Delocalization-Induced Interface Broadening in Strongly Interacting Systems

Kristian Blom,¹ Noah Ziethen,² David Zwicker,² and Aljaž Godec¹,∗

¹Mathematical bioPhysics group, Max Planck Institute for Multidisciplinary Sciences, Göttingen 37077, Germany
²Theory of Biological Fluids, Max Planck Institute for Dynamics and Self-Organization, Göttingen 37077, Germany

(Dated: July 13, 2022)

Most of our current understanding of phase separation is based on ideas that disregard correlations. Here we illuminate unexpected effects of correlations on the structure and thermodynamics of interfaces and in turn phase separation, which are decisive in systems with strong interactions. Evaluating the continuum limit of the Ising model on the Bethe-Guggenheim level we develop a Cahn-Hilliard theory that takes into account pair correlations. For a one-dimensional interface in a strip geometry these are shown to cause an effective interface broadening at interaction strengths near and above the thermal energy, which is verified in the Ising model. Interface broadening is the result of an entropy-driven interface delocalization, which is not accounted for in the widely adopted Flory-Huggins theory. Pair correlations enforce a thermodynamically optimal configuration of defects and profoundly affect nucleation and spinodal decomposition at strong coupling.

Instigated by the seminal works of Cahn and Hilliard [1–3] phase separation—the process through which distinct phases form from a homogeneous mixture—has attracted considerable attention in a variety of fields, incl. physics [4–13], mathematics [14–16], chemistry [17–20], material science [21–23], and recently biology [24–27]. Our basic understanding of phase separation in systems in [28, 29] and out [30, 31] of equilibrium is mostly based on mean field (MF) ideas [32], also known as regular solution [1], Bragg-Williams [33], or Flory-Huggins [34, 35] theory (for recent works see [10–13, 25–27, 36–41]). MF theory neglects correlations whose importance grows with the strength of interactions [4]. For example, capillary wave fluctuations [41, 42] are not captured in MF theories. This raises doubts about whether MF ideas correctly describe the physical behavior of strongly interacting systems [39, 43].

Various refined techniques have been developed beyond the MF approximation, incl. the cavity method [44], random phase approximation [45, 46], self-consistent field theory [47], and field-theoretic approaches close to criticality [48]. Yet, these techniques either do not apply to non-uniform systems or are applicable in a limited range of interaction strengths. As a result, the phenomenology of phase separation in the strong-coupling limit remains largely unexplored and thus poorly understood.

Here we employ the Bethe-Guggenheim (BG) approximation [50–53] that takes into account nearest-neighbor pair-correlations. By directly evaluating the thermodynamic limit of a spatially inhomogeneous nearest-neighbor interacting Ising model, we derive a Cahn-Hilliard theory on the BG level and effectively accounts for the effects of capillary wave fluctuations. We investigate the phenomenology of interfaces and phase condensation, and find at sufficiently strong interactions an effective broadening of interfaces not accounted for by mean field theories. We corroborate the broadening with computer simulations and exact results in the infinite-interaction limit. Furthermore, via numerical simulations of the newly developed Cahn-Hilliard equation [54] we analyze nucleation kinetics, and observe amplified nucleation barriers and a non-monotonic dependence of the interfacial steepness and critical nucleus size on the in-

FIG. 1. (a) Realizations of spin configurations (top) and corresponding instantaneous interfaces (bottom) in a 2D Ising strip with lattice constant Δ and dimensions \((L_x,L_y)\) = (40,120)Δ for different \(J\) obtained from Monte-Carlo simulations (see [49] for details); the circle and dashed line denote the position of the instantaneous interface. (b) Statistics of interface positions derived from simulations (green) and given by Eq. (7) (line) via a mapping onto the Brownian excursion problem (for details see Appendix). (c) Corresponding ensemble averaged concentration profile along the \(x\)-axis alongside theoretical predictions of mean field (MF; red) and Bethe-Guggenheim (BG; blue) theory. (d) Scaled interface steepness \(L_x\varphi'(0)\equiv L_x\partial_x\varphi(x)|_{x=0}\) as a function of \(J\).
interaction strength.

Motivating example: Interface delocalization.—An intriguing phenomenon in strongly interacting systems is interface delocalization [55–66]. Consider a two-dimensional Ising model with ferromagnetic interaction \( J = J/k_B T \) in a strip geometry (i.e. height \( \gg \) length) in the two-phase regime. Imposing periodic boundary conditions in the vertical direction and thermodynamically co-existing phase compositions at the left/right edges the instantaneous concentration of down-spins projected onto the \( x \) direction, \( \varphi_i(x) \), develops an interface (see Fig. S3a), whose position \( x_1 \) is defined implicitly via \( \varphi_i(x_1) = 1/2 \). In the absence of boundary effects shifting an instantaneous interface \( \varphi_i(x_1) \rightarrow \varphi_i(x_1 + dx_1) \) costs no energy. However, \( x_1 \) near the boundaries are entropically penalized, as they allow only for a limited bandwidth of capillary wave fluctuations (see Fig. S3a, top). As a result, we find at moderate \( J \) that the probability density of \( x_1 \), \( p_{int}(x_1|J) \), is peaked at the center (see Fig. S3b, top), whereas at larger \( J \) the amplitude of capillary waves diminishes (see Fig. S3a, center and bottom) and a transition occurs that delocalizes the interface (see Fig. S3b, center and bottom as well as [61–66]). A sharp but delocalized interface becomes effectively broader upon time-order or ensemble-averaging over respective interface positions (see Fig. S3c-d). Exact results in the regime \( J \rightarrow \infty \) have confirmed the interface broadening [61–64], whereas it is known that mean field theories fail to account for the delocalization transition itself remains elusive. This example therefore motivates a deeper and more systematic analysis of interfaces and phase separation in the strong interaction limit.

Cahn-Hilliard theory including pair correlations.—For simplicity, and without much loss of generality, we limit the discussion to two-dimensional systems, i.e. \( x \in \mathbb{R}^2 \). We start from a two-dimensional Ising model with \( N_x^2 \times N_y^2 \) spins \( \sigma_{ij} = \pm 1 \) on a general lattice with physical dimensions \( L_x \) and \( L_y \), respectively (see Fig. S4a). The Hamiltonian reads (in units of \( k_B T \))

\[
\mathcal{H}(\sigma) = -J \sum_{(km)} \sum_{(ln)} \sigma_{km} \sigma_{mn},
\]

where \( \sigma \) is the matrix containing all spin configurations, \( \bar{J} \geq 0 \) is the (isotropic) interaction strength, and \( \langle ij \rangle \) denotes a sum over nearest neighbors. In [49] we also consider anisotropic interactions distinguishing between the horizontal \( J_x \) and vertical \( J_y \) interaction. Our setup should capture the essence of a general binary system with sufficiently short-range interactions.

We divide the lattice into \( N_x^b \times N_y^b \) boxes, with sides \( L_x/N_x^b \) and \( L_y/N_y^b \), respectively and centroids indexed by \( ij \) with \( i \in \{1, \ldots, N_x^b \}, j \in \{1, \ldots, N_y^b \} \). We divide each spin configuration \( \sigma \) in \( N_x^b \times N_y^b \) blocks encoded in the matrices \( \mathbf{b}_{ij} \) with dimension \( \dim \mathbf{b}_{ij} = N_x^b/N_x \times N_y^b/N_y \). Let \( \varphi_{ij}(\mathbf{b}_{ij}) \equiv (\text{dim} \mathbf{b}_{ij})^{-1} \sum_{\sigma_{mn} = \mathbf{b}_{ij}} \left( 1 - \sigma_{mn} \right) / 2 \) denote the concentration of down-spins in block \( ij \) (see Fig. S4a). For any fixed trial configuration of box concentrations \( \hat{\varphi}_{ij} \in [0, 1] \) which we write in the matrix \( \varphi \), the constrained partition function reads \( Z(\varphi) = \sum_{\sigma} e^{-\mathcal{H}(\sigma)} \prod_{i=1}^{N_x^b} \prod_{j=1}^{N_y^b} \mathbf{1}_{\hat{\varphi}_{ij}[\varphi_{ij}(\mathbf{b}_{ij})]} \) where \( \mathbf{1}_{\varphi}[x] \) is the indicator function of \( x \) [70]. Generally, \( Z(\varphi) \) cannot be determined exactly. BG theory provides a variational approach to \( Z(\varphi) \) on the nearest-neighbor pair-correlation level that exactly accounts for the potential energy. Its accuracy is assessed in [49] for system sizes that are amenable to exact solutions. The total free energy density is evaluated as the twofold ordered scaling limit

\[
\tilde{F}(\varphi(x)) \equiv \lim_{N_x^b \to \infty} \lim_{N_y^b \to \infty} \frac{-1}{N_x^b N_y^b} \left( \frac{\ln Z(\varphi)}{(N_x^b/N_x^b)(N_y^b/N_y^b)} \right),
\]

where \( \tilde{F} \equiv F/k_B T \) and we have defined the scaling limits \( \lim_{N_x^b \to \infty} \lim_{N_y^b \to \infty} \to \left[ \right. \lim_{N_x^b \to \infty} \lim_{N_y^b \to \infty} \right] \). The first limit takes the number of spins to infinity while keeping the system \( L_x \), \( L_y \) and box \( L_x/N_x^b \), \( L_y/N_y^b \) sizes constant (see Fig. S4a middle), making \( \phi_{ij}(\mathbf{b}_{ij}) \in [0, 1] \) a continuous function of \( b_{ij} \) in each box. The second limit takes the number of boxes to infinity while keeping \( L_x, L_y \) constant (see Fig. S4a bottom), thereby making the boxes infinitesimally small and rendering \( \hat{\varphi}_{ij} \rightarrow \varphi(x) \) a continuum field. Evaluating Eq. (2) analytically within the BG approximation (see derivation in [49]) leads to a Cahn-Hilliard functional

\[
\tilde{F} = \frac{1}{V} \int_V dx \left[ \tilde{F}(\varphi(x)) + \frac{1}{2} \nabla \varphi(x)^T \kappa(\varphi(x)) \nabla \varphi(x) \right],
\]

\( V \) being the system’s volume and \( \tilde{F}(\varphi) \) and \( \kappa(\varphi) \) the local free energy density and gradient energy coefficient, respectively. In particular we obtain (see also [71])

\[
\tilde{F}(\varphi) = 2\bar{z} \bar{J} [\zeta(\varphi) - 1/4] + (1 - \bar{z})[\Xi(\varphi) + \Xi(1 - \varphi)] + \bar{z}[\Xi(\varphi - \zeta(\varphi))/2 + \Xi(1 - \varphi - \zeta(\varphi))/2 + \Xi(\zeta(\varphi))],
\]

where \( \bar{z} = \text{diag}(\bar{z}_x, \bar{z}_y) \) encodes the average lattice coordinate numbers in the horizontal and vertical direction, \( \bar{z} \equiv \text{Tr} \bar{z} \), \( \Xi(\varphi) \equiv \varphi \ln \varphi \), and \( \zeta(\varphi) \equiv 2\varphi(1 - \varphi)/(1 + [1 + 4(\exp(4\bar{J}) - 1)] \varphi(1 - \varphi))^{1/2} \). The gradient energy coefficient \( \kappa(\varphi) = \text{diag}(\kappa_x, \kappa_y) \) in Eq. (3) is given by

\[
\kappa(\varphi) = \frac{\bar{z} \exp(4\bar{J} - 1) \zeta(\varphi)}{4 \cdot 2\varphi(1 - \varphi) - \zeta(\varphi)}.
\]
analogies are obtained by taking the weak interaction limit
$$\lim_{J \to 0} \tilde{f}(\phi) = f_{\text{MF}}(\phi) + O(J^2)$$
where $f_{\text{MF}}(\phi)$ is given in [49], and
$$\lim_{J \to 0} \kappa(\phi) = \kappa_{\text{MF}} + O(J^2)$$
with $\kappa_{\text{MF}} = 2\tilde{J}$. Note that $\kappa_{\text{MF}}$ is independent of $\phi$, in agreement with
regular solution theory [1]. Notably, $2\zeta(\phi)$ is the thermodynamically optimal number of defects at given $\phi$, while
$$2\phi(1 - \phi) \geq 2\phi(\phi)$$
is the number of defects in a non-interacting system [49]. $\kappa_\phi$ as a function of $\phi$ is shown in
Fig. S4b for the BG and MF theory (blue and red lines, respectively) and highlights a large entropic penalty of
inhomogeneities at $\phi \to 0$ and $\phi \to 1$ (see [49]) that is
not accounted for in MF theory.

Note that Cahn and Hilliard [1] postulated the free energy density to have the form in Eq. (3), whereas here we
derived it from the Ising model. The equilibrium profile minimizes $F_{\text{CH}}$, i.e. it is the solution of $\partial F_{\text{CH}}/\partial \phi(x) = 0$.
We now show that BG and MF theories predict starkly different behavior for moderate and strong interactions—
MF theory fails to account for the interface broadening.

Equilibrium interface profile.—We first focus on the square lattice Ising strip in Fig. S3c ($L_y \gg L_x$) where
the magnetization varies only in the $x$ direction, i.e.
$$\phi(x) = \phi(x).$$
The profile is obtained as the solution of a nonlinear second order differential equation that we
solve numerically by imposing the boundary conditions
$$\phi(\pm L_x/2) = \phi_{\text{min}},$$
where $\phi_{\text{min}} \equiv \inf_{0<\phi<1/2} \tilde{f}(\phi)$ and
$$\phi_+ = 1 - \phi_{\text{min}}$$
denote the co-existing states with compositions set by the left and right minimum of $f(\phi)$, respectively.
We fix the interface location such that $\phi(0) = 1/2$ [72]. Above the critical coupling $J > J_{\text{crit}}$, we
fix $J_{\text{BG,crit}} \equiv \ln (\tilde{z}/(\tilde{z} - 2))/2$ and $J_{\text{MF,crit}} \equiv 1/\tilde{z}$ [71], $\tilde{f}(\phi)$
has two local minima resulting in a nonuniform $\phi(x)$. For
$J \leq J_{\text{crit}}$ the profile is uniform.

Qualitative differences between the profiles predicted by BG and MF theory are seen already in Fig. S3c-
d. In particular, BG concentration profiles depend non-
omonotonically on $J$, which is confirmed by Monte-Carlo (MC) simulations of the Ising model (for simulation de-
tails see [49]), whereas MF interfaces become monotonically steeper. By comparing with Fig. S3b we observe
that interface broadening correlates with interface de-
localization. This is further analyzed systematically in
Fig. S4.

First we inspect in Fig. S4c the interfacial steepness
$\phi'(0)$. In stark contrast to MF theory predicting a steep-
ening interface independent of box size, BG profiles are
non-monotonic in $J$ beyond a sufficient $L_x$ due to
interface delocalization. To verify that this is $no$ arti-
fact we compare our results with the solid-on-solid (SOS)
model for the square lattice Ising strip ($z = 4$) which
becomes exact in the limit $J \to \infty$ and is known to include
interface delocalization [61–65]. The SOS model yields
$$\lim_{J \to \infty} \phi(x) = 1/2 + x/L_x + \sin(2\pi x/L_x)/2\pi [62, 63, 65],$$
and is known to include interface delocalization [61–65].

In Fig. S4d we show the interfacial steepness as a function of $L_x$ for fixed $J$ and find that
the SOS and BG results display the same scaling (see Fig. S4d inset), whereas the MF result is in fact independent
of $L_x$. Further verification is given by the interface
stiffness $\Gamma = 2 \int_{\phi_{\text{min}}}^{\phi_{\text{max}}} |\phi'(x) - \tilde{f}(\phi_{\text{min}})|^{1/2} d\phi$ [1] de-
picted in Fig. S4e. The exact result is shown in the Ap-
pendix, while the SOS model yields $\Gamma_{\text{SOS}} = \cosh (2J) - 1$.
and converges to the exact result for large $\bar{J}$, i.e. $\lim_{j \to \infty} \ln(\Gamma_{\text{SOS}}) \approx 2\bar{J}$. Notably, the BG result is not only considerably more accurate than the MF prediction (see Fig. S4c) but also displays a correct exponential scaling, $\lim_{j \to \infty} \ln(\Gamma_{\text{BG}}) \approx \bar{J}$, in stark contrast to the square-root MF scaling, $\lim_{j \to \infty} \ln(\Gamma_{\text{MF}}) \approx \ln(\bar{J})/2$.

Disentangling interface delocalization.—By exploiting the mapping of instantaneous interface positions onto a Brownian excursion problem (see Appendix, Eq. (7)) we can disentangle interface delocalization from the instantaneous interface width, $\delta$, in the large $\bar{J}$ limit where $\lim_{J \to \infty} f_{\text{int}}(x;\bar{J}) = L^{-1}\mathbb{1}_{|x|<L/2}$ (see derivation in [49]). For $\bar{J} \gg 1$ each instantaneous profile $\varphi(x)$ corresponds to some continuous function $f(x/\delta + b_j)$: $\mathbb{R} \to [0,1]$ obeying $\lim_{x \to \pm \infty} f(x) = (1 \pm 1)/2$, where $\delta > 0$ and $b_j$ describe the width and position of the instantaneous interface $j$. The ensemble averaged profile thus reads $\lim_{x \to \pm \infty} \varphi(x) = L^{-1}\int f(x/\delta + b)\mathbb{1}_{|x|<L/2}db$. We can now straightforwardly compute the interface steepness and find $\lim_{x \to \pm \infty} \varphi'(0) = \delta^{-1}(f(L/2) - f(-L/2))/L$. Finally, taking the large-$L$ limit we obtain $\lim_{x \to \pm \infty} \varphi'(0)$ is $1/\delta$, and have thereby disentangled interface delocalization from the instantaneous interface width $\delta$. This result holds for any continuous integrable $f(x)$ obeying $\int_{-\infty}^{\infty} f(x)dx = (1 \pm 1)/2$. For the Ising strip this yields $\delta_{\text{SOS}} = 0.5$ within the SOS model and $\delta_{\text{BG}} \approx 0.835$ with the BG approximation (see Fig. S4f). Hence, while the ensemble averaged steepness vanishes in the large coupling limit due to interface delocalization, individual realizations maintain a nonzero interface steepness with uniformly distributed interface positions. Importantly, MF theory does not account for delocalization-induced interface broadening and therefore predicts $\delta_{\text{MF}} \to 0$ (see inset of Fig. S4f).

