Hyperuniform monocrystalline structures by spinodal solid-state dewetting

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Materials featuring anomalous suppression of density fluctuations over large length scales are emerging systems known as disordered hyperuniform. The underlying hidden order renders them appealing for several applications, such as light management and topologically protected electronic states. Spontaneous formation of stable patterns could be a viable solution for the implementation of this peculiar class of systems via bottom-up approaches. However, scalable fabrication methods for disordered hyperuniform architectures are missing, limiting their potential application over large-scale. Here we show for the first time that mono-crystalline, semiconductor structures can undergo spinodal solid-state dewetting and that they feature correlated disorder with an effective hyperuniform character. Nano- to micrometric sized structures can be engineered using silicon-on-insulator substrates and Si$_{1-x}$Ge$_x$ deposition. Phase-field simulations explain the underlying non-linear dynamics and the physical origin of the emerging patterns, which rely on solid-state dewetting triggered by elasticity. Our method is a distinct and scalable approach for the controlled self-assembly of monocrystalline, dielectric, disordered hyperuniform metamaterials towards the manipulation of electricity, fluids, and light.

Disordered hyperuniform materials are an emerging class of disordered systems featuring suppression of density fluctuations on large length scales [14]. While not presenting any Bragg peak in diffraction (as a liquid), they have strongly suppressed density fluctuation at long distances (as an ordered crystal). The underlying hidden order results in exotic phenomena such as topologically protected electronic states [5], glassy electronic quantum state transitions [6], Anderson localization of light [7], polarization selectivity [8], lasing [9] and a full photonic band-gap for light propagation [10–13].

In this framework, spontaneously-formed disordered patterns, as those obtained in systems described by Swift-Hohenberg [14] [15] and Cahn-Hilliard equations for phase separation [16], have been recently proposed as possible platforms for implementing hyperuniform materials [17]. In these systems, the presence of long-range interactions and the underlying non-linear dynamics lead to the onset of patterns featuring correlated disorder (deviating from a Gaussian random field [18]). Such patterns have been reported in thin films of polymers [19, 20] and liquid metals [18], where they result by spinodal dewetting (so termed due to the similarities with spinodal decomposition in terms of morphologies). This process consists in the breakup of thin films through the amplification of uniformly distributed surface undulations rather than due to the interface-energy driven retraction of film edges as in usual dewetting processes [19, 21]. This phenomenology has remained mostly inaccessible in the technologically relevant case of semiconductors materials and, more generally, in monocrystalline systems owing to the different dynamics and driving forces at play with respect to the conventional cases of liquid metals and polymers [18, 29].

Here, we report on the annealing of semiconductor monocrystalline thin films that undergo spinodal solid-state dewetting, leading to morphologies with hyperuniform character. Solid-state dewetting of Si$_{1-x}$Ge$_x$ layers deposited on ultra-thin silicon on insulator (UTSOI), can be engineered to control and tune the morphology (connected, disconnected) and typical size (from 100 nm up to 10000 nm) of monocrystalline, atomically smooth nano- and micro-architectures. The morphology of these structures shows deviations from random patterns, as accounted for by a theoretical analysis exploiting Minkowski functionals, thus exhibiting correlated disorder. Three-dimensional phase-field simulations map the experimental observations reproducing their main features as a result of the interplay between elasticity effects and solid-state dewetting. Hyperuniform metrics obtained from the analysis of the spectral density of experimental patterns assess the effective hyperuni-
FIG. 1. Spinodal solid-state dewetting. a) Scheme of the system under investigation: ultra-thin silicon on insulator substrates (14 nm thick Si on 25 nm thick SiO$_2$, buried oxide, BOX) followed by epitaxial deposition of Si$_{1-x}$Ge$_x$ alloys and high temperature annealing in a molecular beam reactor (MBE). b) Scanning electron micrographs (SEM) displaying the morphological evolution (from the edge of the sample, left panel, towards its center, right panel) of 150 nm Si$_{0.7}$Ge$_{0.3}$ on UT-SOI after 4 hours annealing at 800°C (see also the SEM characterization in Supplementary Figure 1 and the optical microscopy in Supplementary File 1 for more information about the temperature gradient and morphology changes). c) Left panel: transmission electron micrograph (TEM) of a dewetted island. Right panel: high-resolution TEM highlighting the interface between the BOX, some pristine UT-SOI and the Si$_{0.7}$Ge$_{0.3}$ (See also the Supplementary Figure 2).

formity [22] and agree with theoretical predictions for patterns obtained by spinodal decomposition via Cahn-Hilliard dynamics [17].

