

Droplets 2019

Book of Abstracts

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DROP IMPACTS: HIGH-SPEED IMAGING OF SPLASHING AND AIR ENTRAPMENT

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This seminar will describe experiments on two aspects of drop impacts on liquid surfaces. First, the splashing of secondary droplets into the air and secondly the entrainment of fine bubbles into the pool. Both aspects are important in many industrial phenomena and even in nature. Only recently have experimental techniques and numerical algorithms been able to resolve the full range of length and time-scales involved during the impact. Experiments using state-of-the-art high-speed video cameras, will be presented at video frame-rates as high as 5 million fps. This is used to identify the origins of the finest splashed droplets, as well as the multitude of air-bubbles trapped between the drop and the pool surface. In particular we probe the dynamics of the ejecta sheet and the sub-micron air-films separating the drop and pool at low impact Weber numbers [1]. Furthermore, close-up imaging reveals how the axisymmetry can be broken on the finest scales [2]. We will also use numerical simulations to show the vortical structures which occur inside the liquid [3]. Finally, new results will be shown for high impact velocities at over 20 m/s, generated by drops free-falling in a 25-m-long vacuum tube. This includes splashing at 1 km/s and convoluted ejecta shapes.

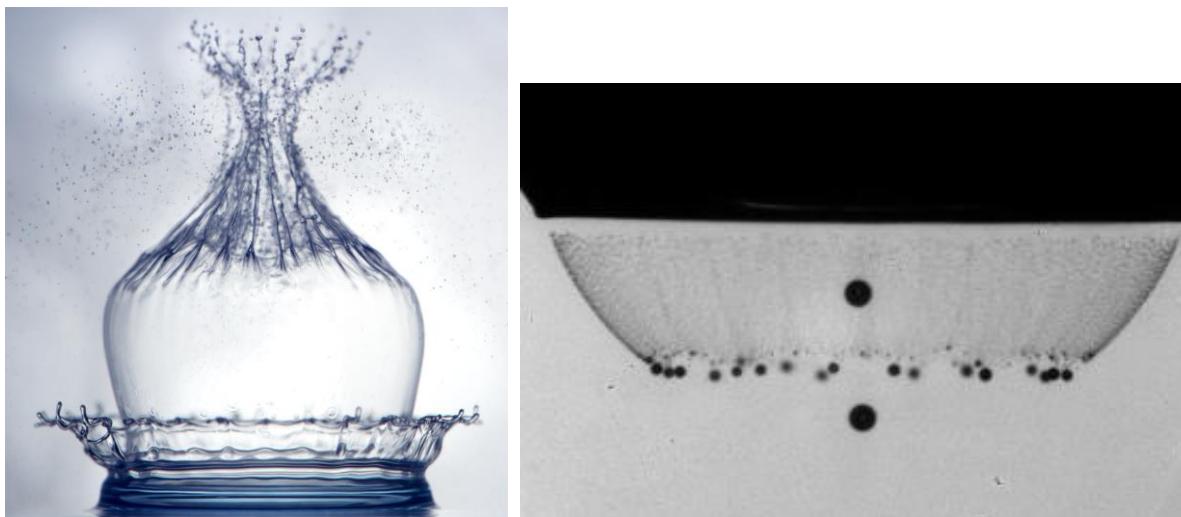


Figure 1: Splashing and air entrapment during drop impacts. Left image courtesy Aljedaani & Langley.

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Photoemission from charged droplets

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Photoelectron velocity-map-imaging (VMI) studies on submicrometer-sized droplets have gained increasing importance for elucidating the properties of low-energy electrons in the condensed phase. An often neglected aspect of droplet VMI is the droplet charge. The presence of an additional electrostatic potential can modify electron transport and lead to a Coulomb barrier to electron escape. To explore and characterize the effects of positive and negative charges on low-energy electron transport we have performed a combined experimental and modeling study. To this end, photoelectron VMI spectra of size-selected droplets were recorded while varying the charge distribution in a controlled way. A probabilistic scattering model has been extended to include both classical and quantum effects of the electrostatic potential on the electron escape process. Good agreement between the experimental results and the extended scattering model is obtained.

DROPLET GENERATION VIA BURSTING, IMPACTING, AND JETTING, WITH SURFACTANTS

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Liquid atomisation processes are widely used to break down a liquid stream into smaller droplets to enhance its mixing with a stagnant phase. These streams may be contaminated with surfactants, whose concentration variations lead to surface tension gradients and Marangoni stresses. Here, we study the effect of surfactant on the complex interfacial dynamics associated with a turbulent jet. We use three-dimensional (3D) direct numerical simulations (DNS) and a hybrid front-tracking/level-set method¹ to capture the dynamics of the complex topological changes in this flow. The numerical method allows the natural tracking of the concentration of interfacial surfactant species and the faithful modelling of its spatio-temporal evolution. Our model also accounts for surfactant solubility and bulk-interface mass exchange. We perform a parametric study of the effect of surfactant properties on the dynamics. The effect of Marangoni stresses is analysed in terms of the mechanisms giving rise to the droplet size distributions depending on the elasticity number. An attempt to understand the interaction between the observed vortical structures accompanying the flow and the regions of elevated surfactant concentration will also be presented.

Countless instances of bursting bubbles over the oceans contribute significantly to the exchange of heat and chemicals with the atmosphere. Understanding bursting bubble physics could be the key to tackling the ever-growing environmental problem. When a bubble is close to a free surface, it forms a hole which leaves an open unstable cavity that undergoes collapse; the change of the interface curvature leads to the formation of a central jet, which breaks into droplets according to the Plateau-Rayleigh instability. The surfactant-free interfacial dynamics are well understood, however, the surfactant-laden bursts are still unexplored. By neglecting gravity, the Laplace number is the only dimensionless control parameter measuring the relative importance of surface tension to viscous forces i.e. $La = \rho\sigma R/\mu^2$, where ρ , μ , σ , and R represent the liquid density, viscosity, surface tension, and the initial radius of the droplet, respectively. The fate of the central jet is analysed with help of a 3D DNS simulations, where the effect number of surfactant related non-dimensional parameter. Results regarding the importance of Marangoni stresses on the jet formation will be discussed.

Finally, we consider the impact of drops on solid and fluid substrates whose rich phenomena have been the source of fascination for decades. Recent experimental work² has investigated the effect of surfactants on “crown” splashing and found that they affect significantly the propagation of capillary waves, the evolution of the crown, and the formation of secondary droplets. Here, we employ 3D DNS to examine drop impacts on thin surfactant-laden films. We couple the hybrid interface-tracking/level-set method for the interfacial dynamics to a convective-diffusion equation for the surfactant concentration to carry out the computations. We vary different surfactant properties (i.e. diffusion, elasticity, and solubility) to study their effect on the phenomena accompanying the drop impact.

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Segregation in multicomponent droplet evaporation

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While the evaporation of a single component droplet meanwhile is pretty well understood, the richness of phenomena in multicomponent droplet evaporation keeps surprising us. In this talk we will show and explain several of such phenomena, namely evaporation-triggered segregation thanks to either weak solutal Marangoni flow or thanks to gravitational effects, and the evaporation of ternary liquid droplet, which can lead to spontaneous nucleation of droplets consisting of a new phase. We will also show how this new phase can be utilized to self-lubricate the droplet in order to suppress the coffee stain effects. The research work shown in this talks combines experiments, numerical simulations, and theory.

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Elastic Ripening and Inhibition of Condensation of Droplets in Gels

Eric R. Dufresne, ETHZ

Inspired by pressing questions in cell biology, we consider the condensation of liquid droplets in polymer networks. We find that bulk elastic stresses dramatically affect every stage in the life of a droplet, from nucleation and growth to ripening and dissolution.

Cavitation for Emulsions

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Liquid – liquid emulsions are widely used in major chemical industries like pharmaceutical, agricultural, cosmetics, paint and food. Emulsions are realised by introducing energy into the desired liquid – liquid systems. Various devices stirrers, rotor-stator systems, and high pressure homogenizers are used for emulsification. Efforts are however being made continuously to develop better ways of realising emulsions (better ways of providing required energy) and better control on resulting emulsion characteristics. There is an increasing interest in using cavitation as a clean, energy efficient method for the production of highly stable emulsion systems. The cavitation process progresses through the formation, growth and collapse of cavities or microbubbles. The cavity implosion results in localized hot spots with very high temperature, pressure and intense shear. High intensity ultrasound, or acoustic cavitation (AC) techniques demonstrate excellent emulsification performance in comparison to more traditional mechanical methods, such as high-speed homogenizers and microfluidizers^{1,2}. A number of examples in open literature have described the successful application of AC to generate highly stable nanoemulsions in water insoluble systems³. Another potentially exciting application area for cavitation assisted emulsification is in the food processing industry, and in particular the dairy industry, where AC has been again found to successfully homogenize particle sizes down to the nano-range⁴. In this talk, we will discuss some of the recent results on using cavitation for emulsions.

Despite the promising results of using AC for emulsions, it should be noted than though AC can be readily used on a laboratory scale, it is difficult to extend their application to commercial scale. Hydrodynamic cavitation (HC) is emerging as an attractive alternative method for the preparation of stable emulsions, offering a range of additional benefits above AC, including; lower energy consumption, continuous flow operation, and potentially easier routes to scale up. HC generated emulsions for food processing applications have been found to achieve comparable droplet size distributions to AC methods⁴. HC systems are commonly based on forcing flow through small restrictions, such as an orifice or venturi, and are thus prone to clogging and contamination in a similar manner to microfluidizers. Recent developments in cavitation reactors based on swirling flows have opened a potential route to realizing the benefits of HC which removes the risk of clogging and erosion⁵. In this work, we present ongoing research on the production of stable oil - water emulsion systems at QUB using both AC and HC methods. In the first part, we will present the results from AC based emulsions to bring out the relative influences of power input, irradiation time and phase concentrations on the final droplet size distributions. The vortex based cavitation devices are then used to compare results from HC based emulsions with those from AC. Characteristics of emulsions prepared with HC will be directly compared with AC methods, highlighting any relative differences in final emulsion properties and total energy consumption. In order to understand the complex multi-phase flow field that develops in vortex based cavitation device, a full 3D, multiphase, transient CFD simulations were carried out. These CFD results were used for quantitatively relating predicted energy dissipation rates with emulsion properties. The presented approach, computational models and results will provide a sound basis for gaining additional insights for enhancing applications of hydrodynamic cavitation to the area of emulsions.

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High-accuracy simulation of free surface flows near finite-time singularities

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Motivated by applications such as ink jet printing, drop-by-drop manufacturing, sprays, and chemical separations, we study the dynamics of breakup and coalescence through high-accuracy simulation, theory, and experiment. In this talk, I will highlight our group's work on accurately capturing the fluid dynamics that takes place in the vicinity of finite-time singularities. The primary focus of the talk will be on simulations of the breakup of surfactant-covered filaments where I will pay special attention to the pinch-off singularity. I will also summarize some of our recent work on the pre- and post-coalescence singularities that arise when two drops or bubbles are driven together and made to merge into one.

Evaporation of nano-suspension drops on soft and structured substrates

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The evaporation of droplets is dictated by the nature of the three phases it is part of i.e. liquid, vapour and solid. The properties of these phases as well as the environmental conditions are known to play a crucial role in the kinetics of evaporation as well as the dynamics of the droplets profile and in particular the three phase contact line. This study investigates the evaporation of sessile pure water and nano-suspension drops on viscoelastic polydimethylsiloxane (PDMS) films. We varied the viscoelasticity of the PDMS films by controlling the curing ratio, and categorised them into three types: stiff (10:1, 20:1, 40:1), soft (60:1, 80:1), and very soft (100:1, 120:1, 140:1, 160:1). The complete evaporation of nano-suspension drops of stiff substrates leads to particle deposition patterns similar to a coffee ring with cracks and deposition tails. On very soft substrates the initial spreading is followed by a mixed mode in which a wetting ridge is pulled up by the vertical component of surface tension at the contact line of the drop. As the evaporation proceeds, the contact line attempts to recede due to the decreasing contact angle, which exerts a horizontal force on the wetting ridge. Due to the viscoelastic nature of the very soft substrates, this horizontal force cannot be countered by the substrates themselves, causing the wetting ridge to move horizontally in a viscous-flow way. The contact line continues to be anchored to the wetting ridge, but recedes relative to the bulk of the substrate, resulting in a mixed mode of evaporation. Complete evaporation of nano-suspension drops on very soft substrates leads to finger-like deposits. Furthermore, we explore the evaporation of nano-suspension drops on micro-structured surfaces. The effect of shape and size of the microstructures is found to significantly alter the base profile of the drops. In some cases, a thin liquid film is found to extend beyond the main drop. The evaporation from this thin film is fed from the drop which acts like a ‘reservoir’ to compensate for evaporation.

Drop impact on superamphiphobic surfaces

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The dynamics of liquid drops impacting superamphiphobic coatings is studied by high speed video microscopy. Superamphiphobic coatings repel water and oils. The coating consists of a fractal-like hydrophobized silica network. Three experimental setups are discussed:

- Impact of mixtures of ethanol-water and glycerin-water drops: The contact time increases with impact velocity, whereas the restitution coefficient decreases. We suggest that the drop temporarily impales the superamphiphobic coating, although the drop completely rebounds.
- Impact of drops on a superamphiphobic mesh: For an impacting water drop, a shower of secondary droplets is produced. Compared to neat steel meshes, superamphiphobically coated meshes produce more monodisperse secondary droplets.
- Impact of an oil drop on an identical sessile drop: We experimentally and numerically investigate the impact and rebound dynamics of an oil drop impacting a sessile oil drop sitting on a superamphiphobic surface as function of velocity and degree of head-on alignment. The simulations quantitatively reproduce all experimentally observed rebound scenarios and allow quantifying the velocity profiles, the energy transfer and the viscous dissipation

Lipid Coated Droplets: From Bubbles to Cells - therapy to diagnostics

Stephen Evans

In the presence of a gas bubble in solution lipids spontaneously assemble at the air/gas boundary presenting a lipid monolayer with the zwitterionic head groups oriented to the aqueous phase. The lipids both stabilize the gas from dissolution and present a biocompatible interface to reduce identification by the immune system. The natural echogenicity of such bubbles combined with their biocompatibility makes them ideal theragnostic agents for ultrasound aided drug delivery. Here we describe our recent approaches for the bubble formation, characterisation and the treatment of cancer.[1]

Microfluidically produced liquid crystal (LC) droplets generate hydrophobic interfaces in aqueous solutions lipids in solution adsorb at these interfaces to produce a lipid monolayer which in turn controls the anchoring of the liquid crystal. Here we describe how interactions with the lipid can lead to transitions in LC alignment and be used to create LC droplet-based sensors for antimicrobial peptides. [2]

Deformability of a droplet/cell in response to mechanical stress depends strongly on the forces experienced by the droplet/cell. Here, we use cell deformability in both shear-dominant and inertia-dominant microfluidic flow regimes to probe different aspects of the droplet and cells. [3]

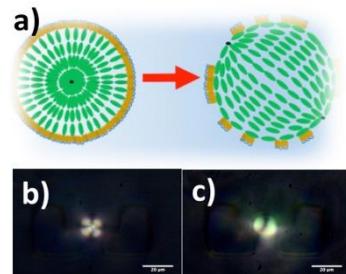


Figure 1 AMP induced transition in LC alignment in LC droplets

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MEASURED VAPOR DISTRIBUTION AND THE DIFFUSIVE, CONVECTIVE, AND VELOCITY FIELDS OF AN EVAPORATING SESSILE METHANOL DROP

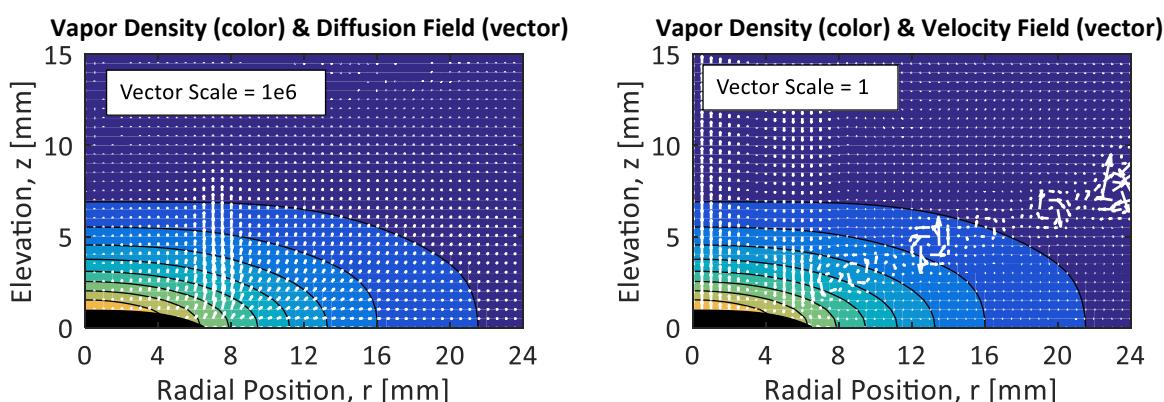
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This paper reports on the vapor transport for an evaporating methanol drop. Methanol was chosen for this study because our evaporation rate measurements were significantly higher than values predicted by the diffusion-limited model, a common model for vapor transport, even though solutal convection is expected to be negligible due to the molecular weight of methanol being approximately equal to that of air. The methanol vapor concentration distribution was measured and then used to compute the diffusion, convection, and velocity fields.

The vapor concentration distribution was measured using infrared spectroscopy and computed tomography, generally following the technique reported in [1]. Since convection alters the spatial distribution of the vapor surrounding the drop, measurements of the vapor distribution provides a qualitative indication of the relative influences of diffusion and convection on the evaporation process. Following the computation of the concentration gradient, from which the diffusive flux field was determined, the evaporation rate was computed according to the technique of Dehaek et al [3]. With knowledge of the evaporation rate and the diffusive flux, mole balance equations for vapor and air were solved for a series of control volumes to determine the convective flux and velocity fields, thereby providing quantitative information about the nature of the vapor transport. The figures below show the measured vapor distribution in a vertical plane with the silhouette of the drop shown in black, and the resulting diffusion field (left) and velocity field (right). As in the case of diffusion-limited evaporation, the diffusive flux is high near the perimeter of the drop. The velocity field indicates a downward flow above the drop perimeter and a series of vortices at larger radii.



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Responding to humidity: Measuring the hygroscopicity of large molecule microdroplets

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Cyclodextrin derivatives, such as 2-hydroxypropyl- β -cyclodextrin, are pharmaceutical excipients thought to help alleviate symptoms of Niemann-Pick disease on inhalation due to improved water solubility and bioavailability over the parent compound. However, the physicochemical properties of cyclodextrins and their derivatives in bulk solution and when aerosolised are ill-defined or unknown. Individual droplets of aqueous HP- β -CD are measured using Raman tweezers to retrieve size and refractive index and determine hygroscopic growth response. Measurements of bulk solution refractive index and density allow prediction of solute properties and super-saturated concentrations of relevance to aerosol. Quantifying physical properties of aerosolised HP- β -CD allows conversion between mass and radius based hygroscopic growth factors and comparison with dynamic vapour sorption measurements and AIOMFAC modelling.

50 YEARS IN SEARCH OF THE DYNAMIC CONTACT ANGLE

T. D. Blake

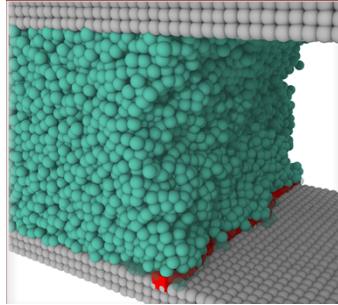
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After introducing some essential basics and a little of the history, the presentation will focus on the molecular-kinetic theory of dynamic wetting (the MKT), first published in 1969 [1,2], and some of the increasing evidence of its relevance to our understanding of how contact lines move across solid surfaces. Much of this evidence will be drawn from molecular dynamics (MD) studies that uniquely illuminate dynamic wetting at the nanoscale.

The MD studies, which have spanned a collaboration of 25 years with Professor Joel De Coninck and his group at the University of Mons, Belgium, have replicated the many of the predictions of the MKT. They have involved simulations of spreading drops, capillary rise, the wetting of fibres, coating and Couette flow with free boundaries, plus investigations of slip and Young's equation. While keeping the properties of the liquid constant, the equilibrium contact angle has been varied from nearly complete wetting to angles similar to those seen in superhydrophobic systems. In each case, the resulting data have agreed with the predictions of the MKT and confirmed expectations such as the dual roles of viscosity and solid–liquid interaction in moderating wetting dynamics, as well as the potential of a localized shear stress to promote it during forced wetting. However, one aspect, the non-linearity predicted by the MKT (and seen in experiments) at high driving forces has yet to be realised in the simulations.

In our most recent work [3], we have investigated the spatial fluctuations of the contact line and the consequent fluctuations in the surface-tension force at the solid-liquid boundary. We have shown that one may use well-established statistical thermodynamics, (the Langevin equation) to predict the *dynamics* of wetting via the MKT by analysing these fluctuations at *equilibrium*. We hope that these studies will open the door to new experimental approaches to discover the private and still hidden life of the dynamic contact angle and the moving contact line.



ACKNOWLEDGEMENTS: J. De Coninck, M. de Ruijter, D. Seveno, E. Bertrand, and J-C. Fernandez-Toledano, without whom much of the progress described above would not have occurred.

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Forming hollow nanoparticle microstructures via double nematic nucleation

Linda S Hirst

Rapid bulk assembly of nanoparticles into microstructures is challenging, but highly desirable for a variety of applications in controlled release, catalysis, and sensing. In recent work our group has developed a new method to form hollow microstructures using a two-stage liquid crystal nucleation process, generating size-tunable closed-cell foams, spherical shells, and tubular networks composed of closely packed nanoparticles. To achieve the structures, mesogen-modified nanoparticles are dispersed in liquid crystal above the nematic-isotropic transition temperature (TNI). On cooling through TNI, nanoparticles first segregate into shrinking isotropic droplets where they locally depress the transition temperature.

On further cooling, secondary nematic domains nucleate inside the nanoparticle-rich isotropic domains, driving the formation of hollow nanoparticle assemblies. We achieve structural differentiation using nanoparticle density and cooling rate. Our methods take advantage of the interaction between particle shape, surface anchoring and liquid crystal elasticity.

Using liquid crystal as a novel anisotropic solvent for nanoparticles opens up a wide range of possibilities for bulk structure templating via phase nucleation and growth.

Atmospheric Droplets: The Role of Interfacial Chemistry in Cloud Droplet Formation and Hygroscopic Growth of Aerosols

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Recent experiments reveal the importance of surface tension depression by interfacial molecules in cloud droplet formation and hygroscopic growth on mixed organic/inorganic aerosols. For realistic mixed inorganic/organic aerosols, the droplet sizes either at activation or at high relative humidity exhibit a more complex relationship than would be predicted based upon bulk solubility alone and points to the important role that surface chemistry plays in droplet growth. I will highlight the main features of this mechanism and discuss current uncertainties in developing a molecular understanding droplet surfaces in the atmosphere.

Why Digital?

Philip Bentley

Digital is generally marketed as the next big thing in printing, a statement bolstered by the extra kudos that comes from the high-tech nature of this technology. This “Silicon Valley” sentiment is also reflected in the teachings of many with vested interests in the technology, that inkjet can print anything. Although this is broadly true if we consider a single drop from a single nozzle, it is quite a different story if we need to print at high speed in production environments. Analogue printing, in comparison, is remarkably tolerant in ink chemistry, a fact that is reflected in the cost of such inks.

The analogue printing vs. digital printing debate is only of interest to those selling such printing machines, the manufacturer simply wants to sell printed product at the largest margins. This presentation looks at whether the analogue or digital printing question is really so cut and dry and discusses in detail a productionised print solution that exemplifies how deeply synergistic the combination of analogue *and* digital can be.

Droplets 2019, Durham UK

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Biextensional viscosity and non stationary elastocapillarity effects in the Impact of viscoelastic drops

When a drop of a viscoelastic fluid hits a solid surface , in minimal dissipations conditions, (achieved using either a small solid target or a repellent surface), it expands radially until reaching a maximum diameter and subsequently recedes. Experiments indicate the presence of two expansion regimes: the capillary regime, where the maximum expansion does not depend on the fluid's zero-shear viscosity, and the viscous regime, where the expansion is reduced with increasing zero-shear viscosity due to viscous dissipation. Two classes of viscoelastic fluids have been investigated: (i) Thinning fluids consisting of solutions of living polymers of various concentration, and (ii) ultra soft self-assembled transient networks, with tunable elastic modulus and relaxation time.

In the viscous regime, we find that the *equibiaxial* viscosity is the appropriate quantity to describe the maximum expansion of both viscoelastic and viscous sheets. For solutions of viscoelastic thinning fluids, shear dissipation is negligible compared to extensional dissipation.

In the capillary regime, the impact experiment allows to evidence a *Deborah-dependent* modulation of the variation of the maximum expansion with impact velocity, that we interpret as a non-stationary elasto-capillarity phenomenon.

SCANNING DROPLET ADHESION MICROSCOPY FOR SURFACE WETTING CHARACTERISATION

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The characterization of wetting properties is usually carried out via measurements of contact angles at the three-phase contact line¹ with contact angle goniometry. Even though contact angle measurement is useful wetting characterization technique, they are prone to substantial inaccuracies, especially for hydrophobic surfaces^{2,3}. Thus, the characterization of such surfaces needs to be more carefully considered³. Here will be addressed the problem and presented a cutting-edge methodology, scanning droplet adhesion microscopy, for evaluating wetting of hydrophobic surfaces with enhanced accuracy based on the adhesion force measurements. The technique allows measurement of small forces with sensitivity down to nanonewton and mapping of wetting properties at microscale spatial resolution⁴ (Figure 1).

