Emergence of Large-scale Patterns in Soft Quasicrystals

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Supplementary Information

Supplementary Note 1: Pattern Formation in Materials with Various Levels of Initial Microstructural Chirality.

Here, we discuss the extended results on pattern formation in materials characterized by a wide range of initial microstructural chirality ($\theta = 45°,\ 15°,\ 10°,\ 4.5°$, and $0°$). For clarity, we refer to each sample explicitly by its chirality angle, such as $\chi^{\theta=15°}$, instead of using the "strong" or "weak" notation. The extended configurations discussed are summarized in Extended Data Table 1, with each configuration defined by the chirality rotation of elliptical voids (θ), normalized major (b/L) and minor (a/L) diameters of the elliptical voids, and the normalized diameter (d/L) of the circular voids. Note that the configurations $\chi^{\theta=45°}$ and $\chi^{\theta=15°}$ correspond directly to the strong (Fig. 3) and weak (Fig. 2) chirality samples reported in the main body of the manuscript, respectively. Additional configurations, including $\chi^{\theta=10°}$, $\chi^{\theta=4.5°}$ and $\chi^{\theta=0°}$, represent results with progressively weaker chirality. One may notice that, aside from the chirality angles, the configurations in Extended Data Table 1 do not share identical void dimension parameters. This discrepancy arises because, given the complex micro-mechanisms, merely specifying a chirality angle does not guarantee large-scale pattern formation. Most configurations with the same θ fail to produce large-scale patterns. For instance, as discussed in Supplementary Note 2, certain configurations with $\theta = 35°$ (Extended Data Fig. 3a) and $\theta = 5°$ (Extended Data Fig. 3b) did not form large-scale patterns even under sufficiently high strains. Therefore, in Extended Data Table 1, the overall material porosity of some configurations is adjusted—if necessary—by modifying the void dimension parameters b/L, a/L and d/L (while largely maintaining their ratios) to achieve successful pattern formation.

In Extended Data Fig. 1, we present the large-scale patterns obtained from the configurations: $\chi^{\theta=45°}$, $\chi^{\theta=15°}$, $\chi^{\theta=10°}$, $\chi^{\theta=4.5°}$ and $\chi^{\theta=0°}$, from top to bottom. The identified characteristic lengths for these large-scale patterns are listed in the last column of Extended Data Table 1, as $L_{cr} = 13(\delta_S)^{-1}L$, $13L$, $13(\delta_S)L$, $13(\delta_S)^2L$, and a "longwave" length (exceeding the sample size) respectively, where L is the prototile side length in the base tiling and $\delta_S = 1 + \sqrt{2}$ is the silver ratio. All results were numerically derived from sufficiently large samples with a size of $W_S = 440L$. The identification of the characteristic lengths was implemented using the same mapping method described in Fig. 4 and verified through the SSIM analysis shown in Fig. 5. Among all configurations, the $\chi^{\theta=45°}$ sample exhibits the smallest characteristic length. Further reductions in the chirality angle θ result in large-scale patterns with an increase in the characteristic length. Interestingly, this set of increasing characteristic lengths fit into a geometric sequence governed by the silver ratio δ_S, expressed with a general formula $L_{cr} = 13(\delta_S)^N L$, where $N = -1,\ 0,\ 1,\ 2,\ \cdots$. Note that, the $\chi^{\theta=0°}$ sample exhibits no large-scale pattern formation within a finite sample size, and thus its characteristic length is regarded as "longwave" ($L_{cr} \to \infty$). Overall, Extended Data Fig. 1 demonstrates that these patterns exhibit strong morphological similarity but develop at different length scales, implying that the reassembly of quasi-crystalline order within the material can occur at various scales depending on the initial chirality of the voids.