The experimental results are illustrated in Fig. 1. Si$_{1-x}$Ge$_x$-based alloys, with variable composition and thickness, are deposited on monocrystalline UT-SOI (diced in 2 cm $\times$ 2 cm samples) in a molecular beam epitaxy reactor (MBE) where they undergo solid-state dewetting under high-temperature annealing (with temperature between 400 and 800°C for periods ranging from 30 minutes to 6 hours).

Keeping the Si$_{1-x}$Ge$_x$ film thickness and annealing time constant leads to a continuous evolution from holes to connected- (noodles-like-) structures and isolated islands by increasing the processing temperature (Fig. 1(b)). In large structures (e.g. obtained from >200 nm thick films of Si$_{0.7}$Ge$_{0.3}$) we observe trading dislocations propagating along the (111) direction typically starting from the UT-SOI/Si$_{1-x}$Ge$_x$ interface (Fig. 1(c)) as expected in crystalline bilayer systems with lattice mismatch. Further details are provided in the Supplementary Information showing that the dewetted structures are monocrystalline and present faceting, terraces and step bunching in 500 nm thick Ge layers after dewetting.

A given morphology (e.g., holes, islands and connected structures) can be obtained in a wide range of Si$_{1-x}$Ge$_x$ thicknesses (from 5 to 700 nm) and compositions (from x = 0.3 up to 1) by an appropriate choice of the annealing temperature and time, providing structures with typical size ranging from hundred nanometers to several microns (Fig. 2). As an example, for a Ge thickness of 50 nm and content x = 1 we can obtain isolated islands or connected structures (Fig. 2 a) and b)). Conversely, a given morphology can be obtained for a different Ge content and thickness (Fig. 2 c) and d) and Supplementary Figure 2).

In contrast with conventional UT-SOI solid-state dewetting, commonly understood in terms of heterogeneous nucleation of holes and surface-diffusion limited kinetics [21, 23–25], we name the process illustrated in Fig. 1 and 2 as spinodal solid-state dewetting due to the simultaneous breakup of the film and to the morphology of the resulting disordered structures. In spite of the different driving forces at play, marked similarities with polymers and liquid metals [14, 15, 18, 26–28] are here observed. For these classes of materials, the spinodal dewetting [21] is triggered by molecular forces and interface potentials [29].

The system illustrated here is heteroepitaxial with a strained bilayer owing to the lattice mismatch between Si$_{1-x}$Ge$_x$ and Si, where these driving forces are not present. However, when considering an ideally infinite Si substrate, this system undergoes an Asaro-Tiller-Grinfeld instability [30–32], where misfit strain, with partial or absent plastic relaxation, triggers surface corrugation [33] and island formation [34, 35]. Trenches between islands form and deepen within the substrate while mass can flow across them. Therefore, no separation between structures nor dewetting occurs while coarsening takes place on a bulk substrate. The key difference in the work
reported here, is the use of an UT-SOI as substrate lying on the amorphous BOX: the trenches formed within the Si$_{1-x}$Ge$_x$ film triggered by elasticity dig into the UT-SOI, suddenly uncover the BOX underneath and there initiate solid-state dewetting. Thus, the onset of the dewetting instability is simultaneous across the sample (as for the trenches in the Asaro-Tiller-Grinfeld instability), differing from the conventional onset of dewetting (as for the trenches in the Asaro-Tiller-Grinfeld instability, see text) to simultaneous breakups.