Spinodal decomposition.—Having established the physical consistency of Eqs. (3-S65) we now address phase separation, and determine the length scales on which inhomogeneities are stable by performing a linear stability analysis on the total free energy density around the uniform concentration profile, $\varphi(x) = 1/2 + \alpha \sin(\pi x)$ with $|\alpha| \ll 1/2$ (the symmetry of the problem imposes odd inhomogeneities). Stable perturbations lower the total free energy density, $\delta F \equiv \bar{F}[\varphi(x)] - \bar{F}[1/2] \leq 0$, yielding an upper bound on stable wavevectors $q \leq q_{\text{crit}} \equiv [\tilde{\Gamma}''(1/2)/\kappa_x(1/2)]^{1/2}$ (see derivation in [49]), where $\tilde{\Gamma}''(\varphi) = \partial^2 \tilde{\Gamma}(\varphi)/\partial \varphi^2$ is the curvature of the free energy barrier. $q_{\text{crit}}$ translates into a critical wavelength $\lambda_{\text{crit}} = 2\pi/q_{\text{crit}}$ above which perturbations are stable. Fig. S4g depicts $\lambda_{\text{crit}}$ as a function of $\bar{J}$ for a square lattice. Similar to Fig. S4c, $\lambda_{\text{crit}}$ displays a non-monotonic trend in the BG theory (blue lines) [73] that is contrasted by a monotonic attenuation in the MF theory (red lines). The interaction strength minimizing $\lambda_{\text{crit}}$ in the BG theory, i.e. $\bar{J}$ allowing for the widest range of stable wavelengths (arrow in Fig. S4g), can be determined exactly and reads

$$J^! = \frac{1}{2} \ln \frac{z + 2\sqrt{z - 1}}{z - 2},$$

with the corresponding $\lambda_{\text{BG}}(J^!)$ given in [74]. The non-monotonicity of $\lambda_{\text{crit}}$ is explained by inspecting how the curvature of the barrier depends on $\bar{J}$. In particular, the BG curvature converges, $\lim_{j \to \infty} \tilde{\Gamma}''(1/2) = 2(2 - z)$ (see Fig. S4g, inset I), whereas the free energy penalty of inhomogeneities increases exponentially, eventually increasing $\lambda_{\text{BG}}$. MF theory overestimates the curvature of the barrier and underestimates the free energy penalty of inhomogeneities, leading to a decreasing $\lambda_{\text{MF}}$.

Implications for nucleation.—We next investigate, in Fig. 3, how interface broadening affects nucleation by determining minimal free energy paths (the reaction coordinate and method are described in Sec. IX of [49]). The inset in Fig. 3a suggests that critical nuclei become less dense and wider as $\bar{J}$ becomes larger. Indeed, we find that correlations captured by BG theory lead to larger critical nuclei (Fig. 3b), shallower interfaces (Fig. 3c), and that the increasing trend with $\bar{J}$ is only captured by BG theory, which is reminiscent of the results shown in Fig. S4. Most importantly, BG theory predicts that the nucleation barrier $\Delta E$ is approximately four times larger than predicted by MF (Fig. 3a), implying a strong reduction of nucleation rates [75-78].

![Figure 3](image-url)

**FIG. 3.** Numerical simulations of critical nuclei of the radially symmetric Cahn-Hilliard equation with the BG (blue) and MF (red) free energy for a hexagonal coordination ($z = 6, \bar{z} = 4$). (a) Free energy difference $\Delta E$ between the critical nucleus and the homogeneous state as a function of the interaction strength $\bar{J}$. The inset shows critical profiles $\varphi(r)$ for three values of $\bar{J}$. (b) Radius $r_{\text{crit}}$ and (c) interfacial steepness $\varphi' r_{\text{crit}}$ of the critical nucleus as a function of $\bar{J}$. To understand why interface delocalization affects nucleation we note that shifting the interface position corresponds to a growing/shrinking nucleus which alters the free energy. Instantaneous interfaces are still affected...
by interface translation and capillary-wave fluctuations. However, in contrast to the strip, distinct instantaneous interface configurations are not iso-energetic. The weighting by the respective free energy of the configuration ultimately gives rise to broadening and thus larger critical nuclei and higher nucleation barriers.

Conclusion.—By directly computing the thermodynamic limit of a spatially inhomogeneous Ising model on general lattices within the Bethe-Guggenheim approximation we derived a Cahn-Hilliard theory that accounts for nearest-neighbor pair-correlations. Strong interactions were shown to give rise to a delocalization-induced interface broadening confirmed by exact results for the two-dimensional Ising model, a strong reduction of nucleation kinetics due to an amplification of the free energy barrier to nucleation, and pair correlations are required to correctly account for them. Pair correlations enforce a thermodynamically optimal configuration of defects and are thus an essential determinant of interfaces and condensates in the strong interaction limit that so far have been overlooked. Our results allow for generalizations to three dimensions and more than two constituents, which will be addressed in forthcoming publications.

Appendix: Statistics of instantaneous interface positions.—Neglecting overhangs, one can map the statistics of instantaneous interfaces onto a confined Brownian bridge problem [66]. The idea is to treat the respective bulk phases as “pure” (i.e. homogeneous) and the interface (i.e. domain wall) as a random walk, which in the continuum limit becomes a Brownian motion with diffusion coefficient $1/4\Gamma$ with the interface stiffness $\Gamma = \sinh(2J + \ln\tanh J)$ and the vertical coordinate $y$ plays the role of time [62]. Periodic boundary conditions in the $y$-direction render the Brownian trajectories Brownian bridges. Accordingly, the probability density of interface positions reads (see derivation in [49])

$$p_{\text{int}}(x; J) = \frac{1}{L_x} \frac{\psi_3(0, e^{-\alpha_J}) - \psi_3(\pi x/L_x, e^{-\alpha_J})}{\psi_3(0, e^{-\alpha_J}) - 1},$$

(7)

where $\alpha_J \equiv \pi^2 L_y^2/4\Gamma L_x^2$ and $\psi_3(a, x)$ is Jacobi's elliptic theta of the third kind. As long as $\tilde{J} \ll 1$ we have $\tilde{\Gamma} \ll 1$ and thus $\alpha_J \gg 1$. Accordingly, Eq. (7) predicts instantaneous interfaces to be localized with a probability density $p_{\text{int}}(x; \tilde{J}) \propto \sin(\pi x/L_x)^2$ (see Fig. S3b, top). Conversely, for sufficiently large $\tilde{J}$ we find $\tilde{\Gamma} \gg 1$ and hence $\alpha_J \ll 1$, and the interface positions become delocalized (see Fig. S3b, center and bottom). In the limit $\tilde{J} \rightarrow \infty$ we find (see [49]) a uniform density $p_{\text{int}}(x; \tilde{J}) \rightarrow L_x^{-1} \chi[x|<L_x]/2$. Notably, when $\tilde{J} \rightarrow \infty$ a Casimir effect appears in addition (see e.g. [79, 80]) that is not captured in Eq. (7), i.e. the entropy due to bulk fluctuations is enhanced near the boundaries giving rise to “peaks” (see Fig. S3b, bottom).

Acknowledgments.—The financial support from the German Research Foundation (DFG) through the Emmy Noether Program GO 2762/1-2 (to AG) and Project ZW 222/3-1 (to DZ), and the Max Planck Society (to DZ and KB), in case of KB in the form of an IMPRS fellowship are gratefully acknowledged.

[1] J. W. Cahn and J. E. Hilliard, J. Chem. Phys. 28, 258 (1958).
[2] J. W. Cahn, J. Chem. Phys. 30, 1121 (1959).
[3] J. W. Cahn and J. E. Hilliard, J. Chem. Phys. 31, 688 (1959).
[4] C. A. Weber, D. Zwicker, F. Jülicher, and C. F. Lee, Rep. Prog. Phys. 82, 064601 (2019).
[5] J. S. Rowlinson and B. Widom, Molecular theory of capillarity (Courier Corporation, 2013).
[6] A. J. Bray, Adv. Phys. 51, 481 (2002).
[7] P. G. de Gennes, J. Chem. Phys. 72, 4756 (1980).
[8] P. Pincus, J. Chem. Phys. 75, 1996 (1981).
[9] K. Binder, J. Chem. Phys. 79, 6387 (1983).
[10] Q. Tang and M. Müller, Phys. Rev. Lett. 126, 028003 (2021).
[11] J. Kirschbaum and D. Zwicker, J. R. Soc. Interface 18, 20210255 (2021).
[12] S. Bo, L. Hubatsch, J. Bauermann, C. A. Weber, and F. Jülicher, Phys. Rev. Research 3, 043150 (2021).
[13] S. Mao, M. S. Chakraverti-Wuerthein, H. Gaudio, and A. Košmrlj, Phys. Rev. Lett. 125, 218003 (2020).
[14] G. Aubert, P. Kornprobst, and G. Aubert, Mathematical problems in image processing: partial differential equations and the calculus of variations, Vol. 147 (Springer, 2006).
[15] M. I. M. Copetti and C. M. Elliott, Numerische Mathematik 63, 39 (1992).
[16] H. Gómez, V. M. Calo, Y. Bazilevs, and T. J. Hughes, Comput. Methods Appl. Mech. Eng. 197, 4333 (2008).
[17] A. W. Adamson, A. P. Gast, et al., Physical chemistry of surfaces, Vol. 150 (Interscience publishers New York, 1967).
[18] Y. Zhao, P. Stein, and B.-X. Xu, Comput. Methods Appl. Mech. Eng. 297, 325 (2015).
[19] Y. Zhao, P. Stein, Y. Bai, M. Al-Siraj, Y. Yang, and B.-X. Xu, J. Power Sources 413, 259 (2019).
[20] A. J. M. Yang, P. D. Fleming, and J. H. Gibbs, J. Chem. Phys. 64, 3732 (1976).
[21] J. Erlebacher, M. J. Aziz, A. Karma, N. Dimitrov, and K. Sieradzki, Nature 410, 450 (2001).
[22] L.-Q. Chen, Ann. Rev. Mater. Sci. 32, 113 (2002).
[23] I. Steinbach, Model. Simul. Mat. Sci. Eng. 17, 073001 (2009).
[24] J. D. Murray, Mathematical Biology I. An Introduction (Springer, 2002).
[25] D. W. Sanders, N. Kedersha, D. S. Lee, A. R. Strom,
W. Hur, J. P. Kemp Jr, M. Tarzia, V. E. Deneke, W. F. Marzuff, R. J. Durono, and S. Di Talia, Dev. Cell 54, 379 (2020).

W. K. Spoelstra, J. M. Jacques, R. Gonzalez-Linares, F. L. Nobrega, A. C. Haagsma, M. Dogterom, D. H. Miejer, T. Idema, S. J. Brouns, and L. Reese, Biophys. J. 120, 1198 (2021).

J. Rogiers and J. Indekeu, EPL (Europhysics Letters) 23, 8 (2016).

V. Drake, J. A. Riback, D. Bracha, J. M. Eefteurs, A. Iwanicki, A. Wang, et al., Cell 181, 306 (2020).

W. L. Bragg and E. J. Williams, Proc. Math. Phys. Eng. Sci. 145, 699 (1934).

S. A. Safran, Statistical thermodynamics of surfaces, interfaces, and membranes (CRC Press, 1998).

J. Stenhammar, A. Tiribocchi, R. J. Allen, D. Marenduzzo, and M. E. Cates, Phys. Rev. Lett. 111, 145702 (2013).

J. Meibohm and M. Esposito, Phys. Rev. Lett. 128, 270401 (2022).

K. Blom and A. Godec, Phys. Rev. X 11, 021029 (2021).

K. Binder, D. P. Landau, and A. M. Ferrenberg, Phys. Rev. Lett. 74, 298 (1995).

K. Binder, M. Müller, F. Schmid, and A. Werner, J. Stat. Phys. 95, 1045 (1999).

K. Binder, D. P. Landau, and A. M. Ferrenberg, Phys. Rev. E 53, 5023 (1996).

A. Ciach, Phys. Rev. B 34, 1932 (1986).

J. Stecki, Phys. Rev. B 47, 7519 (1993).

J. Stecki, A. Maciolek, and K. Olaussen, Phys. Rev. B 49, 1092 (1994).

A. Ciach and J. Stecki, J. Phys. A Math. Theor. 20, 5619 (1987).

E. Albano, K. Binder, and W. Paul, J. Phys. Condens. Matter 12, 2701 (2000).

M. E. Fisher, J. Stat. Phys. 34, 667 (1984).

M. E. Fisher, J. Chem. Soc., Faraday trans. II 82, 1569 (1986).

F. Schmitz, P. Virnau, and K. Binder, Phys. Rev. E 90, 022128 (2014).

R. Lipowsky, D. M. Kroll, and R. K. P. Zia, Phys. Rev. B 27, 4499 (1983).

In the canonical ensemble one is required to introduce an additional constrain inside the sum over $\sigma$ to fix the total number of up and down spins.

K. Blom and A. Godec, Phys. Rev. X 11, 031067 (2021).

We actually fix the ensemble averaged interface position, which is not equal to fixing the position of instantaneous profiles. Thus, the interface location along individual trajectories may still fluctuate.

The non-monotonic dependence of $\lambda_{BG}^*$ on $J$ in fact persist for any background concentration $0 < \varphi_0 < 1$; see [49].

$\lambda_{BG}^*(\bar{J})$ is independent of the uniform background concentration $0 < \varphi_0 < 1$; see [49].

H. Kramers, Physica 7, 284 (1940).

J. Langer, Ann. Phys. 54, 258 (1969).

P. Hänggi, P. Talkner, and M. Borkovec, Rev. Mod. Phys. 62, 251 (1990).

D. Hartich and A. Godec, New J. Phys. 20, 112002 (2018).

A. Mukhopadhyay and B. M. Law, Phys. Rev. E 62, 5200 (2000).

D. M. Dantchev and S. Dietrich, (2022), arXiv:2203.15050.

The vertical dimension in [6] is defined from $y = [-L_y, L_y]$, hence in our terminology there is an additional factor of $1/12$ in the MSD.

The width in [6] is indicated with $w^2$. 

See Supplemental Material at [...] for detailed derivations and auxiliary results.
Supplemental Material for:
Delocalization-Induced Interface Broadening in Strongly Interacting Systems
Kristian Blom\textsuperscript{1,} Noah Ziethen\textsuperscript{2,} David Zwicker\textsuperscript{2} and Aljaž Godeč\textsuperscript{1,\textsuperscript{*}}
\textsuperscript{1}Mathematical bioPhysics Group, Max Planck Institute for Biophysical Chemistry, 37077 Göttingen, Germany
\textsuperscript{2}Theory of Biological Fluids, Max Planck Institute for Dynamics and Self-Organization, 37077 Göttingen, Germany
(\textsuperscript{*}) agodec@mpinat.mpg.de

The sections in this Supplementary Material (SM) are organized in the order they appear in the main Letter. First we present, in Sec. I, a detailed description of Monte-Carlo simulations that are shown in Fig. 1 in the Letter. In Sec. II we proceed with a derivation of the probability density of instantaneous interface positions based on the mapping onto the Brownian bridge problem (the results are briefly shown in Fig. 1b and the mapping explained in the Appendix in the Letter). Sections III-V are devoted to the derivation of the Cahn-Hilliard theory starting from a two-dimensional Ising model using the mean field (Sec. IV) and Bethe-Guggenheim (Sec. V) approximations, respectively. Next, in Sections VI-VII we analyze the field theories by determining the one-dimensional equilibrium concentration profile, interfacial steepness, interfacial width, and critical wavelength of stable perturbations. In Sec. VIII we probe the accuracy of both approximations by comparing them with exact results for system sizes which are amendable to exact solutions. Finally, in Sec. IX we present details on the numerical simulations of nucleation by means of the radially symmetric Cahn-Hilliard equation which are shown in Fig. 3 in the Letter.

CONTENTS

References 5

I. Monte-Carlo simulations of the Ising model 2
   A. Lattice setup and initial configuration 2
   B. Acceptance rate 2
   C. Simulation parameters 3
   D. Equilibration test: Energy fluctuations per spin 3
   E. Benchmark test: Interfacial width and roughening 4
      1. Ensemble averaged concentration profile and the boundary-shift method 4
      2. Interfacial width and weighted linear regression 5

II. Analytical result for the distribution of instantaneous interface position 5
   A. Derivation 5
   B. Convergence to the uniform distribution 6

III. Definitions and initial setup 7
    A. Lattice specification and thermodynamic limit 7
    B. Rewriting the Ising Hamiltonian 7
    C. Coarse-grained lattice observables 7
    D. Hamiltonian and partition function 8

IV. Mean field approximation 9
    A. Approximation of the fraction of defects 9
    B. Optimization w.r.t. $\varphi_{ij} \rightarrow \lim_{N_s} [\cdot]$ 10
    C. $\lim_{N_s} [\cdot] \rightarrow$ optimization w.r.t. $\varphi(x,y)$ 10

V. Bethe-Guggenheim approximation 11
   A. Introduction 11
   B. Pair-distribution Ansatz for the degeneracy of states in a non-uniform system 11
   C. Evaluation of the normalization constant 12
   D. Evaluation of the partition sum 12
   E. Optimization w.r.t. $\varphi_{ij} \rightarrow \lim_{N_s} [\cdot]$ 13
   F. $\lim_{N_s} [\cdot] \rightarrow$ optimization w.r.t. $\varphi(x,y)$ 14

VI. Equilibrium concentration profile 16
I. MONTE-CARLO SIMULATIONS OF THE ISING MODEL

Here we provide details on Monte-Carlo (MC) simulations we used to determine the ensemble averaged concentration profile and histograms of instantaneous interface locations displayed in Fig. 1 in the Letter.

