The physics of 2D microfluidic droplet ensembles

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\begin{abstract}
We review non-equilibrium many-body phenomena in ensembles of 2D microfluidic droplets. The system comprises of continuous two-phase flow with disc-shaped droplets driven in a channel, at low Reynolds number of $10^{-4}$–$10^{-3}$. The basic physics is that of an effective potential flow, governed by the 2D Laplace equation, with multiple, static and dynamic, boundaries of the droplets and the walls. The motion of the droplets induces dipolar flow fields, which mediate $1/r^2$ hydrodynamic interaction between the droplets. Summation of these long-range 2D forces over droplet ensembles converges, in contrast to the divergence of the hydrodynamic forces in 3D. In analogy to electrostatics, the strong effect of boundaries on the equations of motion is calculated by means of image dipoles. We first consider the dynamics of droplets flowing in a 1D crystal, which exhibits unique phonon-like excitations, and a variety of nonlinear instabilities—all stemming from the hydrodynamic interactions. Narrowing the channel results in hydrodynamic screening of the dipolar interactions, which changes salient features of the phonon spectra. Shifting from a 1D ordered crystal to 2D disordered ensemble, the hydrodynamic interactions induce collective density waves and shocks, which are superposed on single-droplet randomized motion and dynamic clustering. These collective modes originate from density–velocity coupling, whose outcome is a 1D Burgers equation. The rich observational phenomenology and the tractable theory render 2D droplet ensembles a suitable table-top system for studying non-equilibrium many-body physics with long-range interactions.
\end{abstract}

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1. Introduction

1.1. Droplet ensembles: a driven-dissipative system

The understanding of non-equilibrium systems lags well behind the advanced theory of systems at thermal equilibrium, since free-energy functionals, whose minimization predicts the system’s behavior, are unavailable (Ruelle, 1999). Such systems exhibit complex patterns and collective dynamics (Cross and Hohenberg, 1993). The existence of underlying principles that govern non-equilibrium systems remains an open question. For systems where the dynamics is dominated by dissipative forces, propagating modes are a priori over-damped, unless sustained by an influx of energy. Driven dissipative systems that have been studied experimentally or theoretically in this context include sedimentation (Ramaswamy, 2001), elastic turbulence (Grosisman and Steinberg, 2000), dusty plasma crystals (Liu and Goree, 2005; Liu et al., 2010; Morfill and Ivlev, 2009), granular media (Aranson and Tsimring, 2006), optically trapped microparticles (Roichman et al., 2007), rotating magnetic discs (Grzybowski et al., 2000), colloidal suspensions in an AC electric field (Pérez and Posner, 2010), vortices in superconductors (Simha and Ramaswamy, 1999) and nucleoprotein filaments (Tlusty et al., 2004).

Non-equilibrium many-body systems with long-range interactions are omnipresent in Nature, for example, stellar motion, electrons conducted in a material, charged particles in plasma and gravitational sedimentation of particles in a fluid. Yet, often in such systems the understanding of collective phenomena from basic inter-particle interactions remains obscure. Quite the opposite, the system of microfluidic 2D droplets reviewed here, exhibiting rich many-body phenomena, is theoretically tractable owing to the linearity of the viscous flow equations, and to the low dimensionality. The close comparison between experiment and theory provides a unique pedagogical zoom into the emergence of collective modes from single-particle force fields. In particular, one can closely examine the intricate interplay between symmetries, boundaries and long-range interactions. This review is, therefore, intended to be an exposition of the basic physics of 2D droplet ensembles, demonstrating collective behavior in a few configurations. We focus on crystalline modes in 1D and on Burgers shock waves in 2D, both fundamental, case-study topics in theoretical physics, however, which have lacked suitable experimental realization. Thus, we suggest 2D microfluidic droplets as a promising experimental and theoretical framework to approach non-equilibrium physics, and the review is aimed to be a comprehensive survey of the essential physics.

The microfluidic droplet ensembles described here resemble the non-Brownian sedimentation of particles settling in a 3D fluid by gravitation (Guazzelli and Hinch, 2011; Ramaswamy, 2001). These particles perturb the flow around them mediating...
1/r long-range hydrodynamic interactions. The particle motion exhibits strong long-range correlations and swirls, which depend on boundary conditions (Brenner, 1999; Segré et al., 1997). Theoretically however, the collective motion remains obscure, due to the divergence in summing of the 1/r forces in 3D, as system size increases (Batchelor, 1972), and the divergence of the resulting fluctuations (Caflisch and Luke, 1985). Observables such as the particle velocity fluctuations, are not fully understood (Brenner, 1999; Caflisch and Luke, 1985; Guazzelli, 2006; Mucha et al., 2004; Ramaswamy, 2001; Segré et al., 1997; Tee et al., 2007, 2002). In contrast, in 2D the hydrodynamic forces decay as 1/r^2, which cures the system from similar divergence. The 2D droplets are a borderline case between “strong” and “weak” long-range systems since their interaction decays with an exponent of 2, which is equal to the dimensionality of the system. In such non-Hamiltonian systems, instead of examining the exponent of the interaction energy, one usually examines the exponent of the dissipation rate, which is proportional to the dipolar force Dauxois et al. (2002). Another such marginal system is that of magnetic dipoles in 3D (Landau and Lifshitz, 1960), which exhibits shape-dependent thermodynamic limits. We discuss analogous shape dependence of the 2D dipolar hydrodynamic interaction in droplet ensembles that leads to velocity–density coupling and an effective Burgers equation. The marginal dimensionality of the system allows us to present a simple first-principles theory for some of the many-body physics, which captures the long-range nature while avoiding the difficulties encountered in higher dimensions.
1.2. Experimental systems: microfluidic droplet generators

In the last decade, microfluidic technology and, specifically, devices for the formation of monodispersed droplets and bubbles, have attracted a growing scientific community working on both their physical and practical aspects (Duffy et al., 1998; Squires and Quake, 2005; Stone et al., 2004). The first studies in this field established a few techniques for droplet generation (Anna et al., 2003; Gañán Calvo and Gordillo, 2001; Cramer et al., 2004; Gañán-Calvo, 1998; Thorsen et al., 2001; Umbanhowar et al., 2000). See Baroud et al. (2010), Christopher and Anna (2007), Gönter and Jensen (2006), Pompano et al. (2011), Seemann et al. (2012), Shui et al. (2007), Stone (2010), Stone et al. (2004), Tabeling (2009), Teh et al. (2008) for comprehensive reviews. Subsequent works focused on the physics of microfluidic droplet formation (Abate and Weitz, 2011; Anna and Mayer, 2006; Castro-Hernández et al., 2009; Christopher et al., 2008a,b; Cubaud et al., 2005; De Menech et al., 2008; Dreyfus et al., 2003; Funfschilling et al., 2009; Garstecki et al., 2006, 2004, 2005c; Van der Graaf et al., 2006; Guillot and Colin, 2005; Hagedorn et al., 2004; Humphry et al., 2009; Jensen et al., 2006; van Steijn et al., 2009, 2010; Suryo and Basaran, 2006; Utada et al., 2008, 2007; Vega et al., 2010; Ward et al., 2005; Willaime et al., 2006; Zhou et al., 2006), breakup of existing droplets flowing into a T junction (De Menech, 2006; Jullien et al., 2009; Leshansky and Pismen, 2009; Link et al., 2004), droplet coalescence (Bremond et al., 2008; Thiam et al., 2009), pattern formation (Cubaud and Ho, 2004; Hashimoto et al., 2007; Hashimoto and Whitesides, 2010; Thorsen et al., 2001), nonlinear dynamics (Barbier et al., 2006; Garstecki et al., 2005a,b; Hashimoto et al., 2008), traffic (Champagne et al., 2010; Cristobal et al., 2006a; Engle et al., 2005; Hashimoto et al., 2007; Hashimoto and Whitesides, 2010), viscoelastic instability of bubble flow (Sullivan et al., 2008), crystallographic structure of droplets (Garstecki et al., 2005a,b; Hashimoto et al., 2008), traffic (Champagne et al., 2010; Cristobal et al., 2006a; Engle et al., 2005; Schindler and Ajdari, 2008), viscoelastic instability of bubble flow (Sullivan et al., 2008), crystallographic structure of droplets in 3D (Shui et al., 2009), and the dynamic structure of foam (Garstecki and Whitesides, 2006; van der Net et al., 2007; Raven and Marmottant, 2006, 2009; Weaire and Drenckhan, 2008).

Apart from the physical interest in microfluidic droplet generators, the ability to produce monodispersed droplets at the 10 µm scale opened the way to a wide range of applications in biology, chemistry and materials science. The common concept in these applications is the compartmentalization of reagents in many small, uniform and isolated sub-volumes with high degree of control over the size, composition and position of each droplet. Each droplet can encapsulate a small number, down to a single entity of interest, such as a cell or a molecule, and act as an individual ‘micro-reactor’, whose volume is in the nanoliter to picoliter range, some 4 to 7 orders of magnitude smaller than a typical 50 µm cell. This minute amount of reagents in many small, uniform and isolated sub-volumes allows for accurate high-throughput measurements and screening at the single cell or gene level, or to scan reaction conditions using minute amounts of reactants. Notably, using droplets as micro-reactors also solves the problem of reagents mixing in the laminar flow regime, which characterizes microfluidic systems (Stremler et al., 2004). The contact of the droplet with the rigid channel boundaries induces flow inside the droplet, which can lead to chaotic advection and dramatically speeds up mixing comparing to a channel without droplets (Burns and Ramshaw, 2001; Cristobal et al., 2006a; Engel et al., 2005; Schindler and Ajdari, 2008), viscoelastic instability of bubble flow (Sullivan et al., 2008), crystallographic structure of droplets in 3D (Shui et al., 2009), and the dynamic structure of foam (Garstecki and Whitesides, 2006; van der Net et al., 2007; Raven and Marmottant, 2006, 2009; Weaire and Drenckhan, 2008).

Applications in biology and biochemistry: One of the marked examples for a biological application of microfluidic droplets is the directed evolution of enzymes, which combines compartmentalization of single cells or genes with droplet sorting to screen for desired mutants within a genetic library of enzymes. Examples include directed evolution for variants of horseradish peroxidase (Agresti et al., 2010), RNA ligase (Paegel and Joyce, 2010) and FeFe Hydrogenase (Stapleton and Swartz, 2010). For a review see Wu et al. (2010). Another notable application of droplets in biology is single-cell assays such as: cell encapsulation (Edd et al., 2008; He et al., 2005a; Köster et al., 2008), enzymatic assays (Huebner et al., 2008a; Schmitz et al., 2009; Shim et al., 2009), screening of human cells (Brouzes et al., 2009; Clausell-Tormos et al., 2008), studying bacteriophage infection (Köster et al., 2009) and bacterial antibiotics susceptibility (Boedicker et al., 2008), detection of low-abundance markers (Joensson et al., 2009), and measuring a single-cell protein expression (Huebner et al., 2007) and hormone response (Barat et al., 2010). Microfluidic droplet have been used also for encapsulation and measurements of multi-cellular organisms, such as C. elegans nematodes (Clausell-Tormos et al., 2008; Shi et al., 2008) and zebrafish embryos (Funfak et al., 2007), PCR reactions of a single template molecule (Beer et al., 2007, 2008; Kiss et al., 2008; Schaerli et al., 2008; Tewhey et al., 2009), scanning of protein crystallographic conditions (Lau et al., 2007; Leng and Salmon, 2009; Shim et al., 2007; Zheng et al., 2005, 2003) and have a potential in drug discovery (Dittrich and Manz, 2006). A particularly interesting biological use of microfluidic droplets is for in vitro protein synthesis using cell-free extract. Protein synthesis was demonstrated from multiple gene copies per droplet (Dittrich et al., 2005) and later from a single copy as well (Courtois et al., 2008). Protein synthesis in such cell-like compartments may lead to the encapsulation of complex genetic
networks in “artificial cells” (Noireaux et al., 2003). For such systems, a more natural choice than water-in-oil droplets would be monodispersed lipid vesicles (Noireaux and Libchaber, 2004), which can also be generated in microfluidic devices based on water-in-oil droplet generators (Matosevic and Paegel, 2011; Osaki et al., 2011; Ota et al., 2009; Shum et al., 2008; Tan et al., 2006) (see Takinoue and Takeuchi (2011) for review).

Applications in chemistry and material science:– The potential of microfluidic droplets to chemistry was first demonstrated by Song et al. (Song et al., 2003) in a series of devices for controlled mixing of reagents into droplets using chaotic advection, measuring the reaction in time, and merging and splitting droplets. Further applications of microfluidic droplets to study chemical reactions include: bromanization reaction inside organic droplets (Cygan et al., 2005), crystallization kinetics (Dombrowski et al., 2007; Lalav et al., 2008), screening solubility diagrams (Laval et al., 2007), measuring nucleation dynamics (Edd et al., 2009), measuring streptavidin–biotin binding kinetics using FRET (fluorescence resonance energy transfer) (Srisa-Art et al., 2008b), and studying the synchronization of the Belousov–Zhabotinsky reaction within droplet ensembles (Delgado et al., 2011; Toiya et al., 2010). In addition, a number of analytical tools where integrated with microfluidic droplet devices, such as Raman spectroscopy (Barnes et al., 2006), confocal Raman microspectroscopy (Cristobal et al., 2006a; Sarrazin et al., 2008), mass spectrometry (Fidalgo et al., 2009; Kelly et al., 2009; Pei et al., 2009), liquid chromatography (Theberge et al., 2010b), electrophoretic analysis (Roman et al., 2008), electrochemical measurements (Han et al., 2009) and surface probing using droplets (Chen et al., 2008). Microfluidic droplets were also used to measure surface tension (Hudson et al., 2005).

The control over the droplets size, shape and composition enables to generate complex materials and microparticles. Examples include the formation of double (Okushima et al., 2004; Utada et al., 2005) and multiple (Abate and Weitz, 2009) emulsions, and a wide range of microparticles: polymeric particles of various shapes (Dendukuri et al., 2005; Nie et al., 2006; Nisisako et al., 2004; Xu et al., 2005), biodegradable (De Geest et al., 2005) and injectable (Shibata et al., 2010) micro-beads, Janus particles (Nisisako et al., 2006; Seiffert et al., 2010; Shepherd et al., 2006), magnetic microparticles (Chen et al., 2009; Hwang et al., 2008), liquid crystals (Hamlington et al., 2007) photonic balls (Kim et al., 2006; Yi et al., 2003) and colloidal crystallization (Gong et al., 2007), nanoparticles (Chen et al., 2005; Frenz et al., 2008b; Karnik et al., 2008; Malloggi et al., 2010; Shestopalov et al., 2004; Song et al., 2008) and even silk microspheres (Breslauer et al., 2010). See Park et al. (2010), Tumarkin and Kumacheva (2009) and Wang et al. (2011) for detailed reviews on microparticles formation.

The microfluidic droplet toolbox:– The high degree of control over microfluidic droplets became possible thanks to an array of new methods for on-chip droplet manipulation. Examples include: droplet storing and incubation (Beer et al., 2009; Boukellal et al., 2009; Edgar et al., 2009; Frenz et al., 2009; Holtze et al., 2009; Huebner et al., 2009; Lalav et al., 2007; Schmitz et al., 2009; Shi et al., 2008; Shim et al., 2007; Tan and Takeuchi, 2007), droplet sorting including active (Abate et al., 2010a; Ahn et al., 2006b; Baret et al., 2009; Fidalgo et al., 2008; Franke et al., 2009; Guo et al., 2010; de Saint Vincent et al., 2008; Zhang et al., 2009) and passive (Chabert and Viovy, 2008; Tan et al., 2004) methods, droplet manipulation (Baroud et al., 2007; Link et al., 2006; Lorenz et al., 2006; Sgro et al., 2007; Yang et al., 2010), and controlled droplet coalescence (Abate et al., 2010b; Ahn et al., 2006a; Bremond et al., 2008; Chabert et al., 2005; Fidalgo et al., 2007; Link et al., 2006; Mazutis et al., 2009; Niu et al., 2009, 2008; Priest et al., 2006; Sarrazin et al., 2007; Song et al., 2003; Tan and Lee, 2007; Thiam et al., 2009). In addition, light-controlled droplet formation was demonstrated using a UV-sensitive surfactant (Diguet et al., 2011) and several approaches were developed for the formation of single droplets “on demand” (Bransky et al., 2009; He et al., 2005a,b; Lorenz et al., 2006).

Finally, we note a body of work that studied and used the hydrodynamic coupling between droplets due to the added hydrodynamic resistance of the droplets themselves. This coupling, for example, introduces nonlinear effects to the distribution of droplets flowing into a T junction (Engl et al., 2005), which can be alleviated by adding a bypass channel across the junction that can transfer the carrier fluid but not the droplets (Cristobal et al., 2006b). Similar bypass channels and ladders were used to synchronize bubbles in parallel channels and used as bits in logical computation (Prakash and Gershenfeld, 2007), and were further analyzed in Schindler and Ajdari (2008). Hydrodynamic coupling between droplets was also exploited in the synchronized formation of droplets in two coupled T junction (Frenz et al., 2008a) as well as in several applications for droplet storing (Boukellal et al., 2009; Huebner et al., 2009; Tan and Takeuchi, 2007). The wide range of applications of microfluidic droplets and the effects of the hydrodynamic coupling between them indicate the importance of understanding the collective dynamics of droplet ensembles.

For further reading on the applications of microfluidic droplets see the following reviews on applications in biology and biochemistry (Chiu and Lorenz, 2009; Goldner et al., 2010; Huebner et al., 2008b; Pompano et al., 2011; Song et al., 2006; Taly et al., 2007; Theberge et al., 2010a; Vyawahare et al., 2010; Zagnoni and Cooper, 2011) and in chemistry and material science (Gönther and Jensen, 2006; Leng and Salmon, 2009; Wang et al., 2011). Microfluidic droplet and bubble generators have been based on three different methods: (a) Breakup in co-flowing streams, in which the dispersed phase and the carrying phase flow in parallel (Cramer et al., 2004; Umbanhowar et al., 2000). (b) Breakup in elongational or stretching dominated flows, also known as flow-focusing, in which the dispersed phase, either gas (Gañán Calvo and Cordillo, 2001) or liquid (Anna et al., 2003) is surrounded by the carrying fluid and squeezed into an orifice. (c) Breakup in cross-flowing streams, in which the two phases meet at a T junction (Thorsen et al., 2001). Our microfluidic devices are based on the latter.

