Electronic Supplementary Material (ESI) for Soft Matter.

Electronic Supplementary Information (ESI) – Commensurability-driven structural defects in double emulsions produced with two-step microfluidic techniques

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Here we provide the caption of Movie S1 and the derivation of four of the generic features of sequences of drops and patterns discussed in the manuscript.

Caption of Movie S1

The movie shows a typical experiment in which an oil-in-water-in-oil double emulsion is formed. The resulting sequence of drops $[N_n]$ is characterized by a succession of two and three inner droplets; in this sequence, N^-=3 and N^+=2. The controlling parameter is T/τ=2.27.

Derivation of properties (A$_3$), (A$_7$), (A$_8$), and (A$_9$)

(A$_7$) *Variations in the number of drops per pattern does not exceed one unity* since P_i can take only two values: $P^{min}=\text{floor}\left(\frac{1}{|\varepsilon|}\right)-1$ and $P^{max}=\text{ceil}\left(\frac{1}{|\varepsilon|}\right)-1$.

As shown in the text, considering the case $N_1=N^-$ one can show that k_i, the index of the $(i+1)$-th N^- found in a sequence of drops $[N_n]$ $(n\in\mathbb{N}^\star)$ is $k_i=\text{ceil}\left(\frac{i}{\varepsilon}\right)+1$ in the case $\varepsilon>0$. When $\varepsilon<0$, a similar approach gives $k_i=\text{ceil}\left(\frac{i}{-\varepsilon}\right)+1$. Hence, $k_i=\text{ceil}\left(\frac{i}{|\varepsilon|}\right)+1$ for any ε. Using this expression and the definition of k_i, P_i reads:

$$P_i = k_{i+1} - k_i - 1 = \text{ceil}\left(\frac{i+1}{|\varepsilon|}\right) - \text{ceil}\left(\frac{i}{|\varepsilon|}\right) - 1. \quad (1)$$

Using the relation $\text{ceil}(x)+\text{ceil}(y)-1\leq\text{ceil}(x+y)\leq \text{ceil}(x)+\text{ceil}(y)$ where x and y are real numbers, one finds:

$$\text{ceil}\left(\frac{i}{|\varepsilon|}\right) + \text{ceil}\left(\frac{1}{|\varepsilon|}\right) - 1 \leq \text{ceil}\left(\frac{i+1}{|\varepsilon|}\right) \leq \text{ceil}\left(\frac{i}{|\varepsilon|}\right) + \text{ceil}\left(\frac{1}{|\varepsilon|}\right). \quad (2)$$

When $\frac{1}{|\varepsilon|}$ is not a natural number, the relation $\text{floor}\left(\frac{1}{|\varepsilon|}\right)=\text{ceil}\left(\frac{1}{|\varepsilon|}\right)-1$ used with Eq. (2) gives $\text{ceil}\left(\frac{i}{|\varepsilon|}\right)+\text{floor}\left(\frac{1}{|\varepsilon|}\right)\leq\text{ceil}\left(\frac{i+1}{|\varepsilon|}\right)\leq\text{ceil}\left(\frac{i}{|\varepsilon|}\right)+\text{ceil}\left(\frac{1}{|\varepsilon|}\right)$. Subtracting $\text{ceil}\left(\frac{i}{|\varepsilon|}\right)$ from each side of this inequality and using Eq. (1), one obtains:

$$\text{floor}\left(\frac{1}{|\varepsilon|}\right) \leq P_i + 1 \leq \text{ceil}\left(\frac{1}{|\varepsilon|}\right). \quad (3)$$

Hence, P_i can be either $P^{min}=\text{floor}\left(\frac{1}{|\varepsilon|}\right)-1$ or $P^{max}=\text{ceil}\left(\frac{1}{|\varepsilon|}\right)-1$.

(A$_8$) $\overline{P}=\frac{1}{|\varepsilon|}-1$ *is the mean number of drops per pattern.*

Using Eq. (1), one can write $\sum_{i=1}^{p} P_i=k_{p+1}-k_1-p=\text{ceil}\left(\frac{p+1}{|\varepsilon|}\right)$-$\text{ceil}\left(\frac{1}{|\varepsilon|}\right)$-$p$. Since $\overline{P}=\lim_{p\to\infty}\frac{1}{p}\sum_{i=1}^{p} P_i$, one obtains $\overline{P}=\lim_{p\to\infty}\frac{1}{p}\left[\text{ceil}\left(\frac{p+1}{|\varepsilon|}\right)-\text{ceil}\left(\frac{1}{|\varepsilon|}\right)-p\right]$. Consequently, as $\lim_{x\to\infty}\frac{\text{ceil}(x)}{x}=1$, one finds $\overline{P}=\frac{1}{|\varepsilon|}$-1.

(A$_3$) and (A$_9$) $\left|\frac{T}{\tau}-\text{round}\left(\frac{T}{\tau}\right)\right|=\mathcal{F}(N^-)$ *and* $\left|\frac{1}{|\varepsilon|}-\text{round}\left(\frac{1}{|\varepsilon|}\right)\right|=\mathcal{F}(P^-)$ *are the fractions of defects and pattern defects, respectively.*

As shown below, demonstrating properties (A$_3$) and (A$_9$) is trivial once we have established that the series $[N_n]$ and $[P_n]$ are made of two consecutive natural numbers. We consider a sequence $[S_n]$ made of such numbers denoted S^- and S^+, that is, the number appearing the less (resp. more) often in $[S_n]$; a direct consequence of the nature of $[S_n]$ is S^+=round($\overline{S}$). The mean value $\overline{S}$ of the sequence reads $\overline{S}=S^+ + \mathcal{F}(S^-)(S^- - S^+)$ where $0\leq\mathcal{F}(S^-)\leq\frac{1}{2}$ is the fraction of S^- found in $[S_n]$. Two situations are possible:

∘ S^+=floor($\overline{S}$) and S^-=ceil($\overline{S}$), so that $\mathcal{F}(S^-)=\overline{S}-S^+$ and $0 \leq \overline{S} - \text{round}(\overline{S}) \leq \frac{1}{2}$.

∘ S^+=ceil($\overline{S}$) and S^-=floor($\overline{S}$), so that $\mathcal{F}(S^-)=S^+-\overline{S}$ and $0 \leq \text{round}(\overline{S}) - \overline{S} \leq \frac{1}{2}$.

As a result, $\mathcal{F}(S^-)=|\overline{S}-\text{round}(\overline{S})|$. By replacing $\overline{S}$ by $\overline{N}$ or $\overline{P}$ in this expression, one respectively demonstrates the properties (A$_3$) and (A$_9$); indeed, $\frac{1}{|\varepsilon|}>2$ in our study so that $\text{round}\left(\frac{1}{|\varepsilon|}-1\right)=\text{round}\left(\frac{1}{|\varepsilon|}\right)-1$.