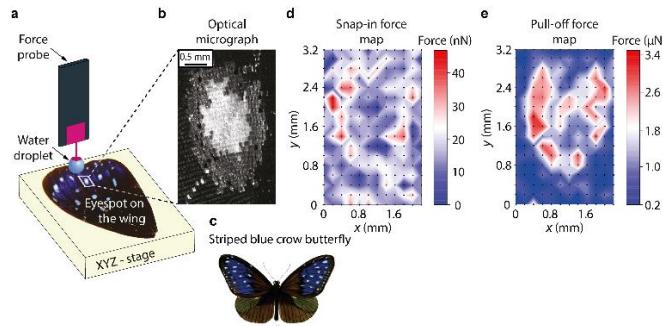


Figure 1. Concept of scanning droplet adhesion microscopy to construct wetting maps showing (a) schematic diagram of the microscope (not to scale), (b) optical micrograph of scanned eyespot area on the wing of (c) striped blue crow butterfly (image by Frederic Moore, PD-1923) with corresponding (d) snap-in and (e) pull-off force maps.

ACKNOWLEDGEMENTS: The authors thank the European Research Council grant ERC-2016-CoG (725513-SuperRepel) and the Academy of Finland Centres of Excellence Programme 2014–2019 for supporting this work.

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DROP IMPACT ONTO A SURFACE COVERED WITH A THIN FILM

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Despite being encountered in various natural and applied cases where a drop impacts a very thin film made of an immiscible liquid (e.g. water impact onto an oil covered surface) there is little experimental or numerical studies available. Experimentally we investigated both the spreading and recoiling phases of the drop impact for such systems. Similar to miscible systems, in the spreading phase of the impact phenomena for when droplet and film are made of immiscible liquids, one observes (see Fig. 1): (i) simple spreading; (ii) crown formation without splashing; and (iii) splashing. However, for a miscible film, a larger impact Weber number (We) and film thickness are needed for the formation of a crown and splashing comparing with immiscible cases. The other difference is that when the film is very viscous (e.g. 100 cSt), the spreading of the drop on an immiscible film is similar to that of drop impact onto a dry surface, unlike the impact onto a miscible viscous film. Contrary to intuition that here will not be a recoiling phase for two miscible liquids, while one expects recoil could be observed for two immiscible liquid, we found that the immiscibility is not a necessary condition for the existence of a recoil phase. The presence of a recoil phase is highly dependent on the value of the interfacial tension between the drop and the film. The larger the interfacial tension difference, the more noticeable the recoiling phase can be. Even a total rebound can be seen for systems with large liquid-liquid interfacial tension values. The occurrence of the total rebound is affected by the substrate wettability, We , film thickness and film viscosity. It was found that the oil film can rupture during the impact process when the substrate has a contact angle $<90^\circ$ with the liquid of the droplet; in such case, the total rebound will be impeded. Instabilities of the lamella (e.g. fingering, crown, and splashing) during the spreading phase also hinder the total rebound. Finally, a total rebound was absent for drop impact onto viscous films (e.g. 100 cSt) due to significant energy desperation during the spreading and recoil phases.

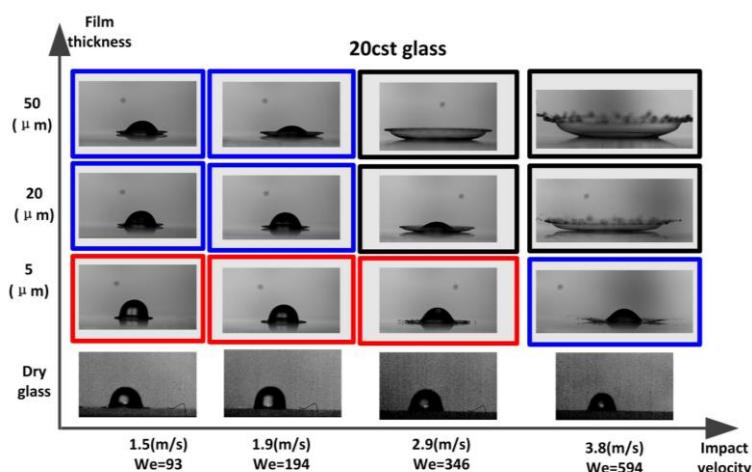


Figure 1 – impact outcome for the spreading phase of water drop impacting a thin oil film of 20 cSt placed on a glass substrate as well as water impact onto a glass surface.

OPTICAL LEVITATION OF LIQUID DROPLETS

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Optical levitation was first demonstrated by Ashkin and Dziedzic [1]. We will here present a versatile experimental system for optical levitation of micron-sized liquid droplets that allows us to control many of the experimental parameters. Two piezo-driven dispensers are used to produce liquid droplets on demand. The charges of the droplets are controlled at the production by applying an electric field on the piezo-dispenser heads and during an experiment by exposing it to ionizing radiation or UV light. The dispensers release droplets into two independently controlled optical traps.

Three dimensional size and position data on droplets in the traps are measured using two orthogonally placed high speed cameras. The electrical charge of the droplets are measured by recording their motion when an electric field is applied. Spectroscopic information about the trapped droplets is obtained by imaging the droplets on the entrance slit of a spectrometer. Finally, the trapping cell can be evacuated, allowing us to investigate droplet dynamics in vacuum.

We present four different experiments. First, we investigate how the rate of droplet coalescence depends upon their charges. Fig. 1 shows the coalescence process between two droplets with a time resolution of 77 μ s. Second, we measure the spectra of two colliding droplets which have been doped with different dyes. The mixing of the dyes is monitored by observing the FRET (Förster Resonance Energy Transfer) signal. Third, we describe the motion of two droplets trapped in the same optical trap. We observe how the droplets move in stable periodic orbits resembling the motion of balls being juggled by a carnival performer. Using a numerical model constructed with Newtonian mechanics and ray optics, we show that the juggling motion arises from particles taking turns eclipsing each other [2]. Finally, we measure the removal of electrons from a trapped droplet at a resolution that is better than a single electron charge, making it to a single-drop Millikan like experiment [3].

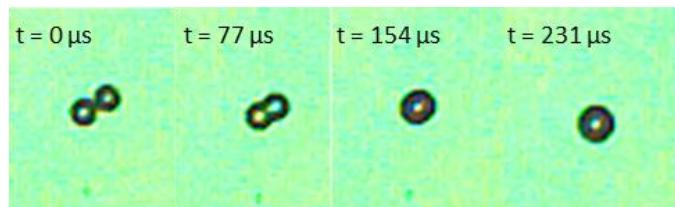


Fig. 1. Coalescence process between two glycerol droplets held in two optical traps when close to each other.

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Droplet growth and collisions due to turbulence and gravity

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This lecture is dedicated to the memory of Jason Reese.

Droplets in a turbulent flow are known to get centrifuged out of vortical regions, and cluster in straining regions. If the flow is accompanied by evaporation and/or condensation, the dynamics may become significantly different. This was shown in two dimensions in a supersaturated ambient [1], where, due to their inertia, solid particles and droplets are rapidly centrifuged out of vortical regions. Due to low availability of condensation nuclei, and hence lower heating from condensation, vortical regions end up slightly colder than straining regions. Interestingly, regions of high strain often lie just outside vortical regions, forming a sleeve around the vortex, and we show that collisions between droplets often occur in such regions, at a rate well in excess [2] of their local number density. We then perform three-dimensional simulations of homogeneous isotropic turbulence in a cubic domain, including inertial droplets and phase change. Initially droplets are distributed homogeneously in a supersaturated environment. We use the second invariant of velocity gradient, Q , to distinguish between straining (negative Q) and vortical (positive Q) regions. In three-dimensional turbulence too, vortical regions are cooler than their surroundings, but the temperature difference is small.

ACKNOWLEDGEMENTS: RG thanks the organisers of Droplets2019 for support to attend this meeting.

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EVAPORATION OF COLLOIDAL DROPS AND FORMATION OF COFFEE RINGS ON POROUS SUBSTRATES

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It is widely known that evaporation of colloidal drops on flat solid substrates may result in the deposition of colloidal particles in the vicinity of the three-phase contact line. This phenomenon is commonly referred to as the coffee ring effect [1]. A similar phenomenon is observed on porous substrates, where the colloidal fluid is initially imbibed into the porous substrate to form a wetted region, and thereafter evaporates. At the end of evaporation process the colloids are preferentially deposited near the periphery of the wetted region [2]. Whereas the mechanism governing the formation of coffee rings on flat substrates is well understood [1], the corresponding effect on porous substrates is still not sufficiently elucidated. A profound understanding of this phenomenon is important for controlling the formation of coffee rings in numerous applications, including inkjet printing, dried spot sampling of biological liquids, and spray coating of paper substrates.

In this work, evaporation of colloidal drops and the formation of coffee rings on porous substrates has been studied experimentally. A dispersion of cationic lipid vesicles was used as a model colloidal liquid. Cellulose fiber filter papers were used as porous substrates. The drops of different sizes were placed on the porous substrate within a glove box chamber with low humidity. A combination of two measurement techniques has been used to obtain insights into the mechanism underlying the coffee ring formation. Evaporation process was observed from the top with a high resolution color camera. The rate of evaporation was simultaneously monitored using an analytical balance. It has been found that the evaporation process took place in two phases: (i) the constant wetted area phase which occurs initially and is accompanied by the formation of coffee rings, and (ii) the phase of fast decrease of the wetted area, which occurs towards the end of evaporation process and is accompanied by a uniform deposition of colloidal particles. The effect of different parameters, such as drop size, colloidal concentration, and substrate porosity, on the size of the coffee rings has been investigated. It has been found that the coffee ring width increases with droplet size for small initial drop volumes and reaches saturation.

ACKNOWLEDGEMENTS: We kindly acknowledge the financial support by the European Commission in the framework of the Marie Curie Initial Training Network “Complex Wetting Phenomena” (CoWet).

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MULTISCALE FLOW ENGINEERING: RE-IMAGINING FLUID DYNAMICS MODELLING

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This keynote talk is a tribute to the late Professor Jason Reese, and describes research in multiscale flow engineering which he pioneered and led.

Imagine sifting salt molecules from seawater, to provide drinking water in countries affected by climate change. Or treating vehicle surfaces to control the air or water flow and reduce fuel costs and carbon dioxide emissions. There are multiple technological areas where leveraging the interactions between the smallest constituents, right down to molecules, would open up exciting opportunities.

Designing these types of multi-scale systems is a grand challenge in science and engineering for the 21st century. A fundamental problem is that current design tools do not account for the physics that determines how these horizon technologies behave. So we cannot cycle through digital prototypes, assessing their performance, to identify the best ones to manufacture.

Our recent advances in engineering analysis in fluid dynamics of non-conventional micro/nanoscales describe methods that can span some 8 orders of magnitude in space, and 10 orders in time - which mean that ideas and designs for new products and processes can start being tested. We are able to use multi-scale innovation to describe new science of water flows in laboratory-scale nanotube membranes [1], thermally-driven rarefied gas flows in microscale pumps with no moving parts [2], thermal capillary waves that drive droplets to coalesce [3], and unravel the limit of superstability of nanobubbles pinned on a surface [4].

ACKNOWLEDGEMENTS: This research is financially supported by the UK's Engineering and Physical Sciences Research Council (EPSRC) via grant nos. EP/N016602/1 and EP/R007438/1, the Royal Academy of Engineering under the Chair in Emerging Technologies scheme and King Fahd University of Petroleum and Minerals, Saudi Arabia.

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Drop coalescence with liquid-liquid interface in the presence of surfactants

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In this work, the effects of surfactants on the coalescence of single drops with liquid-liquid interfaces are investigated. For the studies, high speed imaging, particle image velocimetry and laser induced fluorescence techniques have been used to study the evolution of the interfaces and the velocity fields in the drops. At first, the coalescence of drops with stationary interfaces in two dimensional (Hele-Shaw) and three dimensional cells will be discussed. In the 2D configuration it was found that immediately after the film rupture, the neck growth follows a linear trend for all surfactant concentrations. By assuming interfacial tension values corresponding to twice the surfactant concentration in the bulk, the expanding neck velocities for all surfactant concentrations studied collapse to the same scaling law. The distribution of surfactants on the interfaces during coalescence was further probed with planar laser-induced fluorescence. The two liquid phases had, in this case, matching refractive indices and a fluorescently tagged surfactant was added in the aqueous phase. It was found that during the film thinning, the surfactant molecules were swept outwards by the draining liquid while after the film rupture, the concentration of the surfactants at the tip of the meniscus is indeed high and above its equilibrium value.

Surfactants were found to reduce the partial coalescence region in an Oh-Bo phase map. In the gravity regime of the map, surfactants had a negligible effect on the daughter to mother drop diameter ratio while in the inertia-capillary and in the viscous regimes, an increase in the surfactant concentration resulted in lower drop diameter ratio. Surfactants reduce the interfacial tension while they make the coalescence non-symmetrical, which prevents partial coalescence.

Coalescence was found to be delayed by moving liquid interfaces. A novel flow channel was fabricated that enabled different interface speeds to be studied. It was found that as the interface speed increased the delay in coalescence also increased. This is attributed to the lubrication pressure which develops in the draining film. In addition, with moving interfaces most of the film rupture points appear on the downstream part of the drops.

ACKNOWLEDGEMENTS

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Manipulating droplets with shaped acoustic fields – applications in medical diagnostics and drug delivery

Professor Jon Cooper, University of Glasgow

Microfluidics and droplet technologies are synonymous with the movement of fluids on or around microstructured surfaces, including channels. Flow is often driven by peristaltic pumps or through electrokinetic flows (including electro-osmosis, for example).

Recently, pressure driven flow through the use of surface acoustic waves (SAWs) has attracted much attention. To better control the nature of the acoustic field when using SAWs, we have introduced the concept of using frequency dependent periodic arrays known as phononic crystals within microfluidics. In doing so, we have enabled new "acoustic holograms" that result in waveguiding, reflectors, bandgaps and lenses, that shape the ultrasonic field and create new microfluidic flows.

We are able to demonstrate how we can create interesting droplet manipulations, including droplet centrifugation, cell lysis through shear in droplets, cell separation, liquid heating and atomization, all on disposable "chips".

The applications of this technology are demonstrated through three "real-world" examples, namely (i) sample preparation and identification of malarial parasites in blood; (ii) the measurement of visco-elastic properties in small sample in drops and (iii) the control of the size distribution of droplets during nebulization (a requirement in pulmonary drug delivery, for example).

HOW TRIPLE LINE PINNING AFFECTS WETTING ON TEXTURED SURFACES

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We have explored the kinematics of triple line depinning on textured surfaces. To that aim, we have designed various surfaces, changing periodic surface patterns or texture inclination. Dynamic textures have also been developed for local modulation of the triple line boundary conditions. We have observed the relation between macroscopic contact angle and local triple line kinematics. For a better understanding of the results, numerical models using standard surface energy minimization techniques have also been implemented.

For periodic superhydrophobic surfaces, we have found that the receding contact angle is controlled by point defects of the triple line, which adjusts between consecutive rows through kinks. These kinks mediate line depinning and control the receding contact angle¹. We have also shown that they give rise to surface fraction scaling, as found in the Cassie equation, but also explain the shortcomings of the Cassie equation at low surface fractions². This new kinematics is also useful to understand drop motion on dynamic surfaces, as we have recently demonstrated through magnetically actuated textures.

Finally, we have explored the impact of texture inclination in the Wenzel state. We show that for inclined textures, the triple line can unexpectedly move only “against the nap”, i.e. opposite the direction in which the textures point³. The evolution of the advancing contact angle with inclination can only be explained by a 3D calculation of pinning.

In all these cases, simple rules of mixture do not satisfactorily account for the data and the concept of line pinning has to be called in to reach a successful understanding of dewetting.

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Modelling Drop Dynamics on Liquid Infused Surfaces

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Inspired by pitcher plants, a novel class of functional surfaces, termed liquid infused surfaces (LIS), can be constructed by infusing rough or porous materials with a lubricant. They have been shown to exhibit a wide-range of advantageous surface properties, including self-cleaning, drag reduction, anti-icing and anti-fouling. In this talk I will discuss our recent results modelling the statics and dynamics of drops on these liquid infused surfaces. First, unlike standard wetting problems, our theory shows that the contact angle of a drop on LIS is not uniquely defined by material parameters, but also has a strong dependence on the size of the drop and the pressure of the infusing lubricant [1]. Second, using a novel ternary free energy lattice Boltzmann method to study the dynamics of drops moving across LIS, we observe a rich interplay between contact line pinning and viscous dissipation at the lubricant ridge, which become dominant at large and small apparent angles respectively [2]. Finally, we investigate the motion of drops on LIS with texture gradients. When the lubricant is only partially wetting the solid surface, we will discuss how bi-directional droplet motion may be achieved with the same surface texture.

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SELF-PROPELLED DROPLET TRANSPORT ON SHAPED- LIQUID SURFACES

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The ability to direct droplets on flat surfaces has many practical applications in microfluidics, bio-assay and analytical chemistry. Structured gradient surfaces do offer droplet driving and direction control, but it is necessary to overcome the hysteresis force inherent when any liquid contacts a solid surface. Here, we show that efficient droplet propulsion can be achieved on a shaped-liquid surface where a solid topographic gradient at one length scale provides the base for a smaller length-scale liquid conformal surface. This provides a directional force on a propelling the droplet via an imbalance in the contact angle [1] and high droplet mobility *via* the shaped-liquid surface created using nanoscale roughness imbued with oil [2]. The liquid surface prevents contact between the droplet and the solid and creates liquid on liquid wetting. The resulting surfaces can propel droplets by several times their diameter, as well as against gravity (Figure 1). Furthermore, the strong vertical adhesion of these surfaces, allows impacting droplets to be captured prior to motion (Figure 1b), even when the substrate is completely inverted and the impacting droplets become hanging droplet.

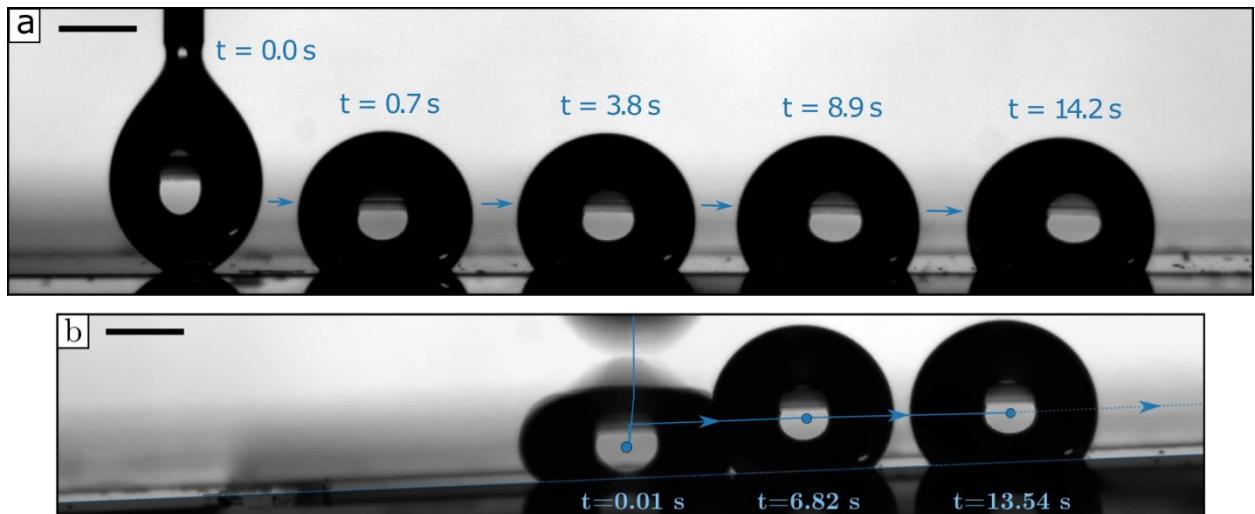


Fig. 1. Droplet self-propulsion on shaped liquid surfaces. (a) Droplet moving by several times its size on a flat surface. (b) Droplet self-propulsion on a tilted surface. Because of the strong vertical adhesion, the droplet is successfully captured (no rebound).

Acknowledgements: This work was supported by the Engineering and Physical Sciences Research Council [grant number EP/P026613/1].

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SYMMETRY SPLITTING OF IMPACTING DROPLETS ON PARTLY WETTING SURFACES.

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Impact of liquid droplets on hard surfaces is of importance in many industrial and natural processes. Although water repellence induced by surface patterning is understood in static systems, its influence in dynamic systems is less obvious. Of interest in this study is the quantitative understanding of droplet shape dependence on surface patterning, which has previously been approached qualitatively.¹

This presentation advances on previous work² with the aim to explain the emergence of fingers in the outer spreading rim in relation to the impact region shape (Fig 1). Impact region shape and size is described as a function of the impact energetics and the microstructure design.

Also of interest in this study are the details of flow present during the early stages of droplet impact on microstructures. Using high speed, high magnification imaging a qualitative description of flow through the microstructure is presented.

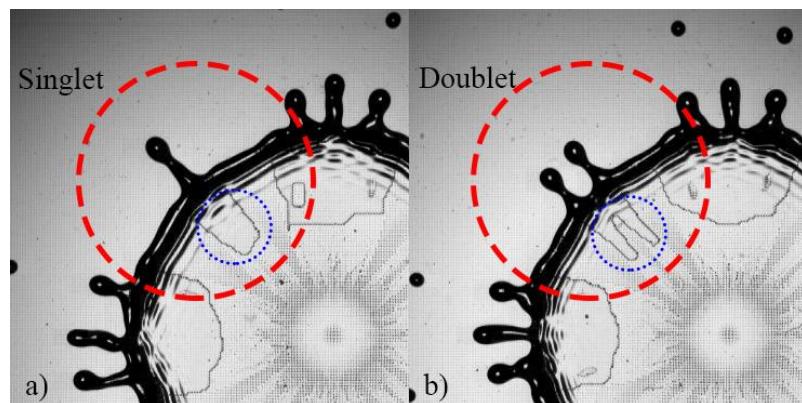


Fig 1: Droplet outcomes on microstructured surfaces consisting of 20 μm square pillars with 60 μm pitch and 15.8 μm height, producing a) a singlet off axis finger, b) a doublet off axis finger.

ACKNOWLEDGEMENTS: The authors thank Royal Society of New Zealand for supporting this work.

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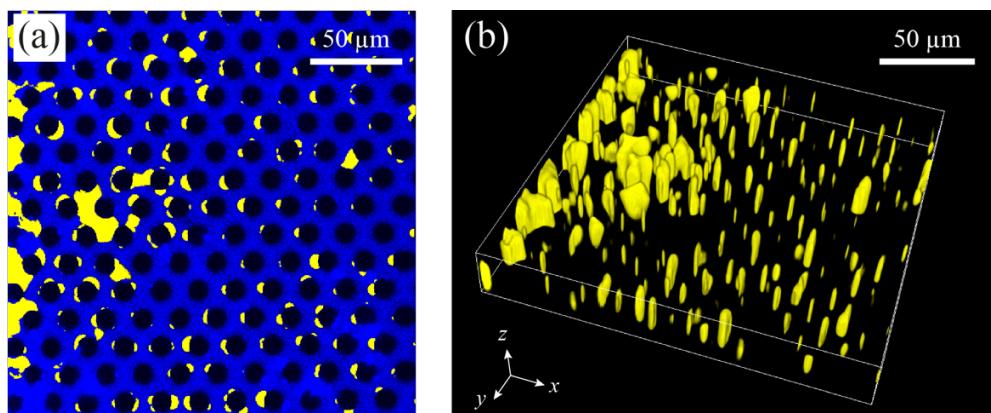
CONTROLLED SOLVENT EXCHANGE IN A POROUS MATERIAL: EXPERIMENT & THEORY

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Abstract: Solvent exchange is a bottom up method to generate micro/nano droplets by nucleation & growth out of an oversaturated solution. In solvent exchange, an oversaturation pulse of a dissolved component, typically an oil, is generated during the mixing of its two miscible solvents, where the good solvent for oil is displaced by the poor one. Here we analytically and experimentally study the solvent exchange in a Hele-Shaw cell type channel with a quasi-2D porous region in the middle. For all the porosities tested, it is found that all of the oversaturation of oil nucleates into droplets and get trapped by the porous region, so that the mixture is no longer oversaturated at the end of the porous region. Solvent exchange is performed with various solvent compositions and flow rates, and the measured total volume of oil is found to depend on the Péclet number Pe with a power law exponent about 0.51. To explain this, we develop a theoretical model based on the ternary phase diagram, where the analytical solution of the total volume of oil is obtained by integrating the composition distribution of oil oversaturation. The total volume of oil is found to increase with Péclet number Pe as $\propto Pe^{1/2}$, where the prefactor can be calculated from the solvent composition. Moreover, we studied how the flow resistance induced by the oil droplets vary with flow rate. These findings may help to improve our understanding and guide further design of solvent exchange.



Confocal images of oil droplets (yellow) in the porous material (black) after solvent exchange (blue is water).

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DROPLET-ACTUATED MICROCHANNEL MIXER

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Mixing and concentration control in microchannels are essential to Lab-on-a-chip devices[1,2]. In this study, droplets are exploited to mix two fluids flowing in parallel to induce intense interfacial disturbance[3]. The proposed mixing mechanism enables us to control the mixing level on demand with a stable operation.

To demonstrate the mixing, water and colored water or particle-dispersed water are mixed in the microchannel. Oleic acid is chosen for the dispersed phase and the droplets are introduced to the mixing channel together with the continuous phase (water). In the mixing channel, the oil droplets strike the mixing front and the interfacial area of two mixing fluids increases as shown in Fig. 1. We found that the striking motion plays a significant role in the mixing and the mixing level can be controlled by adjusting the droplet-striking frequency, channel geometry, and droplet diameter, whereas the confluence angle of inlets for two to-be-mixed fluids has less effect.