A. Lattice setup and initial configuration

We performed MC simulations of the nearest-neighbor interacting ferromagnetic Ising model on the square lattice with size \( (N_x^y = 40) \times (N_y^z \in \{80, 90, 100, 110, 120, 130\}) \) with single spin-flip dynamics in the bulk and two-spin-exchange dynamics at the boundary columns located at \( i = \pm N_x^y/2 \). We considered various values of \( N_y^z \) to benchmark our simulations against known theoretical predictions of the scaling behavior (see Sec. I E). We imposed periodic boundary conditions in the vertical direction (i.e. along the columns) and free boundary conditions in the horizontal direction (i.e. along the rows) whereby we constrained the total magnetization on the left/right boundary (see below). Let \( N_i^x \) with \( i \in \{-N_x^y/2, \ldots, N_x^y/2\} \) denote the number of down spins in column \( i \). To induce a non-uniform concentration profile and in anticipation of known exact results for the bulk concentration values \([1]\) we fixed the number of down spins at the boundaries to be

\[
N_{\pm N_x^y/2}^x = \frac{N_y^z}{2} \left(1 \pm \text{Re}([-\sinh^{-4}(2\tilde{J})]^{1/8})\right),
\]

where \( \tilde{J} \) is the coupling strength. Spins located at the boundaries can exchange only within the same column, and therefore the total number of up/down spins at the boundaries is conserved throughout the simulation. Spins in the bulk are initially prepared in a high-coupling configuration (i.e. aligned) with a vertical interface placed at some random horizontal location in the lattice. Starting from a high-coupling configuration has the advantage that the simulations do not get stuck in frozen sub-optimal states where multiple interfaces are created \([2, 3]\).

B. Acceptance rate

For single spin-flip dynamics let \( \{\sigma_j\}_i^j \) denote the spin configuration obtained by flipping spin \( i \) while keeping the configuration of all other spins fixed, i.e., \( \{\sigma_j\}_i^j \equiv (-\sigma_i, \{\sigma_{j \neq i}\}) \). Moreover, let \( p_i(\{\sigma_j\}_i^j) \) denote the acceptance rate from \( \{\sigma_j\} \) to \( \{\sigma_j\}_i^j \) and \( \Delta \mathcal{H}_i(\{\sigma_j\}) \equiv \mathcal{H}(\{\sigma_j\}_i^j) - \mathcal{H}(\{\sigma_j\}) \) the energy difference associated with the transition. Using the Metropolis algorithm the acceptance rate for the single spin-flip takes the form \([4]\)

\[
p_i(\{\sigma_j\}_i^j) = \min(1, e^{-\Delta \mathcal{H}_i(\{\sigma_j\})}). \tag{S2}
\]

For two-spin-exchange dynamics let \( \{\sigma_j\}_{ik} \) denote the spin configuration upon interchanging the spins \( \sigma_i \) and \( \sigma_k \) while keeping the configuration of all other spins fixed, i.e., \( \{\sigma_j\}_{ik} \equiv (\sigma_i \leftrightarrow \sigma_k, \{\sigma_{j \neq i,k}\}) \). We denote with \( p_{ik}(\{\sigma_j\}) \) the acceptance rate from \( \{\sigma_j\} \) to \( \{\sigma_j\}_{ik} \) and \( \Delta \mathcal{H}_{ik}(\{\sigma_j\}) \equiv \mathcal{H}(\{\sigma_j\}_{ik}) - \mathcal{H}(\{\sigma_j\}) \) denotes the energy difference associated with the transition. Using the Metropolis algorithm the two-spin-exchange acceptance rate reads

\[
p_{ik}(\{\sigma_j\}) = \min(1, e^{-\Delta \mathcal{H}_{ik}(\{\sigma_j\})}). \tag{S3}
\]
FIG. S1. Equilibration test: Energy fluctuations per spin as a function of consecutively stored Monte-Carlo (MC) configurations (see text). In each plot we display the energy fluctuations per spin \((E - E_0)/N\) where \(E_0\) is the ground state energy conditioned on anti-symmetric boundary conditions and \(N = N_x^\sigma N_y^\sigma\) with \(N_x^\sigma = 40\) for a subset of \(10^4\) MC simulations (colored lines). The black solid line indicates the ensemble average energy fluctuation per spin. Plots in the same column have equal \(N_y^\sigma \in \{80, 100, 120, 130\}\), and plots in the same row have equal \(\tilde{J} \in \{0.45, 0.6, 0.95, 1.2\}\).

C. Simulation parameters

For each value of the coupling strength \(\tilde{J}\) and vertical length \(N_y^\sigma \in \{80, 90, 100, 110, 120, 130\}\) we performed \(N_{\text{MC}} = 10^5\) MC simulations, where each individual run contained \(5 \times 10^8\) MC steps. At each \(1.9 \times 10^7\)th MC step we took a snapshot of the configuration and stored the total energy, resulting in 26 (including the initial configuration) snapshots for each simulation run.

D. Equilibration test: Energy fluctuations per spin

To assess whether the MC simulations reached equilibrium we analyzed the energy fluctuations per spin and their corresponding ensemble average. In Fig. S1 we display the energy fluctuations per spin for a subset of \(10^4\) simulations as a function of the MC steps (MCS) for various \(\tilde{J} \in \{0.45, 0.7, 0.95, 1.2\}\) and \(N_y^\sigma \in \{80, 100, 120, 130\}\). In each plot we observe that immediately after the initial snapshot the energy is fluctuating around an average steady state denoted with the black solid line, providing a first indication that the simulations have reached equilibrium (already at the
FIG. S2. Benchmark test: Results in the bottom and top row are derived with and without applying the boundary-shift method, respectively. (a)-(d) Scaling of the interfacial width \( w^2(N_y, \tilde{J}) \) (no boundary shift) and (d) \( \hat{w}^2(N_y, \tilde{J}) \) (boundary shift) w.r.t. the vertical number of spins \( N_y \). Each point is obtained by averaging over \( 2.5 \times 10^6 \) equilibrated configurations. Dashed lines are obtained by weighted linear regression. Colors from light green to dark green correspond to increasing coupling strength \( \tilde{J} \). (b)-(e) Intersection point of the interfacial width at \( N_y^\sigma = 0 \) as a function of \( \tilde{J} \). The standard deviation of each point is estimated with the Jackknife method. In (c) the red and blue lines are the theoretical predictions for the intersection point given in [5, 6], respectively. (c)-(f) Slope of the interfacial width w.r.t. \( N_y^\sigma \) as a function of \( \tilde{J} \). The standard deviation of each point is estimated with the Jackknife method. In (f) the red and blue lines are the theoretical predictions for the slope given in [5, 6], respectively.

first stored configuration). Note that in each plot all energies are initially increasing from zero since we subtract the ground state energy and we initialize the system in a high-coupling configuration which is identical to the ground state.

E. Benchmark test: Interfacial width and roughening

To benchmark the performance of our MC simulations we computed the interfacial width \( w^2(N_y^\sigma, \tilde{J}) \) and compared our results with known theoretical results reported in [5, 6]. The results from [6] predict \( w^2(N_y^\sigma, \tilde{J}) \propto N_y^\sigma/\sinh(\sigma) \) with \( \sigma = 2\tilde{J} + \ln \tanh \tilde{J} \). Analogously, the results from [5] predict \( w^2(N_y^\sigma, \tilde{J}) = N_y^\sigma/12\sigma - c/2\pi\sigma^2 \) with \( c \approx 1 \). Below we explain in detail how we determined the interfacial width and how it compares to the theoretical predictions. The resulting outcomes are shown in Fig. S2 and the comparison with the theoretical results are shown in Fig. S2(e)-(f).

1. Ensemble averaged concentration profile and the boundary-shift method

To compare our results with [5, 6] we need to apply the so-called boundary shift method (see also [7]) where we shift the interface position of each instantaneous concentration profile to the center of the lattice. As a scientific exercise we also consider the resulting outcomes without applying the boundary shift method for which the results are depicted in the top row of Fig. S2. Let \( \hat{\phi}_{i,k} \) be the equilibrated and boundary shifted concentration of down spins in column \( i \in \{-N_y^\sigma/2, ..., N_y^\sigma/2\} \) of the \( k \)th MC simulation run. We define the ensemble average boundary-shifted concentration profile as

\[
\langle \hat{\phi}_i \rangle = \frac{1}{N_{MC}} \sum_{k=1}^{N_{MC}} \hat{\phi}_{i,k}.
\]
From Eq. (S4) we can approximate the mean interfacial width using the central difference method as follows

$$\hat{w}^2(N_x^y, J) = \frac{\sum_{i=-N_x^y/2}^{N_x^y/2-1} \hat{\varphi}_{i+1}^2 - \langle \hat{\varphi}_{i+1} \rangle}{\sum_{i=-N_x^y/2+1}^{N_x^y/2-1} \langle \hat{\varphi}_{i+1} \rangle - \langle \hat{\varphi}_{i+1} \rangle} - \left( \frac{\sum_{i=-N_x^y/2}^{N_x^y/2-1} \hat{\varphi}_{i+1}^2 - \langle \hat{\varphi}_{i+1} \rangle}{\sum_{i=-N_x^y/2+1}^{N_x^y/2-1} \langle \hat{\varphi}_{i+1} \rangle - \langle \hat{\varphi}_{i+1} \rangle} \right)^2. \quad (S5)$$

A similar definition holds for the interfacial width without applying the boundary-shift method which we denote as $w^2(N_x^y, J)$. In Fig. 2(a)-(d) we plot $w^2(N_x^y, J)$ and $\hat{w}^2(N_x^y, J)$ with the green dots as a function of $N_x^y$. Both results show a clear linear trend with $N_x^y$, providing a first validation of the MC simulations. To obtain the variance of $w^2(N_x^y, J)$ – which we use in the next section for weighted linear regression – we used the Jackknife method which is explained below.

### 2. Interfacial width and weighted linear regression

To compare our results with those reported in [5, 6] we need to extract the interception point $\hat{w}^2(J, 0)$ and slope $d\hat{w}^2(J, N_x^y)/dN_x^y$. To obtain both quantities we use weighted linear regression in combination with the Jackknife method. First we determine $\hat{w}^2(0, J)$ and slope $d\hat{w}^2(N_x^y, J)/dN_x^y$ for fixed $J$ while removing one point from the data pool which gives

$$\{\hat{w}^2_j(0, J), \frac{d\hat{w}^2_j(N_x^y, J)}{dN_x^y}\} = \min_{(\alpha, \beta)} \sum_{N_x^y=80, \ldots, 130, \ N_x^y \neq 70+10x} \frac{(\alpha + \beta N_x^y - \hat{w}^2(N_x^y, J))^2}{\text{var}(\hat{w}^2(N_x^y, J))}, \quad (S6)$$

where $j = \{1, \ldots, 6\}$. A similar definition holds for the intersection point and slope without applying the boundary shift method, which we denote as $w^2_j(0, J)$ and $dw^2_j(N_x^y, J)/dN_x^y$, respectively. Finally the Jackknife ensemble averages and variances are given by

$$\hat{w}^2(0, J) = \frac{1}{6} \sum_{j=1}^{6} \hat{w}^2_j(0, J), \quad \text{var}(\hat{w}^2(0, J)) = \frac{5}{6} \sum_{j=1}^{6} (\hat{w}^2_j(0, J) - \hat{w}^2(0, J))^2,$$

$$d\hat{w}^2(N_x^y, J) = \frac{1}{6} \sum_{j=1}^{6} \frac{d\hat{w}^2_j(N_x^y, J)}{dN_x^y}, \quad \text{var} \left( \frac{d\hat{w}^2(N_x^y, J)}{dN_x^y} \right) = \frac{5}{6} \sum_{j=1}^{6} \left( \frac{d\hat{w}^2_j(N_x^y, J)}{dN_x^y} - \frac{d\hat{w}^2(N_x^y, J)}{dN_x^y} \right)^2. \quad (S7)$$

In Fig. S2(e)-(f) we plot $\hat{w}^2(0, J)$ and $d\hat{w}^2(N_x^y, J)/dN_x^y$ together with the standard deviation as a function of $J$. The theoretical results given by [5, 6] are shown with the red and blue lines, respectively. For $J \geq 0.6$ we find a very good agreement between MC simulations and theoretical predictions. Notably, for the slope in Fig. S2(f) we find a remarkable agreement with the results of [6]. For $J < 0.6$ we approach the critical coupling $J_{\text{crit}} \approx 0.441$, where the MC results agree less well with theoretical predictions due to finite-size effects. This is expected since the correlation length diverges around the critical coupling.

### II. ANALYTICAL RESULT FOR THE DISTRIBUTION OF INSTANTANEOUS INTERFACE POSITION

Here we derive the probability density of positions of instantaneous interfaces given by Eq. (7) in the Appendix of the Letter, and we furthermore show the convergence to a uniform distribution in the limit $J \to \infty$.

#### A. Derivation

Let us assume that the instantaneous interface in the two-dimensional Ising strip always separates two homogeneous thermodynamically co-existing phases and that the domain wall behaves like a Gaussian polymer confined between hard walls located at $x = 0$ and $x = L_x$ with a height $L_y \gg L_x$. We parameterize the domain wall as a Brownian motion $\{x_y^J\}_{0 \leq y \leq L_y}$ where $y$ plays the role of time (or contour length in the polymer context). Then the Green’s function of the interface then follows the Edwards equation with absorbing boundary conditions at the walls

$$\partial_y G(x, y|x_0) = D \nabla_y^2 G(x, y|x_0), \quad G(x, 0|x_0) = \delta(x - x_0), \quad G(0, y|x_0) = G(L_x, y|x_0) = 0 \forall y \in [0, L_y]. \quad (S8)$$
where $D$ is the diffusion coefficient and whose general solution is

$$G(x, y|x_0) = \frac{2}{L_x} \sum_{k=1}^{\infty} \sin\left(k\pi x/L_x\right) \sin\left(k\pi x_0/L_x\right)e^{-\pi^2 k^2 D y/L_x^2}. \tag{S9}$$

Particularly interesting is the mean squared displacement (MSD) which is given by

$$\langle (x - x_0)^2 \rangle = \frac{1}{L_x} \int_0^{L_x} dx \int_0^{L_x} dx_0 (x - x_0)^2 G(x, y|x_0) = \frac{4 L_x^2}{\pi^4} \sum_{k=1}^{\infty} \left(-1\right)^{k+1} \left(k^2 \pi^2 - 4\right) \frac{4}{k^4} e^{-\pi^2 k^2 D y/L_x^2}. \tag{S10}$$

For $y \ll D/L_x^2$ we can truncate the exponential inside the sum up to first order in $y$ and obtain $\langle (x - x_0)^2 \rangle \approx 2D y$ as expected for a freely diffusing one-dimensional particle. Now we recall the exact results of Abraham [6] and Fisher [8] who found that for the two-dimensional Ising strip the interface width should scale as $D[81][82] \langle (x - x_0)^2 \rangle \propto y/2\Gamma$ where the proportionality factor includes some lattice length scale and $\Gamma = \sinh \left(2\bar{J} + \ln \tanh \bar{J}\right)$ is the surface stiffness as defined in the Letter. The outcome of [6, 8] allows us to relate the diffusion coefficient $D$ to the surface stiffness

$$D \propto \frac{1}{4 \sinh \left(2\bar{J} + \ln \tanh \bar{J}\right)} \tag{S11}.$$

Under periodic boundary conditions in the $y$-direction the interfaces have an equal position at $y = 0$ and $y = L_y$ (also known as Brownian bridges). In this case the propagator is simply given by $G(x, L_y|x)$. We can now calculate the probability density to have an interface located at position $x$, which upon normalization is given by

$$p_{\text{int}}(x; \bar{J}) = \frac{G(x, L_y|x)}{\int_0^{L_x} G(x, L_y|x) dx} = \frac{1}{L_x} \frac{\vartheta_3(0, e^{-\alpha_J}) - \vartheta_3(\pi x/L_x, e^{-\alpha_J})}{\vartheta_3(0, e^{-\alpha_J}) - 1}, \tag{S12}$$

where $\alpha_J = \pi^2 D L_y/L_x^2$ and $\vartheta_3(a, x)$ is Jacobi’s elliptic theta of the third kind. The second equality can be obtained directly from Eq. (S9) with the definition of Jacobi’s elliptic theta of the third kind. Plugging Eq. (S11) for the diffusion coefficient into Eq. (S12) we obtain Eq. (7) in the Appendix of the Letter, which allows us to analytically compute the distribution of instantaneous interface positions. For any value of the interaction strength as done in Fig. 1b.