A microfluidic T junction droplet generator (Fig. 1, Movie 1) was first introduced by Thorsen et al. (Thorsen et al., 2001) and its droplet formation mechanism was explained by Garstecki et al. (Garstecki et al., 2006) (see Christopher and Anna (2007) for a thorough review). Briefly, both water and oil are under pressure and forced to flow into the T junction. The water finger
Fig. 1. The experimental system. (a) Formation of a 1D microfluidic crystal of water-in-oil droplets at the T-junction between water and oil channels under continuous flow. Main channel width is $W = 250 \mu m$, channel height is $h = 10 \mu m$ and droplet diameter is $\sim 25 \mu m$. All scale bars are $100 \mu m$. (b) A disordered 2D ensemble of droplets formed at a similar device with $W = 500 \mu m$. (c) A scheme of the microfluidic device. Typical main channel width is $W = 40–800 \mu m$ and length 1–2.5 cm. Oil and water channel widths are 50 $\mu m$ and 35 $\mu m$, respectively. The width of the T junction is 20–35 $\mu m$ and the length of each of its arms is 50–300 $\mu m$. The water channel is relatively long to increase its resistance such that it is comparable to the one of the oil inlet (see Appendix A). The resulting droplet radius is typically 6–20 $\mu m$, and $u_d$ is typically 100–400 $\mu m \text{s}^{-1}$. The oil velocity $u_{oil}^\infty$ is typically 4–5 times faster than $u_d$ due to friction. (d) The flow lines around a disc-like droplet between the channel floor and ceiling. The flow is along the $x$ direction. Flow lines were calculated using the unconfined droplet potential.

that enters the junction is subject to three forces, whose balance determines the pinching of the droplet and, hence, its size:

1. The water–oil surface tension is associated with a Laplace pressure jump across the curved water–oil interface, which results in a stabilizing force that pulls the interface upstream.
2. Shear stress is exerted on the water tip by oil flowing in the gap between the interface and the channel wall, which gives rise to a force pushing the water tip downstream.
3. Pressure increase at the junction: As the water tip enters the oil channel, it obstructs the flow of oil, which increases the channel flow resistance. Namely, both the pressure-dropping across the tip and the pressure at the junction increase, which pushes the water–oil interface and narrows the water neck until the droplet pinches off. Following the pinch-off, the water tip recedes but then advances towards the junction to form a new water finger that starts to penetrate the oil channel. Since pinching is determined by a force balance, the droplet formation process is cyclic and the droplets are monodispersed. By setting the channel geometry and flow rates it is possible to control both the size of the droplets and their formation frequency. When the droplets volume is larger than the volume of a sphere whose diameter equals the channel height $h$, the droplets get squeezed between the channel floor and ceiling and are disc-like, rather than spheres. Hence, the geometry of the system is quasi-2D and the droplet disc radius $R > h/2$. In addition, the contact with the channel floor and ceiling induces friction, which renders the droplet flow velocity, $u_d$, slower than the velocity of the surrounding oil, $u_{oil}^\infty$. This velocity difference is essential for the hydrodynamic interactions in such systems.

1.3. Collective modes of droplet ensembles

The imposed flow drives the system far from equilibrium, breaks its translational symmetry and induces a long-range hydrodynamic interaction between the droplets. The flow is heavily dissipative with Reynolds number $Re \sim 10^{-4} – 10^{-3}$,
which implies tractable, linear Stokes flow equations. The quasi-2D geometry – in which one of the dimensions of the confined fluid is much smaller than the other two – significantly simplifies the problem. Theoretically, the quasi-2D Stokes flow is described by the lubrication approximation, in terms of an effective potential flow, which is much simpler than the bi-harmonic equation of 3D Stokes flow. Moreover, in contrast to 3D, the forces in 2D decay as $1/r^2$ dipoles and do not diverge upon summation. Experimentally, the planar geometry, with characteristic length of tens of microns and typical time scale of tenths of a second, render the 2D flow accessible to standard optical microscopy. Overall, these features place microfluidic droplet ensembles as a useful system for studying non-equilibrium many-body physics.

The simplest ensemble exhibiting collective modes is a 1D microfluidic crystal of droplets flowing in the 2D channel. The crystal exhibits acoustic modes, akin to solid state phonons, that propagate at $\sim 100 \mu m s^{-1}$ and frequencies of $\sim 1 Hz$ with unique dispersion relations. In addition, the phonons give rise to a variety of nonlinear instabilities. The phonons and instabilities stem from the hydrodynamic dipolar interaction between the droplets (Beatus et al., 2006). Confining the geometry by narrowing the channel width affects phonon spectra and leads to non-monotonous behavior of the sound velocity. This originates from the interplay between screening of the dipolar forces and the increasing incompressibility of the confined crystal (Beatus et al., 2007). Finally, we shall describe the dynamics of a 2D disordered ensemble, in which droplets move erratically and exhibit dynamic clustering. Superposed on the complex trajectories of single droplets there are collective modes of shock wave and sound that propagate at $\sim 100 \mu m s^{-1}$. These waves obey the 1D Burgers equation owing to local linear coupling between droplet number density and velocity (Beatus et al., 2009).

At first, the observation of waves and shocks at low Reynolds number may seem inconsistent with the notion that oscillations are over-damped in a dissipative medium (Happel and Brenner, 1965; Landau and Lifshitz, 1960; Purcell, 1977), such as in Cicuta et al. (2010), Di Leonardo et al. (2007) and Polin et al. (2006). Note, however, that over-damping is usually discussed in symmetric, non-driven systems, while the microfluidic droplet ensembles described here are driven by a symmetry-breaking flow field. For example, consider a 1D harmonic crystal in the continuum approximation, whose dynamics is described by the 2nd order spatially symmetric equation: $(\rho \partial_t^2 + K \partial_x - \mu \partial_x^2)A = 0$, with the amplitude field $A(x, t)$, mass density $\rho$, elastic constant $K$ and friction $\mu$. When dissipation dominates inertia, all waves are over-damped: $\omega(k) = i(K/\mu)k^2$. However, if a symmetry-breaking field (Ramaswamy et al., 2000), for instance $\xi \partial_x A$, replaces the dispersive term, $K \partial_x$, the equation of motion becomes $((\xi - \mu \partial_x)A = 0$, a first-order wave equation with traveling solutions $\omega(k) = (\xi/\mu)k$. To conclude, when inertia is negligible, waves still persist due to a symmetry-breaking field despite the dissipation, as occurs in the low Reynolds 2D droplet ensembles.

In this paper, we review the many-body physics of 2D droplet ensembles, providing a unified framework. The paper is organized as follows: The flow induced by a single droplet is derived in Section 2 both in an open quasi-2D geometry as well as in a channel confined by sidewalls. In Section 3 we analyze the two-body hydrodynamic interaction between droplets, as well as the collective dynamics of droplet ensembles. Next, we derive the collective behavior of 1D microfluidic crystals (Section 4) and of 2D disordered ensembles of droplets (Section 5). These effects were reproduced also by numerical simulations (Appendix B). Finally, in Section 6 we discuss several open questions that may be further pursued using microfluidic droplet ensembles. Experimental considerations are described in Appendix A.

2. The flow of a single droplet

2.1. Unconfined droplet

2.1.1. Dipoles in potential flow

We model a droplet as a disc squeezed between the channel floor and ceiling with $h \ll R$. The droplet is centered at the origin and moves at velocity $u_0$ with respect to the channel (Fig. 1(d)). The velocity of oil far from the droplet is $u_0^{\infty}$ along the $x$ direction. The $z$ axis is perpendicular to the floor and ceiling, which are located at $z = \pm h/2$ respectively. In this section we assume the channel has no sidewall boundaries, which are considered in the following Section 2.2. The Reynolds number of the moving droplet is given by $Re = u_0h/\eta_0 \sim 10^{-4} - 10^{-3}$, in which $\rho_0 = 0.84 g/cm^3$ is oil density, $\eta_0 = 30 mPa s$ is oil viscosity. The typical length scale $l$ can be chosen either as the channel height $h$ or droplet radius $R$. Here, $R \gtrsim 2h$, such that in both cases Re lies safely in the $Re \ll 1$ limit.

In this low-Reynolds limit, the nonlinear term in the Navier–Stokes equation,

$$\rho_0(\partial_t v + (v \cdot \nabla)v) = \eta_0 \nabla^2 v - \nabla P,$$  

is negligible compared to the dissipation term, and the steady-state incompressible flow around the droplet satisfies the equations:

$$\nabla^2 v = \eta_0^{-1} \nabla P \tag{2}$$

$$\nabla \cdot v = 0 \tag{3}$$

where $v(r)$ is the oil velocity at $r$ and $P(r)$ is the local pressure. The first two boundary conditions (BC) we set are $v = 0$ on the channel floor and ceiling (no slip) and $v = u_0^{\infty} \hat{x}$ very far from the disc.
Fig. 2. The flow within the aqueous interior of droplets was visualized using fluorescent beads. (a) Two images of the same droplet that were taken 50 ms apart. Flow was from left to right. The droplet boundary is marked with a dashed circle. Red and green lines indicate the trajectory of each bead during the 30 ms before the image was taken. The corresponding arrows indicate the direction of each trajectory. The beads marked in red originated in the bottom half of the droplet while the green originated in the top half. Droplet diameter was 18 µm. (b) The velocity profile inside the droplet was measured by tracking a few tens of beads in several droplets. The vector field indicates the local mean velocity of water in the droplet frame-of-reference. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

To find the third BC at the water–oil interface on the droplet’s circumference, we need to consider the flow of water inside the droplet (Fig. 2). The water flow has to satisfy the no-slip BC on the floor and ceiling while on the droplet’s circumference it is entrained by the faster oil. Hence, the water flow has a treadmill pattern, in which water near the water–oil interface moves faster than the droplet’s center of mass and slower elsewhere inside the droplet. The flow profile inside droplets was measured by following fluorescent micro-beads immersed in the water, as shown in Fig. 2. Since the droplet maintains its disc-like shape and there is no flux of oil through the circumference, the boundary condition for the flow of oil on this fluid–fluid interface is, therefore, zero velocity perpendicular to the droplet edge (slip boundary condition). Other cases in which similar (but more elaborated) flow patterns arise include: a spherical droplet moving in ambient fluid (Happel and Brenner, 1965), a ‘plug’ droplet in contact with the channel sidewalls, floor and ceiling (Burns and Ramshaw, 2001; Kinoshita et al., 2007), and a spherical droplet flowing in a channel without touching the boundary (Hudson, 2010). The internal flow in the latter case is due to the velocity gradient of Poiseuille flow.

Farther than \( O(h) \) from the droplet boundary, we can assume that the no-slip condition on the floor and ceiling implies that the velocity variations along the \( z \) direction are much larger than along the \( xy \) plane and that \( \partial^2_z \gg \partial^2_{x}, \partial^2_{y} \) (a.k.a. the Darcy approximation) (Darnton et al., 2001). Within the \( O(h) \) vicinity around the droplet, the validity of this assumption is not guaranteed, since the droplet acts like a moving vertical wall, which may imply large velocity gradients in \( xy \). Nevertheless, this wall is made of fluid surfactant that envelopes the fluid droplet and therefore does not constrain the tangential velocities like a moving solid wall. Indeed, the measured velocity of water near the interface at \( (x, y) \sim (0, \pm R) \) was comparable to \( u_{\infty} - u_{a} \), which suggests that the Darcy approximation is valid closer to the droplet than in the rigid boundary case.

Together with the Stokes equation (2), the approximation of \( \partial^2_z \gg \partial^2_{x}, \partial^2_{y} \) (Darnton et al., 2001) leads to separation of variables, \( \mathbf{v}(x, y, z) = u_{oil}(x, y)f(z) \), such that \( f(z) = (3/2)[1 - (2z/h)^2] \) is a parabolic Poiseuille profile, and

\[
u_{oil}(x, y) = -\frac{h^2}{12\eta_o} \nabla P(x, y)
\]

is the velocity in the \( xy \) plane averaged along the \( z \)-axis. Eq. (4) simply asserts that oil flows from high to low pressure. The pressure defines an effective 2D potential, \( \phi(x, y) \equiv (-h^2/12\eta_o)P \). Together with the incompressibility condition (Eq. (3)) we obtain that the flow in the \( xy \) plane is described by the 2D Laplace equation:

\[
u_{oil}(x, y) = \nabla \phi
\]

\[
\nabla^2 \phi = 0.
\]
Since the flow is effectively 2D, it is convenient to use the complex potential \( w(z) = \phi(z) + \psi(z) \), in which \( \phi = \text{Re}[w] \) is the flow potential used above, \( \psi = \text{Im}[w] \) is the stream function and \( z = x + iy \) (do not confuse with the \( z \) coordinate, which we do not deal with from here on). The components of the oil velocity satisfy the Cauchy–Riemann relations: \( u_x = \partial_y \psi \) and \( u_y = -\partial_x \psi \), which imply that the complex velocity is \( dw/dz = u_x - iu_y \) and that both \( \psi \) and \( w \) satisfy the Laplace equation (Landau and Lifshitz, 1987). In a frame-of-reference moving with the droplet, the boundary condition (BC) for \( \psi \) is \( \psi(z) = \) constant along the droplet boundary at \( |z| = R \). The solution of the Laplace equation that satisfies the BC for the droplet is:

\[
\begin{align*}
\psi(z) &= \delta u_0 \cdot z + w_d(z) \\
\phi_d(z) &= R^2 \delta u_0 \cdot \frac{1}{z},
\end{align*}
\]

in which \( \delta u_0 \equiv (u_0^\infty - u_d) \) is the relative velocity between the droplet and oil. The first term in Eq. (7) corresponds to the imposed main flow of oil (to transform to the channel frame-of-reference, \( w(z) \to w(z) + \bar{u}_d \cdot z \)). The second term \( w_d(z) \) is the potential of a 2D dipole,

\[ d_0 = R^2 \delta u_0, \]

describing the droplet’s perturbation (Tlusty, 2006). The dipolar potential is \( \phi_d(r) = \text{Re}(w_d) = d_0 \cdot x/r^2 \). The perturbation of a single droplet was visualized in Fig. 3 by fluorescent micro-beads suspended in the oil along with the dipolar flow field \( \nabla \phi_d \) superimposed.

The droplet’s dipole is analogous to an electrostatic dipole of two electric charges \( \pm q \) separated by \( \delta x \) along the \( x \) direction, which form a dipole of magnitude \( q \cdot \delta x \). In 2D hydrodynamics, charge has units of 2D flux: \( L^2/T \), hence the dipole magnitude \( d_0 \) - representing a point source and a point sink of oil infinitesimally separated along \( x \) – has units of \( [d_0] = L^3/T \). Intuitively, fluid is pumped into the sink and re-injected at the source, generating a net propagation of the droplet to the right.

The dipolar field decays as \( 1/r^2 \) in contrast to the far flow field of any small particle moving in an unbounded 3D fluid, which decays as \( 1/r \). In the 3D case, the moving particle can be treated as a point source of momentum (momentum monopole), such that the momentum flux through the fluid, which scales as \( \rho v^2 \), is conserved. Namely, for any distance \( r \) from the particle \( \rho v^2 r^2 \) is constant, hence \( v \sim 1/r \). The sphere also displaces fluid mass like a 3D mass-dipole, which stems from mass conservation leading to a description of fluid displacement by mass source–sink pair that forms a mass dipole. However, this effect decays as \( 1/r^3 \), as in electrostatic dipoles in 3D, hence negligible comparing to the momentum monopole (Diamant, 2009; Happel and Brenner, 1965; Landau and Lifshitz, 1987). In contrast, in quasi-2D the translational symmetry along \( z \) is broken by the channel floor and ceiling, hence momentum is not conserved—the floor and ceiling absorb transverse momentum by finite momentum flux, \( \eta_\delta (\partial_z v_z)_{z=\pm h/2} \sim -v/h \neq 0 \), which originates from the Poiseuille flow profile along \( z \) and the momentum–flux density tensor (Eq. (11)). Since the velocity loss is proportional to the velocity itself, namely \( \partial_z \rho v \sim -v/h \) with \( r = (x, y) \), the momentum monopole field decays exponentially as \( r/h \). Hence, since oil mass is conserved, the dominant contribution to the droplet flow field is attributed to mass–displacement, which due to the two-dimensionality results in a mass-dipole (source–sink pair) that decays as \( 1/r^2 \) (Diamant, 2009).

2.1.2. Drag force on a droplet

The drag force \( F_i \) that a flow exerts on a body is given by integrating the momentum–flux density tensor \( \Pi_{ik} \) on the body boundary \( D \) (Landau and Lifshitz, 1987):

\[
F_i = -\int_D \Pi_{ik} dS_k
\]

\[
\Pi_{ik} = \rho \delta_{ik} + \rho_\delta v_i v_k - \eta_\delta (\partial_k v_i + \partial_i v_k).
\]

Here, \( dS \) is a surface element of area \( dS \) perpendicular to the surface and pointing outwards. We work in the channel frame-of-reference, in which the incident oil velocity is \( u_0^\infty \). The first term in (11), \( \rho \delta_{ik} \), results upon integration in a force due to
pressure difference $\delta P \sim R \nabla P$ across the droplet. From Eq. (4) we estimate $\nabla P \sim \eta_0 u^2 / h^2$, hence $F_{\text{press}} \sim \delta P \cdot RH = \eta_0 u R^2 / h$.

Integration over the second term yields a mass flux contribution $F_{\text{flux}} \sim \rho_o u^2 R h$. The third term is the force due to viscous dissipation $F_{\text{visc}} \sim \eta_0 (u/R)(R H) = \eta_0 u h$. Comparing the three forces we get $F_{\text{visc}} / F_{\text{press}} \sim (h/R)^2 \ll 1$ and $F_{\text{flux}} / F_{\text{press}} \sim \text{Re} \ll 1$. Therefore, the dominant term in $\Pi_{\text{drag}}$ is the pressure term $P \delta_R$.

The drag force exerted on the droplet in a potential flow is calculated from the potential $\phi$:

$$ F_{\text{drag}} = -\oint_D P dS = 12 \eta_o h^{-2} \oint_D \phi dS $$

(12)

We write $\phi$ as a combination of the potential in the droplet frame-of-reference $\tilde{\phi}$ and a uniform flow $u_d \cdot x$, such that $\phi = \tilde{\phi} + u_d \cdot x$.