Additionally, we present detailed results for the $\chi^{\theta=10°}$ sample, which is modified from the χ_W configuration (Fig. 2) by reducing the chirality angle to $10°$. Numerical results for this configuration are shown in Extended Data Fig. 2. Specifically, Extended Data Fig. 2a plots the average compressive stress (σ) as a function of strain (ε), while Extended Data Fig. 2b and 2c illustrate the corresponding material patterns and their zoomed-in views at various strain levels ($\varepsilon = 0,\ 0.03,\ 0.04,\ 0.05,\ 0.055,$ and 0.0625). We observe that microstructural transformations propagate significantly faster and extend further in the $\chi^{\theta=10°}$ sample compared to $\chi^{\theta=15°}$ configuration, resulting in an obviously larger pattern ($\varepsilon = 0.0625$ in Extended Data Fig. 2b). By simulations on a sufficiently large sample (Extended Data Fig. 1c), we identify the characteristic length of $\chi^{\theta=10°}$ sample as $L_{cr}^{\theta=10°} = \delta_S L_{cr}^{\theta=15°}$, where $L_{cr}^{\theta=15°}$ is the characteristic length identified for the χ_W sample (Fig. 4). Additionally, compared to $\chi^{\theta=15°}$ configuration, the $\chi^{\theta=10°}$ sample exhibits slightly higher stiffness prior to nucleation, reaches its critical strain earlier, and results in nearly identical stiffness in the post-transformation stage.

Supplementary Note 2: Illustration of Microstructures that do not Develop Large-scale Patterns

In our study, most configurations, explored across a broad microstructural parameter space, fail to develop large-scale patterns due to incomplete progression through three critical stages: i) local nucleation, ii) enough propagation of transformation, and iii) confinement of transformation within distinct domains. Here, we present two typical examples in which large-scale patterns do not emerge.

The sample shown in Extended Data Fig. 3a ($b/L = 0.81$, $a/L = 0.432$, $d/L = 0.75$ and $\theta = 35°$) exemplifies a failure case in which nucleation is initiated but quickly arrested by gridlocks, preventing further propagation. Specifically, as the compressive strain increases from $\varepsilon = 0.05$ to $\varepsilon = 0.08$, densely clustered nucleation spots emerge. The potential propagation paths originating from these spots overlap, leading to strong competition that induces a gridlock effect. This mutual obstruction prevents any single nucleation spot from successfully propagating even at a high compressive strain ($\varepsilon = 0.11$), thereby suppressing large-scale pattern formation.

In contrast, some other microstructures fail to form large-scale patterns due to the absence of localized nucleation upon the initiation of void collapse. Instead, structural collapse is triggered simultaneously across the sample. For example, the sample shown in Extended Data Fig. 3b ($b/L = 0.6675$, $a/L = 0.5$, $d/L = 0.8$ and $\theta = 5°$) undergoes a transition from stable compression to void collapse between $\varepsilon = 0.05$ and $\varepsilon = 0.11$. However, no concentrated nucleation spots are observed; instead, the transformation initiates uniformly across the sample. Consequently, no large-scale patterns were observed in the fully collapsed structure, even at a high enough compressive strain ($\varepsilon = 0.18$).

Supplementary Photo 1: High-resolution photograph of large-scale pattern formation in weak chirality (χ_W) sample at the strain level $\varepsilon = 0.133$.

Supplementary Photo 2: High-resolution photograph of large-scale pattern formation in strong chirality (χ_S) sample at the strain level $\varepsilon = 0.135$.

Supplementary Movie 1: Experimental video of weak chirality (χ_W) sample under equi-biaxial plane-strain compression from $\varepsilon = 0$ to $\varepsilon = 0.133$ (video sped up by a factor of 10).

Supplementary Movie 2: Numerical simulation video of large-scale pattern formation in weak chirality (χ_W) sample from $\varepsilon = 0$ to $\varepsilon = 0.0625$. The left panel shows the full view, while the right panel provides a zoom-in view.

Supplementary Movie 3: Numerical simulation video of large-scale pattern formation in strong chirality (χ_S) sample from $\varepsilon = 0$ to $\varepsilon = 0.0985$. The left panel shows the full view, while the right panel provides a zoom-in view.

Supplementary Movie 4: Numerical simulation video of pattern formations in $\chi^{\theta=45°}$, $\chi^{\theta=15°}$, $\chi^{\theta=10°}$, and $\chi^{\theta=4.5°}$ samples (with sample size of $W_S = 440L$), respectively, from top to bottom.