Supplementary Materials for

Instability mediated self-templating of drop crystals

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The PDF file includes:

Legends for movies S1 to S4
Supplementary Note
Figs. S1 to S7

Other Supplementary Material for this manuscript includes the following:

Movies S1 to S4
S1. SUPPLEMENTARY VIDEO LEGEND

Video 1: **Printing a drop crystal.** Video of the printing experiment with $U_0 = 4.7$ mm/s, $Q = 0.21$ mL/min and $L = 3.0$ mm (top view; video sped up 10 times for the first 3 rows, and then 100 times). The rows are printed in alternating directions. The final snapshot is shown in Fig.1b of the main text.

Video 2: **Wavelength locking.** Breakup of liquid jets next to an acrylic template (view under microscope). The breakup wavelength follows that of the template.

Video 3: **Shift in drop position induced by nozzle motion.** After the drops are formed, they are dragged by the moving nozzle when printing subsequent rows. The change in drop position becomes negligible when the moving nozzle is far away. The video of the unidirectional print is accelerated 5 times and the video of the serpentine print is accelerated 10 times.

Video 4: **Crystal with two populations of monodisperse drops.** Two-step printing process of a crystal with two populations of monodisperse drops. The orange drops are printed next to a solid template first, followed by blue drops printed atop the orange drops.
S2. SUPPLEMENTARY NOTE

S2.1. Convergence to an ordered lattice model.

We develop a model to describe the convergence of wavelengths in the drop crystal reported in the main text (Fig. 4a). This empirical model relies on the observations reported in Fig. 3 and aims at predicting the configuration of a newly printed line \(i\) based on the configuration of the latest printed line \(i-1\). The model is based on the fact that each drop in the row \(i\) typically emerges in-between a pair of drops in \(i-1\), although there are two exceptions to this rule as detailed next.

If the spacing between two consecutive drops in \(i-1\) is \(\lambda < 0.7\lambda^*\), then no drop forms in the row \(i\). For a spacing close to \(\lambda^*\), i.e., \(0.7\lambda^* \leq \lambda < 1.4\lambda^*\), a single drop is produced in \(i\) as mentioned earlier. Conversely, two drops are produced if \(1.4\lambda^* \leq \lambda \leq 2.0\lambda^*\).

Experimentally, we observed that the wavelengths in the breakup pattern of an unbounded jet \((i = 1)\) are typically less than \(2.0\lambda^*\). Therefore, based on the proposed rules, the second row of drops \((i = 2)\) will always have wavelengths in the range \(0.7\lambda^* \leq \lambda < 1.4\lambda^*\).

For simplicity, we assume that a drop formed in \(i\) is equidistant from mother drops in \(i-1\) that lead to its formation (see Supplementary Fig. S3). Therefore, the distance between drops in \(i\) can be obtained as the average value of the two distances between three consecutive mother drops in \(i-1\). Assuming that the list of distances between drops \(i\) is:

\[
\left\{ \ldots, \lambda_{n-3}, \lambda_{n-2}, \lambda_{n-1}, \lambda_n, \lambda_{n+1}, \lambda_{n+2}, \lambda_{n+3}, \ldots \right\} \quad (S1)
\]

then, in line \(i+1\) the distances between drops would be:

\[
\left\{ \ldots, \frac{\lambda_{n-3}}{2} + \frac{\lambda_{n-2}}{2}, \frac{\lambda_{n-2}}{2} + \frac{\lambda_{n-1}}{2}, \frac{\lambda_{n-1}}{2} + \frac{\lambda_n}{2}, \frac{\lambda_n}{2} + \frac{\lambda_{n+1}}{2}, \frac{\lambda_{n+1}}{2} + \frac{\lambda_{n+2}}{2}, \frac{\lambda_{n+2}}{2} + \frac{\lambda_{n+3}}{2}, \ldots \right\} \quad (S2)
\]

This set of values matches the result of the convolution of the kernel \(\left\{\frac{1}{2}, \frac{1}{2}\right\}\) with the list in
Eq. S1, i.e., the pairwise moving average of the list (obtained by computing the means of elements in list taken in blocks of length 2). This process repeats in the subsequent rows. As such, when starting from a noisy distribution of distances whose average is \( \lambda^* \), the system will necessarily converge to distances whose value are virtually uniform and equal to \( \lambda^* \) and thus yield a uniform crystal pattern.

