Go with the flow

A 2006 *Nature Physics* paper reported phonons in a one-dimensional crystal of aqueous droplets traversing a laminar oil flow — putting microfluidics on the map as a tool for unravelling the mechanisms behind regularity in thermodynamically open systems.

Piotr Garstecki and Robert Hołyst

The original motivation for understanding the behaviour of fluids confined to microscale (and smaller) circuits was pragmatic: control over minuscule flows would ease the handling of liquids for the study of chemistry, biology and materials science. Aside from this ‘lab on a chip’ application, the sheer appeal of regular flow and perfectly spaced droplets in meandering microchannels was undoubtedly a driving factor in the development of microfluidics. Still, many physicists were probably unimpressed. Trivial flow at the microscale can generally be interpreted within the linear Stokes flow regime — a fixture in textbooks for many decades now. Yet, it took physicists to not just admire the seemingly obvious regularity, but to marvel at and question the mechanisms behind it.

Writing in *Nature Physics* in 2006, Tsevi Beatus and colleagues described a simple experiment in which a microfluidic device was used to generate a train of evenly spaced water droplets. The droplets were confined to a narrow one-dimensional channel filled with oil, which widened into a quasi-two-dimensional channel, restricting the droplets in one direction only and allowing them to move in the plane of the wide microfluidic channel (Fig. 1). Despite the fact that the droplets moved much slower than the surrounding oil, they remained as ordered as an evenly spaced one-dimensional crystal. Although at first glance it might seem natural that the droplets remained ordered, studies of other similar systems suggest otherwise. For example, whereas two droplets falling simultaneously in a viscous liquid do not interact due to symmetry, introducing a third droplet suffices to curve the trajectories of the drops. How, then, could a train of tens of droplets remain undisturbed?

Beatus et al. noticed small oscillations of the droplets — undulations that could easily be mistaken for simple irregular thermal fluctuations. Two questions then arose. Why were they able to observe a one-dimensional crystal despite the fact that entropy should have dispersed the droplets randomly? And what was the origin of the fluctuations?

A spectral analysis of the collective vibrations revealed unusual dispersion relations markedly different from those observed in harmonic crystals. Thermal fluctuations in an equilibrium one-dimensional crystal should grow with system size and destroy the order. On the other hand, low-Reynolds-number flow dominated by viscous dissipation should damp the oscillations. Beatus et al. showed that the fluctuations — similar to acoustic phonons — were an outcome of the symmetry-breaking flow field. The resulting long-range inter-droplet hydrodynamic interactions emerged as a result of the difference between the fluid flow and the droplet flow in the confined space.

Perhaps the paper’s most inspiring contribution was an example of a tool to study out-of-equilibrium systems. Model systems have always played an important role in physics, although often their role is not immediately recognized. For example, the hard-sphere fluid is the paradigm of the phenomenon of crystallization. Despite the lack of attractive van der Waals interactions, a hard-sphere liquid does crystallize, the process being driven solely by the increase of entropy. George Uhlenbeck, who led a debate in 1957 in the presence of two Nobel Prize winners, proposed a vote, for or against, to determine the existence of crystallization of hard spheres. The vote came out even. We now know that heat release upon crystallization does not preclude a simultaneous increase of entropy, and the hard-sphere model system has greatly contributed to our understanding of many phenomena.

The experiment performed by Beatus et al. has already led to results that may have had a similar reception just ten years ago. One example is the observation of complex two-dimensional flowing-droplet crystals obtained through a rational choice of the shape of the particles, which tunes the hydrodynamic interactions in low-Reynolds-number flow. A similar observation was obtained with spherical droplets in the inertial regime.

Long-range hydrodynamic interactions affect droplets in microfluidics, flocks of bacteria, and the late stages of phase separation. Recent studies have shown that hydrodynamic interactions are responsible for ‘phase transitions’ in flowing suspensions of red blood cells resulting in a reduction of the self-diffusion of hemoglobin in red blood cells. Hydrodynamic coupling in the presence of thermal noise has also been found to contribute to the collective motion of bacterial flagella. The excluded-volume effects and hydrodynamic interactions are sufficient to explain the large reduction in diffusion of macromolecules observed *in vivo*, and may even be responsible for the emergence of the length-scale dependent viscosity in complex fluids, including the living cells.

Physicists need model systems to uncover and attack new problems, even if the solutions are not immediately obvious. A perfect example is the adventure of Enrico Fermi, John Pasta and Stanislaw Ulam, who set out to understand the equipartition of energy in a system of coupled weakly anharmonic oscillators, and found quasiperiodic behaviour that challenged their expectations. Today, their quest has left us with the machinery required to identify the laws of energy-relaxation in non-equilibrium systems — even if the problem itself remains largely unsolved.

Irreversible pathways connecting equilibrium states are not well understood.
news & views

Even less is known about general rules that govern systems far from equilibrium. Beatus et al.\textsuperscript{2} introduced a convenient new system to study off-equilibrium organization. The full implications of controlled hydrodynamic interactions in non-equilibrium systems developed by the authors still remain to be seen. Perhaps this is the beginning of a generation of tools\textsuperscript{11,12} that can equip physicists as they question the mechanisms behind the emergence of order in open dissipative systems — which may not be so obvious after all.

Piotr Garstecki and Robert Hołyst are at the Institute of Physical Chemistry of the Polish Academy of Sciences, Kasprzaka 44/52, 01-224 Warsaw, Poland.

e-mail: garst@ichf.edu.pl; rholyst@ichf.edu.pl

References