Patterns qualitatively similar to the experimental ones are produced by a minimal phase-field model, which accounts for the energetics of surfaces and the relaxation of biaxial (misfit) strain [36] (Fig. 3). The model thus includes both Asaro-Tiller-Grinfeld and dewetting instabilities. It is a degenerate Cahn-Hilliard model coupled with elasticity. The dynamic equation entering this model in non-dimensional form reads

$$\frac{\partial \varphi}{\partial t} = D \nabla \cdot [M(\varphi)\nabla \mu(\varphi)].$$

$\varphi$ is a continuous order parameter, set here to 1 in Si$_{1-x}$Ge$_x$ and UT-SOI and 0 in vacuum, and changing smoothly across the interface over a length $\ell$. $M(\varphi) = (36/\epsilon)\varphi^2(1 - \varphi)^2$ and $D$ is a diffusion coefficient (incorporated in the timescale $t' = t/D$) [36,38]. $\mu(\varphi)$ encodes the total chemical potential and can be derived by the total variation of the free energy of the system with respect to $\varphi$. It reads

$$\mu(\varphi) = \mu_s(\varphi) + \mu_e(\varphi) = \gamma \kappa(\varphi) + \rho_e(\varphi)$$

including surface ($\mu_s$) and elastic ($\mu_e$) contributions with $\gamma$ the isotropic surface-energy density, $\kappa(\varphi)$ the approximation of the curvature in terms of $\varphi$ and $\rho_e(\varphi) = \varepsilon : C(\varphi) : \varepsilon$ the elastic energy density [36,39]. $C(\varphi)$ is the elastic constant tensor and $\varepsilon = \nabla \mathbf{u} + (\nabla \mathbf{u})^T + h(\varphi)\varepsilon_0\mathbf{I}$ the strain tensor, with $\mathbf{u}$ displacement and $\varepsilon_0$ the effective misfit strain between Si$_{1-x}$Ge$_x$ and UT-SOI. Due to its very small thickness, the Si layer is not explicitly considered but its presence is encoded in $\varepsilon_0$ by strain accumulation. $h(\varphi)$ is a smooth function which vanishes...
outside Si_{1-x}Ge_{x} and UT-SOI. ε is determined by solving the mechanical equilibrium equation \( \nabla \cdot \mathbf{\sigma}(\varphi, \mathbf{u}) = 0 \) with \( \mathbf{\sigma} = C(\varphi, \mathbf{u}) : \varepsilon(\mathbf{u}) \) at every step. The amorphous BOX is not modeled but is accounted for by a no-flux boundary condition for \( \varphi \) enforcing a 90° contact angle and allowing for dewetting, without loss of generality \[24, 41\]. Other effects, such as plastic relaxation and intermixing can be treated as changes in the eigenstrain in a mean field approximation. Therefore, similarly to previous studies on similar systems \[42\], which demonstrate good comparison with experimental data under these assumptions, they are not explicitly considered in the model. As our focus is on the emerging morphology, these properties are clearly observed in patterns obtained from thresholded grey-scale images of the morphology. These properties are clearly observed in patterns obtained from thresholded grey-scale images of the morphology.

The simulated evolution of an initially perturbed film is illustrated in Fig. 3 in the initial corrugation amplifies over time. In agreement with the Asaro-Tiller-Grinfeld instability, well describing the early stages of the evolution, the corrugation has a characteristic wavelength \( \ell \) \[46\]. When the trenches reach the substrate, dewetting sets in. Notice that this onset occurs everywhere in the film almost simultaneously. Despite the simplified hypotheses used for the model, the main features of the dewetting dynamics are qualitatively reproduced. This holds true for different film thicknesses, annealing time and temperature (accounted for by diffusion coefficient \( D \), depending on temperature by an Arrhenius law \[42\]).