We then substitute $\tilde{\phi} = \tilde{w} - i \tilde{\psi}$ and $dS = -ih dz$, where $\tilde{w}$ and $\tilde{\psi}$ are in the droplet frame-of-reference. Since $\tilde{\psi}$ is constant along $D$ in the droplet frame, $\oint_D \tilde{\psi} dz = \tilde{\psi} \oint_D dz = 0$. We get from Cauchy’s theorem that the drag force consists of a term proportional to the residues of $w(z)$ inside the droplet, and a contribution due to the uniform flow:

$$ F_{\text{drag}} = 12 \eta_o h^{-2} \left[ \oint_D (u_d \cdot x) dS + \oint_D \tilde{w}(z)(-ih dz) \right] $$

$$ = \frac{1}{2} \xi_d u_d + \xi_d R^{-2} \sum_d \text{Res}(w), $$

(13)

where we used the fact that the poles of both $\tilde{w}(z)$ and $w(z)$ are identical and define the drag coefficient:

$$ \xi_d \equiv 24 \eta_o h^{-1} \pi R^2. $$

(14)

The singularities considered in this paper are all simple, $1/z$ poles, whose residues are the dipoles. The force can therefore be rewritten as a sum over the dipoles $d_i$ inside the droplet,

$$ F_{\text{drag}} = \frac{1}{2} \xi_d u_d + \xi_d R^{-2} \sum_d d_i, $$

(15)

Eqs. (13)–(15) are valid for any flow potential and will be used also to describe an ensemble of droplets, where the interdroplet forces are due to image dipoles reflected into $D$ (Section 3). For a single droplet, $w(z)$ (Eq. (8)) has only one pole at the origin, $d_0/z$, with

$$ d_0 = \text{Res}(w) = R^2 \delta u_0, $$

(16)

hence the drag force on the droplet is:

$$ F_{\text{drag}} = \frac{1}{2} \xi_d u_d + \xi_d R^2 \delta u_0. $$

(17)

The drag force (17) is similar to previous theoretical and experimental results (Dangla et al., 2011; Maruvida and Park, 1996; Rabaud et al., 2011; Taylor and Saffman, 1959), and is a sum of the force exerted by the unperturbed Poiseuille pressure around a droplet carried at the speed of oil (first term) and force due to the velocity difference (second term). A static droplet $u_d = 0$, is subject to the drag force, $F_{\text{drag}} = \xi_d u_0$. While a droplet that moves twice as fast as the oil, $u_d = 2u_0$, experiences no force, $F_{\text{drag}} = 0$, since it perturbs the flow such that the pressure on its boundary is constant. Bubbles filled with low viscosity fluid (gas) may approach this maximal velocity due to negligible friction with the channel.

For typical experimental parameters, $R = 15 \mu m$ and $h = 10 \mu m$ we have $\xi_d = 50 \text{ pN}/(\mu m/s)$, which implies that under a flow with $\delta u_0 = 1000 \mu m/s$ the drag force on the droplet is $F_{\text{drag}} \approx 60 \text{ nN}$ along the flow, assuming a typical ratio of $u_{\text{oil}}^0 / u_d = 4$. For this ratio, the contribution of the second term in Eq. (17) is $6$ times as large as the first term. As we show in Section 3.3, the drag force due to the hydrodynamic interaction between droplets, namely, the typical force on a droplet due to the dipolar flow field of another droplet, is weaker by $1–2$ orders of magnitude than the force exerted on an isolated droplet by the flow (depending on the distance between the two droplets).

### 2.1.3. Friction on a droplet

In addition to the drag force, the droplet is subject to a force of friction due to its contact with the channel floor and ceiling. In the absence of friction, the droplet would have drifted at velocity $2u_{\text{oil}}^\infty$, while in the experiment it flows much slower at about $\frac{1}{4}u_{\text{oil}}^\infty$ to $\frac{1}{4}u_{\text{oil}}^\infty$. As we now show, the flow inside the droplet caused by the boundaries (Fig. 2) induces energy dissipation, which is equivalent to a frictional force of the form $F_{\text{fric}} \propto u_d$. Let $\mathbf{v}(r)$ be the water velocity inside the droplet. The energy dissipation rate of this flow is given by

$$ \dot{\varepsilon} = \frac{1}{2} \eta_o \int (\partial_t \mathbf{v} + \partial_i \mathbf{v})^2 d^3 r, $$

(18)

where the integration over the droplet’s volume (Landau and Lifshitz, 1987). We merely assume that the internal velocity scales with $u_d$ and has a generic dimensionless spatial dependence $\mathbf{g}(r)$, namely $\mathbf{v}(r) = u_d \cdot \mathbf{g}(r)$. It follows from Eq. (18) that
the dissipation rate is \( \dot{\epsilon} = \mu_d \dot{u}_d^2 \), with a friction coefficient \( \mu_d \) that depends on \( \mathbf{g}(\mathbf{r}) \). The friction coefficient must depend also on water viscosity \( \eta_w \), on the channel height \( h \) that affects the shear inside the droplet, as well as on the viscosity ratio \( \eta_w/\eta_o \) (Landau and Lifshitz, 1987). Finally, we equate the energy dissipation rate \( \dot{\epsilon} \) to the work done by the force of friction \( \mathbf{F}_{\text{fric}} \cdot \dot{\mathbf{u}}_d \) and obtain the friction on a single droplet:

\[
\mathbf{F}_{\text{fric}} = -\mu_d \dot{\mathbf{u}}_d, \quad (19)
\]

where the minus sign indicates that friction is opposite in direction to the droplet’s velocity. To calculate \( \mu_d \) exactly one needs to solve for the flow inside the droplet, which is left for elsewhere.

2.1.4. Equation of motion

At the low-Reynolds limit, \( \text{Re} \ll 1 \), inertial effects are negligible and the sum of all forces acting on the droplet must vanish. Hence, drag (Eq. (17)) and friction (Eq. (19)) on the droplet balance out, \( \mathbf{F}_{\text{drag}} + \mathbf{F}_{\text{fric}} = 0 \), which implies:

\[
\mathbf{u}_o^\infty = \left( \frac{1}{2} + \frac{\mu_d}{\xi_d} \right)^{-1} \mathbf{u}_o^\infty \quad (20)
\]

Here we used the additional superscript on \( \mathbf{u}_o^\infty \) to indicate it is the velocity of an isolated droplet. The proportionality constant between droplet and oil velocities reflects the ratio between single-droplet friction and drag coefficients and depends only on geometry \( (R, h) \) and viscosities \( (\eta_o, \eta_w) \). Hence, although we do not calculate nor measure \( \mu_d \) directly, we can still calibrate \( (\frac{1}{2} + \mu_d/\xi_d) \) by measuring \( u_o^\infty / u_w^\infty \), since both \( \xi_d \) and \( \mu_d \) are constants for all droplets, either isolated or not. The calibration is performed once for a given geometry and viscosities and used in the analysis of collective effects and is expressed as

\[
K \equiv \frac{u_o^\infty}{u_o^\infty} = \left( \frac{1}{2} + \frac{\mu_d}{\xi_d} \right)^{-1} \cdot (21)
\]

Using the calibration we estimate for the friction coefficient: Since an isolated droplet typically moves at \( u_o^\infty \approx \frac{1}{4} u_o^\infty \), we have \( \mu_d \approx 3 \frac{1}{2} \xi_d \approx 175 \text{ pN} / (\mu \text{m s}^{-1}) \). Furthermore, using this value we show that thermal effects are negligible in this system, by estimating the droplet’s diffusion coefficient from the Einstein relation \( D = k_B T / \mu_M \), where the mobility, \( \mu_M \equiv F/\mu \), is identical to the friction coefficient: \( \mu_M = \mu_d \). The resulting diffusion coefficient is \( D \sim 10^{-12} \text{cm}^2 \text{s}^{-1} \), about 3 orders of magnitude smaller than the diffusion coefficient of a 1 µm bead in water. Hydrodynamic interactions between micro-beads due to their thermal motion in equilibrium were measured by Cui et al. (Cui et al., 2004), who showed that these interactions lead to anomalous diffusion in quasi-2D. The dipole of each bead is parallel to its instantaneous thermal velocity. The system considered here is essentially different, since the interactions stem from the external flow field that drives the system far from equilibrium and the direction of each dipole is parallel to the imposed flow.

Some of the collective effects reported here – phonons and instabilities – can be understood based on the flow field of the unconfined droplet. The reader interested in these phenomena may skip to Section 3.

2.2. Droplets near boundaries: Confinement

2.2.1. Analogy to electrostatics: method of image dipoles

Until this point we considered the flow around a droplet in a 2D slab of oil without vertical sidewalls, while the rectangular microfluidic channels had a finite width \( W \) with sidewalls at \( y = \pm W/2 \) (Fig. 1). In order to account for the effect of the sidewalls, we impose an additional slip boundary condition (BC) on the Laplace Eq. (6) demanding zero oil flux through the sidewalls. Note that exactly at the sidewalls the actual BC are of no-slip flow due to the viscous shear force of the walls. However, moving even a mere \( h \ll W \) from the walls suffices to render this shear negligible, which allows safe use of slip BC in the Darcy approximation, where \( h \to 0 \). The no-slip BC has no potential-flow solution since the Darcy approximation is invalid. Similar considerations lead to slip BC at the edge of the droplet. When we calculated the flow field of a single droplet without sidewall boundaries, the dipole solution (Eq. (8)) already satisfied the BC at the droplet edge. However, as additional BC are introduced, by considering either sidewalls or more than one droplet, the BC on a droplet’s edges is no longer satisfied by a simple dipole (Eq. (8)). In this section we present a scheme for simultaneously satisfying all BC for a single droplet between two sidewalls, and in the preceding section derive a practical approximation for droplet ensembles.

Just like in electrostatic problems, which are also governed by the Laplace equation, one may eliminate the fluid flux across the boundary using the method of image charges (Kirchhoff, 1845; Lamb, 1932; Milne-Thomson, 1962). This method is used to calculate the electric potential of a charge distribution in the presence of conducting surfaces, and is based on placing image charges of opposite signs to the real charges in order to eliminate the electric field parallel to the surface. In hydrodynamics, one can eliminate the flow field perpendicular to a surface by setting the sign of the image charges to be identical to the real one. The hydrodynamic ‘charges’ are 2D fluid point-sources and point-sinks with potential \( \phi_s = (q/2\pi) \log r \), such that the ‘charge’ \( q \) has units of 2D flux, \( L^2 / T \). This potential conserves the 2D mass flux, as its radial fluid velocity is \( v(r) = \phi_s / \partial r = q/2\pi r \) and the corresponding flux across any round shell around the origin is \( \int v(r) dr = q \). Within this framework, a dipole is a source–sink pair with an infinitesimal separation between them. The
Fig. 4. Image charges method. (a) The 2D flow field of a point-source of fluid (blue) next to a horizontal wall is calculated by summing the source and image (gray) potentials. The image charge is identical to the source and its position is given by a mirror image with respect to the wall. (b) The flow field of a point source near a circular boundary calculated using Kirchhoff’s method. The two images added are a sink at the center of the circle and a source at the reciprocal point $R^2/L$ (gray). The magnitude of both images is identical to the source. (c, e) Using the reflection rule from a straight wall to calculate the flow field of a dipole near a wall. The dipole (blue) is treated as a source–sink pair, hence its image (gray) is identical to the dipole. (d) The flow field of a dipole (blue) near a circular boundary, calculated using Kirchhoff’s method. The image dipole (gray) is inside the circle at the reciprocal point $R^2/L$ from the center, where $L$ is the distance to the real dipole. The magnitude of the image dipole is attenuated by $(R/L)^2$. (f) A schematic representation of the image dipole (gray) generated by a real dipole (blue) due to a circular surface, showing a general case of the real dipole orientation. Note the rotation and attenuation of the image dipole. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Pair potential is the sum of two potentials of the form $\phi_s$ with charges $\pm q$, such that to first order of approximation the sum is a dipole potential (Eq. (8)). The dipole moment has units of hydrodynamic charge times distance, namely, $L^3/T$ (Milne-Thomson, 1962).

To use the method of images we show how to place image charges in order to cancel the flux into the two types of boundaries: a straight sidewall and a circular boundary. We later generalize these two cases and show how to combine image point charges into image dipoles. Image charges are placed according to the following two reflection rules:

(i) For a straight infinite wall and a point charge located at distance $L$ from the wall, an identical image charge is placed at distance $L$ on the other side of the wall, the same as a reflection off a mirror (Fig. 4(a)).

(ii) For a circular boundary of radius $R$ and a point charge $q$ at distance $L > R$, we make use of Kirchhoff’s result (Kirchhoff, 1845; Lamb, 1932; Milne-Thomson, 1962), who placed two image charges inside the circle: (1) a sink of magnitude $-q$ at the center of the circle and (2) an image source $q$ placed at the reciprocal point, $R^2/L$ from the center along the vector pointing at the original charge (Fig. 4(b)). This result applies for both sources ($q > 0$) and sinks ($q < 0$).

To use these two rules for dipoles (source–sink pairs), rather than point charges, we apply each rule twice:

(i) For a straight wall, the magnitude of the image dipole is identical to the real one, but its direction is given by the mirror image of the real dipole. For example, the original dipole in Fig. 4(c), (e) is $d_{or}$ located at $z = -il$, hence, the potential of
its image dipole is given by \( w_{im}(z) = \bar{d}_{im}(z - iL)^{-1} \). The complex conjugation accounts for the rotation of the image dipole upon reflection.

(ii) Applying the reflection rule for a circular boundary for a dipole \( d_{or} \) gives four image charges: identical source and sink that cancel out at the circle's center and an image dipole at the reciprocal point (Fig. 4(d), (f)). The reflection reduces the (infinite) distance between the source and sink of the image dipole, \( d_{im} \), hence its dipole moment decreases by a factor of \((R/L)^2\). The weaker \( d_{im} \) is closer to the circle boundary than \( d_{or} \) and therefore suffices to cancel the influx through this boundary.

The direction of \( d_{im} \) is given by a mirror image of the original dipole \( d_{or} \), such that the mirror is perpendicular to the line connecting \( d_{or} \) and the center of the circle. Fig. 4(f), for example, shows \( d_{or} \) located at \((L, \theta)\) pointing along the \( x \) direction. The orientation of its reflection is, hence, \( 2\theta - \pi \), in addition to taking the complex conjugate of \( d_{or} \). The potential of the image dipole \( w_{im} \) is therefore,

\[
w_{im}(z) = \frac{d_{im}}{z - z_{im}},
\]

\[
d_{im} = -\bar{d}_{or}\left(\frac{R}{L}\right)^2 e^{2i\theta}
\]

with \( z_{im} = e^{i\theta}R^2/L \).

The reflection from a wall is a limiting case of the reflection from an infinitely large circle. Importantly, the induced image dipole is proportional to the velocity field \( \delta u_{or} \) of the original dipole \( d_{im} = R^2 \delta u_{or}(z_{im}) \) measured at \( z_{im} \). We later use the latter result to calculate the force on a droplet as in Eq. (15). In addition, we note that the use of the method of images adds image dipoles, but does not add new boundaries. The only boundaries in the system are the channel sidewalls and the circular boundaries of the real droplets.

In fact, the reflection rule from a circle can be used to calculate the dipole of a single droplet in a different way than the one used to obtain Eq. (8). First, we note that the uniform flow \( u_{oil}\hat{x} \) can be considered as the field between two ‘capacitor plates’. The plates in a 2D system are uniformly charged lines directed along \( y \), such that a positively charged line is located at \( x = -\infty \) and a negatively charged line is at \( x = +\infty \). Both plates have a linear charge density \( \lambda = u_{oil}/2\pi \). Second, we consider a circular droplet boundary at the origin and sum over the images of the plates’ charges that are reflected into the circle. The summation yields the same dipole as obtained from solving the Laplace equation with the droplet’s BC.

In the rest of this section we find the flow field of a droplet between two sidewalls: we first apply the method of images to the simpler case of a droplet near a single wall. This results in an infinite series of images due to back-and-forth reflections between the droplet boundary and the wall. Subsequently we refer to the two sidewalls case, where the three boundaries complicate the enumeration of images, hence an approximated solution is presented.

### 2.2.2. A droplet near a single wall

Using the reflection rules we calculate the flow potential of a droplet at \((0, -L)\) and at distance \( L \) from a single horizontal wall at \( y = 0 \). Fig. 5 shows the stream lines of the calculated flow field, which include the droplet’s dipoles, an increasing number of dipole images and the imposed uniform flow \( u_{oil}\hat{x} \). In Fig. 5(a), (b) we start from the droplet’s dipole at \((x_0, y_0) = (0, -L)\), which satisfies the boundary condition on the droplet but not on the wall. The first reflection off the wall is at \( y_1 = L \) (Fig. 5(c), (d)), whose addition fixes the boundary condition on the wall, though ruins it on the droplet’s surface. The second image is the reflection of \( y_1 \) off the droplet’s boundary to \( y_2 = -L + R^2/(L + y_1) \). The magnitude of this reflected dipole decreases by \( g_2 = (R/2L)^2 \). The third image is the reflection of \( y_2 \) off the wall to \( y_3 = -y_2 > 0 \) and so on, such that the dipole in each even reflection further decreases by the corresponding attenuation factor \( g_n \). The \( y_n \) coordinates and \( g_n \) of all reflections are given by:

\[
y_{2n} = -Rx\left(\frac{\alpha^{2n+2} + 1}{\alpha^{2n+2} - 1}\right)
\]

\[
y_{2n+1} = -y_{2n}
\]

\[
g_{2n} = g_{2n+1} = \prod_{k=0}^{n-1}\left(\frac{R}{L + y_{2n+1}}\right)^2 = \alpha^{2n}\left(\frac{\alpha^2 - 1}{\alpha^{2n+2} - 1}\right)^2
\]

for \( n \geq 0, \chi \equiv \sqrt{(L/R)^2 - 1}, \) and \( \alpha \equiv \chi + L/R \). The sum of the first \( m \) reflections satisfies either the boundary condition on the wall for odd \( m \), or on the droplet for even \( m \). The complex potential of the droplet is:

\[
w_d(z) = R^2 \delta u_0 \sum_{m=0}^{\infty} \frac{g_n}{z - iy_n}
\]

\[
= R^2 \delta u_0 \left(\frac{i\chi}{\ln \alpha}\left[\psi_{1/\alpha} \left(1 - \frac{1}{2 \ln \alpha} \ln \frac{z - R - i\chi}{R + i\chi}\right) - \psi_{1/\alpha} \left(1 + \frac{1}{2 \ln \alpha} \ln \frac{z - R + i\chi}{R + i\chi}\right)\right]\right),
\]

where \( \psi \) is the digamma function.
Fig. 5. The streamlines around a droplet located \( L = 1.25R \) below a single wall was calculated using an increasing number of image dipoles. (a) When only the droplet’s dipole is considered, the flow penetrates the wall, violating boundary conditions (BC). (b) Zooming into the gap between the droplet and the wall shows that the streamlines satisfy the BC on the droplet (gray) but violate them on the wall (red). (c) Adding the first reflection (gray dot) fixes the BC on the wall, but disrupts it on the droplet, as shown in the close-up (d) by the red streamline. This configuration is the same as in Fig. 4(a). (e–f) Adding the second reflection improves the approximation: the droplet BC is satisfied, but there are still streamlines that penetrate the wall, though unseen in the close-up (f). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

in which \( \psi_q(z) \) is the \( q \)-digamma function with \( q = \alpha^{-2} \). This potential is equivalent to the flow potential of two droplets separated by distance \( 2L \) along \( y \). When the droplet is very close to the wall \( (L \to R) \) and after including the uniform flow of the oil, Eq. (26) becomes:

\[
w_d(z) = \pi R \delta u_0 \coth \left( \frac{\pi z}{R} \right).
\] (27)

identical to the solution given by (Milne-Thomson, 1962) for the flow past a cylinder lying on a hard surface. Far from the droplet at \( |z| \to \infty \) the potential approaches \( R^2 \delta u_0 (\pi^2/3)(1/z) \), similar to the potential of an isolated droplet without the wall, but stronger by a factor of \( \pi^2/3 \).