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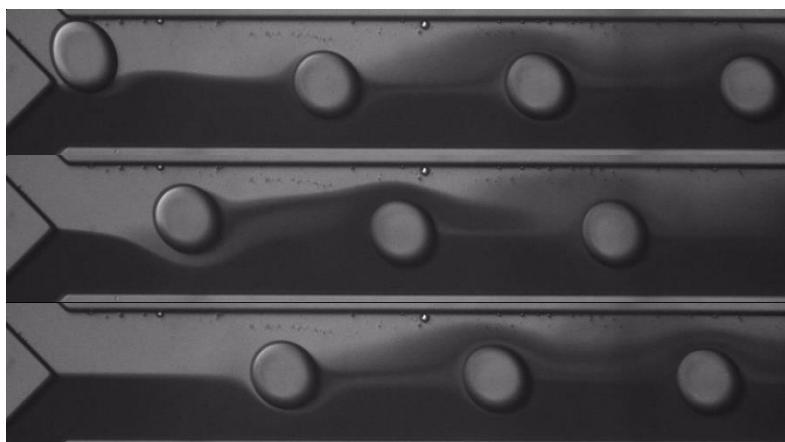


Fig. 1. Successive images of the mixing in a microchannel induced by the droplet injection (channel width = 250 μm).

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THE BASSET-BOUSSINESQ HISTORY FORCE ACTING ON A SPHERICAL DROP

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We consider the Basset-Boussinesq (history) force experienced by a fluid sphere of radius R , viscosity μ_i and density ρ_i moving with relative velocity $W(t)$ in an unbounded fluid of viscosity μ_e and density ρ_e . This force is usually expressed as

$$F_H(t) = 6\pi\mu_e R \int_0^t K(t-t') \frac{dW}{dt'} dt'$$

where expressions of the memory kernel $K(t)$ have been derived for a solid sphere [1,2] and for a spherical bubble [3]. Considering a fluid sphere, these expressions can be considered as the limit at respectively large and small viscosity ratio $\Phi = \mu_e/\mu_i$, respectively. The general expression of the memory kernel K has not been derived so far for a fluid sphere. The solution has been obtained in the Fourier-transform space but the transform from the frequency domain to the time domain can only be achieved under the two limits of solid sphere and spherical bubble [3,4].

We show that the Kernel for a fluid sphere can be described using the kernel obtained for a sphere with a uniform slip along the surface [4,5]

$$K(t^*, \lambda) = \frac{(1+2\lambda/R)^2}{\lambda/R(1+3\lambda/R)} \exp\left[\frac{(1+3\lambda/R)^2}{\lambda^2/R^2} t^*\right] \operatorname{erfc}\left[\frac{(1+3\lambda/R)t^{*1/2}}{\lambda/R}\right]$$

where $t^* = t/t_e$ is the time normalized by the characteristic time of diffusion $t_e = R^2/v_e$, $v_e = \mu_e/\rho_e$ being the kinematic viscosity of the external fluid, and where the slip length λ is related to the viscosity ratio by $\lambda = R/(3\Phi)$. Thanks to direct numerical simulations, we show that such an expression can be used when the characteristic time τ of the flow is less than 0.15 t_e . Thus, combining the analytical expression of the Basset-Boussinesq kernel for a slip sphere and the description of the slip at the interface of a fluid sphere, we are able to describe for the first time the Basset-Boussinesq history force for a fluid sphere whatever the viscosity ratio considered, i.e. for bubbles, drops and particles.

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HYDRODYNAMIC-COLLOIDAL INTERACTIONS OF AN OIL DROPLET AND A MEMBRANE SURFACE

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Membranes offer a superior separation capacity for stable emulsions, which are otherwise difficult to process. However, membrane fouling is always prevalent and requires extensive backwashing and cleaning as part of the operating regime. Oil droplets deform when in close proximity to a membrane, primarily due to permeation drag, which can lead to irreversible deposition on the membrane. Understanding the influence of hydrodynamic stresses and colloidal interactions between droplets and membranes will allow for better engineered membrane surfaces and process conditions. Using a long-wave hydrodynamic description that incorporates wettability via a Derjaguin (disjoining) pressure and electrostatic repulsion via electric double layer (EDL) contributions, we derive a set of equations for the thickness and pressure in the liquid film separating the droplet and the permeable membrane.

Numerical simulations are used to analyse equilibrium states where a zero net force acts on the droplet, to investigate the influence of droplet shape, distance from the membrane, membrane permeability and characteristic parameters of the colloidal interactions on the overall repulsion / attraction towards the membrane surface. A stability phase diagram is constructed in terms of the modified capillary number - accounting for the ratio of the viscous and surface tension forces - and a parameter representing the ratio of the hydrodynamic pressure scale over the electrostatic interaction intensity.

These calculations are used to classify stable (upward deflection, increasing distance from the membrane) vs. unstable drop shapes (downward deflection bringing the droplet closer to the membrane), and investigate conditions leading to irreversible deposition onto the membrane. In particular, the model can describe how membrane properties and emulsion characteristics dictate a 'critical' flux.

ACKNOWLEDGEMENTS: The authors thank the support at the Technion by a fellowship from the Lady Davis Foundation.

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SELF-SIMILAR COALESCENCE OF LIQUID LENSES

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We present a study of the coalescence of liquid lenses, e.g. drops floating on a liquid pool. High-speed imaging is used to experimentally study the initial stages of coalescence. These experiments reveal that the bridge connecting the two lenses exhibits a self-similar growth, in both the viscous and inviscid regimes. The results are compared to similarity solutions for the bridge shape, derived from the one-dimensional thin sheet equations. This is complemented by numerical simulations that provide a detailed picture of the flow inside the lenses.

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ATTRACTION OF SESSILE DROPS OF THE SAME PURE VOLATILE LIQUID

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Controlling the motion of sessile drops has many well rewarding applications in industry. For instance, creating a surface tension gradient on the surface of a heat exchanger improves the total heat transfer by up to three times due to the motion and coalescence of sessile drops (Daniel et al. 2001). In this research we show experimentally that two sessile drops of pure volatile perfectly wetting liquid placed in a close proximity of one another feel each other and attract (See Figure 1) at a velocity significant enough for the drops to coalesce well before they eventually vaporize. Our experiments reveal that the drops attract each other even if, unlike the binary-liquid drops recently studied (Cira et al. 2015), they are made of the same pure liquid. Several perfectly wetting liquids of different volatilities are tested in order to unveil and quantify the mechanisms enabling droplets to communicate. While all recent works on the topic consider vapour-mediated interactions only, we here show that evaporation-induced temperature gradients in the substrate heavily influence this dynamics. For instance, we will describe a hereto unknown “cold-trap resistance” as an effective drag force opposing any motion, like the viscous drag does. The interaction mechanisms described here could hopefully open new directions of research about thermal effects as a mean of self-organizing evaporating/condensing liquid entities on substrates of various shapes and thermal properties.

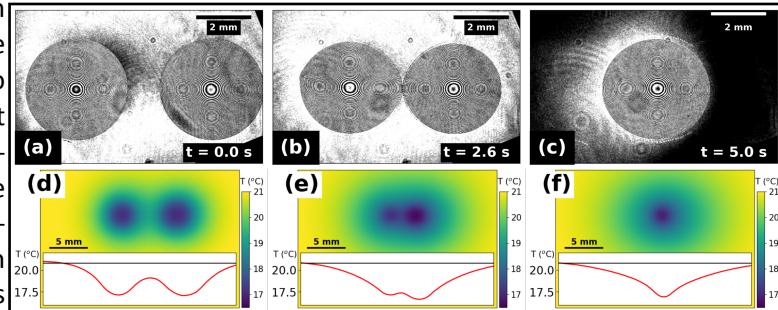


Figure 1- a) Attraction between two sessile hexane drops (initial radii ~ 1.9 mm, inter-apex distance = 5.25 mm on a sapphire substrate, (b) first touching after 2.6 s, (d-f) forming a single drop at 5 s, (d-f) temperature of substrate measured using an Infrared camera for attracting HFE7100 sessile drops

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LONG RANGE INTERACTION OF TWO PL- SESSILE DROPLETS ON A SOLID

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Drops are not isolated in printing and spray-coating processes. Sessile drops at a distance can show long-range attraction across a high surface energy substrate, mediated by their vapour¹, or one can chase the other. Drops landing on a wet film can lead to splashing, bouncing, spreading, a coalescence cascade or instant absorption². Recent work on sessile drops next to each other has shown delayed coalescence of miscible liquids³. This diversity of behaviour is associated with a wealth of dynamics occurring inside the liquid. The fundamental understanding, prediction and control of the behaviour of sessile droplet interactions and coalescence have drawn growing interests from both academic and industrial communities. Sessile drop coalescence may be desirable in generating a continuous track or uniform film – or undesirable, for example, in inkjet printing process where colour blending is to be avoided. Previous research was heavily focused on μL -drops which may not display the same behaviour as the pL- or nL- droplets that are typical in agriculture spraying or inkjet printing. Vapour-mediated drop motion on a substrate is not fully understood, for example, it is not clear or not a precursor film contributes towards droplet mobility. There have been no detailed measurements on internal flows to determine the contribution of Marangoni flows to the drop motion and coalescence in small drops. We also lack knowledge of the influence of substrate on drop behaviour.

At Durham, we developed a rig to study morphology and internal flow of two pL- sessile droplets printed on a solid with a controlled time delay, spacing and composition at industrial printing time and length scales. We investigated the long-range interaction between a drop of ethanol and a drop of water and between a pair of ethylene glycol (EG)/water droplets with varying EG concentration. Plasma cleaned glass was used in both cases and a hexamethyldisilazane-treated glass substrate with controlled receding contact angle (RCA) was also used in the former case. In the case of ethanol/water droplet pair on the high surface energy substrate, we observed that water droplet is repelled within 1 ms after the ethanol droplet lands, with a shortest edge-to-edge distance of the two droplets of 150 μm . We propose the motion is due to the asymmetric adsorption of ethanol vapour into nearby water droplet and the consequent Marangoni stress along the water-vapour interface. The time and length scales agree well with diffusion-control theory. The motion of the water droplet depended on the RCA of the substrate and the time delay between the water and ethanol droplets. Ethanol adsorption to the water droplet had an indiscernible effect on its volume and evaporation rate. However, we observed flow switching inside water droplet in case of pinned contact line on a silanised substrate before and after the disappearance of ethanol droplet. In the case of two droplets of EG/water mixture at a distance, strong attractive motion or chasing is observed under controlled drop composition and time delay. Experiments have been designed to distinguish between the effects of condensation, suppressed evaporation and enhanced evaporation in inducing asymmetric Marangoni flow and droplet motion.

ACKNOWLEDGEMENTS: The authors thank EPSRC under grant EP/N025245/1 for supporting this work.

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MOLECULAR EVENTS KICK-OFF DROPLET COALESCENCE

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Ranging from the formation of thunderstorms in the atmosphere to the printers in our offices, droplet-based systems are ubiquitous in our everyday life. However, until now we have had only partial understanding of the mechanisms by which two or more droplets coalesce to form a larger droplet. The classical notion of coalescence of two droplets is that surface tension drives the process from the beginning. Using computationally expensive molecular simulations, here we show that thermal capillary waves on the droplet surface initiate single or multiple contacts between droplets and coalescence commences in a *thermal regime*. Here the bridges expand linearly in time due to collective molecular jumps near the bridge fronts. In this non-classical regime, surface tension acts to suppress the bridge growth. Transition to the classical hydrodynamic regime only occurs once the bridge radius exceeds a thermal length scale, and so this requires to be considered in hydrodynamic analyses of droplet coalescence.

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Spreading dynamics and contact angle of completely wetting volatile drops

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Abstract

The spreading of evaporating drops without a pinned contact line is studied experimentally and theoretically, measuring the radius of completely wetting alkane drops of different volatility on glass. Initially the drop spreads (increases), then owing to evaporation reverses direction and recedes with an almost constant non-zero contact angle θ ; eventually the drop vanishes at a finite-time singularity. Our theory, based on a first-principles hydrodynamic description, well reproduces the dynamics of and the value of θ during retraction.

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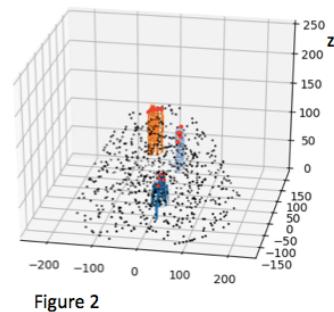
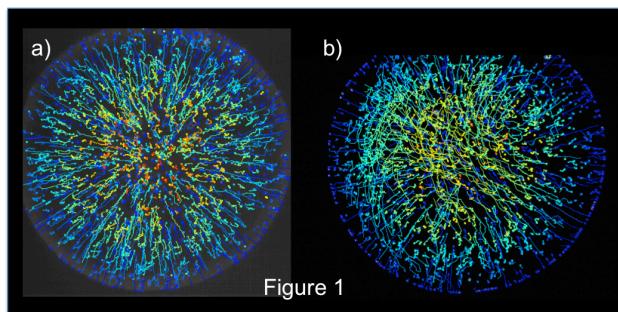
3D PARTICLE TRACKING IN SESSIL EVAPORATING WATER DROPLETS

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Our study comprises the experimental and numerical analysis of the 3D motion of particles in suspension inside evaporating sessile drops of water. The main objective is to track individual particles (1micron polystyrene spheres) throughout the evaporation process to correlate the initial position of suspended particles and their final positions on the sediment. As it has been documented before [1], the process has two characteristic times; the slow, initial part and the fast final part when the drop is a liquid film that rips off the contact line. The experimental observations are made with an inverted microscope equipped with two cameras with different image acquisition rates to capture the events with the two time scales, and an oscillating objective for z multiplane acquisition (see [2]). Microscope images at fixed heights provide information of the particles at a horizontal plane and known time. Tailoring positions of individual particles, the track can be integrated (modified version of Trackmate, ImageJ software [3]). Figure 1 shows the plan view of the particle tracks of 700 particles for initial and final evaporation parts, which lasted 3 and 1.5 min respectively. Traces are rainbow color coded with respect to their average height (red for higher and blue for lower average z positions).



Clearly, the dynamics of the two stages is different with the first being dominated by radial motion and the second by the drag generated by the retreating liquid line after the film pulls away from the contact line. The red traces on the center of Figure 1a) indicate that the particle displacements at the top of the drop are vertical as they are pictured head-on. This is confirmed in Figure 2 where the 3D position of the tracks is displayed. Black dots indicate initial position of particles and color lines are individual tracks during the overall first part evaporation process. Traces that start on the free surface near the top of the drop are nearly vertical (orange lines) while those starting inside the drop have a large radial component. The circles contain the tracers at different horizontal planes and their diameter indicates the spread out of originally neighboring particles. The initial and final diameter of the orange set is approximately the same but the final diameter of the dark blue traces is almost twice as large as the initial one. A numerical simulation of the flow based on the solution of mass and momentum conservation equations with mass loss at the free surface has been performed. The particle tracks are the obtained as streak lines and the agreement is satisfactory.

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Density-Driven Flows in Evaporating Binary Liquid Droplets

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Flow mechanisms and evaporation dynamics of a micro-particle laden sessile droplet deposited on a solid substrate have attracted a lot of interest for the past 20 years because of emerging applications in a wide range of industries such as inkjet printing, additive manufacturing and biomedical assays. Most research focuses on single component droplets, revealing an outward radial flow driven by bulk liquid transport towards a pinned contact line [1]. However, in many applications, droplets contain two or more liquid components of different volatilities, warranting a detailed understanding of the flow driving mechanisms and the resulting deposition patterns in these more complex solutions.

In this work, we report a pioneering development of rotatable optical coherence tomography to image the flow patterns in tilted evaporating binary liquid droplets deposited on substrates with contact angles between 20° and 100°. The evaporation of the droplets proceeds in three distinct stages: chaotic (Stage I), convective (Stage II) and outward radial flow (Stage III). Stage I is characterised by random and strongly circulating vortices, attributed to solutal Marangoni flows driven by the preferential evaporation of the more volatile component (ethanol) [2]. Stage III is characterised by an outward capillary flow of the less volatile single component droplet (water) [1]. Our measurements conclusively demonstrate that, in contrast to the accepted view and conventional calculations of the Marangoni and Rayleigh numbers, the convective flow (Stage II) is induced within the droplets by a difference in density between an outer shell and the bulk of the droplets due to the evaporation of one of the components. We also use gas chromatography to determine the time evolution of the concentration of the more volatile component within the droplet and confirm that a simple analysis of volume data provides the same information. Finally, we establish a flow phase diagram demonstrating the conditions under which the different stages occur.

ACKNOWLEDGEMENTS: A.M.J. E. acknowledges financial support from Nottingham Trent University.

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Drop impact-based energy harvesting using charged hydrophobic polymer surfaces

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Hydrophobic polymer surfaces spontaneously accumulate negative charge upon contact with water. Likewise, rolling drops on hydrophobic surfaces tend to leave traces of negative charge due to a poorly understood triboelectric charge transfer process, which has recently been used to harvest energy from rolling drops. Here, we present an alternative droplet-based energy harvesting process, which relies on drop impact onto polymer surfaces treated by a dedicated charging process [1,2] that deposits substantially higher amounts of stable, permanently trapped charge than previously reported. We quantify the trapped charge density and its life time using electrowetting and measure the resulting electrical current signal upon drop impact. A kinetic model is proposed to describe the charging mechanism. The enhanced charge density enables more efficient conversion of droplet kinetic energy into electric energy. The underlying mechanism is based on transient variations of electrical capacitances due to drop spreading, retraction, and rebound upon impact on the surface. Synchronized high speed video imaging of impacting drops [3] and electrical current measurements confirm the proposed energy conversion mechanism.

ACKNOWLEDGEMENTS: H.W. thanks the National Key R&D Program of China and Guangdong Province Key Laboratory of Optical Information Materials and Technology for supporting this work.

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Two recipes for repelling hot water

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While a hydrophobic microtexture at a solid surface most often reflects rain owing to the presence of entrapped air within the texture, it is much more challenging to repel hot water. As it contacts a colder material, hot water generates condensation within the cavities at the solid surface, which eventually builds bridges between the substrate and the water, and thus destroys repellency. In this talk, we will explore impacts of hot water drops on cold superhydrophobic materials. While both “small” ($\sim 100 \text{ nm}$) and “large” ($\sim 10 \mu\text{m}$) model features do reflect hot drops at any drop temperature and in the whole range of explored impact velocities, surprisingly water does not always bounce on intermediate ($\sim 1 \mu\text{m}$) textures. Hence, we can define two structural recipes for repelling hot water: drops on nanometric features hardly stick owing to the miniaturization of water bridges, while kinetics of condensation in large features is too slow to connect the liquid to the solid at impact.

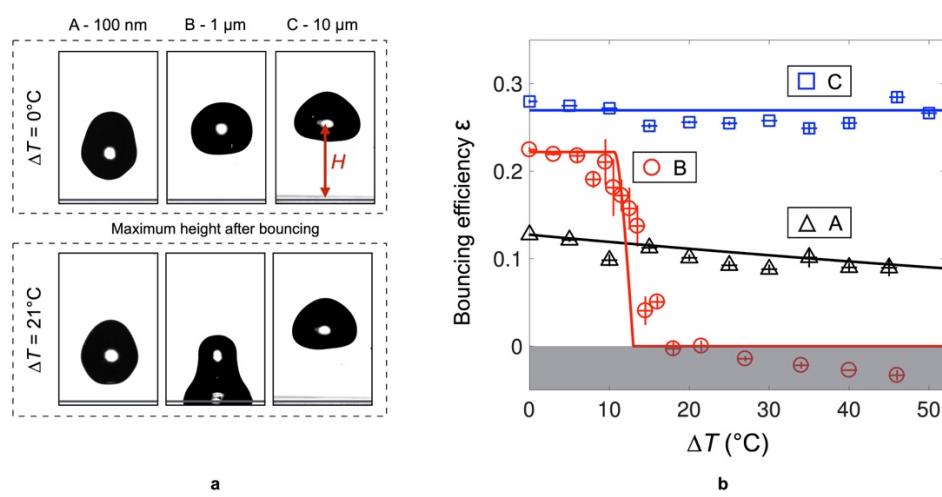


Figure. Bouncing efficiency of hot drops. a. Snapshots of impacting drops ($R = 1.4 \text{ mm}$ and $V = 40 \text{ cm/s}$) at their maximum bouncing height H , for $\Delta T = 0^\circ\text{C}$ (top images) and $\Delta T = 21^\circ\text{C}$ (bottom images) on materials A, B and C with respective pillar heights of about 100 nm, 1 μm and 10 μm . b. Bouncing efficiency ϵ as a function of the temperature difference ΔT for substrates A, B and C, $R = 1.4 \text{ mm}$ and $V = 40 \text{ cm/s}$. Water always bounces on samples A and C, while it gets trapped on B above $\Delta T \approx 15^\circ\text{C}$.

LIQUID BREAK-UP UPON DROP IMPACT NEAR THE EDGE OF AN INCLINED SUBSTRATE

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Raindrop impacts on plant leaves are responsible for the dispersal of several crop diseases [1]. The most likely scenario is that of a raindrop impacting in the immediate vicinity of a pathogen-bearing liquid residue initially present on a leaf. The raindrop would induce the fragmentation of this residue in a myriad of droplets ejected toward neighbour plants [2].

In this talk, we present experiments on the impact of a drop close to the horizontal edge of an inclined substrate. We show that this generic impact configuration shares key features with the more complex impacts on plant leaves. We investigate the formation and break-up of the liquid sheet formed when the liquid expands beyond the edge [3] (cf. figure below). The impact Weber number, the distance from the impact point to the edge, and the substrate inclination are systematically varied. We analyse their influence on the kinematics of both the liquid spreading on substrate and the liquid sheet beyond the edge, as well as on the subsequent statistics of droplet ejection (radius, speed, time of ejection).

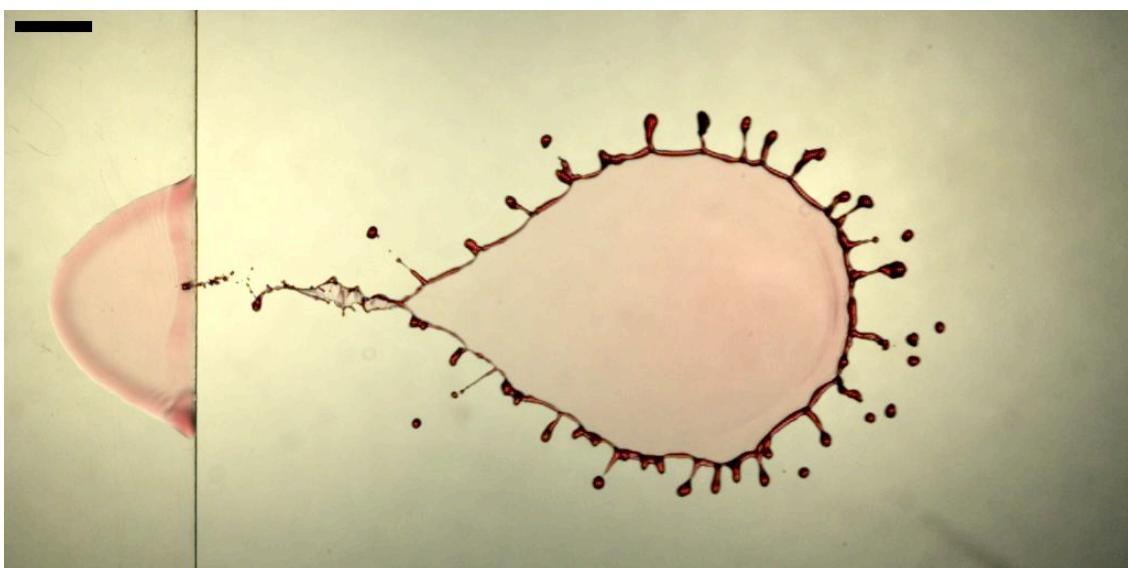


Figure: Impact of a drop (Weber # = 2115) close to the edge of a substrate inclined at 60° downwards (left). A liquid sheet is formed beyond the edge (right), and droplets are ejected from the surrounding rim. The scale bar is 5 mm.

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THIN FILM ACOUSTOFLUIDICS: A NEW PLATFORM FOR LAB-ON-A-CHIP

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This talk will focus on piezoelectric thin films including zinc oxide (ZnO) and aluminium nitride (AlN), which are used for a broad range of lab-on-chip applications such as biosensing, particle/cell concentrating, sorting/patterning, pumping, mixing, nebulisation and jetting. Integrated acoustic wave sensing/microfluidic devices have been fabricated by depositing these piezoelectric films onto a number of substrates such as silicon, ceramics, diamond, quartz, glass, and more recently also polymer, metallic foils and bendable glass/silicon for making flexible devices. Such thin film acoustic wave devices have great potentials for implementing integrated, disposable, or bendable/flexible lab-on-a-chip devices into various sensing and actuating applications. Different acoustofluidic and biosampling functions which can be generated from thin film surface acoustic wave (SAW) devices.

Recent developments in thin film acoustic devices have been focused on: (1) sensors prepared on cheap and commonly used substrates to reduce the cost, or new substrate materials to introduce novel functionality, or flexible substrates for wearable and wireless applications; (2) high frequency sensing with improved sensitivity; (3) shear-wave or Love mode based thin film devices for liquid sensing; (4) Development of various sensing layers/material systems; (5) Integration with acoustic streaming and microfluidics to form the Lab-on-a-chip systems. We foresee that such microsystems are capable of performing the complete task from delivering drops of bio-sample into the device, cell/bacteria separation, manipulation, and tissue generation, through to the delivery of the detection results, using portable, wireless, flexible and remotely controlled devices.