The morphology of the dewetted structures obtained both in the experiments and simulations are analysed by Minkowski functionals \[14, 48, 49\]. These integral geometry measures quantify the topological properties and, in turn, spatial features of 2D patterns that cannot be captured by eye or simple 2D image analysis, such as correlation function or Fourier transform. In particular, we consider the Minkowski functionals of thresholded patterns from digitized grey-scale images of the morphologies \( m_{1,2}(A) \) as function of \( \mu_{0}(A) \), with \( A \) a compact 2D domain as obtained from thresholded height profiles, \( \mu_{0}(A) = F \) with \( F \) area of \( A \), \( m_{1}(A) = (1/2\pi) U \) with \( U \) boundary length of \( A \), and \( m_{2}(A) = (1/\pi) \chi \) Euler characteristic \[28\] (see also Supplementary Information). These quantities for experimental and simulated patterns almost overlap for most of the \( \mu_{0} \) range (Fig. 3(c)), assessing the driving forces at play included in the phase-field simulation as the relevant ones. Different experimental connected and disconnected structures, show a similar behavior for both \( m_{1}(A) \) and \( m_{2}(A) \) (see the range illustrated by dashed lines in Fig. 3(c)).

Spinodal patterns, like those emerging from phase separation and spinodal dewetting, can exhibit disordered hyperuniformity \[17, 22\]. Based on that theoretical prediction, we assess further the correlation properties of the dewetted structures by looking at the spectral density, \( \psi^{*}(|k|) \) with \( k = (k_{x}, k_{y}) \), of their correlation function \( \psi(x,y) \) computed from a given height profile \( Z(x,y) \) and, in particular, at its decay for small wavenumbers (long wavelength) \[17, 22, 50\]. Patterns can be considered to be hyperuniform if \( \psi^{*}(|k|) \sim |k|^\beta \) and \( \beta \geq 4 \) with \( |k|/\bar{k} \rightarrow 0 \) (\( \bar{k} \) is the position of the maximum in the reciprocal space). \( \beta = 4 \) corresponds to a Gaussian random field whereas it is larger in the presence of correlations leading to stronger suppression of long wavelengths oscillations \[3, 4\].

These properties are clearly observed in patterns obtained by spinodal solid-state dewetting discussed above (Fig. 4): \( 5 < \beta < 6 \) is obtained for \( 0.1 < |k|/\bar{k} < 1 \) in most of the analyzed patterns and \( \beta \sim 6 \) when considering sufficiently large images (Supplementary Figure 3).
However, this steep decrease of $\psi^*(|k|)$ is lost for smaller $k$. While for hyperuniform structures $H = \psi^*(0^+)/\psi^*(k)$ should be strictly zero, a $H \leq 10^{-4}$ is accepted to be a fingerprint of hyperuniformity, at least for real systems [3, 4], in particular in the presence of other arguments supporting long-range interactions and correlations [51] and inherent spurious effects arising from image thresholding [17]. We thus consider our emergent patterns by spinodal solid-state dewetting as hyperuniform. Aside, we also remark that, in agreement with recent reports on devices implemented via top-down fabrication [52], connected structures (as those shown in Fig. 1(c)) are less hyperuniform with respect to the broken ones (as those shown in Fig. 3(c)(inset)).

Other natural or artificial systems such as polymer-grafted nanoparticles [63], polymer melt [64] or maximally random jammed packing [65] have been proposed as potential candidates for hyperuniform materials and could be implemented from bottom-up assembly. Theoretical estimates for these systems predict a hyperuniform metrics $H \sim 10^{-4}$. Here we provided evidence that our semiconductor system provides a similar value of this metric rendering it a competitive candidate for realistic applications with disordered hyperuniform materials.

Disordered structures are more robust against fabrication imperfections if compared with the ordered counterpart. As such, in principle, devices based on disordered systems are more apt to be fabricated over large scales via bottom-up methods. However, in spite of the increasing interest in the features emerging from disordered hyperuniform systems, a realistic platform for electricity, light or matter management with hyperuniform materials is still missing. Real effectively hyperuniform devices have been implemented over rather small extensions and with more or less cumbersome top-down approaches [7, 12, 13, 52]. In this respect, our approach is more convenient, as the dewetting instability is independent of the system size and can be implemented in a single fabrication step. The commercial UT-SOI wafers we use are available in a wide range of device and buried oxide thickness, as well as with different $p$ or $n$ type doping, which is important in view of electrically-active disordered hyperuniform metamaterials.