When the droplet is very far from the wall \( (L \gg R) \), all the reflections off the wall can be approximately taken at the position of the first reflection, \( y_{2n+1} = L \). Similarly, all the reflections through the droplet can be taken at its center \( y_{2n} = -L \). The attenuation factor becomes a simple geometric series \( \sum_{n=1}^{\infty} (R/2L)^{2n} \) and the flow potential is the rescaled sum of two dipoles at \( y = -L \) and \( y = L \):

\[
w_d(z) = R^2 \delta u_0 \left( 1 - \frac{R^2}{L^2} \right)^{-1} \left[ \frac{1}{z + iL} + \frac{1}{z - iL} \right].
\] (28)

The force on a droplet near a single wall is obtained by summing over the dipoles inside the droplet (Eq. (15)). The part of the drag force due to the sidewall is, therefore:

\[
\frac{F_{\text{wall}}}{\delta u_0 \xi_d} = \sum_{n=1}^{\infty} g_{2n} = \left( \frac{\chi}{\ln \alpha} \right)^2 \left[ \psi_{\alpha}^{(1)}(2) - 2 \ln \alpha \right].
\] (29)
where $\psi^{(n)}_q(z)$ the $q$-polygamma function used here with $n = 1$ and $q = \alpha^2$. The droplet interacts with its own reflections, which exert a force directed along the flow such that the droplet moves faster than $u_q^\infty$. The resulting overall force on the droplet combines the force on a free droplet (Eq. (17)) and the reflections from the wall $F_{\text{drag}} = \frac{1}{2} \xi u_d + \xi_0 u_0 + F_{\text{wall}}$. Far from the wall, the force is governed by the first image, $F_{\text{wall}} \sim L^{-2}$.

2.23. A droplet confined between two walls

To quantify the degree of confinement of a droplet between two walls we define the confinement parameter $\gamma \equiv 2R/W$. The occurrence of three reflecting surfaces renders the summation over all images unfeasible. To approximate, we first treat the reflections of a point-like dipole from the two parallel walls, ignoring the droplet BC. These reflections form an infinite array of image dipoles perpendicular to the flow, along the $y$ direction (Lamb, 1932). After calculating the flow potential of the array $w_{\text{array}}(z)$, we rescale it by a factor $C$,

$$w_d(z) = C \cdot w_{\text{array}}(z), \quad (30)$$

to account for the boundary conditions on the finite-size droplet, which yields the approximated droplet potential.

The construction of the image dipole array induced by a dipole, $d_0 = R^2 \delta u_0$, located at the center of the channel, at $(x_d, y_d) = (0, 0)$, is illustrated in Fig. 6(a) as we add the images one by one: The image at $y = W$ cancels the flow penetrating the upper wall, but when the image at $y = -W$ is introduced, the combined field of the three dipoles penetrates both walls. We then add the image at $y = 2W$ to cancel the field of the $y = -W$ image on the upper wall, and add the image at $y = -2W$ to cancel the field of the $y = W$ on the bottom wall and so on, until an infinite array is constructed. If the droplet position is not at the center of the channel, $y_d \neq 0$, then the image array splits into two interlaced arrays of lattice constant $2W$, such that one is displaced by $y_d$ and one by $-y_d$ with respect to the array of the centered droplet (Fig. 6(b)).

Each dipole in the infinite array can be considered as a pair of positive and negative mass charges separated along the $x$ direction. Hence, far from the droplet, where $x \gg W$, the infinite array of dipoles can be seen as a charged parallel plate capacitor along $y$, with one plate consists of all the positive charges and the other plate of all the negative ones. The electrostatic field outside such a continuous capacitor vanishes. However, close to the capacitor the discreteness of its charges dictates that the field along the $x$ direction leaks out and decays exponentially as $\exp(-2\pi x/W)$ (Cui et al., 2002; Feynman et al., 1963). This hints that the flow field of a dipole confined between two sidewalls is also exponentially screened. Screening along the $x$ direction is a general property of the Laplace equation, in which boundary condition along a certain axis induces exponential dependence of the solution along the perpendicular axis.

When the droplet is at the center of the channel, $(x_d, y_d) = (0, 0)$, summation over the infinite array yields:

$$w_{\text{array}}(z) = R^2 \delta u_0 \left[ \frac{1}{z} + \sum_{n=1}^{\infty} \left( \frac{1}{z + inW} + \frac{1}{z - inW} \right) \right] = \frac{\pi R^2 \delta u_0}{W} \coth \left( \frac{\pi z}{W} \right). \quad (31)$$

To find $w_{\text{array}}(z)$ for the general case $y_d \neq 0$, we add up the potentials of two arrays (Fig. 6(b)) with lattice constant $2W$ (instead of $W$), such that the first starts at $y_d$ and the second at $(W - y_d)$. The resulting potential is:

$$w_{\text{array}}(z) = \frac{\pi R^2 \delta u_0}{W} \coth \left( \frac{\pi z}{W} \right) \left[ 1 - \frac{\sin(\pi y_d/W)}{\sinh(\pi z/W)} \right]^{-1}. \quad (32)$$

We use two methods to find the scaling factor $C$: When the droplet is far from both walls ($|y_d| \ll \frac{W}{2} - R$), we can assume that the reflections of all images in the infinite array fall at the droplet’s center with an attenuation factor proportional to their squared distances from the droplet. These reflections are then reflected back across the walls to form an additional array of attenuated reflections, which overlaps the original array, and so on. The sum of attenuated arrays of images results in a scaling factor $C$ (Eq. (30)):

$$C = \left[ 1 - \left( \frac{\gamma W}{4} \right)^2 + \frac{1}{3} + \sec^2 \frac{\pi y_d}{W} \right]^{-1}. \quad (33)$$

To approximate the general case, where the droplet is not necessarily very far from both walls, we calculate the scaling factor $C$, based on a method by Lamb (Lamb, 1932) that is generalized here for $y_d \neq 0$. We examine the overall potential, $w_{\text{oil}}(z) + w_d(z) = w_{\text{oil}}(z) + C \cdot w_{\text{array}}(z)$. Without walls, we have $C(\gamma = 0) = 1$, consistent with Eqs. (7), (8), (31) and (33) in the $W \to \infty$ limit. For $\gamma > 0$, we make use of the similarity of the flow around the array of image droplets to the potential flow perpendicular to a row of round pillars (Lamb, 1932). Since the fluid is incompressible and the total flux is fixed by the imposed $u_q^\infty$, the flow between the pillars is faster comparing to the single pillar case. Hence, we expect $C$ to increase with $\gamma$ and term $C$ the “incompressibility factor” (Beatus et al., 2007). We calculate $C$ using the fact that the two points at the north and south poles of the droplet, $(0, y_d - R)$ and $(0, y_d + R)$, belong to the same stream line, which has to split
and recombine in order to bypass the droplet. The stream lines are \( \psi(z) = iy \cdot u_\infty + C \cdot \text{Im}[w_{\text{array}}(z)] \) and by demanding \( \psi(iy_0 - iR) = \psi(iy_0 + iR) \) at the poles we obtain:

\[
C = \left( \frac{4}{\pi y} \right)^2 \left[ \cot(\pi y/4) - \frac{\sin(\pi y/2)}{\cos(\pi y/2) + \cos(2\pi y/4)} \right]^{-1}.
\]

(34)

In the \( y \to 0 \) limit, both (33) and (34) approach \( C \simeq 1 + (\pi y/4)^2 \left[ 1 + \sec^2(\pi y_0/W) \right] \). The dependence of \( C \) on \( y \) (Eq. (34)) clarifies when we set the droplet at the middle of the channel, \( y_0 = 0 \), for which \( C = (\pi y/2) \tan(\pi y/2) \). Under very weak confinement, \( y \to 0 \), we get \( C = 1 \) as in the unconfined potential (Eq. (8)), while under tight confinement, \( y \to 1 \), \( C \) diverges, consistent with the pillars intuition. The confined droplet potential is, therefore:

\[
\phi_d(z) = C \cdot \text{Re}[w_{\text{array}}(z)].
\]

(35)

The flow lines calculated using this potential are shown in Fig. 7. The asymptotic spatial dependence of the confined flow field \( \nabla \phi_d \) at the center of the channel \( (x_d, y_d) = (0, 0) \) is shown below for \( r = (x, 0) \):

\[
\begin{align*}
\partial_x \phi_d(x) & \sim \delta u_0 \gamma \tan(\pi y/2) \exp(-2\pi y x/W), & x \gg W \\
& \sim \gamma \tan(\pi y/2) \exp(-2\pi y x/W), & x \ll W, \\
\partial_y \phi_d(x) & \sim \delta u_0 \gamma \tan(\pi y/2) \exp(-\pi x/W), & x \gg W \\
& \sim \gamma \tan(\pi y/2) \exp(-\pi x/W), & x \ll W.
\end{align*}
\]

(36)

Close to the droplet, where \( x \ll W \), velocity decays as \( x^{-2} \) similarly to the unconfined dipole. However, for \( x \gg W \) the field is determined by two competing effects: an exponential spatial decay and the divergence of the incompressibility factor, \( \tan(\pi y/2) \), due to confinement. Under tight confinement \( (y \to 1) \) this divergence overcomes the screening. We note, though, that when \( y \sim (1 - h/R) \) the exact nature of the divergence may be affected by the sidewall boundaries since the flow is no longer an effective potential flow in 2D. Importantly, the screening length for the flow field in the \( y \) direction, \( W/\pi y \), is doubled compared to the \( x \) direction, which results in a substantial difference between the corresponding velocity components. Fig. 8 shows the oil velocity components \( v_x \) and \( v_y \) induced by a confined diplet located at \( (x_d, y_d) = (0, -R/200) \) under several confinement values, and normalized by the natural velocity scale in the system \( c_0 = K\delta u_0 \). Since \( y_d = 0 \) gives \( v_y = 0 \) along the midline \( y = 0 \), we set \( y_d \) to a nonzero value small enough for the droplet to fit into a tightly confined channel with \( y \leq 0.99 \). For \( y \leq 0.1 \) both velocity components are practically identical to the unconfined field with a \( 1/x^2 \) decay. Experimentally, this result indicates that for \( y \leq 0.1 \), the unconfined dipole field is a good approximation. For higher \( y \), the interplay between the exponential screening and the incompressibility factor results in a remarkable difference between \( v_x \) and \( v_y \). As \( y \) increases, \( v_x \) is heavily screened and only under extremely tight confinement \( (y \sim 0.99) \) the divergence of the incompressibility factor picks up to dominate. In contrast, since the screening length of \( v_y \) is double, the
incompressibility factor overcomes the spatially-dependent screening within range of a few droplet radii for all the shown values of \( \gamma \).

Very close to the droplet the approximated flow field deviates from the actual one, especially under tight confinement. Specifically, the calculated streamline that envelops the droplets forms a "lens" that meets the droplet's boundary only at its north and south poles (Fig. 7(b)). This streamline splits and recombines at two points outside the droplet, encircling an area of quiescent fluid that moves along with the droplet. The lens implies tangential discontinuity of the velocity field that is feasible in a potential flow. In the viscous flow of the experiment, which is only effectively a potential flow, such pattern was not observed as it would have implied infinite shear stress.

The equation of motion of a confined droplet is obtained from balancing drag and friction as in the unconfined case (Eq. (20)), \( F_{\text{drag}} + F_{\text{fric}} = 0 \). The drag force in a confined channel is: \( F_{\text{drag}} = \frac{1}{2} \xi_d u_d + C \xi_d^2 \delta u_0 \) (Eq. (13)). The first term due to the uniform oil flow is not affected by confinement. The \( C \geq 1 \) factor in the second term stems from the pole of \( w_d(z) \) at the droplet's center ("self pole"), which was scaled by \( C \). The larger drag can be seen as the result of the interaction of the droplet with its image dipoles, whose dipolar flow fields increase the oil velocity at the droplet's position.

The equation of motion reads \( u_d = K u_{\infty}^{\text{oil}} \), and the proportionality constant, \( K = C/[(C - 1/2) + \mu_d/\xi_d] \), can be calibrated as before by measuring the ratio \( u_{\infty}^{\text{conf}}/u_{\infty}^{\text{oil}} \) for an isolated droplet. How confinement affects the velocity of an isolated droplet is evident from the expression

\[
\frac{u_{\infty}^{\text{conf}}}{u_{\infty}^{\text{unconf}}} = \frac{K_{c=1}}{K_{c=1}} = 1 + \left(1 - \frac{1}{C}\right) \left(\frac{1 - 2 \mu_d/\xi_d}{1 + 2 \mu_d/\xi_d}\right). \tag{37}
\]

For high friction droplets (\( \mu_d > \frac{1}{2} \xi_d \)), such as the present measurements, confinement causes the droplet to move faster than an unconfined droplet. Low friction droplets (\( \xi_d < \frac{1}{2} \xi_d \)) move slower. For both regimes, at a limit of plug flow (\( \gamma \rightarrow 1, C \rightarrow \infty, K \rightarrow 1 \)) the droplet approaches the oil velocity, \( u_{\infty}^{\text{conf}} \rightarrow u_{\infty}^{\text{oil}} \).

To conclude, we derived the flow field of a single droplet flowing in a quasi-2D channel (\( h \ll R \)). Under the Darcy approximation the flow is effectively described as a 2D potential flow, such that the droplet's velocity perturbation is given by \( \nabla \phi_d \). We showed that the flow potential of an unconfined droplet is a 2D dipole and used this result to calculate the potential of a droplet confined between two sidewalls as an infinite sum of image dipoles. The confined flow field is determined by an interplay between screening of the sidewalls and the divergence of the incompressibility factor. In addition, we derived the two forces acting on a droplet: drag and friction. Since \( \text{Re} \ll 1 \) drag and friction balance out, leading to the droplet equation of motion (Eq. (20)), which sets a linear ratio between the droplet velocity and the local oil velocity.

3. The hydrodynamic interaction between droplets

3.1. The two-body hydrodynamic force and flow potential

To calculate the two-body hydrodynamic interaction, one needs to find the flow potential \( w(z) \) around two droplets by solving the Laplace equation \( (6) \), while demanding zero-flux through the boundaries of both droplets. Using the method of
Fig. 8. The interplay between screening and incompressibility. The spatial dependence of the droplet’s flow field is plotted for different values of the confinement parameter \( \gamma \). The longitudinal and transversal velocities, \( v_x \) and \( v_y \), induced by the droplet are calculated along the channel midline \((x > 0, y = 0)\) and shown in logarithmic scale. The droplet position is \((x_d, y_d) = (0, -R/200)\). We set \( y_d \neq 0 \) to get \( v_y \neq 0 \). Distance along \( x \) was normalized by \( R \) and the velocities by \( c_0 \).

(a) The exponential decay of \( v_x \) has screening length \( W/2\pi \), such that the divergence of the incompressibility factor dominates only under very tight confinement \( \gamma \ll 0 \). (b) In contrast, since the screening length of \( v_y \) is halved, the incompressibility effect becomes dominant even when \( \gamma \geq 0.25 \).

Fig. 9 shows the two-body flow potential obtained by considering an increasing number of image dipoles.

To calculate the two-body force, consider two droplets: droplet \( A \) at \((L, \theta - \pi)\) and droplet \( B \) at \((L, \theta)\). Let \( d_A \) and \( d_B \) be the lowest-order dipoles associated with the droplets (Eq. (9)). The positions of the \( n \)th image dipoles inside each droplet are \( z_n^A = y_{2n} e^{i\theta} \) for droplet \( A \) and \( z_n^B = -y_{2n} e^{i\theta} \) for \( B \), with the same \( y_{2n} < 0 \) used for a droplet near a single wall (Eq. (24)).

To find the orientation of each image dipole we notice a property of the reflection operation between two circular boundaries, such that all image dipoles inside \( A \) that originated from \( A \) have the same orientation as \( d_A \). In addition, all image dipoles inside \( A \) that originated from \( B \) have the same orientation angle \([\text{arg}(-d_B) + 2\theta]\) as in Eq. (22). The same property holds for the reflections inside \( B \). It is convenient to define a mirror operation \( M(d) \equiv -e^{2i\theta}d \), and the following sums that are based on the \( g \) coefficients from Eq. (25):

\[
G_{\text{even}} \equiv \sum_{k=0}^{\infty} B_{2k} = \left( \frac{x}{\ln \alpha} \right)^2 \left[ \frac{1}{4} \psi^{(1)}(\frac{1}{2}) - \ln \alpha \right]
\]

\[
G_{\text{odd}} \equiv \sum_{k=0}^{\infty} B_{2k+1} = \left( \frac{x}{\ln \alpha} \right)^2 \left[ \frac{1}{4} \psi^{(1)}(1) - \ln \alpha \right],
\]

where \( \psi^{(n)}(z) \) is the \( q \)-polygamma function used here with \( n = 1 \) and \( q = \alpha^4 (\chi = \sqrt{(L/R)^2 - 1} \) and \( \alpha = \chi + L/R \) as above).