ACKNOWLEDGEMENTS: UK EPSRC EP/P018998/1.

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RECIPROCATING MOTION OF FEMTOLITER DROPLETS BETWEEN TWO LIQUID INTERFACES

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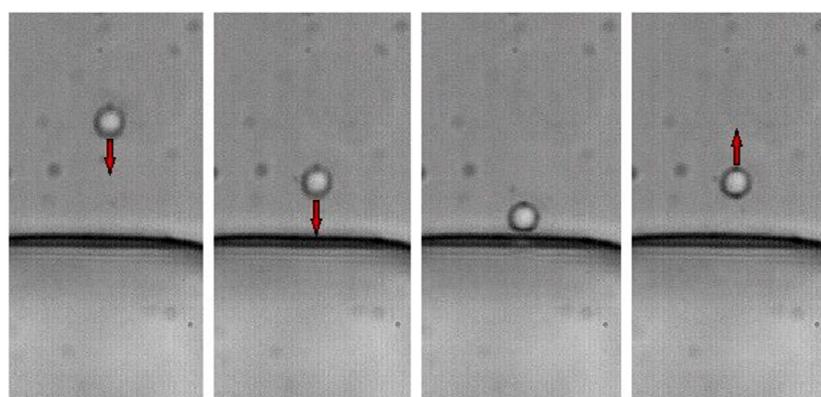
We demonstrate a handling scheme for femtoliter droplets that is based on the reciprocating motion of the droplets between two oil/aqueous interfaces. The aqueous droplets move in a microchannel filled with oil between two interfaces to aqueous phases. The motion is driven by a DC electric field between the interfaces. Under suitable conditions [1], upon touching an interface, a droplet does not merge but bounces back. This is demonstrated in the time-lapse images below that show a 5 μm diameter droplet bouncing back from the interface. The arrows indicate the direction of motion. As a result, a droplet can perform a reciprocating motion between the two interfaces. We characterize the regimes in parameter space where this type of motion occurs.

Upon touching an oil/aqueous interface, a droplet exchanges a tiny amount of dissolved species with the liquid reservoir. We study the mass transfer to the droplet by imaging the transfer of a fluorescent dye contained in one of the aqueous reservoirs. After a few cycles, the dye concentration inside the droplet reaches an asymptotic value, indicating that the amount of dye taken up at one interface equals the amount released at the other interface. That way the droplets qualify as tiny semi-batch reactors, a function that could be important for mimicking the metabolism of cells. The mass transfer to/from the droplets is size selective: While dissolved molecular dyes are able to pass the bridge forming between a droplet and an aqueous reservoir, 500 nm polystyrene beads are rejected. The size selectivity could open new avenues for the selective transfer of smallest sample amounts.

ACKNOWLEDGEMENTS: The authors acknowledge the support from the LOEWE CompuGene Project, funded by the Hessian Ministry of Science and Art.

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TRANSPORT OF AQUEOUS DROPLET PLACED OVER OIL BASED FERROFLUID SPIKES IN PRESENCE OF A MAGNETIC FIELD

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Droplets can be used as carrier vehicles for the transportation of biological and chemical reagents¹. Magnetic field based manipulation being non-contact and biocompatible can be advantageous over other active manipulation techniques. An oil based ferrofluid splits into array of spikes due to the presence of a vertical magnetic field². We report the transportation of aqueous droplets over oil-based ferrofluid spikes in presence of a magnetic field. Our study reveals that transportation phenomena is governed by the interplay of three different forces – magnetic force F_m , frictional force F_f and interfacial tension F_i , which is expressed in terms of the force ratios, $k_m = (F_f/F_m)$ and $k_i = (F_f/F_i)$. Based on the values of the force ratios k_m and k_i , three different regimes – steady droplet transport, spike extraction and magnet disengagement, are identified. It was found that steady droplet transport is observed for $k_m \leq 1$ and $k_i \leq 1$, whereas extraction of spikes is observed for $k_m \leq 1$ and $k_i > 1$, and magnet disengagement is observed for $k_m > 1$. In the steady droplet transport regime, velocity of the aqueous droplet U_{ds} was found to be dependent on the volumes of the aqueous droplet V_w and FF droplet V_{FF} . In the spike extraction regime, the spike extraction distance L_{se} was found to vary with V_w , V_{FF} and the magnet velocity U_{ms} .

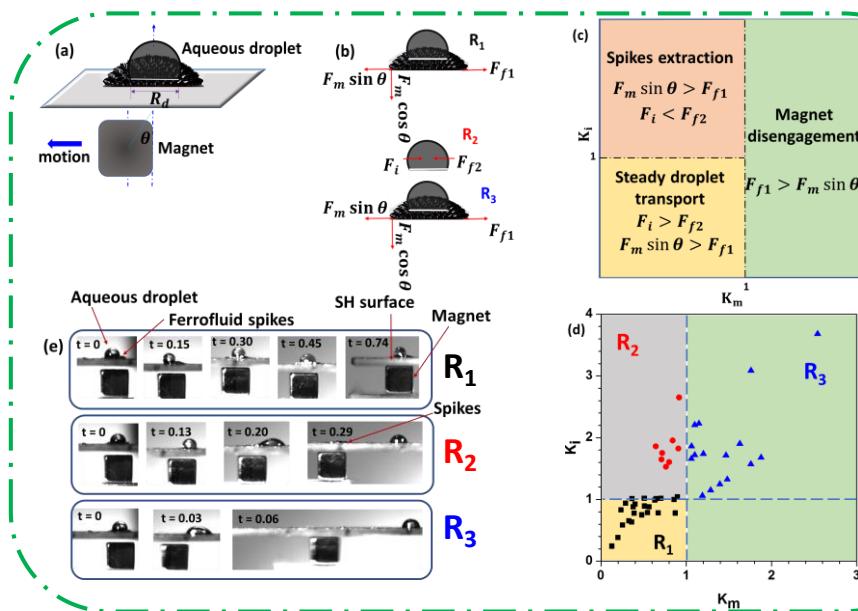


Figure 1 (a) Schematic of the experimental setup, (b) Force analysis, (c) Regimes of droplet transport, (d) Regimes obtained from experimental data, (e) Experimental images of various regimes

ACKNOWLEDGEMENTS : The authors would like to thank I.I.T. Madras (MEE/15-16/843/RFTP/ASHS) for providing the financial support which enabled this work.

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VAPOUR POINT-SOURCE CONTROL AND MANIPULATION OF EVAPORATING DROPLETS

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The manipulation and control of evaporating droplets are important for phenomena as diverse as printing, thin-film deposition and self-assembly. Typical approaches to perform such tasks rely on controlling the intrinsic physical, chemical and/or geometrical properties of the droplet and its environment at the onset of the evaporation process. Here, I will show a novel experimental technique to generate recirculating Marangoni flows within the droplet during evaporation. In particular, the deterministic emergence and real-time control of these flows can be achieved within the evaporating droplet by relying on an external point source of vapour positioned near the droplet's surface. Depending on the droplet's initial composition, these flows can be either used to control in-situ deposition of materials when the droplet is sessile [1] or, alternatively, they can be used to induce controllable 2D manipulation of a moving droplet for applications in printing, materials deposition and for controllable reactions [2].

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THE UNDERSIDE OF A LEIDENFROST DROP ON A BATH

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The Leidenfrost effect, when a drop levitates on its own vapor over a hot solid surface, can also be observed over hot baths of non-volatile liquids [1-2]. Compared to the classical Leidenfrost effect on solid substrates, the liquid bath presents three major differences: the substrate is atomically smooth, deformable, and fluid. As a consequence of such fluidity, heat transfer through the bath to the drops is most certainly dominated by convection and not by just conduction as in the solids. Here, we undertake an experimental and numerical study of the flow in the bath of silicone oil V20 induced by an overlying Leidenfrost drop. We highlight that the structure of this flow is far from being universal. In particular, the sense of circulation in a toroidal vortex formed under the drop is found experimentally and confirmed theoretically to depend on the nature of the liquid that makes the drop. We show that this is due to a shift in a complex and delicate interplay between three mechanisms pulling in different directions: the local cooling of the bath by the drop gives rise to both (i) a buoyancy action and (ii) Marangoni stresses, whereas the vapor escaping from the gap between the drop and the bath exerts (iii) a shear action on the bath surface. Whatever the structure of the convection, its efficiency for heat transfer through the bath is readily confirmed in numerical simulations.

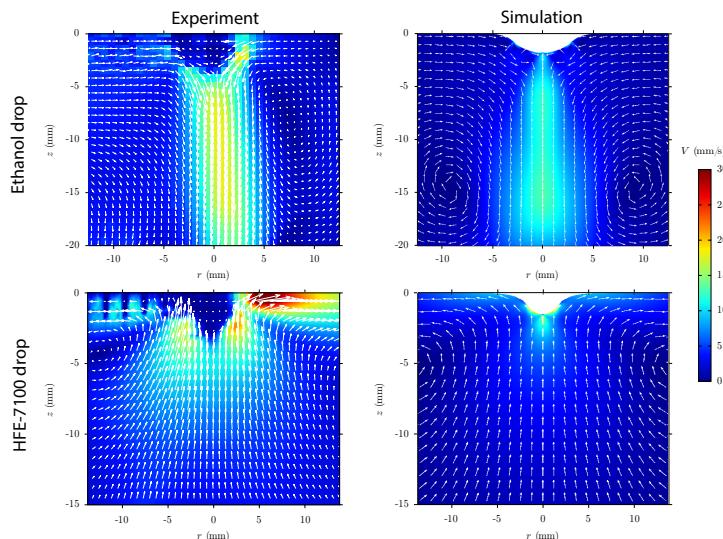


Figure: Experimental (left) and numerical (right) flow fields in the bath under levitating ethanol (top) and HFE-7100 (bottom) Leidenfrost drops, both of radius $R/l_c=2$ and subject to a superheat $\Delta T=19^\circ\text{C}$.

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PARTICLE MIGRATION IN INKJET-PRINTED DROPLETS

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The evaporation of sessile droplets can be a useful method to pattern substrates, with inkjet printing technology being particularly good at the selective deposition of functional materials. A predictive understanding of formulations is necessary in order to design systems in which the internal flows generated during drying do not lead to undesirable non-uniform morphologies.¹ Composition or temperature gradients across the liquid-vapour interface have been shown to induce Marangoni flows which can redistribute suspended material,² however studies have mainly taken place on microlitre droplets.

Here we report experiments on the internal flows of inkjet-printed picolitre droplets in which high-speed cameras are used to follow the trajectories of light-scattering tracer particles and record the droplet profile. Solutal Marangoni flows are generated in a selection of solvent mixtures and solutions however at these smaller length-scales different morphologies are observed. Instead of obtaining uniform deposits, particles are seen to migrate across flow streamlines³ to collect in groups in ethanol-water mixtures, ethylene glycol-water mixtures and sucrose, lactose, sodium chloride and sodium nitrate solutions, demonstrating the prevalence of particle migration in a disparate range of chemical systems. A weak particle-size dependence to the migration is noted and a diffusiophoretic mechanism proposed.

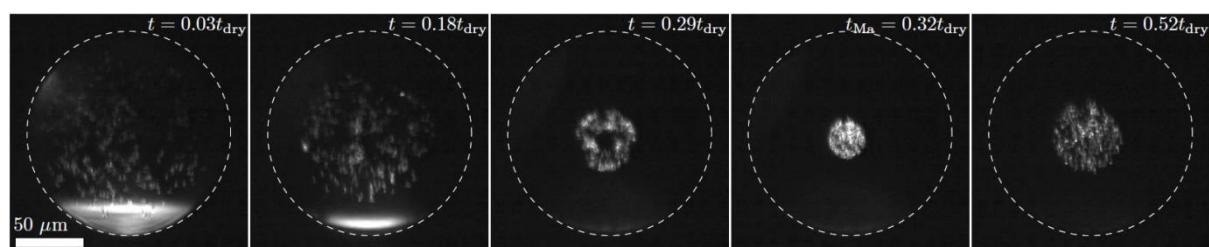


Figure 1. Particle migration towards the centre of an evaporating droplet of a 50:50%v ethanol-water mixture at $RH = 50\%$ on a cleaned glass coverslip. The dashed line is the position of the contact line. t_{Ma} is the time when the Marangoni flows ended and the collected group had its minimum radius while the overall drying time of the droplet was 2.4 s.

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SURFACE TENSIONS OF PICOLITER DROPLETS WITH SUB-MILLISECOND SURFACE AGE

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Aerosols are key components of the atmosphere and play important roles in many industrial processes. Because aerosol particles have high surface-to-volume ratios, their surface properties are especially important to their reactivity and cloud droplet forming potential. However, direct measurement of the surface properties of aerosol particles is challenging. In this work, we describe a new approach to measure the surface tension of picoliter volume droplets with surface age <1 ms by resolving their dynamic oscillations in shape immediately after ejection from a microdroplet dispenser (Figure 1). Droplet shape oscillations are monitored by highly time resolved (500 ns) stroboscopic imaging (Figure 1b). These shape oscillations follow a damped oscillator (Figure 1c), the frequency of which (Figure 1d) gives the surface tension. Droplet surface tension is accurately retrieved across a wide range of droplet sizes (10-25 μm radius) and surface ages (down to \sim 100 μs). The approach is validated for droplets containing sodium chloride, glutaric acid, and water. Experimental results from the microdroplet dispenser approach are compared to complementary measurements of the surface tension of 5-10 μm radius droplets with aged surfaces using a holographic optical tweezers approach and predictions of surface tension using a statistical thermodynamic model. These approaches combined allow investigation of droplet surface tension across a wide range of droplet sizes, compositions, and surface ages.

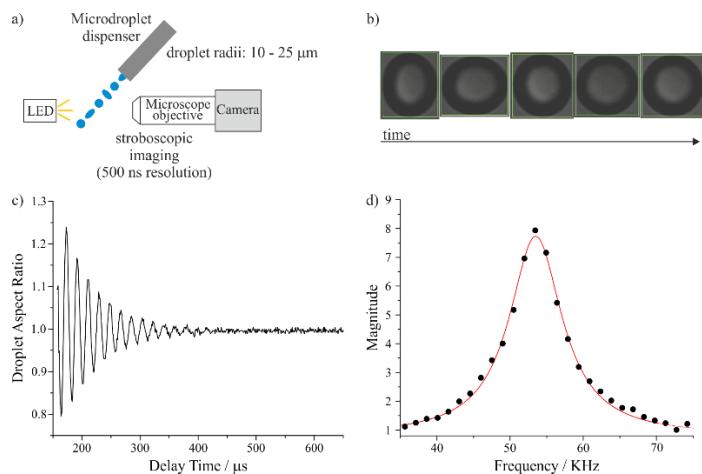


Figure 1. a) Schematic of the microdroplet dispenser setup. b) Images of droplet shape shortly after ejection. c) Droplet aspect ratio vs. time after droplet formation illustrates the damped oscillations in shape. d) A Fast Fourier Transform of the aspect ratio plot gives the oscillation frequency, which allows accurate retrieval of surface tension.

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TEMPORAL EVOLUTION OF MULTI-ORDER STIMULATED RAMAN SCATTERING IN DROPLET

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Nonlinear Raman scattering proceeds under an intense light field of the incident and the Raman scattering lights. A liquid droplet provides the intense light field because it acts as a high-quality optical cavity. We previously found that a colliding droplet acts as an optical cavity with higher quality factor than the single droplet by using carbon tetrachloride (CCl_4) as sample liquid, where multi-order stimulated Raman scattered light emerges with significant intensity.[1] In the present study, we investigated the mechanism of the multi-order Raman scattering from temporal evolution of the stimulated Raman scattered light in the single/colliding droplet.

Liquid droplets of CCl_4 were produced by a set of piezo-driven nozzles. The single/colliding droplet was irradiated with a second harmonic of a Q-switched Nd:YAG laser for excitation of the stimulated Raman scattering. The Raman scattered light passed through a long-pass filter for removal of the Rayleigh scattering, and divided into two components for a simultaneous measurement of the collision image and the spectrum. For the measurement of the temporal profile, the Raman scattered light was monochromated and detected by a photomultiplier.

Figure 1 shows the stimulated Raman spectrum obtained from the single and the colliding CCl_4 droplet. For single droplet, peaks in the spectrum are assignable to an integer multiple of 460 cm^{-1} , the wavenumber of the v_1 mode of CCl_4 . For colliding droplet, on the other hand, peaks appear at Raman shift with linear combinations of the vibrational modes of CCl_4 , such as v_1 (460 cm^{-1}), v_2 (240 cm^{-1}), v_3 (770 cm^{-1}), and v_4 (340 cm^{-1}). The Raman bands are denoted as $[a\ b\ c\ d]$, where $a - d$ represent the number of the $v_1 - v_4$ modes, respectively, included in the band.

Figure 2 shows the temporal profile of the Raman bands observed from the single and the colliding droplet. From the single droplet, higher-order Raman scattered light emerges with longer delay time from the incident laser. From the colliding droplet, on the other hand, the higher order Raman scattered light originating from the v_1 mode emerges with almost no delay time, whereas those originating from v_2 and v_4 modes have delay of several nanoseconds which is significantly smaller than the delay of the single droplet. These results are explained by assuming the multi-order Raman scattering proceeds after building up light field of the fundamental and the most intense Raman mode inside the droplet.

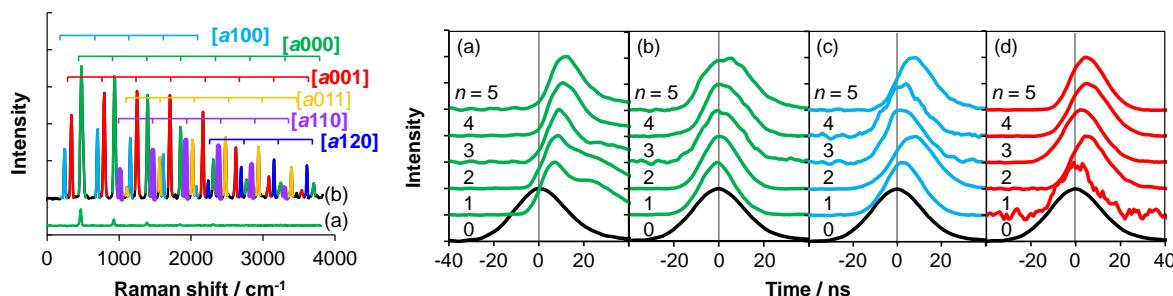


Figure 1 Raman spectra of single (a) and colliding (b) CCl_4 droplet.

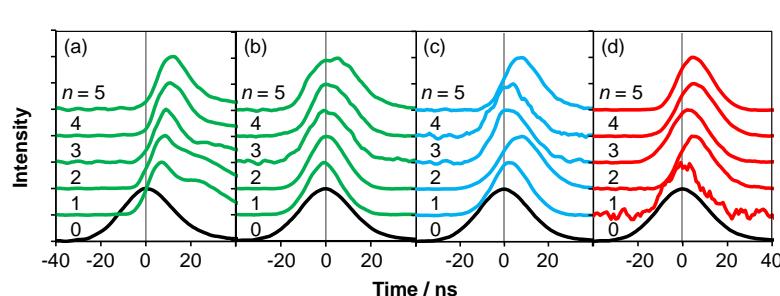


Figure 2 Time profiles of multi-order Raman bands of nv_1 mode obtained from single (a) and colliding (b) CCl_4 droplet, and those of v_2+nv_1 (c) and v_4+nv_1 (d) modes from colliding CCl_4 droplet.

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CONCENTRATION-DRIVEN ACOUSTIC INSTABILITY IN AEROSOLS

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A temperature gradient imposed on a dense aerosol may trigger an acoustic instability which, in turn, may accelerate the coalescence and agglomeration of droplets. Thermoacoustic instability, i.e. a critical temperature gradient imposed on a solid-gas interface initiating spontaneous self-sustained oscillations, has been intensively studied in the context of undesired acoustic waves produced in combustion chambers. Recent work^{1,2} demonstrated how modifying the working fluid to contain water vapour dramatically decreases the temperature gradient required for triggering this instability. In this process a thin film of liquid covers the solid such that a liquid-gas interface is formed, from which water evaporates and condenses according to the local equilibrium with the surrounding gas. In the present work we speculate on the feasibility of a similar process, only here we consider a uniform, three-dimensional structure of droplet in contact with the gas rather than a thin film.

In the considered configuration, the liquid-gas interface is discontinuous such that the gas is not constantly in contact with its source/sink. Unlike a thin film interface, droplets are free to move with the surrounding oscillating gas, with their entrainment factor largely determined by the droplet diameter³. A minimum droplet size of $\sim 1 \mu\text{m}$ is required, below which droplets are practically fully entrained and the instability is unlikely to occur.

As a first approximation, the droplets formation is thought of as only slightly deviating from a close packing of spheres, hence $d/L = 1 - \varepsilon$ with d and L the droplet diameter and distance between neighbouring droplets, respectively, and $\varepsilon \ll 1$. A linear stability analysis is performed, seeking the critical temperature gradient and the resonant frequency that would trigger the instability. The results demonstrate the possibility of a relatively low temperature gradient initiating acoustic oscillations in a dense aerosol.

ACKNOWLEDGEMENTS: The authors acknowledge the support from the Nancy and Stephen Grand Technion Energy Program.

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ADAPTIVE WETTING

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The dynamic process of liquids that wet or dewet surface is not only ubiquitous in everyday life. It is also of key importance in many technological applications. Existing work mostly focuses on the influence of the topography and composition of rigid and inert substrates. Here, we discuss the wetting of adaptive surface. Adaptive substrates change their physico-chemical properties due to the presence of a liquid or its vapor. As it turns out, not only specifically designed substrates adapt, but adaptation to the liquid is a rather general phenomenon. Adaptation can explain contact angle hysteresis.

We propose a first order kinetic model to describe dynamic contact angles of such adaptive surfaces. The model is general and does not refer to a particular adaptation process. The aim of the proposed model is to provide a quantitative description of adaptive wetting and to link changes in contact angles to microscopic adaptation processes. By introducing exponentially relaxing interfacial energies and applying Young's equation locally, we predict a change of advancing θ_a and receding contact angles θ_r depending on the velocity of the contact line. Even for perfectly homogeneous and smooth surfaces, a dynamic contact angle hysteresis is obtained. As possible adaptations we discuss changes in the surface state reconstruction of polymers or monolayers, diffusion and swelling, adsorption of surfactants, reorientation of liquid molecules or the formation of an electric double-layer.

Crossover of Dynamic Wetting Regimes: a Molecular Dynamics Study

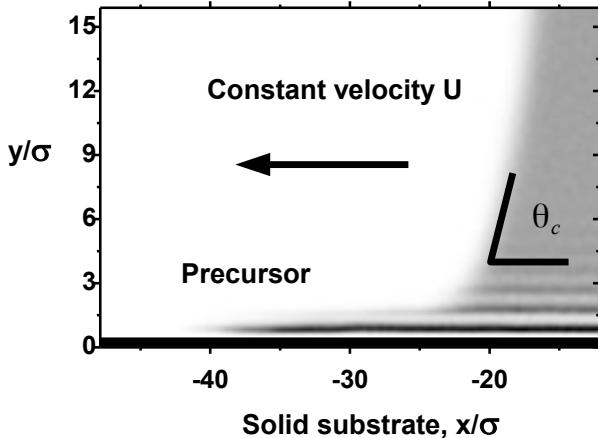
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The dynamic spreading regime in the complete wetting case of advancing contact line motion features a transition when the contact line speed approaches a critical value from above and an advancing precursor film starts to appear. In the study, which is a sequel of our previous investigation of the dynamic contact angle mechanism, **ACS Nano**, (2016), **10**: 6045-6053, we analyze a crossover between the two dynamic wetting states, with and without the precursor film, using molecular dynamics simulations. We investigate, from the first microscopic principles, characteristic features of the onset of the precursor film and its relationship with the dynamic contact angle. Remarkably, though could be expected, we observed that even when the precursor film was well developed, the dynamic contact angle θ_c (as is illustrated) was still defined by the whole area covered by the precursor film.

Density distribution at the contact line region in a steady state



Out-of-balance contact-line force in a steady state

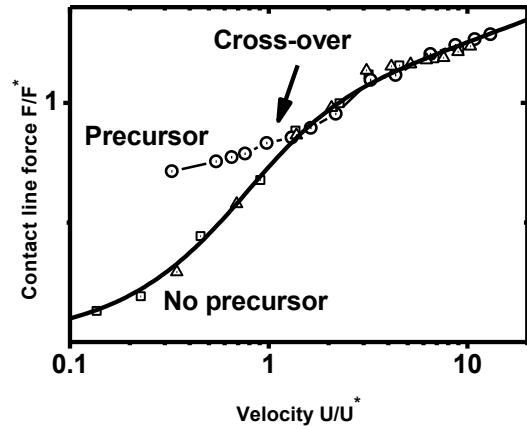


Figure 1: Illustration of the onset of the **precursor** film in advancing contact line motion - **MDS**. **Density distribution** (liquid is shown in dark colour) in complete wetting case and the out-of-balance **contact line force** $F/F^* = \gamma_{GS} - \gamma_{LS} - \gamma \cos \theta_c$ as a function of normalized velocity U/U^* in complete wetting (static angle $\theta_S = 0$, precursor present) and incomplete wetting (static angle $\theta_S > 0$, no precursor) cases.

Keywords: wetting, nano-scale, contact line, macroscopic boundary conditions, molecular dynamics simulations.