### B. Convergence to the uniform distribution

Next we prove that Eq. (S12) converges to the uniform distribution in the limit $\bar{J} \to \infty$. We first introduce $q \equiv e^{-\alpha_J}$ and rewrite

$$\vartheta_3(\pi x/L_x, e^{-\alpha_J}) = 1 + 2 \sum_{n=1}^{\infty} e^{-n^2 \alpha_J} \cos(2\pi n x/L_x) = \sum_{n=-\infty}^{\infty} q^n \left(e^{i 2\pi x/L_x}\right)^n, \tag{S13}$$

Since the term in the brackets is independent of $\bar{J}$, $\alpha_J \geq 0$ and because $\lim_{\bar{J} \to \infty} \alpha_J = 0$ (uniformly), saying $\lim_{\bar{J} \to \infty} e^{-\alpha_J}$ is equivalent to $\lim_{q \to 1} q$. We now use the asymptotic result for $q \uparrow 1$ [9]

$$\sum_{n=-\infty}^{\infty} q^n \left(e^{i 2\pi x/L_x}\right)^n \approx \sqrt{\frac{\pi}{-\ln q}} \exp\left(\frac{\pi^2 x^2}{L_x^2 \ln q}\right), \tag{S14}$$

where $\approx$ stands for asymptotic equality, i.e. $A \approx B$ stands for $A/B \to 1$. Note that Eq. (S14)$\gg 1$ since $0 < x < L_x$. Let us now rewrite Eq. (S12) as

$$p_{\text{int}}(x; \bar{J}) = L_x^{-1} \frac{\vartheta_3(0, e^{-\alpha_J}) - \vartheta_3(\pi x/L_x, e^{-\alpha_J}) + 1}{\vartheta_3(0, e^{-\alpha_J}) - 1} = L_x^{-1} - \frac{\vartheta_3(\pi x/L_x, e^{-\alpha_J}) - 1}{\vartheta_3(0, e^{-\alpha_J}) - 1} \tag{S15}$$

We can now evaluate the limit of Eq. (S15) using Eq. (S14) to find

$$\lim_{\bar{J} \to \infty} p_{\text{int}}(x; \bar{J}) \approx L_x^{-1} - L_x^{-1} \lim_{q \to 1} \exp\left(\frac{\pi^2 x^2}{L_x^2 \ln q}\right) = L_x^{-1} - L_x^{-1} \lim_{\alpha_J \to 0} \exp\left(-\frac{\pi^2 x^2}{L_x^2 \alpha_J}\right) = L_x^{-1} \tag{S16}$$

for $0 < x < L_x$, while we have $p_{\text{int}}(0; \bar{J}) = p_{\text{int}}(L_x; \bar{J}) = 0, \forall \bar{J}$. Applying a translational shift $x \to x - L_x/2$ yields the resulting uniform distribution mentioned in the Letter.
III. DEFINITIONS AND INITIAL SETUP

This section describes the initial setup for the derivation of the field theories based on the mean field (MF) and Bethe-Guggenheim (BG) approximation in Sec. IV and Sec. V, respectively.

A. Lattice specification and thermodynamic limit

We consider $N^x_\sigma \times N^y_\sigma$ classical spins $\sigma_{ij} = \pm 1$ with $(i,j) \in \{(1, \ldots, N^x_\sigma), \{1, \ldots, N^y_\sigma\}$ arranged on a two-dimensional lattice with a given topology and horizontal and vertical length ($L_x, L_y$). The lattice spacings $\delta_x$ and $\delta_y$ in the horizontal and vertical direction, respectively, are $\delta_x = L_x/N^x_\sigma$ and $\delta_y = L_y/N^y_\sigma$. Let $\mathbf{b}_{ij}$ with $(i,j) \in \{(1, \ldots, N^x_b), \{1, \ldots, N^y_b\}$ denote a box inside the lattice containing $\dim \mathbf{b}_{ij} = N^x_\sigma/N^x_b \times N^y_\sigma/N^y_b$ spins. Consequently the horizontal and vertical length of each box is given by $(l_x, l_y) = (L_x/N^x_b, L_y/N^y_b)$. The thermodynamic limit is defined as the scaling limit in which we take the number of spins/boxes to infinity while simultaneously keeping the lattice length fixed, i.e.

$$\lim_{N^x_\sigma, N^y_\sigma \to \infty} N^x_\sigma \cdot \delta_x \cdot N^y_\sigma \cdot \delta_y = \text{const.}, \quad \lim_{N^x_\sigma, N^y_\sigma \to \infty} N^x_b \cdot \delta_x \cdot N^y_b \cdot \delta_y = \text{const.},$$

where $\mathbf{N}_\sigma$ and $\mathbf{N}_b$ denote the thermodynamic limit of the spins and box numbers, respectively. Finally, the average lattice coordination number is denoted by $\bar{z} = ||\bar{z}||_1$, where $\bar{z} = (\bar{z}_x, \bar{z}_y)^T$ denote the average lattice coordination numbers in the horizontal and vertical direction.

B. Rewriting the Ising Hamiltonian

Here we start with a derivation that relates the Ising Hamiltonian given by Eq. (1) in the Letter to the number of defects in the lattice. Since the Ising Hamiltonian goes over nearest-neighbor terms, we can rewrite Eq. (1) in the Letter exactly as

$$H(\sigma) = -J \sum_{(km)} \sum_{(ln)} \sigma_{kl} \sigma_{mn} = \tilde{J}(N_{\uparrow\downarrow} - N_{\uparrow\downarrow}^{-} - N_{\downarrow\uparrow}^{-}),$$

(S18)

where $N_{\uparrow\downarrow}, N_{\downarrow\uparrow}$, and $N_{\uparrow\downarrow}^{-}$ denote the total number of up-up, down-down, and up-down (defects) spin pairs, respectively. Notice that every up-up spin pair consists of two up spins, and every up-down spin pair consists of a single up and a single down spin; hence, $2N_{\uparrow\downarrow} + N_{\downarrow\uparrow}^{-} \approx \bar{z} N_{\uparrow\downarrow}$, where $N_{\uparrow\downarrow}$ is the total number of up spins and $\bar{z}$ the average lattice coordination number. A similar reasoning applies to down-down spin pairs, resulting in the general relations

$$N_{\uparrow\downarrow}^{-} \approx (\bar{z} N_{\uparrow\downarrow} - N_{\downarrow\uparrow})/2, \quad N_{\downarrow\uparrow} \approx (\bar{z} N_{\downarrow\uparrow} - N_{\uparrow\downarrow}^{-})/2$$

(S19)

Note that Eq. (S19) is exact (i.e. $\approx \rightarrow =$) for infinite lattices and lattices with periodic boundary conditions. Plugging Eq. (S19) into Eq. (S18) and using that $N = N_{\uparrow\downarrow} + N_{\downarrow\uparrow}$ we obtain

$$H(\sigma) \approx \tilde{J}(2N_{\downarrow\downarrow}^{-} - \bar{z} N/2),$$

(S20)

where (as before) equality is restored for infinite lattices and lattices with periodic boundary conditions. Hence, we have re-written the sum over nearest neighbor spins in terms of the total number of defects $N_{\uparrow\downarrow}^{-}$. In the next sections we will formally define the number of defects in and between the coarse-grained boxes $\mathbf{b}_{ij}$ which we defined in III A. This allows us to define the number of defects at a given position in the lattice, which will eventually result in a position dependent Hamiltonian.

C. Coarse-grained lattice observables

Recall that $\dim \mathbf{b}_{ij} = N^x_b/N^x_\sigma \times N^y_b/N^y_\sigma$ denote the total number of spins inside a box. Let us define the concentration of down spins in box $\mathbf{b}_{ij}$ as

$$\varphi_{ij}(\mathbf{b}_{ij}) \equiv (\dim \mathbf{b}_{ij})^{-1} \sum_{(m,n) \in \mathbf{b}_{ij}} (1 - \sigma_{mn})/2.$$  

(S21)
Furthermore we define the total fraction of intra-box defects inside \( b_{ij} \) in the horizontal and vertical direction given by

\[
\zeta^{x}_{ij}(b_{ij}) \equiv (z_{x} \dim b_{ij})^{-1} \sum_{(m,n) \in b_{ij}} \sum_{(nk)} |\sigma_{mn} - \sigma_{kn}|/2, \tag{S22}
\]

\[
\zeta^{y}_{ij}(b_{ij}) \equiv (z_{y} \dim b_{ij})^{-1} \sum_{(m,n) \in b_{ij}} \sum_{(nk)} |\sigma_{mn} - \sigma_{mk}|/2, \tag{S23}
\]

where \( \langle nk \rangle \) denotes a sum over nearest neighbors. Finally we define the total fraction of inter-box defects between neighbouring boxes in the horizontal and vertical direction, respectively given by

\[
\xi^{x\pm}_{ij} (\{ b_{ij}, b_{i\pm 1j} \}) \equiv (z_{x} \dim b_{ij})^{-1} \sum_{(m,n) \in b_{ij}} \sum_{(k,n) \in b_{i\pm 1j}} |\sigma_{mn} - \sigma_{kn}|/2, \tag{S24}
\]

\[
\xi^{y\pm}_{ij} (\{ b_{ij}, b_{ij\pm 1} \}) \equiv (z_{y} \dim b_{ij})^{-1} \sum_{(m,n) \in b_{ij}} \sum_{(nk \in b_{ij\pm 1}} |\sigma_{mn} - \sigma_{mk}|/2, \tag{S25}
\]

where one needs to take into account of the lattice boundary conditions upon summing over neighbouring boxes around the boundaries.

**D. Hamiltonian and partition function**

We now write the nearest neighbour Ising Hamiltonian given by Eq. (S20) in terms of coarse-grained lattice variables Eqs. (S21)-(S25). Let the spin interaction strength in the horizontal and vertical direction be given by \( \beta J_{x}, \beta J_{y} \), where \( \beta \equiv 1/k_{B}T \) is the inverse thermal energy. For sake of generality we also introduce an external field \( \mu \equiv \beta \mu \) which couples to all “down spins” (i.e. \( \sigma_{ij} = -1 \)). We decompose the Hamiltonian in Eq. (S20) into a sum over intra- and inter-box contributions

\[
\tilde{H}(b) = \dim b_{ij} \sum_{i=1}^{N_{x}^{z}} \sum_{j=1}^{N_{y}^{z}} [\tilde{H}_{\text{inter}}(\{ b_{ij}, b_{i\pm 1j}, b_{ij\pm 1} \})/2 + \tilde{H}_{\text{intra}}(\{ b_{ij} \}) - C], \tag{S26}
\]

where the intra- and interbox Hamiltonian densities are given by

\[
\tilde{H}_{\text{inter}}(\{ b_{ij}, b_{i\pm 1j}, b_{ij\pm 1} \}) \equiv \bar{z}_{x} \tilde{J}_{x} \xi^{x\pm}_{ij}(\{ b_{ij}, b_{i\pm 1j} \}) + \xi^{x-}_{ij}(\{ b_{ij}, b_{i-1j} \}) + (x \leftrightarrow y), \tag{S27}
\]

\[
\tilde{H}_{\text{intra}}(\{ b_{ij} \}) \equiv \bar{z}_{x} \tilde{J}_{x} \xi^{x\pm}_{ij}(\{ b_{ij} \}) + (x \leftrightarrow y) - \tilde{\mu} \varphi_{ij}(\{ b_{ij} \}), \tag{S28}
\]

and the constant is given by \( C \equiv (z_{x} \tilde{J}_{x} + z_{y} \tilde{J}_{y})/2 \). The term \( (x \leftrightarrow y) \) in Eqs. (S27)-(S28) denote that the same term given in front follows but with \( x \) and \( y \) interchanged. Note that the factor 1/2 for \( \tilde{H}_{\text{inter}} \) in Eq. (S26) accounts for the double counting over interbox contributions. Plugging Eq. (S26) in combination with Eqs. (S27) and (S28) into the partition function \( Z = \sum_{\{ \sigma_{ij} \}} \exp (-\tilde{H}(b)) \) leads to the following exact expression

\[
Z = \sum_{b_{ij}} \prod_{i=1}^{N_{x}^{z}} \prod_{j=1}^{N_{y}^{z}} \mathcal{Z}(\{ b_{ij}, b_{i\pm 1j}, b_{ij\pm 1} \})
= \sum_{\{ b_{ij} \}} \prod_{i=1}^{N_{x}^{z}} \prod_{j=1}^{N_{y}^{z}} e^{-\dim b_{ij}[\bar{z}_{x} \tilde{J}_{x}(\xi^{x\pm}_{ij}(\{ b_{ij} \}) + (\xi^{x-}_{ij}(\{ b_{ij}, b_{i+1j} \}) + \xi^{x-}_{ij}(\{ b_{ij}, b_{i-1j} \})) + (x \leftrightarrow y) - \tilde{\mu}\varphi_{ij}(\{ b_{ij} \})] - C} \tag{S29}
\]

where in the first line we have defined the partition function per box \( \mathcal{Z}(\{ b_{ij}, b_{i\pm 1j}, b_{ij\pm 1} \}) \). The evaluation of the configurational sum over all possible box configurations \( \{ b_{ij} \} \) constitutes a difficult – if not impossible – task. However,
the Hamiltonian density inside the sum solely depends on the five lattice variables \((\varphi_{ij}, \zeta_{ij}^x, \zeta_{ij}^y, \zeta_{ij}^{x\pm}, \zeta_{ij}^{y\pm})\), \((i, j) \in \{1, \ldots, N_X^2 \} \times \{1, \ldots, N_Y^2 \}\). Therefore, we can interchange the configurational sum by a sum over all possible values of these five lattice variables and introduce a degeneracy of states \(\Psi(\varphi_{ij}, \zeta_{ij}^x, \zeta_{ij}^y, \zeta_{ij}^{x\pm}, \zeta_{ij}^{y\pm})\) which accounts for the multiplicity of configurations with equal lattice variables. Hence

\[
Z = \sum_{\varphi_{ij}} \sum_{\zeta_{ij}^x} \sum_{\zeta_{ij}^y} \sum_{\zeta_{ij}^{x\pm}} \sum_{\zeta_{ij}^{y\pm}} N_X^2 N_Y^2 \prod_{i=1}^{N_X^2} \prod_{j=1}^{N_Y^2} \Phi(\varphi_{ij}, \zeta_{ij}^x, \zeta_{ij}^y, \zeta_{ij}^{x\pm}, \zeta_{ij}^{y\pm}) e^{-\text{dim } b_{ij}[z_x J_x(\zeta_{ij}^x) + (\zeta_{ij}^{x+} + \zeta_{ij}^{x-})/2] + (z + y) - \bar{\mu} \varphi_{ij} - \zeta], \quad (S30)
\]

Note that Eq. \((S30)\) remains an exact expression as long as the degeneracy of states \(\Psi\) is evaluated correctly. We know that the degeneracy of states should obey the following relation

\[
\sum_{\zeta_{ij}^x} \sum_{\zeta_{ij}^y} \sum_{\zeta_{ij}^{x\pm}} \sum_{\zeta_{ij}^{y\pm}} \Psi(\varphi_{ij}, \zeta_{ij}^x, \zeta_{ij}^y, \zeta_{ij}^{x\pm}, \zeta_{ij}^{y\pm}) = \text{dim } b_{ij}\int_{\varphi_{ij}} \Psi(\varphi_{ij}), \quad (S31)
\]

since that is the amount of possible box configurations to place \(\varphi_{ij}\) down spins in a box that contains \(\text{dim } b_{ij}\) spins in total. We will use Eq. \((S31)\) later to consistently approximate the degeneracy of states. In the remainder of this SM we will evaluate Eq. \((S30)\) based on two different approximation schemes: the mean field (MF) and Bethe-Guggenheim (BG) approximations. The difference between the MF and BG approximation arises from a different treatment of the degeneracy factor, which will have important consequences for the resulting free energy density.