The resulting stable patterns are similar to what has been reported in soft matter and liquids metals [14, 15, 18, 20, 25]. However, compared with these systems, the use of $\text{Si}_1-x\text{Ge}_x$ alloys on an insulating substrate (the buried oxide) offers invaluable advantages for device fabrication (e.g. electrical and optical isolation from the substrate toward the implementation of opto-electronic devices).

Solid-state dewetting allows tuning the typical size (and spacing) of the structures permitting, in principle, to use them for applications in photonics at visible and near infrared frequencies [7, 12, 13, 52]. The use of $\text{Si}_{1-x}\text{Ge}_x$ alloys having high refractive index ($n_{1000\text{ nm}} \sim 4$) and low absorption losses ($k_{1000\text{ nm}} \sim 0.17-10^{-4}$) while being monocrystalline and atomically smooth, can provide superior performances with respect to soft materials ($n_{1000\text{ nm}} = 1.5$) and metals (e.g. for gold $k_{1000\text{ nm}} \sim 6.3$) for light manipulation.

In conclusion, we identified an elasticity-driven instability in strained, thin films undergoing solid-state dewetting, enabling the formation of spinodal patterns that exhibit effective hyperuniformity. We have shown that spinodal solid-state dewetting is a very effective process in forming correlated disorder with tunable morphology (holes, connected structures, and isolated islands), composition ($\text{Si}_{1-x}\text{Ge}_x$ alloys with Ge content from $x = 0.3$ to $1$), and typical size (from 100 nm to 10000 nm). Finally, the choice of an UT-SOI substrate and $\text{Si}_{1-x}\text{Ge}_x$ alloys, which are common and cost-effective materials, opens the path to fruitful industrialization for implementing electronic, photonic, and microfluidic hyperuniform devices on 300 nm wafers.

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SUPPLEMENTARY INFORMATION
Hyperuniform monocrystalline structures by spinodal solid-state dewetting
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Methods

All the dewetted structures are obtained starting from the same ultra-thin, silicon-on-insulator wafer (UT-SOI, double polished, oriented along the (001) direction): 14 nm of Si on 25 nm of SiO2 (buried oxide, BOX) on bulk Si. The UT-SOI is first cleaned in a HF solution (∼5%) for 20 s in a glove box under nitrogen atmosphere in order to remove the native oxide. Then, it is introduced in the ultra-high vacuum of a molecular beam epitaxy (MBE) reactor (static vacuum ∼10−10 Torr). The samples undergo a first annealing flash at 600 °C for 30 s in order to remove any trace of native oxide. SiGe alloys of variable thickness are deposited on the UT-SOI and annealed at temperature ranging from 400 °C to 800 °C in order to tune the dewetted morphologies (e.g. island or connected nano-architectures). After solid-state dewetting, the samples are investigated via atomic force microscopy (AFM) in non-contact mode, high resolution scanning electron microscopy (SEM), and transmission electron microscopy (TEM).

Morphology tuning

The morphology of the dewetted islands can be tuned in a wide range of SiGe thickness and Ge content adjusting annealing temperature and time. As an example, in Figure 1 in the main text is reported the case of temperature change for a given deposited thickness and annealing time. In Figure S1 a more precise characterization of the same sample is provided. This is obtained on an individual sample observing the evolution of the layer morphology from the sample edge towards its center. In this case the nominal annealing temperature was 800°C as measured with a pyrometer at the center of the sample. Owing to the holder that clamps the sample at its edges, the temperature on the sides is lower than at the sample center. This allows to monitor the effect of temperature while not changing all the other conditions (SiGe thickness, annealing time). Provided that at the sample edges the dewetting is barely visible (a few holes) we estimate a temperature at the edge of about 600°C.