Controlling Bubbles with Electric Fields

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The wetting of a surface by a droplet of a dielectric liquid can be controlled using surface fabricated interdigitated electrodes and an applied voltage to create an electric field which decays with depth of penetration into the liquid.¹ This interface-localized liquid dielectrophoresis (L-DEP) introduces a dielectrophoretic energy stored in the liquid near the interface in addition to the interfacial energies from the liquid-solid, solid-liquid and liquid-vapor interfaces. Thus, an applied voltage can be used to control the wetting of the surface – an effect called dielectrowetting.¹⁻³ Here, we show that in the complementary situation of an air-bubble attached to a surface immersed in a dielectric liquid, the extent of wetting can also be fully controlled including inducing bubble detachment. We then control the vertical position of the bubble close below the surface and induce bubble reattachment to the surface. The thickness of the liquid film preventing reattachment is shown to have a logarithmic dependence on the magnitude of the applied voltage. We describe these experimental results by developing a theory that considers the energy balance due to liquid-dielectrophoresis and the buoyancy of the liquid-bubble system. This work provides a new method of bubble control that has the potential to control flow in channels and to manage dry patches in heat exchangers.

We acknowledge funding from the UK EPSRC (EP/K014803/1, EP/K015192/1, EP/R036837/1 and EP/R042276/1).

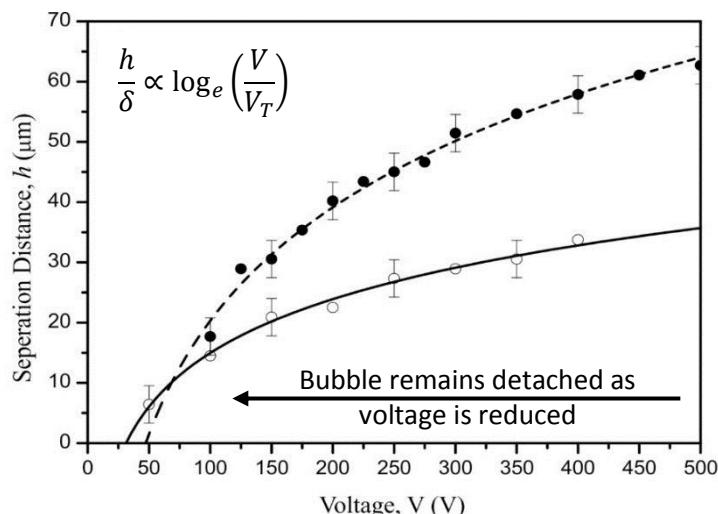


Figure 1: Experimental data and fit for reattachment for two electric field penetration depths, δ .

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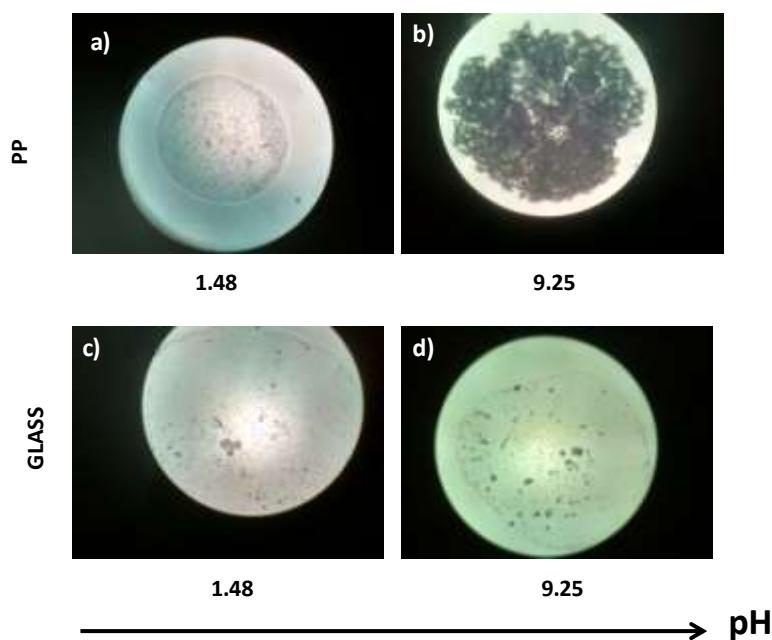
ROLE OF pH AND SUBSTRATE ON DRYING PATTERNS OF LAPONITE DROPLET

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Disc shaped Laponite RD particles in aqueous solution have a net negative charge on the faces and a positive charge along the rim with a pH of 9.7. Changing the pH of the aqueous Laponite solution will determine whether the Laponite particles arrange themselves in a repulsive glass phase or form a cage-like gel structure. When droplets of Laponite solution having different pH are allowed to evaporate, the arrangement of the particles during drying is further perturbed by drift and diffusion currents of mass transfer due to uneven evaporation flux from the droplet surface and Marangoni effects. In this work we report the interesting patterns left by drying droplets of aqueous Laponite with different pH dried on a polypropylène substrate, Figs.(a) and (b). The pH was varied from 1.5 to 13 by adding measured volumes of HCl and NaOH. In highly acidic pH a prominent coffee-ring was observed , while in a strong basic pH, salt crystals on Laponite particles, gathered at the centre of the dried droplet. Similar studies were done on glass substrate with quite different results of dried patterns as shown in Figs.(c) and (d). This proves that substrate effects couple with the role of pH in the determination of the final pattern. We offer an explaination of the different patterns on the basis of intermolecular forces of repulsion between the partides, the attractive van der Waal's forces and the resultant Debye length of the electric double layer surrounding each particle.



ACKNOWLEDGEMENTS: SH thanks DST, India for supporting this work through INSPIRE scholarship.

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EMBEDDING, REBOUND AND TUNNELING OF LIQUID DROPLETS IMPACTING ONTO FREELY SUSPENDED FLUID FILMS

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We have demonstrated experimentally that microdroplets shot onto freely suspended smectic films can interact with the films in three different ways, depending on the impact speed [1,2]. They are trapped and embedded in the film when they are sufficiently slow. At intermediate impact speeds they are reflected, and at high speeds they tunnel the film, become encapsulated with a thin closed layer of film material. They leave the film intact in all three scenarios. The tunneling of thin smectic films can be employed to create smectic shells, i. e. micrometer sized droplets of an immiscible liquid encapsulated by a thin smectic layer, which can be collected in another liquid [2].

Extending these experiments to millimeter sized droplets, we are able to study droplet impact on smectic films over huge parameter ranges, varying the Weber, Bond and Reynolds numbers over nearly three orders of magnitude, kinetic energies over 6 orders of magnitude and time scales over four orders of magnitude, thus providing a comprehensive description of droplet impact and interactions for different droplet and film materials.

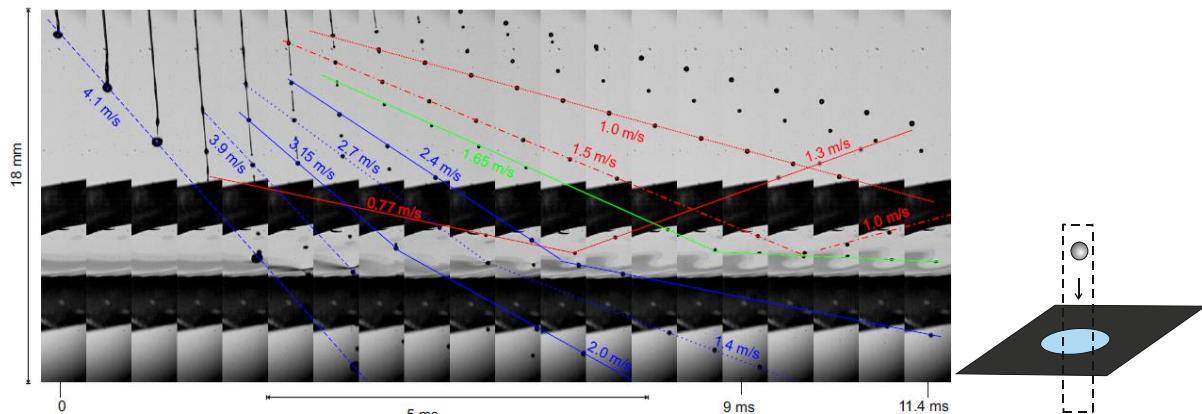


Figure 1: Time series of large droplets impacting on a freely suspended smectic film. The drawing at the right hand side schematically sketches the geometry, the dashed rectangle indicates the region that was selected for the image series. The images show droplets of different sizes in the range of few hundred microns to about one millimeter, impacting on a smectic film with different velocities. Fast droplets can tunnel the film, slow ones are reflected. The trapping and embedding regimes exchange their roles for these large droplets as compared to microdroplets.

ACKNOWLEDGEMENTS: This study was funded by DFG within project STA 425/44-1 .

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ACID-SOAP COMPLEXES IN LEVITATED ATMOSPHERIC AEROSOL PROXIES: HUMIDITY AND OZONE RESPONSE

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Unsaturated fatty acids, such as oleic acid, are a major constituent in organic atmospheric aerosols. The main sources of oleic acid include marine and cooking emissions. The phase state of organic aerosols has been shown to be of importance to the reactivity of organic aerosols towards atmospheric oxidants such as ozone, implying a more significant effect on aerosol atmospheric lifetimes with implications for weather and pollution. Being surface active, fatty acids can self-assemble in water into a range of lyotropic liquid crystal (LLC) phases and we have previously shown this in levitated droplets of oleic acid/sodium oleate/water atmospheric aerosol proxies.¹

Here we present the observation of a crystalline oleic acid/sodium oleate acid-soap complex in our levitated aerosol proxy. Simultaneous Small-Angle/Wide-Angle X-ray Scattering (SAXS/WAXS) and Raman spectroscopy was used on our levitated particle to probe the structural (fig. 1) and chemical changes in the acid-soap complex during atmospherically-relevant humidity changes and exposure to ozone. The micro-focus capability on the I22 SAXS beamline at the Diamond light source (UK) made it possible to resolve structural differences within the particle on humidification. This has revealed an implied viscosity gradient within a humidifying acid-soap complex particle due to the formation of a micellar phase in the outer regions of the particle. Diffusion and viscosity gradients within atmospheric aerosol particles and particle proxies have been theorised and observed before. This is the first time this has been observed using SAXS.

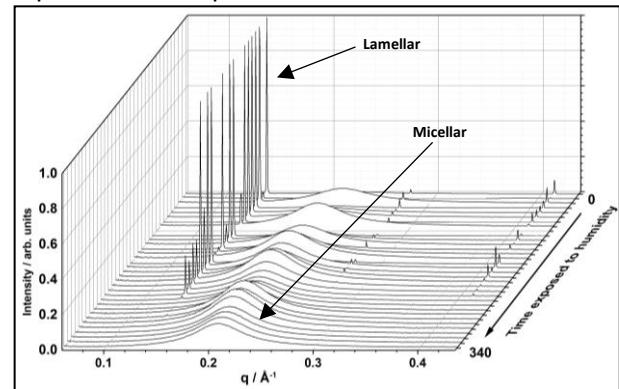
Ozonolysis of the levitated acid-soap complex revealed a significant decrease in reaction rate compared to pure oleic acid, it was also possible to see structural changes in the Raman spectrum as a result of ozonolysis; showing the degradation of the acid-soap complex with time exposed to ozone (fig. 2).

In summary, our lab-based observation of a crystalline acid-soap complex in a levitated particle and its response to ozone and an atmospherically-relevant humidity change has added to the dynamic nature of our atmospheric aerosol proxy. A humidity-dependent phase (and viscosity) change was observed and has implications for the atmospheric lifetime of unsaturated fatty acid aerosols and urban climate.

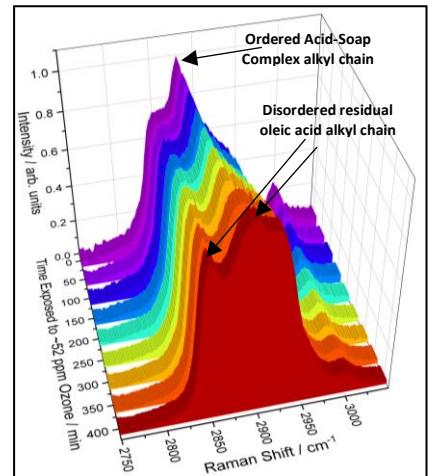
ACKNOWLEDGEMENTS: This work was supported by NERC grant no. F4026713. Diamond Light Source (UK), Prof. Nick J. Terrill (Diamond) and Dr Andy D. Ward (Central Laser Facility) are acknowledged.

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[Figure 1. The changing 1D SAXS pattern of the outer region of a levitated acid-soap complex during exposure to >90% relative humidity]



[Figure 2. Evolution of the Raman spectrum of a levitated acid-soap complex during exposure to ~52 ppm O₃. Change in –CH₂ stretching region]

THE DISJOINING PRESSURE IN A DROPLET ON A SPHERICAL SOLID PARTICLE: DFT RESULTS

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The effects of internal inhomogeneity of a thin spherical liquid droplet formed on a wettable solid particle are manifested through the appearance of its disjoining pressure [1]. For a flat thin liquid film between a solid substrate and undersaturated vapor, the disjoining pressure is defined as the difference in the normal component of the pressure tensor in the film and the bulk pressure in the liquid phase at given chemical potentials and temperature. However, because the normal component of the pressure tensor in the liquid film with curved interfaces depends on the location inside the film, it is not completely clear how to define the disjoining pressure for such films. It is possible to use a thermodynamic route [1] and to take the disjoining pressure in spherical films to be the same as in flat films of the same thickness. A rigorous analysis of the mechanical equilibrium condition of a curved wetting liquid film on a solid substrate established a relationship [2] between the normal component of the pressure tensor inside the curved film in the vicinity of the solid substrate, the Laplace pressure of the film, and the difference in bulk pressures in the liquid and gas phases for given molecular chemical potentials. This relation can be considered as a mechanical route to find the disjoining pressure. However, it was still unclear how close to the solid core one should choose the local value of the normal component of the pressure tensor and how the thermodynamic and mechanical routes for the disjoining pressure are consistent in reality.

We have shown here that the answer can be found with the help of the density functional theory (DFT) for a Lennard-Jones fluid with the Carnahan-Starling hard-sphere contribution. Using the square-gradient variant of the DFT at different values of the condensate chemical potential, we have found density profiles for thin spherical liquid films around the solid core and the density peak in the vicinity of the core, which exceeds the bulk value for the liquid density and weakly depends on the value of the condensate chemical potential. Our analysis of the profiles for density and normal and tangential pressure in the central part of the thin droplet on a solid core demonstrates overlapping of surface layers of solid-liquid and liquid-vapor interfaces. We have shown that the disjoining pressure for a flat thin liquid film on a solid substrate in the undersaturated vapor can be related to the disjoining pressure in small spherical droplets on completely wettable solid cores in supersaturated vapor found through the thermodynamic and mechanical routes. The disjoining pressure of the spherical liquid films depends on the internal and external radii of the film.

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AEROSOL OPTICAL PROPERTIES DURING THE FORMATION OF BROWN CARBON AEROSOL

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Light absorbing organic aerosol is referred to as brown carbon (BrC). Light absorption by BrC is highly uncertain, with BrC composed of a diverse range of organic species that absorb light at UV-visible wavelengths <500 nm. BrC formed from α -dicarbonyl gas (such as glyoxal and methylglyoxal) reacting with ammonium-containing particles has been of recent interest,¹ with glyoxal and methylglyoxal estimated to contribute significantly to atmospheric organic aerosol.² Notably, BrC can be formed through aqueous droplet chemistry or *via* heterogeneous reaction on particle surfaces.

We use laser-based spectroscopy in laboratory studies of BrC aerosols, including for reactions in aqueous droplets or *via* heterogeneous reactions on solid particle surfaces. Photoacoustic and cavity ring-down spectroscopy afford non-contact measurements of aerosol absorption (σ_{abs}) and extinction (σ_{ext}) cross sections respectively. A suite of spectrometers measure σ_{abs} and σ_{ext} at the visible wavelengths 405, 514 and 658 nm. Laboratory-generated seed particles of ammonium sulfate (AS) passed to a reaction volume where the humidity and concentration of α -dicarbonyl gas was controlled. The aerosol reacted for ~15 minutes prior to size-selection with a differential mobility analyser and subsequent measurement of σ_{abs} and σ_{ext} . From these measurements, we determined the intrinsic microphysical properties of real and imaginary refractive index. Fig. 1(a) shows the wavelength dependence in the measured σ_{abs} for the reaction of methylglyoxal with aqueous AS particles, with the BrC formation rate in aerosols (minutes) accelerated compared to that in bulk solutions (>10 days). Fig. 1(b) shows the σ_{abs} for BrC formed *via* the heterogeneous reaction of glyoxal on solid AS particles, with BrC formed only when the ambient humidity is >30%, but remains below the deliquescence humidity of AS. Our current effort is focussed on the design of an injection flow tube reactor with the capability of measuring the UV photobleaching of BrC aerosols.

ACKNOWLEDGEMENTS: MIC thanks the Royal Society of Chemistry/Analytical Chemistry Trust Fund Tom West Fellowship for support. JMH and MIC thank the Natural Environment Research Council for support *via* grant NE/L013797/1. The Met Office funded experimental measurements.

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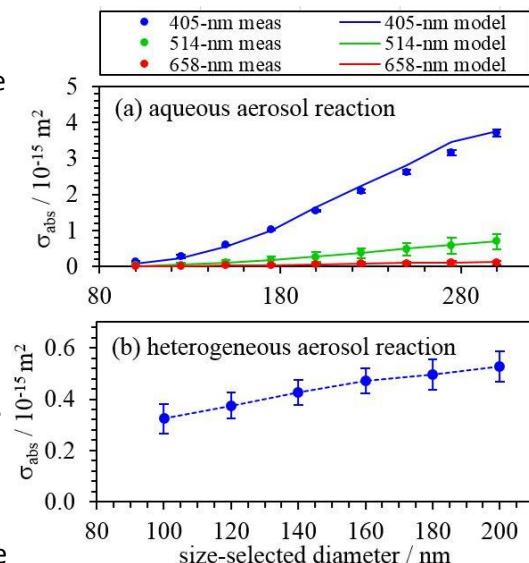


Fig. 1 - (a) The measured size-dependent σ_{abs} for BrC formed from the reaction of methylglyoxal with aqueous AS particles. Measurements are fit to Mie theory (solid lines), in combination with the measured σ_{ext} for retrieval of n -RI and k -RI. **(b)** The measured size-dependent σ_{abs} for BrC formed from the reaction of glyoxal on solid AS particle surfaces.

CRYSTALLINE VS. AMORPHOUS: PREDICTING AND CONTROLLING PARTICLE FORMATION IN RAPIDLY EVAPORATING AEROSOL MICRODROPLETS

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The drying of droplets in the aerosol phase to form solid microparticles is of fundamental importance to a range of industries, particularly spray-drying wherein powdered products for pharmaceuticals, food and cosmetics are produced from rapidly drying an aerosolised feed solution. The resulting particle size, morphology and degree of crystallinity, whilst crucial for the desired product, can be very sensitive to the drying process, and hence to the processing conditions. In this work we are studying the propensity for crystallisation in a rapidly evaporating droplet, to understand what governs crystallisation or amorphous particle formation in a spray-dryer.

Using an Electrodynamic Balance (EDB) we levitate a single liquid droplet (radius $\sim 25 \mu\text{m}$) and collect the elastic light scattering pattern from a 532 nm laser that illuminates the droplet. Using the scattering phase function and the geometric optics approximation we can calculate the droplet radius throughout the drying process and distinguish between crystallisation and the formation of an amorphous particle.¹ We can thus measure the drying kinetics and propensity to crystallise for droplets containing a range of components (inorganic, organic or mixtures, e.g. NaNO₃, KNO₃, NaCl, lactose, sucrose) in different drying conditions. Using SEM imaging analysis, we can relate the final dry-particle morphology to the evaporation kinetics. By the addition of co-components to the initial droplet solution we can induce, prevent or delay crystallisation in an evaporating droplet of an aqueous inorganic salt (see Fig. 1). We thus present a step closer to ultimately predicting and controlling product morphology and degree of crystallinity in the products of spray-drying processes.

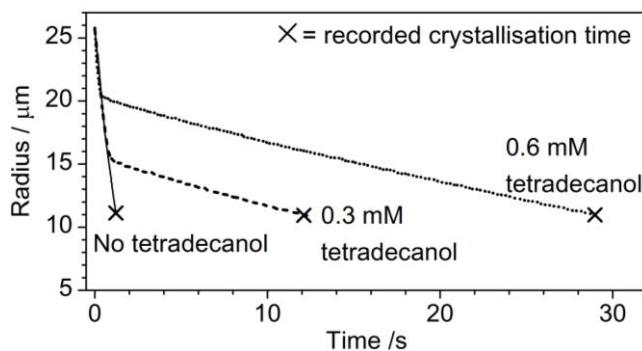


Figure 1: An example of a technique to delay crystallisation in a salt solution droplet in the EDB. A droplet containing NaCl (10% wt/wt), ethanol (45%) and water (45%) evaporates and crystallises at $\sim 11 \mu\text{m}$ but with increasing amounts of a small addition of surface-active alcohol which reduces the evaporation rate, we delay the crystallisation time.

ACKNOWLEDGEMENTS: The authors acknowledge the funding support from the EPSRC under grant code EP/N025245/1.

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NUMERICAL SIMULATIONS OF INKJET PRINTING PROCESSES

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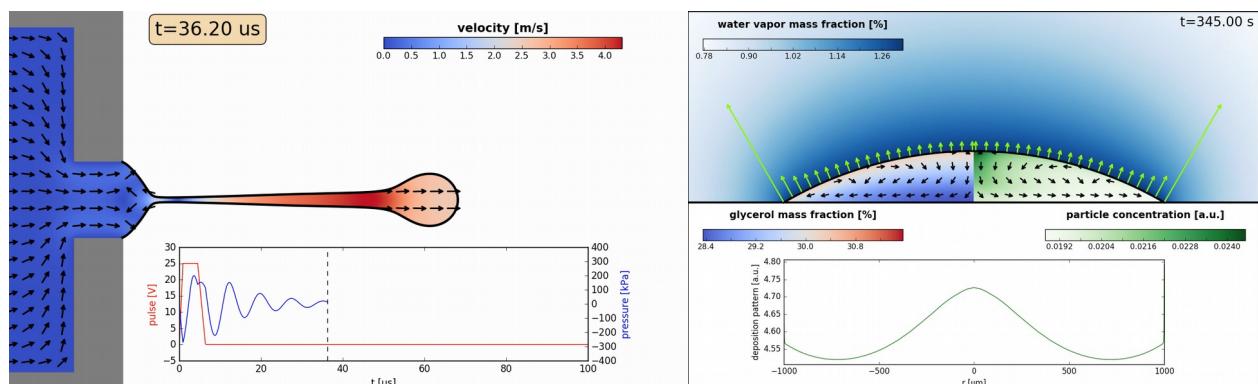
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Nowadays, when you can easily order any kind of book or even customized photo books with a few clicks online, print-on-demand solutions become more and more important, for which ink-jet printing is an ideal candidate. This process, however, demands controlling the relevant processes, i.e. the jetting of droplets and the subsequent evaporation and absorption dynamics, which is even more complex due to the fact that ink is constituted of a mixture of different liquids, surfactants and pigments.

Using a sharp-interface ALE finite element method, we numerically investigate all the aspect relevant in ink-jet printing. We show how a short pause in jetting can result in clogged nozzles due to solvent evaporation and how mixture droplets evaporate and coalesce on the paper. Furthermore, the relevance of surfactants is addressed, helping to control the Marangoni flow to avoid undesired effects and leading to a perfect final printout.



ACKNOWLEDGEMENTS: This work is part of an Industrial Partnership Programme of the Foundation for Fundamental Research on Matter (FOM), which is financially supported by the Netherlands Organisation for Scientific Research (NWO). This research program is co-financed by Océ-Technologies B.V., University of Twente, and Eindhoven University of Technology.

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DEFORMING FREE SURFACE OF SOLUTION DRIED ON A SUBSTRATE WITH CIRCLE-PATTERNED BANK STRUCTURE: NUMERICAL STUDY

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A mathematical model describing drying behavior of a solution droplet deposited on a flat surface was proposed to predict the morphology of the formed solid film. The model includes the fluid dynamics, heat transfer, and mass transfer, and also considers wettability of the substrate and deformation of the free surface [1].

This model is applied into the system where a solution dries on a substrate with circle-patterned bank structure (Figure 1). The radius (a) and height (b) of a hollow cylinder are 2.25 μm and 45 μm , respectively. N, N'-bis(3-methylphenyl)-N, N'-bis(phenyl)-benzidine (TPD) is chosen as a typical organic semiconductor. Anisole is chosen as solvent. Initial mass fraction of solute is 0.02. Contact angle (Ψ_c) is assumed to be 60°.

The effect of solutal Marangoni forces on drying behavior is mainly discussed. Figure 2 shows time variation of liquid frame. R and Z are defined as r/a and z/a , respectively. Dashed lines represent the results for the case where Marangoni forces are neglected. The free surface keeps a near-spherical cap during drying. The center of the free surface reaches near the bottom of substrate when the droplet volume decreases to 50% of the initial volume (i.e. $V/V_0=0.5$). On the other hand, solid lines represent the results where Marangoni forces are considered. At $V/V_0 < 0.6$, Marangoni flows near the meniscus largely deform the whole of the free surface. The deformation develops the effective convective flows to transfer the solute mass as shown in Figure 3, which depicts the time variation of area density profile of solute. It is found that the shape of density profile at $V/V_0 = 0.2$ is similar to the experimental morphology of the solid film formed under the same condition [2].

Consequently, the present calculations reveal that solutal Marangoni flows play an important role in formation of solid film.