### IV. MEAN FIELD APPROXIMATION

#### A. Approximation of the fraction of defects

On the MF level we introduce the following approximation of the fraction of defects between two boxes \(b_{ij}\) and \(b_{nn}\):

\[
\zeta_{MF}(\varphi_{ij}, \varphi_{mn}) \equiv (\varphi_{ij} - 1 - \varphi_{mn})(1 - \varphi_{ij})/2. \quad (S32)
\]

In the MF level we thus approximate the number of defects between boxes \(b_{ij}\) and \(b_{nn}\) by the product of the spin down concentration in box \(b_{ij}\) and spin up concentration in box \(b_{nn}\), and vice versa. Making the substitutions \(\zeta_{ij}^x, \zeta_{ij}^y \to \zeta_{MF}(\varphi_{ij}, \varphi_{ij})\), \(\zeta_{ij}^{x\pm} \to \zeta_{MF}(\varphi_{ij} \pm 1, \varphi_{ij})\), and \(\zeta_{ij}^{y\pm} \to \zeta_{MF}(\varphi_{ij}, \varphi_{ij} \pm 1)\) we see that the energy density inside the exponent of Eq. \((S30)\) becomes independent of the variables \((\zeta_{ij}^x, \zeta_{ij}^y, \zeta_{ij}^{x\pm}, \zeta_{ij}^{y\pm})\). Therefore we can immediately use Eq. \((S31)\) to perform the four inner sums. This results in the MF partition function

\[
Z_{MF} \equiv \sum_{\varphi_{ij}} \prod_{i=1}^{N_X^2} \prod_{j=1}^{N_Y^2} Z_{MF}(\varphi_{i\pm j}, \varphi_{i\mp j}, \varphi_{i\pm j \pm 1}) = \sum_{\varphi_{ij}} \prod_{i=1}^{N_X^2} \prod_{j=1}^{N_Y^2} \left(\frac{\text{dim } b_{ij}}{\text{dim } b_{ij}}\right) e^{-\text{dim } b_{ij}[z_x J_x(\zeta_{MF}(\varphi_{ij}, \varphi_{ij}) + (\zeta_{MF}(\varphi_{i+1j}, \varphi_{ij}) + \zeta_{MF}(\varphi_{i-1j}, \varphi_{ij})/2) + (z + y) - \bar{\mu} \varphi_{ij} - \zeta], \quad (S33)
\]

where \(Z_{MF}(\varphi_{i\pm j}, \varphi_{i\mp j}, \varphi_{i\pm j \pm 1})\) is the MF partition function per box. To evaluate the sum over \(\varphi_{ij}\) in Eq. \((S33)\) we employ the maximum term method and take the maximum term of the continuous summand in the thermodynamic (scaling) limit. To that end we introduce the MF free energy density

\[
\tilde{f}_{MF}(\varphi_{i\pm j}, \varphi_{i\mp j}, \varphi_{i\pm j \pm 1}) \equiv \lim_{N^*} \left[\left(-\text{dim } b_{ij}\right)^{-1} \ln \left(Z_{MF}(\varphi_{i\pm j}, \varphi_{i\mp j}, \varphi_{i\pm j \pm 1})\right)\right]
\]

\[
= \Xi(\varphi_{ij}) + \Xi(1 - \varphi_{ij}) - \bar{x}_{ij} J_x[\zeta_{MF}(\varphi_{ij}, \varphi_{ij}) + (\zeta_{MF}(\varphi_{i+1j}, \varphi_{ij}) + \zeta_{MF}(\varphi_{i-1j}, \varphi_{ij})/2) + (z + y) - \bar{\mu} \varphi_{ij} - \zeta], \quad (S34)
\]

where we have used Stirling’s approximation \(\ln (n!) = \Xi(n) = n - N \ln (n) + O(\ln (n))\) with \(\Xi(n) \equiv n \ln (n)\) to evaluate the logarithm of the binomial coefficient. Note that so far we have only taken the thermodynamic limit of the spins. This makes \(\varphi_{ij} \in [0, 1]\) a continuous variable, as well as \(\zeta_{MF}(\varphi_{i\pm j}, \varphi_{ij}) \in [0, 1/4]\) and \(\zeta_{MF}(\varphi_{i\pm j \pm 1}, \varphi_{ij}) \in [0, 1/4]\). Upon considering the thermodynamic limit of the number of boxes we can employ two different strategies:

1. First optimize \(\tilde{f}_{MF}(\varphi_{i\pm j}, \varphi_{i\mp j}, \varphi_{i\pm j \pm 1})\) over \(\varphi_{ij}\) and finally apply \(\lim_{N^*} \left[\int \phi_{i\pm j}, \varphi_{i\mp j}, \varphi_{i\pm j \pm 1}\right]\) and then optimize the resulting free energy functional.

Below we carry out both and show that they give equivalent results for the resulting concentration profile. Only the second strategy, however, leads to a Cahn-Hilliard type free energy functional.
B. Optimization w.r.t. $\varphi_{ij} \to \lim_{N^y \to \infty}$

Using the maximum term method we need to find the $\varphi_{ij}$ which renders $Z_{MF}(\varphi_{i+1,j}, \varphi_{ij}, \varphi_{ij+1})$ maximal in the thermodynamic limit of the boxes. This is equivalent to finding the local minima of the local free density given by Eq. (S34), yielding the equation

$$\partial_{\varphi_{ij}} \left[ \tilde{I}_{MF}(\varphi_{i+1,j}, \varphi_{ij}, \varphi_{ij+1}) + \sum_{k=\pm 1} \left( \tilde{I}_{MF}(\varphi_{i+k+1,j}, \varphi_{i+kj}, \varphi_{i+kj+1}) + \tilde{I}_{MF}(\varphi_{i+1j+k}, \varphi_{ij+k}, \varphi_{ij+k+1}) \right) \right] = 0,$$  \hspace{1cm} (S35)

where $\partial_{\varphi_{ij}} \equiv \partial/\partial \varphi_{ij}$. Note that besides $\tilde{I}_{MF}(\varphi_{i+1,j}, \varphi_{ij}, \varphi_{ij+1})$ four extra terms enter Eq. (S35) since they also contain an explicit dependence on $\varphi_{ij}$. The solution to Eq. (S35) can be cast into the following set of difference equations

$$\tilde{z}_x \tilde{J}_x (\varphi_{i+1,j} - 2\varphi_{ij} + \varphi_{i-1,j}) + \tilde{z}_y \tilde{J}_y (\varphi_{ij+1} - 2\varphi_{ij} + \varphi_{ij-1}) = 2(\tilde{z}_x \tilde{J}_x + \tilde{z}_y \tilde{J}_y)(1 - 2\varphi_{ij}) - \ln(1/\varphi_{ij} - 1) - \tilde{\mu},$$  \hspace{1cm} (S36)

for $(i, j) \in \{(1, ..., N^x_y), (1, ..., N^y_y)\}$. Now we can carry out the scaling limit of the boxes, for which we introduce the following notation:

$$\lim_{N^x \to \infty} [\varphi_{ij} = \varphi(i\ell_x, j\ell_y)] \equiv \varphi(x, y) \forall (x, y) \in A,$$

$$\lim_{N^x \to \infty} [\varphi_{i+1,j} = \varphi(i\ell_x \pm \ell_z, j\ell_y)] \equiv \lim_{\ell_z \to 0} \varphi(x \pm \ell_z, y) \forall (x, y) \in A,$$

$$\lim_{N^x \to \infty} [\varphi_{i\pm 1,j} = \varphi(i\ell_x \pm \ell_z, j\ell_y)] \equiv \lim_{\ell_z \to 0} \varphi(x, y \pm \ell_y) \forall (x, y) \in A,$$  \hspace{1cm} (S37)

where $A = [-L_x/2, L_x/2] \times [-L_y/2, L_y/2]$. Applying $\lim_{N^x \to \infty} [\cdot]$ to both sides of Eq. (S36) we obtain the following partial differential equation

$$\tilde{z}_x \tilde{J}_x \tilde{J}_x^{2} \varphi(x, y) + \tilde{z}_y \tilde{J}_y \tilde{J}_y^{2} \varphi(x, y) = 2(\tilde{z}_x \tilde{J}_x + \tilde{z}_y \tilde{J}_y)(1 - 2\varphi(x, y)) - \ln(1/\varphi(x, y) - 1) - \tilde{\mu}, \forall (x, y) \in A,$$  \hspace{1cm} (S38)

where we have used that $\lim_{\ell_z \to 0} [\varphi(x \pm \ell_z, y) - 2\varphi(x, y) + \varphi(x - \ell_z, y)] = \tilde{J}_x^2 \varphi(x, y)$ and $\lim_{\ell_y \to 0} [\varphi(x, y \pm \ell_y) - 2\varphi(x, y) + \varphi(x, y - \ell_y)] = \tilde{J}_y^2 \varphi(x, y)$. Upon specifying the boundary conditions the solution to Eq. (S38) extremizes the MF partition function in the thermodynamic limit.

C. $\lim_{N^y \to \infty} [\cdot] \to$ optimization w.r.t. $\varphi(x, y)$

To apply the thermodynamic limit of the boxes to Eq. (S34) we first add and subtract $\tilde{z}_x \tilde{J}_x \tilde{J}_x^{2} \varphi(x, y)$ inside the third term, and similarly we add and subtract $\tilde{z}_y \tilde{J}_y \tilde{J}_y^{2} \varphi(x, y)$ inside the fourth term. Next we use Eq. (S37) to get

$$\lim_{\ell_z \to 0} [\tilde{I}_{MF}(\varphi(x \pm \ell_z, y), \varphi(x, y)) - 2\tilde{I}_{MF}(\varphi(x \pm \ell_z, y), \varphi(x, y)) + \tilde{I}_{MF}(\varphi(x - \ell_z, y), \varphi(x, y))] = \tilde{J}_x^2 (1 - \varphi(x, y)) \varphi(x, y),$$

$$\lim_{\ell_y \to 0} [\tilde{I}_{MF}(\varphi(x, y \pm \ell_y), \varphi(x, y)) - 2\tilde{I}_{MF}(\varphi(x, y \pm \ell_y), \varphi(x, y)) + \tilde{I}_{MF}(\varphi(x, y - \ell_y), \varphi(x, y))] = \tilde{J}_y^2 (1 - \varphi(x, y)) \varphi(x, y).$$  \hspace{1cm} (S39)

Inserting the result of Eq. (S39) into Eq. (S34) we obtain the following result in the thermodynamic limit

$$\lim_{N^y \to \infty} \tilde{I}_{MF}(\varphi_{i+1,j}, \varphi_{ij}, \varphi_{ij+1}) = \tilde{I}_{MF}(\varphi(x, y)) + (1 - \varphi(x, y))(\tilde{z}_x \tilde{J}_x \tilde{J}_x^{2} \varphi(x, y) + \tilde{z}_y \tilde{J}_y \tilde{J}_y^{2} \varphi(x, y))/2,$$  \hspace{1cm} (S40)

where the MF local free energy density is defined as

$$\tilde{I}_{MF}(\varphi) \equiv \Xi(1 - \varphi) + 2(\tilde{z}_y \tilde{J}_y + \tilde{z}_x \tilde{J}_x) [\varphi(1 - \varphi) - 1/4] - \tilde{\mu} \varphi.$$  \hspace{1cm} (S41)

Finally we can construct the MF free energy density functional which is given by

$$\tilde{F}_{MF}(\varphi(x, y)) \equiv \lim_{N^y \to \infty} \left[ \sum_{i=1}^{N^x_y} \sum_{j=1}^{N^y_y} \tilde{I}_{MF}(\varphi_{i+1,j}, \varphi_{ij}, \varphi_{ij+1}) \right]$$

$$= \frac{1}{L_x L_y} \int_{(x,y) \in A} \tilde{I}_{MF}(\varphi(x, y)) + (1 - \varphi(x, y))(\tilde{z}_x \tilde{J}_x \tilde{J}_x^{2} \varphi(x, y) + \tilde{z}_y \tilde{J}_y \tilde{J}_y^{2} \varphi(x, y))/2 dxdy$$

$$= \frac{1}{L_x L_y} \int_{(x,y) \in A} \tilde{I}_{MF}(\varphi(x, y)) + \tilde{z}_x \tilde{J}_x \tilde{J}_x^{2} (\partial_x \varphi(x, y))^2/2 + \tilde{z}_y \tilde{J}_y \tilde{J}_y^{2} (\partial_y \varphi(x, y))^2/2 dxdy.$$

\hspace{1cm} (S42)
where in the last line we carried out a partial integration (P.I.) and used zero-flux boundary conditions \( \partial_y \varphi(x, y) |_{y = \pm L_y/2} = \partial_x \varphi(x, y) |_{x = \pm L_x/2} = 0 \) which we will assume in later sections. The profile \( \varphi(x, y) \) which constitutes a stationary point of Eq. (S42), i.e. \( \delta \tilde{F}_{MF}[\varphi(x, y)]/\delta \varphi(x, y) = 0 \), is obtained by solving the corresponding Euler-Lagrange equation

\[
\tilde{J} \frac{\partial^2 \varphi(x, y)}{\partial x^2} + \tilde{J}_y \frac{\partial^2 \varphi(x, y)}{\partial y^2} = \partial_x \varphi(x, y) \tilde{H}_{MF}(\varphi(x, y)), \quad \forall (x, y) \in A.
\]  

(S43)

Plugging Eq. (S41) into Eq. (S43) finally results in Eq. (S38).

V. BETHE-GUGGENHEIM APPROXIMATION

A. Introduction

Before we start with the actual calculation we provide some intuition about the BG approximation. The basic idea is to derive a closed-form expression for the degeneracy factor in Eq. (S30) by placing pairs of spins onto the lattice. Imagine that we have a given number of spin pairs where \( N_{\uparrow\uparrow}, N_{\downarrow\downarrow}, \) and \( N_{\uparrow\downarrow} \) denote the number of up-up, down-down, and up-down spin pairs. Note that \( N_{\uparrow\downarrow} = N_{\downarrow\uparrow} \). The total number of distinct lattice configurations for fixed \( N_{\uparrow\uparrow}, N_{\downarrow\downarrow}, \) and \( N_{\uparrow\downarrow} \) is simply given by

\[
\Psi_{BG} = \frac{(N_{\uparrow\uparrow} + N_{\downarrow\downarrow} + N_{\uparrow\downarrow})!}{(N_{\uparrow\uparrow})!(N_{\uparrow\downarrow}/2)!^2(N_{\downarrow\downarrow})!}!
\]

(S44)

where the factor 1/2 in the denominator takes account of the symmetry of \( N_{\uparrow\downarrow} \). Eq. (S44) comprises the main essence of the BG approximation. For an even number of up-down spin pairs the term \((N_{\uparrow\downarrow}/2)!\) is well defined. However, when \( N_{\uparrow\downarrow} \) is odd we are forced to consider the generalized factorial (i.e., Gamma function) which then yields

\[
\Psi_{BG} = \frac{\Gamma(N_{\uparrow\uparrow} + N_{\downarrow\downarrow} + N_{\uparrow\downarrow} + 1)}{\Gamma(N_{\uparrow\uparrow} + 1)\Gamma(N_{\uparrow\downarrow}/2 + 1)^2\Gamma(N_{\downarrow\downarrow} + 1)},
\]

(S45)

where \( \Gamma(n) = (n - 1)! \) for \( n \in \mathbb{Z}^+ \). To express Eq. (S45) purely in terms of \( N_{\uparrow\downarrow} \) and \( N_{\downarrow\uparrow} \) we use the relations given by Eq. (S19). Finally, we plug the resulting expression for \( \Psi_{BG} \) back into Eq. (S30) and perform the sum over \( N_{\uparrow\downarrow} \) using the maximum-term method. To account for a non-uniform concentration profile we need to construct the degeneracy of states given by Eq. (S45) for each of the individual boxes \( b_{ij} \) defined in Sec. III A. This is an elaborate task since we must distinguish between spin pairs within the box and those located at the borders of adjacent boxes. Furthermore, we must also distinguish between horizontal and vertical spin pairs to account for an (in general) anisotropic coupling. In the next section we will do this in a formal way. Thereafter in the remaining sections we evaluate the partition sum with the maximum term method, and finally take the thermodynamic (scaling) limit.

B. Pair-distribution Ansatz for the degeneracy of states in a non-uniform system

On the BG level we apply a different technique and approximate the degeneracy factor in Eq. (S30) as follows

\[
\Psi_{BG}(\varphi_{ij}, \varphi_{i\pm 1,j}, \varphi_{ij\pm 1}, \xi^x_{ij}, \xi^y_{ij}, \xi^{x\pm}_{ij}, \xi^{y\pm}_{ij}) = N_{BG}(\varphi_{ij}, \varphi_{i\pm 1,j}, \varphi_{ij\pm 1}) \tilde{\Psi}_{BG}(\varphi_{ij}, \varphi_{i\pm 1,j}, \varphi_{ij\pm 1}, \xi^x_{ij}, \xi^y_{ij}, \xi^{x\pm}_{ij}, \xi^{y\pm}_{ij}),
\]

(S46)

where \( N_{BG}(\varphi_{ij}, \varphi_{i\pm 1,j}, \varphi_{ij\pm 1}) \) is a normalization constant left to be determined, and the unnormalized part reads

\[
\tilde{\Psi}_{BG}(\varphi_{ij}, \varphi_{i\pm 1,j}, \varphi_{ij\pm 1}, \xi^x_{ij}, \xi^y_{ij}, \xi^{x\pm}_{ij}, \xi^{y\pm}_{ij}) = \tilde{\Psi}_{BG,\text{intra}}(\varphi_{ij}, \xi^x_{ij}, \xi^y_{ij}) \tilde{\Psi}_{BG,\text{inter}}(\varphi_{ij}, \varphi_{i\pm 1,j}, \varphi_{ij\pm 1}, \xi^{x\pm}_{ij}, \xi^{y\pm}_{ij}).
\]

(S47)

Here we have split the degeneracy factor into a intra- and interbox contribution, respectively given by

\[
\tilde{\Psi}_{BG,\text{intra}}(\varphi_{ij}, \xi^x_{ij}, \xi^y_{ij}) = \psi_x(\varphi_{ij}, \varphi_{i\pm 1,j}, \varphi_{ij\pm 1}, \xi^x_{ij}, \xi^y_{ij}),
\]

(S48)

\[
\tilde{\Psi}_{BG,\text{inter}}(\varphi_{ij}, \varphi_{i\pm 1,j}, \varphi_{ij\pm 1}, \xi^{x\pm}_{ij}, \xi^{y\pm}_{ij}) = \prod_{\pm} \psi^{1/2}_x(\varphi_{ij}, \varphi_{i\pm 1,j}, \xi^{x\pm}_{ij}) \psi^1_y(\varphi_{ij}, \varphi_{ij\pm 1}, \xi^{y\pm}_{ij}).
\]

(S49)
The functions $\psi_{x,y}(a, b, c)$ which enter Eqs. (S48)-(S49) is given by
\[
\psi_{x,y}(a, b, c) \equiv [\hat{\Gamma}_{x,y}(1 - a - c)\hat{\Gamma}_{x,y}(b - c)\hat{\Gamma}_{x,y}(a - b + c)]^{-1/2},
\]
with $\hat{\Gamma}_{x,y}(w) \equiv \Gamma(\varepsilon_{x,y} \dim b_{ij}(w)/2 + 1)$, and $\Gamma(w)$ being the generalized factorial (i.e. Gamma function). Eq. (S50) can be derived by counting the number of degenerate configurations upon distributing pairs of spins over a lattice (see also [10–12]). The functions $\psi_{x}(\varphi_{ij}, \varphi_{j1}, \xi_{ij}^{\pm}, \xi_{ij}^{y \pm})$ and $\psi_{y}(\varphi_{ij}, \varphi_{ij+1}, \xi_{ij}^{y \pm})$ account for distributing pairs of spins inside a single box in the horizontal and vertical direction, respectively. While $\psi_{x}(\varphi_{ij}, \varphi_{i1+1}, \xi_{ij}^{x \pm})$ and $\psi_{y}(\varphi_{ij}, \varphi_{ij+1}, \xi_{ij}^{y \pm})$ account for distributing pairs of spins between two neighbouring boxes in the horizontal and vertical direction, respectively.