In these notations, the hydrodynamic forces that the droplets exert on each other are:

\[
\frac{f_{BA}}{\xi_d R^{-2}} = G_{\text{even}} d_A + G_{\text{odd}} M(d_B)
\]
Fig. 9. Streamlines around two droplets calculated using different number of reflections \( N_{\text{ref}} \), from 0 to 3 (a–d) respectively. Blue dots indicate the main dipole of each droplet and gray dots represent reflected image dipoles. The streamlines that violate the boundary condition on the droplets are shown in red. For \( N_{\text{ref}} \geq 3 \) there are still streamlines that cross the droplet boundaries. They are not seen in the figure since this effect is minor. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

\[
\frac{f_{AB}}{E_d R^{-2}} = G_{\text{even}} d_B + G_{\text{odd}} \mathcal{M}(d_A),
\]

in which \( f_{AB} \) is the force that \( B \) exerts on \( A \). If \( d_A = d_B \), namely, the original dipoles move in the same direction and velocity, we get \( f_{AB} = f_{BA} \). The forces are identical both in magnitude and sign. Interestingly, the inter-droplet interaction does not satisfy Newton’s third law of reciprocal actions, since the viscous flow does not conserve momentum.

Similarly to the two-body interaction, the two-body flow potential can be obtained by summing the potentials of the two dipoles and all their images:

\[
w_{AB}(z) = \sum_{n=0}^{\infty} \left[ \frac{g_{2n}}{z - z^n_A} d_A + \frac{g_{2n+1}}{z - z^{n+1}_A} \mathcal{M}(d_B) + \frac{g_{2n}}{z - z^n_B} d_B + \frac{g_{2n+1}}{z - z^{n+1}_B} \mathcal{M}(d_A) \right]
\]

\[
= \frac{\chi e^{-i\theta}}{2R \ln \alpha} \left\{ \left[ \psi_q \left( \frac{1}{2} - \tau \right) - \psi_q \left( \frac{1}{2} \right) \right] d_A + \left[ \psi_q \left( 1 - \tau \right) - \psi_q \left( 1 \right) \right] \mathcal{M}(d_B) \right. \\
\left. - \left[ \psi_q \left( \frac{1}{2} + \tau \right) - \psi_q \left( \frac{1}{2} \right) \right] d_B - \left[ \psi_q \left( 1 + \tau \right) - \psi_q \left( 1 \right) \right] \mathcal{M}(d_A) \right\},
\]

with \( q = \alpha^{-4} \) and

\[
\tau \equiv \frac{1}{4 \ln \alpha} \ln \frac{z/R - \chi e^{i\theta}}{z/R + \chi e^{i\theta}}.
\]

A series solution for the 2D potential flow around two cylinders was also found by Mazur (Mazur, 1970), who used consecutive conformal maps. In this context it is worthwhile to mention the solution of Stimson and Jeffery for the viscous flow around two spheres (Stimson and Jeffery, 1926). Unfortunately, generalizing the method of reflections for three droplets or more is too cumbersome, since it involves enumerating the reflections created by more than two surfaces. This calls for a systematic approximation scheme.

### 3.2. Approximating the ensemble flow potential

We approximate the two droplets potential by considering only the first reflection of each droplet off the surface of the other (Fig. 9(b)). This scheme can be readily generalized for an ensemble of \( N \) droplets, such that the overall flow potential \( w(z) \) consists of the imposed-flow potential, a sum over the single-droplet potentials \( w_n(z) \) of the \( N \) droplets (Eq. (8)) and a sum over \( N(N - 1) \) first-order reflections \( w_{jn}(z) \) of each droplet off the surfaces of the remaining ones:

\[
w(z) = \bar{u}_{oil} z + \sum_{n=1}^{N} w_n(z) + \sum_{n=1}^{N} \sum_{j \neq n} w_{jn}(z).
\]
Fig. 10. Hydrodynamics interaction. The flow around three droplet was visualized by fluorescent micro-beads in the oil. The flow was from left to right. Superimposed are the drag forces exerted on the middle droplet by its two neighbors, along with their dipolar fields. Both neighbors exert drag forces with longitudinal component directed against the flow, reflecting the broken symmetry of the system.

The potential $w_{jn}(z)$ is due to the reflection of the $j$'th droplet off the $n$'th droplet, which induces a dipole $d_{jn}$ at $z_{jn}$,

$$w_{jn}(z) = \frac{d_{jn}}{z - z_{jn}}, \quad d_{jn} = \text{Res}[w_{jn}] = - \frac{\delta u_0 R^4}{r_{jn}^2} e^{2i\theta_{jn}},$$

in which $r_{jn} = |z_j - z_n|$, $\theta_{jn} = \text{Arg}(z_j - z_n)$ and the position of the reflected dipole is $z_{jn} = z_n + e^{i\theta_{jn}} R^2/r_{jn}$. The reflected dipole $d_{jn}$ is proportional to $\delta u_{jn}$, the induced flow field of the $j$'th droplet at $z_{jn}$, namely, $d_{jn} = R^2 \delta u_{jn}$.

3.3. Approximating the two-body interaction in an ensemble

The drag force on the $n$'th droplet consists of a term proportional to the sum of the residues (i.e., the sum of dipoles) inside the droplet, $\sum_{j \neq n} \text{Res}[w_{jn}] = d_0 + \sum_{j \neq n} d_{jn}$, and the uniform flow contribution $\frac{1}{2} \xi_d u_d$ (Eqs. (13)–(15)). The force exerted by the $j$'th droplet on the $n$'th one is:

$$f_{jn} = \frac{\xi_d}{R^2} \cdot d_{jn} = \xi_d \delta u_{jn}.$$  

The hydrodynamic interaction is proportional to the dipolar flow field of the $j$'th droplet as measured at reflected dipole $z_{jn}$. If droplets are far enough from each other ($r_{jn} \gg R$), then $z_{jn} \simeq z_n$ and

$$f_{jn} = \xi_d \delta u_{jn} = \xi_d \nabla \phi_d(r_j - r_n).$$  

The interaction decays as $1/r_{jn}^2$ and its dipolar shape is shown in Fig. 3. The direction of $\nabla \phi_d$ in Eq. (45) is the phase of $d_{jn}$ in Eq. (44). The forces are illustrated in Fig. 10, in which the dipolar fields of two droplets are superimposed on the measured flow field, showing the drag forces on the middle test-droplet.

In an ensemble of droplets the drag force on the $n$'th droplet (Eq. (13)) is given by summing over its interaction with all the other droplets that contribute $N - 1$ poles inside the droplet and adding the “self-pole” for its interaction with the uniform flow field. Eq. (45) indicates that one could derive the inter-droplet interaction by assuming that the droplets are far enough from each other ($r_{jn} \gg R$) such that the velocity field of droplet $j$ at $z_n$ is approximately uniform, and calculating the drag force of this uniform field (17).

Under the assumption that the droplets are sufficiently far from each other and, therefore, only the first reflections matter, the overall hydrodynamic force exerted on the $n$'th droplet is:

$$\mathbf{F}_{\text{drag}} = f_0 + \sum_{j \neq n} f_{jn} = \xi_d \cdot (\delta u_0 + \frac{1}{2} u_d + \sum_{j \neq n} \delta u_{jn}).$$  

where $f_0 = \frac{1}{2} \xi_d u_d + \xi_d \delta u_0$ is the drag due to the driving flow (Eq. (17)). By equating the forces, $\mathbf{F}_{\text{drag}} = -\mathbf{F}_{\text{fric}} = \mu_d \mathbf{u}_d$, we obtain the equation of motion of the $n$'th droplet in an ensemble:

$$\mathbf{u}_d(r_n) = K \mathbf{u}_{oil}(r_n)$$

(47)
The equation of motion (47)–(48) is analogous to that of an isolated droplet (20)–(21) where the oil velocity $u_{\text{oil}}(r)$ results from the imposed linear flow ($u_{\text{oil}}^\infty$) along with the perturbations of all the other ($j \neq n$) droplets. Note that under confinement $\phi_0$ depends explicitly on the positions of the two interacting droplets and not on their difference.

To get an idea about the accuracy of the first-order approximation, consider two droplets with distance $L$ between them (Fig. 9). The first reflection is attenuated by $R(L)$ with respect to the initial dipole and each subsequent reflection is further attenuated by $\mathcal{O}(R/L)^2$ with respect to the preceding one. For typical values of $R = 15 \mu m$ and $L = 75 \mu m$, the correction of the second-order reflection to the drag force is only 4% of the first-order. To this accuracy, the typical velocity perturbation that each droplet induces on the other, assuming typical values of $u_{\text{oil}}^\infty = 1250 \mu m s^{-1}$, $u_{\text{air}}^\infty = 250 \mu m s^{-1}$, is $\delta u \approx 40 \mu m s^{-1}$ with $L$ taken along the $x$ direction. The interaction drag forces on both droplets are the same in magnitude and direction $F = \xi_0 \delta u \approx -2.25 nN$, which is weaker by more than an order of magnitude than the force exerted by the main flow on each droplet (Section 2.1.2). According to the equation of motion (47), the interaction reduces the velocity of each droplet by $K \delta u \approx -8 \mu m s^{-1}$ with respect to an isolated droplet moving at $u_{\text{air}}^\infty$.

4. One-dimensional crystal: phonons, confinement and instabilities

The simplest droplet ensemble is a one-dimensional microfluidic crystal—an infinite ordered array of droplets with radius $R$ and constant distance $a$ between adjacent droplets (Fig. 1(a)) (Beatus et al., 2006). The droplet lattice positions are, therefore, $\{na, 0\}_{n=-\infty}^{\infty}$, and their actual positions $(x_n, y_n)$ may deviate from the lattice. We first consider the crystal in the unconfined limit $R \ll W$, which is practically met for $\gamma \leq 0.1$. The equation of motion for the position of the $n$th droplet $z_n = x_n + iy_n$ is obtained using Eqs. (8), (42), (44) and (47):

$$\ddot{z}_n = u_{\text{air}}^\infty - K \sum_{j \neq n} \frac{R^2 \delta u_0}{(z_n - z_j)^2}. \tag{49}$$

4.1. Collective drag reduction: the peloton effect

If the crystal has no perturbations, $z_n = na$ and $\dot{y}_n = 0$, but the droplet longitudinal velocity $\dot{x}_n$ is slower than $u_{\text{air}}^\infty$ and depends on $a$ due to the dipolar interactions between the droplets (Fig. 1(a), inset). Since all droplets are equivalent in this case, $\dot{x}_n$ is the same for all $n$ and identical to the crystal velocity $u_d(a)$. The dipolar forces exerted on the $n$th droplet by all the other droplets are directed opposite to the direction of flow and increase in magnitude as $a$ decreases, since the dipolar force decays as $1/r^2$. Therefore, $u_d(a)$ decreases as the crystal spacing decreases. Equivalently, with respect to the oil, the crystal moves faster as $a$ decreases, a result known as collective drag reduction. This is similar to sedimenting particles (Ramaswamy, 2001) as well as to a pack of cyclists (peloton), where riding closely helps them to reduce drag and ride faster. We, therefore, term this phenomenon the ‘peloton effect’. We find $u_d(a)$ self-consistently from Eq. (49) for a crystal without perturbations ($z_n = na$):

$$u_d(a) = u_{\text{air}}^\infty \left[ 1 + \frac{\pi^2}{3} \left( \frac{R}{a} \right)^2 \left( \frac{u_{\text{air}}^\infty - u_{\text{oil}}^\infty}{u_{\text{oil}}^\infty} \right) \right]^{-1}. \tag{50}$$

A measurement of the peloton effect (Fig. 11) was done by forming 1D crystals with different values of $a$ and measuring the mean longitudinal velocity of the droplets (Beatus et al., 2006). A similar effect was reported by Mcwhirter et al. (2009), who simulated the flow of deformable blood cells in a narrow 3D tube.

4.2. Phonon-like acoustic modes in a 1D unconfined crystal

To consider small fluctuations of the droplet positions around their lattice points, we define the displacement of the $n$th droplet as $\delta z_n = z_n - na$ and work in the frame-of-reference of the crystal that moves at $u_d(a)$. Assuming small deviations $|\delta z_n| \ll a$ we expand the equations of motion (49) to first order in $\delta z_n$ and obtain first-order wave equation:

$$\ddot{\delta z}_n = \frac{3C_s}{\pi^2 a} \sum_{j=1}^{\infty} \frac{\delta z_{n+j} - \delta z_{n-j}}{j^3}. \tag{51}$$

where the sound velocity $C_s$ is:

$$C_s = \frac{2\pi^2}{3} \left( \frac{R}{a} \right)^2 K \delta u_0. \tag{52}$$
The wave equation (51) has both longitudinal (along $x$) and transversal (along $y$) modes, whose dispersion relations are obtained by substituting a plane-wave $\exp[i(kx - \omega t)]$ with wave-number $k$ and frequency $\omega$:

$$\omega_x(k) = -\frac{6C_s}{\pi^2 a} \sum_{j=1}^{\infty} \frac{\sin(jka)}{j^3}$$

$$\omega_y(k) = -\omega_x(k).$$

The wave equation (51) describes phonon-like vibrational excitations that propagate along the crystal, with decoupled longitudinal and transversal modes. The dispersion relations are real, $\text{Im}[\omega(k)] = 0$ for all $k$ values, corresponding to linearly stable oscillations without damping. The sound velocity $C_s$ is the velocity in the long wavelength limit ($k \to 0$), where the equations of motion reduce to simple, first-order wave equations: $\partial_t x_n = C_s \partial_x x_n$ and $\partial_t y_n = -C_s \partial_x y_n$. In these wave equations long longitudinal waves propagate backwards at $C_s$ and long transversal waves propagate forward at the same velocity. The sound velocity $C_s$ (Eq. (52)) reflects the two-body hydrodynamic interaction: the term $R^2 \delta u_0$ is the magnitude of the single-droplet dipole, $1/a^2$ stems from the $1/r^2$ decay of the interaction and the summation over all interaction along the crystal results in a harmonic series which contributes the numeric factor, finally, the ratio $K = u_0^\infty / u_0^\infty$ couples the droplet velocity to the oil velocity in the equation of motion.

The dispersion relations were measured by following the moving crystal in its frame-of-reference, tracking the positions of droplets in time and applying Fourier transforms in space and time to these data (Movie 2). Thus, the power-spectra of both longitudinal and transversal vibrations in terms of $k$ and $\omega$ were obtained. In order to apply Fourier transforms it is required to measure the deviations of the droplets, $(\delta x, \delta y)$, from their unperturbed crystal positions $(na, 0)$. In the $y$ direction this is trivial, since the whole crystal is, by definition, at $y = 0$. Nevertheless, since the channel and crystal were never perfectly aligned with the camera, we calculated a linear fit of the droplets positions $(x_n, y_n)$ and subtracted the fitted $y$ value from each $y_n$. In the $x$ direction it is hard to determine the unperturbed positions of the moving, vibrating crystal. For this purpose we defined a new observable as the longitudinal distance between adjacent droplets $x_n' \equiv x_{n+1} - x_n$. Here $x'$ is the vibrating function for which we calculate a power-spectrum and $n$ is the spatial lattice coordinate for the discrete Fourier
Fig. 12. Intensity plots of the logarithm of power-spectrum of longitudinal (a) and transversal (b) waves as a function of wave-vector and frequency (k, ω). The corresponding dispersion relations (c, d) are the peaks of the power spectrum. The skewed sine-like curve is due to the hydrodynamic interactions. The red line in the theoretical calculation for ω(k) with no adjustable parameters. The black straight line ω(k) = −udk is due to droplet deflections by stationary defects along the channel. (e, f) are images of longitudinal (e) and transversal (f) waves. Scale bars are 50 µm. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

The measured power-spectra are shown in Fig. 12(a), (b), and sample images of longitudinal and transversal phonons are shown in Fig. 12(e), (f). The colors in the power-spectra plots represent the amplitude of each combination of (k, ω) shown in a log10 scale. The peaks of the resulting power-spectra define the phonons dispersion relations ω(k) (Fig. 12(c), (d)). The main feature of the dispersion is a skewed sine-like curve with the following properties: (a) Both dispersion relations are anti-symmetric in k, ω(k) = −ω(−k). (b) The longitudinal and transversal dispersion relations are identical in magnitude and form, but have opposite signs, ωx(k) = −ωy(k). Consequently, the group velocity ∂ω/∂k has the same inversion property. (c) Around k = 0 we have ωx(k) = −Csk and ωy(k) = Csk, which corresponds to acoustic waves with a sound velocity Cs ≈ 250 µm s−1. Long longitudinal waves travel backwards at velocity Cs, against the direction of flow, and long transversal waves travel forward, along the flow, also at Cs. (d) Close to the edges of Brillouin zone at k = ±π/a the acoustic waves travel oppositely to the long waves at velocity Cs/2 and have zero frequency. These shortest waves correspond to the pairing (longitudinal) wave and the zigzag (transversal) wave. (e) Standing waves appear at the middle of Brillouin zone and not at its edges, as occurs in a 1D harmonic crystal. The standing waves are the modes with highest frequency in the system ~1 Hz. The standing wavelength is not an integer number of droplet but is about 4.732a. The theoretical predictions for ω(k) (Eq. (53)) were calculated for the experimental results with no fit parameters. They are plotted in red lines in Fig. 12(c), (d), showing very good agreement with the measurement.
In both longitudinal and transversal spectra, the largest amplitudes of the phonon are found around $k = 0$ and $k = \pi / a$. The transversal amplitudes are about 2 orders of magnitude weaker than the longitudinal ones. Had the transversal amplitude been higher the crystal would likely break-up as often observed (Section 4.5). The lower transversal amplitudes are also related to the fact that the transversal spectra are typically more noisy than the longitudinal ones, since errors in droplet tracking become more significant as oscillation amplitudes decrease.