From optical microscopy on a similar sample (200 nm Si0.7Ge0.3 on UT-SOI after 4 hours annealing at 800°C), by scanning over 2 mm from the edge, we estimate that the morphology change occurs in about 0.5 mm. Thus, in the central part of the sample, the morphology is the same thus providing homogeneous properties. This image is provided as a separate high-resolution Supplementary File 1, allowing to zoom-in to better see the fine details of the dewetted structures. It was obtained with a Leica optical microscope with a 100× magnification objective lens in dark field configuration.

Dislocations in large structures from thick Si0.7Ge0.3 dewetting

The presence of strain between thick Si0.7Ge0.3 layers and the underlying silicon on insulator wafer potentially leads to plastic relaxation and formation of dislocation. This is very evident also from scanning electron micrographs from large islands.

As an example, in Figure S2 a) we show the case of 500 nm Ge on UT-SOI after 6 hours annealing at 800°C. The overall morphology is very similar to that observed in thinner samples (as shown in the main text). The large size of the islands allows to highlight the presence of facets, terraces and step bunching (Figure S2 b-d)). From the same analysis is evident the presence of a dislocation network emerging on the islands surface.

Atomic force microscopy of dewetted samples

For a precise assessment of the disorder properties (e.g., hyperuniform character) of the dewetted structures we use atomic force microscope, high-resolution and large images. We performed this characterization over
Phase-Field model

A phase-field model that accounts for both surface and elastic energy is considered. It is based on the model introduced in Refs. [S1], where a continuous order parameter $\varphi \equiv \varphi(\mathbf{x})$ that is set to be $\varphi(\mathbf{x}) = 1$ for $\mathbf{x}$ in the solid phase (SiGe, Si), $\varphi(\mathbf{x}) = 0$ for $\mathbf{x}$ in the vacuum, and with a continuous transition in between well described by

$$
\varphi(\mathbf{x}) = \frac{1}{2} \left( 1 - \tanh \left( \frac{3d(\mathbf{x})}{\epsilon} \right) \right). \quad (S1)
$$

$\epsilon$ is the interface thickness between solid and vacuum phase. $d(\mathbf{x})$ corresponds to the signed distance from the surface of the solid phase, represented as the 0.5 level set of $\varphi$. The model is based on the free-energy functional

$$
F = \int_{\Omega} \left( \frac{\epsilon_0}{2} | \nabla \varphi |^2 + \frac{1}{\epsilon} B(\varphi) \right) d\mathbf{x} + \int_{\Omega} \rho_e(\varphi) d\mathbf{x}, \quad (S2)
$$

with $\Omega$ the 3D simulation domain and $\mathbf{x} \in \Omega$. The first integral in Eq. (S2) accounts for the surface energy with $B(\varphi) = 18\varphi^2(1-\varphi)^2$ and $\gamma_0$ the isotropic surface energy. The second integral in Eq. (S2) accounts for the elastic energy with

$$
\rho_e = \mu h(\varphi) \sum_{ij} \varepsilon_{ij}^2 + \frac{\lambda}{2} h(\varphi) \left( \sum_i \varepsilon_{ii} \right)^2. \quad (S3)
$$

the elastic-energy density, assuming isotropic media, with $\lambda$ and $\mu$ the Lame constants, $h(\varphi) = \varphi^3(6\varphi^2 - 15\varphi + 10)$ a function smoothly vanishing when approaching the vacuum phase, and $\varepsilon$ the strain tensor

$$
\varepsilon = \nabla \mathbf{u} + (\nabla \mathbf{u})^T - \varepsilon_0, \quad (S4)
$$

with displacement $\mathbf{u}$ and eigenstrain $\varepsilon_0$, which encodes the permanent deformation due to the mismatch between the lattice constant of the epilayer (SiGe), $a_e$, and of the substrate (Si) $a_s$ such as $\varepsilon_0 = (a_e - a_s)/a_s$, and

$$
\varepsilon_0 = -h(\varphi)\varepsilon_0 I. \quad (S5)
$$

The strain tensor entering Eq. (S3) is determined by assuming mechanical equilibrium, namely by solving