C. Evaluation of the normalization constant

The normalization constant $\mathcal{N}_{BG}(\varphi_{ij}, \varphi_{i1+1}, \varphi_{ij+1})$ is determined by the normalization equation Eq. (S31). To evaluate the four sums over the lattice variables ($\xi_{ij}^{x}, \xi_{ij}^{y}, \xi_{ij}^{x \pm}, \xi_{ij}^{y \pm}$) on the left hand side of Eq. (S31) we take the thermodynamic limit of the spins – thereby making the lattice variables continuous – and employ the maximum term method. This results in the following arguments
\[
\zeta_{BG}(\varphi_{ij}, \xi_{ij}) \equiv \sup_{\xi_{ij}} \lim_{N \to \infty} N^{-1} \ln \left( \psi_{x,y}(\varphi_{ij}, \varphi_{i1+1}, \xi_{ij}^{x,y}, \xi_{ij}^{x \pm}, \xi_{ij}^{y \pm}) \right) = \varphi_{ij} (1 - \varphi_{ij}),
\]
\[
\xi_{BG}^{x}(\varphi_{i1+1}, \varphi_{ij}) \equiv \sup_{\xi_{ij}} \lim_{N \to \infty} N^{-1} \ln \left( \psi_{x,y}^{1/2}((\varphi_{ij}, \varphi_{i1+1}, \xi_{ij}^{x \pm})) \right) = \varphi_{i1+1} (1 - \varphi_{ij}),
\]
\[
\xi_{BG}^{y}(\varphi_{ij+1}, \varphi_{ij}) \equiv \sup_{\xi_{ij}} \lim_{N \to \infty} N^{-1} \ln \left( \psi_{x,y}^{1/2}((\varphi_{ij}, \varphi_{ij+1}, \xi_{ij}^{y \pm})) \right) = \varphi_{ij+1} (1 - \varphi_{ij}).
\]

To obtain Eqs. (S51) we have used Stirling’s approximation for the Gamma function $\ln (\Gamma(w)) = \Xi(w) - w + O(\ln w)$ for $\text{Re}(w) > 0$ with $\Xi(w) \equiv w \ln (w)$. Plugging Eqs. (S51) into Eq. (S46), and finally solving Eq. (S31) for the normalization constant yields
\[
\mathcal{N}_{BG}(\varphi_{ij}, \varphi_{i1+1}, \varphi_{ij+1}) = \left( \prod_{ij} \psi_{ij}^{1/2}(\varphi_{ij}, \varphi_{i1+1}, \varphi_{ij+1}, \xi_{ij}^{x+y}, \xi_{ij}^{x \pm}, \xi_{ij}^{y \pm}) \right),
\]
where we have suppressed arguments of $\xi_{BG}$ and $\xi_{BG}^{x+y \pm}$ for convenience, which we henceforth do in all following sections. With the normalization constant evaluated explicitly we can now determine the partition function given by Eq. (S30).

D. Evaluation of the partition sum

Similarly to our previous calculation for the normalization constant, we take the thermodynamic limit of the spins and approximate the four sums over the lattice variables ($\zeta_{ij}^{x}, \zeta_{ij}^{y}, \xi_{ij}^{x \pm}, \xi_{ij}^{y \pm}$) with the maximum term method. The extremizing arguments read
\[
\zeta_{BG}^{x}(\varphi_{ij}, \xi_{ij}) \equiv \sup_{\xi_{ij}} \lim_{N \to \infty} N^{-1} \ln \left( \psi_{x,y}(\varphi_{ij}, \varphi_{i1+1}, \xi_{ij}^{x,y}, \xi_{ij}^{x \pm}, \xi_{ij}^{y \pm}) \right) = \frac{2\zeta_{BG}}{\Phi_{x,y}(0, \zeta_{BG})},
\]
\[
\xi_{BG}^{x}(\varphi_{i1+1}, \xi_{ij}) \equiv \sup_{\xi_{ij}} \lim_{N \to \infty} N^{-1} \ln \left( \psi_{x,y}^{1/2}(\varphi_{ij}, \varphi_{i1+1}, \xi_{ij}^{x \pm}) \right) = \frac{2\xi_{BG}^{x \pm}}{\Phi_{x}(\varphi_{ij} - \varphi_{i1+1}, \xi_{ij}^{x \pm})},
\]
\[
\xi_{BG}^{y}(\varphi_{ij+1}, \xi_{ij}) \equiv \sup_{\xi_{ij}} \lim_{N \to \infty} N^{-1} \ln \left( \psi_{x,y}^{1/2}(\varphi_{ij}, \varphi_{ij+1}, \xi_{ij}^{y \pm}) \right) = \frac{2\xi_{BG}^{y \pm}}{\Phi_{y}(\varphi_{ij+1} - \varphi_{ij}, \xi_{ij}^{y \pm})},
\]
with
\[
\Phi_{x,y}(a, b) \equiv 1 + a \gamma_{x,y} + [\delta_{a,0} + \text{sgn}(a)] \left( (1 + a \gamma_{x,y})^{2} + 4b \gamma_{x,y} \right)^{1/2},
\]
where $\text{sgn}(x) = \pm 1$ for $\pm x > 0$ and $\text{sgn}(0) = 0$, and we defined the auxiliary function $\gamma_{x,y} \equiv \exp (4 \hat{J}_{x,y}) - 1$. Note that for $\hat{J}_{x,y} = 0$ one obtains $\zeta_{BG}^{x} = \zeta_{BG}$ and $\xi_{BG}^{x \pm} = \xi_{BG}^{x \pm}$, as expected. With the sum over the lattice variables
(ζ_{ij}, ϕ_{ij}, ξ_{ij}, \bar{z}_{ij}^\pm, \hat{\xi}_{ij}^\pm) evaluated the BG partition function takes the following form

\[ Z_{BG} \equiv \sum_{\varphi_{ij}} N^x_{\varphi} N^y_{\varphi} Z_{BG}(\varphi_{i\pm1,j}, \varphi_{ij}, \varphi_{ij\pm1}) \]

\[ = \sum_{\varphi_{i}} N^x_{\varphi} N^y_{\varphi} \left( \prod_{b_{ij}} \right) \Psi_{BG}(\varphi_{ij}, \varphi_{i\pm1,j}, \varphi_{ij\pm1}, \hat{\xi}_{BG}, \bar{z}_{BG}^\pm) \exp \left\{ \sum_{b_{ij}} b_{ij} (z_{ij} J_{BG} + (\bar{z}_{ij}^+ + \bar{z}_{ij}^-)/2 + (x \leftrightarrow y) - \bar{\mu}_{ij} - c) \right\}, \]  

(S55)

where we have suppressed the arguments of \( \hat{\xi}_{BG} \) and \( \hat{\xi}_{BG}^\pm \) for convenience. Similarly to the MF calculation, we determine the sum over \( \varphi_{ij} \) in Eq. (S55) by employing the maximum term method in the thermodynamic limit of the spins. To that end we write down the BG free energy density

\[ f_{BG}(\varphi_{i\pm1,j}, \varphi_{ij}, \varphi_{ij\pm1}) \equiv \lim_{N^x} \left[ -(\text{dim } b_{ij})^{-1} \ln (Z_{BG}(\varphi_{i\pm1,j}, \varphi_{ij}, \varphi_{ij\pm1})) \right] \]

\[ = \frac{\tilde{z}}{x} J_{BG} (\tilde{\xi}_{BG}^x + (\tilde{\xi}_{BG}^x + \tilde{\xi}_{BG}^-)/2) + (\tilde{x} \leftrightarrow \tilde{y}) - \bar{\mu}_{ij} - c \]

\[ + \left( \tilde{z}/8 \right) \sum_{\pm} \Xi(1 - \varphi_{ij} - \tilde{\xi}_{BG}^x + \Xi(\varphi_{i\pm1,j} - \tilde{\xi}_{BG}^x) + \Xi(\varphi_{ij} - \varphi_{ij\pm1} + \tilde{\xi}_{BG}^x) + \Xi(\varphi_{ij}) \right] \]

\[ + \left( \tilde{y}/8 \right) \sum_{\pm} \Xi(1 - \varphi_{ij} - \tilde{\xi}_{BG}^y + \Xi(\varphi_{ij\pm1} - \tilde{\xi}_{BG}^y) + \Xi(\varphi_{ij}) \right) \]

\[ - \left( \tilde{z}/8 \right) \sum_{\pm} \Xi(1 - \varphi_{ij\pm1} + \Xi(\varphi_{ij})) \right] - \left( \tilde{y}/8 \right) \sum_{\pm} \Xi(1 - \varphi_{ij}) \right) \]

\[ + \left( 1 - 2\tilde{z}/4 \right) \left( \Xi(\varphi_{ij}) + \Xi(1 - \varphi_{ij}) \right) \]  

(S56)

where we recall that \( \Xi(x) \equiv x \ln(x) \) and \( \tilde{z} = ||b||_1 \). Comparing Eq. (S56) and Eq. (S34) we notice that the BG free energy density has considerably more terms than its MF counterpart due to the functional form of the degeneracy factor. As with the MF calculation we will consider two different strategies for carrying out the optimization over \( \varphi_{ij} \).

### E. Optimization w.r.t. \( \varphi_{ij} \rightarrow \lim_{N^x} \) [\( \cdot \)]

Extremizing Eq. (S55) over \( \varphi_{ij} \) in the thermodynamic limit of the spins is equivalent to finding the local minima of the BG free energy density given by Eq. (S56), yielding the equation

\[ \partial_{\varphi_{ij}} \left[ f_{BG}(\varphi_{i\pm1,j}, \varphi_{ij}, \varphi_{ij\pm1}) + \sum_{k=1}^{N^x_{\varphi}} \left( f_{BG}(\varphi_{i+k\pm1,j}, \varphi_{i+k,j}, \varphi_{i+k\pm1,j}) + f_{BG}(\varphi_{i\pm1,j+k}, \varphi_{ij+k}, \varphi_{ij+k\pm1,j}) \right) \right] = 0. \]  

(S57)

Note that when we take the partial derivative of the BG local free energy density w.r.t. \( \varphi_{ij} \), we can use

\[ \partial_{\varphi_{ij}} f_{BG}(\varphi_{i\pm1,j}, \varphi_{ij}, \varphi_{ij\pm1}) = \partial_{\tilde{\xi}_{BG}^x} f_{BG}(\varphi_{i\pm1,j}, \varphi_{ij}, \varphi_{ij\pm1}) = 0, \]

(S58)

since both \( \hat{\xi}_{BG}^x \) and \( \hat{\xi}_{BG}^y \) are obtained by minimization of the BG free energy density. This renders the evaluation of Eq. (S57) a relatively easy task and results in the following recurrent set of difference equations

\[ \frac{\tilde{z}}{8} \sum_{\pm} \ln \left( \frac{1 - \varphi_{ij} - \tilde{\xi}_{BG}^x(\varphi_{i\pm1,j}, \varphi_{ij})}{\varphi_{ij} - \varphi_{ij\pm1} + \tilde{\xi}_{BG}^x(\varphi_{i\pm1,j}, \varphi_{ij})} \right) - \ln \left( \frac{\varphi_{ij} - \tilde{\xi}_{BG}^x(\varphi_{ij\pm1}, \varphi_{ij})}{\varphi_{ij\pm1} - \varphi_{ij} + \tilde{\xi}_{BG}^x(\varphi_{ij\pm1}, \varphi_{ij})} \right) \]

\[ = \frac{\tilde{z}}{4} \left[ \ln \left( \frac{\varphi_{ij} - \tilde{\xi}_{BG}^y(\varphi_{ij\pm1}, \varphi_{ij})}{1 - \varphi_{ij} - \tilde{\xi}_{BG}^y(\varphi_{ij\pm1}, \varphi_{ij})} \right) + (x \leftrightarrow y) + \left( 1 - \tilde{z} \right) \ln \left( \frac{\varphi_{ij} - \tilde{\xi}_{BG}^x(\varphi_{ij\pm1}, \varphi_{ij})}{1 - \varphi_{ij} - \tilde{\xi}_{BG}^x(\varphi_{ij\pm1}, \varphi_{ij})} \right) \right]. \]  

(S59)

For a one-dimensional concentration profile \( \varphi \), a similar equation has been derived in [11] – see Eqs. (31)-(33) therein – where the solution is obtained (only) around the critical point. Here we will proceed with applying the thermodynamic limit of the boxes to Eq. (S59) using Eq. (S37). To obtain the thermodynamic limit we calculate the following terms:

\[ \lim_{l_x \to 0} \left( \sum_{x} \ln(1 - \varphi(x,y) - \tilde{\xi}_{BG}^x(\varphi(x\pm l_x, y), \varphi(x,y))) - 2\ln(1 - \varphi(x,y) - \tilde{\xi}_{BG}^x(\varphi(x,y), \varphi(x,y))) \right)/l_x^2 = \]

\[ \left( \frac{\tilde{z}_{BG}^{x(1,0)} \varphi(x,y) + \tilde{\xi}_{BG}^x}{\varphi(x,y) + \tilde{\xi}_{BG}^x - 1} \right) \partial_x^2 \varphi(x,y) - \left( \frac{\tilde{z}_{BG}^{x(1,0)} \varphi(x,y) + \tilde{\xi}_{BG}^x}{\varphi(x,y) + \tilde{\xi}_{BG}^x - 1} \right)^2 \partial_x \varphi(x,y) + \tilde{\xi}_{BG}^x (\partial_x \varphi(x,y))^2, \]  

(S60)
where  $\hat{\xi}^{(m,n)}_{\text{BG}} \equiv \partial^m_a \partial^n_b \hat{\xi}_{\text{BG}}(a,b)$, and we have used that $\hat{\xi}^\pm_{\text{BG}}(a,a) = \hat{\xi}_{\text{BG}}(a,a)$. Upon interchanging $x$ with $y$ the results of Eqs. (S60)-(S63) also apply to the $y$-direction. Note that the blue terms in Eqs. (S60) and (S61) are added by hand, and therefore also need to be added to the RHS of Eq. (S59). The purple terms in Eqs. (S62) and (S63) directly cancel, and therefore do not need to be added to the RHS. Summing up all the contributions we obtain the following expression

$$\bar{\xi}_x (S60) + (S61) + (S62) + (S63) = \kappa_x(\varphi(x,y)) \partial_x^2 \varphi(x,y) + \kappa'_x(\varphi(x,y))(\partial_x \varphi(x,y))^2/2,$$

where we have defined the function

$$\kappa_{\text{BG},x}(\varphi) \equiv \frac{\bar{\xi}_x(e^{4J_{x,y}} - 1)}{4\sqrt{1 + 4(e^{4J_{x,y}} - 1)}(1 - \varphi)}$$

which applies in the domain $(x,y) \in A$ with $A = [-L_x/2, L_x/2] \times [-L_y/2, L_y/2]$. Recall that $(x \leftrightarrow y)$ denotes that the same term given in front follows but with $x$ substituted by $y$, and $\hat{\xi}_{\text{BG}}$ is given by the first equation in Eqs. (S54). Eq. (S66) is the BG equivalent of the MF PDE given by Eq. (S38).