A secondary feature of the dispersion relations is a straight line $\omega(k) = -u_d k$, which stems from stationary defects along the channel. In the frame of the moving crystal the defects seem to be moving backwards. These defects deflect the droplets, creating waves that travel at velocity $-u_d$ for all $k$ values. Hence, this obvious linear dispersion is not related to the hydrodynamic interactions between the droplets.

The summation in Eq. (53) is over the interactions with the $j$'th neighbors on the right and left sides, as in the equations of motion (51). To show the importance of the number of neighbors considered in this sum we plotted the wave velocities $\partial \omega / \partial k$ for long waves ($k \to 0$) and short waves ($k = \pi / a$), assuming the interaction cuts off after a finite number of neighbors (Fig. 13). When taking into account only nearest neighbor interactions both velocities are around $0.6 C_s$ since the only term in $\omega(k)$ is $\sin(ka)$, which has equal slopes at $k = 0$ and $k = \pi / a$. As the summation includes more distant neighbors, the velocities of the long and short waves approach $C_s$ and $\frac{1}{2} C_s$ respectively, thus regaining the skewness of $\omega(k)$. The approach, however, is rather slow. For example, to reach $0.95 C_s$ the summation of the long waves velocity must include the 12 nearest neighbors, which demonstrates the importance of long-range interactions in the crystal.

### 4.2.1. Intuition for long ($k \to 0$) and short ($k = \pi / 2$) waves

An intuitive explanation for the velocity of longitudinal waves is provided by the pelotoneffect. Consider a crystal in which a pack of droplets are more densely spaced than the remaining crystal as shown in Fig. 11(b). Owing to the peloton effect, this pack moves slower than the rest—droplets behind the pack catch up, whereas droplets at the front escape. The denser part of the crystal, therefore, travels opposite to the flow, resulting in a traveling density wave. A similar argument explains why long transversal waves travel along the flow (Beatus et al., 2006).

Simple configurations of dipoles also clarify why the shortest modes of the crystal, the zigzag and pairing waves, have zero frequency (Fig. 14). These modes are shown, for example in Figs. 12(e) and 19 respectively. First, note that the transversal forces on a test droplet (Fig. 14(a), (b), gray) due to its two neighbors (red and blue) in the isosceles triangle configurations cancel out. It follows that an infinite zigzag wave can be decomposed into similar red–blue pairs such that the total transversal force on a given test droplet is zero. For example, in the zigzag in Fig. 14(e) the transversal forces exerted on the test droplet by the droplets located $\pm a$ from it cancel and so do the forces by the pairs at all odd multiples of $a$: $\pm 3a$, $\pm 5a$ and so on. The droplets at the even positions do not exert any transversal force since they are at same $y$ position as the test droplet. This argument can be applied to any droplet in the infinite zigzag wave using either one of the configuration in Fig. 14(a) or (b), which implies that all droplets are equivalent and are not subject to any transversal force. Therefore, given an infinite zigzag wave it does not oscillate and has zero frequency.

Similar argument is given for the longitudinal pairing wave: In the two triplets shown in Fig. 14(c), (d) the test droplet is subject to the same sum of longitudinal forces exerted by its two neighbors. The reason is that the dipolar force is proportional to $-x^{-2}$, independent whether the test droplet is on the right or the left side of the corresponding neighbor. In Fig. 14(c) the blue neighbor is closer and exerts a stronger force to the left than the red neighbor. In Fig. 14(d) the distances flip but the sum of forces is the same. Hence, given an infinite pairing wave (Fig. 14(f), (g)), it can be decomposed into similar red–blue pairs, such that the longitudinal drag force exerted on any given droplet has the same magnitude and direction.
opposite to the flow. Therefore, all droplets move slower by the same amount and are stationary with respect to each other, which implies zero frequency.

The intuition given here is valid for infinite crystals, while in finite crystals we observe instabilities related to the two shortest modes, for example, near the droplet formation area (Section 4.5).

4.2.2. Symmetry properties of $\omega(k)$

The anti-symmetry of the dispersion relation $\omega(k) = -\omega(-k)$ stems from the broken symmetry of the system, which is induced by the imposed flow field $u_{\text{oil}}^\infty \hat{x}$. The transformation $k \rightarrow -k$ is equivalent to the inversion $x \rightarrow -x$ in real space, and since the equations of motion invert their sign under the spatial inversion, so does $\omega(k)$ upon changing $k \rightarrow -k$. For example, if a wave with a given wavevector $k$ propagates forward at velocity $\partial^2 k \omega > 0$, then the wave with $-k$ will propagate at the same direction and velocity. This behavior is opposite to the propagation of waves in harmonic crystals, where the equations of motion are invariant under inversion and the waves with $\pm k$ travel in opposite directions.

The $xy$ inversion property of the unconfined dispersion relations $\omega_x(k) = -\omega_y(k)$ is related to the translation invariance of the dipolar interaction without sidewalls ($W = \infty$). For small displacements $(\delta x_n, \delta y_n)$ this invariance implies: $\delta x_n \propto \sum_{j \neq n} \delta(\partial_x \phi_d(r_n - r_j)) \approx \sum_{j \neq n} (\delta x_n - \delta x_j) \partial^2_x \phi_d((n - j)\alpha)$ and similarly $\delta y_n \propto \sum_{j \neq n} (\delta y_n - \delta y_j) \partial^2_y \phi_d((n - j)\alpha)$. Since $\phi_d$ satisfies the Laplace equation $\partial^2 \phi_d = -\partial^2 \phi_d$ (equivalently, since $w(z)$ is analytic), the equations of motion for $(\delta x_n, \delta y_n)$ are identical up to a sign and, hence, so are the corresponding dispersion relations. For finite $W$ this is no longer the case, as shown in the following section.

4.3. The phonons under confinement

Under confinement, the sidewalls break the translational invariance along $y$ and the force between two droplets depends explicitly on their $y$ coordinates, not only on the difference $r_n - r_j$. We, therefore, expect that under confinement $\omega(k)$ will not have the $xy$ inversion property (Beatus et al., 2007).
Fig. 15. Microfluidic crystals under confinement. (a) Droplet formation in confinement of $\gamma = 0.62$. Note the elongated droplet that fills the channel connecting the T-junction and the wider main channel. The droplet blocks the connecting channel, thereby increasing its resistance to flow. In this geometry, only after the droplet enters the main channel can the next droplet emanate. This effect introduces additional coupling between the crystal and channel parameters. (b) Longitudinal waves in $\gamma = 0.58$. (c) Transversal waves in $\gamma = 0.46$. Scale bars are 100 $\mu$m.

The inter-droplet force changes dramatically as a result of the interplay between hydrodynamic screening and the divergence of the incompressibility factor (Eq. (36), Fig. 8). Moreover, the longitudinal and transversal forces have different screening lengths and the interaction between droplets is no longer translational invariant since the droplet’s flow field explicitly depends on its $y$ coordinate. To study the effects of confinement on the phonon spectra we calculate the droplets’ equations of motion, as before, by using Eqs. (47) and (48) with the confined flow potential (Eq. (35)). After expanding in small perturbations ($\delta x, \delta y$) $\ll a$ we obtain:

$$\dot{x}_n = -2B \sum_{j=1}^{\infty} (\delta x_{n+j} - \delta x_{n-j}) \coth(\pi j \beta) \text{csch}^2(\pi j \beta)$$

$$\dot{y}_n = \frac{B}{2} \sum_{j=1}^{\infty} (\delta y_{n+j} - \delta y_{n-j}) [3 + \cosh(2\pi j \beta)] \text{csch}^2(\pi j \beta).$$

where $B \equiv c_0(\pi^2 R/W^2) \tan(\pi \gamma/2)$ is related to the incompressibility factor (Eq. (34)), $\gamma \equiv 2R/W$ as before, $\beta \equiv a/W$ and $c_0 = k\delta_0$. In contrast to the unconfined crystal, the confined wave equations of the two polarizations are substantially different from each other due to the breaking of translational invariance. Substituting propagating plane solutions we obtain the confined dispersion relations:

$$\omega_x(k) = -4B \sum_{j=1}^{\infty} \sin(jk a) \coth(\pi j \beta) \text{csch}^2(\pi j \beta)$$

$$\omega_y(k) = B \sum_{j=1}^{\infty} \sin(jk a) [3 + \cosh(2\pi j \beta)] \text{csch}^2(\pi j \beta).$$

Unlike the unconfined dispersion relations, here the inversion property between $\omega_x$ and $\omega_y$ does not hold. Rather, we have $|\omega_x(k)| \leq |\omega_y(k)|$, since for all $k$, every term in the sum of $\omega_x(k)$ is smaller or equal to the corresponding term in $\omega_y(k)$.

To measure the phonon spectra of crystals under different degrees of confinement, a series of microfluidic devices were fabricated with different widths of the main channel, from $W = 250 \mu$m down to 40 $\mu$m (Fig. 15). A single device could not suffice to obtain an arbitrary combination of $R$, $a$ and $\gamma$. These crystal parameters are coupled through the droplet formation process as well as the flow resistance of the output channel due to the droplets (Engl et al., 2005; Sullivan and Stone, 2008). In addition, the T junction parameters had to be tuned, namely its width and the length of the channel connecting the junction and the output channel (Fig. 15(a)), which affect droplet size and formation frequency. Specifically, a shorter connecting channel results in smaller $a$, since each droplet evacuates the connecting channel faster, thus reducing its flow resistance and enabling the formation of the next droplet.

The spectra of confined crystals from $\gamma = 0.09$ to 0.9 were measured by moving in frame with the crystal at $u_d$ and tracking the droplet positions as before. The measured phonon spectra are shown in Fig. 16, along with the theoretical predictions of the confined (Eq. (55)) and unconfined (Eq. (53)) dispersion relations. Under weak confinement of $\gamma = 0.09$ (Fig. 16(a), (b)) the longitudinal and transversal dispersion relations were identical to those of unconfined crystals, namely, anti-symmetric, skewed, sine-like curves, $\omega_x(k) = -\omega_y(k)$, with slope $C_i = 165 \mu$m s$^{-1}$ at $k = 0$ and $-C_i/2$ at $k = \pm \pi/a$. The measured spectra coincide with the theoretical predictions for both unconfined and confined crystals, confirming that confinement effects are negligible at least for $\gamma \leq 0.09$ (Fig. 8).

Under tighter confinement, the general shape of the sine-like curve remained. However, the longitudinal–transversal inversion property was broken: $|\omega_x(k)| < |\omega_y(k)|$ for all $0 < |k| < \pi/a$ (Fig. 16(c)–(g)). This effect is summarized in Fig. 16(h), where we plotted the longitudinal and transversal sound velocities, $C_{i,x}$ and $C_{i,y}$, normalized by the theoretically computed unconfined $C_i(\gamma = 0)$. For the longitudinal phonons, $C_{i,x}$ decreased as $\gamma$ increased. For $\gamma \geq 0.45$ the longitudinal sine-like curve fell below our detection limit (data not shown). Oppositely, for the transversal phonons $C_{i,y}$ increased with $\gamma$. The amplitude of the phonons decreased exponentially with $\gamma$ (Fig. 16(i)), essentially undetectable for $\gamma > 0.63$. The computed dispersion relations are superimposed on the experimental data (Fig. 16). The theory recapitulates the breaking of the $xy$ anti-symmetry and fits the data satisfyingly without any adjustable parameters. The model is somewhat more
Fig. 16. Confined phonons spectra. (a-g) Transversal (left column) and longitudinal (right column) phonon power-spectra for increasing values of the confinement parameter $\gamma$ (shown on each plot). Color code represents the logarithm of the amplitude. White curves show our theory for $\omega(k)$. Black curves indicate the corresponding theoretical $\omega(k)$ for unconfined crystals ($\gamma = 0$). Dashed line is $\omega(k) = -\nu k$. (h) $C_s(\gamma)/C_s(\gamma = 0)$ for both polarizations. $C_i(\gamma = 0)$ was calculated using Eq. (53) with the same flow parameters as the corresponding confined crystal. (i) Mean amplitude of the transversal modes as a function of $\gamma$. 
accurate for transversal modes than for longitudinal ones. We note that according to this model $\zeta$ diverges as $\gamma \to 1$ as well as the frequencies of oscillations. In this limit the droplets block the channel (plug-flow) and since the oil is incompressible, it is difficult to push liquid through the narrow necks. In order to excite a phonon, it is required to move droplets as well as transfer oil through the necks in the opposite direction. Hence, as $\gamma \to 1$ oil incompressibility implies that phonons are harder to excite and expected to have reduced amplitudes, which is consistent with our observations (Fig. 16(i)).

4.4. Sources of phonon excitation

Both with and without confinement, the measured spectra show that the phonons are excited at all wavelengths. The reason for phonon excitation cannot be thermal noise, since the droplets are too large and over-damped such that thermal fluctuations are utterly negligible (see the estimation of the droplet thermal diffusion coefficient in Section 2.1.4). The sources of noise are most probably: (a) stationary defects along the channel that, as shown above, deflect the droplets and excite traveling waves of all wavelengths that propagate at $-u_d$; (b) the asymmetry at the droplet formation area, where the crystal is effectively semi-infinite, results in force asymmetry in both longitudinal and transversal directions. This force asymmetry reflects in droplet motion and may even induce crystal instability as shown below; and (c) the droplet formation process itself, namely the periodic, sudden injection of droplets perturbs the system.

4.5. Nonlinear instabilities of the 1D crystal

The crystal exhibits instabilities, which occur upon large-amplitude fluctuations $\delta x, \delta y \sim a$ and lead to breaking of the crystalline order. Since the crystal is linearly stable ($\text{Im}[\omega(k)] = 0$), these instabilities can only be understood when considering coupling between different vibrational modes. Instabilities appeared both with and without confinement, though here we refer to instabilities we studied in practically unconfined crystals. The instabilities are related to shortest modes of the crystal: the longitudinal pairing and transversal zigzag waves. These modes are special also because they have finite velocity of $\pm C_s/2$ with respect to the crystal but zero frequency (Section 4.2.1). A similar phenomenon occurs in 2D dusty plasma crystals that are also governed by dipolar interactions: crystal instability and melting arise in this system from the nonlinear interaction between two longitudinal and transversal phonons that have the same wavenumber and frequency (Couèdel et al., 2010; Ivlev et al., 2003; Liu et al., 2010; Sheridan, 2008; Thomas and Morfill, 1996).

4.5.1. The 1 + 3 instability

In most experiments, as the crystal was flowing downstream from the droplet-formation area, phonons amplitude increased in time. Initially, the increase is due to some excitation (Section 4.4) and when amplitude is large enough, nonlinear effects of the dipolar interaction become significant and further increase the amplitude. In the 1 + 3 instability a local fluctuation grows in the middle of the crystal, far from the T junction, and breaks the crystal (Fig. 17(a), Movie 3). Interestingly, in most experiments, in which the crystal broke far from the T junction, this local fluctuation had a distinct structure of a single droplet followed by an oblique row of three droplets. In addition, the 1 + 3 structure had typical dynamics of a few oscillations with increasing amplitude that led to breakup after 1–2 s.

As a first step towards tracing its origin, we used our numerical simulation (Appendix B) to reproduce the 1 + 3 instability under controlled initial conditions. For example, we calculated the dynamics of a crystal with two initial wave packets—a short droplet sequence carrying a pairing wave followed by a similar pack with a zigzag wave, with a gap of a few unperturbed droplets in between the two packets (Fig. 17(b)). The packets propagate in velocity $\sim C_s/2$ towards each other and their collision resulted in a 1 + 3 instability. In fact, we noticed that similar breakup can be obtained also without the pairing wave packet, due to nonlinear interactions at the rear edge of the zigzag packet. The nonlinear interactions independently generate a longitudinal mode that interacts with the rest of the zigzag package, leading to a similar 1 + 3 breakup.

4.5.2. “Surface” melting

Our theory so far considered an infinite crystal although experimentally it is finite, as it extends from the T junction at $x = 0$ to the outlet of the channel. Even without perturbations the force balance on a droplet near the T junction is different than far from this point of asymmetry. Far enough from the formation zone the crystal can be treated as approximately infinite and the force on a droplet is given by summing the contributions of its neighbors from both sides, as calculated for the peloton effect (Eq. (50)). The jth neighbor to the left side of the droplet and its counterpart on the right, both exert the same longitudinal force opposite to the flow, which decays as $(ja)^{-2}$. The sum of all interactions is directed against the flow and hence slows down the droplet comparing to an isolated one. However, the leftmost droplet at $x = 0$ has only neighbors on its right, therefore, the force exerted on it by the other droplets is halved and this droplet moves faster than droplets in the middle. Indeed, droplets near the formation point move faster, as seen in Movie 3: closer to the T junction inter-droplet distances are larger, and since their formation rate is constant, it implies they move faster. Applying the same argument for the other end of the crystal, where the rightmost droplet has neighbors only to its left, we see that, again, the last droplet moves faster. Using the properties of the harmonic series, we verified that a finite crystal well-approximates an infinite one,
Fig. 17. Crystal breakup. (a) Subsequent images, taken 0.3 s apart, of a local fluctuation in a 1D crystal (1 + 3 droplets, marked). The fluctuation grows and leads to crystal breakup. Scale bars are 100 µm. (b) Similar dynamics was obtained in a numerical simulation of the nonlinear equations of motion. In this example, the initial condition was a low-amplitude zigzag wave assigned to the green droplets and a low-amplitude pairing wave assigned to the red droplets. The two packets propagated in $\sim C_s/2$ towards each other, and their nonlinear interaction leads to breakup. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fig. 18. “Surface” melting. (a–c) Crystal instability at the tailing edge that occurred upon stopping droplet formation and maintaining the flow of oil. The instability appeared as a longitudinal pairing wave and wandering motion in the transversal direction. Scale bars are 100 µm. (d) This instability was also obtained in numerical simulation.

when considering the droplets farther then $\sim 60a$ from its two edges (about a frame width in our experiments), since the sum of interaction forces on the 60’th droplet is weaker by 1% than on a droplet in an infinite crystal.