### S2.2. Self-correction of local defects.

We demonstrate the self-correcting ability of drop crystals through three scenarios with different types of intentionally seeded defects. In the first two scenarios, we increased the size of a single drop (Fig. S4a) or removed a drop completely (Fig. S4b), without changing the wavelength of the pattern. In the last scenario, we removed a drop and shifted its neighbor so as to convert three natural wavelengths into two (Fig. S4c). The left panels in Fig. S4 show the initial configurations after the artificial defects were seeded. The final snapshots after more rows were printed are shown in the right panels in the figure. The dashed lines outline the region affected by the initial defect based on our kernel model. Unlike the schematic picture in Fig. S3, the actual affected region is not an upright triangle if neighboring rows in the crystal lattice do not have an out-of-phase relation. The triangular region will be tilted depending on the actual phase relationship between the rows.

### S2.3. Drop crystal lattice pattern.

When we print successive rows one after the other, the drops produced in prior rows serve as template and seed perturbations in the subsequent rows. The overall crystal pattern evolves as new rows are added until a final state is reached. In Supplementary Fig. S5, we illustrate this evolution
by plotting the angle $\theta$ as a measure of relative drop position between neighboring rows. We note that the ultimate lattice structure is the result of two contributions: first, the initial phase difference $\Delta$ between consecutive rows and second, the shift $s$ a drop experiences after being printed.

In Supplementary Fig. S6, we show that the initial phase difference $\Delta$ increases with the jet length $\ell$. The ratio $\Delta/\lambda_f$ appears to be an increasing function of $\ell$ which plateaus at a value close to 0.5. This limiting value corresponds to the case where a drop is formed equidistant to the two adjacent drops in the prior row. Note that this long jet limit is similar to the out-of-phase mode observed in the simultaneous breakup of multiple liquid threads.

In terms of the shift in drop position $s$ caused by nozzle movement, we find that the prior rows are affected to different extents. In Supplementary Fig. S7a, we show that the shift $s$ of a drop decays with its distance from the new row. When the moving nozzle is far enough, there is negligible shift in drop position. Experimentally, we also find that data with different inter-row spacing $L$ collapse onto a single curve. In Supplementary Fig. S7b, we further show that $s$ increases with the nozzle speed $U_0$. Hence, the printing parameters $U_0, Q$ and $L$ can be adjusted to change $\Delta$ and $s$. This will in turn lead to the change in relative drop position and the unit cell of the ultimate lattice, enabling the fabrication of different crystal patterns.
FIG. S1. **Size of drops between main drops.** (a) Photographs of jet breakup with initial radius $h_0 = 0.63$ mm and templates with different forcing wavelength $\lambda_f (L = 5h_0)$. (b) Radius of the biggest drops between main drops, $R^\dagger$, relative to radius of main drops, $R$, plotted as a function of the frustration ratio, $\lambda_f/\lambda^*$. 
FIG. S2. **Printing a uniform drop lattice using a template.** A sequence of photographs showing a regular lattice of drops formed by printing multiple lines parallel to a template. In each row, the spacing between drops follows the wavelength of the template $\lambda_f = 7.0$ mm. Printing parameters: $U_0 = 46.8$ mm/s, $Q = 3.50$ mL/min, $L = 4.0$ mm.
FIG. S3. **Kernel model for the defect correcting process.** A cartoon showing a single wavelength defect with 1.6 times of the natural value in the first row. The wavelengths in row $i$ are determined by the pairwise averages of the neighboring wavelengths in row $i - 1$.

FIG. S4. **Self-correction of defects.** Correction of defects in drop crystals in three example scenarios: (a) correct wavelengths with a bigger than average drop, (b) correct wavelengths with one drop missing, (c) two consecutive incorrect wavelengths (each being equal to 1.5 times of the natural value). The dashed lines outline the region affected by the initial defects. In these experiments the printing direction is from right to left in all rows.
FIG. S5. **Evolution towards the lattice pattern.** (a) In this unidirectionally printed drop lattice (i.e., nozzle moves in the same direction in each row), the position of drops relative to those in the previous row (measured in terms of the angle $\theta$) changes as more rows are printed. (b) Changes in $\theta$ across the rows. The ultimate lattice pattern is settled when the drops are far from the new row and thus unaffected by the nozzle movement.
FIG. S6. **Initial phase difference.** The initial phase difference $\Delta$ between the newly printed row of drops and its neighboring row (or solid template) plotted as a function of the observed jet length $\ell$. The colored symbols (resp., empty circles) represent data from printing a single row next to another row of drops (resp., an acrylic template with $L = 3.4$ mm).

FIG. S7. **Drops dragged by moving nozzle.** (a) The shift in the lateral drop position $s$ versus the distance between the drop and the newly printed row, $d$. The nozzle speed is constant $U_0 = 7.2$ mm/s. (b) The shift $s$ depends on $U_0$. The spacing between the rows is kept constant $L = 2.35$ mm.