$$
\nabla \cdot \sigma = 0, \quad (S6)
$$

with

$$
\sigma = \lambda \text{Tr}(\varepsilon) I + 2\mu \varepsilon. \quad (S7)
$$

The evolution law for $\varphi$ approximating mass transport by surface diffusion is given by the conservative gradient flow

$$
\frac{\partial \varphi}{\partial t} = D \nabla \cdot \left( M(\varphi) \nabla \mu \right), \quad (S8)
$$

with $M(\varphi) = (36/\epsilon)\varphi^2(1-\varphi)^2$ a degenerate mobility restricted to the surface and $D$ the effective diffusion coefficient. The latter expected to depend on temperature by an Arrhenius law, encoding the activation of surface diffusion at high temperature. Without loss of generality it is here set to 1. The effect of $D$ is incorporated in the timescale such as $t' \to t/D$. $\mu = \delta F/\delta \varphi = \mu_s + \mu_e$ is determined by

$$
g(\varphi)\mu_s = -\epsilon_0 \nabla^2 \varphi + \frac{\gamma_0}{\epsilon} B'(\varphi) \quad (S9)
$$

with $g(\varphi) = 30\varphi^2(1-\varphi)^2$ an additional degeneracy improving the description of surface diffusion for relatively large $\epsilon$ [S1][S3]. A contact angle of $90^\circ$ at the substrate (BOX) is enforced by a no-flux boundary condition for the order parameter $\varphi$ [S4]. The model presented so far allows for reproducing the main experimental features reported in this work. Model extensions can be considered for modeling of the substrate or of a second solid phase [S5][S7], alloys [S8], surface-energy anisotropy leading to surface faceting [S9][S11] and different contact angles [S4].

Simulations were performed by employing the parallel Finite Element toolbox AMDiS [S12][S13], allowing for time adaptivity and mesh refinement at the solid-vacuum interface. The equations governing the evolution of $\varphi$ are solved as a system of two second-order partial differential equation, namely for $\varphi$ and $\mu$, and discretized by a semi-implicit integration scheme (see Ref. [S1][S4][S14]). Periodic boundary conditions are imposed at the lateral boundaries for $\varphi$, while no-flux Neumann boundary conditions are imposed elsewhere. Strain fields at equilibrium are computed by solving Eq. (S6) for $\mathbf{u}$ at each timestep. Periodic boundary conditions are imposed along in-plane directions, while a Dirichlet boundary condition with $\mathbf{u} = 0$ is imposed at the bottom of the simulation domain.

Fig. [S4] shows different outcomes according to the wavelengths encoded in the initial perturbation. In agreement with the literature, wavelengths smaller than the characteristic length of the Asaro-Tiller-Grinfeld (ATG) instability must be considered to account for the proper development of the instability for an unpatterned film. The outcome of simulations with initial perturbation containing large wavelength only, may suggest however a way to obtain striped patterns by a proper preparation of the substrate.

Pattern Analysis

The analysis of patterns through Minkowski functionals follows the work reported in Ref. [S15]. The Minkowski functionals, $m_i(A)$ with $A$ a compact 2D do-
m_0(A) = \int_A \int d^2x \\
m_1(A) = \frac{1}{2\pi} \int_{\partial A} dx \\
m_2(A) = \frac{1}{2\pi^2} \int_{\partial A} \frac{1}{R} dx \quad (S10)

with the measure \(dx\) and \(R\) the radius of curvature of \(\partial A\). These functionals delivers the following information:

\[ m_0(A) = F \]

\[ m_1(A) = \frac{1}{2\pi} \]

\[ m_2(A) = \frac{1}{2\pi^2} \]

with \(F\) the area of \(A\), \(\chi\) the Euler characteristic. We used the discretized Minkowski formulation and the marching cube/marching square algorithm for grey-scale images implemented in the software enclosed with Ref. [S15]. In the main text, the analysis of patterns emerging from experiments and simulations is compared to the Minkowski functionals obtained in the case of a Gaussian Random Field. This corresponds to a field with Gaussian probability density functions which can be obtained by sum of a large number of plane waves with uniformly distributed phases. The Minkowski functionals for a Gaussian Random Field (GRF) are given by the following expressions [S15].