To experimentally isolate the effects of asymmetry at the trailing edge, the crystal was cut abruptly by stopping droplet formation while maintaining the flow of oil. The truncated, flowing crystal exhibited a longitudinal pairing wave that advanced from the edge downstream and a wandering motion in the transversal direction (Fig. 18, Movie 4). Pair formation results from the longitudinal asymmetry and the peloton effect: the first (leftmost) droplet has fewer neighbors to slow it down, thereby it catches up with the droplet ahead to form the first pair. The interaction within the pair slows it down with respect to the crystal. When the pair lags further behind, the third droplet becomes effectively the first droplet of the remaining crystal, thus a cascade of pair-formation ensues. The transversal wandering results from sensitivity to small transversal fluctuations of the leftmost droplet. Any small deflection grows because the first droplet is pushed in the same direction by all the other droplets, with no neighbors behind it to cancel these forces. At the same time, the first droplet pulls the second one off the crystal axis. This deflection avalanche advances and the entire crystal drifts aside. This instability was also obtained in simulation.
4.5.3. The zigzag instability

Another instability caused by the asymmetry at the formation point is the zigzag instability, in which the crystal bifurcates into two parallel crystals (Fig. 19, Movie 5). Similarly to the transversal wandering in the “surface melting” instability, a small transversal deflection of the leftmost droplet is increased due to the transversal force imbalance. In contrast to the previous instability, here droplets are formed continuously and when the next droplet emanates, it is pushed transversally in the opposite direction by the (now) second droplet, and so on. Hence, small perturbations near the T junction excite a zigzag mode there. In order to explain the growth of the zigzag amplitude, consider the transversal forces exerted on the droplets within a small-amplitude zigzag wave that already exists near the formation area. The trailing-end asymmetry implies that closer to the formation area, zigzag amplitude grows faster (Beatus et al., 2006).

The growth of the zigzag mode can be also obtained assuming nearest-neighbors interactions in a low-amplitude zigzag wave. By linearizing the transversal force between the first and second droplets we get that the transversal distance between them satisfies \( \delta_t (y_2 - y_1) \approx \left[ 2(R^2/a^2)K_0u_0 \right] (y_2 - y_1) \), namely, it grows exponentially until the next droplet emanates. The exponential growth can be expressed also as a function of the distance \( x \) from the formation point as \( A(x) = A_0 \exp[2(R^2/a^2)(1 - K)x] \) with \( A_0 = A(x = 0) \). Fig. 20 shows simulation results of crystals with different values of \( a \) that were subject to random noise at the formation point. Plotting the spatial growth \( dA/dx \) from the simulation shows it scales as \( 1/a^3 \) as expected from the nearest-neighbors calculation. Notably, a finite microfluidic crystal has another area of asymmetry at its leading end, where droplet exit the main channel. Using a similar argument it can be shown that at the leading end zigzag amplitude decreases at the same slope, which is also captured in simulation.

5. Two-dimensional disordered ensemble: shock waves and sound

The phonons are entirely tractable theoretically due to the first-reflection approximation, the 1D crystalline order and the small fluctuations of droplets motion. Furthermore, the theoretical dispersion relations fit the experiment with no adjustable parameters and even the nonlinear instabilities of the crystal can be understood by considering a superposition of dipolar fields. In contrast, the many-body problem is far more complex when dealing with the dynamics of a 2D disordered ensemble. In such ensembles, individual droplets move erratically, with large amplitude fluctuations and dynamic clustering, in which droplet clusters form and break dynamically (Fig. 21, Movie 6).

2D droplet ensembles were generated using microfluidic devices with a main channel wider than previously described (\( W = 500–800 \mu m \)), while droplet size was roughly the same. The confinement parameter was, therefore, relatively small \( \gamma \leq 0.05 \) and the channel was able to accommodate more than 20 droplets across its width. We characterize the 2D
Fig. 21. Two-dimensional disordered ensemble of droplets. (top) Droplets flowing in a 500 µm width channel with mean area fraction $\rho_0 = 0.23$. To determine $\rho_0$, the area fraction was averaged over all frames in a given experiment. Red vectors show the velocity of some of the droplets with respect to the mean droplet velocity $u_d$. Typical droplet velocity was $\sim 100$ µm s$^{-1}$ and oil velocity was $\sim 400$ µm s$^{-1}$. Scale bar is 100 µm. (bottom) The 1D density profile $\rho(x, t)$ corresponding to the image. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

ensemble by its mean number density of droplets $n_0 \equiv N/A$ and mean area fraction $\rho_0 \equiv n_0 \pi R^2$, where $N$ is the number of droplets in a large channel area $A$, typically a frame size. $\rho_0$ was varied between 0.06 to 0.6 by setting the water and oil pressures.

5.1. Velocity–density coupling

Since the 2D ensemble is disordered, the simplest way to theoretically analyze its collective dynamics is within a mean-field approach, in which we neglect its discrete nature and the rather complex dynamics of individual droplets and droplet clusters. We, therefore, treat the ensemble as a smooth distribution of droplets with an average number density of $n_0$ droplets per unit area. We first assume that the density is uniform and calculate the effect of this ensemble on a single test-droplet at $r_t = (x_t, 0, y_t)$. Namely, we calculate the velocity change along the flow, $\Delta u_x$, of the test droplet due to its interactions with a slab of a uniform droplet distribution, which extends between $x_L < x_t$ and $x_R > x_t$ along the flow and across the whole channel along $y$ (Fig. 22).

In the weak confinement limit $\gamma \ll 1$ we have $C = 1 + O(\gamma^2)$, which is valid across most $y$ values. Very close to the sidewalls $C = \frac{1}{2} + O(\gamma^2)$ but this occurs only within a distance of $O(R)$ from each wall, hence it is negligible. Since $C \approx 1$, the flow potential of the droplets in the uniform distribution is well approximated by the array potential $w_{array}$ (Eq. (32)). The array potential is a summation over the infinite array of image dipoles resulting from the reflections of a single droplet

\[ w_{array} = \sum_{i=1}^{\infty} \frac{1}{4\pi} \frac{\rho_0}{x_i - x_t} \]
between two sidewalls (Fig. 6, Eq. (31)). The flow potential of the whole uniform distribution is a sum of the \( w_{\text{array}} \) potentials of all its droplets. If we consider each \( w_{\text{array}} \) as its constituents: one actual dipole and its infinite series of image dipoles, we find that the slab of uniform droplet distribution is equivalent to an infinite slab of unconfined dipoles that extends from \(-\infty\) to \(\infty\) along \(y\) and from \(x_l\) to \(x_b\) along \(x\). Namely, each reflection of a uniform droplet distribution is also a uniform distribution of image dipoles (Fig. 22). We neglected the fact the original distribution avoids a disc of radius 4\(R\) centered around the test droplet. To account for this 'hole' one should subtract its contribution to \(\Delta u_x\) by considering its reflections. However, the hole's contribution is of the order of \(-\pi K \delta t_0 y^2\) and, therefore, negligible.

Under the first-reflection approximation we calculate \(\Delta u_x\) by integrating the contributions of the dipoles in the uniform distribution (using Eqs. (43), (47) and (48)):

\[
\Delta u_x = -KR^2 \delta t_0 \rho_0 \times \left[ \int_{-\pi}^{\pi} \frac{1}{2R} \int_{-\pi}^{\pi} \frac{1}{2R} \int_{-\pi}^{\pi} \frac{1}{2R} \int_{-\pi}^{\pi} \cos \frac{2\theta}{r^2} rdrd\theta \right] = c_0 \rho_0,
\]

with \(L_1 = x_b - x_l\), \(L_2 = x_l - x_t\) and \(c_0 = K \delta t_0\) as before. This calculation is accurate within \(O(y^2)\), since we neglected the contribution of the 'hole'.

Eq. (56) constitutes a linear coupling between the velocity change of the test droplet and the ensemble density. Similar coupling appears also in non-Brownian sedimentation (Batchelor, 1972; Brenner, 1999). Interestingly, the result is independent of \(W, y_t\) as well as of \(x_l\) and \(x_b\), which implies that it is valid locally and depends only \(\rho_0\) in a slab of width 4\(R\) centered around the test droplet. In fact, a similar calculation shows that even if the rest of the channel has different droplet density, \(\Delta u_x\) is still determined only by \(\rho\) in the slab surrounding the test droplet. Hence, we can express the local material velocity as a function of the local density: \(u(x, t) = u_0^0 + c_0 \rho(x, t)\), in which \(u_0^0\) is, as before, the velocity of an isolated droplet.

The independence of \(\Delta u_x\) of the channel width is quite peculiar, since at the \(W \to \infty\) limit it does not coincide with \(\Delta u_x, \text{unconf}\), the velocity change due to a uniform droplet distribution in an unconfined, infinite channel. To calculate \(\Delta u_x, \text{unconf}\) we set for simplicity \(x_l = y_t = 0\), integrate \(\partial_t \phi_0\) from Eq. (8) over the whole channel excluding a 4\(R\) disc around the test droplet and get that the velocity change vanishes due to symmetry:

\[
\Delta u_x, \text{unconf} = c_0 \rho_0 \int \int \frac{y^2 - x^2}{(x^2 + y^2)^2} dx dy = 0.
\]

This can be seen also from the symmetry of the unconfined dipole by dividing the droplet ensemble into pairs such that the velocity contribution of each pair to the test droplet cancels out. Consider, for example pairs of the form \((\pm|x_0|, \pm|y_0|)\) and \((\pm|y_0|, \pm|x_0|)\), as shown in Fig. 23(a) for \(y_0 = 0\). However, under confinement the symmetry within each pair is broken due to screening, such that the pair contributes a net positive velocity to the test droplet. For example, in Fig. 23(b) the field of the red \((|x_0|, 0)\) droplet at the test droplet's position weakens comparing to the unconfined case, while the field of the blue \((0, |x_0|)\) droplet strengthens due to the incompressibility factor. The source of the velocity–density coupling is, therefore, the sidewall boundaries and this coupling has singularity at the \(W \to \infty\) limit. Namely, for any finite \(W\) the coupling exists and is independent on \(W\). Nevertheless, without sidewalls \((W \to \infty)\) the coupling vanishes. This singularity is a marginal case of the known divergence of the long-range interactions in 3D (Caflisch and Luke, 1985; Ramaswamy, 2001). This kind of singularity is analogous to the dependence of polarizability tensor of dielectric bodies on their shape (Landau and Lifshitz, 1960). In our case, the slab is effectively infinite in the direction perpendicular to the walls and this asymmetry leads to the density–velocity coupling which is absent in the unconfined channel.

5.2. 1D Burgers equation

Since \(\Delta u_x\) is determined by the density in a narrow slab around the test droplet and is uniform across the slab, we apply an additional simplification by reducing the 2D ensemble into a 1D density profile \(\rho(x, t)\) along the flow (Fig. 21). Namely,
The density of the 1D density profile is described by the renowned 1D Burgers equation:

\[ \partial_t \rho + \partial_x (u \rho) = D[\rho], \]

in which \( u \) is the advective flux and \( D \) is a diffusion operator that accounts for the random motion of individual droplets (see Section 5.3 below). We discuss diffusion further below and show it can be neglected. We substitute into Eq. (58) the linear velocity–density coupling \( u(x, t) = u_d^\infty + c_0 \rho(x, t) \). We then transfer to the frame moving at \( u' \equiv (x - u_d^\infty t)/2c_0 \) and obtain that the dynamics of the 1D density profile is described by the renowned 1D Burgers equation:

\[ \frac{\partial \rho}{\partial t} + \rho \frac{\partial \rho}{\partial x'} = D[\rho]. \]

This equation was introduced in the 1930s by J.M. Burgers as a simplified version of the Navier–Stokes equation (Burgers, 1974) and has been used to study diverse non-equilibrium, nonlinear phenomena in turbulence (Bec and Khanin, 2007), cosmology (Zel’Dovich, 1970) and interface dynamics (Kardar et al., 1986). To the best of our knowledge there are only a few experimental systems that were shown to be governed by this equation: second sound in liquid helium (Goldner et al., 1993; Kitabatake and Sawada, 1978), dusty plasma (Ghosh et al., 2003) and the recently reported colloidal suspensions driven by an AC electric field (Pérez and Posner, 2010). Recently, Burgers shock waves were also observed in dense 1D microfluidic crystals and were theoretically explained from the dipolar interactions between the droplets (Champagne et al., 2011).

The solutions of the Burgers equation are shock waves with sharp front and rarefaction at the rear, as shown in Fig. 24. The velocity of a Burgers shock is obtained from mass conservation across its front and depends on the mean density at both its sides: \( u_{\text{shock}} = c_0 (\rho_1 + \rho_2) \) (Burgers, 1974). Under small perturbations \( \rho(x, t) = \rho_0 + \delta \rho(x, t) \) with \( \delta \rho \ll \rho_0 \), the Burgers equation becomes a first-order wave equation \( (\partial_t + c_0 \partial_x) \delta \rho = 0 \) with traveling wave solutions that propagate forward at a sound velocity \( c_s = c_0 \rho_0 \) in the frame moving at the mean droplet velocity \( u_d = u_d^\infty + c_0 \rho_0 \).

5.3. Burgers dynamics measurements

Once a 2D droplet ensemble was being continuously generated and filling the main channel, a pack of droplets was followed by moving the microscope stage at their mean velocity \( u_d \). Subsequently, the droplets’ trajectories were extracted using the ‘Abadiscope’ algorithm (Beatus et al., 2006). Droplet density was non-uniform on many length scales and individual droplets exhibited random motion due to the hydrodynamic interactions and dynamic clustering. Averaging over many single-droplet trajectories revealed that at short time scales \( t < 0.5 \) s droplets motion was ballistic over distances of \( \sim R \), and at longer time scales the droplets exhibited diffusion and super-diffusion: Perpendicular to the flow, droplet trajectories were diffusive \( (\Delta y^2 = D_y t, D_y \sim 300 \mu\text{m}^2\text{s}^{-1}) \) with a symmetric velocity distribution, while along the flow their trajectories were super-diffusive \( (\Delta x \propto t^\nu, \nu \approx 1.4) \) with an asymmetric velocity distribution (data not shown) (Beatus et al., 2009).

Similar diffusive dynamics were observed in the quasi-2D sedimentation of hard spheres (Rouyer et al., 2000) and in dusty plasma (Liu and Goree, 2008). Faster longitudinal diffusion was also measured for red blood cells flowing in a quasi-2D microfluidic channel (Higgins et al., 2009). We chose to model the super-diffusion operator in the Burgers equation (59)
as $D[ρ] = [D_0/4c_0^2]ρ^{2/ν+1}$ (Spohn, 1993) with $D_0 \approx 75 \, μm^2 \cdot s^{-1}$ and $ν = 1.4$ (for other formulations see (Metzler and Klafter, 2000; O’shaughnessy and Procaccia, 1985)). Nevertheless, super-diffusion can be neglected comparing to the advection term in the Burgers equation, since $D_0/[4λc_0^2λx] \approx 0.01$. The width of the shock front, namely, the characteristic length along which the density reaches maximum value at the front is $D(∂/∂t)(c_0Δρ)$, where $Δρ$ is the front amplitude (Burgers, 1974).

The 1D density profile $ρ(x, t)$ was extracted by measuring the local area fraction in discrete narrow slices perpendicular to the flow with $Δx$ of a few droplet radii. The mean velocity of droplet within a slice, $U(x, t)$ was found to increase linearly with slice density: $U(x, t) = U_0 + λc_0ρ(x, t)$, such that the velocity–density coupling constant differed from the theory by a numerical factor $λ$ between 1 and 2 (Fig. 25(a)), in which $U_0 \approx 100 \, μm \cdot s^{-1}$ and $c_0 \approx 80 \, μm \cdot s^{-1}$). Specifically, $λ \approx 1.8 ± 0.2$ for low mean densities of $ρ_0 < 0.3$, and for $ρ_0 > 0.3$, $λ$ decreases towards $λ \approx 1$ for $ρ_0 = 0.6$.

Plotting the space–time diagram of $ρ(x, t)$ revealed large-scale fluctuations, hundreds of microns long, propagating along the flow, as measured in the frame-of-reference moving at $U_0$ (Fig. 26(b)). The power spectrum of $ρ(x, t)$ showed a linear dispersion relation $ω(k) = c_λ k$ with a sound velocity of $c_λ = 10 – 100 \, μm \cdot s^{-1}$ and frequencies of less than $0.5 \, Hz$ (Fig. 25(c)). The sound velocity scaled with density, $c_λ \approx 2c_0ρ_0$ and was independent of $W$ (Fig. 25(d)). The results of the linear coupling, density waves and their sound velocity are identical to the prediction of the mean-field theory within a factor of 1 to 2.

To examine the dynamics of large density fluctuations, a short pulse of 200–300 droplets was generated followed (Fig. 26, Movie 7). The pulse self-organized into a shock wave structure with a discontinuous density jump at the front, and a rarefaction at the rear that increased with time. The front line was perpendicular to the flow and its density decreased in time. The front propagated at $u_{shock} = 145 \, μm \cdot s^{-1}$ and kept its asymmetric shape although individual droplets changed their relative position within the pack and despite their diffusive motion. This dynamics is predicted by the Burgers equation. Specifically, the shock in Fig. 26 had $ρ_L \approx 0.4–0.5$, $ρ_R \approx 0$, $U_0 \approx 100 \, μm \cdot s^{-1}$, $c_0 \approx 75 \, μm \cdot s^{-1}$, and $λ \approx 1.5$ estimated from the measured dependence of $λ(ρ_0)$. This implies a velocity–density coupling of $λc_0 \approx 112 \, μm \cdot s^{-1}$, which according to the theory implies $U_{shock} = 145–155 \, μm \cdot s^{-1}$. The shock was supersonic, as it was faster than the speed of sound in the (very dilute) medium ahead $c_λ \approx U_0$. The front width was narrow, which corroborates that diffusion was negligible and, indeed, $D_0/(c_0Δρ) \approx 1 \, μm$.

Traces of shock waves were found also in experiments with continuous droplet formation as dense, small-scale fronts $\sim 100 \, μm$ long in the $x$ direction. These fronts formed spontaneously, propagating for 1–2 s before spreading out (not shown) (Beatus et al., 2009). The shorter lifetime comparing to the full shock wave (Fig. 26) is quantitatively consistent with Burgers dynamics, in which shock lifetime decreases with the overall shock width (Fig. 24).