**F.** $\lim_{k \to \infty} [\cdot] \rightarrow \text{optimization w.r.t. } \varphi(x,y)$

Applying the thermodynamic limit to Eq. (S66) in the $x$-direction we need to keep track of the following terms:

$$\lim_{l_x \to 0} [\sum_{\pm} \hat{\xi}^\pm_{\text{BG}}(\varphi(\pm l_x, y), \varphi(\pm l_x, y)) - 2\hat{\xi}_{\text{BG}}(\varphi(0, y), \varphi(0, y))]/l_x^2 \xrightarrow{P.1} -(\partial_x \varphi(x,y))^2 \hat{\xi}_{\text{BG}}^{(1,1)},$$

$$\lim_{l_x \to 0} [\sum_{\pm} (\varphi(\pm l_x, y) - 2\varphi(\varphi(x, y)))/l_x^2 = (\partial_x \varphi(x,y))^2/\varphi(x,y) + \partial_x^2 \varphi(x,y)\ln(\varphi(x, y)) + 1 \xrightarrow{P.1} 0,$$

$$\lim_{l_x \to 0} [\sum_{\pm} (1-\varphi(\pm l_x, y) - 2(1-\varphi(\varphi(x, y)))/l_x^2 = (\partial_x \varphi(x,y))^2/(1-\varphi(x, y)) - \partial_x^2 \varphi(x,y)\ln(1-\varphi(x, y)) + 1 \xrightarrow{P.1} 0,$$
\[ \lim_{l_x \to 0} \left| \sum \Xi(\xi_{BG}^+(\varphi(x \pm l_x, y), \varphi(y, x)) - 2\Xi(\xi_{BG}^+(\varphi(x, y), \varphi(y, x))) \right| / l_x^2 \]

\[ = -(\partial_x \varphi(x, y))^2 \frac{\xi_{BG}^+(x \pm 1)}{\xi_{BG}^+(x)} - (\partial_x \varphi(x, y))^2 \frac{\xi_{BG}^+(1)}{\xi_{BG}^+(x) + 1}, \quad (S70) \]

\[ \lim_{l_y \to 0} \left| \sum \Xi(\varphi(x \pm l_x, y) - \xi_{BG}^+(\varphi(x \pm l_x, y), \varphi(y, x)) - 2\Xi(\varphi(x, y) - \xi_{BG}^+(\varphi(x, y), \varphi(y, x))) \right| / l_y^2 \]

\[ = -(\partial_y \varphi(x, y))^2 \frac{\xi_{BG}^+(x \pm 1)}{\xi_{BG}^+(x)} - (\partial_y \varphi(x, y))^2 \frac{\xi_{BG}^+(1)}{\xi_{BG}^+(x) + 1}, \quad (S71) \]

\[ \lim_{l_x \to 0} \left| \sum \Xi(\varphi(x \pm l_x, y) - \xi_{BG}^+(\varphi(x \pm l_x, y), \varphi(y, x)) - 2\Xi(\varphi(x, y) - \xi_{BG}^+(\varphi(x, y), \varphi(y, x))) \right| / l_x^2 \]

\[ = -(\partial_x \varphi(x, y))^2 \frac{\xi_{BG}^+(x \pm 1)}{\xi_{BG}^+(x)} - (\partial_x \varphi(x, y))^2 \frac{\xi_{BG}^+(1)}{\xi_{BG}^+(x) + 1}, \quad (S72) \]

where we have immediately carried out a partial integration – since each term arises inside an integral – and used zero-flux boundary conditions \( \partial_x \varphi(x, y) |_{x = \pm L_x / 2} = 0 \) to express everything in terms of \( (\partial_x \varphi(x, y))^2 \). Next we add up all the blue terms in Eqs. (S70)-(S73) and use the first equation of Eqs. (S54) to show that

\[ \ln \left( \frac{1 - \varphi(x, y) - \xi_{BG}^+(\varphi(x, y), \varphi(y, x))}{\xi_{BG}^+(y)^2} \right) = 4J_x, \quad (S74) \]

Therefore all the blue terms in Eqs. (S70)-(S73) exactly cancel with Eq. (S67) upon plugging them back into Eq. (S56). Adding up all the purple terms in Eqs. (S70)-(S73) gives the following result

\[ \frac{\xi_{BG}^+}{8} (\text{S70}) + (\text{S71}) + (\text{S72}) + (\text{S73}) = \frac{1}{2} \kappa_{BG,x}(\varphi(x, y))(\partial_x \varphi(x, y))^2. \quad (S75) \]

Upon interchanging \( x \leftrightarrow y \) the same results applies to the \( y \)-direction. Putting the results back into Eq. (S56) and adding/subtracting those terms which have been added by hand in Eqs. (S67)-(S73) we finally obtain the BG free energy density in the scaling limit of the lattice

\[ \lim_{N_y} N_x \left[ \hat{F}_{BG}(\varphi_{i \pm 1, j}, \varphi_{i, j \pm 1}) - \hat{F}_{BG}(\varphi_{i, j}, \varphi_{i, j}) \right] = \hat{F}_{BG}(\varphi_{i, j}, \varphi_{j, i}) (\partial_x \varphi(x, y))^2 + \frac{\kappa_{BG,x}}{2} (\partial_y \varphi(x, y))^2, \quad (S76) \]

where \( \kappa_{x,y}(\varphi) \) is defined in Eq. (S65), and the BG local free energy density is defined as

\[ \hat{F}_{BG}(\varphi) = 2[\hat{z}_x J_x \hat{\xi}_{BG}^+ + \hat{z}_y J_y \hat{\xi}_{BG}^+] - C/4 - \hat{\mu}(\varphi) + (1 - \hat{\xi})(\Xi(\varphi) + \hat{\Xi}(1 - \varphi)) \]

\[ + \frac{\hat{z}_x}{2} \hat{\Xi}(\varphi - \hat{\xi}_{BG}) + \frac{\hat{z}_y}{2} [\Xi(\varphi - \hat{\xi}_{BG})] + \frac{\hat{z}_y}{2} \Xi(\varphi) + \Xi(1 - \varphi - \hat{\xi}_{BG}) + 2\Xi(\hat{\xi}_{BG})]. \quad (S77) \]

Finally the BG free energy density functional is given by

\[ \hat{F}_{BG}[\varphi(x, y)] \equiv \lim_{N_x} \left[ \left( N_x N_y \right)^{-1} \sum_{i=1}^{N_x} \sum_{j=1}^{N_y} N_y \hat{F}_{BG}(\varphi_{i \pm 1, j}, \varphi_{i, j \pm 1}) \right] \]

\[ = \frac{1}{L_x L_y} \int_{(x,y) \in A} \left[ \hat{F}_{BG}(\varphi(x, y)) + \frac{\kappa_{BG,x}}{2} (\partial_x \varphi(x, y))^2 + \frac{\kappa_{BG,y}}{2} (\partial_y \varphi(x, y))^2 \right] dx dy \quad (S78) \]

where we have carried out partial integration with zero-flux boundary conditions \( \partial_x \varphi(x, y) |_{x = \pm L_x / 2} = 0 \) and \( \partial_y \varphi(x, y) |_{y = \pm L_y / 2} = 0 \). Note that the coordinates \( x \) in the Letter have been written in units of the box size \( (L_x, L_y) \),
which is equivalent to setting \( l_x = l_y = 1 \) in Eq. (S78). The profile \( \varphi(x, y) \) which constitutes a stationary point of Eq. (S78), i.e. \( \delta F_{BG} [\varphi(x, y)] / \delta \varphi(x, y) = 0 \), is obtained by solving the corresponding Euler-Lagrange equation

\[
\frac{\partial^2}{\partial x^2} \varphi(x, y) + \frac{\partial^2}{\partial y^2} \varphi(x, y) = \partial_x \varphi(x, y) \frac{\partial \varphi(x, y)}{\partial x} + (x \leftrightarrow y) = \partial_x \varphi(x, y) \frac{\partial \varphi(x, y)}{\partial x}.
\]  

Plugging Eq. (S77) into Eq. (S79) finally results in Eq. (S66).

VI. EQUILIBRIUM CONCENTRATION PROFILE

Here we consider a concentration profile which only varies in the \( x \) direction, i.e. \( \varphi(x, y) = \varphi(x) \forall x \in [-L_x/2, L_x/2] \). The equilibrium profile \( \varphi(x) \) is an extremum of Eqs. (S42) and (S78) for the MF and BG approximation, respectively. Here we will derive analytical expressions for the interfacial steepness, interfacial width (according to the Cahn-Hilliard definition), and prove the broadening of the BG equilibrium profile.

A. Results within Mean Field theory

For a one-dimensional concentration profile Eq. (S43) reduces to a second order autonomous ODE. Therefore we can directly obtain the interfacial steepness \( \varphi_{MF}^\prime(x) \), which reads

\[
\varphi_{MF}^\prime(x) = \pm \sqrt{2(\tilde{\varphi}_{MF}(\varphi_{MF}(x)) - \tilde{\varphi}_{MF}(\varphi_{MF,min})/\tilde{x} J x},
\]  

where we have set the integration constant to \( C_1 = -\tilde{\varphi}_{MF}(\varphi_{MF,min}^\pm) \) with \( \varphi_{MF,min}^\pm \equiv \inf_{0 \leq \varphi \leq 1/2} \tilde{\varphi}_{MF}(\varphi) \) and \( \varphi_{MF,min}^\pm = 1 - \varphi_{MF,min}^\pm \) such that the term inside the square root on the RHS is always positive and \( \lim_{x \to \pm \infty} \varphi_{MF}(x) = 0 \). The location of the global minimum of the uniform MF free energy density can be written as \( \varphi_{MF,min}^\pm \equiv (1 \pm |s|)/2, \) where \( s \in [-1, 1] \) is given by the nonzero solutions to the so-called transcendental mean field equation \[10\]

\[
s = \tanh ((\tilde{x} J x + \tilde{y} J y)|s + \tilde{\mu}).
\]  

Below the critical coupling for \( \tilde{x} J x + \tilde{y} J y \leq 1 \) and for \( \tilde{\mu} = 0 \) the only solution to Eq. (S81) is given by \( s = 0 \), resulting in \( \varphi_{MF,min}^\pm = 1/2 \). Above the critical coupling for \( \tilde{x} J x + \tilde{y} J y > 1 \) there exists two nonzero solutions resulting in \( \varphi_{MF,min} < 1/2 \). Now let us focus on the isotropic case with a vanishing external field, i.e. \( \tilde{J}_x = \tilde{J}_y = \tilde{J} \) and \( \tilde{\mu} = 0 \), and consider the interfacial steepness at \( x = 0 \). Based on the imposed boundary conditions we know that \( \varphi_{MF}(0) = 1/2 \), and therefore the interfacial steepness at \( x = 0 \) reads

\[
\varphi_{MF}^\prime(0) = \pm \sqrt{2(\tilde{\varphi}_{MF}(1/2) - \tilde{\varphi}_{MF}(\varphi_{MF,min})/\tilde{x} J}. \]  

For a square lattice \( (\tilde{s} = 4, \tilde{x}_{z} = 2) \) Eq. (S82) is shown in Fig. (2)c in the Letter with the red solid line. For \( \tilde{s} J \leq 1 \) we have \( \varphi_{MF,min} = 1/2 \), and therefore we find \( \varphi_{MF}^\prime(0) = 0 \). This results is expected since there is no phase separation for this range of the coupling strength. To obtain the interfacial width as defined by Cahn and Hilliard (see Eq. (2.25) in [13]) we simply need take a line tangential to the slope of the concentration profile at \( x = 0 \) and determine the crossing points of this line with the bulk concentration values as depicted in Fig. S3a. This leads to the expression

\[
l_{MF,CH} = (\varphi_{MF,min}^+ - \varphi_{MF,min}^-)/\varphi_{MF}^\prime(0),
\]  

where we insert Eq. (S82) for \( \varphi_{MF}^\prime(0) \) with the positive sign. We see that for \( \tilde{s} J \leq 1 \) we have \( l_{MF,CH} \to \infty \). Now let us consider an infinite coupling strength. In this limit the nonzero solutions to Eq. (S81) are trivially given by \( s = \pm 1 \forall \tilde{x} > 0 \), and therefore we obtain

\[
\lim_{J \to \infty} \varphi_{MF}(0) = \lim_{J \to \infty} \pm \sqrt{2(\tilde{\varphi}_{MF}(1/2) - \tilde{\varphi}_{MF}(0 \vee 1)/\tilde{x} J}\tilde{J} = \lim_{J \to \infty} \pm \sqrt{2(\tilde{s} J/2 - \ln (2))/\tilde{x} J} = \pm \sqrt{\tilde{s}/\tilde{x}}.
\]  

Hence, in the infinite coupling limit the interfacial steepness converges to a maximum finite nonzero value. This value for the MF interfacial steepness is also reported in Fig. (2)c in the Letter. Furthermore, the interfacial width decreases and converges to the value

\[
\lim_{J \to \infty} l_{MF,CH} = \lim_{J \to \infty} (\varphi_{MF,min}^+ - \varphi_{MF,min}^-)/\varphi_{MF}^\prime(0) = \sqrt{\tilde{s}/\tilde{x}}.
\]  

(S85)
B. Results within Bethe-Guggenheim theory

Similar to the MF analysis Eq. (S79) reduces to a second order autonomous ODE for a one-dimensional concentration profile. To obtain the interfacial steepness we first rewrite the LHS of Eq. (S79) as

\[ \kappa_{BG,x}(x)\varphi'_{BG}(x) + \kappa_{BG,x}(\varphi_{BG}(x))^2/2 = \frac{1}{2\varphi'_{BG}(x)} \frac{d}{dx}[\kappa_{BG,x}(\varphi'_{BG}(x))^2]. \]  

(S86)

Taking the term \(1/\varphi'_{BG}(x)\) to the RHS of Eq. (S79) and using the fact that \(\varphi_{BG}(x)(\partial_{BG}(\varphi_{BG}(x))/\partial \varphi_{BG}(x)) = df_{BG}(\varphi_{BG}(x))/dx\), we can integrate both sides over \(x\), resulting in the first-order autonomous ODE

\[ \frac{1}{2}\kappa_{BG,x}(\varphi'_{BG}(x))^2 = f_{BG}(\varphi_{BG}(x)) + C_1, \]

(S87)

where \(C_1\) is an integration constant. From Eq. (S87) we can directly readout the interfacial steepness

\[ \varphi'_{BG}(x) = \pm \sqrt{2(f_{BG}(\varphi_{BG}(x)) - f_{BG}(\varphi_{BG,min}))}/\kappa_{BG,x}(\varphi_{BG}(x)), \]

(S88)

where we have set the integration constant \(C_1 = -f_{BG}(\varphi_{BG,min}^{\pm})\) with \(\varphi_{BG,min}^{\pm} = \inf_{0 \leq \varphi \leq 1/2} f_{BG}(\varphi)\) and \(\varphi_{BG,min}^{\pm} = 1 - \varphi_{BG,min}^{\pm}\). The integration constant is chosen such that the term inside the square root on the RHS is always positive and to impose a vanishing derivative at the boundaries. Now let us focus specifically on the isotropic case with a vanishing external field, i.e. \(J_x = J_y = \tilde{J}\) and \(\mu = 0\). The location of the global minimum \(\varphi_{BG,min}^{\pm}\) of the local BG free energy density can be written as \(\varphi_{BG,min}^{\pm} = \chi_{\varphi}/(1 + \chi_{\varphi})\), where \(\chi_{\varphi} \in [0, \infty)\) is given by the nontrivial solutions (i.e. \(\chi_{\varphi} \neq 1\)) to the transcendental equation [10]

\[ \chi_{\varphi} - e^{2\tilde{J}x}(\chi_{\varphi}^{-1/2} - e^{-\tilde{\mu}/\chi_{\varphi}^{1/2}}) - 1 = 0. \]

(S89)

Below and at the critical coupling \(\tilde{J} \leq \ln (\tilde{\varphi}/(\tilde{\varphi} - 2))/2\) Eq. (S89) has one trivial solution \(\chi_{\varphi} = 1\), resulting in \(\varphi_{BG,min}^{\pm} = 1/2\). Above the critical coupling there exists two nontrivial solutions, resulting in \(\varphi_{BG,min}^{\pm} \neq 1/2\). Eq. (S89) cannot be solved analytically for general \(\tilde{\varphi}\) but is explicitly solvable for a triangular, square, and hexagonal lattice, which gives

\[ \varphi_{BG,min}^{\pm}_{\tilde{\varphi} = 3 \mid \tilde{\mu} = 0} = \begin{cases} \frac{1}{2}, & 0 \leq \tilde{J} \leq \ln (3)/2 \\ \frac{1}{2} \left[ 1 \pm e^{2\tilde{J}((e^{2\tilde{J}} + 1)(e^{2\tilde{J}} - 3))^{1/2}} \right] 2(e^{2\tilde{J}} \sinh (\tilde{J} - 1)) \right], & \tilde{J} \geq \ln (3)/2 \end{cases}, \]

\[ \varphi_{BG,min}^{\pm}_{\tilde{\varphi} = 4 \mid \tilde{\mu} = 0} = \begin{cases} \frac{1}{2}, & 0 \leq \tilde{J} \leq \ln (2)/2 \\ \frac{1}{2} \left[ 1 \pm e^{2\tilde{J}(e^{2\tilde{J}} - 4)^{1/2}} e^{2\tilde{J} - 2} \right], & \tilde{J} \geq \ln (2)/2 \end{cases}, \]

\[ \varphi_{BG,min}^{\pm}_{\tilde{\varphi} = 6 \mid \tilde{\mu} = 0} = \begin{cases} \frac{1}{2}, & 0 \leq \tilde{J} \leq \ln (3/2)/2 \\ \frac{1}{2} \left[ e^{4\tilde{J}} + (e^{4\tilde{J}} + 4)^{1/2} + \sqrt{2}(e^{4\tilde{J}}(e^{4\tilde{J}} + 4)^{1/2} + e^{4\tilde{J}} - 6)^{1/2} \right]^6, & \tilde{J} \geq \ln (3/2)/2 \end{cases}. \]

(S90)

For \(\tilde{\mu} \neq 0\) Eq. (S89) is also explicitly solvable for a triangular and square lattice but the final expression is less compact (see for example Eq. (D16) in [10] for \(\tilde{\varphi} = 4\)). Plugging (S90) into Eq. (S88) and noting that \(\varphi(0) = 1/2\) we obtain closed-form expressions for the interfacial steepness at \(x = 0\). Similarly, using the definition given by Eq. (S83), we obtain the Cahn-Hilliard interfacial width for the BG approximation. Results for the interfacial steepness are shown in Fig. (2)c in the Letter with the blue lines and display a strong non-monotonic trend w.r.t. \(\tilde{J}\). The broadening of the profile is in sharp contrast to the conclusion drawn by Cahn and Hilliard who write in [13]: “The interface between two coexisting phases is diffuse and its thickness increases with increasing temperature until at the critical temperature \((T_c)\) the interface is infinite in extent.” (p266) Recall that \(\tilde{J} = J/k_BT\), and therefore an increase in temperature corresponds to a decrease in \(\tilde{J}\). To proof that broadening is a general effect regardless of the lattice we take the strong coupling limit of Eq. (S88). For \(\tilde{\varphi} > 2\) and \(\tilde{J} \to \infty\) the nontrivial solutions to Eq. (S89) are approaching \(\chi_{\varphi} \to 0\) and \(\chi_{\varphi} \to \infty\), resulting in \(\varphi_{BG,min}^{\pm} \to 0 \lor 1\). Plugging this into Eq. (S88) together with \(\varphi_{BG}(0) = 1/2\) we obtain

\[ \lim_{\tilde{J} \to \infty} \varphi_{BG}(0) = \lim_{\tilde{J} \to \infty} \pm \sqrt{2(2\tilde{\varphi} \tilde{J} - \tilde{J} \ln (e^{2\tilde{J}} + 1) + (\tilde{\varphi} - 2) \ln (2))/2x \sinh (2\tilde{J})} = 0. \]

(S91)
FIG. S3. (a) Representation of the Cahn-Hilliard interfacial width $l_{\text{CH}}$ used in Eq. (S83). Here we used the concentration profile for a hexagonal ($\bar{z} = 6$) lattice obtained with the BG approximation. (b) Critical wavevector obtained with the MF approximation Eq. (S97) for a square lattice. The black line represents the MF spinodal and the black dot the MF critical point $\bar{\kappa}_{\text{MF}} = 1/4$. (c) Critical wavevector obtained with the BG approximation Eq. (S99) for a square lattice. The black line represents the BG spinodal and the black dot the BG critical point $\bar{\kappa}_{\text{BG}} = \ln(2)/2$.