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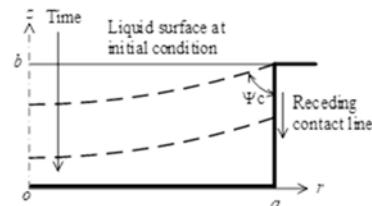


Figure 1 Computational domain

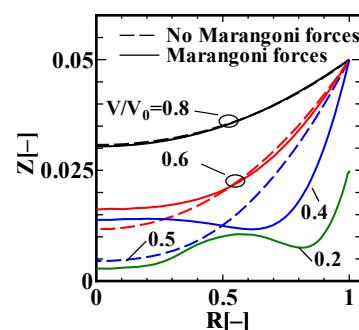


Figure 2 Change in free surface profile with time

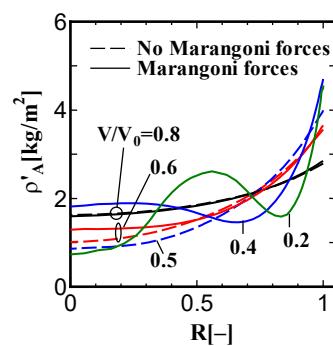


Figure 3 Change in area density profile with time

THE EFFECT OF A PRECURSOR FILM ON EVAPORATING INKJET DROPLETS WITH SURFACTANTS

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A common method for solving the contact line singularity of an evaporating sessile inkjet droplet is by assuming a thin liquid film that covers the substrate. The existence of this precursor film and a disjoining pressure enables the combination of the no-slip condition with the moving contact line. An assumption often made in literature is that the pressure in the precursor film can be set equal to zero. It is argued that the Laplace pressure in the macroscopic part of the droplet is negligible due to the large curvature and that thus an equilibrium solution results in a constant zero pressure^[1]. However, also if there is significant curvature in the droplet, it is still commonly assumed that the pressure in the precursor film is negligible^[2,3]. This causes a pressure discontinuity at the contact line. It is shown that this discontinuity has significant influence on flow profiles in evaporating inkjet droplets, both with and without surfactants.

Using a lubrication model, it is demonstrated that the zero-pressure condition results in only flow towards the contact line, but that without this assumption a secondary flow towards the droplet apex is formed. Similarly, with the addition of insoluble surfactants a circulating Marangoni flow is formed for the zero-pressure condition, but without this condition the flow splits in a branch flowing towards the droplet apex and a part recirculating back towards the contact line. For cases with soluble surfactants even richer behavior is observed. For a zero-pressure condition only a circulating Marangoni flow is found, but without this assumption the flow direction reverses completely with occasional secondary circulations.

The additional effects that are found when the zero-pressure condition is dropped are explained by the fact that the droplet system tends towards an equilibrium pressure. Without the zero-pressure condition, the precursor film functions as a reference pressure according to which the droplet adjusts its pressure and thus curvature. This tendency towards a ‘target curvature’ causes a fluid flow towards the droplet apex.

This qualitative difference with and without a zero-pressure condition shows that the type of precursor film one considers has significant influence on the flow behavior in evaporating inkjet droplets.

ACKNOWLEDGEMENTS: This work is part of an Industrial Partnership Programme of the Foundation for Fundamental Research on Matter (FOM), which is financially supported by the Netherlands Organisation for Scientific Research (NWO). This research program is cofinanced by Océ-Technologies B.V., University of Twente, and Eindhoven University of Technology.

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SURFACTANTS AND JETTING BEHAVIOUR IN INKJET PRINTING

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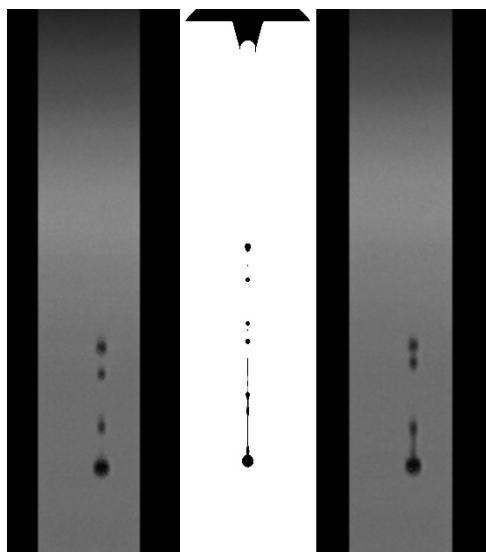
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A key challenge in developing new applications of inkjet technology is to produce inks that can be jetted to form individual droplets and to transport functional components needed for the application. The development of mathematical models that allow fluid jetting behaviour to be determined as a function of fluid properties would allow optimisation to be carried out in-silico before creating the inks and verifying the performance.

Surfactants are often added to aqueous inks in order to modify the surface tension. However, the rapid expansion of the free surface during the fast jetting process means local areas of the surface will be depleted of surfactants leading to surface tension gradients, the effects of which on ink behaviour in jetting are unknown..



In this work, experimental studies of the jetting behaviour with and without the addition of surfactants are presented.

In parallel we are developing a finite element based numerical simulation of inkjet break-up and drop formation in the presence of surfactants.

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DROPLET ELECTROWETTING IN A WEDGE GEOMETRY

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The motion of a fluid front subject to electric stresses is a fundamental aspect of the process of electrowetting. Here, we use an experimental setup consisting of a pair conducting plates that form a wedge¹ to study the response of the leading and trailing fronts of a droplet upon electrowetting actuation. We observe two distinct dynamic regimes corresponding to high and low droplet mobilities, which we model using a recently-developed lattice-Boltzmann method². Our results provide evidence that the motion of a liquid front during electrowetting exhibits dynamic transitions between two limiting regimes : a stable forced wetting regime, where the droplet has a true contact line that advances on the solid, and an entrainment regime, where a thin film of the ambient fluid prevents direct contact.

ACKNOWLEDGEMENTS: The authors thank the UK's Engineering and Physical Sciences Research Council (EP/R036837/1) and the Dutch Science Foundation NWO (VICI 11380) for supporting this work.

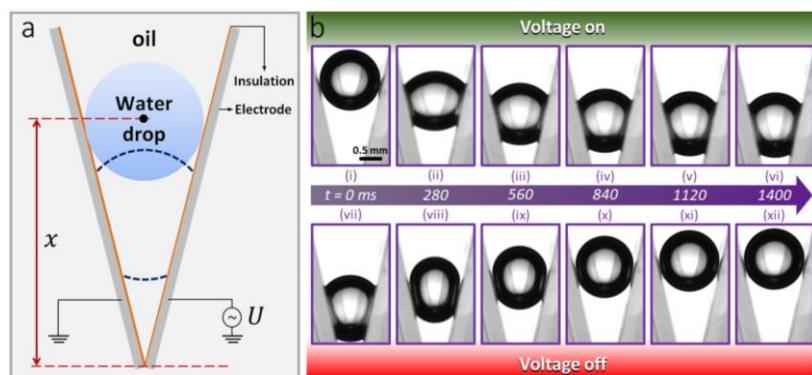


Fig. 1: Electrowetting and dewetting of a droplet in a wedge geometry

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DROPLET DYNAMICS ON ROUGH SURFACES

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We undertake a combined numerical and analytical approach to study the motion of partially wetting droplets on surfaces decorated with smoothly varying topographical features. This work is based on the associated thin-film equation for the evolution of the droplet thickness $h(\mathbf{x}, t)$, which accounts for the effects of viscous dissipation, capillarity, slip, and surface heterogeneities, cast in the form

$$h_t + \frac{\sigma}{3\mu} \nabla \cdot [h(h^2 + 3\lambda^2) \nabla \nabla^2(h + s)] = 0,$$

where μ and σ are the fluid's viscosity and surface tension, respectively, $s(\mathbf{x})$ describes the topographical features of the substrate and λ is the slip length, which allows us to alleviate the non-integrable stress singularity that would occur at a moving contact line.¹

A matched asymptotic analysis is used to extend an earlier study on two-dimensional droplets moving on rough surfaces² to the arguably more realistic three-dimensional setting. Our methodology is based on a recent work on the dynamics of droplets on ideally smooth and chemically heterogeneous surfaces and pertains in the limit $\lambda \rightarrow 0$.³ For sufficiently long timescales, the quasi-static approximation can be invoked to deduce a lower-dimensional surrogate model to describe the evolution of nearly circular contact lines.

This model constitutes a system of differential equations for the harmonics of the contact line and is reminiscent of the Cox–Voinov law¹ supplemented with higher-order corrections. Noteworthy is also that this analytical approach may be straightforwardly adapted for other contact line models, by suitably choosing the microscopic scale and formally linking it with the dynamics in the vicinity of the contact line.⁴

A number of representative cases are discussed, demonstrating that the model typically exhibits very good agreement with accurate solutions to the full problem. A hybrid numerical scheme combining the boundary integral method and the relative merits of the surrogate model is also proposed, which offers improved agreement with the predictions of the full model for strongly deformed contact lines, whilst requiring considerably fewer computing resources.

ACKNOWLEDGEMENTS: We acknowledge financial support from the Engineering and Physical Sciences Research Council (EPSRC) of the UK through the grant EP/P505453.

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MORPHOLOGY AND MECHANICAL PROPERTIES OF LIQUID-AIR INTERFACES CONFINED BY NONWETTING NANOPORES

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The entrapped air cavities in microscopic structures of superhydrophobic surfaces are well-known. Still lacking is, however, a detailed microscopic study of the morphology and mechanical properties of the liquid-air interfaces confined by these nonwetting structures and correlating them to the wetting behavior of the surface. In this study, we imaged the three-dimensional morphology of the water-air interfaces on a series of nonwetting porous surfaces (hydrophobized porous alumina with pore diameter ranging from $\sim 50\text{ nm}$ to $\sim 600\text{ nm}$) beneath a water drop using a high-resolution atomic force microscopy. It was found that water invades partly into the nanopores and the invasion depth is determined by the surface structures. The water-air interfaces are stable during imaging, though they could be deformed under increasing forces. We show that this deformation is reversible under decreasing forces. Furthermore, we determined the stiffness of the water-air interfaces from the force-distance curves and found its size-dependence, which can be correlated to the transition from bouncing to sticking of impinging macroscopic droplets on the nanoporous surfaces. The present work quantitatively resolves the morphology and mechanical property of liquid-air interface at nanoscale, which are correlated to the static and dynamic wetting of structured surfaces.

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**WETTING DYNAMICS AND THE LEIDENFROST TRANSITION
OF LIQUID DROPS IMPACTING ON A HOT PLATE**

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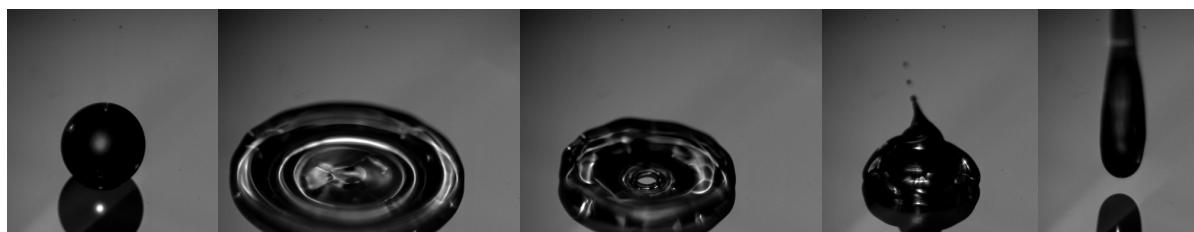
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Most of us are acquainted with the Leidenfrost effect from dancing water drops in a sufficiently hot pan: such drops levitate on a stable layer of their own vapour, thereby evaporating much slower than any contacting drop. Similar effects are highly relevant for, e.g., cooling of hot metals or, conversely, efficient drop removal for corrosion avoidance in industrial processes. In those cases, the drops will impact onto the surfaces at considerable velocities. Due to the short time scale of drop impact, the vapour layers get much thinner, leading to touchdown at increased temperatures compared to the case of gentle deposition. The determination of the *dynamic* Leidenfrost point via the final impact outcome or side view imaging becomes impossible.

We combine high-speed Total Internal Reflection (TIR) measurements with high-speed phase contrast X-ray data obtained at the Advanced Photon Source at Argonne National Laboratory to determine the dynamic Leidenfrost point for a number of liquids, which appears to be at much higher temperatures than previously anticipated. For a range of Weber number, we derive that statistically, lamella rupture is a clear signature of and initial, short-lived contact of the drop with the hot substrate. Here, hole formation is absent for all Leidenfrost drops. This finally allows us to determine the dynamic Leidenfrost point with simple shadowgraphy imaging.



Ethanol drop (2mm diam.) impacting at $v=0.89$ m/s on a hot sapphire plate (250°C). After brief contact, a central hole forms during drop retraction. Times: 0 ms, max. spreading 4 ms, visible hole at 7 ms, jet from collapse at 9 ms, rebound at 19 ms after impact.

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INFLUENCE OF SUBSTRATE ROUGHNESS ON BLOOD DROP IMPACT STAINS

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A map of passive drip stains is presented in Figure 1, the pictures of the stains are taken after drying¹. Figure 1 is therefore a visual representation showing the appearance of dried passive drip stains, from a top view, ordered according to their impact velocities and the roughness properties of the real surfaces. The surfaces correspond to those often encountered on a crime scene.

The influences of roughness and velocity on splashing and deformation of drip stains will be examined in detail. Deformation and splashing are still visible after drying. The bulging outer rim is also visible on the dried drip stains, whereas it was not visible when the stains are photographed before drying. This map shows all the noticeable features resulting from physical mechanisms and could therefore be used as a tool to classify dried drip stains patterns found on a crime scene. By photographing drip stains found on a crime scene an approximative height of fall of those stains could be deduced with a protocol that will be described during the presentation.

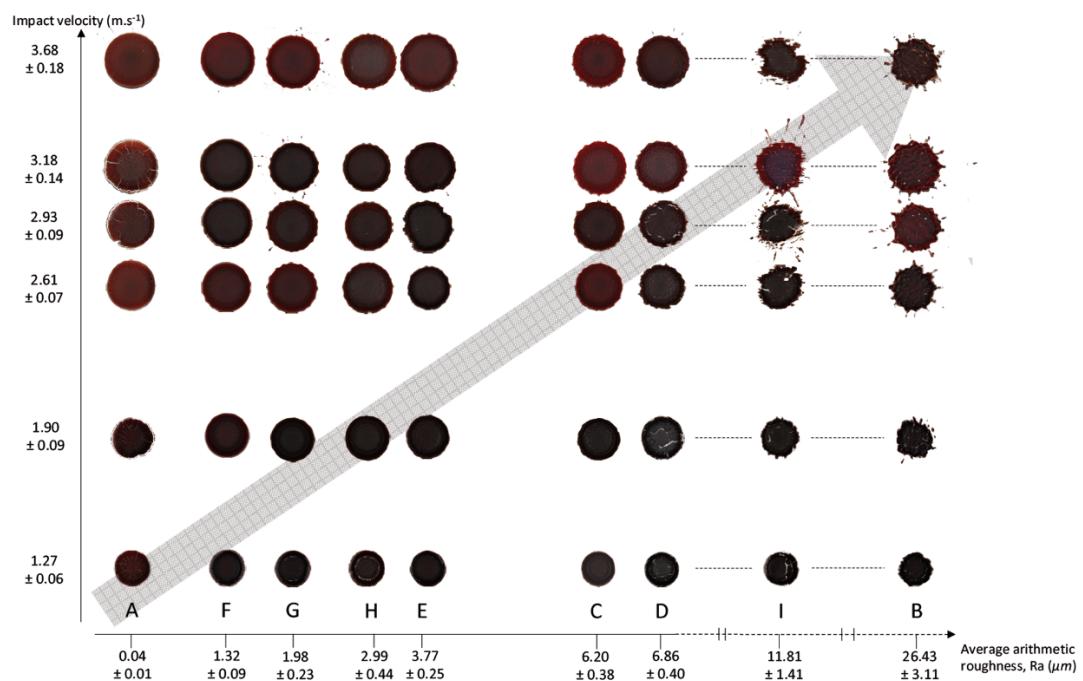


Figure 1: Recorded pictures after drying of passive drip stains of chapter 2. The bulging outer rim is visible for smooth surfaces, while deformation of the stain is increasing with velocity, from bottom to top, and with rough real surfaces (cf table 2.2), from right to left (scaled axis).

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USING WAGNER THEORY TO PREDICT EARLY-TIME JET PROPERTIES IN LIQUID-LIQUID IMPACT PROBLEMS

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Droplet impact problems have a wide range of applications throughout real-world phenomena and industrial processes ranging from inkjet printing to aerosol formation to soil erosion. Due to the violent displacement of liquid free surfaces and the associated rapid topological changes on short timescales and disparate lengthscales, impacts are notoriously difficult to model theoretically, numerically and experimentally.

With the advent of ever more powerful cameras and computers, we have a wealth of resources at hand to investigate impact phenomena in greater detail and at earlier times. However, since impact problems are highly nonlinear and complex, it is desirable to use mathematical modelling to help predict certain properties, such as the location of the root of the splash jet (or ejecta), enabling us to focus our numerical or experimental investigations on a subset of the full impact problem.

Wagner theory – an inviscid, incompressible model that neglects the roles of surface tension, gravity and the surrounding air – was developed in relation to ship-slaming applications. However, much of the model is readily extended to droplet impacts. In this analysis we perform a comprehensive Wagner analysis of droplet impacts for general droplet radii and impact speeds. We derive leading-order predictions for the location, thickness and velocity of the root of the high-speed jet, as well as predictions for the leading-order jet thickness and velocity.

We compare the predictions to direct numerical simulations performed in the open-source software package Gerris, which allows for a high-level of local grid refinement, necessary for problems with such a wide range of spatial scales. We discuss what the theory predicts well, as well as where it struggles to predict the impact dynamics. In the former cases, we discuss possible uses for the theory, while in the latter cases, we discuss extensions to Wagner theory to improve the predictions.

ACKNOWLEDGEMENTS: R.C. was partly supported by EPSRC grants EP/K041134/1 and the Mathematical Institute at the University of Oxford. R.C. also acknowledges the infrastructure support and resources provided by the Imperial College London High Performance Computing Service.

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The roles of droplet size and viscosity in binary droplet collisions

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Droplet collision is an important phenomenon in both natural and industrial processes. In most spray applications, e.g. spray drying, combustion engines, etc, droplets usually vary in size and viscosity. This work experimentally investigates the roles of droplet size and liquid viscosity in binary droplet collisions using 2% and 4% of hydroxypropyl methylcellulose solutions in water. Monodisperse nozzles were used to generate the droplets, while the collision events were captured using high-speed imaging techniques. The collision outcomes – namely bounding, coalescence, reflexive separation, and stretching separation – were mapped in the parameter space of Weber number (We) and the impact parameter (offset) for the two solutions. Surprisingly, for equal-sized collisions, the regime maps are not universal in terms of scale (droplets size). The work reported here finds that reducing the size of the droplets enhances the coalescence regime by shifting the bouncing regime towards lower We , the reflexive separation regime toward higher We , and the stretching separation regime toward higher impact parameter. This is attributed to the fact that, for a given value of We , the smaller droplets have lower Reynolds number thus it is more difficult for them to achieve separation compared to the larger droplets. Moreover, due to higher capillary pressure, the small droplets have higher penetration potential through the air layer that causes the bounding, and hence easier coalescence. Collisions of droplets with a size ratio of 0.6 shift the separation regimes even more than the small droplets collisions. This is attributed to the break in the dynamics' symmetry, which leads to more mixing and hence more viscous dissipation. Increasing the viscosity also enhances the coalescence regime by suppressing the separation regimes. Thus, in applications where the droplets are relatively small and possess high viscosity the coalescence regime is expected to dominate collision behaviour.

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SPRAYS FROM DROPLETS IMPACTING A MESH

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In liquid spray applications, the sprays are often created by the formation and destabilization of a liquid sheet or jet. The disadvantage of such atomization processes is that the breakup is often highly irregular, causing a broad distribution of droplet sizes. As these sizes are controlled by the ligament corrugation and size, a monodisperse spray should consist of ligaments that are both smooth and of equal size. A straightforward way of creating smooth and equally sized ligaments is by droplet impact on a mesh. In this work we show that this approach does however not produce monodisperse droplets, but instead the droplet size distribution is very broad, with a large number of small satellite drops. We demonstrate that the fragmentation is controlled by a jet instability, where initial perturbations caused by the injection process result in long-wavelength disturbances that determine the final ligament breakup. During destabilization the crests of these disturbances are connected by thin ligaments which are the leading cause of the large number of small droplets. A secondary coalescence process, due to small relative velocities between droplets, partly masks this effect by reducing the amount of small droplets. Of the many parameters in this system, we describe the effect of varying the mesh size, mesh rigidity, impact velocity, wetting properties, keeping the liquid properties the same by focusing on water droplets only. We further perform Lattice Boltzmann modeling of the impact process that reproduces key features seen in the experimental data.

ACKNOWLEDGEMENTS: This work is part of the Industrial Partnership Program Hybrid Soft Materials that is carried out under an agreement between Unilever Research and Development B.V. and the Netherlands Organisation for Scientific Research (NWO). A.M.M., D.D., and J.C. acknowledge the support by the Swiss National Science Foundation (Project no. 200021 175793). The computational resources were provided by the Swiss National Supercomputing Center (CSCS) under project number s823. The authors thank Emmanuel Villermieux for fruitful discussions.

Levitating droplet over a moving wall: mechanism and position control

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When a droplet is deposited onto a moving wall, the droplet can steadily levitate (Ref. 1). An air film between the moving wall and the levitating droplet plays an important role in the steady levitation since lubrication pressure generated inside the air film sustains the droplet's weight. We investigate the steady air film in order to clarify the mechanism of the droplet levitation over the moving wall. First, we discuss whether lubrication theory can be basically applied to the air film for the calculation of lubrication pressure. Pressure distributions and flow fields inside the air film are computed by numerical simulations under two kinds of governing equations. These results verify that lubrication theory can be applied due to negligible inertia force inside the air film. Next, we consider a typical process of determining the steady shape of the air film based on the local pressure balance at the droplet's bottom. In addition, we used our figures to control the position of the droplets as shown in Figure 1. We systematically investigate the mechanism of this method.

ACKNOWLEDGEMENTS: This work was supported by JSPS KAKENHI grant no. 17H01246.

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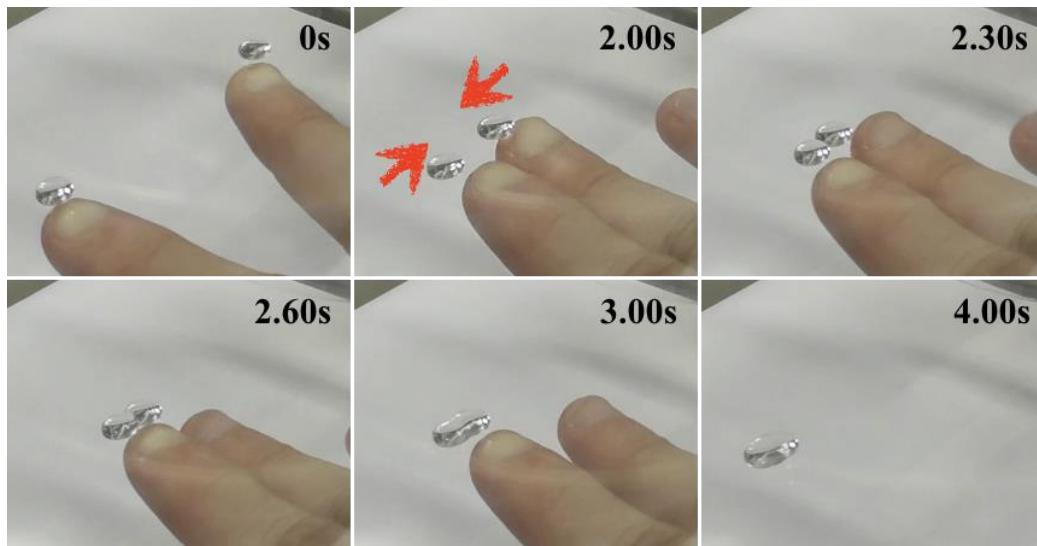


Figure 1. Position control of levitating droplets by fingers.

