus we can determine the constant,

$$C = \frac{3}{16} \frac{\epsilon^2 R^2}{h^{*2} \tilde{t}^{1/4}}. \quad (\text{C.26})$$

Substituting the Equations C.26 and C.25 in Equation C.20 and solving for R , the spreading radius can be obtained

$$R_s = \left(\frac{2^{10}}{3^5 \pi^3} \right)^{1/8} \left(\frac{g \rho V^3 t}{\mu} \right)^{1/8}, \quad (\text{C.27})$$

with the constants simplifying to 0.779.

C.3 Materials and Methods

Figure C.3 illustrates the schematic of the experimental setup used to determine the viscosity of the fluid. A known initial volume, V (10 ml, 25 ml, or 50 ml), of a viscous fluid was swiftly poured onto a flat surface. The

pouring time and the spreading time was recorded using a stopwatch. A smartphone positioned directly above the flat surface recorded the radial pancake-spreading of the fluid until the spreading slows down. A checker square board measuring $4\text{ mm} \times 4\text{ mm}$ was used to calibrate the recorded video. In the case of highly viscous fluid, the small volume of fluid adhering to the measuring cup is subtracted from the initial known volume. The experiment was repeated with various initial volumes of the fluids to ensure measurement accuracy. The recorded video was analyzed using simple image processing techniques in the MATLAB (refer to the Supplemental Material) to determine the increase in the fluid front's radius. With the radius of propagation of the fluid front known, the viscosity of the fluid can be theoretically predicted using Equation C.8. This method of determining the viscosity is of value to the beginning undergraduate students, whereas the detailed theory associated can be appreciated by more experienced students with a background in fluid mechanics and mathematics.

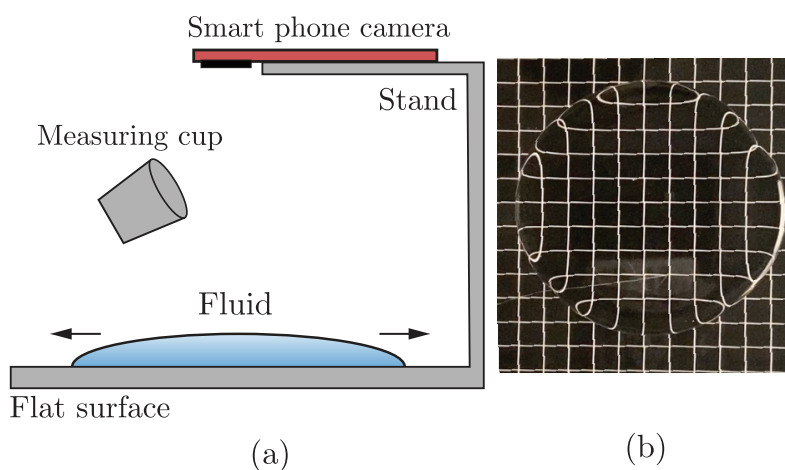


Figure C.3: (a) Experimental setup; (b) Sample image taken from the experiments.

The fluid viscosity determined from the experiments was then compared with that of the values measured with a rheometer (*ARES-G2 Rheometer, TA instruments*) using a cone-plate geometry (40 mm, 0.04 rad cone plate angle with a gap of $24\text{ }\mu\text{m}$). The surface tension of the fluids was measured with a drop shape analyzer (*KRUSS Scientific Instruments*). If interested, students can determine the surface tension of the fluid using a smartphone [226]. The physical properties of the fluids used here are summarized in Table C.1.

Fluids	ρ (kg/m ³)	σ (mN/m)	Re	Bo	μ_m (Pa.s)	μ_r (Pa.s)
Pure glycerin	1230	63.44	0.0448	4100.92	1±0.1	0.99
Corn syrup (dilute)	1347	56.57	0.0213	3339.20	0.5±0.2	0.60
Corn syrup (viscous)	1610	53.76	0.000675	919.54	20±0.3	20

Table C.1: ρ - density; σ - surface tension; Re - Reynolds number; Bo - Bond number; μ_m - measured viscosity from pancake-spreading experiments; μ_r - viscosity from the rheometer measurement. The ‘dilute’ and ‘viscous’ correspond to 70% and 90% (by weight) of corn syrup in water mixture, respectively.

C.4 Results and discussion

When the fluid with known volume is placed onto a horizontal surface, the fluid spreads radially due to gravity. However, the viscous forces resist the flow. It is important to understand that this resistance is due to the internal friction within the fluid. By balancing the buoyancy and viscous forces, viscosity can be determined as discussed in the Section C.2. The spreading of the fluid has two regimes; (1) during pouring and (2) spreading after the pouring is stopped. Thus, the Equation C.8 can be split into two parts,

$$R(t) = R_p + R_s, \quad (\text{C.28})$$

where

$$R_s = 0.779 \left(\frac{g\rho V^3 t}{\mu_m} \right)^{1/8} \quad \text{for } t > t_p. \quad (\text{C.29})$$

Here, R_p is the final radius of the fluid after it is poured onto a horizontal surface, R_s is the spreading radius of the fluid due to its own weight, and t_p is the pouring time of the fluid.