\[ m_0^{\text{GRF}}(A) = \frac{1}{2} \left[ 1 - \text{erf} \left( \frac{\rho - \rho_0}{\sqrt{2}\sigma} \right) \right] \]

\[ m_1^{\text{GRF}}(A) = \frac{k}{\sqrt{8\pi}} e^{-\frac{1}{2}(\rho-\rho_0)/\sigma} \]

\[ m_2^{\text{GRF}}(A) = \frac{k^2}{\sqrt{2\pi^3\sigma^2}} (\rho - \rho_0) e^{-\frac{1}{2}(\rho-\rho_0)/\sigma} \]

Following [S15] we set \(k = 1\), \(\sigma = 1\), \(\rho_0 = 0\). A numerically generated Gaussian Random Field and its analysis are shown in Fig. S5.

The analysis of the hyperuniform nature of the patterns emerging by the solid-state dewetting process has been performed following Refs. [S16–S18]. The height profile of the structures, \(H(x, y)\), is considered by its auto-correlation function \(\psi_H(x, y)\). \(\psi_H^*(k_x, k_y)\) denotes the 2D Fourier transform of \(\psi_H(x, y)\) with \(k_x, k_y\) the components of the (2D) wave vectors. \(\psi^*(r)\) and \(\psi^*(|k|)\) denote the same quantity evaluated as radial distributions in the corresponding domains, with \(r = \sqrt{x^2 + y^2}\) and \(|k| = \sqrt{k_x^2 + k_y^2}\) the distance from the center of the frame of reference in the real and reciprocal space, respectively. Length scales in the real and reciprocal space are scaled according to the characteristic length emerging in the pattern. This consist of \(\hat{k}\) and \(\lambda = 2\pi/\hat{k}\) in the reciprocal and real space, respectively. With this choice the quantities are evaluated with respect to the physical feature of the pattern and are independent from image resolution and size, provided that these are large enough to resolve \(\lambda\). To assess the hyperuniformity character of the patterns the decay of \(\psi^*(|k|)\) for \(\psi^*(|k| \to 0)\) is evaluated in detail. Patterns can be considered to be hyperuniform if \(|\psi^*(|k|)| \leq |k|^4 \) and \(\psi^*(|k|)/\psi^*(\hat{k}) \leq 10^{-4}\) for \(|k| \to 0\) [S16, S17].
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FIG. S1. Scanning electron micrographs (SEM) displaying the morphological evolution (from the edge of the sample, left panel, towards its center, right panel) of 150 nm Si$_{0.7}$Ge$_{0.3}$ on UT-SOI after 4 hours annealing at 800°C. Another example from a different sample is provided as a separate high-resolution Supplementary File 1 where optical microscopy show that the change in morphology occurs over about 0.5 mm from the edge.

FIG. S2. a) Scanning electron micrograph of 500 nm Ge on UT-SOI after 6 hours annealing at 800°C. b) Detail of a dewetted island with large facets. c) Blow up of the area highlighted by a white square in b). d) Detail of facets, terraces and step bunching.
FIG. S3. Large-scale atomic force microscope image, $40 \times 40 \, \mu m^2$ with a $2048 \times 2048$ points, of 25 nm Ge deposited on UT-SOI and annealed at $480^\circ C$ for 45 minutes. This image was used for Minkowski analysis of experimental patterns and for the hyperuniformity analysis.
FIG. S4. Different simulated pattern depending on the wavelength(s) encoded in the initial perturbation, $\lambda_i$ with respect to the characteristic length of the ATG instability $\ell$. a) $\lambda_i < \ell$, b) $\lambda_i > \ell$.

FIG. S5. Numerically generated Gaussian random field (GRF) and its analysis.