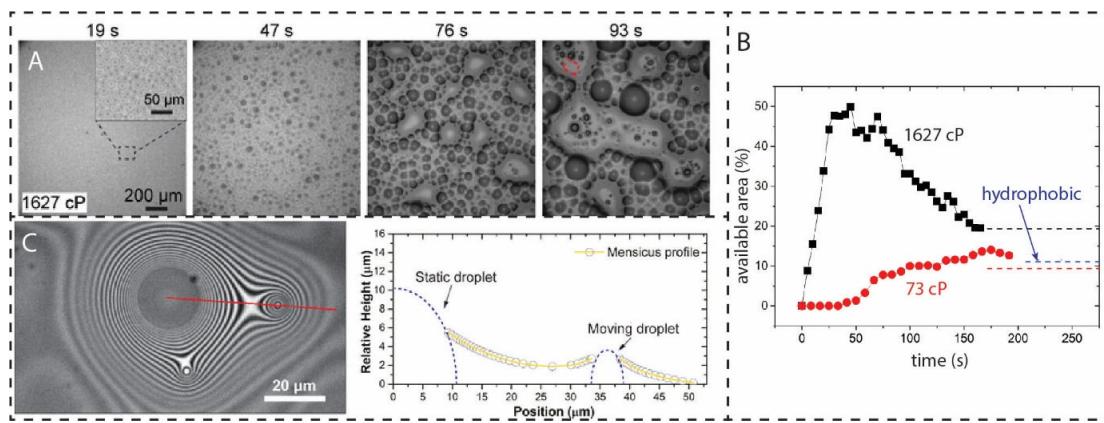
CAPILLARY-INDUCED DROPLET MOBILITY DURING CONDENSATION ON THIN LUBRICANT FILMS

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Condensation is ubiquitous in nature in many industrial applications, including water harvesting, power generation, and thermal management of power electronics and medical equipment. As compared to traditional filmwise condensation, dropwise condensation on lubricant-infused surfaces (LIS) can lead to an order-of-magnitude increase in heat transfer rates.¹ Small droplets ($D \leq 100 \mu\text{m}$) account for nearly 85 % of the total heat transfer and droplet sweeping plays a crucial role in clearing nucleation sites, allowing for frequent re-nucleation. We showed previously that the droplet size distribution during condensation on LIS follows that of droplets condensing on solid hydrophobic surfaces for $D \geq 10 \mu\text{m}$, but is higher for smaller droplets.^{2,3} Here we show using high-speed microscopy and interferometry that the nucleation rate density is more than an order of magnitude higher on LIS than on solid hydrophobic surfaces. Furthermore, we show that the initially uniform lubricant film redistributes during condensation, resulting in lubricant-rich and lubricant-poor regions (fig. A). While droplet nucleation is limited to lubricant-poor regions (Fig. B), their high mobility compensates for the reduced area-availability. Governed by lubricant height gradients (Fig. C), microdroplets as small as 2 μm in diameter undergo rigorous and gravity-independent self-propulsion, travelling distances multiples of their diameters at velocities up to 1100 $\mu\text{m}/\text{s}$. Although macroscopically the movement appears to be random, we show that on a microscopic level capillary attraction due to asymmetrical lubricant menisci causes this gravity-independent droplet motion. Combining experiments and modeling, we show that the maximum sliding velocity is inversely proportional to the lubricant viscosity and is strongly dependent of the droplet sizes. This novel and non-traditional droplet movement is expected to significantly enhance the sweeping efficiency during dropwise condensation, leading to higher nucleation and heat transfer rates.



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The speed of rolling droplets

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We analyze the near-rolling motion of two-dimensional nonwetting drops down a gently inclined plane. Inspired by the scaling analysis of Mahadevan and Pomeau [[Phys. Fluids 11, 2449 \(1999\)](#)], we focus upon the limit of small Bond numbers, $B \ll 1$, where the drop shape is nearly circular and the internal flow is approximately a rigid-body rotation except close to the flat spot at the base of the drop. Our analysis reveals that the leading-order dissipation is contributed by both the flow in the flat-spot region and the correction to rigid-body rotation in the remaining liquid domain. The resulting leading-order approximation for the drop velocity \mathcal{U} is given by $\mu\mathcal{U}/\gamma \sim \alpha/2B \ln \frac{1}{B}$, wherein μ is the liquid viscosity, γ the interfacial tension, and α the inclination angle.

DROPLET DYNAMICS ON CONICAL SUBSTRATES

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Droplet motion on conical substrates is a phenomenon observed extensively in nature and has since been adopted in industrial applications. It has been suggested as a mechanism responsible for: dew drops forming on spider webs¹, aiding cacti to remain hydrated in the desert² and even repelling water from the legs of the common water strider³. This motion has been exploited in the petroleum industry where micron sized droplets of oil can be removed from water using arrays of cones⁴. Surfaces with conical micro-posts have shown promising results in enhancing water repellent surfaces⁵. Despite these promising technologies there has not until very recently been interest in the dynamics of droplet on such surfaces.

We study, both experimentally and theoretically, the dynamics of droplets on conical surfaces. The curvature gradient of the surface induces a pressure gradient that leads to spontaneous propulsion of the droplets. Normally, contact line pinning would prevent or impede this effect. However, by lubricating the substrate the effects of pinning are reduced and the droplet is free to move. We derive the curvature induced force acting on the droplet and balance this with the viscous dissipation occurring in the lubricating wedge. We find that at suitable late times, the droplet's displacement from the apex of the cone, $s \sim t^{1/4}$ and verify this prediction experimentally.

ACKNOWLEDGEMENTS: The authors thank the Royal Society and European Research Council for supporting this work.

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FILLING THE GAP IN BOUNCING DYNAMICS

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Depending on physical parameters, impacting bodies on a liquid surface have been shown to sink, become trapped or rebound off the interface [1]. Here we focus on the latter of these scenarios (see Fig. 1 below), when surface tension plays an important role, driving the dynamics towards non-trivial regimes of relevance to many drop-bath and fluid-structure interaction contexts.

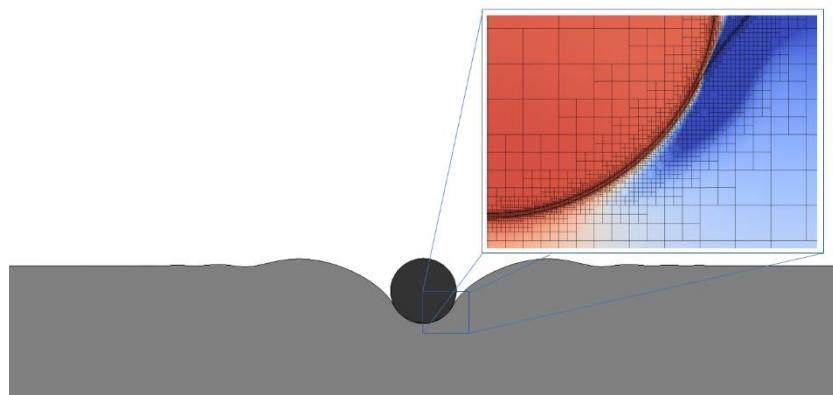


Figure 1. Direct numerical simulation of the impact process, with the inset highlighting the vertical velocity and underlying adaptive mesh structure.

A partial differential equation model starting from the Navier-Stokes equations can be derived to represent the motion of the free surface [2]. This reduced linear model is coupled to the motion of the impactor (a superhydrophobic solid sphere in the first instance), yielding promising results when compared to experiments without the need for fitting parameters. By construction however the model ignores nonlinear effects and air cushioning.

In this work we present a synergistic approach between a generalised modelling framework, state-of-the-art direct numerical simulations based on the volume-of-fluid method, as well as recent experimental advances in drop generation [3]. These allow us to explore a wide parameter spectrum and characterise the dependence of the normal coefficient of restitution and contact time on the impact velocity, radius, and relative densities of the impactor and liquid pool in both solid-liquid and liquid-liquid cases. The combined efforts are directed towards enhancing predictive capabilities and gaining a detailed understanding of this challenging multi-scale impact problem.

ACKNOWLEDGEMENTS: The authors would like to thank the EPSRC (project EP/N018176/1) and the Brown OVPR Seed Award for financial support for the experiments.

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DEWETTING OF POLYMER MICRODROPLETS WITH STRONG SLIP.

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Classical hydrodynamic models predict that infinite work is required to move a three-phase contact line over a the surface of a solid. Assuming a slip boundary condition, in which the liquid slides against the solid, such an unphysical prediction is avoided. Motivated by the results of dewetting experiments with spherical cap-shaped polystyrene microdroplets [1], we study the relaxation of the droplet shape to its new equilibrium using a boundary integral approach [2]. Besides a velocity independent microscopic contact angle we assume that the dynamics of the liquid is described by the steady Stokes equation with a Navier slip boundary condition at the solid. We find that slip has a strong influence on the evolution of the droplet contour, both on the transient nonspherical shapes and the contact line dynamics. The scaling of the contact line radius at early and late time observed in our numerical solutions are in good agreement with scaling predictions for the evolution of the droplet contour as well as the experimental results .

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A LATTICE-BOLTZMANN MODEL OF ELECTROCAPILLARITY

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Dielectrophoresis and electrowetting have become widely used techniques for controlling and manipulating small amounts of liquids. Applications of dielectrophoresis include the transport, and separation of liquids and other particles of different electric permittivity. Examples of electrowetting applications are electronic paper displays, adjustable lenses, and lab-on-a-chip devices.^{1,2} In both cases, the underlying phenomena can be encompassed under electrocapillarity, the interaction of electric fields with multiphase systems where the effects of surface tension are comparable with electrostatic forces. Usually, one of the phases is a conducting liquid surrounded by an ambient dielectric or two liquids of different electric permittivities. Fundamental aspects in electrocapillarity are still open for investigation; this mainly concerns the dynamical aspects, for example, the motion of contact lines and the shape that the liquid interface acquire in the presence of electric stresses.^{2,3} This is important since most of the applications rely a precise control of the liquids and by placing electrodes in clever arrays. Simulations on electrocapillarity are timely as they may provide insights to address these aspects. In this contribution, we propose a simple lattice-Boltzmann method that is capable of simulating electrocapillarity. We use a binary fluid model that includes capillary phenomena and extend the algorithm to include the forces produced by electric fields. The electric field and the charges are derived from a potential function, which we obtain by a relaxation method.⁴ We first validate our method by comparison against the experimental observations. Then, we examine the morphology of droplets under dielectrophoretic stresses.

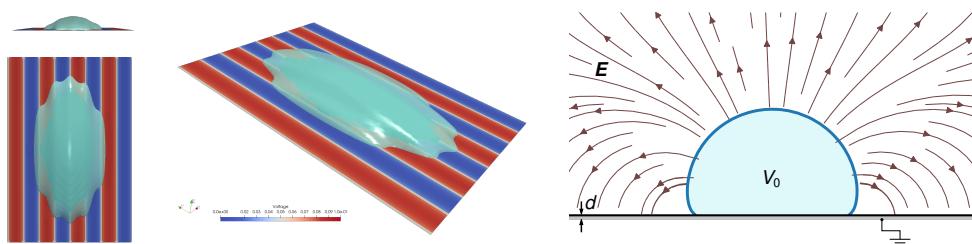


Figure 1: Example of simulations of droplet spreading by dielectrophoretic forces (left) and electrowetting (right).

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INTERFACIAL PARTICLE ACCUMULATION IN THE DRYING-TEARDROP EFFECT

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Salts can be found in different forms in almost any evaporating droplet in nature, our homes and in laboratories. The transport processes in such - apparently simple- systems differ strongly from 'sweet' evaporating droplets¹ since the liquid flows in the inverse direction due to Marangoni stresses at the liquid-gas interface. The inverted flow pattern that takes place in such salty droplets is revealed using 3D particle tracking measurements to quantify the full three-dimensional flow in evaporating droplets of sodium chloride². Contrary to what is typically reported, the flow inversion does not prevent the coffee-stain effect; on the contrary, particles accumulate, get trapped in the vicinity of the liquid-gas interface and are consequently advected towards the contact line along the interface. In this work, we make use of confocal imaging to quantify the accumulation process and the growth of the particle interfacial deposits for different salt concentrations along the droplet's interface. The experimental results are contrasted with numerical simulations that capture the solvent evaporation, the evaporation-induced liquid flow and the quasi-equilibrium liquid-gas interface.

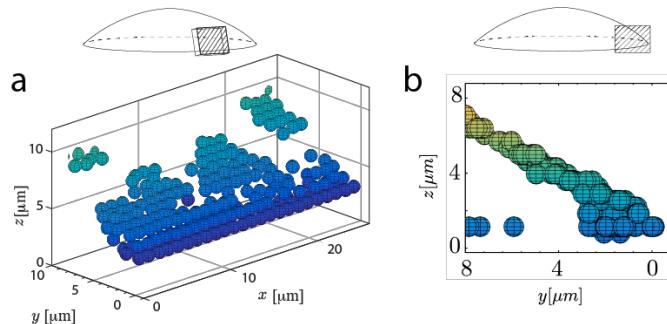


Figure 1. Confocal reconstructions of 1- μm -diameter polystyrene particles accumulating at the interface of a salt solution droplet (initial concentration NaCl 5mM) during its evaporation.

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Evaporation processes of sessile droplet and liquid film: Research from the ground to space

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Abstract

The evaporation processes on the ground and in space have recently attracted more scientific interests owing to engineering applications as efficient ways of heat management. In space, the phase change coupling with typical interfacial phenomena of heat and mass transfer will play main role in the process of evaporation, while which are still absent of comprehensive understanding in space where the influence of gravity (i.e. natural convection, buoyancy) can be minimized. In present paper, both droplet evaporation and liquid film evaporation are investigated on the ground and in space by our research team in recent 10 years. We got the space experimental results of droplet and film evaporation in microgravity condition in recent two space flight missions of Chinese Satellite SJ10 and 1st Cargo Spacecraft TZ-1 launched in 2016 and 2017, respectively. The compared results showed that due to the gravity-free constraints, the height of droplets in space would be higher than that on ground for same injection volume. The average evaporation rate of FC-72 droplet in space is much slower than the ground tests in the same working conditions (temperatures, volume and pressures), which was thought of the large contribution of buoyancy convection to evaporation on the ground. On the ground, we performed an experimental study of thermocapillary convection of a ~2mm deep layer of 0.65cSt silicon oil confined in an open rectangular pool. The evaporation rate and the thermal field at evaporating interface were examined by the laser co-focal displacement meter and the infrared thermography. Depth of the liquid layer maintained quasi-changeless with the evaporation rate and liquid-injection rate approximately equal. According to its instability to hydrothermal waves and a steady multicellular, the evaporation processes are divided into three stages. Thermal instabilities at the evaporation interface are shown in Figure 1 for three different stages. Comparative experiments were carried out with different working fluids (FC-72 and HFE7500).

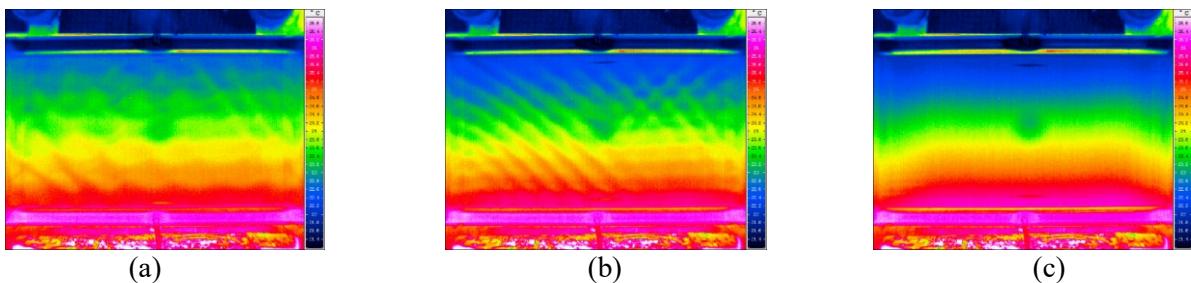


Figure 1: Thermal field of three different stages: (a) Oscillatory multicellular; (b) Hydrothermal waves; (c) Static thermal convection

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COMPETITIVE EVAPORATION OF MULTIPLE DROPLETS

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An asymptotic model is derived for the competitive diffusion-limited evaporation of multiple thin sessile droplets under the assumption that the droplets are well separated. Comparisons are made with numerical solutions of the full governing equations in order to verify the accuracy of the model; in particular, the model is found to perform well even outside its anticipated range of applicability, up to and including the limit of touching droplets. The "shielding" effect" of other droplets is demonstrated, and the model is used to investigate the effect of this shielding on droplet evolutions and lifetimes, as well as on the coffee-ring effect.

Interface dynamics of nano-filaments

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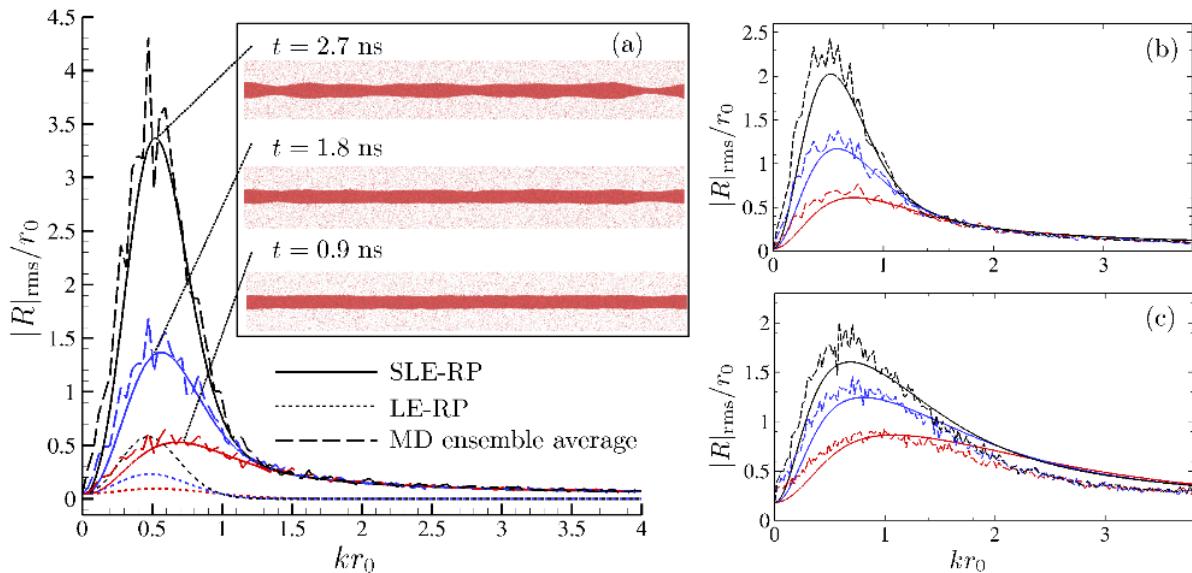
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Interface dynamics of liquid nano-filaments and their subsequent disintegration into droplets are key elements in many future nano-technologies. In this work, they are studied by molecular dynamics simulations (MD) and stochastic lubrication equations (SLE). In the linear stage, MD simulations are performed that clearly indicate that the classical Rayleigh-Plateau (RP) instability mechanism is violated at the nanoscale. A new theoretical framework based on the SLE is developed that captures MD data and explains why (i) the classical model fails to predict the dominant disturbance mode on interfaces (Rayleigh's result) and (ii) the sharp stability criterion (Plateau's result) is lost. For nonlinear dynamics study, a computational approach is introduced to solve the SLE numerically, which is validated by MD. The SLE simulation is used to investigate the limits of applicability of a similarity solution proposed for this system.

ACKNOWLEDGEMENTS: This work was supported by the Leverhulme Trust (Research Project Grant) and the EPSRC (grants EP/N016602/1, EP/P020887/1 & EP/P031684/1).

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A Decomposition of Droplet Simulation Using Molecular Dynamics

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Nowhere in fluid dynamics do the limits of continuum computational fluid dynamics (CFD) modelling become more apparent than a droplet on a surface. It is here that the application of molecular dynamics (MD) can provide unique insights. Although it is possible to model droplets at the molecular scale, as in Fig 1a, it is not clear if MD simulation of these nano-droplets will ever be large enough to behave in a similar manner to their macroscale counterparts.

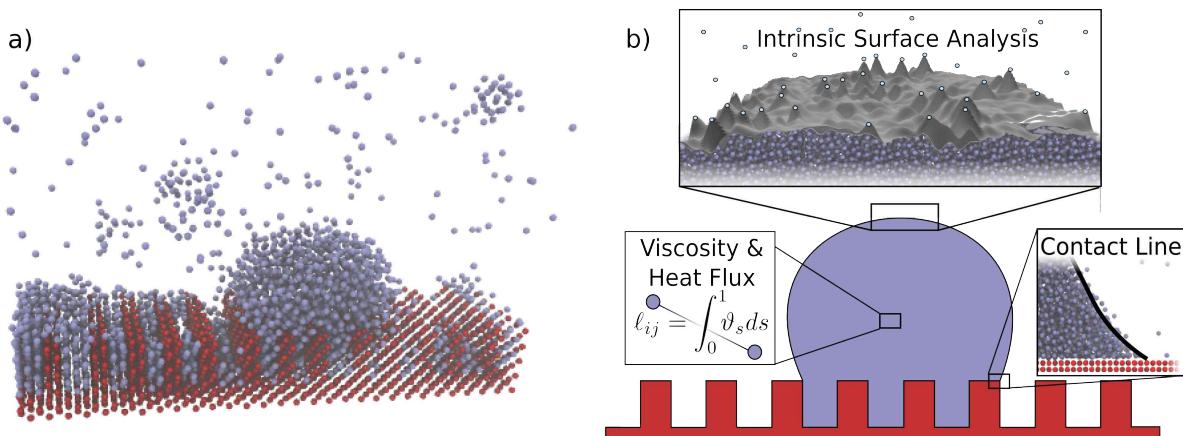


Figure 1 - a) Full MD droplet and b) multiscale model using MD for key details

As the most fundamental classical method, MD captures the full underlying structure of the fluid so that complex phenomena are reproduced with no additional models. In this talk we present molecular models for each of the key physical processes in an evaporating droplets, Fig 1b, including the moving contact line between liquid, vapour and gas [1], the liquid-vapour interface [2] and the bulk fluid properties [3]. We discuss how we use MD to extract the key empirical coefficients for use in CFD, including surface tension, Fourier's law, viscosity and a model for the contact line.

However, the holy grail for multi-scale modelling is to bring these various cases together in a single model which uses molecular detail only where it is needed. We finish with a discussion of progress in this direction [4], and discuss domain decomposition coupling with specially designed software, www.cpl-library.org, to link MD and CFD together for multiphase flow.

ACKNOWLEDGEMENTS: E. R. Smith was supported by EPSRC Grant EP/L020564/1

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A NEW LATTICE BOLTZMANN APPROACH TO THIN FILM HYDRODYNAMICS

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We propose a novel approach to the numerical simulation of thin film flows, based on the lattice Boltzmann method [1,2,3]. We outline the basic features of the method and show in which limits the expected thin film equation is recovered. To validate our method we perform several benchmark cases including the evolution of the Rayleigh-Taylor instability, the relaxation of a droplet to its equilibrium contact angle and the sliding of a droplet on an inclined plane. In Fig. 1 we show rendered data of an out of equilibrium droplet. The relaxation follows the predicted behaviour according to the Cox-Voinov law [4], see Fig. 2. Concerning the sliding we observe a linear relation between the capillary and Bond number which is in agreement with theory and experimental observations.

Finally, we will address the effect of thermal fluctuations on thin film dewetting. We will show preliminary results on the early time evolution of the spectrum of height fluctuations in a spinodally dewetting film, and on the morphology of dewetting patterns [5].

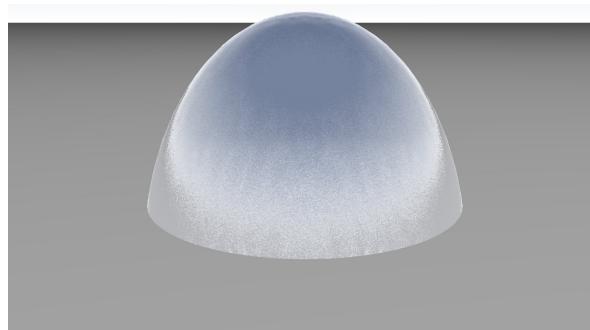


Figure 1. Initial droplet shape, rendered data

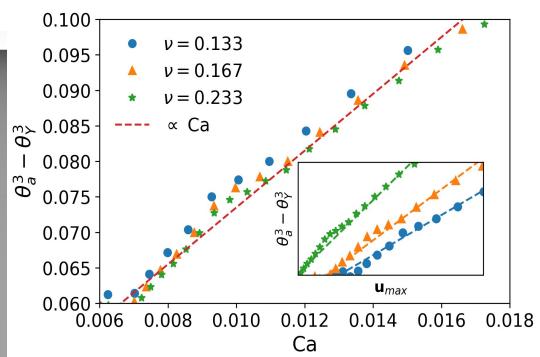


Figure 2. Cox-Voinov spreading law

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THERMOCAPILLARY-DRIVEN FLOWS IN PURE WATER DROPS ON A LOCAL HOT-SPOT

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Phase change and flows in liquid drops are omnipresent in nature and fundamental in industrial processes such as inkjet printing, coating, biomedical analysis as well as thermal management. Considering the progression of densely-integrated microelectronic chips, drop-based cooling is a potential technique for efficient heat removal due to phase change heat transfer. Conventionally, many experimental/numerical studies have investigated water or organic drops on uniformly-heated substrates, while high heat fluxes in local areas i.e. hot-spots are often observed in aforementioned problems. In the present paper, combining infrared thermography with particle imaging, we present thermal and flow visualisation of pure water drops evaporating on a hot-spot. The hot-spot was produced by locally heating the substrate directly below the centre/edge of the drop using a laser. When heating the centre of the drop, a ring-like hot region appeared along the periphery, eventually breaking into two counter-rotating vortices which travelled azimuthally. As heating went on, the vortices began to merge and split in an oscillatory manner. On the other hand, edge heating resulted in more directional vortices with a distinct oscillatory pattern: rather, the vortices swung around the main heat conduction path (from the heating spot to the other side). The origin of these patterns was identified by calculating and comparing the dimensionless Marangoni and Rayleigh numbers, which showed the dominance of the Marangoni effect. This fact was further supported by a second set of experiments where the same flow patterns were observed when the system was inverted i.e. a pendant drop heated from above. Besides heating locations, the effect of heating power on the strength of Marangoni instabilities was also discussed.