So we find a vanishing interfacial steepness at $x = 0$ for any lattice with $\bar{z} > 2$ in the strong coupling limit. For the interfacial width we find

$$\lim_{\bar{\kappa} \to \infty} l_{\text{BG},\text{CH}} = \lim_{\bar{\kappa} \to \infty} (\phi_{\text{BG,min}}^+ - \phi_{\text{BG,min}}^-)/\phi_{\text{BG}}(0) = \infty.$$

VII. LINEAR STABILITY ANALYSIS

Here we determine the length scales on which inhomogeneities of the concentration profile are stable. We consider a concentration profile of the form $\phi(x) = \phi_0 + a \sin(q \cdot x)$ with $q = (q_x, q_y)^T$ and $|a| \ll \min(\phi_0, 1 - \phi_0)$. A sinusoidal perturbation is taken to agree with the odd boundary conditions which we imposed for Eq. (S43) and (S79). Expanding the local free energy density and gradient energy coefficient around the homogeneous state up to second order gives

$$\bar{f}(\phi(x)) = \bar{f}(\phi_0) + a \sin(q \cdot x) \bar{f}'(\phi_0) + \frac{1}{2} a^2 \sin^2(q \cdot x) \bar{f}''(\phi_0) + O(a^3),$$

$$\frac{1}{2} \nabla \phi(x)^T \kappa(\phi(x)) \nabla \phi(x) = \frac{1}{2} a^2 (q^T \kappa(\phi_0) q) \cos^2(q \cdot x) + O(a^3),$$

where $\bar{f}'(\phi_0) \equiv \partial_\phi \bar{f}(\phi)|_{\phi_0}$ and $\bar{f}''(\phi_0) \equiv \partial_\phi^2 \bar{f}(\phi)|_{\phi_0}$. Now we want to find out when a sinusoidal perturbation decreases the total free energy compared to the uniform concentration profile. Plugging Eqs. (S93) and (S94) into Eq. (3) in the Letter and subtracting the free energy density of the uniform concentration gives

$$\bar{F}[\phi(x)] - \bar{F}[\phi_0] = \frac{L_y}{2L_x L_y} \int_{-L_x}^{L_x} \int_{-L_y}^{L_y} \left[2a \sin(q \cdot x) \bar{f}'(\phi_0) + a^2 (\sin^2(q \cdot x) \bar{f}''(\phi_0) + \cos^2(q \cdot x) q^T \kappa(\phi_0) q) \right] dx dy$$

$$= \frac{a^2}{4L_x L_y} \left(L_x L_y \left(\bar{f}''(\phi_0) + q^T \kappa(\phi_0) q\right) - (q_x q_y)^{-1} \sin(q_x L_x) \sin(q_y L_y) \left(\bar{f}''(\phi_0) - q^T \kappa(\phi_0) q\right)\right)$$

$$= \frac{a^2}{4} \left(\bar{f}''(\phi_0) + q^T \kappa(\phi_0) q\right) + O\left(\frac{a^3}{L_x L_y}\right),$$

where in the last line we have taken the large system-size limit $(L_x, L_y) \to \infty$. To decrease the total free energy the RHS of Eq. (S95) must be negative. Note that $q^T \kappa(\phi_0) q \geq 0$, and therefore only $\bar{f}''(\phi_0)$ can make the RHS negative. The region where $\bar{f}''(\phi_0) < 0$ in the $(\phi_0, J)$-plane is called the spinodal region, and therefore this process is also known as spinodal decomposition. When $\bar{f}''(\phi_0) < 0$ there is an upper bound on stable wavevectors which is given by

$$q^T_{\text{crit}} \kappa(\phi_0) q_{\text{crit}} = -\bar{f}''(\phi_0)$$
For a one-dimensional perturbation with \( q_y = 0 \) this translates to \( q_{\text{crit}} = (\tilde{f}'(\phi_0)/\kappa_x(\phi_0))^{1/2} \). The critical wavelength given by \( \lambda_{\text{crit}} = 2\pi/q_{\text{crit}} \) provides a lower bound on stable wavelengths. We will now determine the properties of \( q_{\text{crit}} \) and \( \lambda_{\text{crit}} \) for the MF and BG approximation.

### A. Results within Mean Field theory

Taking the MF local free energy density and square gradient coefficient defined in Eqs. (S40)-(S41) and plugging them into \( q_{\text{crit}} \) gives the following result

\[
q_{\text{crit, MF}} = \sqrt{-\frac{\tilde{f}'(\phi_0)}{\kappa_{\text{MF}}(\phi_0)}} = \sqrt{\frac{4(\bar{z}_x J_x + \bar{z}_y J_y) - 1/(\phi_0(1 - \phi_0))}{\bar{z}_x J_x}}. \tag{S97}
\]

For isotropic interaction strength \( \tilde{J}_x = \tilde{J}_y = \tilde{J} \) and inside the spinodal region \( \bar{z}, \tilde{J} \geq 1/(4\phi_0(1 - \phi_0)) \) the MF critical wavevector is monotonically increasing with \( \tilde{J} \) and for \( 0 < \phi_0 < 1 \) converges to

\[
\lim_{\tilde{J} \to \infty} q_{\text{crit, MF}} = 2\sqrt{\bar{z}/\bar{z}_x}. \tag{S98}
\]

In Fig. S3b we plot Eq. (S97) for a square lattice with isotropic interaction strength. The critical wavelength \( \lambda_{\text{crit, MF}} \) decreases monotonically with \( \tilde{J} \) and converges within the aforementioned range to the value \( \lim_{\tilde{J} \to \infty} \lambda_{\text{crit, MF}} = \pi \sqrt{\bar{z}_x/\bar{z}} \). In Fig. (2)g in the Letter we show the MF critical wavelength for a square lattice with the red line.

### B. Results within Bethe-Guggenheim theory

The BG local free energy density and square gradient coefficient are defined in Eqs. (S65) and (S77). For convenience we immediately take the isotropic interaction strength \( \tilde{J}_x = \tilde{J}_y = \tilde{J} \). Plugging the results for the second derivative of the local free energy density (see Eq. (B17) with \( \tilde{h} = 0 \) in [10]) into \( q_{\text{crit}} \) gives

\[
q_{\text{crit, BG}} = \sqrt{-\frac{\tilde{f}'(\phi_0)}{\kappa_{\text{BG}}(\phi_0)}} = \sqrt{\frac{2(\bar{z} - 2)(1 + 4(e^{4\tilde{J}} - 1)\phi_0(1 - \phi_0))\bar{z}_x - 2\bar{z}_x \phi_0(1 - \phi_0)(e^{4\tilde{J}} - 1)}{\bar{z}_x \phi_0(1 - \phi_0)(e^{4\tilde{J}} - 1)}}. \tag{S99}
\]

Inside the spinodal region \( J \geq \ln \left( (\bar{z} - 1 - \phi_0(\bar{z} - 2))(1 + \phi_0(\bar{z} - 2))/((\bar{z} - 2)^2\phi_0(1 - \phi_0)) \right)/4 \) (see Eq. (16) with \( \tilde{h} = 0 \) in [10]) the BG critical wavevector has a non-monotonic trend and for \( 0 < \phi_0 < 1 \) converges to the value

\[
\lim_{\tilde{J} \to \infty} q_{\text{crit, BG}} = 0. \tag{S100}
\]

In Fig. S3c we plot Eq. (S99) for a square lattice with isotropic interaction strength. Similarly the critical wavelength diverges, i.e. \( \lim_{\tilde{J} \to \infty} \lambda_{\text{crit, BG}} = \infty \). Hence for \( 0 < \phi_0 < 1 \) there exist no finite stable wavelength perturbations in the strong interaction limit. The coupling strength \( J^1(\phi_0) \) where \( q_{\text{crit, BG}} \) is maximal – and therefore \( \lambda_{\text{crit, BG}} \) minimal – is given by

\[
J^1(\phi_0) = \frac{1}{4} \ln \left( 1 + \frac{\bar{z}(2 + \sqrt{\bar{z}} - 1) - 2}{(\bar{z} - 2)^2\phi_0(1 - \phi_0)} \right). \tag{S101}
\]

For \( \phi_0 = 1/2 \) we recover Eq. (6) in the Letter. Remarkably, the maximum of \( q_{\text{crit, BG}} \) – and therefore the minimum of \( \lambda_{\text{crit, BG}} \) – is independent of the uniform concentration value \( \phi_0 \) and reads upon plugging Eq. (S101) into Eq. (S99)

\[
q_{\text{crit, BG}}^{\text{max}} = \frac{2[\bar{z} - 2]}{\sqrt{\bar{z}_x}} \sqrt{\frac{(\bar{z}(1 + \sqrt{\bar{z}} - 1 + \sqrt{\bar{z}/4}) - 1)^{\frac{1}{2}} - \bar{z}/2}{\bar{z}(2 + \sqrt{\bar{z}} - 1) - 2}}. \tag{S102}
\]

The minimum wavelength is easily obtained by \( \lambda_{\text{crit, BG}}^{\text{min}} = 2\pi/q_{\text{crit, BG}}^{\text{max}} \). In Fig. (2)g in the Letter we depict the BG critical wavelength for a square lattice with the blue line. The coupling values where \( \lambda_{\text{crit, BG}} \) attains a minimum is indicated with the blue arrow.
FIG. S4. Relative error between the exact and approximated partition function obtained with the (a) MF and (b) BG approximation for increasing number of spins and various values of the coupling strength $\tilde{J} = \{0, 1, 2, 3\}$. The relative error Eq. (S104) is determined for a square lattice composed of $(N_x^\sigma = 3) \times (N_y^\sigma = \{3, \ldots, 15\})$ spins with periodic boundary conditions in the vertical and anti-symmetric boundary conditions in the horizontal direction, respectively. The total fraction of down spins is fixed to $\hat{\psi} = 1/2$.

VIII. ERROR ANALYSIS OF THE APPROXIMATE PARTITION FUNCTIONS IN FINITE SYSTEMS

To probe the accuracy of the MF and BG approximations we compare their partition functions given by Eqs. (S33) and (S55) with exact results for the partition function of finite systems. We limit our error analysis to a one-dimensional concentration profile, conform with the majority of results discussed in the Letter. For a uniform concentration profile an error analysis between the MF and BG approximation is provided in [10] (see Fig. (11)). For a lattice composed of $N_x^\sigma \times N_y^\sigma$ spins, let $\varphi = (\varphi_1, \ldots, \varphi_{N_x^\sigma})$ be a vector containing the concentration of down spins in each column of the lattice. The total concentration of down spins in the lattice is given by $\varphi = |\varphi||/N_x^\sigma$. The exact partition function for a fixed concentration profile along the columns is denoted with $Z(\varphi)$ and can be computed via

$$Z(\varphi) = \sum_{\sigma} e^{-\tilde{\mathcal{H}}(\sigma)} \prod_{i=1}^{N_x^\sigma} 1_{\varphi_i} [\varphi_i],$$

(S103)

where we recall that $\sigma$ denotes the matrix containing all spin configurations, $1_x [z]$ is the indicator function of $x$, and $\mathcal{H}(\sigma)$ is given by Eq. (1) in the Letter. Note that in the Letter we define a partition function for a fixed concentration of down spins in disjoint boxes $b_{ij}$, whereas here we consider the concentration of down spins in each column of the lattice. The relative error between $Z_{BG, MF}(\varphi)$ and $Z(\varphi)$ for a fixed total concentration of down spins $\hat{\varphi}$ is defined as

$$\epsilon_N(\hat{\varphi}) = \left( \frac{\sum_{\sigma} Z(\varphi) \left( 1 - \frac{\ln(Z_{MF, BG}(\varphi))}{\ln(Z(\varphi))} \right) 1_\varphi [\varphi]}{\sum_{\sigma} Z(\varphi) 1_\varphi [\varphi]} \right).$$

(S104)

Eq. (S104) is defined such that differences between $Z(\varphi)$ and $Z_{BG, MF}(\varphi)$ attain the largest weight for thermodynamically stable configurations. In Fig. S4 we plot the relative error for the (a) MF and (b) BG approximation for a finite square lattice composed of $(N_x^\sigma = 3) \times (N_y^\sigma = \{3, \ldots, 15\})$ spins with anti-symmetric and periodic boundary conditions in the horizontal and vertical direction, respectively. Upon increasing the number of spins in the vertical direction we see that the relative error of the BG approximation decreases towards zero regardless of the coupling strength, whereas the MF approximation saturates to a nonzero value (note that the small system size gives rise to a marked even-odd dependency). For $\tilde{J} = 0$ both approximations are exact and therefore have zero relative error. The improvement of the BG approximation with increasing $N_y^\sigma$ is due to the fact that Eq. (S55) is obtained through a variational principle which is applied in the thermodynamic scaling limit (see for example Eq. (S54)). The MF approximation on the other hand becomes worse with increasing $N_y^\sigma$ due to the approximation for the fraction of defects given by Eq. (S32).

IX. NUMERICAL SIMULATIONS OF THE RADIIALLY SYMMETRIC CAHN-HILLIARD EQUATION

We study nucleation based on radially symmetric concentration profiles $\varphi(r)$ in two dimensions. Since critical profiles correspond to stationary points of the free energy $\tilde{F}$ given by Eqs. (S42) and (S78), we next determine minimal free
energy paths between the homogeneous state and large droplets. We use a measure for the mass concentrated in the nucleus, \(N[\varphi] = \int \tanh(w(\varphi - 1/2))dV\) with \(w = 10\), as a reaction coordinate and determine the profile \(\varphi(r)\) that minimizes \(\tilde{F}\) for a given value \(N_0\) of the constraint using a Lagrange multiplier \(\lambda\). We thus minimize the constrained free energy

\[
\tilde{F}_\lambda[\varphi, \lambda] = \tilde{F}[\varphi] - \lambda(N[\varphi] - N_0)
\]

by evolving the corresponding partial differential equations

\[
\begin{align*}
\partial_t \varphi &= \Lambda_D \nabla^2 \delta \tilde{F}_\lambda \\
\partial_t \lambda &= -\Lambda_L \frac{\delta \tilde{F}_\lambda}{\delta \lambda}
\end{align*}
\]

which corresponds to conserved and non-conserved dynamics with mobilities \(\Lambda_D = 10^2\) and \(\Lambda_L = 10^4\), respectively. Using this procedure, we determine the profile \(\varphi(r)\) with Neumann boundary conditions that optimizes \(\tilde{F}_\lambda\) for each value \(N_0\) of the constraint, which yields the minimal free energy path. The profile with the largest free energy \(\tilde{F}\) corresponds to the saddle point and thus to the critical nucleus that we sought. The corresponding profiles \(\varphi(r)\) are shown and analyzed in Fig. 3 in the Letter. Here, the nucleation barrier \(\Delta E\) is given by the difference of the energy of the critical nucleus to the energy of the homogeneous state.

[1] C. N. Yang, Phys. Rev. 85, 808 (1952).
[2] V. Spirin, P. L. Krapivsky, and S. Redner, Phys. Rev. E 63, 036118 (2001).
[3] K. Barros, P. L. Krapivsky, and S. Redner, Phys. Rev. E 80, 040101 (2009).
[4] N. Metropolis, A. W. Rosenbluth, M. N. Rosenbluth, A. H. Teller, and E. Teller, J. Chem. Phys. 21, 1087 (1953).
[5] G. Münster and M. Cañizares Guerrero, J. Stat. Phys. 182, 1 (2021).
[6] D. B. Abraham, Phys. Rev. Lett. 47, 545 (1981).
[7] M. Müller and G. Münster, J. Stat. Phys. 118, 669 (2005).
[8] M. P. A. Fisher, D. S. Fisher, and J. D. Weeks, Phys. Rev. Lett. 48, 368 (1982).
[9] X. Wang, Communications in Mathematical Analysis 7, 50–54 (2009).
[10] K. Blom and A. Godec, Phys. Rev. X 11, 031067 (2021).
[11] J. Parlange, J. Chem. Phys. 48, 169 (1968).
[12] R. Kikuchi and J. W. Cahn, J. Phys. Chem. Solids 23, 137 (1962).
[13] J. W. Cahn and J. E. Hilliard, J. Chem. Phys. 28, 258 (1958).