The 1D Burgers description captures quite accurately the collective modes of the 2D ensemble – density and shock waves – despite the mean-field and superposition approximations. Furthermore, the model ignores many degrees of freedom that
**Fig. 26.** Burgers shock waves. (a) Shock wave evolution in time shown at 6 s intervals in the frame of the channel (solid lines). Scale bars are 100 µm. The pulse was generated by keeping the oil pressure constant and changing the water pressure $P_w$ as follows: (1) To empty the main channel from droplets $P_w$ was decreased below the critical pressure $P_c$, under which oil starts to back-flow into the water channel. (2) $P_w$ was increased above $P_c$, which forced the water towards the T junction until droplet formation had started. (3) To stop droplet formation, $P_w$ was decreased below $P_c$. (b) The corresponding density profile $\rho(x, t)$ for each snapshot (bars). The black line shows the solution of the Burgers equation for the experimental initial condition with $u_0 = 100 \mu m s^{-1}$ and velocity–density coupling of $2c_0 = 150 \mu m s^{-1}$.

**Fig. 27.** Non-uniformities along $y$ perpendicular to the flow for $\rho_0 = 0.5$ and $W = 500 \mu m$. (Top) Density profile along $y$ shown by the probability density to find a droplet at a given $y$. The peaks are $R$ and $3R$ away from the sidewalls. (Bottom) Mean longitudinal-velocity profiles along $y$.

arise from the 2D, random and discrete nature of the system, such as transversal droplet velocity (Fig. 21). These “hidden variables” are reflected, for example, in the measured variance of the linear velocity–density coupling (Fig. 25(a)) as well as in direct measurements of the density and longitudinal-velocity profiles along the $y$ direction (Fig. 27). Both profiles are non-uniform: the probability density of $\rho(y)$ has high peaks next to the sidewalls and the velocity profile $u_x(y)$ is above average around $y = \pm W/4$ and below average elsewhere. Near the sidewalls, at $y = \pm W/2$, $u_x(y)$ is slowest, probably because in these denser regions the droplets assume a peloton-like configuration, thus down slow each other. Nevertheless, $u_x(y)$ is small comparing with $u_d$: its largest difference gives $\delta u/u_d \sim 0.05 - 0.1$ and across the central part of the channel we have $\delta u/u_d \leq 0.02$. The small variations along $y$ do not waive the effective 1D description by the Burgers equation.

### 6. Summary and outlook

This paper summarizes recent experimental and theoretical work on the physics of microfluidic droplet ensembles in quasi-2D channels, at low Reynolds number of $10^{-4} - 10^{-3}$. We studied the emergence of many-body, collective phenomena from single-particle dynamics in a system far from equilibrium, focusing on a 1D crystal and a 2D disordered ensemble of droplets. The observed collective modes – phonons, nonlinear crystal instabilities, sound waves and shock waves – are understood within a unified theoretical framework based on hydrodynamic interactions of single-particle dipolar fields. The simplicity of the experimental system with accessible time and length scales establishes as a convenient test-bed for further
investigation of non-equilibrium physics. Here, we discuss several open questions, which stand out for further research in our opinion.

The propagation of 1D crystalline modes in a 2D channel can be taken to yet unexplored scenarios. One could study the 1D crystal modes beyond the linear description of small amplitude fluctuations, namely, understanding its nonlinear dynamics. This may include studying interactions between eigenmodes, for example by temporal excitation of specific modes. A nonlinear amplitude-equation formalism may be developed for a complete description of mode propagation. There are interesting points in $k$ space that call for further investigation: (i) the region of $\partial \omega / \partial k = 0$ around the middle of Brillouin zone, where higher order collective excitations (quasi-particles) may arise (Cross and Hohenberg, 1993); (ii) the peculiar $\omega = 0$ zigzag instability and surface melting, both at $k = \pi / a$. Next, a regime of Anderson localization could be realized by embedding random defects in the channel as quenched disorder that impedes propagating waves. Finally, by reducing the lateral channel dimensions, in the limit of exponentially screened hydrodynamic interactions, one could address models of non-equilibrium particle exclusion with short-range interactions (Levine et al., 2005).

Much remains to be explored in 2D ensemble motion. First, the simplified effective 1D droplet density profile is an opportunity to revisit many untested features and predictions associated with Burgers dynamics. For example, one could experimentally study 1D Burgers turbulence (Frisch and Bec, 2000) and shock wave interactions by controlling their formation at given intervals and magnitude.

Second, the mean field description, though successful in elucidating the origin the shocks and density waves, cannot account for cluster dynamics and fluctuations in 2D. It would be interesting to measure the statistical properties of 2D clustering dynamics, which appears to be in steady-state, namely, cluster size distributions, lifetime, and transition rates. From the hydrodynamics standpoint clustering poses a theoretical challenge since a far-field approximation does not hold and hence multiple reflections are required. At a larger scale, the 2D ensemble may exhibit large scale collective swirls and vorticity, as in 3D particle sedimentation. To investigate such collective fluctuations, the lateral channel dimensions could be extended to the centimeter scale to include $10^2$–$10^3$ droplets across the channel (our current channel width accommodated roughly 20 droplets). Presumably, the system would then exhibit large-scale 2D turbulent flow, and it would be particularly interesting to study the cascade of energy transfer between modes at different length scales. In contrast to 3D where such large-scale swirls are difficult to explain theoretically, the 2D problem is likely to be tractable leading to new modes for these ensembles. Light scattering may be used to examine fluctuations of large ensembles in case particle tracking is limited to small ensembles.

The 2D droplet ensemble could be used as an experimental platform for studying the micro-rheology of complex fluids, namely the relation between their microscopic structure and macroscopic rheological properties such as viscosity. This active field studies, for example, the phenomena of shear thinning and shear thickening in non-Newtonian fluids (Cheng et al., 2011; Larson, 2001; Vermant and Solomon, 2005; Wagner and Brady, 2009). In the microfluidic setup, the effective water-in-oil emulsion viscosity could be measured by relating its mean velocity, including both oil and droplets velocities, with the applied pressure gradient. It would be interesting to study the effective viscosity as a function of the droplets density and micro-structure such as clusters, sound waves, shock waves etc.

Finally, a long standing controversial question relates to the existence of a variational principle in non-equilibrium statistical physics. To this end, we propose to expand the experimental methodology beyond single-particle tracking to include a measurement of the dissipation rate as well as a means to apply spatial and temporal perturbation (Niven, 2009). Then, perturbing a steady-state motion of a droplet ensemble, observing the collective response while monitoring the overall dissipation rate, may provide clues on the existence of optimization rules underlying the collective modes. The physics of 2D droplet ensembles is awaiting further discoveries.

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Appendix A. Materials and methods

A.1. Fabrication and materials

The microfluidic devices were fabricated using standard soft lithography (Duffy et al., 1998; Squires and Quake, 2005). Reusable casting molds were made of SU-8-2007/2010 negative photoresist (Microchem, Newton MA) using optical photoplotter masks (Suron, Israel). Channels were prepared by casting PDMS elastomer (poly-dimethylsiloxane, Sylgard-184, Dow Corning) on the molds and curing at 80 °C for 1 h. After separating the cured PDMS from the mold and punching holes at the inlets and outlet, the PDMS was reversibly attached to a clean PDMS-coated glass slide that constituted the channel floor. The slide was prepared by spin-coating PDMS at 3000rpm for 30 s following by the same curing procedure. Irreversible sealing was done by oxidizing both surfaces in a plasma chamber (March Plasmod) for 50 s at 150 W under
1–2 mtorr pressure of oxygen and argon in a 1:1 ratio. Subsequently, we attached the channels onto the slide and cured overnight at 80 °C.

We used light mineral oil (Sigma, M5904, viscosity $\eta_o = 30$ mPas, density $\rho_o = 0.84$ g/ml) with 2% (w/w) span-80 surfactant (Sigma) and deionized double-distilled water (Millipore, 18 MΩ). The surface tension between water and the oil–surfactant solution was 3.5 mN/m, as determined by the pendant droplet method (Ramè-Hart Inc. RH2001 goniometer). Mineral oil is one of the few oils fully compatible with PDMS (Lee et al., 2003). Other compatible oils, such as fluorocarbon oils, are typically less viscous, which would result in smaller drag coefficient of the droplets and, hence, weaker hydrodynamic interaction and weaker collective effects. Fluids were kept in 1 ml plastic syringes and connected to the device by tygon tubes and blunt-end needles. The flow was driven by nitrogen pressure typically between 4 and 25 psig, such that water and oil pressures were controlled by precise pressure regulators (T100, Control Air, Amherst NH). The flow was visualized by adding fluorescent polystyrene micro-beads (diameter 0.4 µm) either to the water (Fig. 2) or to the oil (Fig. 3).

A.2. Design considerations

To design a functional and stable droplet generator we used a set of heuristic constraints on the channels’ geometry. We estimate the water and oil fluxes in the device given its geometry and the applied pressures by approximating the channels network to an electrical DC circuit, in which electric resistance is analogous to hydrodynamic resistance to flow (Cristobal et al., 2006a; Jousse et al., 2005; Schindler and Ajdari, 2008). The resistance of a channel of length $L$ and rectangular cross section $h \times W$ and $h \ll w$ is well approximated by $R \approx \left(12\eta h^3 W / \rho 1 + 0.63h / W \right)$ (Happel and Brenner, 1965), and the pressure difference $\Delta P$ is related to the flux $Q$ by Ohm’s law $\Delta P = QR$. In addition, we assume for simplicity that water and oil mix at the T junction and that the resistance of the main channel is determined by oil viscosity alone, rather than by a complicated emulsion viscosity.

According to our observations, the width of the channels at the T junction is the most important parameter that determines droplet size. Hence, we determine the junction width to be of the order of the chosen droplet diameter. In our devices, the T junction width was smaller than the main channel width $W$, such that immediately after its formation, the droplet blocked the T junction (plug flow), which significantly increased the T junction resistance and prevented the formation of the next droplet. Once a droplet left the T junction and entered the main channel it became disc-like, decreasing the junction resistance and enabling the formation of the next droplet. Since there was typically no more than one droplet in the junction, the length of the channel between the junction and the main channel might limit the frequency of droplet formation and needs to be considered in design.

Given the chosen working pressures and channel geometry, we calculated the fluids fluxes and velocities, and iteratively changed the geometrical parameters to suit the flow requirements. Importantly, to improve the stability of the device with respect to pressure and resistance variations, we set the water and oil inlet resistances, $R_{water}$ and $R_{oil}$ to $R_{water} \sim R_{oil} \geq 2R_{main}$, where $R_{main}$ is the main channel resistance. The improved stability stems from the larger pressure range at which there is no back-flow of oil into the water inlet and vice versa: $(1 + R_{oil}/R_{main})^{-1} < \rho_{water}/\rho_{oil} < (1 + R_{water}/R_{main})$. Note that resistance variations are inherent to the experiment, since $R_{main}$ depends on the number of droplets in the main channel as well as on their configuration.

A.3. Optical microscopy of droplet ensembles

The device was mounted on an inverted microscope (Olympus, IX-71) with a motorized stage (UMR5.25 and CMA-25CC, Newport Corp.). After oil and water flows were stabilized to give the sought droplet size, frequency and flow velocity, the droplets were recorded using a digital camera (PCO, SensiCam-QE, maximal resolution 1376 × 1044). To sample many droplets, we used a 10x lens with a 0.5x adapter, which gave a frame width of 1.75 mm. Typical frame rate was 20–35 Hz. In the ‘peloton effect’ measurements (Section 4.1) the device was static with respect to the microscope. In all other experiments, where power-spectra of vibrations or density-fluctuations were measured, the sampling time had to be increased to get a sufficient resolution in $\omega$, which was done by tracking a pack of droplets in motion, by moving the device at velocity $u_d$ with respect to the microscope. To measure oil velocity, once the set of measurements was completed for a given device, we changed the oil to the oil–beads mixture and recorded the flow in fluorescence mode under the same pressure conditions.

A.4. Data analysis

The sequence of images acquired in the experiment was analyzed using a precise tracking algorithm (the Moses–Abadi algorithm, the ‘Abadiscope’), implemented in Matlab. The algorithm locates the droplets in each image, finds the positions of their centers and tracks the trajectory of each droplet in time. To extract the spectra of crystal vibrations it is crucial to accurately determine the positions of the droplets’ centers. The algorithm works, therefore, in two stages. First, the location of the droplet is determined to low accuracy on the basis of the contrast between the droplet boundary and the background. Next, the center is found with higher accuracy by calculating an optimal fit between the image of the droplet and a ring with a Gaussian intensity profile along its radius. Using this method, it is possible to locate a droplet’s center to sub-pixel accuracy, estimated as tens of nanometers.
Appendix B. Numerical simulation

Our simulation is based on numerical solution of the nonlinear equations of motion of all droplets in an ensemble of \( N \) droplets. As in the derivation of these equations, the main assumption of the simulation is that the ensemble flow potential is given by superposition of single-droplet potentials. The motion of each droplet is described by two ordinary differential equations (ODE) for its \( x \) and \( y \) coordinates that together form a set of \( 2N \) coupled nonlinear ODEs, in which each droplet interacts with all the others. The equations were solved using Matlab’s ODE solver. This numerical scheme considers only the far-field dipolar interaction between the droplets and is, hence, much simpler and less accurate than other numerical methods for calculating the dynamics of colloidal suspensions, such as Stokesian dynamics (Baron et al., 2008; Sierou and Brady, 2001; Swan and Brady, 2007), dissipative particle dynamics (Hoogerbrugge and Koelman, 1992) and fluid particle dynamics (Tanaka and Araki, 2000). However, it successfully reproduces the experimental dynamics when the droplets do not touch each other, as does our theory. In cases where droplets can touch each other, such as in 2D disordered ensembles, the simulation reproduces many, but not all, of the experimental features (see Appendix B.3 below).

B.1. Boundary conditions

In the direction of flow we set either periodic boundary condition or experimental-like conditions, where we defined a channel of finite length with droplets injected from one side and disappearing on the other. In the \( y \) direction we imposed either open or confined geometry, for which we respectively used the unconfined single-droplet potential (8) or the confined one (35). When solving for confined droplets we had to prevent the droplets from penetrating the sidewalls, since the confined potential is invalid for \( |y| > \frac{1}{2}W - R \) and does not include any restoring force off the sidewalls. For this purpose we defined a sidewalls restoring force acting on the \( n \)’th droplet:

\[
F_{\text{wall},n}^{\pm} = f_0 \left[ 1 - \tanh \left( \frac{\frac{1}{2}W - R \pm y_n}{l_0} \right) \right] \hat{y},
\]

where \( \pm \) indicates the repulsion from the sidewalls at \( \pm \frac{1}{2}W \). The force scale is \( f_0 = \xi_d \cdot (100 \, \mu \text{m s}^{-1}) \), equivalent to the drag force of an incident flow with velocity 100 \( \mu \text{m s}^{-1} \) relatively to the droplet. The restoring force smoothly turns across a scale of \( l_0 = 0.1 \, \mu \text{m} \). In addition, in simulations where droplets might touch each other, such as for dense 2D ensembles or crystal instabilities, we had to impose a hard-disc repulsion between droplet to prevent interpenetration. The single-droplet potential is invalid inside the droplet and does not induce a repulsive force. The repulsion we imposed between the \( n \)’th and \( j \)’th droplets was similar to the sidewalls repulsion:

\[
F_{\text{HD},n,j} = f_0 \left[ 1 - \tanh \left( \frac{|r_n - r_j| - 2R}{l_0} \right) \right] \cdot \hat{r}_{nj},
\]

where \( \hat{r}_{nj} \) is a unit vector connecting the centers of the two droplets.

B.2. Initial conditions and noise

To excite the collective modes of the 1D crystal it is required to introduce some perturbation, either through an initial condition or external noise. We used two types of initial conditions: (a) assigning a random amplitude along \( x \) and \( y \) for each droplet with respect to its crystal position, and (b) defining the initial configuration in \( k \)-space (usually using only one or two wavenumbers) or in real-space (Fig. 17(b), for example). The sources of noise we used were (a) formation noise—random offsets given to the position of each droplet as it appears at the formation area, and (b) embedded defects, represented as dipolar fields with fixed positions at the channel frame-of-reference. Such defects are equivalent to stationary pillars in the channel. When simulating 2D ensembles we usually randomly assigned the droplets initial positions across the whole channel or parts of it. In some cases we set the initial positions according to an experimental configuration. In all cases, initial positions were selected such that the droplets do not interpenetrate.

B.3. Results

The simulation quantitatively reproduced the experimental dynamics: The phonons, crystal instabilities (Figs. 17, 18 and 20) as well as the sound waves in the 2D ensemble (Fig. 28). The recapitulation of the 2D ensemble dynamics is interesting, since the superposition approximation is expected to fail when droplets touch each other. Nevertheless, we note that the values of \( c_s \) and the velocity–density coupling are a bit lower than in the experiment, although rather close to the theory with \( \lambda = 1 \). The simulation failed to reproduce the exact dynamics of Burgers shock waves—while in the experiment the front line remained perpendicular to the flow, in the simulation droplets at the front converged towards the middle of the channel (not shown). However, the dynamics of the 1D profile \( \rho(x) \) exhibited the expected Burgers dynamics.
Fig. 28. Simulation of a 2D ensemble of 955 droplets, $\rho_0 = 0.30$, $R = 10 \, \mu m$, $W = 500 \, \mu m$, $u_c = 100 \, \mu m \, s^{-1}$ and $u_{oil} = 400 \, \mu m \, s^{-1}$. The channel length was 2000 $\mu m$ with periodic boundary conditions along $x$. Simulation time was 60 s. (a) The mean longitudinal velocity of droplets across vertical slices of length 4$R$ plotted vs. slice density (blue dots). Errorbars show the standard deviation around the mean. Solid line is a linear fit with slope 50 $\mu m \, s^{-1}$.

(b) Density (top) and mean longitudinal velocity (bottom) profiles along $y$. The top plot shows the probability density to find a droplet at a given $y$. (c) Space–time diagram of $\rho(x, t)$. Color encodes for local density. (d) The power spectrum of $\rho(x, t)$ shown in (c) in units of $(\mu m \, s)^{-2}$. Power is concentrated along a line with linear dispersion $\omega = c_s k$ and sound velocity $c_s = 30 \, \mu m \, s^{-1}$. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Appendix C. Supplementary data

Supplementary material related to this article can be found online at http://dx.doi.org/10.1016/j.physrep.2012.02.003.

References

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