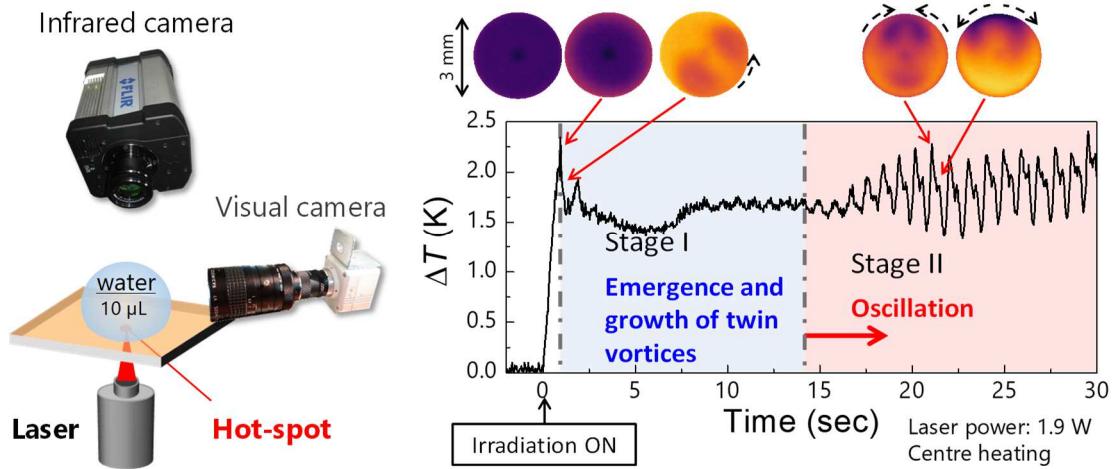


FIG. Schematic of experimental setup (left) and representative result for the mid-power centre-heating case (right). The chart presents the temporal evolution of the maximum temperature difference obtained from the IR images.

DROPLET PROPULSION AND DIRECTION CONTROL ON A PLANAR SURFACE USING A SELECTIVE LEIDENFROST EFFECT

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The Leidenfrost effect is a bulk effect, involving the super-heating of fluids to the point whereby they are suspended by their own vapour layer and thus thermally insulated from the heat source below, thereby reducing the evaporation rate. The bulk nature of this effect means that its properties have not been used in many practical applications on the scale of droplets due to the high energy input required. Previous work has shown that the Leidenfrost effect can be achieved selectively on a planar surface using micron-scaled features, which are created photolithographically, providing energy savings at the micro-scale, macro-scale and also in the time domain via current pulsing¹. In the macro scale, Leidenfrost propulsion has been achieved by super-heating brass structures in the form of herringbones², ratchets³ and herringbone-ratchets⁴. By taking the original serpentine patterns, used to selectively heat the planar substrate to create the Leidenfrost effect, and positioning them in to a herringbone structure to replicate herringbone² and herringbone ratchet⁴ structures, droplet propulsion and directional control can be achieved using a selective Leidenfrost effect. In this work, instead of using physical structures to control the vapour flow under the droplets, the selective heating of a planar surface uses thermal gradients to direct the vapour flow. Furthermore, the central region of the design has been modified to provide a lower current density region, which makes the centre of the Leidenfrost region cooler than the regions on either side, thus trapping the fluid droplet in the centre of the pattern. Interestingly, the direction of propulsion is in the opposite direction to that of the macro brass structure. It is believed that the hot regions directly above the serpentines have thicker vapour regions and convection occurs between the heated and unheated regions, causing the vapour flow to occur in parallel to the direction of the serpentines.

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Quantized effective viscosity of dense monodisperse emulsions in microchannels

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The rheology of pressure-driven flows of dense monodisperse emulsions in microchannels is investigated by means of mesoscopic lattice Boltzmann simulations, capable of handling large collections of droplets, in the order of several hundreds. The simulations reveal that the fluidisation of the emulsion is quantized, i.e. it proceeds through a sequence of discrete steps, characterized by yielding events whereby layers of droplets start rolling over each other, thus leading to sudden drops of the effective viscosity. It is shown that such a discrete fluidisation is robust against loss of confinement, namely it persists also in the regime of small ratios of the droplet diameter over the microchannel width. We also develop a simple phenomenological model which predicts a linear relation between the effective viscosity of the emulsion and the product of the confinement parameter (global size of the device over droplet radius) and the viscosity ratio between the disperse and continuum phases. The model shows excellent agreement with the numerical simulations.

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CONTROLLED SHAPING OF SESSILE MAGNETIC DROPLETS

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Controlled shaping of droplets is used in digital microfluidics to manipulate droplets for mixing and sensing [1]. Droplets can be shaped through the application of electromagnetic fields, which exert a force on ions or electric and magnetic dipoles in the droplet. This force can be calculated using the electromagnetic stress tensor, which is a powerful tool derived from first principles of electromagnetism and thermodynamics [2]. The electromagnetic stress tensor in vacuum has been applied to a limited set of well-defined problems such as electrowetting, a technique where surface energies of the substrate are electrostatically modified [3].

Electric actuation techniques are well-studied and widely adopted [1]. In contrast, relatively little work exists on magnetic actuation techniques, such as the actuation of paramagnetic salt solutions, which has been demonstrated on superhydrophobic surfaces [4]. Since magnetic actuation techniques require the presence of magnetic dipoles, they open a range of new possibilities for sensing and measurement applications.

Here, we explicitly apply the electromagnetic stress tensor to a sessile droplet in a homogeneous magnetic field and derive a relationship for the change in droplet shape as a function of applied magnetic field. We validate this relationship experimentally, by measuring the change in shape of a paramagnetic sessile droplet in a magnetic field and showcase its potential by demonstrating lateral movement and mixing of paramagnetic droplets [5]. Our results demonstrate the controlled shaping and manipulating of magnetic droplets while highlighting the fundamental physics involved in these interactions.

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SEPARATION OF VAPOR FROM LIQUID IN ELECTRICALLY DRIVEN LIQUID FILM FLOW BOILING

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The enhancement of pool boiling, evaporation, and condensation heat transfer is of crucial importance to many industries, including the heating ventilating air conditioning (HVAC), power, process, electronics, and aerospace industries. A variety of enhancement techniques have been researched and implemented to a limited range of industrial applications. These techniques are commonly referred to as passive and active means of heat transfer enhancement.

Electrohydrodynamic (EHD) phenomenon involves the interaction between flow fields and electric fields in a dielectric fluid medium. EHD conduction pumping is primarily driven by the Coulomb force acting on free space charges which are redistributed to the vicinity of the energized electrodes. Free charges are formed due to the imbalance in the dissociation and recombination of neutral electrolytic species in the dielectric fluid. Proper asymmetric design of the electrodes generates net axial flow motion, pumping the fluid [1]. EHD conduction pumps can be used as the sole driving mechanism for small-scale heat transport systems and have a simple electrode design, which allows them to be fabricated in exceedingly compact form (down to micro-scale). EHD conduction is also an effective technique to pump a thin liquid film in the presence and absence of gravity.

Dielectrophoresis is a translational motion of neutral matter in a non-uniform electric field [2]. The non-uniform electric field results in field induced polarization of vapor bubbles or droplets in the medium. Unlike the Coulomb force (which acts on free charges), the DEP force acts on the polarized charges and can be used to influence vapor bubble motion during nucleate boiling or droplet motion during condensation.

This presentation will provide the experimental results of active augmentation of heat transfer with the application of external electrical fields in boiling. Specifically, EHD conduction pumping is combined with dielectrophoresis extraction force in liquid film flow boiling [3]. The results show remarkable enhancements exceeding 1000% in boiling heat transfer coefficient at a given superheat when both mechanisms are used simultaneously. The experimental data are important for applications in thermal management in terrestrial and space conditions. Select results in the absence of gravity will be presented as well.

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Self-propelling Droplet Shells Stabilized by Liquid Crystal Topology

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Liquid shells e.g. double emulsions, vesicles etc. are susceptible to interfacial instability and rupturing when driven out of mechanical equilibrium. This poses a significant challenge for the design of liquid shell based micro-machines, where the goal is to maintain stability and dynamical control in combination with motility. Here we present our solution to this problem with controllable self-propelling liquid shells, which we have stabilized using the soft topological constraints imposed by a nematogen oil. We demonstrate, through experiments and simulations, that anisotropic elasticity can counterbalance the destabilizing effect of viscous drag induced by shell motility, and inhibit rupturing. We analyse their propulsion dynamics, and identify a peculiar meandering behaviour driven by a combination of topological and chemical spontaneously broken symmetries. Based on our understanding of these symmetry breaking mechanisms, we provide routes to control shell motion via topology, chemical signalling and hydrodynamic interactions

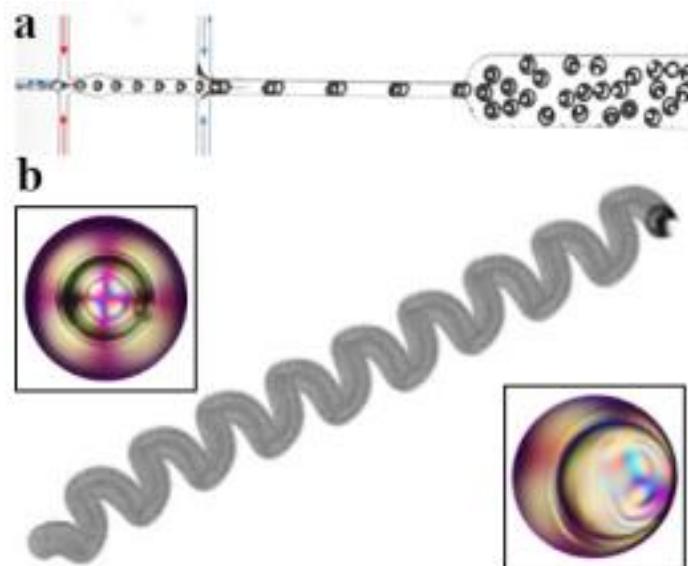


Fig. 1. **a.** Microfluidic production of monodisperse double emulsions in a flow focusing cross-junction configuration. **b.** Shark-fin meandering of a swimming shell. Top-left and bottom-right insets show example images of shells under cross-polarized microscopy before and during swimming respectively.

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EVAPORATION OF A SINGLE DROPLET IN A CIRCULAR WELL

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When a coffee drop dries on a surface, it leaves behind a solid deposit along the edge. The coffee particles which are initially uniformly dispersed throughout the entire drop, becomes concentrated near the edge, giving rise to the well-known ‘coffee-ring effect’¹. Such a ring deposit can occur in any type of solution or colloidal suspension of particles used in many industrial applications. In the coating and printing industry, the challenge is to prevent the formation of ring-like deposit in order to obtain uniform coatings or homogeneously dried ink drops.

The principles behind the evaporation of droplets, which play a key role in inkjet printing used in the manufacture of OLED displays, must be understood in order to control and predict the deposits left behind when a drop is printed onto a patterned substrate.

Single droplets of solvents with different volatility were printed inside a circular well and the influence of substrate temperature on droplet drying profile was investigated. An interferometry technique was used to provide real-time information on the changing droplet profile during drying. Droplets of different solvents printed in circular wells showed different behaviour during drying. The shape of the droplet profile (U- or W-shape) was found to be influenced by the physical properties of the solvents and by the aspect ratio of the well. Breward et al.² have recently developed a lubrication model to describe the evaporation of a liquid droplet inside a well. The model identifies a single dimensionless parameter which depends only from the capillary number and the aspect ratio of the well and discriminates between different drying regimes. The experimental data of single droplets evaporation, acquired for different solvents and different temperatures of the substrate, will be used to verify the correspondence between the dimensionless parameter and the different drying regimes.

ACKNOWLEDGEMENTS: The author thanks EPSRC and Merck Chemical Ltd for supporting this work.

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SELF AMPLIFYING CRYSTALLIZATION IN THIN LIQUID FILMS

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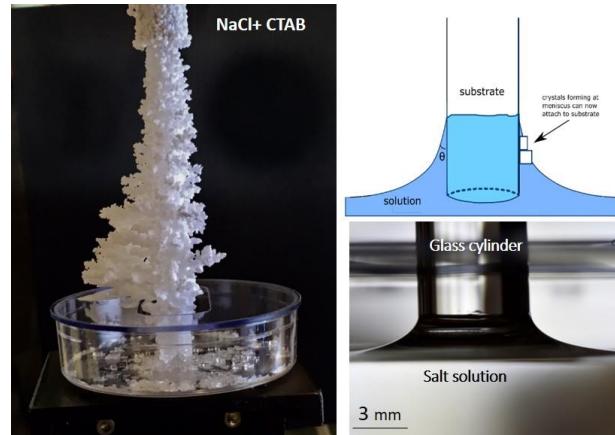
Salt creeping consists of the precipitation and growth of crystals from evaporating salt solutions far from the solution boundary, i.e. the contact line with a solid surface. It is both a fascinating phenomenon and a major nuisance whenever salt solutions are present and evaporate. The latter can cause weathering of artworks, sodification of soils and corrosion of outdoor electronics. Indeed, because of the spreading of salt solution due to creeping, even a well-protected surface can corrode in a very short time. Even though salt creeping occurs quite commonly, the mechanisms involved in the process are still poorly understood^{1,2} and a quantitative description of the creeping mechanism and its universality with respect to different salts is still missing.

We have investigated the dynamics of vertical salt creeping on glass substrates in contact with different aqueous salt solutions during evaporation. With our novel experimental approach⁴, we are able to quantify the time evolution of the contact angle of the salt solution meniscus, as well as the amount of crystals growth (i.e. creeping) on the solid substrate.

We show that there exists a critical contact angle below which salt creeping occurs, provided also the nucleation of multiple crystals is favoured. The precipitation of new crystals happens ahead of the contact line by the meniscus that progressively advances over the crystals forming also nanometric precursor films. High-resolution Scanning Electron Microscopy reveals the existence of nanocrystals very far from the macroscopic limit of the creeping zone.

The increase of the evaporative area in this way, causes an exponential increase of the crystals mass in time. The self-amplifying process then results in spectacular three-dimensional crystals at macroscopic distances far from the solution.

These findings also allow us to control the creeping by using surfactants as crystallization modifiers. We show that the addition of surfactants to the salt solution have an opposite influence on creeping dictated by their role as crystallization modifiers rather than their impact on the wetting properties:



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SPREADING AND CONTRACTING THREE-COMPONENT DROPLETS

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When a binary droplet is placed on a surface, inhomogeneous evaporation (1) can cause surface tension gradients that lead to enhanced spreading (2) or droplet formation, even on a fully wetting surface (3). Introduction of additional components can lead to much richer dynamics as seen in Ouzo and Whiskey droplets. (4,5).

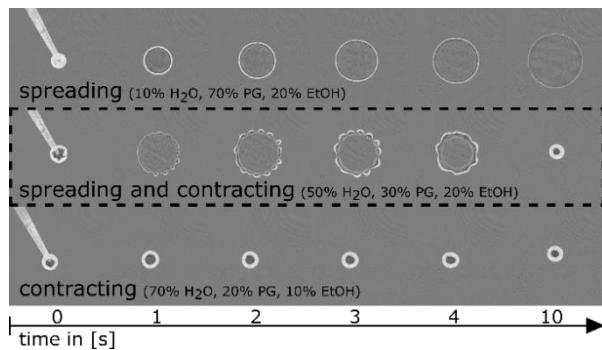


Figure 1: Timeseries of the three different dynamics observed in ternary droplets. We focus on the spreading and contracting dynamic only seen in ternary droplets.

Here we explore the Marangoni convection of a fully miscible, three-component system of water, ethanol, and propylene glycol on a completely wetting substrate. Evaporation of ethanol (most volatile and lowest surface tension) results in a higher surface tension around the perimeter of the droplet and causes rapid spreading. After ethanol has mostly evaporated, water evaporation (next most volatile component and highest surface tension) from the perimeter reverses the surface tension gradient. This induces a reversal in the direction of Marangoni stress and causes the droplet to contract.

We investigate the ternary parameter space of compositions at different relative humidities to understand how the interplay of Marangoni flow, capillary flow, and evaporation give rise to different droplet dynamics. Such a ternary self-spreading and contracting droplet may have applications in cleaning high energy surfaces.

ACKNOWLEDGEMENTS: The authors thank the Rowland Fellowship Program for support of this work.

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CROWN-TO-SPLASH TRANSITION OF IMPINGING HIGH-FREQUENCY ETHANOL DROPLET TRAIN ON HEATED SURFACE

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Impingement of droplets onto a solid surface is ubiquitous in nature. Many efforts have been made to understand the intricate hydraulic, at some point multi-phase, liquid-solid interfacial phenomenon from different perspectives including droplet or droplet train impingement with or without boiling [1,2]. The various impingement patterns such as spreading, splashing, rebound, deposition, crown formation and propagation, levitation, jetting, and so on have been observed. Two distinct transitions of hydrodynamic patterns during the impingement of high-speed and high-frequency continuous ethanol droplet train on a heated titanium surface has been experimentally observed with three boiling regimes, namely, aggregation and crown periphery instability, splashing and stable crown, and stable splashing angle show in Figure 1. The transitions are further proved by the variation of the steady-state but temperature-dependent wetting area. The crown strongly depends on the surface temperature. It is quantitatively characterized by its periphery diameter and height. In the stable splashing angle regime, a sharp shift of the splashing angle from decreasing to increasing is found at the surface temperature of 319 °C. It is found that the transition of the normalized splashing angle occurs at a value of a dimensionless factor incorporating the physical properties of the liquid, the hydrodynamic parameters of the droplet and the normalized superheat of the solid surface. The correlation can be employed to suggest the emergence of the Leidenfrost phenomenon for the droplet train impingement systems.

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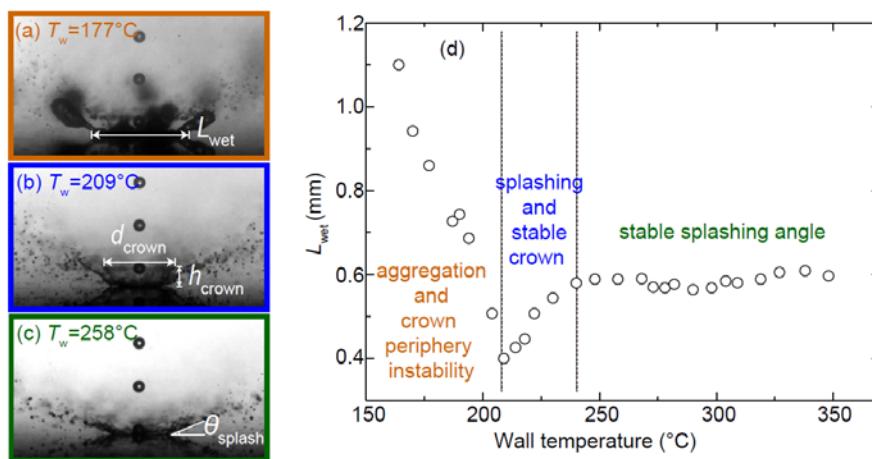


Figure 1 The typical steady-state hydrodynamic patterns for the regime of (a) the wetting length, (b) the stable crown and (c) the splashing angle. (d) shows the wetting length vs the surface temperature.

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WHERE ‘UNPREDICTABILITY’ SHADES OFF : THE TUNNELLING OF BOUNCING DROPLETS

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Classical point mechanics offers a fairly simple view of the crossing of a barrier of potential. A particle always crosses a potential barrier if its kinetic energy K is larger than the potential energy E_p of the barrier. On the contrary, if $K < E_p$, it will always be reflected by the barrier. In quantum mechanics, the crossing of a barrier of potential is not as simple : a particle with $K < E_p$ has a non-zero probability of crossing the barrier. This is called the tunnelling effect. Conversely, a particle with $K > E_p$ also has a non-zero probability of being reflected.

The random character of the crossing of a repulsive barrier by a macroscopic object was reported by A. Eddi in 2009 [1]. These objects, *the walkers*, are millimeter-sized oil droplets bouncing on a vertically vibrated oil bath. Successive rebounds create waves on the surface of the bath, which in turn guide the droplet: walkers therefore represent a tight association of a particle and a wave at the macroscopic scale [2,3,4]. When a walker is sent to the repulsive barrier, it may pass through it or be reflected -- apparently randomly.

In this work, we revisit the tunnelling of walkers with unprecedented experimental accuracy in order to understand where and how the unpredictability of the interaction outcome (crossing vs. reflexion) shades off. We show that this outcome is not directly related to the kinetic energy of the walker. Instead, a variation of the fast dynamic (vertical rebounds) during interaction with the barrier is responsible for the apparent randomness [5]. These experimental results are compared to those of a numerical simulation of the quantum tunnelling effect. We determine the trajectory of the quantum particle launched towards a barrier of potential thanks to measurements of position and momentum at regular time intervals. We observe that, similarly to walkers, the ‘unpredictability’ of the interaction outcome of a quantum particle is unveiled at the foot of the potential barrier.

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AIR ENTRAPMENT DURING THE IMPACT OF VISCOUS DROPS ONTO THIN VISCOUS FILMS

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When a liquid drop approaches a solid or liquid surface, the impact is cushioned by a thin layer of air. A stagnation point below the center of the drop prevents the air within the layer from completely escaping. Instead the pressure within the layer becomes sufficient to form a dimple in the drop ultimately entrapping an air bubble. For low viscosity liquids (~ 1 cSt), contact between the drop and the surface occurs around a ring marking the perimeter of the dimple. In the case of higher viscosity drops impacting on solid surfaces, Langley *et al.* [1] found that the drop glides over an extended layer of air thinner than 160 nm, initially avoiding contact between the liquid and solid all together. Eventually localized contacts between the drop and solid appear at random locations within this gliding layer.

Herein, we investigate drops ranging in viscosity between 10 and 10^6 cSt impacting onto a thin film of the same liquid at 1 - 5 m/s. Using ultra-high-speed interferometry at frame rates up to 5 million frames per second, we measure the evolution of the thickness of the intervening air disc and the ultimate entrapment of an air bubble. As with impacts onto solid surfaces, the drop glides on a thin layer of air. Since liquid surfaces are molecularly smooth, there are substantially fewer localized contacts when compared with impacts onto solid surfaces. Figure 1 shows bottom view images from the impact of a 100,000 cSt drop onto a thin film. The centerline height of the dimple decreases as the drop glides on an air layer. Notice the absence of local contacts in (b). The centerline height of the air disc is weakly dependent on the viscosity and follows the scaling for impacts onto solid surfaces when scaled by half of the impact velocity.

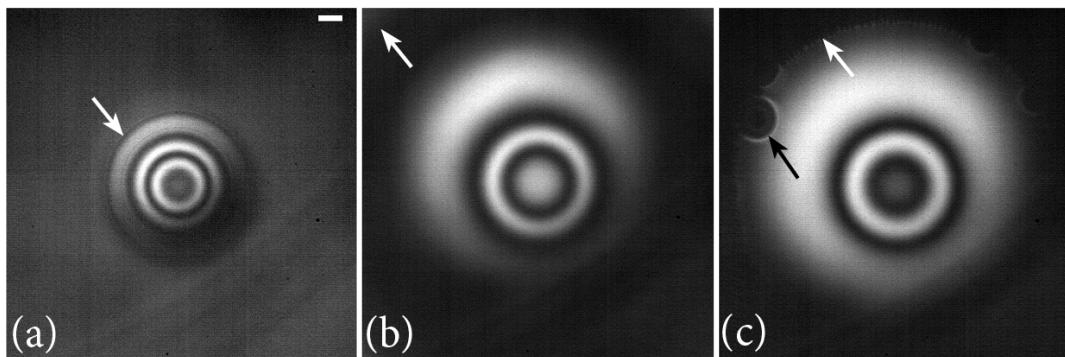


Figure 1. Bottom view interferograms from the impact of a 100,000 cSt silicone oil drop onto a thin film of the same liquid ($R_b = 1.8$ mm, $V = 1.24$ m/s). Panels are spaced by 240 microseconds. The white arrows mark the outer edge of the thin air layer. After the dimple is initially formed (a), the drop glides on a thin film of air less than 160 nm thick in (b). Eventually the air film ruptures and the central bubble contracts in (c). There are also some localized random contacts between the drop and film at later stages (black arrow)

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WETTING DYNAMICS AND ADHESION OF THERMOPLASTIC POLYMERS ON GLASS

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Wetting dynamics drive numerous technological processes involving liquids in contact with solid substrates with a wide range of geometries. The spreading dynamics of organic liquids and liquid metals at, respectively, room temperature and above 1000 °C have been studied extensively, both experimentally and numerically. However, much less attention has been paid to the wetting behavior of molten drops of thermoplastic polymers, despite its importance in the fields of polymer composites, injection moulding, and fused deposition modelling for example. There is even no knowledge about the relevancy of the classical dynamic wetting theories, i.e. the hydrodynamic approach and the molecular-kinetic theory to model the behavior of these complex liquids. This work therefore investigates the spreading dynamics of three polymers, polypropylene (PP), maleic anhydride-grafted polypropylene (MAPP), and polyvinylidene fluoride (PVDF) on glass slides, at temperatures ranging between 200°C and 260°C. These polymers were chosen based on the fact that they have different physico-chemical interactions with this substrate. The PP/glass substrate interaction should be dominated by van der Waals interaction. In the case of PVDF, additionally to the van der Waals forces, many hydrogen bonds can be created between PVDF and the OH groups on the substrate, owing to the electronegativity of the fluorine group. In addition to physical interactions, MAPP has the capability to form chemical bonds with glass via the reaction between maleic anhydride (MA) groups and OH groups presented on the glass surface [1], [2]. These measurements are completed by experiments performed with mixtures of PDMS oils of different viscosities acting as prototypes of the polymers. The study emphasizes the complex wetting behavior of these liquids controlled at the same time by the surface topography and the possible segregation of short polymer chains at the contact-line[3], [4].

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WETTABILITY-INDEPENDENT DROPLET TRANSPORT BY BENDOTAXIS

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When a drop is confined in a thin channel with deformable walls, a combination of bending and capillarity causes a pressure gradient that, in turn, results in the spontaneous movement of the liquid. Surprisingly, the direction of this motion, which we refer to as *bendotaxis*, is always the same, regardless of the wettability of the channel; *bendotaxis* may therefore be a useful means of transporting droplets on small scales, with various technological applications. This talk will present details of macroscopic experiments and a simple mathematical model used to study this motion, focussing in particular on the time scale associated with the motion, and we discuss the implications of these results.

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