By fitting the experimental data to the Equation C.29, the viscosity of the fluid can be determined. Figure C.4 shows the radial spreading of pure glycerin as a function of time, shifted by the pouring time, for different initial volumes. The solid lines represent the spreading radius, R_s , predicted from the theory by adjusting the viscosity value such that an agreement with the experimental data is achieved. For simplicity, only the results of the pure glycerin are presented here. The viscosity of the fluids determined from the experiments are compared with the viscosity determined from the rheometer in the Table C.1. By repeating

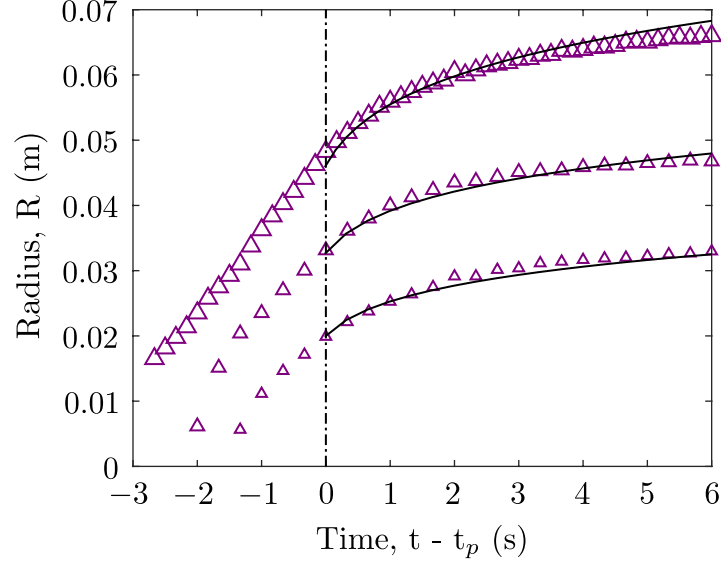


Figure C.4: Propagation of the fluid front as the fluid is poured onto a horizontal surface. The origin for time is shifted by the pouring time. The size of the purple triangles corresponds to the initial volumes: 10 ml, 25 ml, and 50 ml of pure glycerin, respectively. The solid lines represent the theoretical R_s determined by adjusting the viscosity value.

the experiments multiple time with different initial volumes, the accuracy of the measurement can be improved.

By normalizing the radius and time as follows,

$$R^* = \frac{R}{V^{1/3}}, \text{ and} \quad (C.30)$$

$$T^* = \frac{(t - t_p)\rho g V^{1/3}}{\mu},$$

the different initial volumes of various fluids collapse into a single curve as shown in Fig. C.5. When the fluid is propagating due to the own weight, the radius of the fluid front follows the scaling law (solid line in the Fig. C.5),

$$R(t) \sim t^{1/8}, \quad (C.31)$$

which is in accordance to the prediction of Equation C.29.

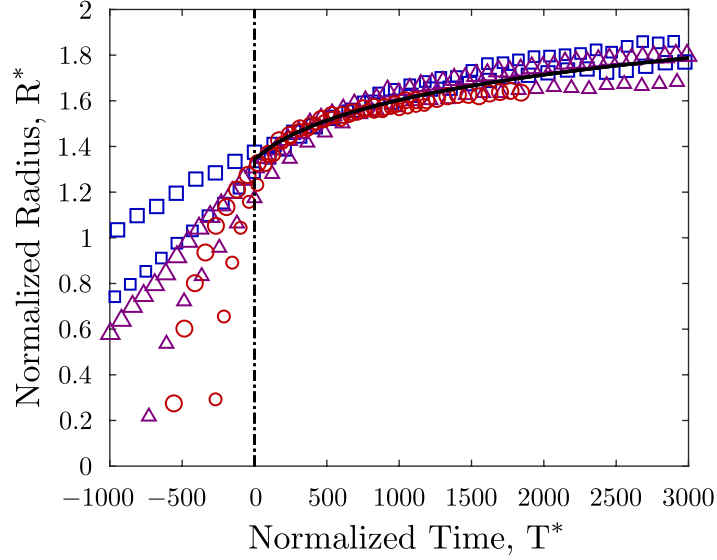


Figure C.5: Normalized radius of the fluid front as a function of normalized time. The red, purple, blue colors correspond to the increasing viscosity of the fluids considered and the increasing size of the markers correspond to 25 ml and 50 ml of the respective fluids. The solid line represents the normalized theoretical R_s determined by adjusting the viscosity value.

C.5 Conclusions

Here, we proposed a simple method to determine the viscosity by observing the speed of a viscous volume spreading over a horizontal surface. The propagation of the fluid front due to the balance between the buoyancy and viscous forces was used to predict the viscosity by fitting the experimental data to the Huppert's model [222]. From this simple 'pancake' experiments, the students can visually grasp that viscosity is a property that offers a resistance to flow.